

Enabling Cost-Scalable Mass Personalization through Hybrid Additive Manufacturing–
Electroforming and Data-Driven Process Modeling

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Abstract

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Mass personalization in metal manufacturing requires processes that accommodate unrestricted geometric variability while preserving cost scalability and surface quality. Additive manufacturing enables geometric freedom but is limited in material and surface reliability, whereas electroforming provides high precision and parallel metal deposition but is sensitive to electrolyte condition and process variability. This thesis addresses this challenge by integrating a hybrid additive manufacturing–electroforming process architecture with a data-driven framework for adaptive electroforming control.

In this work, a hybrid workflow was developed in which additively manufactured polymer molds enable low-cost personalization, while electroforming provides scalable, high-precision metal fabrication. Experimental validation confirmed the feasibility of this approach for producing personalized metal components in a cost-efficient manner. However, sustaining consistent surface quality within this hybrid process requires systematic control of electroforming behavior as the electrolyte evolves during repeated use.

To support this requirement, a comprehensive experimental and data-driven framework was established. An open dataset for additive-free copper electroplating was constructed, comprising 114 electrolyte baths across multiple CuSO_4 concentrations, aging stages, and pulse–reverse deposition conditions. Analysis showed that surface roughness and gloss cannot be reliably predicted using static process parameters or conventional bath measurements alone.

An operational bath-state representation was therefore introduced, inferred from measurable physicochemical bath properties and dynamic electroplating pulse-response features. Machine-learning models were trained to predict surface properties, with Random Forest providing the most robust performance. Scenario-based modeling demonstrated that data-driven adjustment of electroforming parameters can maintain target surface quality within defined chemical and operational bounds, enabling adaptive control within the hybrid manufacturing workflow.

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Contribution of Authors

Chapter 4 of this thesis, entitled **“Low-Cost Manufacturing of High-Precision Personalized Flexures by a Hybrid 3D Printing–Electroforming Technique,”** is based on a peer-reviewed journal article published in *The International Journal of Advanced Manufacturing Technology*. **Reproduced with permission from Springer Nature.**

The experimental design, hybrid manufacturing methodology, electroforming experiments, data analysis, interpretation of results, and manuscript preparation were conducted by the author of this thesis.

Dr. Rolf Wüthrich (supervisor) contributed through scientific supervision, guidance on experimental design, and critical review of the manuscript.

Zhaohan Zheng provided limited technical guidance related to selected 3D printing process steps during the early stages of the work. Her contribution was advisory in nature and did not include experimental execution, data analysis, modeling, or manuscript writing.

All other aspects of the work, including conceptualization, implementation, validation, and documentation, were carried out by the author.

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Symbols

Me	Metal
I_a	Anodic current
I_c	Cathodic current
i_a, i_c, j	Current density
i^0	Exchange current density
t	Time
z	Metal ion charge
F	Faraday's constant
I	Current
Δn	Metal moles
E	Electrochemical cell potential
E°	Standard electrode potential
R	Gas constant
T	Absolute temperature
n	Number of electrons transferred in the reaction
M_e	Concentrations of metal ions at the cathode
M_e^+	Concentrations of metal ions at the anode
Cu	Copper
H_2SO_4	Sulfuric acid
Cu_2SO_4	Copper sulfate
K	Angular stiffness
E	Young modulus
e	Hinge thickness
r	Radius of the circular profile
w	Flexible pivot width
N_{3DP}	Total number of molds
T_{3DP}	3D printing time (in days) per mold
T_{EF}	Electroforming time (in days) per part
T_{Total}	Total fabrication time per part
C_{3DP}	Printing cost per mold assembly
CPH_{3DP}	Hourly rate of 3D printing
CPH_{EF}	Hourly rate of electroforming
R_a	Arithmetic average roughness
R_q	Root mean square roughness
R_z	Maximum height roughness
GU	Gloss units
G_{20}	Gloss at 20°
G_{60}	Gloss at 60°
Q_{dep}	Deposition charge
Q_{pol}	Polishing charge
E_{dep}	Deposition potential
E_{pol}	Polishing potential

$i_{dep-pulse}$	Mean current for deposition phase
$i_{pol-pulse}$	Mean current for polishing phase
$t_{dep-pulse}$	Total pulse time for deposition phase
$t_{pol-pulse}$	Total pulse time for polishing phase
t_{pulse}	Total pulse period
A	Amperes
S	Seconds
$^{\circ}C$	Degree Celsius
H^{+}	Hydrogen ions
Cu^{2+}	Copper ions
SO_4^{2-}	Sulfate ions
M	Molar
mV	miliVolt
K	Number of neighbors
C	Regularization constant
ε	Insensitive margin
γ	Kernel width

Abbreviations

AM	Additive Manufacturing
IoT	Internet Of Things
AI	Artificial Intelligence
EDL	Electrical Double Layer
DC	Direct Current
PRC	Pulse-Reverse Current
PEG	Polyethylene Glycol
SPS	Sulfopropylsulfonate
MEMS	Micro-Electro Mechanical Systems
OLAP	Online Analytical Processing
CV	Cyclic Voltammetry
EIS	Electrochemical Impedance Spectroscopy
CVS	Cyclic Voltammetric Stripping
ML	Machine Learning
RL	Reinforcement Learning
DDPG	Deep Deterministic Policy Gradient
RMS	Reconfigurable Manufacturing Systems
FDM	Fused Deposition Modeling
EBM	Electron Beam Melting
Wire-EDM	Wire Electric Discharge Machining
CNC	Computer Numerical Control
WJC	Water Jet Cutting
ABS	Acrylonitrile Butadiene Styrene
SGB	Spray Glue Bonding
HPB	Hot-Press Bonding
CAP-3DP	Conductive Aerosol Painting – 3D Printing
FEA	Finite Element Analysis
IPA	Isopropyl Alcohol
PLA	Polylactic Acid
CE	Counter Electrode
RFE	Recursive Feature Elimination
KNN	K-Nearest Neighbors
RF	Random Forest
SVR	Support Vector Regression
GPR	Gaussian Process Regression
MI	Mutual Information
ANOVA	Analysis Of Variance
MAE	Mean Absolute Error
RBF	Radial Basis Function

1. Introduction

1.1 Background and motivation

Modern manufacturing owes much of its efficiency to mass production, a paradigm built on the ability to fabricate large quantities of identical components with predictable quality and steadily declining unit cost. This economic model works because the substantial initial investment in tooling, setup, and process optimization is distributed across high production volumes. As long as products remain uniform, the marginal cost of producing additional units continues to drop^{1,2}. Today, however, industries increasingly face the expectation to deliver products tailored to individual users³. Whether for performance, aesthetics, ergonomics, biomedical fit, or functional customization, demand is shifting toward highly individualized components rather than standardized ones^{4,5}. This shift fundamentally challenges the economic logic of mass production⁶. When every part differs in geometry or specification, the fixed costs associated with design changes, calibration, and verification no longer scale efficiently. The traditional mechanisms that reduce cost with volume begin to break down.

These emerging pressures highlight a need for manufacturing strategies capable of delivering both personalization and economic scalability. Achieving such a combination requires a rethinking of how production processes are designed, configured, and controlled.

1.2 From mass production to mass personalization

Mass production and mass personalization represent two ends of a wide manufacturing spectrum. In mass production, the focus is on minimizing variability and maximizing throughput. In contrast, personalization embraces variability as a core requirement. Intermediate concepts such as mass customization attempted to bridge this divide by offering modular variations, but they still rely on a limited set of predefined options and consequently do not accommodate unrestricted geometric freedom^{7,8}.

Personalization changes the fundamental assumptions that underpin most industrial processes. With each new part potentially differing in form, traditional manufacturing workflows, originally built for stability and repetition, face frequent disruptions. Setups must be adapted, quality-control requirements increase, and previously scalable fixed costs become repetitive. As a result, the unit cost often remains high or may even increase as production scales, reversing the economic trend that makes mass production viable.

Thus, the central question motivating much of today's manufacturing research is how to engineer processes that can absorb geometric variability without the penalties typically associated with it.

1.3 Limitations of existing approaches for high-variation manufacturing

Several Industry 4.0 technologies have attempted to address the challenges associated with producing diverse or individualized components. Digital-twin systems, for example, provide real-time virtual representations of factory operations, enabling rapid simulation and adjustment^{9–14}. Cyber-physical frameworks enhance communication between machines, sensors, and decision layers, and reconfigurable manufacturing systems allow equipment to be physically restructured when different tasks are required^{15,16}.

Despite their advantages, these technologies primarily improve responsiveness and coordination rather than fundamentally altering cost scaling. High design variability still forces frequent process adjustments, and hardware-based flexibility often introduces new layers of cost or operational complexity.

Additive manufacturing (AM) is frequently cited as a natural fit for personalization because it can fabricate arbitrary geometries directly from digital models without specialized tooling^{17,18}. The main limitation in the context of mass personalization is that the cost per part does not decrease as production volume increases. Unlike conventional mass production, where tooling and setup costs are amortized over many identical units, mass-personalized workflows often keep geometry-specific steps (design preparation, setup, and processing) tied to each individual part. As a result, scaling the number of unique parts mainly scales total effort, leaving the cost per part effectively fixed, rather than benefiting from volume-driven cost reduction. Yet AM carries its own constraints: build speeds are slow, deposition is inherently serial, machine time is expensive, and post-processing is often required. Numerous techno-economic studies have shown that AM rarely achieves decreasing cost per part with increasing volume unless parts are highly repetitive or machine capacity is near full utilization, conditions that contradict the realities of personalization^{19,20}.

Overall, existing approaches offer adaptability but do not resolve the core economic mismatch between personalization and scalable cost reduction.

1.4 Hybrid manufacturing as a pathway to scalable personalization

One promising direction is to combine complementary manufacturing technologies so that the strengths of one compensate for the limitations of another. Additive manufacturing excels at producing customized molds or mandrels quickly and inexpensively. Electroforming, in contrast, is unmatched in its ability to deposit high-quality²¹, high-precision²² metal features and, importantly, to process many parts in parallel²³.

A hybrid workflow that uses AM to generate personalized polymer mandrels and electroforming to metallize them introduces a compelling architecture:

- Geometry changes no longer require new metal tooling.
- AM provides the needed design freedom at minimal cost.

- Electroforming supplies excellent accuracy, surface properties, and batch scalability.
- Parallelism in electroforming allows costs to decline with part volume, even when designs differ.

This hybrid strategy formulates a new way to think about personalization: instead of reducing variation, one designs the process architecture such that variation does not incur additional cost or at least minimize the the impact.

1.5 Electroforming variability and the challenge of bath evolution

Although electroforming is highly promising for scalable personalization, it remains operationally complex and strongly influenced by evolving chemical and physical conditions during use. Copper-ion concentration, acid composition, conductivity, temperature, and the presence of impurities or additives can all shift as the bath undergoes repeated deposition cycles. These gradual changes alter mass transport, nucleation behavior, and current distribution, ultimately influencing the microstructure, thickness uniformity, and surface finish of the deposited metal²⁴⁻²⁹.

In high-volume industrial production, where part geometries are fixed, baths can be tightly monitored and adjusted to maintain consistent deposition behavior. In a personalized setting, however, these assumptions no longer hold. Because every new geometry redistributes current differently and interacts with the bath in unique ways, the same electroforming parameters may yield inconsistent results from one part to the next. Bath aging exacerbates this unpredictability, making it difficult to maintain stable quality over time.

1.6 Need for intelligent, data-driven control of electroforming

Given the complexity and time dependence of electroforming, achieving reliable performance under personalization requires adaptive and predictive control tools. Recent advances in data analytics and machine learning demonstrate strong potential for modeling electrochemical processes, uncovering relationships that are not easily accessible through first-principles analysis³⁰⁻³⁸. These tools can detect subtle changes in bath state, correlate them with observed outcomes, and infer how process parameters should be modified to maintain desired deposition characteristics.

By analyzing how surface characteristics and deposition behavior respond to different bath states and operating conditions, it becomes possible to construct a digital framework capable of:

- identifying indicators of bath aging,
- predicting surface quality outcomes,

- recommending or adjusting process parameters in real time,
- and extending bath usability while minimizing maintenance.

Transforming electroforming from an experience-driven craft into a data-informed, adaptive process is essential for enabling it to function as a reliable platform for mass personalization.

1.7 Problem statement

As a result, this thesis addresses two fundamental and interconnected problems that currently prevent electroforming from functioning as a scalable and economically viable platform for mass personalization.

Problem 1 – Creating a Manufacturing Process Architecture Where Cost Decreases With Volume Despite Personalization

Personalized production traditionally leads to increased per-unit cost because each design change introduces new tooling requirements, calibration steps, and process adjustments. Unlike mass production, where unit cost decreases with volume, there is currently no manufacturing architecture that enables cost-down behavior when every part is geometrically unique.

Accordingly, the first research problem is:

Can electroforming be integrated with complementary manufacturing technologies, such as additive manufacturing, to create a hybrid process in which the cost per personalized part decreases as production volume increases, similar to the economics of mass production?

Problem 2 – Establishing Reliable Digital Control for Electroforming Under Bath Aging

Even with an economically scalable architecture, electroforming cannot support mass personalization without consistent and predictable process control. Electroforming performance is highly sensitive to bath aging, ion depletion, and additive degradation. These factors cause deposition quality to drift over time, making traditional empirical methods insufficient.

Thus, the second research problem is:

How can data-driven and predictive control approaches be developed to ensure stable, high-quality electroforming outcomes, independent of bath aging, so that personalized components can be produced reliably at scale?

Combined Research Challenge

To develop both a scalable hybrid manufacturing process and a reliable digital control framework that enable electroforming to produce personalized micro metal parts with decreasing cost per unit and consistent quality across evolving bath states and geometries.

1.8 Research objectives

This thesis pursues two overarching objectives corresponding to the two major research problems identified.

Objective 1 – Develop a novel manufacturing approach that integrates additive manufacturing with electroforming to enable the production of mass-personalized micro metal parts, with a cost per part that decreases as production volume increases.

This includes:

- *Evaluating the geometric and functional capabilities of AM-fabricated molds.*

A key objective of this study is to evaluate the suitability of additively manufactured molds as foundational elements for producing personalized metal micro-components. This requires a systematic assessment of their geometric fidelity, dimensional stability, and functional adequacy. The research will determine whether low-cost polymer molds can accurately reproduce intricate features, maintain structural integrity throughout handling and electrochemical processing, and provide interfaces compatible with subsequent metal deposition. This evaluation establishes the conditions under which printed molds can reliably act as precision formers and how their characteristics may influence downstream mechanical or assembly requirements.

- *Demonstrating the fabrication of personalized micro-mechanisms.*

A second component of this objective is to demonstrate the feasibility of fabricating personalized micro-mechanisms through the hybrid use of additive manufacturing and electroforming. This involves selecting representative designs with varying geometric and functional complexity and examining how effectively the hybrid process can accommodate such variations. The research will investigate the ability of the workflow to produce compliant structures, articulated elements, or multi-axis assemblies while preserving the geometric intent specified in the initial digital model.

- Analyzing how hybridization influences the cost structure and how unit cost scales with production volume.

The final task under this objective is to analyze how integrating additive manufacturing with electroforming impacts the overall cost structure of producing personalized components. This includes examining how design variability, mold throughput, and parallel electroforming influence unit-cost trends as production volume increases. The goal is to determine whether the hybrid process can achieve decreasing per-part cost despite producing unique geometries and to identify the operational factors that enable mass-production-like scaling in a personalized manufacturing environment.

Objective 2 – Develop a systematic approach for adapting electroforming process parameters to the evolving state of the plating bath, thereby ensuring stable and reproducible part quality in aged electrolyte conditions.

This includes:

- Constructing and curating a multi-bath electroforming dataset.

The first task under this objective is to construct and curate a comprehensive multi-bath electroforming dataset that captures the variability inherent in electrolytes of different ages, compositions, and usage histories. This requires systematic data collection across multiple baths, including measurements of chemical properties, physical characteristics, pulse-level electrical inputs, and resulting surface outputs. The dataset will serve as the foundation for quantitative analysis and model development, ensuring that the subsequent framework reflects realistic process variability rather than idealized or single-bath behavior.

- Identifying signatures of bath aging and correlating them with surface outcomes.

A second task is to identify measurable indicators that characterize how the electrolyte evolves over time. This involves analyzing trends in chemical and physical bath properties and determining which variables reliably signal changes in bath condition. The research will examine how these indicators relate to deviations in deposited surface properties, enabling the definition of bath-aging signatures that can be incorporated into predictive models.

- Developing predictive models capable of estimating deposited surface properties from the current electroforming bath state.

The next component focuses on developing machine learning models capable of estimating deposited surface properties from the current state of the electroforming bath. These models will be trained to map relationships between bath characteristics, process parameters, and resulting surface metrics such as roughness or gloss. The objective is to generate predictive tools that can anticipate the quality of a deposition cycle before it occurs, thereby reducing dependency on empirical trial-and-error adjustments.

- Developing predictive models that can adjust or recommend process parameters to maintain desired deposition performance as the bath ages.

The final task is to develop predictive machine learning models that recommend or adjust process parameters in response to bath aging. This requires analyzing how different pulse settings, waveform configurations, and operating conditions interact with the measured bath state. The resulting models will be designed to identify parameter combinations that maintain required deposition performance despite electrolyte degradation. This capability forms the basis of an adaptive control strategy intended to stabilize surface quality, extend bath usability, and support reliable production of personalized components.

1.9 Significance of the study

The significance of this research lies in its contribution to bridging a long-standing gap between the economic efficiency of mass production and the design freedom required for mass personalization, an area repeatedly highlighted as unresolved in the manufacturing literature^{5,6}. While advances in cyber-physical systems, digital twins, and reconfigurable manufacturing have improved adaptability, they have not delivered scalable cost reduction when every component is geometrically unique^{9–14}. This thesis introduces a hybrid manufacturing architecture that directly addresses this limitation by combining additive manufacturing and electroforming in a manner that restores cost-down behavior while accommodating unlimited geometric variation.

A second major contribution is the development of one of the first systematically structured electroforming datasets explicitly designed to study bath variability, aging effects, and process parameters-deposition outcome interactions. Existing electroforming studies typically rely on controlled, single-bath experiments and therefore cannot capture the complex, multi-source variability seen in real manufacturing environments^{39–43}. By integrating data from multiple baths, compositions, and aging stages, this work provides a foundation for reproducible research and future benchmarking in a field where open datasets are nearly nonexistent.

Furthermore, the research establishes a quantitative framework for identifying and predicting the progression of bath aging, an issue historically managed through qualitative operator experience

rather than measurable indicators ²⁴. By uncovering relationships between chemical drift, electrical response, and resulting surface properties, the study enables a more rigorous and science-based understanding of bath evolution, offering industry tools for improved electrolyte maintenance and lifetime prediction.

The thesis also introduces predictive, machine-learning models capable of estimating key surface properties of electroformed parts from the current bath state. While machine learning is gaining traction in electrochemical sciences, its application to electroforming, especially in the context of aging baths and personalization, remains limited ⁴⁴. The models developed here simultaneously address multiple surface outputs, offering a more comprehensive view of part quality than the single-response approaches commonly reported in the literature.

Finally, the integration of these predictive tools into an adaptive parameter-recommendation framework demonstrates tangible industrial benefits. The ability to adjust pulse parameters in response to bath aging supports consistent quality, reduces scrap rates, minimizes downtime, and extends bath usability. These advantages are crucial for enabling the economic sustainability of personalized manufacturing systems, where variability is inherent and traditional process-control strategies are inadequate. Together, these contributions establish a scientific and technological foundation that moves electroforming toward becoming a digitally controlled, scalable platform suited for the next generation of high-variation manufacturing.

2. Literature

2.1 Mass personalization: A modern manufacturing approach

2.1.1 What does mass personalization mean in modern manufacturing?

The first manufacturing paradigm introduced in the 18th century was craft production, which resulted in the costly fabrication of products based on customer desires with very low productivity⁶. Then, during the past century, the mass production paradigm was raised to reduce fabrication costs, notably by spreading the manufacturing overhead over a Large number of identically produced parts¹. Although mass production has indisputable advantages, such as increased productivity and reduced production costs, it suffers from a lack of flexibility⁴⁵. Mass customization, a terminology first introduced in academia in 1987, is a natural evolution of mass production⁴⁶. Mass customization allows the customer to customize the product by choosing from a number of predefined features. Fabrication lines are then set up to allow the production of these customized products.

However, today, personalized products are demanding more in the manufacturing sector³. Unlike traditional customization, in personalization, customers are actively involved in the design process. This allows them to go beyond preset features and collaboratively fabricate products that fit their individual preferences. Accordingly, this shift leads to production characterized by low volume and high diversity.

Mass personalization, as an evolving concept, represents a departure from traditional customization, particularly within the Industry 4.0 landscape. This paradigm enables manufacturers to move from segment-based product variations to truly individualized offerings, tailoring products and services to meet each customer's specific needs and preferences. The distinction between mass customization and mass personalization is crucial; while the former focuses on producing goods that cater to a defined group of customers, the latter aims to create unique products for each customer, thereby enhancing individual satisfaction and engagement^{7,8}. Figure 2.1 shows the relationship between quantity, cost, and variety of products for the recent manufacturing paradigms.

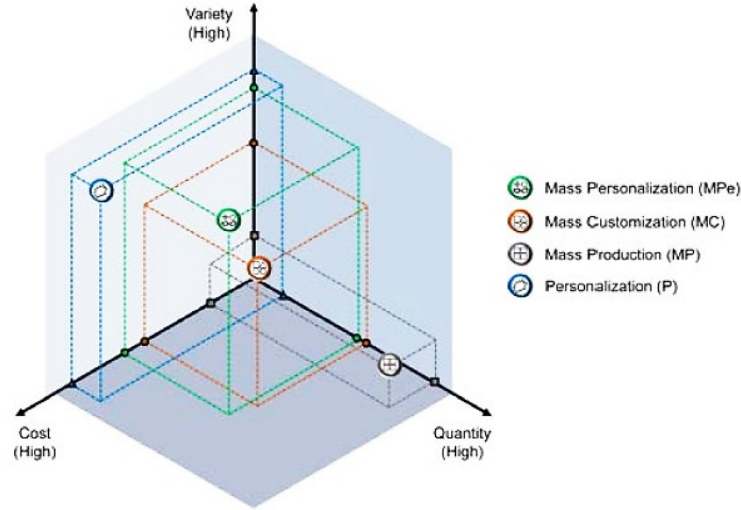


Figure 2.1 Schematic Quantity - Variety - Cost relationship in different manufacturing paradigms

At the core of mass personalization is the integration of advanced technologies, particularly information technology, which facilitates the collection and analysis of vast amounts of customer data. This data-driven approach enables businesses to deeply understand individual customer preferences and to respond with tailored offerings. Unlike mass customization, which often relies on predefined parameters to create variations of a product, mass personalization leverages real-time data to create products that can be uniquely designed for each customer⁸. This transformation is particularly evident in industries such as automotive, where manufacturers are beginning to shift from offering customizable options to providing fully personalized vehicles that reflect the specific desires of individual buyers⁸.

The advent of Industry 4.0 technologies, including the Internet of Things (IoT), artificial intelligence (AI), and advanced manufacturing techniques such as 3D printing, plays a pivotal role in enabling mass personalization. These technologies allow for greater flexibility in production processes and facilitate collaboration in product design between manufacturers and customers. For instance, 3D printing not only supports the creation of unique products but also contributes to sustainability by reducing waste and minimizing the carbon footprint associated with traditional manufacturing methods^{7,47}. This capability aligns with the broader goals of sustainable consumption, as it encourages the production of goods that are not only personalized but also environmentally responsible.

Moreover, the concept of mass personalization extends beyond individual products to encompass the entire value-creation network. It involves horizontal integration across various stakeholders,

vertical integration within manufacturing systems, and end-to-end engineering throughout the product lifecycle. This holistic approach ensures that personalized products are not only designed with the customer in mind but are also produced and delivered efficiently, thereby enhancing overall customer satisfaction and loyalty ⁴⁷. The implications of this transformation are profound, as businesses that successfully implement mass personalization can differentiate themselves in competitive markets and foster deeper connections with their customers.

The subsequent sections delve into the benefits and challenges that mass personalization presents in modern manufacturing. By exploring both the advantages and potential obstacles of mass personalization, the following provides a comprehensive understanding of its transformative impact on the manufacturing landscape.

2.1.2 How does mass personalization benefit consumers and manufacturers?

Mass personalization in the manufacturing sector possesses several benefits, such as improved operational efficiency, consumer fulfillment, and competitive positioning. This technique enables industries to develop personalized items at a scale comparable to traditional mass production while integrating the advantages of both paradigms. The following explains the major benefits of mass personalization, substantiated by relevant studies.

Cost-effective individualized production. One of the fundamental benefits of mass personalization is the ability to suit unique consumer requests while being cost-effective. Companies can produce personalized items at lower prices by embracing sophisticated manufacturing technologies such as additive manufacturing and digital twins. This allows manufacturers to offer personalized products while maintaining manufacturing costs comparable to mass-produced items ^{48,14}. For instance, companies can employ modular designs that allow for quick feature modification, avoiding the requirement for major retooling and setup costs ⁴⁹.

Active engagement with customers. Another significant advantage of mass personalization is improved customer engagement and fulfillment. Mass personalization empowers customers to actively participate in the design process, resulting in products that better reflect their preferences and lifestyles. This interaction promotes devotion to the company in addition to increasing customer fulfillment since customers feel a greater connection to items that are adapted to their unique needs ⁵⁰. According to research, when customers participate in the customization process, they are more likely to evaluate the product's value favorably, which can lead to increased sales and returning customers ⁵¹.

Increased flexibility. Manufacturers are able to adjust quickly to market changes and customer preferences in mass personalization. In an era when consumer preferences change quickly, the ability to adjust manufacturing lines to accommodate new designs or features with minimal downtime or cost increases is critical ⁵². This flexibility is especially valuable in areas like fashion and consumer electronics, where trends can shift overnight, needing a quick reaction from producers ⁵³.

Improved sustainability. Mass personalization contributes to improved inventory management and reduced waste. Traditional manufacturing often results in costly overproduction and excess inventory. Mass personalization, on the other hand, enables a more demand-driven approach. Products are manufactured in response to individual consumer demands. This change reduces the risk of unsold inventory, matches production with real market demand, and improves overall efficiency. Manufacturers can implement just-in-time sustainable production strategies to shorten lead times and optimize the utilization of resources ⁴⁸.

Optimized efficiency. The implementation of digital technology in mass personalization improves operational efficiency. This happens through enhanced data analytics and decision-making abilities. Manufacturers can gather and evaluate data on consumer preferences and manufacturing processes. Consequently, they can make more informed decisions that improve product design and manufacturing operations. This data-driven strategy enables continual improvement in product offers and operational methods, strengthening firms' competitive edge in the marketplace. Currently, cloud-based platforms and powerful analytics tools provide real-time monitoring and modification of manufacturing processes. These tools allow manufacturers to adapt quickly to consumer requests and market dynamics ^{7,54}.

Increased profitability. Mass personalization can also lead to increased profitability for manufacturers. By offering customized products that command higher prices due to their unique value propositions, companies can enhance their profit margins. The ability to differentiate products in a crowded marketplace through personalization can also create a competitive advantage, allowing manufacturers to capture market share from competitors who may be slower to adapt to consumer preferences. This strategic positioning is particularly important in industries where consumers are prepared to pay extra for products tailored to their unique needs ⁵⁵.

Mass personalization, in summary, provides several benefits, including cost savings, higher customer satisfaction, production flexibility, better inventory management, and increased profitability in the manufacturing sector. Manufacturers can successfully manage the intricacies of contemporary markets while providing tailored, high-quality products that accommodate the varied demands of customers.

2.1.3 What are the challenges of mass personalization in the context of Industry 4.0?

The emergence of Industry 4.0 has significantly transformed manufacturing paradigms, particularly through the lens of mass personalization or individualization. This transformation promises enhanced customer satisfaction and operational efficiencies, but it also presents challenges that must be addressed to harness its full potential. The following examines the major challenges of mass personalization in the context of Industry 4.0, substantiated by relevant studies.

Complexity. The complexity of manufacturing processes presents a significant challenge. Expansion of product variety necessitates innovative solutions in quality assurance and process improvement ⁵⁶. To meet unique customer needs, companies face the dual pressures: maintaining low production costs and ensuring high customization levels. This led to a fundamental conflict

between customization and cost efficiency ⁵⁷. Additionally, this complexity can increase production steps with longer lead times and undermine the competitive advantage that mass production traditionally offers ⁵⁸. Therefore, flexibility in manufacturing processes is essential, requiring companies to develop solutions that can accommodate a broader range of product types ⁵⁶. This, in turn, necessitates a robust framework for managing production details while upholding quality standards.

Investment, integration, and cybersecurity risks. The integration of advanced technologies poses additional challenges. Industry 4.0 and personalized manufacturing rely heavily on the seamless connectivity of various components, including autonomous robots, cloud computing, and additive manufacturing ⁵⁹. However, achieving this level of integration requires significant investment in infrastructure, technology, and a skilled workforce capable of operating and maintaining these systems. The transition to a digitally connected manufacturing environment can also introduce vulnerabilities, particularly concerning cybersecurity. As companies become more reliant on interconnected systems, the risk of cyberattacks increases. This can jeopardize not only individual businesses but also the entire supply chain ⁵⁹.

Unemployment. Another critical challenge is the potential for mass unemployment as automation and digitalization reshape the workforce landscape. The transition to Industry 4.0 and personalization necessitates a workforce equipped with new skills to operate and maintain advanced manufacturing technologies. This shift raises concerns about the adequacy of current educational and training programs to prepare workers for the demands of a highly automated environment. Additionally, the societal implications of such a transition, including economic disparities and the potential for job displacement, must be carefully considered and addressed ⁵⁸.

Supply chain challenges. The concept of mass personalization also introduces complexities in supply chain management. As companies seek to provide tailored products to individual customers, they must navigate the details of managing diverse inventories and production schedules ⁵⁷. This complexity can lead to increased operational costs and logistical challenges, particularly for smaller manufacturers who may lack the resources to implement sophisticated supply chain solutions. Furthermore, the need for real-time data and persistent information throughout the value chain is critical for achieving the desired level of personalization. However, establishing such systems can be resource-intensive and technically challenging ⁵⁹.

Mindset shift barriers. In addition to operational challenges, there are also significant cultural and organizational hurdles to overcome. The successful implementation of Industry 4.0 and mass personalization requires a shift in organizational mindset toward embracing innovation and collaboration among humans, machines, and technologies ⁵⁸. This cultural shift can be difficult to achieve, particularly in traditional manufacturing environments where established practices and resistance to change may hinder progress. Companies must foster a culture of continuous learning and adaptability to thrive in this new industrial landscape ⁶⁰.

Environmental concerns. Furthermore, the environmental implications of mass personalization and Industry 4.0 cannot be overlooked. While these advancements hold the potential for more sustainable manufacturing practices, they also raise concerns about resource consumption and waste generation. These concerns are associated with increased production variability ⁶⁰. The

challenge lies in balancing the benefits of personalization with the need for sustainable practices that minimize environmental impact. Companies must adopt holistic approaches that consider the entire lifecycle of products, from design to disposal, to ensure that mass personalization contributes positively to sustainability goals ⁶⁰.

Updating regulations. Lastly, the regulatory landscape presents another layer of complexity. As Industry 4.0 technologies evolve, existing regulations may become outdated or insufficient to address new challenges related to data privacy, intellectual property, and labor rights. Policymakers must work collaboratively with industry stakeholders. This collaboration develops frameworks that support innovation for mass personalized production and protects the rights and interests of all parties involved ⁶¹.

Scaling the cost. In mass personalization, the central economic bottleneck is the weak (or absent) economy of scale: because each unit can be geometrically unique, a non-trivial share of work remains order-specific (e.g., design/data preparation, validation, setup decisions, inspection planning, and process adjustments). This prevents fixed costs and learning effects from being amortized across identical parts in the same way as conventional mass production, so total effort tends to grow roughly with the number of unique units, keeping cost per part nearly constant even as volume increases. The mass-customization literature repeatedly frames this as a core implementation challenge: unless the workflow is redesigned around modular product architectures and reconfigurable processes that limit variety-induced complexity, customization can quickly erode productivity and cost competitiveness. Additive manufacturing is often proposed as a technical enabler because it removes dedicated tooling, but cost models and techno-economic studies show AM is typically most competitive in low-to-medium volumes and is highly sensitive to machine-time, post-processing, and capacity utilization, factors that further constrain cost-down with volume when every build is effectively different ^{6,19,62,63}. This work directly targets this cost-scaling bottleneck in mass personalization by developing a workflow designed to reduce the effective cost per part as production volume increases. This motivation and the proposed approach are presented in more detail in Chapter 3.

While the potential benefits of mass personalization and Industry 4.0 are substantial, the challenges associated with their implementation are equally significant. From operational complexities and workforce implications to cultural shifts and regulatory considerations, addressing these challenges will be crucial for realizing the full potential of this new industrial paradigm. As industries navigate this transformative landscape, a concerted effort to innovate, collaborate, and adapt will be essential for success.

2.2 Integrating additive manufacturing and electroforming: Potentials and challenges

2.2.1 The rise of additive manufacturing and electroforming

The advent of AM and electroforming in modern manufacturing reflects a paradigm change in production methods. This change is driven by technological breakthroughs and shifting market

needs. Additive manufacturing, commonly referred to as 3D printing, involves building components layer by layer based on digital models, allowing for remarkable design flexibility and individualization. This technique has expanded from its early uses in fast prototyping to embrace a wide range of industrial applications. It can produce end-use parts, tools, and intricate geometries that are challenging or impossible to create through traditional manufacturing processes^{17,18}.

One of the primary motivators for the emergence of additive manufacturing is its capacity to decrease material waste and improve sustainability. Traditional subtractive manufacturing methods often involve removing material from a larger block, leading to significant waste. In contrast, additive manufacturing creates components by adding material precisely where it is required, resulting in improved resource efficiency^{64,65}. This attribute is consistent with the increased emphasis on sustainable manufacturing techniques, as companies seek to minimize their environmental impact while being profitable⁶⁶. Additionally, the capability to manufacture parts as needed eliminates the necessity for extensive inventories, resulting in additional cost savings and enhanced efficiency⁶⁷.

Electroforming, a related technology, complements additive manufacturing by facilitating the production of highly detailed and precise metal components. This process involves depositing metal onto a sacrificial conductive 3D-printed mold or substrate. It can manufacture intricate shapes and fine features that are essential in the automotive, aerospace, and electronics sectors. The integration of additive manufacturing and electroforming can lead to hybrid manufacturing processes that leverage the strengths of both technologies, resulting in parts that are not only complex in design but also robust in performance^{68,23}.

As businesses embrace digital transformation and novel production processes in Industry 4.0, the combination of additive manufacturing and electroforming is likely to play a critical role in determining the future of manufacturing.

2.2.2 Advances in the integration of additive manufacturing and electroforming

One of the emerging and transformative concepts in modern manufacturing is hybrid manufacturing. This approach allows manufacturers to combine existing traditional processes, such as electroforming, with newly developed technologies like AM. By integrating these processes, manufacturers can leverage the strengths of both techniques to overcome limitations inherent in using them individually. This integration has some benefits for industries that are listed below:

Complex personalized geometries AM excels in producing complex, intricate 3D printed molds that are challenging or impossible to accomplish using conventional machining techniques. Electroforming complements this by enabling the precise deposition of metal atoms onto these 3D-printed molds. By combining AM to fabricate detailed molds with electroforming metal deposition, manufacturers can create highly personalized metal parts featuring complex internal geometries⁶⁹.

Enhanced material properties Electroforming adds significant value by depositing a low-stress thick, uniform layer of metal onto AM-fabricated parts. This process improves the mechanical strength and durability of the components, addressing the limitations of some 3D-printed materials. This approach also enables the manufacturing of composite structures using different materials tailored to the desired properties of personalized parts, allowing for greater personalization and optimization of performance ^{69,70}.

Improved surface finish and eliminated post-processing Electroforming provides a high-quality and smooth surface finish without requiring any post-processing. Consequently, the resulting components show both aesthetic appeal and functional precision. This is particularly advantageous in applications requiring precision, such as electronics or medical devices ^{69,71}.

Electrical conductivity The electroforming process allows for the creation of lightweight components with excellent electrical conductivity from non-conductive or semi-conductive 3D printed substrates. This makes it ideal for industries like electronics, where metallic components with precise electrical properties are crucial ⁷².

Cost and material efficiency The combination of AM and electroforming in low-volume or personalized production minimizes material waste and reduces production costs by using resources efficiently. It also allows for the use of cheaper materials in the AM phase, with a higher-value metal layer added via electroforming only where needed ^{68,71}.

Scalability for microfabrication Integration of these technologies enables consistent production of high-precision microfabricated parts ⁶⁹. This is particularly advantageous for sectors such as aerospace and medical devices, where small-scale, high-precision components are essential ⁶⁸.

Mass production. This integration allows for the simultaneous electroforming of multiple molds with customized dimensions and geometries in the same electrolyte bath. Additionally, while electroforming is being carried out on the current batch, the next set of molds can be 3D printed, optimizing mass production efficiency ^{68,69}.

2.2.3 Challenges in integrating additive manufacturing with electroforming

Although combining additive manufacturing and electroforming presents a promising low-cost route for fabricating complex personalized parts, several technical challenges remain and must be addressed to ensure high-quality outcomes. Key challenges include:

Material compatibility. Electroforming primarily includes metal deposition, while AM processes frequently employ a broader range of materials, including polymers, ceramics, and composites. Ensuring compatibility between the initial AM-built substrate and the electroformed metal layer

can be challenging, as differences in thermal expansion, adhesion properties, conductivity, and structural integrity may lead to delamination or defects in the final product ⁷³.

Part removal without damage. Electroformed parts must be separated from the 3D-printed mold without causing damage. This can be especially challenging with complex geometries or fine structures, as mechanical removal can break or distort the electroformed part ²³.

Surface quality and adhesion. The success of electroforming largely depends on the substrate's surface quality, as rough or inconsistent surfaces can result in uneven metal deposition. AM processes, such as powder-bed or fused deposition modeling, may produce substrates with high surface roughness. Thus, additional post-processing steps, particularly polishing or coating, are often required to prepare AM surfaces for electroforming, adding time and cost ⁷⁴.

Dimensional control and precision. Electroforming deposits metal in a layer-by-layer fashion, which can affect dimensional accuracy if not carefully controlled. Integrating this process with AM, which also builds in layers, requires precise coordination to prevent dimensional inconsistencies. Variability in the deposition rate during electroforming may further complicate achieving tight tolerances, especially for intricate geometries ²³.

Quality control and inspection. Monitoring the quality of hybrid 3D printed-electroformed parts is complex, as each process introduces potential defects. 3D printed molds may contain porosity or layer misalignment ⁷⁵, while electroforming can introduce issues such as voids, inclusions, or uneven thickness ^{76,27}. Ensuring the final product's integrity demands advanced inspection techniques, including non-destructive testing, which can increase the overall production cost ⁷⁷.

In addition to the inherent challenges of electroforming into the 3D printed molds, electroforming also presents its own set of limitations and quality concerns. One significant issue is the instability of the electroforming process over time, particularly when applied to large-scale production and mass personalization. As the electroforming bath ages, variations in the electrolyte composition and operational factors can lead to inconsistencies in the thickness, mechanical properties, and surface finish of the electroformed parts ⁷⁸. These challenges are discussed in greater depth in the following sections, where potential solutions and optimization strategies for improving stability and reproducibility in electroforming are thoughtfully explored.

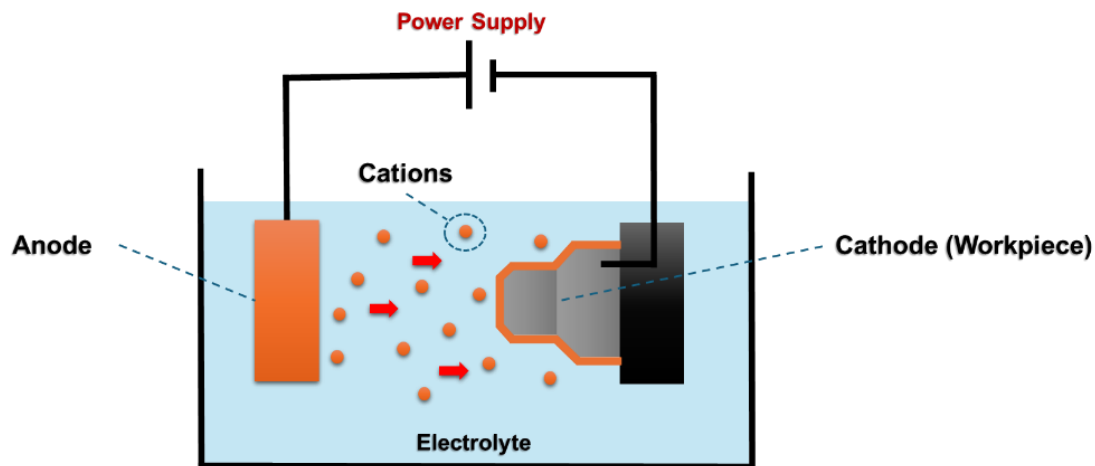
The research conducted in this field so far suggests that personalized parts can, in principle, be fabricated using electroforming within additive-manufactured molds. This approach offers significant potential for creating custom metal components, particularly in applications requiring precision and detail. However, several open questions remain regarding the overall feasibility of this method, mainly in the context of fabricating personalized parts. Critical concerns include achieving consistent quality, maintaining the required dimensional accuracy, and ensuring that electroformed components meet the necessary mechanical and surface finish specifications. Further studies are needed to address these challenges and optimize the process for industrial-scale applications.

2.3 Electroforming process: Potentials and challenges

2.3.1 Definition of electroforming and electroplating

Electroforming is a technique that employs electrodeposition, similar to electroplating, to produce intricate and precise metallic components. This process is particularly advantageous in producing components that are hard to machine conventionally. It can provide elevated dimensional precision repeatability, low surface roughness, and the capability to tailor mechanical properties ²³. The general principle of electroforming, which will be described in detail later, is the electrochemical deposition of metal ions from the solution onto a conductive substrate, where these ions are reduced at the cathode to form a solid metal layer. Figure 2.2 shows a typical electroforming cell ⁷⁹.

The primary difference between electroforming and electroplating lies in their applications. While electroplating is utilized to coat an existing object with a thin metallic layer for decorative or protective purposes, electroforming produces a standalone metallic object. This is achieved by depositing metal onto a mold or mandrel of the desired shape, ultimately producing a functional or decorative component as a separate entity ⁶⁹.



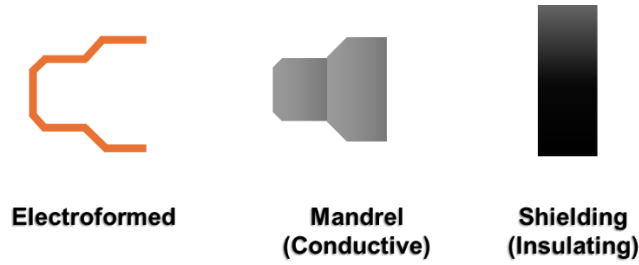


Figure 2.2 Electroforming cell

2.3.2 Mechanism of electroforming

The fundamental principles of electroforming originated in electrochemistry, particularly the movement of electrons between electrodes in an electrolytic medium. When a metal is introduced into an electrolyte solution, spontaneous dissolution takes place under certain conditions. During this dissolution, metal atoms detach from the crystalline lattice and ionize, forming positively charged cations that diffuse into the electrolyte. This anodic dissolution results in an accumulation of cations near the metal surface, creating a positive charge in the surrounding electrolyte. At the same time, the departure of these cations leaves an equivalent negative charge on the metal surface, maintaining electrostatic balance. Electrostatic forces subsequently act to draw the cations back to the negatively charged surface of the metal. This interaction between the attractive and repulsive forces establishes a dynamic equilibrium, which is mathematically illustrated by the following equation ⁸¹:



The double arrow denotes that equilibrium is attained when the forward and reverse processes occur at the same rate. This means that anodic dissolution (or oxidation), where metal atoms move into the electrolyte, and cathodic deposition (or reduction), where electrostatic attraction pulls ions back to the metal surface, occur simultaneously, maintaining a dynamic balance between both directions ⁸⁰. This equilibrium is fundamental to understanding the electrochemical behavior of metals in electrolytic environments, as it governs the kinetics of both dissolution and deposition processes.

The process of anodic dissolution and cathodic metal deposition includes the transfer of charge, resulting in the production of an electric current. Anodic current (I_a) is considered positive, while cathodic current (I_c) is negative, following standard convention. Depending on the scenario, either the total current or the current density (current per unit area) may be relevant. Current density is usually denoted as i_a or i_c , and sometimes as j .

At equilibrium, no net reaction takes place because the rates of anodic dissolution and cathodic deposition are balanced. This equilibrium can be mathematically expressed as follows.

$$|I_c| = |I_a| = I^0 \text{ or } |i_c| = |i_a| = i^0 \quad 2.2$$

The exchange current density, represented as i^0 , indicates the rate at which the dynamic equilibrium of these reactions occurs, as shown in equation 2.1.

However, when two metal electrodes are placed in an electrolyte and linked to a current source, the equilibrium state will be disturbed due to the varying rates of anodic dissolution and cathodic deposition, as outlined in equation 2.1. These processes no longer occur at equal rates, with the current source driving the system away from equilibrium. In this electrical circuit, one electrode functions as the cathode, where metal deposition occurs, while the other acts as the anode, where metal is dissolved to replace the ions removed from the solution during deposition. The deposition and dissolution rates are influenced by the voltage applied by the external current source. The reaction rates at both electrodes are accelerated by an increase in voltage, which boosts the rate of metal deposition at the cathode and the rate of metal dissolution at the anode ⁸¹.

The cathodic deposition process follows Faraday's law when the electrodes are connected to a current source. This principle posits that the deposited metal mass on the cathode is precisely correlated with the number of electrons involved in the electrochemical reaction, indicating a proportional relationship between the two factors. This relationship can be expressed mathematically as below ⁷⁹:

$$\Delta n = \frac{It}{zF} \quad 2.3$$

The formula shows a relationship between the time (t), metal ion charge (z), Faraday's constant (F), and current (I) to calculate the change in the amount of metal moles (Δn) deposited. The precise control of the thickness of the electrodeposited layer can be achieved by altering the current, duration, and electrolyte concentration ⁷⁹.

Furthermore, the Nernst equation is utilized in electrodeposition to determine the necessary potential for depositing a particular metal under specific conditions, as shown in equation 2.4 ⁷⁹.

$$E = E^\circ - \frac{RT}{nF} \ln \frac{M_e}{M_e^+} \quad 2.4$$

In this equation, E stands for the electrochemical cell potential, whereas E° represents the standard electrode potential, which indicates the likelihood of a half-cell in an electrochemical system to either accept or donate electrons. The gas constant is symbolized by R , the absolute temperature by T , and the number of electrons transferred in the reaction by n . The concentrations of metal ions at the cathode and anode are represented by the terms M_e and M_e^+ , respectively. This Equation is instrumental in determining the thermodynamic viability of an electrochemical reaction.⁷⁹

A fundamental aspect of electrodeposition is the transport and replenishment of metal cations at the cathode, as they are continually deposited from the solution. The efficiency of this process is greatly affected by the rate at which new ions and necessary uncharged particles move from the bulk solution to the cathode surface. The hydrodynamic surroundings around the cathode have a significant impact on this rate.

There are three primary mechanisms⁸¹ responsible for the transport of ions to the electrode. Figure 2.3 illustrates these mechanisms, focusing on the formation of the electrical double layer (EDL) when an object interacts with a fluid. The EDL consists of two charged layers: the first, known as the surface charge, is formed by ions chemically bonded to the object's surface. The second, or diffuse layer, contains free ions loosely associated with the surface, which move in the fluid due to electric forces and thermal motion. The ion-transporting mechanisms can be elaborated more as follows:

- **Migration:** This occurs when the applied voltage across an electrolysis cell creates a potential gradient, causing anions to move toward the anode and cations toward the cathode. Despite this, the low velocity of ion movement and interactions with the solvent generally make the contribution of migration to overall ion transport relatively small.
- **Diffusion:** This occurs when charged or uncharged species move across the diffuse layer toward the electrode surface, driven by a concentration variation or chemical potential. The electrode reaction uses up these species, leading to a concentration variation within the electrode interface and the solution, which encourages additional diffusion to restore the depleted species.
- **Convection:** This refers to the movement of reactants with the electrolyte, as opposed to their movement through it, and occurs through stirring, workpiece motion, or natural circulation due to density differences. While convection enhances mass transport by bringing the solution to the diffuse layer near the electrode, the thickness of this layer, which governs ion diffusion, is reduced with stronger agitation.

Additionally, the zeta potential (the electric potential difference between the stationary fluid layer attached to the object's surface and the moving fluid beyond it) is a key parameter. Together with the double-layer structure, the zeta potential is critical for understanding electrochemical behavior at interfaces, such as electrodes, where ion interactions heavily influence deposition processes⁸². Each of these mechanisms plays a crucial role in determining the overall ion flux and, consequently, the rate of electrodeposition.

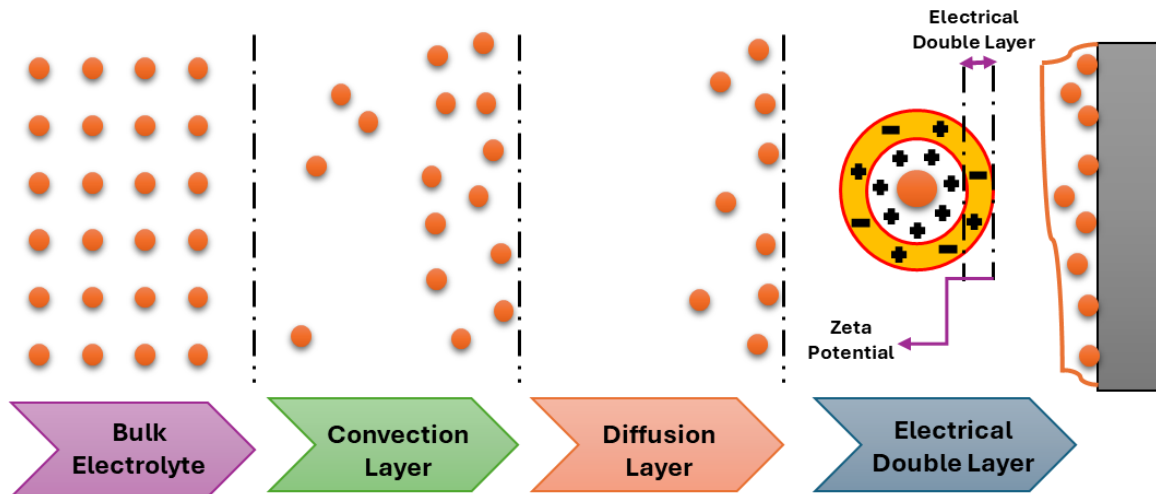


Figure 2.3 The mechanism of electroforming

Despite nearly two centuries of research into ion transport and electrodeposition mechanisms in thin-layer electroplating, electroforming remains a highly challenging process. While significant progress has been made in understanding and modeling these fundamental aspects, electroforming introduces additional challenges that require precise control over factors such as current density, cell potential, electrolyte composition, electrode characteristics, and temperature. The goal is not just to manage the deposition rate but to achieve thick metal layers (often exceeding 1 mm) while ensuring high surface quality and material properties^{83,24}.

As the thickness of the deposits increases, the complexity of the process grows. Extended deposition times can cause fluctuations in these parameters, affecting the uniformity, deposition rate, and quality of the metal layer. For instance, variations in electrolyte composition or temperature over time may further influence the mechanical properties and structural integrity of the electroformed metal⁸⁴.

To ensure optimal results, it is essential to continuously monitor and adjust key process parameters throughout the deposition period. This ongoing optimization helps maintain consistent control over quality, particularly in processes requiring homogeneous thick layers or high precision.

2.3.3 Electroforming parameters and their effects

Improving the electroforming process demands an in-depth understanding of the critical variables and their effects on the final surface quality and material properties of the deposited layer. These parameters can be grouped into the following categories:

Electrode characteristics play a significant role in the electroforming process, as they directly influence the quality and uniformity of the deposited metal. The key factors include:

Size: The dimensions of the electrode affect the current distribution and overall deposition rate, especially for nano and micro-scale applications, as *Chen et al*⁸⁵ mentioned in their study.

Shape and Geometry: Geometries can result in non-uniform deposition, necessitating precise control to achieve consistent coatings. Research demonstrates that the corners and edges of electrodes, for example, tend to exhibit higher deposition rates compared to the center regions due to increased current densities in these areas⁸⁶.

Electrode Material: The material composition of the electrode affects its conductivity and interaction with the electrolyte. When depositing materials with lower electrical conductivity than the main electrode substrate, there is a reduction in current density that correlates directly with the deposited layer thickness⁸⁷.

Surface Finish: Roughness of the electroformed layer increases as the substrate's surface roughness rises. On the other hand, electroforming thickness, quality, and hardness tend to increase as the electrode's surface roughness decreases⁸⁸.

Surface Cleanliness: A clean surface is crucial for ensuring proper adhesion and uniform deposition, as contamination can disrupt the electroforming process. Different impurities and cleaning conditions impact electrochemical reactions, ultimately affecting the quality of the deposited layer⁸⁹.

Positioning and Movement: The positioning and movement of the electrodes in the electrochemical cell impact the electric field, current distribution, mass transport, and deposition uniformity^{90,91}.

Current-potential characteristics are crucial in controlling the electroforming process and can be managed using different techniques, each offering specific advantages based on the desired deposition outcomes. These techniques include:

Galvanostatic: In this method, the current is kept constant while the potential varies. It allows for a controlled and consistent deposition rate, but the changing potential can affect the deposit quality, especially over long durations⁹².

Potentiostatic: The potential is held constant while the current adjusts to maintain this condition. This approach provides precise control over the deposition potential, which can be critical for achieving desired surface properties. However, it may lead to varying deposition rates^{93,94}.

Direct Current (DC): A constant flow of current is applied throughout this process. Continuous application of direct current results in a steady deposition. While simple to

implement, it can cause issues like dendritic growth, uneven deposition, and lower hardness, particularly in thicker electroforms ⁹⁵.

Pulsed Current: Pulsing the current on and off at regular intervals helps improve mass transport and often reduces internal stress in the deposit. This method can enhance deposit quality and uniformity, making it ideal for thick layers. Higher current densities can also be achieved compared to the DC method ⁹⁵.

Pulse-Reverse Current: Pulse-reverse current (PRC) enhances the quality of metallic coatings by alternating between cathodic and anodic phases, leading to improved deposit morphology and reducing defects like voids and rough grains. This process refines the diffusion layer, lowers internal stress, and increases current density, making coatings more durable. PRC also boosts hardness and decreases porosity, making it adaptable for various materials and alloy systems ⁹⁵.

Electrolyte characteristics are critical in determining the overall efficiency and quality of the electroforming process. These parameters must be carefully controlled to achieve optimal deposition and surface properties:

pH: According to a study done by *Boubatra et al.*⁹⁶, changing the pH can affect the electrochemical behavior of the deposition process and change the way metal ions nucleate and grow onto the cathode surface. Lower pH shifts the reduction onset toward more cathodic potentials. This can boost deposition rate but often leads to rough, granular films. However, higher pH produces more compact and uniform deposited layers, reflecting improved surface morphology.

Viscosity: Increased viscosity can slow down the movement of metal ions to the cathode, resulting in slower deposition rates and potentially leading to uneven surface morphology. However, lower-viscosity electrolytes enhance ion mobility and increase the deposition rate by thinning the diffusion layer. This can improve the conductivity and deposition quality ⁹⁷.

Conductivity: As *Kang et al.* ⁹⁸ demonstrated in their study, increased electrolyte conductivity improved the movement of metal ions toward the cathode. This resulted in faster deposition of the metal layer, improved surface morphology, and reduced defects in the deposited layers.

Concentration: The composition of the electrolyte can influence the deposition behavior drastically. For instance, increasing sulfuric acid concentration enhances hydrogen evolution at the cathode. This is often linked to higher conductivity, and it can slow deposition rates and worsen surface morphology ⁹⁹. As a result, the deposition roughness is highly sensitive to the acid concentration ⁸⁰. Similarly, metal ion concentration must be carefully optimized. Excessively low metal ion concentration decreases the conductivity and deposition rate. This inefficient deposition process increases the production time and cost. On the other hand, extremely high concentration results in poor deposition quality (rough and dendritic deposits), even if deposition accelerates ¹⁰⁰.

Additives and Impurities: Additives can be categorized into three main types: accelerators, levelers, and suppressors, each serving distinct functions that enhance the deposition process. An electrolyte formulated with the right amount of additives can promote uniform and defect-free deposition. It also ensures the desired surface texture and finish, enhancing the overall reliability of the electrodeposition process ¹⁰⁰. *Cho et al.* ¹⁰¹ reported that the combination of polyethylene glycol (PEG) as a suppressor and sulfopropylsulfonate (SPS) as an accelerator in copper electrodeposition led to improved filling of microstructures, resulting in smoother surfaces and reduced defects. However, it is essential to balance the concentrations of these additives, as excessive amounts can lead to adverse effects, such as increased internal stress ¹⁰². Impurities, particulates, and unwanted by-products from the electrolyte can also affect the physical properties, surface roughness, and thickness of deposited metal ⁸⁰. Continuous filtration and purification of the electrolyte is required to ensure uninterrupted plating operations while efficiently removing impurities.

Temperature: As the temperature increases, the kinetic energy of the ions in the electrolyte rises, leading to enhanced diffusion rates and increased nucleation rates, which can result in larger grain sizes and altered surface morphologies ¹⁰³. For instance, *Gunpum et al.* ¹⁰³ observed that higher solution temperatures led to the formation of larger pyramidal-like structures in the deposited metal, indicating a correlation between temperature and the agglomeration of metal particles. Furthermore, *Popczyk and Łosiewicz* ¹⁰⁴ reported that decreased temperatures could promote the formation of porous nickel coatings, which are characterized by their unique electrochemical behavior during hydrogen evolution reactions. However, excessively high temperatures may also lead to undesirable effects, such as increased internal stresses and reduced deposit quality ¹⁰⁵.

Electrolyte flow: In static electrolytes, the limited movement of ions can lead to concentration gradients near the electrode surface, which may result in uneven deposition and increased surface roughness due to localized depletion of metal ions. Contrarily, flowing electrolytes enhance mass transport, promoting a more uniform distribution of metal ions and reducing the likelihood of defects such as voids and dendrites in the deposited layers ¹⁰⁶. For example, *Zeng et al.* ¹⁰⁷ noted that sufficient agitation of the electrolyte can help maintain a consistent temperature and concentration, further optimizing the electrodeposition process and enhancing the mechanical properties of the coatings.

Process parameters listed below are critical in controlling the electroforming process. These parameters are interconnected and must be carefully adjusted to achieve high-quality, defect-free electroforms.

- **Current Density:** Variations in current density influence electrodeposited morphology and mechanical properties. At higher deposition current densities, the grain size of the coating tends to increase. This occurs because, at lower current densities, deposition proceeds more slowly, allowing sufficient time for particles to reach and distribute uniformly at the cathode. Such uniform incorporation promotes dispersion hardening and the formation of harder surfaces. In contrast, elevated current densities accelerate the deposition rate but reduce process control, often leading to rapid crystal growth and non-uniform particle distribution within the matrix ¹⁰⁸.

- **Electrode Potential:** Increasing the electrode potential typically accelerates the electrochemical reactions, leading to increased deposition rates; however, it can also result in undesirable effects such as increased roughness and the formation of defects like cracks or voids in the deposited layer. On the other hand, lower potentials may promote smoother deposits but can slow down the deposition rate, potentially leading to poor adhesion and inadequate coverage of the substrate; this nuance is important for optimizing the process. As *Wang et al.*¹⁰⁹ mentioned in their study, higher potentials (> 2.8 V) can cause dendritic and void-ridden deposits even though the deposition is faster. In contrast, lower potentials (< 2.8 V) can produce uniform, void-free structures. This confirms that a trade-off between deposition rate and surface integrity is necessary when selecting the appropriate potential.
- **Current (Faraday) Efficiency:** The current efficiency is the ratio of the actual amount of deposited material to the theoretical amount based on the applied charge. It measures how efficiently the applied current is being used for metal deposition. High current efficiency indicates that a greater proportion of the supplied current contributes to the deposition process rather than being wasted on side reactions, which can lead to defects and poor surface quality¹¹⁰. For example, *Rigsby et al.*¹¹¹ demonstrated that the current efficiency for cobalt deposition is significantly influenced by factors such as pH and mass transport, highlighting the importance of optimizing these parameters to enhance deposition efficiency and surface characteristics. In contrast, low current efficiency can result in increased roughness and porosity of the deposited layers. The excess current may promote undesirable reactions, such as hydrogen evolution, which competes with metal deposition¹¹².
- **Duty Cycle:** It refers to the proportion of the pulse-on time relative to the total cycle duration. A higher duty cycle typically increases ion migration and boosts nucleation rates, leading to more uniform and denser deposits¹¹³. *Saravanan and Mohan*¹¹³, for example, observed that at a 40% duty cycle, enhanced ion migration improved nucleation rates, resulting in a more consistent microstructure in chromium deposits. Lower duty cycles, in contrast, can hinder adequate ion replenishment during deposition. This often produces rougher surfaces and inferior mechanical properties due to larger grain formation and increased porosity¹¹⁴. Duty cycles can affect internal stress within the deposited layers, with higher duty cycles generally reducing residual stress, thus enhancing coating adhesion and durability¹¹⁵.

The role of process parameters such as current density, duty cycle, and temperature has been extensively analyzed. *Ding et al.*¹¹⁶ emphasized the challenges associated with void formation in high-aspect-ratio microstructures during nickel electrodeposition, advocating for optimized plating conditions to enhance the quality of electroformed structures. Additionally, *Han et al.*¹¹⁷ proposed the use of pulse-reverse current techniques to eliminate nanovoids in metallic nanostamps, demonstrating that optimizing current profiles can lead to significant improvements in the quality of electroformed components.

Overall, the collective insights from these studies underscore the importance of optimizing various process parameters and employing innovative techniques to enhance the electroforming process.

2.3.4 Potentials and challenges of electroforming

Electroforming presents significant advantages in advanced manufacturing, yet it also involves challenges that must be managed to ensure optimal performance and product quality. To highlight some of the potential applications of the electroforming process, the following examples can be identified:

- High-precision manufacturing: Electroforming enables the production of highly precise components with exceptional dimensional accuracy. This is achieved through an atom-by-atom deposition process that enables precise control of material morphology and structure^{23, 22}. This precision is crucial in applications requiring tight tolerances, such as micro-electromechanical systems (MEMS) and advanced electronic devices. For instance, *Tsao* highlighted the application of photo-assisted electroforming in producing MEMS via high-precision micro-scale patterning¹¹⁸.
- Complex design manufacturing: Another key benefit of electroforming is the fabrication of intricate geometries and thin-walled structures without the need for joints or seams, which can be limiting in traditional manufacturing methods. This characteristic is particularly advantageous for producing items like stents and other biomedical devices, where structural integrity and biocompatibility are essential¹¹⁹.
- Mass production of parts with desired properties: Electroforming has the potential to produce parts in parallel, which can be widely used to mass-produce high-precision metal micro-parts. It also allows for the manufacturing of parts with tailored mechanical properties. Manufacturers can influence the microstructure of deposited materials by adjusting electroforming parameters such as current density and electrolyte composition, leading to improved mechanical performance, including enhanced strength and ductility²¹. This tunability is particularly advantageous for applications requiring specific material characteristics, such as wear resistance or fatigue strength.
- Electrolyte as a tool: In electroforming, the electrolyte itself functions as a universal tool (the shape of the part is defined solely by the mandrel immersed in the bath). To change part geometries, one simply swaps the mandrel, without needing new tooling. For example, *Hamed et al.*²³ demonstrate this flexibility via additively manufactured molds to fabricate multi-level, free-form metal parts in a single bath.

Despite the aforementioned advantages of electroforming, certain disadvantages remain as well. As an example, electroforming baths age from extensive exploitation in the mass-production processes. The "aging" term refers to the inconsistency of the electrolyte composition and the gradual changes in its key properties, such as metal (or acid) concentration, which may result in inconsistent electroforming quality⁷⁸. The issue of electrolyte degradation becomes even more pronounced in mass personalization, where multiple parts with varying conditions are electroformed simultaneously. While it is possible in mass production to monitor the key bath properties and conduct electrolyte filtering/changing when necessary, in a mass personalization paradigm where various parts are produced all the time, the same strategy reaches its limits due to increased costs of bath replenishment and disposal.

Frequent adjustments to the bath can also lead to instability in electroforming parameters, which ultimately reduces the quality and consistency of the final products. This issue underscores the necessity of precise control and continuous monitoring of the bath composition and electroforming process to ensure reproducible results. Such control is particularly critical when fabricating personalized parts with defined material properties or surface finishes. In the following section, a thorough analysis of this topic will be presented, covering the impact of electrolyte aging and how it affects the overall surface quality and production costs of the components.

Nevertheless, in the context of mass personalization, automation and quality control of the electroforming process face several challenges, including:

- Artisanal nature of electroforming and optimization challenges: The phrase "artisanal" describes a hands-on process that depends on the skill and expertise of the person doing it. This often leads to one-of-a-kind, handmade items. In industrial settings, it opposes automated or standardized approaches, highlighting craftsmanship and precise adjustments instead of mass production. Mass personalization necessitates process flexibility. While digital processes such as AM or CNC machining can adapt through software adjustments, electrochemical processes remain predominantly artisanal, especially in controlling the process parameters and dealing with the changes in electrolyte composition ²⁴. The heavy dependence on operators' expertise presents a major obstacle to automating electroforming operations and transitioning to mass production and personalized manufacturing strategies. To achieve mass personalization, processes must be optimized for higher efficiency, faster production speeds, and reduced reliance on manual labor. Streamlining electroforming processes calls for adopting rapid and cost-effective parameter optimization methods. Additionally, ensuring product uniformity in mass personalization requires reproducibility and consistent quality, which can be achieved through standardized workflows and automated quality control systems.
- Process unpredictability: One of the main difficulties is the inherent unpredictability associated with electroforming, which can result in inconsistent outcomes. This unpredictability is often worsened by variations in the electrochemical environment, making it challenging to achieve consistent results. For example, *Park et al.* ²⁵ found that, without corrective measures, electroforming often results in thicker deposits at the edges compared to the center, due to uneven current distribution and ion transport conditions.
- Quantity of influencing process parameters and their interactions: The parameters set during the electroforming process, such as current density, voltage, and electrolyte composition, heavily influence the control over the process. The type and concentration of the electrolyte used, for example, can significantly affect the electroforming process. Variations in these parameters can result in changes in the deposition rate and the morphology of the electroformed materials, ultimately impacting the performance of the final product ²⁶. Lack of precise control (operators may employ varying techniques and have distinct preferences in how they approach the process optimization) may lead to

undesired defects, which can be detrimental to the functionality of electronic components²⁷.

- Complications from external factors: The electroforming process is often influenced by external factors like temperature, adding complexity to the process. For instance, temperature fluctuations can alter the electrolyte's viscosity, impacting ion transport and deposition rates²⁸. Moreover, impurities or variations in the electrolyte composition can lead to undesired side reactions, undermining the integrity of the electroformed structures²⁹. Measuring and understanding these interactions can be challenging, especially when they depend on the subjective assessments of the operator.
- Modeling and data gaps in capturing electrochemical dynamics: The collection of data and mechanisms for feedback in electroforming are frequently restricted or completely nonexistent. It is challenging to make well-informed decisions for process optimization without accurate and thorough datasets on key process parameters and the consequent surface traits. The absence of data hinders the capability to adjust electroforming operations, ultimately impacting the uniformity and quality of the end products. Additionally, despite the progress in modeling and simulation methods, the application of these models in real-world electroforming processes is still constrained. Several current models fail to consider the intricate dynamics of the electrochemical setting, resulting in disparities between expected and observed outcomes^{120, 121}.

As mentioned earlier, studies have focused on optimizing electroforming parameters to achieve specific mechanical properties and surface finishes in electroformed parts. These studies have primarily looked at the initial process parameters to improve the deposition process. However, the dynamic state of the plating bath, which can significantly impact the consistency and quality of the electroformed layer⁷⁸, has been largely overlooked. Changes in bath composition over time, such as variations in metal ion concentration or the presence of impurities, are often not considered when optimizing electroforming conditions during the process.

2.4 Electrolyte Ageing

2.4.1 What does electrolyte aging mean?

The term "electrolyte aging" in electroforming and electroplating processes signifies the progressive deterioration of the electrolyte solution over time due to chemical reactions and environmental exposure⁴⁰. This aging process can lead to the:

- a) Accumulation of impurities: The bath becomes contaminated by foreign particles, dissolved gases, or ions resulting from equipment corrosion or a dirty environment.
- b) A buildup of by-products: Occurs because of the reduction or oxidation of impurities or metal ions in the bath.
- c) Decrease of active species concentration: As the electrochemical reactions continue, the metal and additive ions concentration in the bath decreases, causing a reduction in deposition efficiency and potential changes in the system's electrochemical behavior^{41, 42}.

2.4.2 How does electrolyte aging affect electroformed parts?

The surface quality and mechanical properties of electroformed parts can be significantly impacted by the aged electrolyte. Electrolyte degradation over time can lead to several adverse effects due to the amount of impurities, byproducts, and active species concentration. These effects can be:

- Surface roughness: Accumulation of impurities and by-products in the electrolyte can lead to uneven deposition. Therefore, surface roughness or defects on the electroformed parts increase due to the introduction of more impurities to the bath¹²².
- Decreased deposition efficiency: As metal ions are consumed, the concentration of active ions in the electrolyte decreases due to aging. As a result, the rate of deposition slows down as the concentration of active ions in the electrolyte decreases¹²³.
- Altered grain structure and mechanical properties: Over time, when the electrolyte is used without filtering and refreshing, impurities and foreign particles become part of the metal matrix. This can affect the microstructure of the deposited metal and result in decreased ductility, heightened brittleness, or reduced tensile strength. As a result, the part becomes more susceptible to failure under stress. For instance, *Kao et al.*¹²⁴ demonstrated that impurities in electroplated copper foils can lead to variations in grain size, which directly influence the mechanical strength of the material. Therefore, electrolyte aging can alter the mechanical properties of the electroformed material.

- Corrosion resistance: Impurities and changes in electrolyte composition due to aging lead to poor corrosion resistance in the electroformed part. This is especially critical in applications where the parts are exposed to corrosive environments. For instance, *Sobolev et al.*¹²⁵ noted that coatings formed in molten salt, which typically have fewer impurities compared to aqueous electrolytes, exhibit higher polarization resistance, indicating better corrosion resistance due to the lack of detrimental impurities.
- Increased defect density: Contaminants and inconsistencies in the electrolyte can result in an increased number of defects, like pits, voids, or cracks in the electroformed parts. Such defects can potentially affect the overall reliability and performance of the final product. As an example, *Harry et al.*¹²⁶ highlighted that the presence of faceted impurity particles can lead to increased defect density in electrodeposited materials and consequently compromise the structural integrity of the final product. *Frenck et al.*¹²⁷ also observed that impurities can nucleate defects during lithium electrodeposition. This indicates that similar mechanisms could apply to other metals where impurities disrupt the deposition layer.
- Color or appearance changes: The alteration in concentration and accumulation of impurities due to electrolyte aging can cause discoloration and other aesthetic flaws in electroformed parts, significantly affecting their visual appeal. For instance, *Anwar et al.*¹²⁸ noted that variations in the electrolyte composition can lead to changes in the surface morphology of zinc-nickel films, which can manifest as discoloration or uneven finishes.
- Poor adhesion or delamination: The presence of impurities and contaminants in the aged electrolyte during electrodeposition can significantly undermine the adhesion of the deposited layer to the substrate, increasing the risk of delamination or flaking. Impurities can disrupt the electrochemical processes that are crucial for forming a strong bond between the coating and the substrate. For example, *Zhang et al.*¹²⁹ reported that impurities in the electrolyte directly affect the grain structure of electroplated copper, leading to weak interfaces that are more prone to delamination. These impurities can move to grain boundaries, disrupt the uniform growth of the deposited layer, and weaken adhesion. Additionally, *Kao et al.*¹²⁴ highlighted that impurities co-deposited with the metal can reside at crystalline boundaries, further compromising adhesion and increasing the likelihood of defects such as voids and cracks in the electroformed parts. Similarly, *Chen et al.*¹³⁰ noted that the introduction of nonmetallic impurities, such as carbon and sulfur, during the electrodeposition process can decrease the etching resistance of the deposited layers, leading to poor adhesion and heightened vulnerability to delamination.

As a result, regular monitoring and maintenance of the electrolyte are crucial for maintaining high-quality electroformed parts, as they lead to minimizing impurities and ensuring that the chemical composition of the electrolyte remains within optimal ranges.

2.4.3 What are the impacts of electrolyte aging on the electroforming costs?

The electrolyte's aging in electroforming procedures can significantly affect costs, both directly and indirectly. These impacts encompass:

- Escalated maintenance costs: In mass personalization, the electrolyte deteriorates more frequently due to the introduction of various components with differing conditions into the bath. Consequently, to preserve the electrolyte's effectiveness and plating quality, increased frequency of monitoring, filtering, and purifying operations is necessary. For instance, the breakdown of additives results in defects like voids and rough surfaces in the deposited layers, necessitating regular assessments and adjustments to the electrolyte composition^{131,132}. Regular monitoring allows for timely interventions that can mitigate the adverse effects of aging, ensuring consistent outcomes⁴¹. However, this leads to increased operational costs due to the need for specialized equipment, chemicals, and labor.
- Elevated fresh bath/material replacement costs: The deterioration of plating baths markedly elevates the consumption of raw materials owing to the degradation of electrolyte constituents and the formation of byproducts. As an example, when the bath ages, additives break down, leading to a decline in electrodeposition quality and necessitating the addition of fresh materials to compensate for the loss of effectiveness^{131,132}. Moreover, the presence of impurities and byproducts in aged baths can alter the electrochemical properties, prompting the need for more frequent replenishment of the electrolyte to maintain performance standards¹³². Consequently, the overall demand for raw materials escalates as manufacturers strive to mitigate the adverse effects of aging in electrodeposition processes¹³². The incorporation of new materials or the complete replacement of an electrolyte bath can be expensive, particularly in extensive operations, owing to the significant expense of the electrolyte solution and the necessary downtime for bath or fresh metal substitutions.
- Energy inefficiency: Aging electrolytes in electrodeposition processes can significantly reduce deposition efficiency, necessitating longer electrodeposition times or higher currents to achieve the same results. As the electrolyte ages, its composition and effectiveness deteriorate, leading to a decrease in the rate of metal deposition¹³³. This inefficiency often compels operators to increase the electrodeposition current to maintain desired deposition rates, which can further exacerbate energy consumption and operational costs¹³⁴. For instance, studies have shown that the quantity of metal deposited is directly related to the duration of electrodeposition, and as the efficiency declines, longer durations are required to achieve the same thickness of deposition¹³⁵. This increases energy consumption, raising operational costs related to electricity usage.
- Lower production efficiency with reduced yield and rework cost: As previously stated, electroforming bath aging (due to changes in concentration and impurity buildup) can result in low-quality deposits, such as increased surface roughness¹³⁶, emergence of

different surface defects ¹²⁶, or uneven thickness ¹²². The low quality of the deposits calls for later modifications or rejection of the electroformed components, which would, in turn, increase the disposal costs. Furthermore, it results in prolonged component production cycle durations. Deliveries will be postponed due to the reduced overall throughput of the electroforming operation. Costs ultimately increase as additional labor is needed to achieve production objectives.

Overall, the challenges associated with controlling and mitigating electrolyte aging often add up over time. Therefore, it is critical to monitor electrolyte conditions and develop measures to extend bath life, estimate bath remaining life, decrease waste, and improve the electroforming process.

2.4.4 How can electrolyte aging impact the environment?

Electrolyte aging in electroforming operations can have a variety of environmental consequences, mostly owing to the accumulation of contaminants and by-products in the electrolyte, which need careful treatment and disposal. The main environmental consequences can be mentioned as:

- Increased waste and environmental effects: As the electroforming bath ages, a buildup of impurities such as metal hydroxides, byproducts from electrochemical processes, and foreign particles is formed. These compounds can make the electrolyte hazardous, resulting in a higher amount of polluted waste. Improper disposal of this material can be destructive to the environment, particularly if it includes dangerous metals or compounds ^{137, 138}. In Addition, aged bath solutions ought to be treated before disposal according to environmental regulations. Treating and neutralizing this waste from an aged electrolyte requires the use of extra chemicals and energy. These waste treatment technologies frequently generate secondary byproducts that must also be managed. Depending on the pollutants in the electrolyte, more extensive treatment processes may be required, which increases the environmental effect ^{137, 138}.
- Chemical leakage and hazardous spills: The accumulation of certain chemicals or degradation products in the electrolyte also increases the risk of leakage if waste is not appropriately controlled. When waste from aging electrolytes enters natural water systems or soil, it has the potential to negatively impact local ecosystems. Toxic metals and chemicals can endanger aquatic life, diminish biodiversity, and impact plants and animals in contaminated places. Long-term exposure to these pollutants can cause ecosystem imbalances ¹³⁷. Carbon, oxygen, and sulfur, which are commonly found in organic additives and substrate materials, can co-deposit with the metal, adding to the electrodeposition solution's instability ¹³⁹. This raises the possibility of leaks, spills, or

unintentional discharges, which can pose major environmental dangers, especially in locations where the electrolyte contains heavy metals.

- Higher carbon footprint: As electrolyte aging reduces the efficiency of the electroforming process, larger energy inputs are frequently necessary to maintain acceptable output rates¹³⁴. This increased energy consumption can result in higher carbon emissions, particularly in countries where electricity is generated from nonrenewable sources such as coal or natural gas.
- Air pollution: During electroforming activities, aging electrolytes can emit volatile chemicals like carcinogens or hydrogen gas, particularly if the solution is not well maintained (the plating temperature, for example)¹⁴⁰. The emission of these volatile compounds into the environment can pollute the air and endanger workers and surrounding communities.

The environmental impacts of electrolyte aging in electroforming processes underscore the necessity for continuous monitoring systems to enhance waste management, treatment, and recycling plan forecasting. The electroforming sector may significantly mitigate its environmental impact by employing sustainable practices, including technologies to assess bath lifespan, adjusting active ion concentrations, and minimizing waste.

2.4.5 Review of current studies on electroforming in aged electrolyte

As mentioned earlier, numerous industrial studies have highlighted the importance of process monitoring in preserving the quality of the surface in electrodeposited products^{78, 41}. While these studies touch on key electrodeposition/electrolyte parameters that should be tracked, they tend to be concise and do not provide readers with a thorough understanding of the complexities involved. Over the years, companies specializing in plating have developed deep expertise and distinctive techniques, primarily due to the expertise of experienced operators with decades of practice. This specialized knowledge offers these companies a market advantage, which is why they treat it as a closely guarded trade secret, withholding such information to preserve their market position.

A review of academic publications in this field shows a notable lack of detailed research on electroforming in aging electrolytes and its influence on the surface condition of the finished products. The existing studies are limited in scope, both in terms of the process variables examined and the specific surface characteristics targeted. An examination will be conducted on several significant gaps and limitations in the existing literature.

Unclear explanation for what constitutes an aged electrolyte

It is often stated that the solution is considered aged when electrodeposition outcomes start to differ from the desired specifications. However, a comprehensive understanding of how the properties of the electroforming electrolyte evolve with extended use is lacking. The very limited research on electrolyte aging has primarily focused on the dissolved metal and additives concentration in the solution, with only a few studies exploring other bath characteristics like pH⁴³ over time. Without a thorough grasp of the electrolyte degradation process, it is challenging to adjust process parameters to preserve the quality of electroformed parts.

A need for advanced and effective real-time methods to analyze electrolytes efficiently

To date, only a restricted number of investigations concerning the in-line quality assessment of plating baths and the prediction of necessary maintenance intervals are available in the open literature^{24, 32, 33, 78, 91, 141–147}.

One of the challenges related to the composition changes in plating baths is accurately measuring the concentration of essential species¹⁴⁵. To address this, researchers have proposed various solutions, one of which involves using a model approach based on mass balance equations to predict the remaining useful life of electrolytes. However, determining the phenomenological coefficients necessary for these models proves challenging, as they vary over time due to changes in part geometries¹⁴⁷. Another study proposed combining fuzzy logic with Online Analytical Processing (OLAP) methods to design systems capable of monitoring features such as chemical concentration in electroplating while enhancing operational efficiency and decision-making. Yet, this approach still suffers from the complexity of model tuning and the need for precise parameter settings¹⁴².

Other approaches attempted to employ cyclic voltammetry (CV), a traditional method, to estimate the concentration of the electroplating bath. Still, it does not address the inherent limitation of requiring regular bath sampling and an offline approach³³.

Restricted scope of electrolyte conditions and corresponding electroformed outcomes

As previously mentioned, most studies have focused on examining the effects of process parameters such as current density, applied potential, and concentration of various additives on the final electroformed parts. However, there is no clear understanding of the relationship between electrolyte conditions, such as metal ion concentration, conductivity, and viscosity as it ages, and the quality of electroformed components. For instance, *Gabrielli et al.*⁴² conducted a comprehensive study on the aging of copper interconnect deposition baths, employing Electrochemical Impedance Spectroscopy (EIS) to analyze the impact of chlorides on the deposition mechanism. Their findings indicate that at elevated current densities, the concentration of CuCl becomes the predominant species on the electrode surface. This observation is crucial as it highlights the significance of the CuCl to Cu metal deposition reaction mechanism. The authors

utilized a kinetic model to elucidate the relationship between chloride ions and copper deposition, demonstrating that the presence of chlorides not only influences the deposition rate but also alters the morphology of the deposited copper. This study underscores the necessity of understanding the chemical dynamics within the deposition bath to optimize the electrodeposition process.

*Moffat et al.*⁴¹ explored the effects of accelerator aging during copper electrodeposition. Their research focuses on the role of thiourea as an accelerator in the copper plating process. The authors noted that oxidative thiol-disulfide reactions, which are well-documented in protein chemistry, also play a significant role in copper plating chemistry. By examining the significant impact of accelerators on the copper deposition reaction, the study employed a strategy akin to Cyclic Voltammetric Stripping (CVS) analysis to probe the underlying chemistry of the accelerators. Their findings suggest that the aging of the deposition bath can significantly alter the efficiency and quality of the copper deposits, emphasizing the need for careful monitoring and control of the deposition environment.

Moreover, *Hao et al.*⁴³ demonstrated that, despite extensive research on various formulations and methods for achieving consistent black nickel coatings, a considerable gap remains in understanding the influence of bath aging on the coating quality. They also emphasized that maintaining stable pH levels in the plating bath is a crucial aspect of the electroplating process. Traditionally, boric acid has been employed as a pH buffer to ensure its stability, which is vital for producing high-quality coatings. However, the disposal of boric acid-containing solutions presents significant environmental challenges, such as soil toxicity and elevated disposal costs. These issues raise concerns about the sustainability of current practices and underscore the need for alternative pH management methods that do not rely on boric acid.

These studies offer fundamental insight into how the solution (especially the additive concentration and pH) aging influences mass transport phenomena within the electrolyte, leading to a decline in the quality of the electroplated part. However, their primary focus is on identifying the ideal electrolyte formulation that extends its efficiency and minimizes deposition defects. No study has been conducted on other aspects of bath characteristics and their potential impact on varying electroplated quality outcomes in these papers. This is a critical area of study, particularly in the context of mass personalization, where the demand for parts with different desired mechanical and surface properties is increasing.

Unavailable database for electroforming in aged bath

A skilled operator might be able to gauge the suitable parameters for the electroforming process according to the state of the bath. However, this method tends to be subjective, time-intensive, and limited in terms of the range of parameters explored. The continual changes in the plating bath add further difficulty to this task. One of the goals of this research, as emphasized earlier, is to use data-driven techniques to automatically optimize the electroforming parameters while the electrolytes are aging. In this context, a comprehensive review of the research that has utilized models to forecast final surface properties or determine optimal electroforming parameters in aged baths is essential.

However, to the author's best knowledge, there is no attempt published to develop a tool able to analyze and optimize the electroforming parameters at the current state of electrolyte, as there is an ongoing process. Considering the current lack of information regarding the optimization of electroforming process parameters in aged solutions, and the critical role this information plays in ensuring consistent outcomes and minimizing the electroforming costs, a thorough investigation is necessary. This research should rely on an extensive database of experimental data, covering a broad spectrum of surface qualities and electrolyte conditions. The findings should be evaluated using various assessment techniques, and the potential for applying the insights to new operational scenarios must also be explored.

2.5 Quality control in electroforming

The consistency and quality of electroforming outcomes are directly influenced by how well the process is managed ¹⁴¹. Since electrolytes degrade significantly with repeated use in mass production, poor control over the process can lead to unpredictable and uneven results ³³. To preserve the efficiency of the plating electrolyte as it ages, several techniques are employed in electroforming operations. These typically include monitoring specific bath properties, removing accumulated sludge, and regularly decanting the electrolyte to maintain its effectiveness.

2.5.1 Removing electrolyte sludge

The management of electrolyte sludge in electrodeposition processes is a significant concern due to the environmental and operational challenges it poses. Electrolyte sludge, often a byproduct of metal finishing operations, contains a mixture of heavy metals, organic compounds, and other contaminants that can lead to environmental pollution if not properly treated. There are various methods for the removal and management of electrolyte sludge, supported by relevant literature.

One effective approach to managing electrolyte sludge is through the recovery of costly metals contained within it. For instance, studies have demonstrated that nickel can be recovered from electroplating sludge using acid leaching methods, which can subsequently be processed to form nickel-iron alloys ¹⁴⁸.

In addition to metal recovery, the treatment of electrolyte sludge can also involve the utilization of various chemical processes. For example, the application of alkaline treatments has been explored to stabilize and immobilize heavy metals within the sludge, thereby reducing their leachability and potential environmental impact ¹⁴⁹. This approach can transform hazardous sludge into a more stable form suitable for safe disposal or even for use in construction materials, as demonstrated in studies where electroplating sludge was incorporated into ceramic bricks ¹⁵⁰.

Another innovative method for managing electrolyte sludge is through the use of biological processes. Bioleaching, which employs microorganisms to extract metals from sludge, has gained attention as a sustainable alternative to traditional chemical methods ¹⁵¹.

Furthermore, advanced filtration techniques, such as nanofiltration and membrane processes, have been investigated for their effectiveness in treating electroplating wastewater and sludge. These methods can selectively remove heavy metals and other contaminants, thus improving the quality of the effluent and reducing the sludge volume generated during the treatment process ¹⁵².

Despite the advantages of these methods, they come with challenges, including high costs, equipment maintenance, scalability issues, slow processing times, and potential environmental risks. Each method has limitations in efficiency and sustainability, making it crucial to evaluate their suitability based on specific operational requirements.

As a result, the importance of continuous monitoring and optimization of the electrodeposition process cannot be overstated. By monitoring and adjusting electrolyte composition, the generation of sludge can be controlled, and the quality of the electroplated products can be enhanced.

2.5.2 Decanting the electrolyte

Replacing aged electroforming baths with a new electrolyte or adding/removing particular components to reestablish the correct composition is the most straightforward and widely used technique for rejuvenating aged electrolytes. In practice, the bath is typically used until the deposition quality starts to decline. At that stage, about 20% (or less) is extracted and substituted from the solution with a new electrolyte to preserve the ideal balance of chemical substances, a process known as decanting.

In the realm of mass personalization, the objective is to minimize additional interventions in the aging bath while ensuring that it maintains the required properties for producing personalized parts, based on its current condition. It is more cost-effective and time-efficient to avoid fully or partially replacing the bath with a fresh solution. Therefore, there is a need to find an approach to reduce operational disruptions and optimize resource utilization as the electrolyte ages.

2.5.3 Monitoring bath properties

Monitoring the physical and chemical properties of the electroplating bath is essential for ensuring the quality and performance of the electroplated coatings^{145, 153}. Each property, as listed below, can significantly influence the electroplating process and the characteristics of the deposited layer.

pH

The pH level of the electroplating bath plays a vital role in determining the deposition rate, coating quality, and stability of the electrolyte¹⁵⁴. Maintaining the correct pH range is crucial for ensuring metal ions remain soluble and that complexing agents work effectively¹⁵⁵. In nickel electroplating, for instance, a pH that is too low can lead to increased hydrogen evolution, which negatively impacts the deposit quality¹⁵⁶. Therefore, consistent monitoring of pH is necessary to maintain optimal deposition conditions and avoid coating defects, especially in aging electrolytes.

Electrical conductivity

The electrical conductivity of the electroplating bath is another crucial parameter that influences the current distribution and the overall efficiency of the electroplating process¹⁵⁴. Variations in conductivity can signal changes in ion concentration or the absence/presence of inorganic impurities within the bath^{154 157}. Consistent monitoring of conductivity is essential for evaluating the electrolyte's condition and maintaining optimal plating performance.

Temperature

The temperature of the electroplating bath significantly affects the electrochemical reaction kinetics and the solubility of metal salts. Elevated temperatures typically enhance the deposition rate but may also introduce greater internal stresses in the deposited layer¹⁵⁸. In nickel plating, for example, a temperature around 50°C is commonly advised to strike a balance between deposition speed and coating quality¹⁵⁶. Regular temperature monitoring is essential to ensure stable and consistent plating conditions¹⁵⁴.

Viscosity

The changes in temperature, concentration of ions, and level of impurities in the electroplating bath can change the viscosity and adversely affect the quality of the deposited layer. Impurities can lead to defects such as pitting, roughness, and poor adhesion^{122,136}. Regular testing for viscosity and concentration of impurities is necessary to maintain the integrity of the electroplating process and to ensure high-quality coatings.

Specific gravity

Specific gravity is an indicator of the overall composition of the bath, including the concentration of dissolved solids such as metal salts, additives, and impurities¹⁵⁴. Monitoring specific gravity helps to assess bath aging and maintain the desired chemical balance for consistent electrodeposition¹⁴⁷.

Concentration of metal ions and additives

The concentration of metal ions in the bath is a critical factor that impacts both the deposition rate and the quality of the electroplated layer. Low metal ion levels can result in slow deposition and uneven coatings, while excessively high concentrations can lead to a faster deposition rate and rougher surfaces^{159,134}. Regular monitoring of metal ion

concentration is essential to maintain the electrolyte's optimal composition and ensure high-quality deposits. Additives also play a key role in modifying the bath's characteristics and the properties of the electroplated layer. They can improve the brightness, smoothness, and hardness of the coating, as well as control grain size and reduce internal stresses^{101,160,161}. Maintaining an appropriate balance in the concentrations of additives is vital, as excessive or insufficient levels can lead to detrimental effects, such as internal stress and a reduction in the mechanical properties of the deposits¹²⁹.

Agitation rate

The agitation of the electroplating bath affects the mass transport of metal ions to the cathode surface, influencing the formation rate, uniformity, and quality of the deposit¹⁵⁴. Insufficient agitation can lead to localized depletion of metal ions, resulting in uneven coatings. Monitoring the agitation rate helps to maintain a consistent supply of metal ions and to enhance the overall efficiency of the electroplating process^{162,163}.

Bath level

Controlling the liquid level or volume in plating tanks is crucial for several reasons. A sufficient volume of solution is needed to operate pumps, submerge components or sensors, and carry out the plating process effectively. Keeping the solution level stable also helps maintain consistent concentrations of the solution's components¹⁵⁴.

In conclusion, monitoring the characteristics of the electrodeposition bath is critical for producing high-quality electroplated coatings. Manufacturers can improve the performance of deposited layers by systematically examining the aforementioned characteristics.

2.5.4 Quality control challenges

The main challenge in controlling bath quality for electroforming is the need to measure and adjust key parameters like metal ion concentration, electrical conductivity, and temperature. These factors are essential for ensuring the quality of deposited components, especially when mass-producing personalized parts. In summary, these challenges can be listed as follows:

On-site monitoring of bath properties

Electroforming requires precise control of bath parameters such as metal ion concentration, additives, temperature, pH, and electrical conductivity. Variations in any of these can result in flaws such as uneven deposition, rough surfaces, or poor mechanical characteristics in the produced component ¹⁵⁴. Monitoring these parameters on-site can be difficult, necessitating precise instrumentation and frequent modifications to keep the bath within ideal limits ¹⁴⁴.

Operator and skill dependency

Controlling the electroforming bath generally relies on the operator's expertise and judgment. Operators must often check the status of the bath and alter settings based on visual cues or empirical knowledge, a process known as the "art of electroforming". However, this might bring discrepancies, especially if several operators are controlling the same bath, because each individual may have a somewhat different method or perception of the bath state. This diversity might result in uncertain product quality ¹⁵⁴.

Maintaining consistency in an aging bath

As the electrolyte ages, its composition gradually changes owing to metal depletion, additive breakdown, and the buildup of byproducts ^{41,42}. These shifts make it difficult to maintain a constant bath condition, resulting in differences in the electroforming process and the quality of the finished pieces. Determining the optimal time to refresh the bath or modify the electrolyte composition is critical, but it may be challenging without exact, real-time data.

Cost of bath replenishment

Electroforming baths are expensive to maintain because of the high amounts of electrolytes required, particularly in industrial settings ¹³⁸. Regular replacement of metal ions and additives is required to keep the bath functioning properly; however, refilling huge volumes of electrolytes is expensive. Frequent bath replacement in mass personalization is impractical and expensive, both in terms of materials and time wasted during the replenishment process.

Uncertainty in electrolyte quality

As the electroforming process continues, the electrolyte's quality deteriorates, affecting the stability and uniformity of the manufactured components ⁴⁰. Insufficient monitoring or variability in operator expertise can introduce uncertainty in determining bath conditions, complicating efforts to achieve consistent and reliable outcomes. This challenge is further amplified in mass personalization, where different parts with distinct quality requirements must be electroformed in a shared bath.

Long-term environmental impact

As mentioned above, electroforming generates waste products that are difficult to neutralize and dispose of due to the presence of heavy metals and other toxic substances. The environmental effect of electroforming processes is gaining attention, since the disposal of sludge, rinse water, and wasted electrolytes creates both hazardous substances and regulatory issues ^{137, 138}. Strict environmental standards must be followed, and failure to properly handle these wastes can have serious legal and financial consequences for facilities.

Maintaining electrolyte quality in a cost-effective and ecologically friendly manner is critical for creating high-quality electroformed products while guaranteeing long-term process viability in mass personalization.

2.5.5 Quality assurance employing data-driven methods

Maintaining consistent surface quality in electroforming becomes increasingly challenging as the electrolyte evolves during repeated use. In the mass-personalization context, where hundreds of unique components may be manufactured under varying conditions, small deviations in bath chemistry, current efficiency, or electrode condition can lead to substantial differences in roughness, gloss, and mechanical performance. Conventional process control depends heavily on operator experience and trial-and-error adjustments, making it difficult to reproduce a prior surface finish once the bath has aged or to establish suitable parameters for a geometry or finish that has never been manufactured before. As product variety expands and production speeds increase, this reliance on tacit knowledge becomes unsustainable. A transition toward data-driven decision-making is therefore essential for ensuring quality, reducing scrap, and enabling personalization without prohibitive cost escalation.

The volume, diversity, and temporal evolution of electroforming data far exceed what human operators can reliably interpret. Digital technologies (continuous monitoring, logging, and

analytics) offer the ability to structure and learn from this information, transforming domain experience into quantitative insight. Machine learning (ML) in particular can extract complex, nonlinear relationships between bath state, process parameters, and finished surface properties that are not evident through empirical reasoning. By leveraging historical data and predictive models, ML can reduce the need for repeated chemical replenishment experiments, minimize trial-based parameter exploration, and support autonomous quality assurance as the bath ages, capabilities that become indispensable in mass personalization, where each part may demand unique surface attributes.

Existing data-driven modeling efforts in electroplating

A review of the literature shows a growing, yet still fragmented, effort to apply ML and optimization techniques across different electrochemical systems.

- Bath-chemistry optimization and additive analysis

*Katirci and Tekin*³⁰ applied NSGA-II to zinc electroplating baths, demonstrating that evolutionary multi-objective optimization can balance coating quality and operating efficiency with relatively low computational complexity. *Katirci and Danaci*³¹ extended data-driven optimization to nickel electroplating, using artificial neural networks to model the influence of organic additives and classify coating quality from visual measurements.

ML has also been used to monitor and quantify additive concentrations in complex electrolytes. *Ellis et al.*³² developed a neural-network-based method for determining accelerator, suppressor, and leveler concentrations in copper plating, overcoming the limitations of conventional CVS analysis and achieving sub-1% accuracy. Their work demonstrates that ML can resolve multiple overlapping electrochemical signatures from voltammetric data, an early example of non-destructive, data-driven bath characterization.

Similarly, *Yoon and Kim*³³ used neural networks to identify halide ions and multiple PEG species directly from a single voltammogram. Their study illustrates that ML can distinguish subtle variations in additive chemistry even in highly complex multi-component $\text{CuSO}_4/\text{H}_2\text{SO}_4$ electrolytes, which traditional analytical techniques struggle to achieve.

- Improving electroforming process outcomes

ML has also been explored for enhancing electroforming processes themselves. *Hsu*³⁴ introduced an integrated optimization framework combining BP neural networks, desirability functions, and tabu search to improve stamper quality in optical media manufacturing. By optimizing multiple electroforming responses simultaneously, the procedure reduced defect

rates from >10% to ~4.76% and demonstrated the benefits of data-driven parameter exploration in complex multi-output systems.

Reinforcement-learning approaches have also begun to emerge. *Goulart and Pereira*³⁵ developed an autonomous pH control system for electroplating wastewater using RL, demonstrating that data-driven control can outperform PID regulation in nonlinear and time-varying chemical systems. Although their work targets wastewater treatment rather than plating quality, it illustrates the viability of autonomous learning-based controllers in electrochemical environments.

- Efforts focused on surface-property prediction

More recent ML applications aim at predicting surface quality directly. *Sun et al.*³⁶ modeled defect occurrence, colour classification, and coating mass in boric-acid-free black nickel plating. Despite promising results from Adaboost, Random Forest, and ML models, the authors report significant limitations: narrow interpolation validity, poor extrapolation as bath conditions shift, and a requirement for large standardized datasets. *Katirci and Danaci*³¹ proposed a three-stage pipeline, Mask R-CNN segmentation, ML prediction, and NSGA-II optimization, to tune organic additives in nickel plating using Hull-cell imagery. While powerful for decorative systems where visual cues dominate, the method is not transferable to additive-free or pulse-controlled deposition systems. *Orabi et al.*³⁷ used process-mining and XGBoost to detect anomalous executions across large industrial zinc-plating lines. Their approach excels at early deviation detection but does not link anomalies to surface outcomes or infer underlying bath state. *Ren et al.*³⁸ integrated MLP predictors with a deep deterministic policy gradient (DDPG) controller for black-nickel plating optimization. Although showing promising virtual-to-real transfer, the method remains constrained to a narrow set of bath features and optimizes only current density, without modeling multi-output surface properties.

- Traditional statistical modeling in electroforming

Classical modeling efforts are scarce and limited in scope. *Behagh and Tehrani*¹⁶⁴ used full factorial experiments, regression, and Pareto ANOVA to model Ra, Rq, Rz, and ACL as functions of voltage, time, concentration, and filler content. Their study confirms that different roughness parameters respond to different dominant variables. However, the dataset includes only 16 experimental conditions, relies on linear and low-order models, and excludes bath aging, pulse dynamics, and multi-output interactions, highlighting the broader data-scarcity and generalization limitations of non-ML approaches.

Systematic evidence of field-wide limitations

Despite the diversity of methods, these studies address only isolated aspects of electroplating. Most focus on a single chemistry, a limited set of process parameters, or a single output (corrosion rate, thickness, additive content). A comprehensive systematic review by *Maphanga et al.*⁴⁴ reinforces this point: out of 5,851 publications screened over two decades, only 22 studies worldwide met the criteria for ML-based electroplating optimization, illustrating the global scarcity of robust data-driven models.

Their review identifies several structural barriers:

- Severe data scarcity and lack of standardization: Electroplating datasets are typically small, expensive to generate, and process-specific, making generalizable ML models difficult to train.
- Single-output prediction focus: Most models predict only thickness, deposition rate, or corrosion resistance, ignoring critical linked surface properties such as roughness, gloss, and brightness.
- No modeling of bath aging: None of the reviewed studies account for electrolyte evolution, impurity accumulation, or electrode conditioning over time, despite these being dominant factors in industrial variability.
- Poor extrapolation: Existing models perform well only within narrow interpolation ranges and fail when bath conditions drift, or new geometries are introduced.
- Chemistry-specific approaches: Methods developed for Ni–P, Ni–W, decorative nickel, or electroless systems do not transfer to copper electroforming or pulsed-current systems.

Taken together, historical work, recent studies, and systematic reviews reveal a consistent pattern: current data-driven electroplating research is promising but fundamentally narrow and fragmented. Across the literature, existing models rely on small, chemistry-specific datasets, target only a single surface output at a time, and assume static or idealized bath conditions. Most approaches ignore the interaction between bath-state evolution and pulse-current dynamics, struggle with extrapolation or out-of-distribution scenarios, and do not capture the coupled behavior of roughness, gloss, and deposition characteristics. As a result, current methods fall short of providing reliable or generalizable insights that can support production settings where variability and aging effects dominate.

Despite decades of work, no existing study offers the kind of holistic, physics-informed, multi-output predictive framework required for modern electroforming. There is currently no approach capable of simultaneously linking evolving bath chemistry, pulse-current waveform behavior, and resulting surface roughness, gloss, and deposition efficiency, particularly in copper electroforming, where electrolyte aging plays a central role and mass personalization demands coordinated control of multiple surface-quality metrics. This gap underscores the need for a unified modeling strategy

that integrates bath-state measurements, physicochemical variables, and pulse-dynamics to provide actionable predictions across diverse operating conditions.

3. Towards scalable and cost-efficient mass personalized production

3.1 Introduction

For more than a century, modern manufacturing systems have been shaped by the principles of mass production. A key economic aspect of these systems is that the unit cost of production decreases as the number of manufactured parts increases. Fixed costs such as tooling, setup, and overhead are distributed over a large number of identical parts, enabling significant cost reductions once production scales up ¹. This cost–volume relationship highlights the industrial models that have enabled standardized products to be manufactured efficiently and economically at large scales.

However, industrial trends are shifting toward increasing product diversity and personalization. This shift is driven by changing consumer expectations, rapid advances in digital technologies, and the desire for competitive differentiation ^{45,3}. In such scenarios, production volumes are typically low, product designs are frequently modified, and the economies of scale characteristic of mass production are no longer guaranteed. In many cases, unit costs remain constant or even increase with personalization.

The central challenge is therefore to develop manufacturing strategies that retain economic scalability (decreasing cost per part when increasing production volume), while enabling high levels of personalization. Achieving this requires rethinking not only manufacturing technologies but also process architectures and the way production systems are organized.

3.2 From mass production to mass personalization

Mass production represents one end of the manufacturing spectrum: standardized products manufactured in large volumes with significant economies of scale. At the opposite end lies mass personalization, in which individualized products are manufactured at scale, often with each unit being unique or specifically adapted to customer requirements ⁴⁶.

An intermediate concept, mass customization, emerged in the late 1980s ⁴⁶. In mass customization, customers choose among predefined modules or configurations, and production lines are optimized to accommodate this limited set of variations efficiently. While this allows some degree of variety, it does not permit arbitrary design changes.

In mass personalization, a most recent manufacturing trend, customers actively participate in shaping the design rather than selecting from predefined options ¹⁶⁵. This leads to frequent design

modifications, small batch sizes, and highly diverse production. These characteristics introduce additional costs into manufacturing processes. The combined effect of these factors is that the cost per part in mass personalization does not decrease with volume in the same way as in mass production. This economic mismatch is one of the main barriers to making personalization viable at an industrial scale.

3.3 The need for a new process architecture

While advances such as cyber-physical systems, digital twins, and reconfigurable manufacturing systems have improved flexibility and responsiveness^{166,167}, these technologies often increase indirect costs. Workforce training, data infrastructure, cybersecurity, and integration complexity can all add substantial overhead. Incremental improvements to existing processes are therefore insufficient to resolve the fundamental economic challenges posed by personalization.

Instead, what is required are new process architectures that intelligently combine existing manufacturing methods to achieve both flexibility and scalability. Such architectures must exploit complementarities between different technologies to parallelize workflows, reduce non-recurring costs, and maintain high throughput even when product designs vary significantly.

The key idea is to combine manufacturing technologies strategically so that the flexibility needed for personalization is preserved while introducing mechanisms that recover the cost-reduction behavior typical of mass production.

3.4 Literature landscape: attempts to scale personalization

The transition toward highly individualized production brings fundamental economic and technological challenges, chief among them the escalating cost per unique part as product variation increases. Over the past decade, researchers have proposed numerous strategies to improve manufacturing adaptability, resource utilization, and responsiveness; however, few directly target the reduction of cost per part in scenarios where each unit differs. The literature reveals a critical gap: existing methods enhance flexibility and automation but rarely achieve economic scalability under high variability.

A major research stream has focused on the deployment of digital-twin and cyber-physical frameworks within Industry 4.0 architectures. These systems establish a continuous digital representation of the physical process to optimize production planning, maintenance, and quality control. *Soori et al.*¹³ reviewed digital-twin applications in smart manufacturing, showing measurable improvements in process monitoring and predictive maintenance that indirectly influence productivity and resource efficiency. *Leng et al.*¹⁰ expanded this concept by demonstrating that real-time synchronization between virtual and physical layers allows reconfiguration of complex production systems with minimal downtime. Similarly, *He et al.*⁹ emphasized the role of twin-driven manufacturing for sustainable, intelligent production, noting

its potential for lowering operational inefficiencies but acknowledging that explicit cost models remain largely absent. These works collectively strengthen process adaptability but stop short of demonstrating how per-unit cost behaves when every component produced is unique.

Parallel developments have sought to translate these digital capabilities into physical production flexibility through modular and reconfigurable systems. *Koren et al.*¹⁶, *Mehrabi et al.*¹⁶⁸, and *Hu et al.*¹⁶⁹ introduced the principles of reconfigurable manufacturing systems (RMS) as a means to retool production lines quickly for new designs. These studies verified reductions in changeover time and ramp-up delay, but capital intensity and integration complexity often offset the economic gains when batch sizes fall below ten units. Consequently, while RMS increases responsiveness, it does not guarantee a sustainable decrease in per-part cost at high levels of personalization.

More recent attention has turned to AM as a route to individualized geometry without dedicated tooling. AM is inherently suited for customization because it fabricates parts directly from digital models; yet its economic performance remains constrained by slow build rates, high machine amortization, and limited material throughput. *Baumers et al.*¹⁷⁰ quantified the total cost structure of AM and concluded that the cost per unit only declines when machine capacity utilization is maximized. *Ivkic et al.*¹⁷¹ expanded this by analyzing the cost–benefit profile of “additive-manufacturing-as-a-service,” demonstrating that machine time and energy consumption dominate cost even when parts are printed on demand. *Cokyasar et al.*¹⁷² investigated capacity-allocation models across distributed AM networks and confirmed that throughput scheduling can improve profitability but requires a steady inflow of diverse yet combinable jobs. *Luft*¹⁸ proposed a quantitative framework linking flexibility to cost efficiency but acknowledged that most results remain theoretical rather than empirically validated in individualized production. *Framinan et al.*¹⁷³ surveyed operations-research approaches in AM and highlighted that while scheduling and capacity optimization are advancing, studies capturing the economic trajectory of cost per part under extreme variation are virtually nonexistent.

Further techno-economic research continues to refine cost modeling. *Rintala et al.*¹⁷⁴ conducted a systematic review of 255 studies, indicating that the economically viable use cases for additive manufacturing remain limited, which in turn constrains the real-world relevance and applicability of many of the advanced optimization models developed for the field. *Thomas and Gilbert*’s NIST analysis¹⁹ remains a cornerstone reference, identifying energy consumption, cycle time, and utilization as the governing factors for AM cost behavior. Together, these findings show that even advanced optimization and scheduling strategies cannot overcome the fundamental cost drivers of build time and equipment depreciation. Across these contributions, the same pattern persists: additive and digital technologies successfully expand design freedom but fail to replicate the cost efficiency of conventional serial production.

The combined evidence suggests that existing Industry 4.0 and AM strategies have effectively addressed operational challenges, such as information latency, setup complexity, and reconfiguration effort, but still fail to deliver a measurable reduction in unit cost under high-variation production. Even studies claiming economic benefits typically rely on mixed builds, geometric repetition, or modular product families rather than truly unique geometries. Consequently, the economic scalability of fully personalized manufacturing remains unresolved.

This structural shortfall exposes a clear research gap: there is no empirical, adaptive framework that quantifies and optimizes cost scaling in highly individualized production. Most studies emphasize flexibility, responsiveness, or system intelligence, but only a few explicitly investigate how unit cost evolves when each part differs. The literature, therefore, advances technical capability more than economic understanding, leaving open the key question of how to sustain or reduce per-part cost as design variability increases.

Within this context, additive manufacturing emerges as the most promising avenue toward economically viable personalization because of its intrinsic geometric freedom and tool-less operation. Nevertheless, existing research focuses largely on build quality and process performance rather than systematic cost-scaling behavior across diverse production conditions. This indicates that AM possesses significant untapped potential to address the cost-scaling challenge, yet realizing that potential demands more integrated approaches that go beyond flexibility to directly target economic scalability.

3.5 Strategic concept: hybridization for scalable personalization

A promising solution to the personalization cost-scaling challenge is to hybridize additive manufacturing with electroforming. Additive manufacturing, particularly fused deposition modeling (FDM), can rapidly fabricate customized polymer molds without incurring extra costs for each new design. Electroforming can then deposit metal onto these molds to create high-precision parts.

The critical insight is that mold fabrication can be parallelized across multiple low-cost 3D printers, while electroforming can be performed in centralized batches. By printing several personalized molds simultaneously and electroforming them together, the cost per part decreases as the batch size increases, even if every part is geometrically different.

This approach enables manufacturers to recover the cost-down behavior of mass production not by standardizing the product, but by standardizing and parallelizing the manufacturing steps. It represents a conceptual shift in how personalization and scalability can be reunited.

3.6 Conclusion

This chapter has outlined the economic and technological challenges associated with mass personalization. It has shown how personalization disrupts the classical cost–volume relationship and why existing technological solutions often fail to address this issue at its root.

A review of current literature highlights the absence of manufacturing strategies that combine personalization with cost-scaling behavior similar to mass production. The hybridization of additive manufacturing and electroforming offers a promising pathway toward scalable and economically viable personalized manufacturing.

The following chapter presents an experimental demonstration of this concept:

“Low-cost manufacturing of high-precision personalized flexures by a hybrid 3D printing-electroforming technique.”

This work illustrates how hybrid manufacturing can be structured to enable high-precision, low-cost, and scalable mass personalization.

4. Low-cost manufacturing of high-precise personalized flexures by a hybrid 3d printing-electroforming technique ⁶⁸

4.1 Introduction

During the past century, mass production reduced fabrication costs noticeably by spreading the manufacturing overhead over a large amount of identical produced parts ¹. Although mass production has indisputable advantages, such as increased productivity and reduced production costs, it suffers from flexibility ⁴⁵. Mass customization, a terminology first introduced in academia in 1987, is a natural evolution of mass production ⁴⁶. Mass customization allows the customer to customize the product by choosing among existing options. Fabrication lines are then set up to allow the production of these customized products.

However, nowadays, manufacturing industries face more and more demands for personalized products ³. In personalized products, unlike customized products, customers are actively involved in the design and do not limit themselves to predefined options. As a result, production is characterized by low volume combined with high diversity. Current production lines are, in general, not able to handle such demands. The manufacturing industry has to adapt to this new reality. Digital manufacturing, or Industry 4.0, is one of the promising approaches to address this challenge ¹⁷⁵. In digital manufacturing, cyber-physical systems are deployed toward achieving the needed flexibility. These systems enable quick and efficient adaptation of the production lines to the customer design inputs ¹⁷⁵. However, changing design inputs generate extra production costs, which can be classified following the 6M methodology of Ishikawa (Figure 4.1).

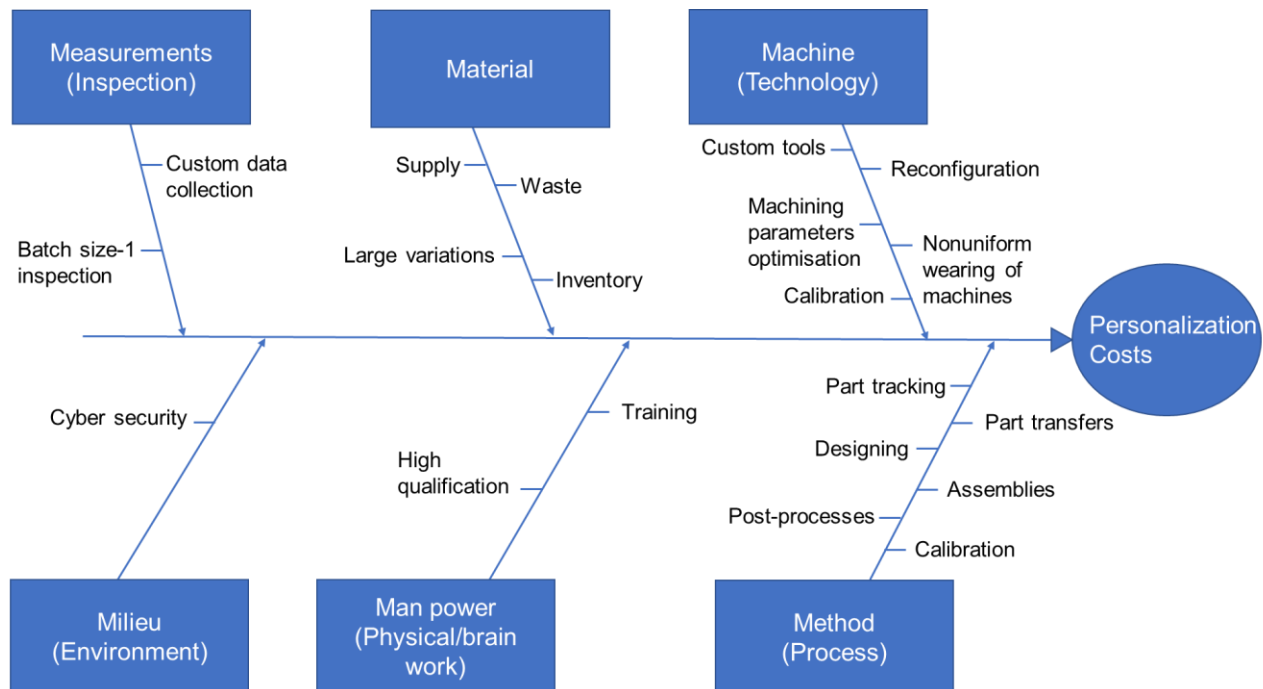


Figure 4.1 Additional costs related to personalized part production categorized according to the 6Ms of Ishikawa: 1) Machine (technology); 2) Method (process); 3) Material (Includes Raw Material, Consumables, and Information.); 4) Man Power and Mind Power; 5) Meas

In the measurement category, extra manufacturing costs appear from the fact that small batches of unique products are produced in mass personalization. Compared to mass production, inspection by sampling can no longer be used to ensure the products meet the set requirements. An increased number of inspection operations will be required adding extra costs to the product ¹⁷⁶.

The usage of cyber-physical systems and digital technologies comes with a high risk of cyberattacks, adding extra manufacturing costs in the milieu category for deploying counter-measures ¹⁷⁷.

Leaving the freedom to the customer to actively influence the design causes additional material costs. Since there is no predefined design, the required material is less predictable. As a result, it becomes challenging for industries to obtain the needed material at reasonable prices and difficult to set up a sustainable supply chain ¹⁶⁵.

The deployment of cyber-physical systems and digital technologies will require new skills from the employees. This requirement causes further manufacturing costs in the man-power category in the form of training and higher salaries to attract highly qualified personnel ^{178,179}.

Changes in design generate as well extra manufacturing costs in the machining processes. For example, custom tools required to fabricate a personalized part can no longer be used over a large number of products. Reconfiguration, re-calibration, and process parameters optimization also

bring additional manufacturing costs that can hardly be distributed over a significant quantity of products, as was the case in mass production. Further, since the designs are not defined in advance, achieving a uniform wearing of tools and machines during production is more challenging, which may reduce the lifetime of systems and add extra maintenance costs¹⁸⁰.

Most importantly, manufacturing personalized products increases the costs of manufacturing processes such as assembling, part transferring, designing, and post-processing. Since customers come with their inputs and consequently, the product design is changed over and over, there is no universal procedure, for example, for assembling or transferring the parts between different manufacturing cells and calibrating the machining processes. As a result, manufacturing time and costs required to manage the processes increase^{166,167,181}. The usage of cyber-physical systems can significantly help to address these challenges. However, it is still essential to develop novel manufacturing processes in favor of reducing the effects of the above-mentioned costs in personalized product fabrication.

In summary, mass personalization's challenge is to allow factories to massively produce personalized products (with designs not defined in advance) such that the above-discussed manufacturing costs can be spread among a high production volume, reducing the cost per part as in mass production.

As a first step, existing processes can be revisited and rethought in order to adapt them to this new reality. Novel approaches can also emerge from combining established processes in innovative ways better suited for producing unique parts.

In this study, AM and electroforming processes have been selected to illustrate how combining existing manufacturing technologies can help reduce costs per part as the production volume increases in the context of mass personalization. A hybrid technique combining these processes allows taking advantage of the capability of producing complex geometries by additive manufacturing and fabricating massively, in parallel, parts by electroforming with no extra costs due to design changes by the customer. The manufacturing of flexure mechanisms has been selected as an example of mass personalized manufacturing of complex micro designs. As a demonstrator, the fabrication of a double compensated X-Y stage is selected.

Further, the scalability of the proposed method for mass personalization is discussed by comparing two scenarios: one with low-cost and one with high-end 3D printers. The cost analysis shows that the manufacturing time and cost per part are reduced by increasing the number of deployed 3D printers. In this way, as in mass production, it becomes possible to reduce the cost per part as production volume increases, and this, regardless of design changes by the customer, by increasing the number of manufacturing cells.

4.2 Mass personalization of flexures

4.2.1 Flexures

Flexure mechanisms are jointless, monolithic mechanical devices providing smooth and high-resolution motion guidance ¹⁸². A further advantage is that being monolithic, they require no assembly operation. Flexure mechanisms play a crucial role in high-precision positioning systems ¹⁸³. For example, flexures are used as components in manufacturing systems of semiconductors, in electrostatic actuators of microelectromechanical systems, in manipulators for biological cells, in positioning stages of atomic force microscopes, or in micro-assembly manipulators ^{184,185}. By implementing flexure hinges rather than conventional bearings in joints, issues such as friction, hysteresis, stick-slip motion, lubrication, and assembly can be eliminated ¹⁸⁵. In addition, flexure hinges can be applied in micro-scale applications without any maintenance needs ¹⁸⁶.

The choice of the manufacturing process is a crucial step in fabricating flexure mechanisms. Flexures tend to be manufactured monolithically to reduce their fabrication costs by avoiding assembly operations and achieving high-precision systems ¹⁸⁷. There are a variety of manufacturing processes used in industry for the fabrication of flexure mechanisms, such as jig boring ¹⁸³, additive manufacturing by electron beam melting (EBM) ¹⁸⁵, wire electric discharge machining (Wire-EDM) ¹⁸⁶, laser cutting ¹⁸⁶, metal stamping ¹⁸⁶, computer numerical controlled (CNC) milling ¹⁸⁶, water jet cutting (WJC) ¹⁸⁷ or direct metal/polymer 3D printing ¹⁸⁸.

However, these manufacturing processes have limitations when fabricating high-precision personalized flexure mechanisms. Water jet cutting fabricates flexure hinges with tapered walls ¹⁸⁷. Conventional machining methods like CNC milling are limited by flexure hinge size ¹⁸⁶. 3D printing of polymers results in low strength and consequently limited fields of applications for flexure mechanisms. Porous surface finishes in 3D printed metal parts cause poor mechanical performance and decrease the accuracy of mechanical motions in flexure mechanisms ¹⁸⁸. In addition, available 3D printers are not capable of manufacturing highly precise flexures with thicknesses thinner than typically 0.5 to 1 mm ¹⁸⁹. Wire-EDM fabricates flexure mechanisms with high-quality surfaces and cuts ¹⁸⁶. However, Wire-EDM can cause heat-affected zones and thermal stresses on the metal workpiece due to the high-temperature spark discharges used for machining ¹⁹⁰. As a direct consequence, costly machining process optimization is needed in order to reach the required quality ¹⁹¹. Even once the machining process is optimized, Wire-EDM remains an expensive machining process, mainly due to numerous needed setup steps ¹⁸⁷. As such, especially considering the costly optimization process, wired-EDM becomes too expensive in the context of low batch size personalized part fabrication.

4.2.2 A possible approach for mass personalization of flexures

Presently used manufacturing methods for flexures are not well suited for mass personalization as design changes requested by the customer translate into extra manufacturing costs. The fabrication of flexures by electroforming in 3D printed molds could be a potential approach to fabricating personalized flexures. Indeed, the additive manufacturing of the mold is achieved without extra

costs due to design changes. The possibility to combine several of these molds in a single plating batch allows parallelizing the workflow and, in this way, reduce the manufacturing costs per part regardless of design changes.

Employing molds to electroform parts is applied in MEMS fabrication processes¹⁹². Electroforming is a reproducible machining technology with several advantages, among them accuracy¹⁹³. 3D printing of polymer parts is an accessible and economical method for rapidly fabricating custom parts with complex geometries⁷². A significant advantage in combining these two processes is that electroforming in 3D printed molds can partially be parallelized since several personalized parts can be electroformed in the same plating batch. The combination of 3D printing and electroplating has recently gained increased interest in academia. Electroplating 3D printed plastic mandrels is an emerging hybrid manufacturing technology that fabricates low-cost, personalized, lightweight, and durable plastic parts with metal skin. Numerous industries, such as automotive, power, aerospace, dental, and health companies, currently employ this hybrid technique to reduce manufacturing costs.

Both electroplating and electroforming are performed by electrodeposition. However, in electroforming, the mandrel is removed after the solid metal part is formed, whereas, in electroplating, the mandrel is not removed, and the finished product is a part with a thin metal layer. So far, most studies have been conducted on electroplating or electroforming on external surfaces of 3D printed parts rather than electroforming in 3D printed molds.

Electroforming in 3D-printed molds presents some challenges. First of all, a uniform, highly electrically conductive surface is required to perform electroforming¹⁸⁵. Further, removing the electroformed part without breaking it must be possible. Several studies have investigated methods to provide a conductive surface on a 3D printed polymer mold. They can be categorized into three main approaches.

The first approach, as investigated by *Ryosuke Matsuzaki et al.*⁷⁰, is to electroform in a mold on top of a metal substrate. A metal plate is employed as a base on which the molds are 3D printed. The electroformed part is afterward detached mechanically from the substrate and the mold. However, the mechanical release of the electroformed parts is labor intensive and presents significant design limitations. In particular, fine structures, such as those required by high precision flexures, are not possible as they will be damaged during the mechanical release from the base and mold.

The second approach is printing conductive surfaces using conductive filaments. Simultaneously 3D printing of conductive composites and non-conductive thermoplastics was suggested by *Kristin Angel et al.*⁷². For selective electrodeposition, the authors used a fused filament printer with multiple extruders to print conductive and non-conductive regions concurrently. Although this technique can print conductive molds with complex geometries (adding significant flexibility to the design), it has difficulty producing a highly conductive surface for subsequent electroforming⁷². Regardless of the filament's printing direction, the electrical resistance of the printed parts is high compared to metal substrates or coated surfaces. In addition, 3D printing parts with two filaments having different mechanical and thermal characteristics (conductive and non-conductive

filaments) result in lower printing quality and, eventually, non-uniform surfaces after electroforming.

The third approach is to employ a polymer substrate, such as acrylonitrile butadiene styrene (ABS), and make it conductive by electroless coating or applying a conductive paint. For instance, the usage of electroless plating for metallization on FDM parts was investigated by *Eßbach et al.*¹⁹⁴ and *Equbal et al.*¹⁹⁵. Still, this method has limitations, such as the requirement for several surface preparation steps prior to plating and the most critical one: the conductivity of 3D printed parts highly depends on the electroless bath substances and the FDM part's surface roughness. Also, selective electrodeposition is not possible unless additional masking techniques are deployed. This is a serious design limitation in the context of molds, as only the bottom of the mold is allowed to be conductive in order to obtain high-quality electroformed parts. Using conductive paints is proposed by *Saroj Kumar Padhi et al.*¹⁹⁶. Various metalization techniques, such as applying conductive silver paint, painting of annealed chemically reduced silver colloidal solution, and painting of aluminum-seeded paste, were investigated to apply a thin layer of metal on the outer surface of plastic 3D printed parts. The results showed that silver paint has the best performance in metal forming.

The studies mentioned above show that personalized parts can potentially be fabricated by electroforming in 3D printed molds. However, open questions remain in order to prove the feasibility of fabricating high-precision personalized parts with this approach. Fine delicate structures, such as thin micro-hinges required by high precision flexures, cannot be mechanically detached from the mold without being damaged. Releasing the part from the mold is possible by dissolving the mold instead of breaking it mechanically. However, this approach rules out the possibility of building the electroformed part directly on a highly conductive metal plate. Instead, a structure formed by a dissolvable material coated by an electrically conductive layer is required. Therefore, an assembly of a dissolvable mold, a conductive layer, and a dissolvable base substrate is essential to electroform high-precision personalized metal parts. The present paper presents possible solutions to fabricate such assemblies.

The key question is to determine the capability of this approach to fabricate mass personalized fine delicate, precision structures. So far, the primary focus of the published studies presenting the forming of metal parts on 3D printed mandrels was on the fabrication methods rather than the investigation of which precision can be achieved. A relevant question of practical importance is investigating to which extent it is possible to fabricate high-precision personalized structures. The present paper shows that it is feasible to fabricate micro-hinges in the size range required for relevant applications of flexures.

In the present work, three different approaches are investigated for electroforming in 3D printed molds by comparing their ability to manufacture a dissolvable personalized mold while providing a conductive surface required for the electroforming process. The fabrication of hinges with dimensions relevant to applications in high precision personalized flexures is demonstrated. The proposed approaches are validated by the fabrication of a double compensated X-Y micro-positioning stage.

4.3 Experimental

4.3.1 Mold fabrication

3D printing was performed on an Ultimaker 2+ FDM printer with black ABS filaments since ABS allows the fabrication of precise 3D parts with low thermal expansion ¹⁹⁷. A 250 μm nozzle in conjunction with adaptive layers (to reduce build time) was employed for 3D printing. To avoid the usage of extra masking techniques, an approach consisting of fabricating the molds by an assembly of a conductive "substrate" and an "upper mask" was chosen (Figure 4.2). Three different techniques have been compared to fabricate the mold assemblies. In all cases, conductive paint was used to provide a conductive layer.

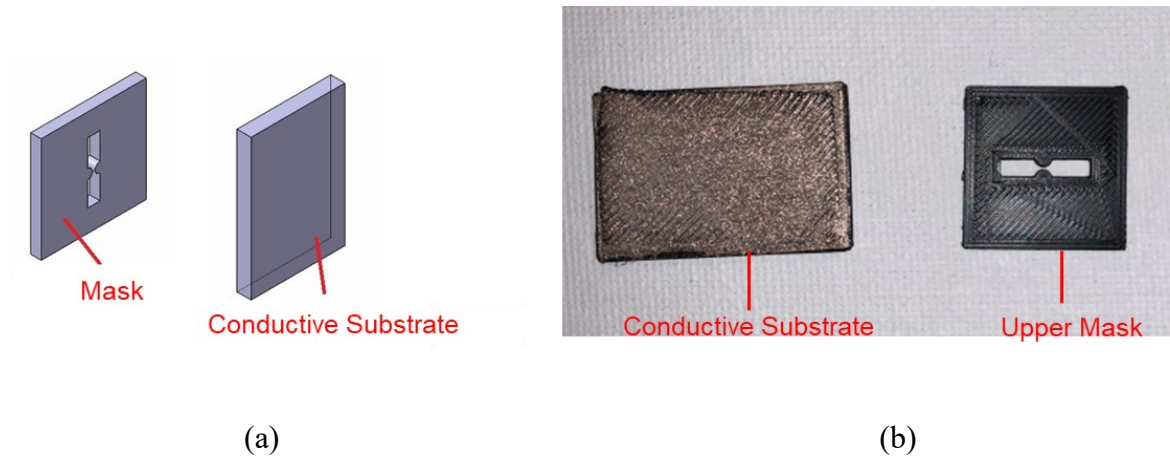


Figure 4.2 a) The primary components of molds are the mask and substrate. b) Example of a 3D printed substrate and 3D printed mask

Spray glue bonding (SGB) molds

In spray glue bonding, the mask and substrate are printed separately. Afterward, the substrate is sprayed with conductive silver paint (MG Chemicals 842AR-140G). Once the conductive paint is thoroughly dried, a 3M Super 77 Multipurpose Spray Adhesive glue is sprayed on the surface of the mask. In the last step, mechanical pressure is applied on top of the mask to bond it with the conductive substrate.

Hot-press bonding (HPB) molds

The mask and substrate are printed separately in hot-press bonding, as in the SGB method. First, the substrate is sprayed with conductive paint. Once the paint is thoroughly dried, the mask and the conductive substrate are hot-press bonded together in a furnace by applying a pressure of 26 KPa at 115 °C for 3 hours ¹⁹⁸.

Conductive aerosol painting – 3D printing (CAP-3DP) molds

In conductive aerosol painting - 3D printing (CAP-3DP), the mask is 3D printed directly on the conductive substrate surface (Figure 4.3). Once the substrate is entirely 3D printed, aerosol silver paint is applied. After the paint application, the mask is printed on the conductive substrate without removing the substrate from the 3D printer build plate.

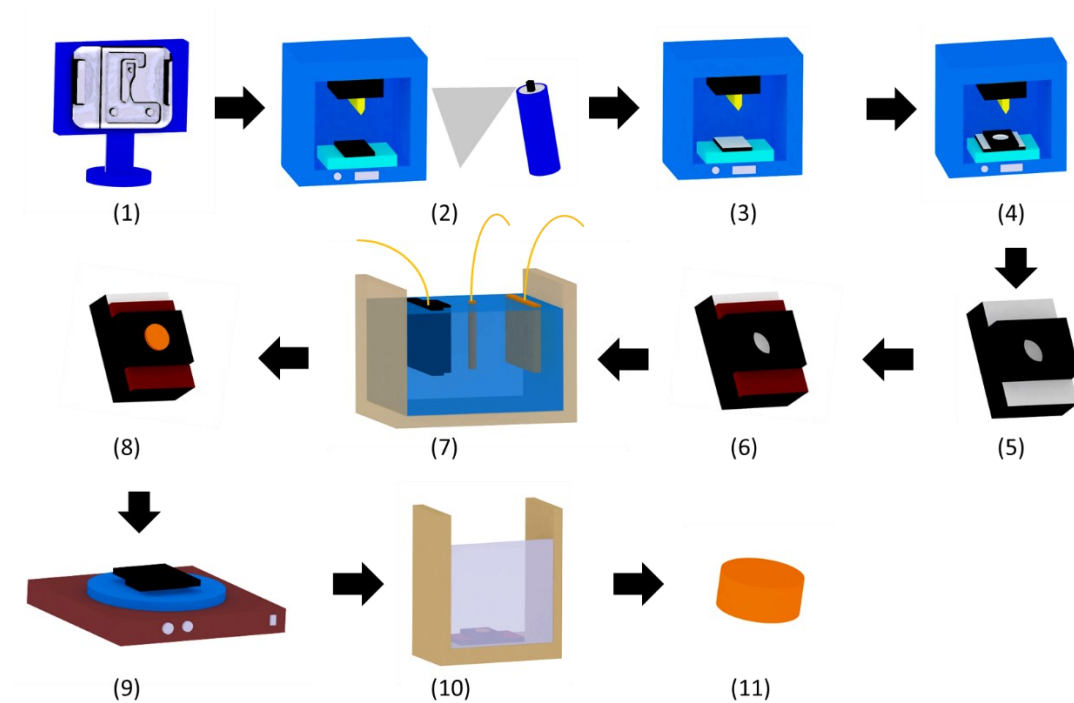


Figure 4.3 Manufacturing steps for personalized metal parts by the CAP-3DP method; 1) CAD design; 2) 3D printing of substrate and conductive aerosol painting; 3) 3D printed conductive substrate; 4) 3D printing of the mask on the conductive substrate; 5) Conductive mold assembly; 6) Sealing of the mold; 7) Electroforming in the conductive mold; 8) Electroformed part inside the mold; 9) Optional polishing of the electroformed part while still assembled inside the mold; 10) Dissolving ABS mold in acetone; 11) Final electroformed metal part

4.3.2 Electroforming process

The outer surfaces of the mold assembly were sealed by varnish to avoid electrodeposition on non-desired areas. A rectangle-shaped copper sheet (8 cm x 16 cm) is used as a counter electrode. In order to have optimal mass transfer and homogenization of copper ions during the electroforming process, the mold assembly is moved back and forth by a robotic arm parallel to the counter electrode. A thin copper rod placed near the mold assembly at a distance of approximately 2 mm served as a pseudo-reference electrode. Electroforming is performed in a 1M Sulfuric Acid (H_2SO_4) and 0.5M Copper Sulfate (Cu_2SO_4) solution. In addition, the electrolyte is stirred by a magnetic rod to homogenize the solution and avoid a non-uniform surface finish¹⁹⁹. Pulsed plating is used where deposition pulses (-150mV vs. Cu) and polishing pulses (+100mV vs. Cu) are alternated. Deposition pulses last until a charge of 1.33 C/cm^2 (corresponding to a thickness of about 1 micron) is passed in the electrochemical circuit. Polishing pulses last for 10% of the deposition charge. Adapt in-line the pulse duration by monitoring the electrical charge is essential in achieving high-quality thick deposits in additive-free plating baths²⁰⁰.

4.3.3 Mold releasing

Following the electroforming process, the electroformed mold was removed from the bath, rinsed with deionized water, and dried by blowing air. Subsequently, the rinsed and dried plated mold was submerged in acetone for at least 24 hours, after which the electroformed part could be detached effortlessly from the mold as the ABS was almost entirely dissolved in acetone. The final cleaning of the part was done in acetone and deionized water.

4.4 Results and discussions

4.4.1 Flow diagram of manufacturing personalized metal parts by CAP-3DP technique

Figure 4.3 illustrates the manufacturing steps of personalized metal parts, where the CAP-3DP method is used for fabricating the conductive mold assembly. Following the manufacturing of the mold assembly by the CAP-3DP approach, the assembly is sealed, and the part is electroformed. As the metal part is still firmly mounted inside the mold assembly, it can be mechanically polished as needed (either to obtain the desired surface finish and/or to adjust the part thickness). This presents a significant advantage in production as no extra steps are required to mount the part prior to mechanical polishing. Subsequently, the metal part is released by dissolving the mold assembly in acetone.

4.4.2 Fabrication of mold assembly

In this study, three different methods have been investigated to electroform metal parts in 3D-printed molds. The mold assembly is made of two components: a flat ABS substrate coated with conductive paint and a 3D-printed mask.

The first two methods investigated were spray glue bonding (SGB) and hot-press bonding (HPB). A significant advantage of employing these two methods is that the upper mask and the conductive substrates are fabricated separately. This means that both processes can be optimized independently in a production environment, and there is considerable freedom in choosing the substrate (for example, it is not required to 3D print the substrate). However, the main drawback is the bonding operation. Bonding the mask with glue to the conductive substrate is a time-consuming process in the SGB method. Further, avoiding glue leaking in undesired areas is challenging since the mask is pressed on top of the conductive substrate. In the case of the HPB method, as the mask is bonded to the conductive 3D printed substrate by hot pressing, geometrical changes happen in the mold assembly (typically 15 % and more).

To address the issues of the SGB and HPB methods, a third method, conductive aerosol painting – 3D printing (CAP-3DP), was investigated. Bonding operations (gluing or hot-pressing) are eliminated in the CAP-3DP method as the mask is printed directly on the conductive painted substrate.

Although the CAP-3DP method addresses glue leakage and geometrical deformation during hot-pressing, several aspects need special considerations and optimization. The key concerns are dimensional errors caused by the warping of the substrate, optimization of printing pause operation required for the application of the conductive paint, and the type of conductive paint. As a consequence, five key parameters must be chosen correctly:

- Substrate thickness
- Pause duration after paint application (in order to let dry the paint)
- Nozzle-substrate distance after the application of the paint
- Type of conductive paint
- Mold-substrate contact areas

It is well known that in 3D printing, warping causes dimensional changes. To avoid warping issues, thick enough substrates must be printed (typically a minimum of 3 mm thick). In case the substrate is not printed, but rather an existing ABS part is used, this issue is no longer present.

The printing pause duration needs to be optimized. A long enough printing pause duration is required in order to let the conductive paint semi-dry. Indeed, after too short pause times, when the mask is printed on the substrate, the 3D printer nozzle is able to move the wet paint, block narrow geometrical shapes, cause non-conductive spots on the substrate surface, and decrease

surface quality. On the other hand, ABS will not bond correctly on the painted substrate after excessively long pause times. The painted surface will be too dry to allow bonding with the ABS, affecting the upper mask dimensions and 3D printing quality. Pause times must be adjusted in function of the substrate thickness. Experiments showed that 3 mm thick substrates require a longer pause time than a 1 mm thick substrate to dry the paint correctly (as the thickness affects heat transfer from the build plate to the paint). Typically, a pause time of about 10 seconds on thin substrates and a pause time of 20 seconds for 3 mm substrates were found to be adequate.

The printed mask on the painted substrate can move in case of insufficient bonding between the paint layer and the printed mask, causing dimensional error during 3D printing. Sufficiently large contact areas between mold and substrate must be considered to minimize the occurrence of mentioned defect when designing the upper mask of the assembly (Figure 4.4). This requirement limits the design of flexures to be fabricated.

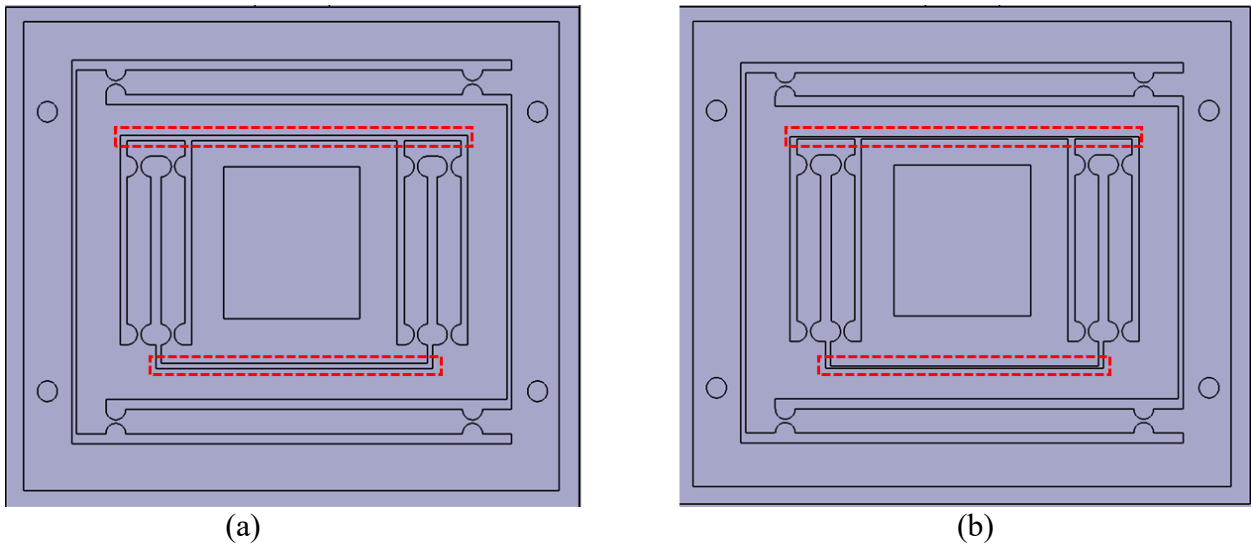


Figure 4.4 Mask and substrate with the same design but different contact areas: (a) a design that worked and (b) a design that failed.

Experiments showed that the choice of the conductive paint and 3D printer nozzle also affects 3D printing quality (a 250 μm was selected after several trials and errors). Various conductive paints (conductive copper paint, conductive silver-coated copper aerosol paint, and conductive silver aerosol paint) have been tested. Among all these paints, silver aerosol paint has the highest electrical conductivity (essential for the electrodeposition process), dries fastest, and results in the best printing quality of the mask (perpendicular 3D printed walls and minimal dimensional errors). In addition, silver paint demonstrated better surface quality after electroforming compared to other conductive paints¹⁹⁶.

4.4.3 Application: Fabrication of micro-hinges

Various types of hinge cut-out profiles are used in the design of flexures, including V-shaped²⁰¹, circular, right-circular, elliptical, parabolic, and hyperbolic hinges²⁰². Among all these profiles, circular flexure hinges have been chosen in this study.

Under small enough deformations to remain in linear conditions, the angular stiffness K [N·m/rad] of a circular hinge is given by²⁰³:

$$K \approx \frac{2Ew}{9\pi} \sqrt{\frac{e^5}{r}} \quad 4.1$$

where E is the Young modulus of the material, e is the thickness, and r is the radius of the circular profile. The flexible pivot has a width of w (Figure 4.5). Equation 4.1 shows that the geometric parameter affecting most of the bending stiffness is the hinge thickness, e . In order to maximize the stiffness in the other degrees of freedom, the width w has to be maximized. Studies show that the rotation performance of circular hinges is improved by increasing r while decreasing e and w ¹⁸⁶. To avoid stress concentration at the neck, the ratio $\frac{r}{e}$ should be typically greater than five²⁰³.

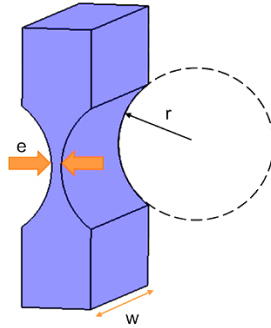


Figure 4.5 Circular flexure hinge

Typical values for e in circular flexure hinges used in high-precision applications range from 100 to a few hundred micrometers. For example, *P. Gräser et al.*²⁰⁴ proposed a high-precision and large-stroke X-Y micropositioning stage based on serially arranged compliant mechanisms with flexure hinges thickness of $e = 100 \mu\text{m}$. *S. H Chang et al.*²⁰⁵ fabricated a high-resolution micropositioner using flexure hinges with $e = 250 \mu\text{m}$. Also, *T.C. Tseng et al.*²⁰⁶ designed a monolithic aspherical mirror bender with the help of circular flexure hinges with $e = 300 \mu\text{m}$. *T. Cheng et al.*²⁰⁷ worked on a trapezoid-type sick-slip piezoelectric linear actuator using the circular flexure hinges with $e = 500 \mu\text{m}$. *Qingsong et al.*²⁰⁸ studied an XY parallel micromanipulator with circular hinge thickness $e = 1 \text{ mm}$.

In order to illustrate the CAP-3DP method capabilities and limitations for the fabrication of micro-parts, flexure hinges with different hinge thicknesses e , covering the typical values used in applications, have been manufactured and characterized. Figure 4.6 shows the manufacturing of circular flexure hinges with various thicknesses e (140, 170, 200, 230, and 260 μm). The thickness w of the formed part was 730 μm . The main limitation in reaching such small dimensions is the 3D printer resolution. Figure 4.7 shows a micrograph of an electroformed flexure hinge with the minimum achieved hinge thickness $e = 140 \mu\text{m}$ by the CAP-3DP method on our laboratory Ultimaker 2+.

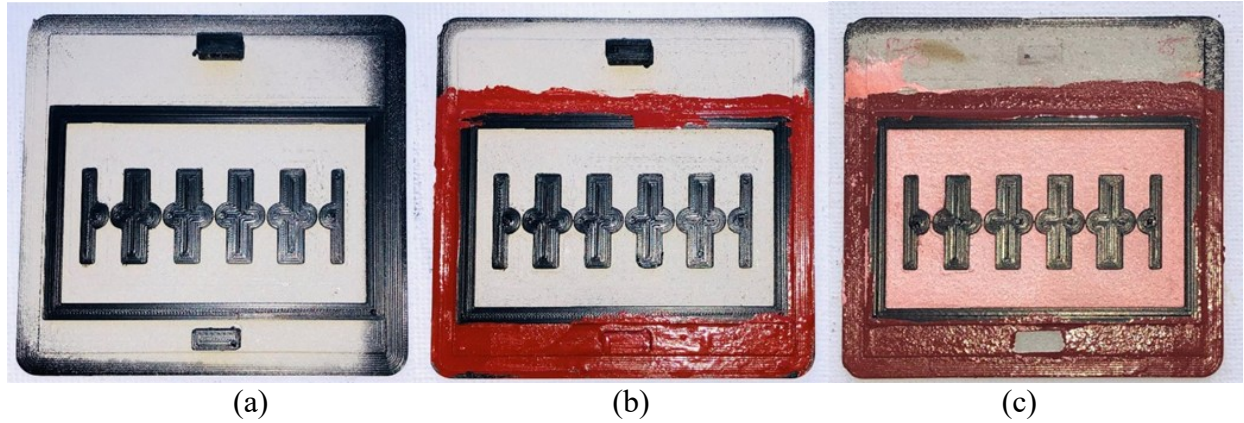


Figure 4.6 a) CAP-3D printed circular flexure hinges with various thicknesses e (140, 170, 200, 230, and 260 μm) and width of $w = 730 \mu\text{m}$. b) Sealed mold before electroforming and c) Electroformed CAP-3D printed mold.

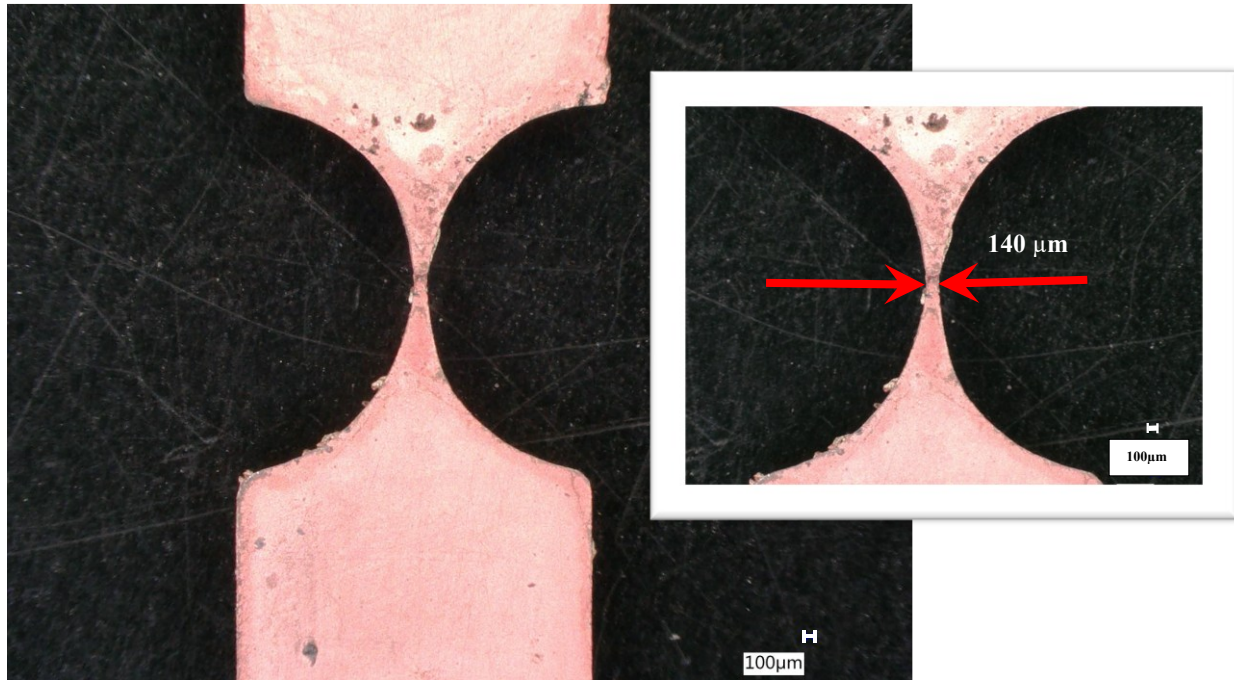


Figure 4.7 Picture of an electroformed flexure hinge with the smallest achieved thickness e equal to 140 μm .

4.4.4 Mechanical characteristics of the CAP-3DP electroformed hinges

In order to characterize the mechanical deformation of the electroformed CAP-3DP parts, the following experiment was conducted. A single hinge with thickness $e = 330 \mu\text{m}$ was manufactured according to the proposed CAP-3DP method (Figure 4.8).

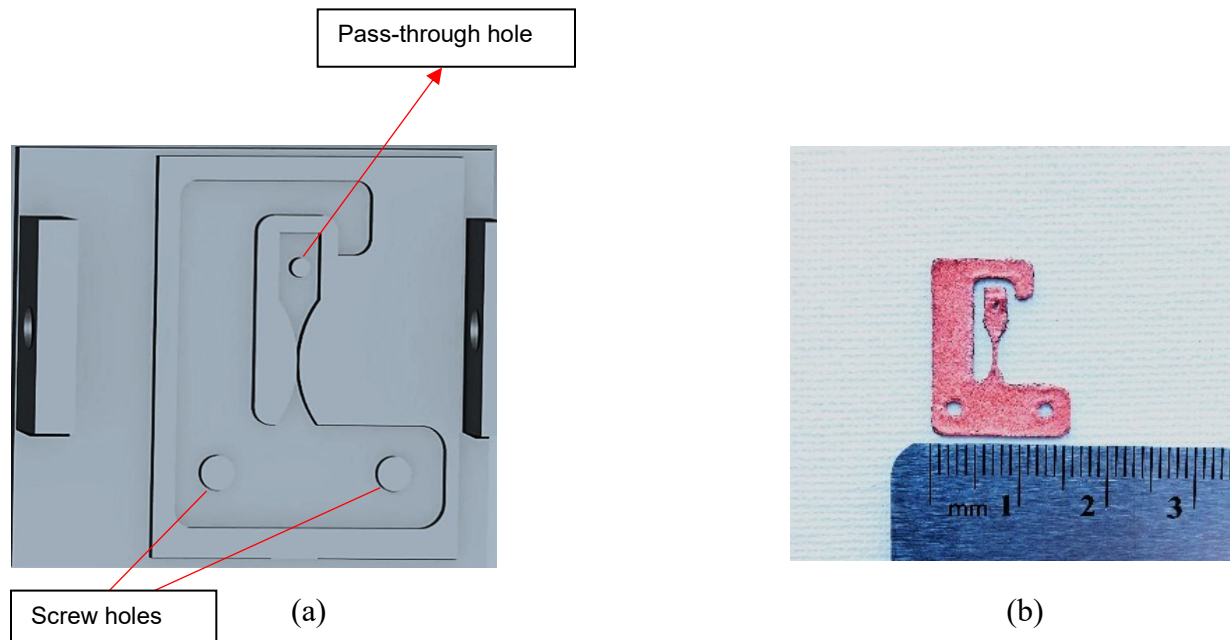
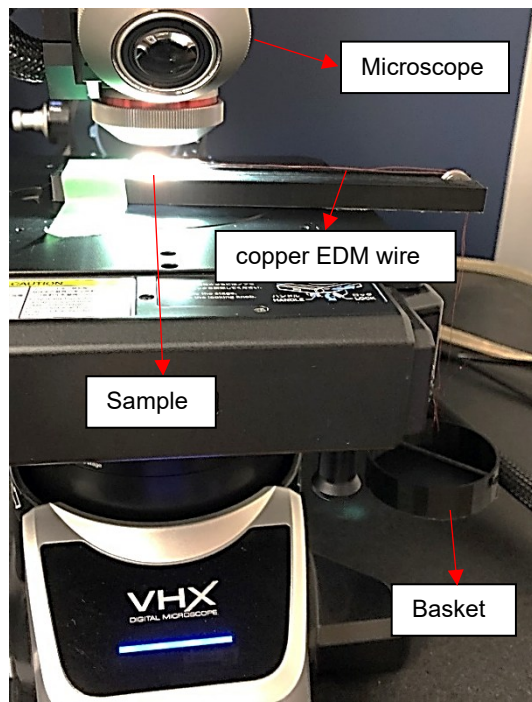
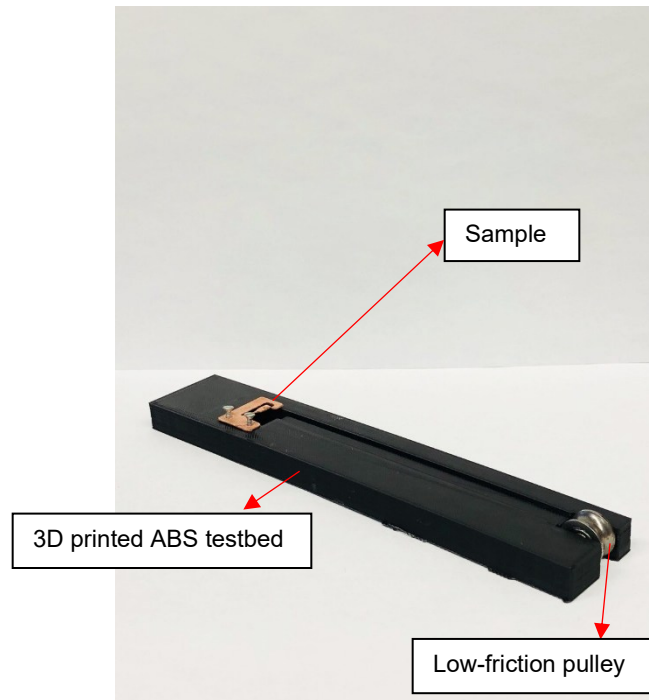


Figure 4.8 Hinge with thickness $e = 330 \mu\text{m}$ and width $w = 500 \mu\text{m}$ a) CAD design and b) Final electroformed part

An in-house setup was used to illustrate the mechanical deformation of the electroformed hinge (Figure 4.9.a). First, the electroformed CAP-3DP hinge was fixed with screws on a 3D printed ABS testbed (Figure 4.9.b). Then, a 3D printed ABS basket was attached to the test part with a rigid copper wire over a low friction pulley. An optical microscope captured the angular deformation of the electroformed hinge when weights from 0 to about 41 grams were added in 24 steps (in each step, an extra 1.7 grams of mass was added). Figure 4.10.a shows the measured mechanical deformation in the function of the applied moments to the electroformed hinge. A maximum moment of $1.5 \text{ mN}\cdot\text{m}$ can be applied to the electroformed hinge before reaching the plastic region.



(a)



(b)

Figure 4.9 a) Setup to characterize mechanical deformation of the electroformed hinge under a microscope b) The electroformed hinge is screwed on an ABS 3D printed testbed allowing mechanical deformation by applying various weights through a copper wire attached to the hinge and passed over a low-friction pulley

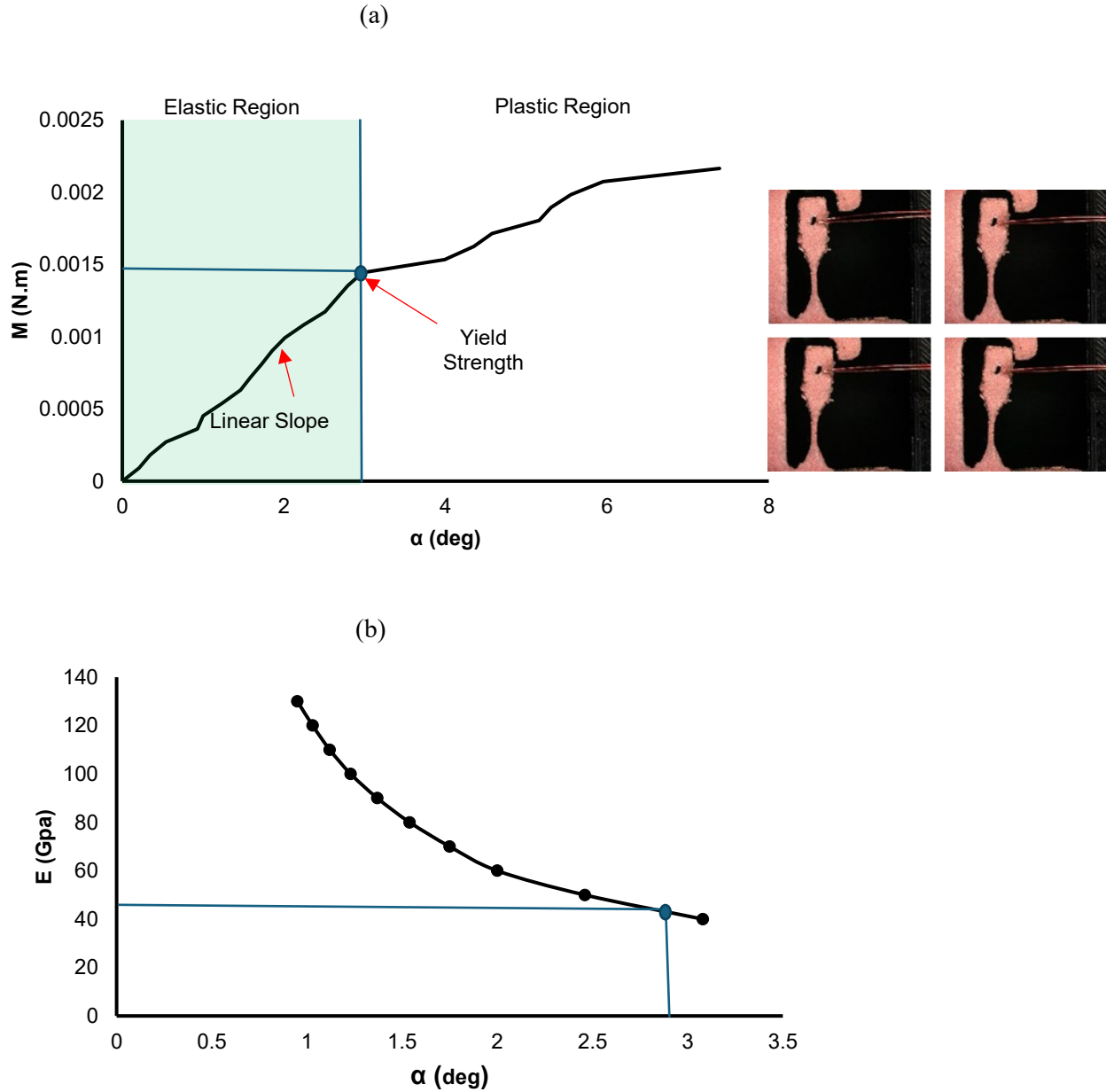


Figure 4.10 a) Measured angular deformation α of the electroformed hinge in the function of the applied moment M . Linear and plastic deformation regions are clearly visible. b) Deformation angles computed by Finite Element Analysis for various Young modulus for an applied moment of 1.5 mN·m. (The blue point pictured in both figures is a reference point to compare the experiment and FAE results in the same conditions)

Experimental results are compared with Finite Element Analysis (FEA) using the same design and material and applying a moment of 1.5 mN·m. The mechanical properties of electroformed parts can noticeably change by varying bath composition and plating parameters¹⁹⁹. In addition, it is well known that electrodeposited metal has a lower Young modulus than the corresponding bulk

metal²⁰⁹. Consequently, for the simulations, values from 40 to 130 GPa are used for the Young modulus (the Young modulus for bulk copper is typically about 120 GPa.). From fig. 10b shows that FEA simulations reproduce experimental values when using a Young modulus of 45 GPa.

4.4.5 Application: Fabrication of an XY flexure stage by electroforming of CAP-3DP molds

Fig. 11 shows a manufactured double compensated XY-stage through the CAP-3DP method with minor mechanical polishing of the top surface. The hinge thickness e was 150 μm , and the electroformed thickness of the XY flexure stage was 0.75 mm. Figure 4.11.c shows various positions of the X-Y stage under deformation (starting from the most left-bottom position and ending with the most right-top position). The deformations remain linear over the entire range. This final application illustrates how the proposed method can monolithically fabricate complex high-precision designs.

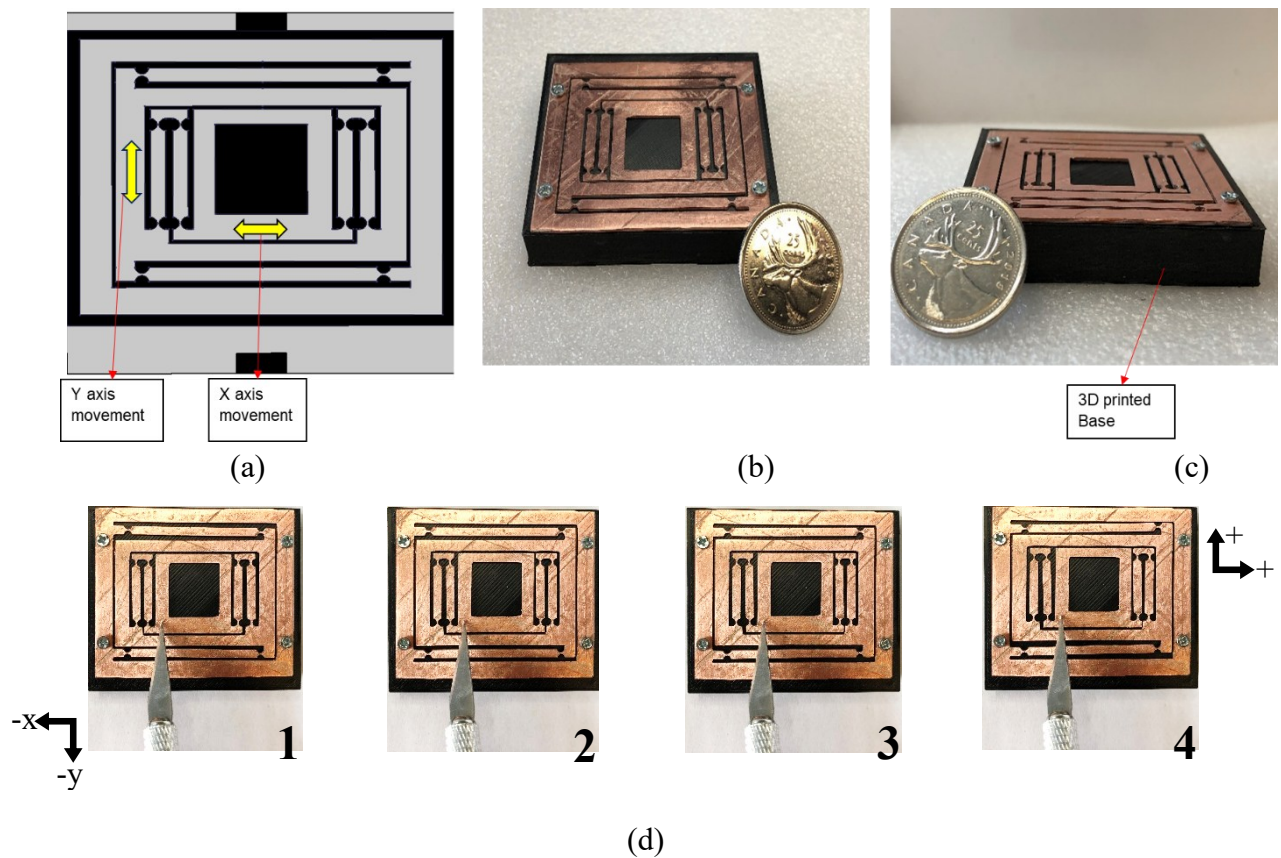


Figure 4.11 Electroformed X-Y double compensated flexure stage: a) CAD design; b,c) fabricated stage using CAP-3DP method and fixed on a 3D printed base; d) deformation of the electroformed XY flexure stage, from left-bottom position to the right-top position.

4.5 Cost analysis

This section investigates the manufacturing time and costs per part to fabricate personalized parts in the function of deployed 3D printers (number and investment cost of printers). In the studied scenario, it is assumed that the printing of the mold assemblies takes place during the time required to electroform a batch of parts. The aim is to provide a continuous workflow. The time is calculated as a function of the number of used 3D printers. Two different scenarios are considered to calculate manufacturing costs:

- deploying high-end 3D printers for manufacturing high-precision parts
- deploying low-price 3D printers for low-precision parts

In the final step, the cost per part is calculated as a function of the hourly rate of 3D printing.

Let t be the average printing time, in hours, per mold with one 3D printer (to simplify the analysis, an average build time t for all molds is used. A more detailed analysis would have to consider situations where not all molds require identical print times). The electroforming time is considered to be equal to 4 days, the time required for the X-Y stage of section 3.5. Therefore, there are four days available to print molds to complete a batch to be able to complete the process without interrupting the fabrication process. In the present scenario, we assume only 8 hours of 3D printing per day. The total number of molds, N_{3DP} , that can be printed in four days is given by:

$$N_{3DP} = \frac{32}{t}n \quad 4.2$$

where n is the number of 3D printers. Thus, 3D printing time (in days) per mold is:

$$T_{3DP} = \frac{t}{8n} \quad 4.3$$

These N_{3DP} printed molds will be electroformed in a single batch over four days. Therefore, the electroforming time (in days) per part is:

$$T_{EF} = \frac{t}{8n} \quad 4.4$$

The total fabrication time per part, in days, is:

$$T_{Total} = \frac{t}{4n} \quad 4.5$$

The printing cost per mold assembly is given by:

$$C_{3DP} = t \cdot CPH_{3DP} \quad 4.6$$

where CPH_{3DP} is the hourly rate of 3D printing. As electroforming happens over 24 hours per day, the cost of electroforming per mold assembly is:

$$C_{EF} = \frac{4 \times 24}{N_{3DP}} CPH_{EF} = \frac{3t}{n} CPH_{EF} \quad 4.7$$

where CPH_{EF} is the hourly rate of electroforming (which includes costs from the plating bath, energy consumption, electrodes, operation, and amortization). The total cost per part is then given by:

$$C_{Total} = t \left(CPH_{3DP} + \frac{3}{n} CPH_{EF} \right) \quad 4.8$$

The accuracy of 3D printing primarily relies on the deployed 3D printers. Two hourly rates of CPH_{3DP} for printing are considered. For a high-end 3D printer required for high-precision applications, a 200\$/h rate is used, and a 1\$/h rate for a hobbyist 3D printer for applications with low accuracy requirements. The electroforming cost rate, CPH_{EF} , is roughly estimated since it is

not straightforward to estimate precisely electroforming costs (such as labor, material, utility, and waste disposals) ²¹⁰. A value of 17\$/h for CPH_{EF} is estimated based on an hourly labor wage of 16\$ and initial equipment costs of 30,000\$ (which results in about 1\$/h for a five-years amortization period) as these costs account for the bulk (96%) of the total costs ²¹⁰. Note that this cost is probably an overestimation as it is not required to have an operator present during the full electroforming time ($4 \times 24 = 96$ hours) and further one operator can take care of several plating baths in parallel. Figure 4.12 and Figure 4.13 show the 3D printing, electroforming, and total time and cost per part for numerous 3D printers printing in parallel with an average printing time t of 2 hours per mold.

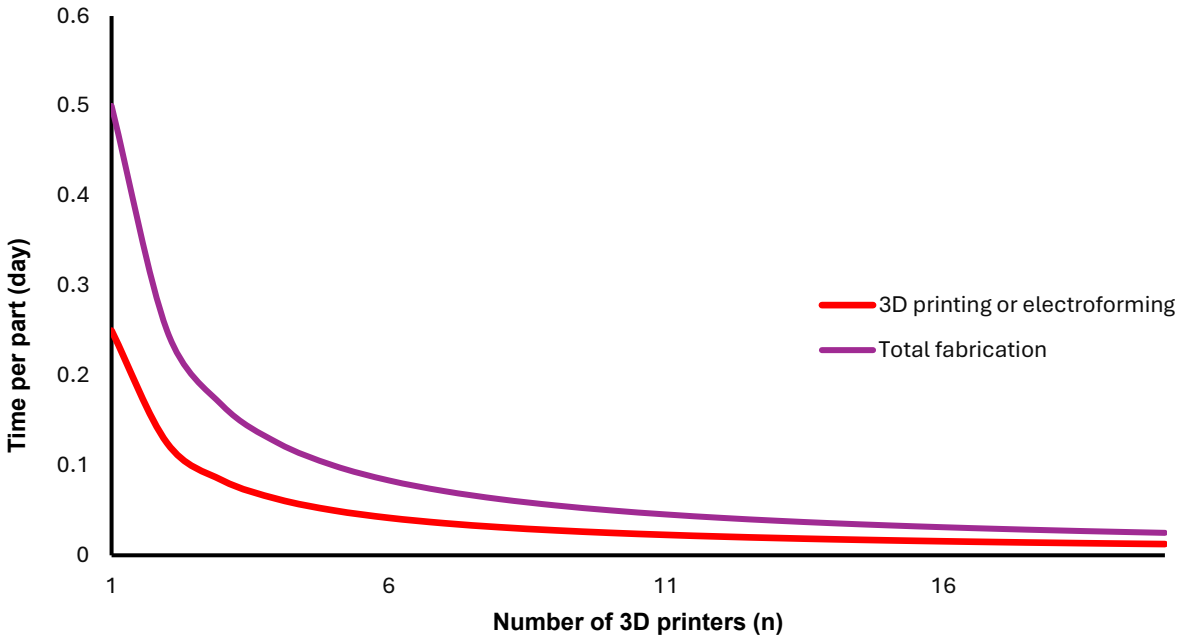


Figure 4.12 3D printing, electroforming, and total fabrication time (in days) per part vs. the number of employed 3D printers (The 3D printing and electroforming curves coincide since the same number of printed molds will be electroformed in a single batch within a set timeframe)

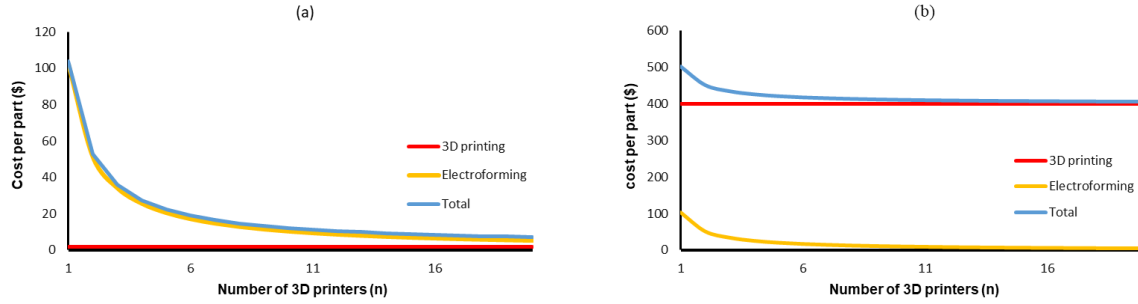


Figure 4.13 3D printing, electroforming, and total fabrication cost per part for two types of 3D printers a) Low-cost printer: the hourly rate of 3D printing is 1 \$/h, and b) High-end printer: the hourly rate of 3D printing is 200\$/h. A fixed electroforming cost hourly rate of 17\$/h is used for both scenarios.

Fig. 12 indicates that the total manufacturing time can be noticeably decreased (about 94%) when sixteen 3D printers are printing in parallel compared to a single 3D printing. Moreover, fig. 13 shows that the total cost per part highly depends on CPH_{3DP} , the hourly rate of printing, when high-end 3D printers are used. However, by increasing the number of employed 3D printers, the total cost per part drops significantly for both conditions. For low-cost 3D printers, the total manufacturing cost is essentially driven by the cost of electroforming. In contrast, for high-end printers (for ten printers and more), the cost per part is limited by the hourly rate of 3D printing.

A comparison between final manufacturing costs per part through the proposed scenarios shows that the manufacturing cost of personalized X-Y stages will be lower than 10\$ per part (about a 90% decrease compared to the single 3D printing) by deploying various low-end 3D printers and lower than 420\$ for high-end machines. The final cost per part for both scenarios is significantly lower than manufacturing the same personalized parts through available techniques such as Wire-EDM. Further, figures indicate that for the low-price 3D printer scenario, manufacturing costs would be most affected by the electroforming cost rate rather than the 3D printing cost rate.

4.6 Conclusion

Fabricating personalized parts causes extra manufacturing costs. This study demonstrated that by using a hybrid manufacturing process, combining 3D printing and electroforming, it is possible to obtain a low-cost mass fabrication method of personalized metallic precision micro parts. The main challenge to address is the process of electroforming inside a non-conductive 3D printed plastic mold. Therefore, three approaches were compared. Among the investigated methods, the direct 3D printing of molds on a conductive painted substrate revealed improved outcomes compared to the other two approaches: glued and hot-pressed assemblies.

Simple shapes, such as micro dimensional hinges (with hinge thicknesses of 140, 170, 200, 230, and 260 μm), and complex shapes, like a double compensated X-Y flexure stage (with a hinge thickness of 150 μm and part thickness of the of 0.75 mm) were manufactured as a demonstration. Results reveal the feasibility of manufacturing high-precision functional micro metallic parts.

Furthermore, it was shown how the mass fabrication costs and time per personalized metallic micro parts significantly decreased by deploying 3D printing and electroforming processes in parallel. As a personalized product example, the fabrication of a double-compensated X-Y stage through the proposed method was discussed. In this case, the total fabrication time per part is reduced from 12 hours when employing a single 3D printer (by assuming 3D printing time of 2h per mold with a single printer) to about 45 minutes when sixteen 3D printers are deployed during the time required for electroforming a batch of 3D printed molds. The total fabrication cost per part when using a single hobbyist 3D printer (assuming a 1\$ hourly rate for 3D printing and 17\$ hourly rate for electroforming) is 100\$ (mainly electroforming cost), which can be reduced to less than 10\$ when sixteen 3D printers are deployed. These numbers indicate that manufacturing time and cost per part are decreased by over 90% when more than sixteen low-cost 3D printers are printing in parallel while the former batch of 3D printed molds is being electroformed.

5. Electroforming process variability in mass personalized production

5.1 Introduction

In traditional industrial manufacturing, processes are designed for repeatability and stability. Parameters are standardized, equipment is finely tuned, and large production volumes ensure statistical consistency. However, as manufacturing transitions toward personalization, where each part can differ in design, geometry, or functionality, process control becomes increasingly complex. This shift introduces new challenges in ensuring reproducibility, particularly in electroforming, a process that is highly sensitive to the electrochemical environment.

Electroforming depends on maintaining equilibrium within the electrolyte bath, where minor variations in composition, temperature, or agitation can alter nucleation, grain growth, and surface morphology^{78, 103, 107}. Standardization and continuous monitoring easily mitigate such variations in mass production, where all parts are identical and processed under uniform conditions. In contrast, the production of personalized or small-batch parts involves continuously changing boundary conditions. This variability disrupts the steady-state assumptions upon which conventional electroforming control methods are based^{26, 27}. As a result, deposits formed under seemingly identical conditions may differ in morphology, uniformity, or mechanical properties, highlighting the inherent sensitivity of the process to subtle environmental changes.

5.2 Bath quality and its influence on electroforming behavior

The electrolyte bath serves as the central component of the electroforming process, governing ion transport, deposition rate, and overall deposit quality. The term "bath quality" encompasses several physical and chemical characteristics, including copper ion concentration, sulfuric acid ratio, conductivity, temperature, density, pH, viscosity, and impurity levels. Over time, these properties evolve due to metal ion depletion, additive consumption, pH drift, or contamination. This gradual transformation, commonly referred to as bath aging, directly affects current efficiency, nucleation kinetics, and deposit microstructure^{40, 122, 123}.

In large-scale industrial operations, bath aging is compensated through continuous monitoring and periodic regeneration. However, such procedures are impractical in mass personalized production, where intermittent use amplifies the effects of degradation. As the electrolyte composition changes, even slightly, its electrochemical response shifts, leading to variations in deposit thickness, surface roughness, and internal stress⁷⁸. The same set of process parameters can therefore yield different outcomes depending on the bath's instantaneous state.

This dependency on bath quality represents one of the key barriers to reproducible electroforming in a personalized manufacturing context. Understanding and compensating for this variability requires a deeper characterization of how the electrolyte evolves during repeated use and how those changes influence the electrochemical behavior of the system.

5.3 The challenge of personalization

The shift from standardized production to personalization alters the very nature of process control. In mass manufacturing, every part shares identical geometry, ensuring predictable current distribution, temperature fields, and hydrodynamic conditions. These consistencies allow for strict parameter control and highly stable outcomes.

In contrast, the manufacturing of personalized parts introduces variability in parts geometry, size, and surface characteristics. These variations influence the local electric field and current density distribution, which in turn affect the rate and uniformity of metal deposition. A part with sharp edges, deep cavities, or variable thicknesses can experience localized over-deposition or under-deposition even under globally identical process settings. The result is a system where geometric variability couples with bath variability, producing nonlinear and often unpredictable effects on the final deposit^{86, 33}.

Moreover, small-batch or one-off production means that process optimization through repetition is limited. In industrial production, feedback from thousands of identical parts allows fine-tuning of process parameters to minimize defects. In mass personalized electroforming, each new design requires renewed parameter tuning, and the same bath chemistry may behave differently from one component to the next. The interplay between these factors, bath aging, geometry-dependent current distribution, and limited repeatability, creates a highly dynamic environment that challenges conventional empirical optimization.

5.4 Understanding variability: A system-level perspective

The complexity of electroforming variability can be understood as the interaction of three evolving subsystems:

The Bath: A chemical environment that changes over time due to depletion, by-product accumulation, and pH shifts.

The Process: The electrochemical parameters (current density, waveform, agitation, and temperature) that drive deposition.

The Part: The geometric and surface features that determine how the local electric field and ion flux are distributed.

Each subsystem influences and is influenced by the others. Variations in bath composition alter the effective conductivity, which modifies the current distribution on the part. In turn, local differences in current density change ion consumption rates, further accelerating bath evolution. This dynamic coupling means that even small deviations can compound, leading to noticeable differences in deposition quality.

A key implication is that achieving reproducibility in mass personalized electroforming requires not only precise control of process parameters but also real-time awareness of the bath state. Without understanding the evolving condition of the electrolyte, parameter tuning becomes reactive rather than predictive. The lack of such predictive capability has historically limited the scalability of electroforming for individualized or small-batch production.

5.5 Motivation for a data-driven framework

Given the inherently dynamic and coupled nature of electroforming, traditional empirical approaches are insufficient to ensure consistent quality under personalized production conditions. Models based purely on static parameter optimization fail to capture the time-dependent and nonlinear relationships between bath state, process variables, and resulting deposit properties.

A data-driven framework offers a path forward. By systematically collecting and structuring experimental data across multiple baths and operating conditions, it becomes possible to uncover statistical correlations and latent relationships that describe process behavior. Such a framework allows for the identification of hidden variables (such as indicators of bath aging or instability) that are not directly observable through routine measurements²¹¹.

This approach aligns with the broader trend in smart manufacturing, where data acquisition, analysis, and prediction replace empirical trial-and-error. In the context of personalized electroforming, data-driven modeling can provide insight into how bath conditions evolve, how they affect deposition outcomes, and how process parameters can be adaptively adjusted to maintain quality despite variability.

5.6 Framework for addressing process variability

The need to address process variability motivates the research framework developed in the subsequent chapters of this thesis. These chapters are designed to progressively build understanding and capability from capturing variability, to characterizing it, to ultimately controlling it.

To illustrate the logical flow of this framework, Figure 5.1 presents the overall methodology linking data acquisition and predictive modeling. It highlights how systematic experimentation feeds into model-based prediction of process parameters, forming a closed learning loop for adaptive electroforming control.

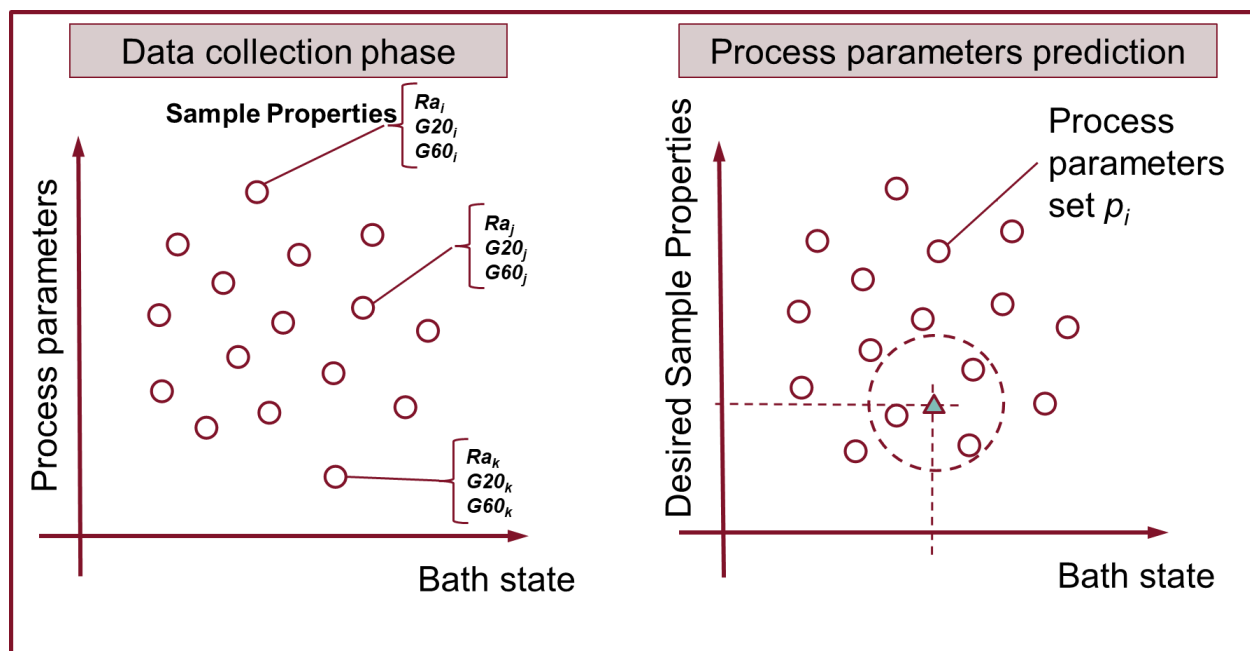


Figure 5.1 Schematic representation of the overall methodological framework. The left panel illustrates the data collection phase, where numerous experiments are performed under different bath and process conditions to populate the electroforming dataset. The right panel represents the process parameters prediction phase, where the accumulated data are analyzed to predict suitable process parameters for achieving target sample properties in given bath conditions, thereby enabling adaptive optimization in mass personalized electroforming.

This framework unfolds through three interdependent research directions that progressively address the challenge of electroforming variability:

Chapters 6 and 7 – Methodology and open dataset of electroforming:

These two chapters detail the systematic acquisition and structuring of a dataset encompassing a range of electroforming experiments under varying bath and process conditions. The dataset serves as a foundation for statistical analysis and machine learning, enabling reproducible and open research in this domain.

Chapter 8 – Search for bath state:

This chapter investigates the identification and quantification of bath-related indicators based on the constructed Open Dataset, capturing the evolving state of the electrolyte. It explores how

measurable parameters can represent the dynamic “state” of the bath and how these indicators correlate with deposition outcomes.

Chapter 9 – Search for process parameters:

Building upon the previous chapters, this section focuses on developing predictive and adaptive strategies for optimizing electroforming parameters. The objective is to establish relationships that enable reliable control despite bath aging, ultimately advancing the concept of adaptive, data-informed electroforming.

Together, these three research directions constitute an integrated methodology aimed at transforming electroforming from a process constrained by empirical uncertainty into a controllable, data-driven system suitable for low-cost, high-precision, and personalized production.

5.7 Conclusion

This chapter outlined the core problem motivating this research: the intrinsic variability of electroforming in personalized manufacturing contexts. Variations in bath composition, part geometry, and electrochemical conditions combine to create an unpredictable process environment that cannot be fully managed through traditional means.

Recognizing these challenges provides the rationale for adopting a systematic, data-driven approach. By decomposing the problem into three complementary objectives (dataset development, bath state identification, and process optimization), this thesis establishes a coherent path toward reproducible, adaptive control of electroforming.

In the following chapters, this framework is implemented and demonstrated, illustrating how quantitative analysis and data-centric modeling can transform electroforming into a predictable and scalable process for the era of personalized manufacturing.

6. Methodology

6.1 Introduction

This chapter outlines the integrated experimental and data-driven framework developed to investigate how electrolyte bath evolution influences copper electrodeposition and to build predictive models capable of estimating surface-quality outcomes and process parameters under varying bath conditions. The methodology was intentionally designed to generate a large, diverse, and high-quality dataset while maintaining strict experimental control, enabling both mechanistic interpretation and machine-learning analysis.

The experimental setup combines a precision three-electrode electrochemical system, custom ECTK control scripts, and a robotic linear-motion mechanism that introduces consistent and programmable mass-transport enhancement. This configuration ensures repeatable deposition conditions while allowing the process to be systematically perturbed across different electrolyte compositions, pulse-reverse parameters, and bath-aging states.

A second key component of the methodology is the structured acquisition of multi-modal data. Each electrodeposition trial produces (i) bath-state indicators such as specific gravity and electrical conductivity, (ii) time-resolved current waveforms that capture the dynamical behavior of the electrolyte during plating, and (iii) detailed post-deposition surface measurements including roughness parameters (R_a , R_q , R_z) and gloss values at multiple measurement angles. Together, these datasets provide a comprehensive view of how the bath evolves and how those changes translate into measurable differences in surface morphology and optical appearance.

The final component of the methodology involves constructing a machine-learning pipeline capable of learning relationships between process inputs, bath metrics, and resulting surface properties.

6.2 Experimental framework and execution steps

6.2.1 Sample and electrolyte solution preparation

This study investigates the electrodeposition of copper from an acidic electrolyte. In industrial applications, copper is typically deposited from a bath composed of sulfuric acid and a copper salt. In this work, copper sulfate (CuSO_4) was used as the metal ion source, and sulfuric acid (H_2SO_4) served as the supporting electrolyte. This electrolyte system is widely adopted due to its high conductivity, stability, and effectiveness in delivering uniform copper coatings.

A total of 360 copper bars, each pre-polished to a mirror finish and protected on the front surface with a plastic film (Kocour Co., USA), were sectioned to dimensions of 76.5 mm × 20 mm × 0.45 mm. These specimens were prepared for the electrodeposition process as outlined below:

- Removing the protective plastic film.
- Placing 13 mm × 19 mm label stickers.
- Coating the area around the labels and the back of the bars with insulating varnish to limit the exposed surface to approximately 245 mm² (label area).
- Leaving varnished samples at room temperature to allow the varnish to dry completely.
- Removing the labels.
- Rinsing the exposed area with isopropyl alcohol (IPA) and deionized water to ensure no adhesive residue remains.
- Cleaning for 10 minutes in a 10% v/v acetic acid in deionized water solution.
- Rinsing again with deionized water to remove any remaining acid.
- Drying with a stream of compressed air.

Following the completion of the electrodeposition tests, the workpieces were carefully and thoroughly rinsed with deionized water to ensure the complete removal of any residual acidic contaminants remaining on the surface. This rinsing step was critical to prevent interference with subsequent analyses. After rinsing, the samples were dried under a stream of compressed air. Once fully dried, the specimens were subjected to comprehensive surface characterization to evaluate the effects of the deposition process.

Four initial electrolyte baths were formulated by dissolving copper sulfate (CuSO₄) at concentrations of 0.1 M, 0.3 M, 0.5 M, and 0.7 M in a constant background of 1 M sulfuric acid (H₂SO₄). These specific concentrations were selected to cover a broad but practical range of copper ion availability commonly encountered in industrial and laboratory-scale electrodeposition processes. The lower limit of 0.1 M allows for the study of mass transport limitations at dilute conditions, while the upper limit of 0.7 M ensures sufficient ion availability for high-rate deposition without exceeding solubility limits. This range enables a comprehensive assessment of the effect of Cu²⁺ concentration on nucleation behavior, deposition rate, and film morphology under otherwise constant acidic conditions.

6.2.2 Electrodeposition process

Electrochemical deposition and bath aging experiments were carried out using a standard three-electrode setup controlled by a potentiostat (Model EC301, Stanford Research Systems). The system was operated via an in-house developed control software (EC301, ECTK), referenced from <https://sourceforge.net/projects/ectk/>, which enables advanced command execution, automated scripting, and real-time data acquisition. This setup allowed for precise manipulation of potential waveforms and ensured high reproducibility and flexibility in tailoring the deposition protocols for a range of experimental conditions.

Copper bars were employed as the working electrodes in all tests. A high-conductivity copper sheet, sourced from McMaster-Carr and machined to dimensions of 100 mm × 76.5 mm × 0.81 mm, was used as the counter electrode, providing a stable and uniform return path for current. A thin copper rod served as a pseudo-reference electrode and was positioned at a fixed distance of 2 mm from the working electrode to ensure reliable and repeatable potential measurements. The reference electrode dimensions (152 mm in length and 1.5 mm in diameter) were selected to maintain geometric consistency and minimize potential drift, thereby contributing to stable electrochemical conditions throughout the deposition process.

As illustrated in Figure 6.1, a dual-arm robotic system composed of two CNC Z-axis module supports (ANNOY TOOLS) was custom-engineered to facilitate mounting, demounting, and secure holding of the workpieces during the electrodeposition process. In addition to providing mechanical stability, the system enabled programmable and precise linear motion of the working electrode along the X-axis. This configuration introduced controlled reciprocating movement of the working electrode, enhancing mass transport and promoting a more uniform distribution of copper ions within the electrolyte. The reciprocating motion, maintained at a constant linear velocity relative to the counter electrode, improved ion replenishment at the electrode surface, reduced concentration gradients, and contributed to more homogeneous film growth. The robotic arm system was interfaced with a Raspberry Pi 3 Model B+ (1.4 GHz 64-bit quad-core processor, 1 GB RAM), which was programmed to execute and control the electrode motion throughout the experiments. The inter-electrode distance between the working and counter electrodes was fixed at 20 mm to ensure consistent current density and electrochemical environment.

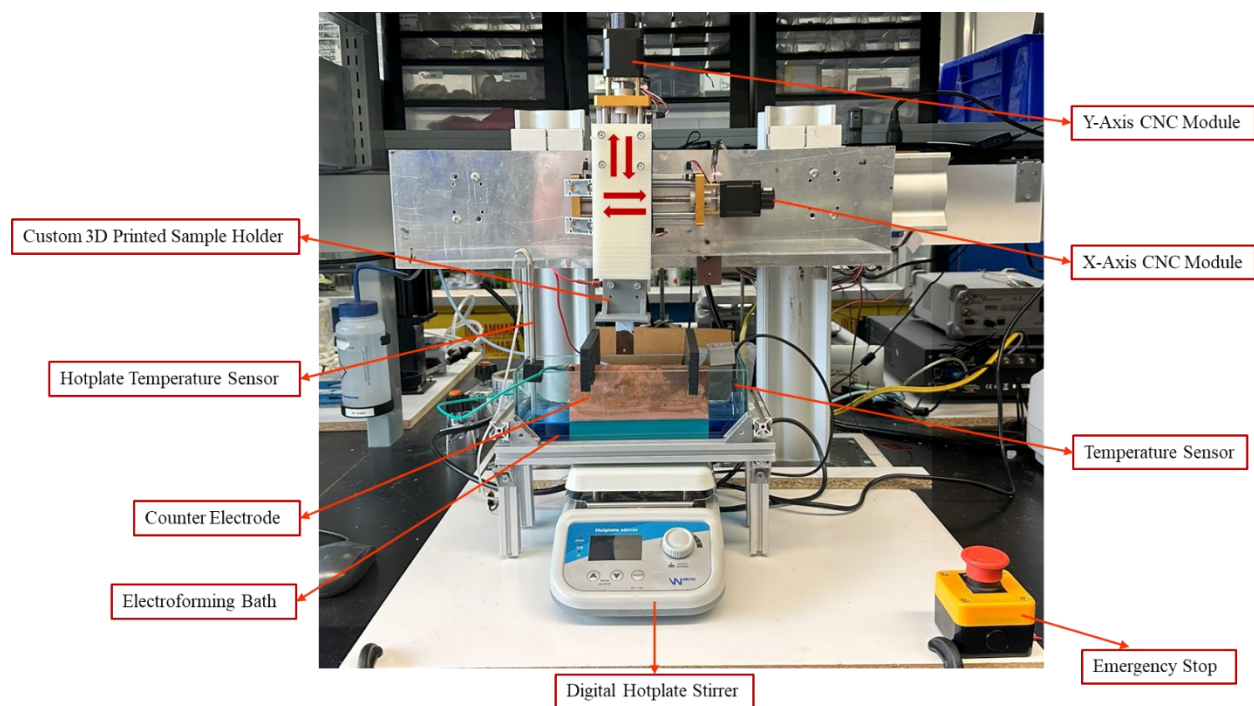


Figure 6.1 The dual-arm robotic system for electrodeposition tests

A custom electrode holder, fabricated from polylactic acid (PLA), was designed using computer-aided design software and manufactured via FDM on a Bambu Lab X1 Series 3D printer. This holder was engineered to securely house both the copper working electrode and the pseudo-reference electrode, ensuring precise and repeatable alignment within the electrochemical cell. PLA was selected for its mechanical stability, ease of prototyping, and chemical resistance under the operating conditions of the experiments.

The electrolyte bath was mounted on a digital hotplate-stirrer unit (Walter Products), equipped with an external temperature probe to maintain thermal stability. Throughout the experiments, the solution temperature was precisely regulated at 26 °C, while agitation was provided by a magnetic stir bar. This controlled stirring was essential to promote uniform ion distribution, enhance mass transport, and minimize concentration polarization near the electrode surface.

Electrodeposition was conducted using a pulse-reverse pulse plating technique implemented via custom ECTK scripts. The protocol involved alternating cathodic deposition pulses and anodic polishing pulses to regulate the surface morphology and structural properties of the copper films.

Temperature monitoring was conducted in parallel using a Robojax 100 cm waterproof DS18B20 stainless steel digital temperature sensor with a PCB adapter. The sensor was integrated into the same Raspberry Pi system used for robotic control and was programmed to continuously log temperature data in real time. This setup ensured precise thermal tracking throughout each

experiment and enabled correlation of thermal fluctuations with electrochemical performance, bath stability, and deposit quality. The sensitivity and measurement error of the temperature sensor are presented in Figure 6.2. This characterization was performed to assess the sensor's accuracy and reliability under the experimental conditions. The data confirm that the sensor operates within an acceptable error margin, ensuring precise temperature monitoring throughout the electrochemical processes.

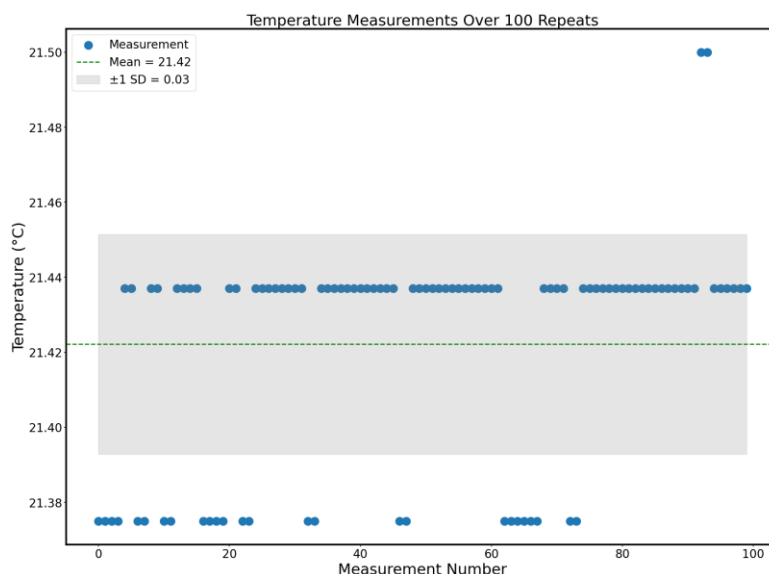


Figure 6.2 The sensitivity and measurement error of the temperature sensor over 100 repeats

6.2.3 Bath characterization

Specific gravity and electrical conductivity were selected as the primary indicators for assessing the electrolyte condition during copper electroplating in sulfuric acid due to their direct relationship with bath composition, ease of measurement, and strong sensitivity to ionic and concentration changes. The electrolyte, composed solely of CuSO_4 and H_2SO_4 without additives, maintains a highly acidic environment ($\text{pH} \approx 0$), eliminating the need to monitor pH as a variable. In such acidic media, pH remains constant throughout the process and therefore provides little diagnostic value regarding bath aging or compositional drift.

Electrical conductivity serves as a direct measure of the total ionic strength and mobility of charged species, primarily Cu^{2+} and SO_4^{2-} ions, within the electrolyte ²¹². Any significant change in conductivity can be attributed to depletion of copper ions through electrodeposition, accumulation of impurities, or evaporation-related concentration effects. Because it responds immediately to compositional or temperature fluctuations, conductivity is an effective and rapid indicator for

identifying changes in the electrochemical environment that might influence deposition quality or efficiency.

Specific gravity complements conductivity by reflecting the overall concentration of solutes in the bath ²¹³. As copper ions are reduced and plated onto the cathode, the density of the remaining solution changes accordingly. Tracking specific gravity allows for quantitative inference of bath concentration and helps detect gradual aging effects such as ion depletion, acid dilution, or buildup of nonconductive by-products. Together, conductivity and specific gravity provide a comprehensive, non-invasive monitoring approach that captures both the ionic and bulk concentration characteristics of the electrolyte.

Prior to each electrodeposition experiment, the specific gravity and electrical conductivity of the electrolyte bath were measured once the bath temperature stabilized at 26 °C. A handheld density meter from METTLER TOLEDO™ was employed to determine the specific gravity, providing a rapid and accurate assessment of bath density. Electrical conductivity measurements were conducted using a Thermo Scientific Orion benchtop conductivity meter, ensuring high-precision monitoring of ionic strength.

Additionally, the viscosity of the sulfuric acid-based bath was not included as a diagnostic parameter because it remained effectively constant throughout all experiments. Given the absence of organic additives and the stable operating temperature range, the electrolyte's viscosity exhibited negligible variation, rendering it an uninformative indicator of bath evolution.

6.2.4 Sample characterization

Weight measurement

Before and after each electrodeposition experiment, the samples were cleaned following the procedure detailed in Section 6.2.1 to ensure the removal of any surface contaminants or loosely adhered material. Subsequently, the samples were weighed using an OHAUS PIONEER® precision balance to verify the mass of the deposited copper. This gravimetric measurement served as a cross-validation of the deposition process, confirming that the deposited mass of approximately 0.019 g was consistent with the value predicted from the cumulative cathodic charge of 23.4 C/cm², as calculated using Faraday's law.

Surface roughness measurement

The surface roughness of the electrodeposited samples was evaluated following each experiment using a Mitutoyo Surftest SJ-210 Series-178, a contact-type portable surface roughness tester known for its high precision and reliability in field and laboratory settings. To ensure representative and statistically meaningful results, roughness measurements were performed at a minimum of 10 randomly selected points within the exposed deposition area of each sample.

The cut-off length for each measurement was carefully chosen based on the expected roughness scale, in accordance with profilometry best practices, to ensure the accurate capture of surface features and to avoid filtering artifacts.

Three standard roughness parameters (R_a , R_q , and R_z) were recorded for each measurement. According to the manufacturer's specifications²¹⁴:

- **R_a (Arithmetic Average Roughness)** represents the arithmetic mean of the absolute deviations of the surface profile from the central mean line over the measurement length.
- **R_q (Root Mean Square Roughness)** is defined as the square root of the mean of the squared deviations of the profile height from the mean line, providing a more sensitive indicator of peaks and valleys compared to R_a .
- **R_z (Maximum Height Roughness)** corresponds to the vertical distance between the highest peak and the deepest valley within a single sampling length, capturing the extreme topographical variations on the surface.

As illustrated in Figure 6.3, thicker electroplated layers require a longer deposition time. Based on this observation, a target thickness of approximately 8 μm was selected, corresponding to an average deposition duration of about 25 minutes per sample. Although lower thicknesses can be achieved more quickly, they introduce significant challenges in accurately measuring surface roughness and gloss, as the extremely thin coatings tend to reflect the initial topography of the copper substrate rather than the characteristics of the deposited layer. Conversely, plating thicker samples demands substantially longer deposition times, meaning that collecting even a small number of samples can take several months, which limits experimental throughput and repeatability.

Nevertheless, surface roughness was still considered an essential evaluation parameter because it directly reflects the microstructural evolution of the deposit and provides critical information about the uniformity, grain growth, and nucleation behavior during electroplating. Unlike gloss, which primarily indicates optical appearance, roughness reveals the underlying physical changes caused by bath composition, aging, and process parameters, making it essential for understanding and correlating surface morphology with plating quality.

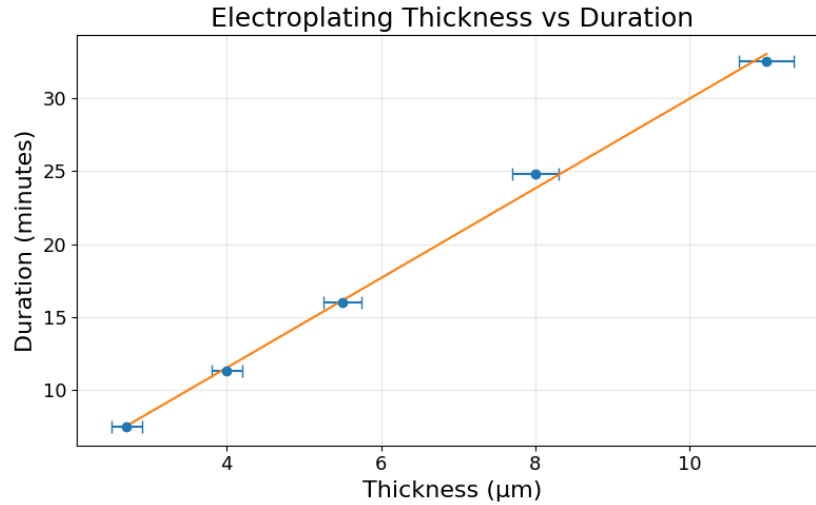
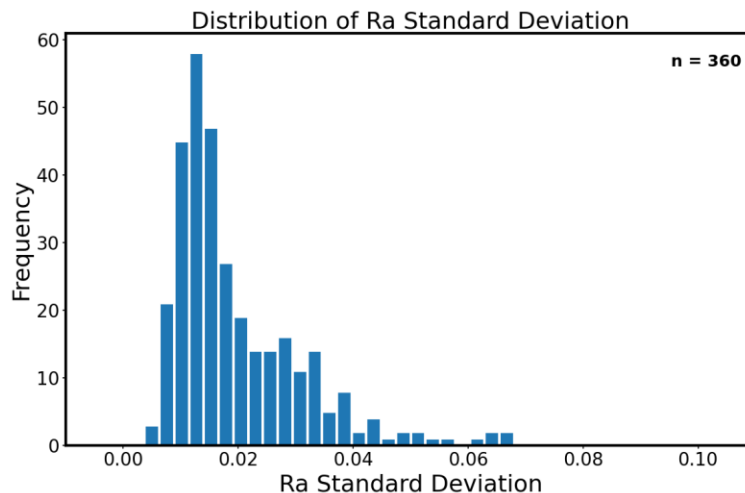


Figure 6.3 Comparing the total duration of electroplating when the plating thickness increases

Because the electroplated layer was thin (approximately $8\ \mu\text{m}$), the roughness measurements were performed using a fixed stylus force and displacement, as controlled by the instrument settings, to ensure a constant probe–surface distance. This approach minimized the applied contact pressure and prevented mechanical damage or partial removal of the copper layer, which would otherwise lead to erroneous roughness values. After each measurement, the scanned surface area was inspected under an optical microscope to verify the absence of surface damage. Any measurements associated with detectable damage to the plated surface were excluded from the analysis.

The standard deviation of surface roughness parameters measured after electrodeposition across 360 samples is presented below (Figure 6.4). This statistical analysis provides insight into the variability and consistency of the surface morphology under the applied deposition conditions. Evaluating the standard deviation of key roughness metrics (R_a , R_q , and R_z) helps assess the reproducibility of the process and identify potential sources of non-uniformity across samples.



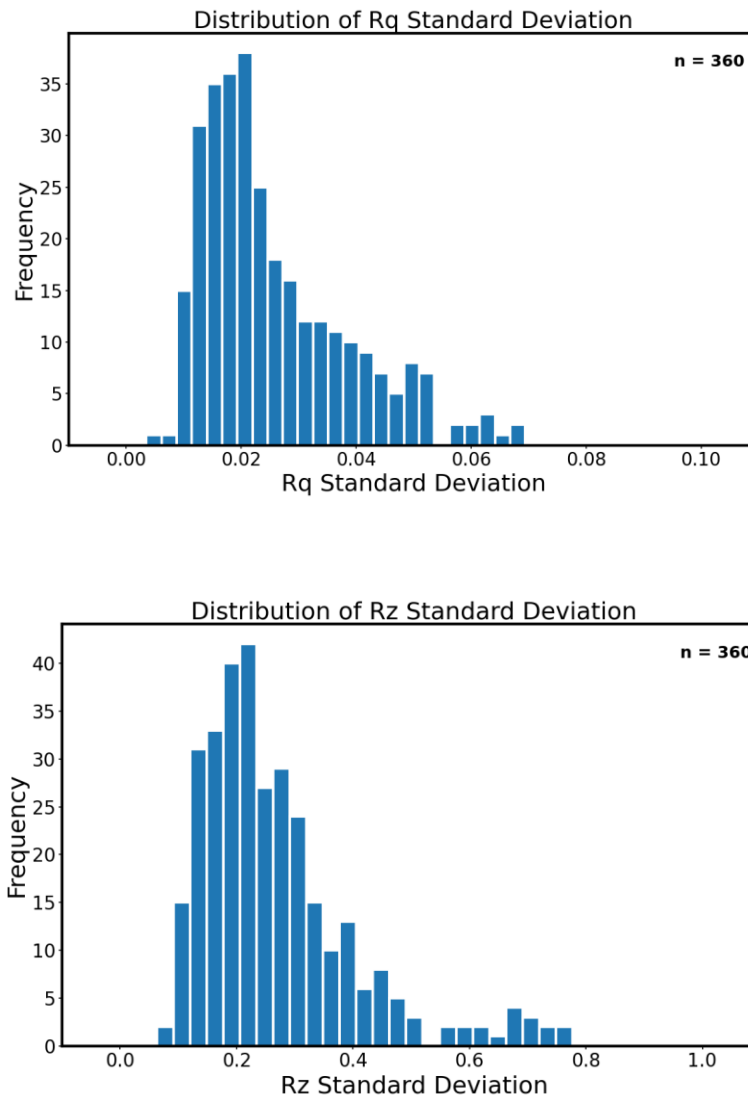


Figure 6.4 The standard deviation of surface roughness parameters measured after electrodeposition tests (360 samples)

Surface gloss measurements

In this study, a Miza Trigloss Meter was employed to evaluate the gloss level of the copper bars following the electrodeposition process. The Trigloss Meter operates on the principle of specular reflectance, a fundamental concept in optics that describes how light reflects from smooth surfaces. When a beam of light strikes a surface, it can either scatter in multiple directions (diffuse reflection) or reflect uniformly at an angle equal to but opposite to the angle of incidence (specular reflection).

reflection)(Figure 6.5). Gloss meters are designed specifically to measure this specularly reflected light, which is highly sensitive to the smoothness and microstructure of the surface.

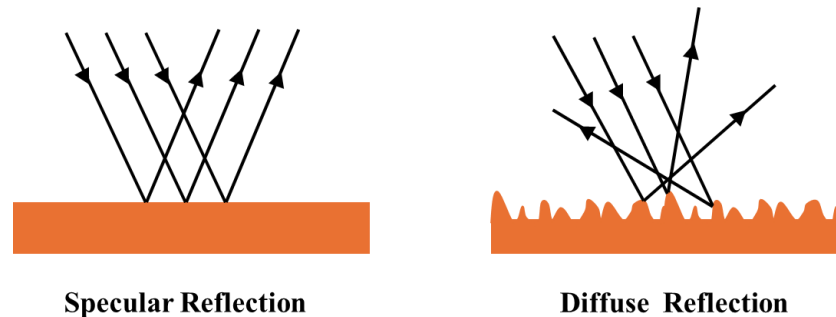


Figure 6.5 Specular vs diffuse reflectance

At the core of the Trigloss Meter's functionality is a calibrated light source, typically an LED or tungsten lamp, that emits a beam at a fixed angle (20° , 60° , or 85° , depending on the selected geometry). The beam is directed onto the test surface through a precision aperture to ensure uniform intensity and directionality. The surface reflects a portion of the incident light, and a photodetector, positioned at the mirror angle (equal to the angle of incidence but on the opposite side), captures the intensity of the reflected light (Figure 6.6). The gloss value is calculated by comparing the detected reflected light intensity against a reference standard, typically a highly polished black glass with a known refractive index (generally $n = 1.567$). At a 60° measurement angle, this reference is defined to have a gloss value of 100 gloss units (GU). Surfaces reflecting more light than this standard can record values greater than 100 GU, indicating extremely high gloss (e.g., polished ceramics).

Each measurement angle in the Trigloss Meter serves a specific purpose:

- **20°** is used for high-gloss surfaces, offering increased sensitivity to small variations such as micro-defects or polishing inconsistencies.
- **60°** is a general-purpose geometry for assessing medium-gloss finishes and is widely used across industrial applications.
- **85°** is reserved for low-gloss or matte surfaces, where minimal specular reflection occurs.

Internally, the gloss meter includes systems for calibration, digital signal processing, and temperature compensation, ensuring accurate and stable readings.

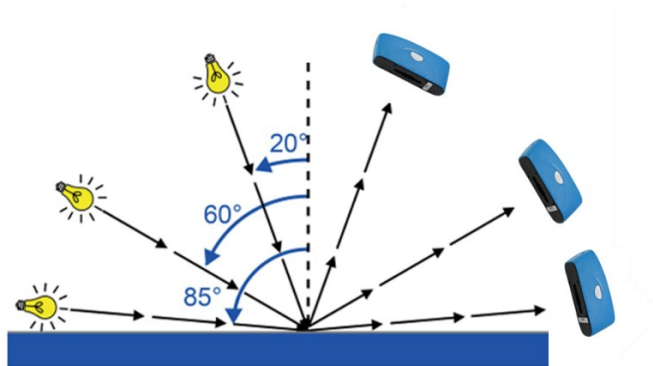


Figure 6.6 Surface gloss measurement angles

Given that the copper surfaces ranged from medium to high gloss after treatment, measurements were primarily conducted using the 20° (G20) and 60° (G60) geometries, which are most appropriate for detecting subtle variations and surface enhancement at these gloss levels. As Table 6.1 shows, the use of these two angles allowed for a detailed assessment of surface reflectivity and uniformity, providing critical insight into the effectiveness of the electrodeposition and polishing parameters. Figure 6.7 also shows three examples of samples located in three main ranges of gloss values.

Measurement Angle	Gloss Range	Typical Use
20°	High gloss	Polished metals, coatings
60°	Medium gloss	General-purpose surfaces
85°	Low gloss	Matte finishes, textured surfaces

Table 6.1 Gloss measurement angles and their application ranges

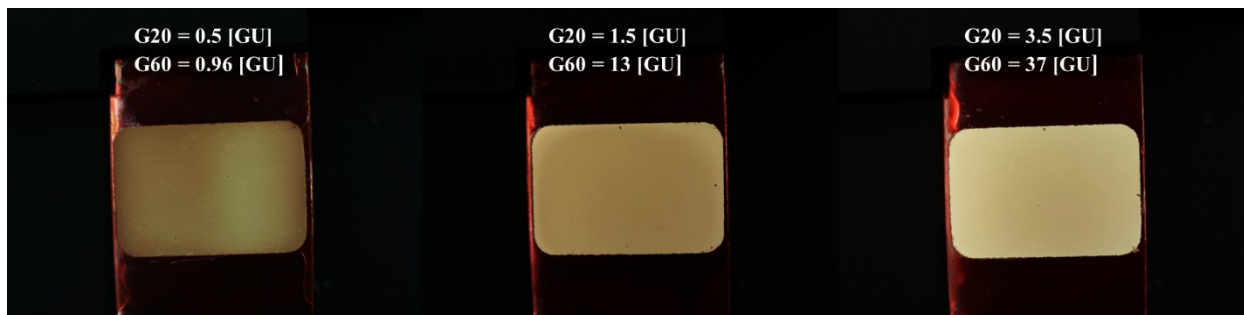
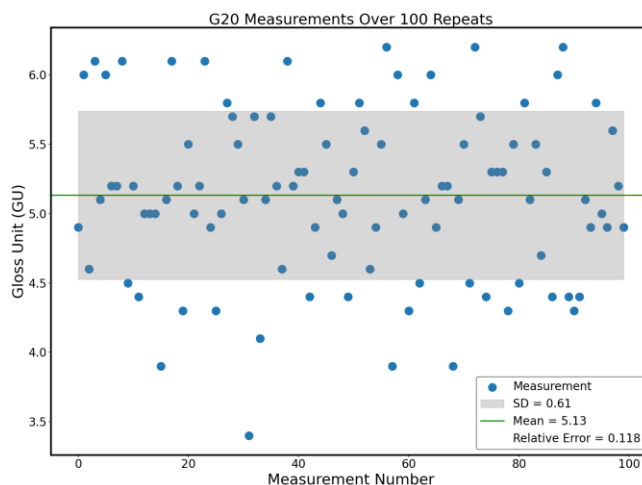


Figure 6.7 The examples of surface gloss for different ranges (low, medium, high)

The accuracy of gloss measurements at 20° and 60° geometries was evaluated by performing 100 repeated measurements on a representative electrodeposited copper surface for each angle (Figure 6.8). This repeatability test allowed for the assessment of measurement precision and instrument stability under consistent conditions. The resulting data provided statistical validation of the gloss meter's performance and confirmed the reliability of the readings obtained at these two angles, which were primarily used due to the medium-to-high gloss nature of the electrodeposited copper surfaces.



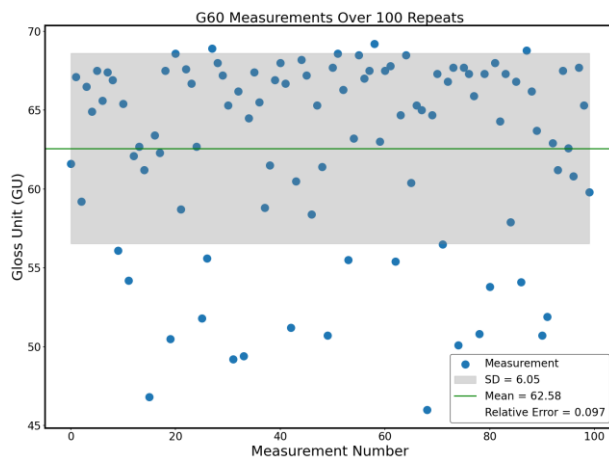


Figure 6.8 Measurements on a representative electrodeposited copper surface for 20° (G20) and 60° (G60) angles

6.3 Data-driven model formulation

The development of the machine learning model in this study followed a multi-step workflow, illustrated in Figure 6.9, which integrates experimental data processing, statistical analysis, and algorithmic modeling. The foundation of the model is a dataset compiled from electrodeposition experiments, where each data entry reflects a distinct combination of deposition parameters and electrolyte conditions. In addition to final surface characteristics, the dataset includes time-resolved process data, specifically current intensity, which was continuously monitored and recorded using the ECTK software.

To manage this data, an open-source environment, Jupyter Notebook, was used as the main interface for analysis, enabling the combination of live code execution, data visualization, and annotation in a single platform. Raw input files were read and organized into tabular form using the Pandas library²¹⁵, which provided structured access through its DataFrame and Series formats. These formats made it possible to clean the data, fill in missing entries, normalize scales, and convert process logs into a format suitable for statistical modeling.

Rather than directly using all collected variables, a refined feature selection step was applied to extract the most informative inputs. This was achieved through both automated selection algorithms and expert-driven filtering based on knowledge of the electrochemical system, as discussed in Chapter 7.

Following this, a series of regression models were built using the Scikit-Learn package²¹⁶. Multiple algorithms were tested and evaluated to determine which produced the most reliable and interpretable outcomes. To assess model performance and generalization, predictions were

validated against new electrodeposition trials performed in fresh bath solutions with varying preparation and aging profiles not included in the original dataset.

Throughout this process, additional open-source Python libraries were essential. NumPy ²¹⁷ supported vectorized operations and numerical calculations, while Matplotlib ²¹⁸ was used to generate comparative plots, visualize trends, and communicate the structure of the data and results. These tools enabled a comprehensive analysis pipeline that transformed raw sensor data into a predictive machine-learning framework. Full details of model architecture, training, and evaluation are presented in Chapter 8.

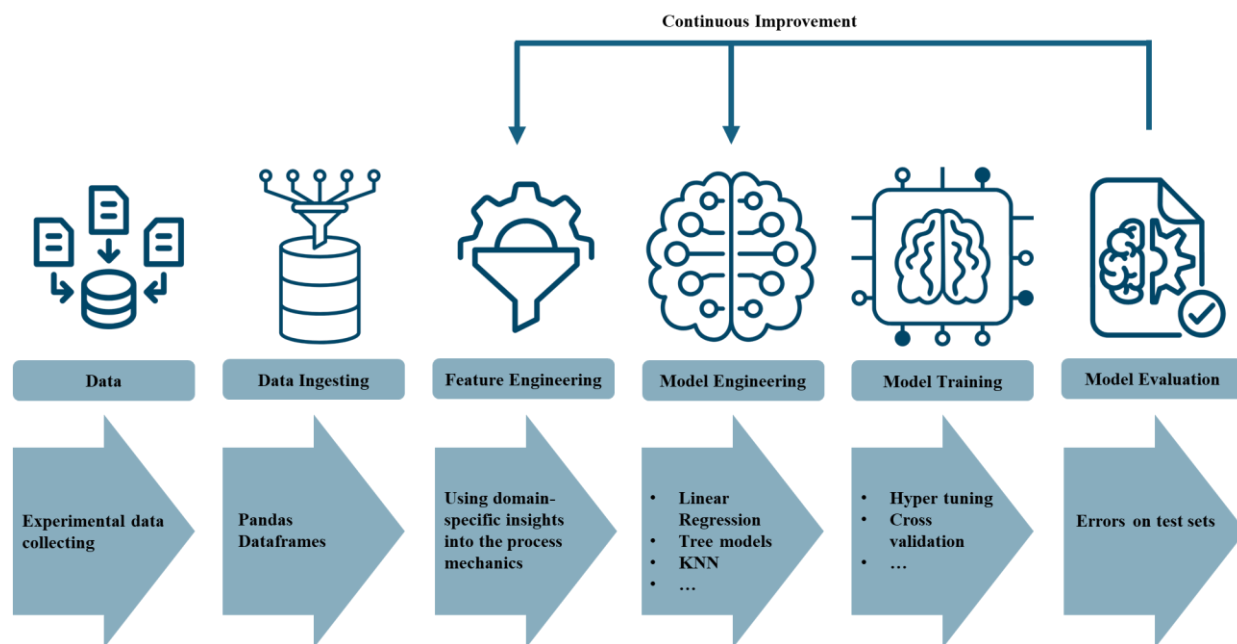


Figure 6.9 Data-driven model formulation steps

7. Open dataset for electroplating

7.1 Introduction

As manufacturing evolves toward mass personalization, the traditional paradigms of process standardization and repeatability are being challenged. Electroforming, long valued for its precision and ability to reproduce fine details, holds great potential for producing personalized metallic components at microscale and mesoscale dimensions. However, when the process is adapted for low-volume or one-of-a-kind production, its inherent sensitivity to bath composition, electrode properties, and electrochemical stability becomes a major obstacle to consistency. Unlike mass production, where conditions are carefully standardized and continuously monitored, personalized manufacturing involves parts with different surface characteristics, smaller bath volumes, and frequent contaminations and interruptions. These variations amplify the influence of bath aging and local field fluctuations, resulting in significant process variability and loss of predictability in deposit quality.

In traditional mass-production environments, process reliability is often sustained through the accumulated experience of skilled operators. Their intuitive adjustments, such as compensating for small deviations in current density, temperature, or bath composition, allow stable operation despite gradual process drift. However, this human-centered control strategy becomes ineffective in the context of mass personalization. Each new part alters the electrochemical environment, rendering experience less transferable and operator intuition less reliable. To achieve reproducible quality across personalized products, control must therefore shift from subjective expertise to objective, data-driven decision-making supported by systematic monitoring and predictive modeling.

To overcome these limitations, a deeper, data-centric understanding of electroforming behavior is essential. Empirical tuning of parameters is no longer sufficient when each new design introduces a different set of electrochemical boundary conditions. What is needed instead is a quantitative framework that captures the relationships between bath state, process parameters, and deposition outcomes, enabling adaptive and predictive control across personalized production scenarios. The creation of a structured and openly accessible electroplating dataset represents the first concrete step toward this objective.

This open dataset systematically compiles experimental data from a broad range of electroplating conditions designed to simulate the diversity encountered in personalized manufacturing. It provides a unified foundation for analyzing how process inputs, such as pulse parameters and agitation, interact with bath characteristics, including CuSO_4 concentration, pH, conductivity, and specific gravity. By capturing both the physicochemical process conditions and the resulting deposit properties, the dataset enables quantitative investigation into reproducibility, aging effects, and bath-state dynamics. Such a resource is critical for developing machine-learning models capable of predicting surface properties under unseen or evolving process conditions and tuning process parameters based on desired surface morphologies.

Beyond its analytical value, the dataset addresses a key practical limitation in mass personalization: the lack of transferable process knowledge. In conventional electroforming, process optimization is highly context-dependent and often lost when conditions or parts change. The structured dataset presented here transforms this empirical knowledge into a form that can be shared, reused, and generalized across different setups and part configurations. It thus contributes to the long-term goal of making electroforming a data-driven, adaptive, and scalable process compatible with the principles of Industry 4.0 and smart manufacturing.

The organization of this chapter reflects this dual objective of documentation and analysis. Section 2 provides a general summary of the dataset and its hierarchical structure. Section 3 outlines the electroplating process parameters used during data collection, while Section 4 describes the measured and derived variables grouped into deposition, pulse, and bath categories. Section 5 examines the statistical distribution of measured quantities to characterize dataset diversity, followed by Section 6, which explores correlations between process parameters and surface properties. Sections 7 and 8 focus on the temporal evolution of the electrolyte bath and the comparative effects of fresh and aged states on deposition outcomes under identical conditions. Section 9 evaluates how counter-electrode conditions influence reproducibility and deposition efficiency.

Together, these sections establish the open electroplating dataset as both a scientific contribution and a strategic enabler for mass personalization. It bridges empirical experimentation and intelligent process modeling, forming the empirical basis upon which adaptive and predictive electroforming systems can be built, an essential step toward reproducible, low-cost, and personalized manufacturing.

7.2 Dataset summary

Before getting into the detailed analysis and parameter descriptions, this section provides an overview of the Open Electroplating Dataset to help the reader understand its scope and structure. The dataset was designed to capture the variability of electroforming across a wide range of physicochemical bath conditions representative of personalized manufacturing. It serves as the experimental foundation for all subsequent correlation and modeling analyses presented in this study.

Average number of measurements conducted for dataset construction	
Number of investigated baths	114
Total number of conductivity/specific gravity measurements	228
Typical number of samples electroplated per solution	3-5
Total number of samples	360
Total number of roughness/gloss measurements	5760

Table 7.1 A summary of the open electroplating dataset

As Table 7.1 indicates, a total of 114 baths with distinct physicochemical properties were prepared and characterized prior to electroplating. For each bath, conductivity and specific gravity were measured to record its initial state. In each bath, an average of three to five samples were electroplated under identical process parameters to evaluate reproducibility. This procedure produced a total of 360 electroplated samples. Following deposition, the surface roughness and gloss of every sample were measured, resulting in approximately 5760 individual measurements across all baths.

Together, these data provide a comprehensive foundation for understanding how bath composition and aging affect electroplating reproducibility under fixed process conditions. The next sections describe in detail the experimental parameters, measurement variables, and statistical distributions that form the analytical basis of the open dataset.

7.3 Electroplating process parameters during dataset construction

As discussed earlier in Methodology (Chapter 6), all electroplating experiments in this study were performed using a pulse–reverse current method, which alternates between cathodic (deposition or -) and anodic (polishing or +) phases within each cycle. This approach was selected for its ability to produce smoother, denser, and more uniform deposits compared with conventional direct-current or unidirectional pulse plating. During the cathodic (-) phase, copper ions are reduced and deposited onto the substrate surface. In the subsequent anodic (+) phase, a controlled reverse current partially dissolves surface asperities and reduces nodular growth, thereby improving smoothness and reflectivity. This cyclic process enhances surface leveling and grain refinement while mitigating concentration polarization and local ion depletion. Such precise control of growth and dissolution dynamics makes the pulse–reverse pulse method particularly suitable for generating reproducible data across multiple electrolyte conditions.

The total transferred charge per experiment was maintained constant at 23.4 C/cm², ensuring comparable deposit thickness among all samples. The number of cycles was adjusted according to the selected pulse durations and frequencies, preserving the same overall charge input. By defining

both the deposition charge (Q_{dep}) and polishing charge (Q_{pol}) within each pulse cycle, the process could be fine-tuned to study how different cathodic–anodic ratios influenced surface morphology and gloss. This method provided a consistent experimental framework capable of isolating the effects of bath composition and aging from process-related variability. The full range of parameters used during dataset construction is summarized in Table 7.2, which defines the operational space applied throughout all 114 baths.

Parameters	Values
Deposition Charge (Q_{dep})	-0.1 C/cm ² to -26 C/cm ²
Deposition Potential (E_{dep})	-110 mV to -800 mV
Polishing Charge (Q_{pol})	0-90% of Deposition Charge
Polishing Potential (E_{pol})	100 mV
Agitation	Magnet stirring fixed on 400 rpm
pH	≈ 0
Reciprocating movement of the sample	Fixed on 4 (mm/s) in the range of ± 30 mm
Distance between sample and counter electrode	Fixed on 2 cm
Number of cycles for each experiment	Calculated based on total cumulative charge of 23.4 C/cm ²

Table 7.2 Electroplating process parameters used during the construction of the open dataset.

The Q_{dep} ranged from -0.1 C/cm^2 to -26 C/cm^2 , representing variations in cathodic energy input rather than differences in deposit thickness. This range was selected to evaluate how the rate of charge transfer and local current density affect nucleation kinetics and surface appearance under similar total electroplated thicknesses. The deposition potential (E_{dep}) varied between -110 mV and -800 mV , corresponding to the potential region where copper reduction dominates, and hydrogen evolution remains minimal. Together, these parameters govern the cathodic portion of each pulse cycle and control the growth rate, surface morphology, and overall energy balance of the process.

During the reverse phase, the Q_{pol} was varied from 0 % to 90 % of the deposition charge, while the polishing potential (E_{pol}) was maintained at 100 mV. Adjusting the anodic-to-cathodic ratio allowed systematic control over surface smoothing and brightness. This configuration provided a controlled way to analyze how partial anodic dissolution affects gloss and roughness parameters, forming the basis for later correlation studies.

Hydrodynamic and geometric parameters were standardized throughout all experiments to minimize uncontrolled variability. Magnetic stirring was maintained at 400 rpm to ensure

homogeneous ion distribution and to prevent localized depletion zones. The pH of the electrolyte remained close to zero due to the high sulfuric-acid concentration (1 M), providing a stable, highly conductive medium dominated by Cu^{2+} and SO_4^{2-} ions. The distance between cathode and anode was fixed at 2 cm, maintaining a uniform electric field, while the sample reciprocating movement (held by the dual-arm robotic system) speed was kept constant at 4 mm/s (within a ± 30 mm working range) to provide mild convection without excessive turbulence. These fixed geometric and hydrodynamic conditions ensured that variations observed across samples could be attributed primarily to changes in bath composition and electrochemical aging rather than external disturbances.

Overall, the pulse–reverse pulse method offered precise control over deposition and dissolution dynamics, enabling the generation of high-quality, reproducible data suitable for quantitative analysis. The combination of well-defined pulse parameters, controlled hydrodynamics, and standardized geometry provided a reliable framework for comparing electroforming behavior across 114 distinct baths. The following section presents the measured properties and experimental variables that describe the relationship between electrochemical conditions, bath characteristics, and resulting surface properties.

7.4 Measured properties and experimental variables

7.4.1 Deposition characterization

The organization of the measurements within the dataset was designed to ensure both consistency and statistical reliability. The detailed procedures for measuring gloss and surface roughness, including the instruments and test configurations, have already been described in the Methodology chapter. The present section, therefore, focuses on how these measurements were structured, processed, and stored for dataset construction. The overall layout of the measurement workflow is illustrated in Figure 7.1.

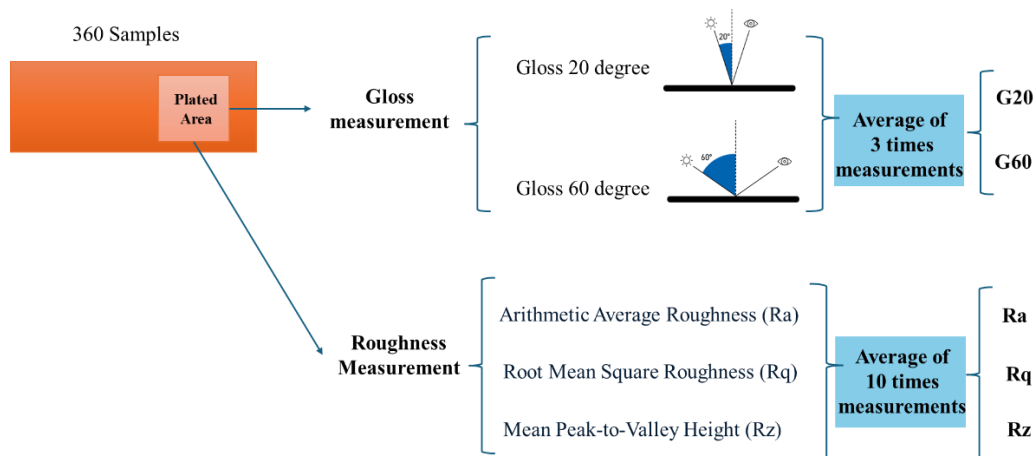


Figure 7.1 Schematic representation of the electroplated area and the associated measurement protocol.

Each copper sample contained a defined electroplated area, produced using the pulse–reverse process. All measurements were performed exclusively within this plated region to ensure that the data represented only the deposited copper layer, excluding any influence from the substrate or uncoated regions. The outer edges were systematically avoided to eliminate variations associated with non-uniform current distribution and to ensure uniform comparison among all samples.

For each electrolyte bath, between three and five samples were electroplated under identical process parameters. On every sample, surface gloss and roughness were evaluated following the same measurement sequence. The surface gloss was measured three times on the plated area, and the mean value of these readings was recorded as the representative GU for that sample. These values quantify the reflective quality of the electroplated surface, allowing direct comparison of surface brightness among different baths.

The surface roughness was measured ten times on the same plated area to assess repeatability and local variability. From these scans, three statistical descriptors were extracted: R_a , R_q , and R_z , corresponding respectively to the arithmetic average height, the root mean square height, and the mean peak-to-valley distance across the measured profile. The mean value of each parameter was stored in the dataset as the characteristic roughness of the sample, while the standard deviation of the ten scans was also retained as an indicator of within-sample variation.

This structured and repetitive measurement protocol ensured high confidence in the reproducibility and comparability of the results. Across the 360 electroplated samples, these repeated measurements produced 5760 individual data points that were consolidated into averaged and variance-based values for each feature.

7.4.2 Pulse characterization

As mentioned earlier, in pulse–reverse electroplating, each cycle consists of two sequential phases: a cathodic deposition pulse in which copper ions are reduced onto the surface, followed by an anodic polishing pulse that partially dissolves surface irregularities. The duration of each phase was automatically determined by the total target charge of that phase, corresponding respectively to Q_{dep} and Q_{pol} . The system alternated continuously between these two stages until the cumulative transferred charge reached 23.4 C/cm^2 for each sample. This approach provided a consistent overall energy input while allowing the current and time responses to vary with process conditions.

For every sample, the current–time response of each individual pulse was recorded and analyzed. From these curves (Figure 7.2), two quantities were extracted for every pulse:

1. The average current (i_{dep_pulse} and i_{pol_pulse}) is calculated over the active duration of the pulse for each phase.
2. The total pulse time (t_{dep_pulse} and t_{pol_pulse}) represents the full duration required to reach the specified charge.

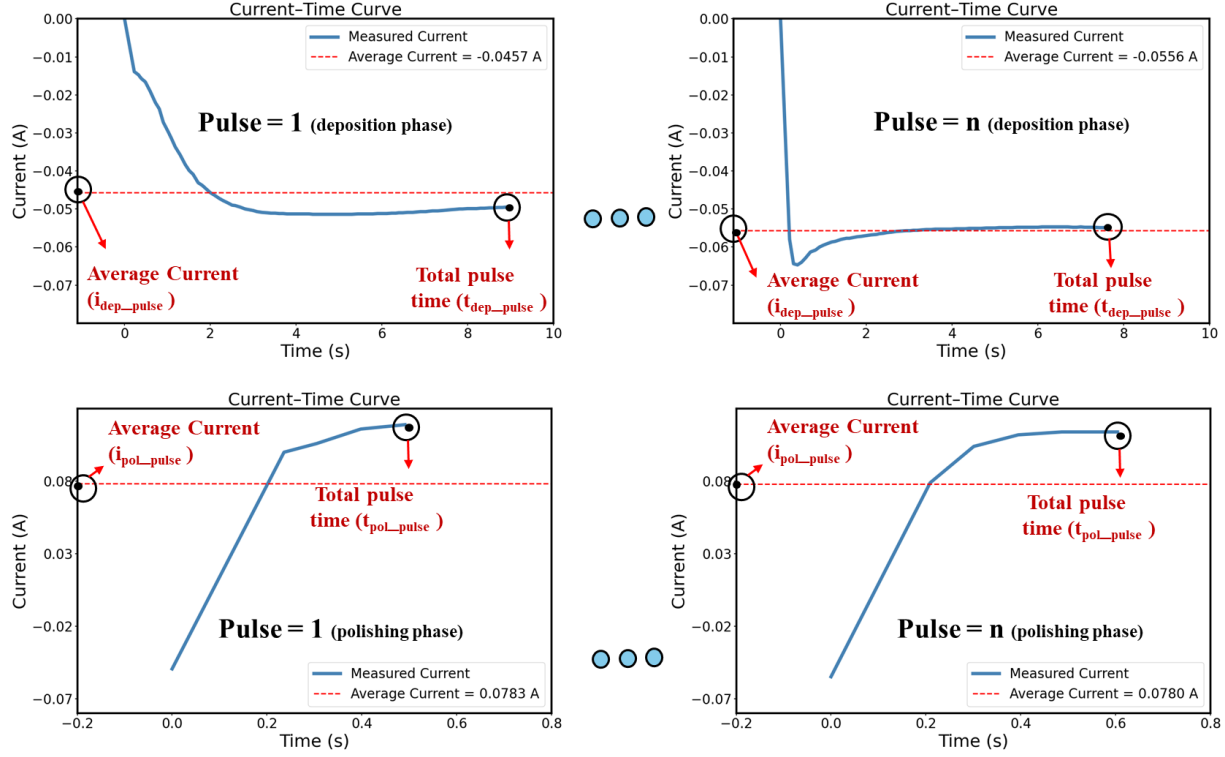


Figure 7.2 Characterization of pulses (deposition and polishing phases) and statistical parameters derived from current-time data.

Because the number of pulses differed among samples (depending on the ratio of deposition to polishing charges), the resulting data for each sample contained a variable-length series of $i_{\text{dep_pulse}}$ (or $i_{\text{pol_pulse}}$) and $t_{\text{dep_pulse}}$ (or $t_{\text{pol_pulse}}$) values. To ensure consistency across the dataset, these pulse-wise data were summarized into six statistical descriptors:

- Minimum, average, and maximum of the mean current values ($\min_i_{\text{dep_pulse}}$, $\text{avg_}i_{\text{dep_pulse}}$, $\max_i_{\text{dep_pulse}}$, $\min_i_{\text{pol_pulse}}$, $\text{avg_}i_{\text{pol_pulse}}$, $\max_i_{\text{pol_pulse}}$); and
- Minimum, average, and maximum of the corresponding pulse durations ($\min_t_{\text{dep_pulse}}$, $\text{avg_}t_{\text{dep_pulse}}$, $\max_t_{\text{dep_pulse}}$, $\min_t_{\text{pol_pulse}}$, $\text{avg_}t_{\text{pol_pulse}}$, $\max_t_{\text{pol_pulse}}$).

These features were calculated separately for the deposition and polishing pulses and, when relevant, combined to generate equivalent statistics for the total pulse period (t_{pulse}) as:

- $\min_t_{\text{pulse}} = \min_t_{\text{dep_pulse}} + \min_t_{\text{pol_pulse}}$
- $\text{avg_}t_{\text{pulse}} = \text{avg_}t_{\text{dep_pulse}} + \text{avg_}t_{\text{pol_pulse}}$
- $\max_t_{\text{pulse}} = \max_t_{\text{dep_pulse}} + \max_t_{\text{pol_pulse}}$

All current values were expressed in amperes (A) and pulse durations in seconds (s).

The resulting variables capture the essential characteristics of the electrochemical waveform while providing a compact description of the temporal and energetic behavior of each plating condition. Each pulse represents a distinct electrochemical event, where the average current indicates the effective rate of charge transfer and the pulse duration reflects the kinetic and mass-transport constraints of the system. Variations in these quantities across successive pulses naturally arise from differences in bath conductivity, ionic concentration, and electrolyte aging. As the bath evolves, achieving the same target charge requires longer pulse durations or lower average currents, and these subtle variations collectively encode the electrochemical state of the system.

By summarizing these pulse responses through their minimum, average, and maximum values, the dataset transforms variable-length time-series data into a uniform and interpretable form. This statistical representation preserves the dominant features governing the deposition–polishing dynamics while eliminating signal noise and inconsistencies associated with pulse count or sampling rate. The resulting descriptors (average current, pulse duration, and their variability) thus provide a meaningful link between the electrochemical process input and the resulting deposit characteristics. The dataset encapsulates the key information of each transient waveform in a fixed-size structure, optimized for machine-learning applications, rather than storing every waveform in its entirety. This allows for predictive modeling of surface quality outcomes and supports correlation analyses with bath state, deposition parameters, and the impacts of electrolyte aging, which are addressed in later sections.

7.4.3 Electrolyte bath characterization

i. Electrolyte properties

The characterization of the electrolyte bath represents a critical component of the open electroplating dataset, as it defines the physicochemical state under which all deposition experiments were carried out. While the detailed measurement procedures have been described in the Methodology chapter, this section explains how these bath-related variables were recorded and incorporated into the dataset. The key measured properties are summarized in Table 7.3.

Before Electroplating	
Measured properties	Electrical Conductivity
	Copper Sulphate Concentration
	Specific gravity
During Electroplating	
Measured properties	Temperature

Table 7.3 Summary of electrolyte properties measured before and during electroplating.

Prior to each electroplating run, three fundamental physicochemical parameters were quantified: electrical conductivity, copper sulfate concentration, and specific gravity. These quantities together characterize the ionic composition, solute content, and transport capability of the electrolyte.

- **Electrical conductivity** serves as a direct measure of the total ionic strength and mobility of charge carriers, reflecting how efficiently current can be transmitted through the solution.
- **Specific gravity** provides a complementary indicator of total solute concentration and indirectly tracks the relative balance of CuSO_4 and H_2SO_4 species.
- **Copper sulfate concentration** was taken to be equal to that of the freshly prepared initial bath for all experiments, despite repeated use of the electrolyte, in order to represent the nominal availability of copper ions participating in the electrodeposition process.

Each of these parameters was recorded before every experimental session, resulting in 114 individual bath characterizations across the entire dataset. Together, they capture the gradual evolution of the electrolyte composition during repeated use (transitioning from fresh to aged states) and provide a quantifiable means to correlate bath quality with deposition performance.

During electroplating, the temperature of the bath was maintained at a controlled level between 26 °C and 27 °C. This narrow range ensured stable ion mobility and minimized thermal fluctuations that could otherwise introduce variability in current density or deposition rate. Since temperature remained effectively constant throughout all experiments, it was treated as a fixed process condition rather than a variable parameter within the dataset.

Overall, these bath measurements define the electrolyte state descriptors that serve as the foundation for subsequent analysis. Their evolution across successive baths enables the examination of how solution degradation affects electrochemical performance and surface properties, topics explored in detail in the following sections.

ii. Electrolyte variants

To evaluate the effect of electrolyte composition on electroplating behavior, four $\text{CuSO}_4\text{--H}_2\text{SO}_4$ bath formulations were prepared with initial CuSO_4 concentrations of 0.1 M, 0.3 M, 0.5 M, and 0.7 M, while maintaining constant sulfuric acid concentration at 1M. The initial physicochemical properties of these baths are summarized in Table 7.4.

Solution	CuSO ₄ Concentration	Electric Conductivity	Specific Gravity
	Error: ± 0.0002 mol/L	Error: ± 0.4 mS/cm	Error: ± 0.001 g/L
1	0.1	388	1.075
2	0.3	356	1.104
3	0.5	323	1.136
4	0.7	309	1.158

Table 7.4 Initial CuSO_4 concentrations and corresponding conductivity and specific-gravity values for the four investigated baths.

As shown in Table 7.4, increasing CuSO_4 concentration produces a clear rise in specific gravity and a slight decrease in electrical conductivity. This inverse relationship originates from the fundamental differences in ionic charge transport between the species present in the electrolyte. In the $\text{CuSO}_4\text{--H}_2\text{SO}_4$ system, the sulfuric acid component is highly dissociated, generating a large number of light and highly mobile hydrogen ions (H^+) that dominate the overall conductivity of the solution. When the CuSO_4 concentration is increased, part of the acid fraction is effectively replaced by copper ions (Cu^{2+}) and sulfate ions (SO_4^{2-}). Although this increases the total solute concentration (thereby raising the solution's specific gravity), the newly introduced ions exhibit significantly lower mobility compared to protons due to their greater mass and stronger solvation

in the aqueous medium. As a result, the total conductivity of the solution decreases slightly despite the higher ionic content.

Additionally, elevated CuSO_4 levels enhance ion–ion interactions and increase solution viscosity, both of which contribute to reduced ionic mobility. These effects become more pronounced at higher molarities, where partial association between Cu^{2+} and SO_4^{2-} ions or complex formation can further hinder charge transport. Consequently, the observed combination of higher specific gravity and modestly lower conductivity reflects a balance between ionic concentration and mobility, governed by both solute composition and molecular-scale transport dynamics.

The four selected CuSO_4 concentrations (0.1, 0.3, 0.5, and 0.7 M) therefore establish a controlled and reproducible range of electrolyte chemistries. Together, they capture the transition from highly conductive, acid-dominant solutions to denser, copper-rich electrolytes where ionic motion is increasingly restricted. This range forms a reliable foundation for examining how electrolyte composition influences current response, pulse behavior, and the resulting surface characteristics of the deposited layers.

iii. Bath-state mapping

As mentioned earlier, the dataset includes the measured conductivity and specific gravity values for all 114 electrolyte baths used throughout the experimental campaign. Figure 7.3 presents a comprehensive mapping of these measurements, illustrating how the two physicochemical parameters vary across all recorded baths. Each point in the figure corresponds to a distinct bath composition and condition at the time of electroplating, collectively representing the complete range of electrolyte states captured in the dataset.

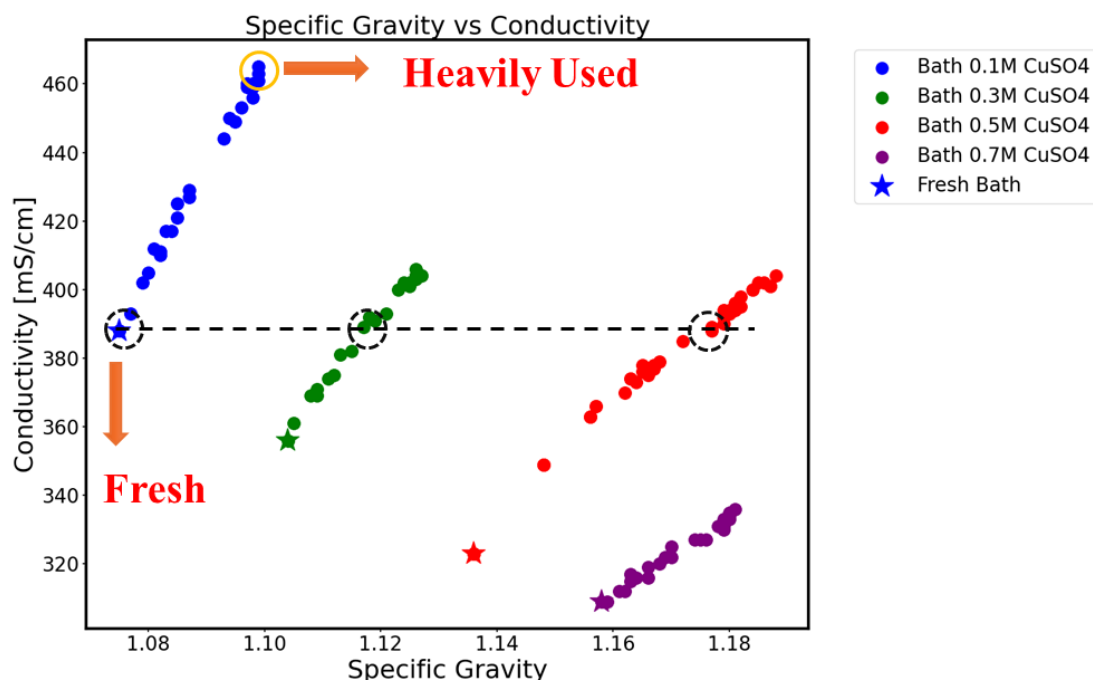


Figure 7.3 Mapping of bath state based on measured conductivity and specific gravity for the four investigated CuSO_4 concentrations.

Although a detailed analysis of bath evolution is presented later in this chapter (Section 7), the mapping in Figure 7.3 already highlights a key observation: neither electrical conductivity nor specific gravity alone can reliably represent the true state of the bath. As shown in the figure, different CuSO_4 concentrations can exhibit similar conductivity values at distinct stages of use. For example, a fresh low-concentration bath (0.1 M) and a heavily used high-concentration bath (0.3 and 0.5M) may overlap in conductivity despite having fundamentally different ionic compositions. The same limitation applies to specific gravity, which may increase due to higher solute concentration or byproduct accumulation during aging.

This overlap emphasizes that a multidimensional approach (combining both conductivity and specific gravity) is necessary to distinguish electrolyte states accurately. The two parameters together provide a more complete description of the bath's physicochemical condition, capturing the combined effects of ion concentration, mobility, and degradation products. This joint interpretation forms the basis for the bath-state analysis presented in Section 8, where the temporal evolution and its influence on deposition quality are examined in detail.

7.5 Distribution of measured variables

The complete dataset encompasses a total of 360 electroplated samples, each associated with a corresponding set of bath, process, and surface-quality descriptors. To verify that the

measurements adequately represent the entire operational space, the distribution of the main variables (electrolyte properties, process temperature, and surface outcomes) was analyzed. The resulting plots and summary tables provide insight into the internal consistency, variability, and balance of the dataset.

7.5.1 Bath property distributions

The histograms in Figures 7.4 –7.6 depict the distribution of specific gravity, electrical conductivity, and bath temperature across all recorded samples.

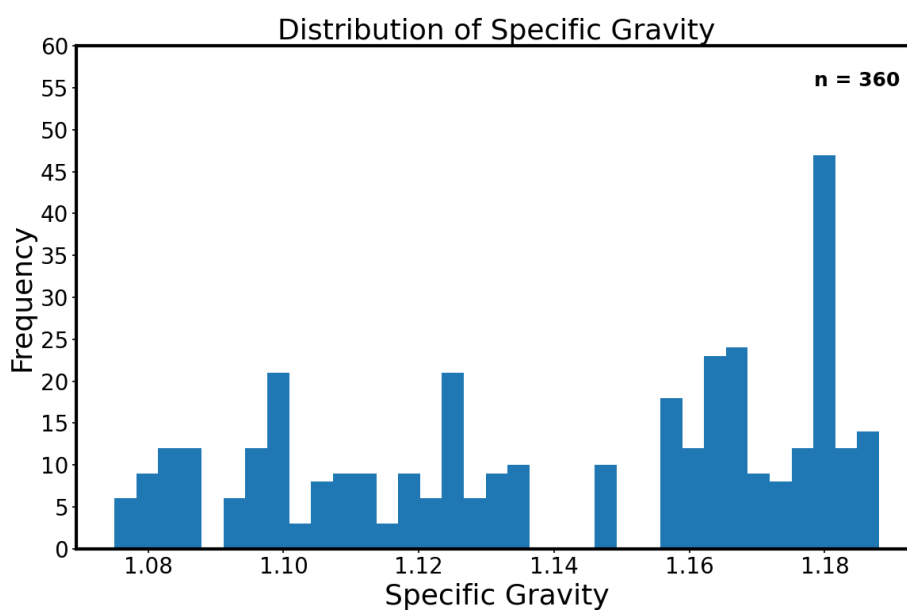


Figure 7.4 Distribution of specific gravity values for all electrolyte baths in the dataset.

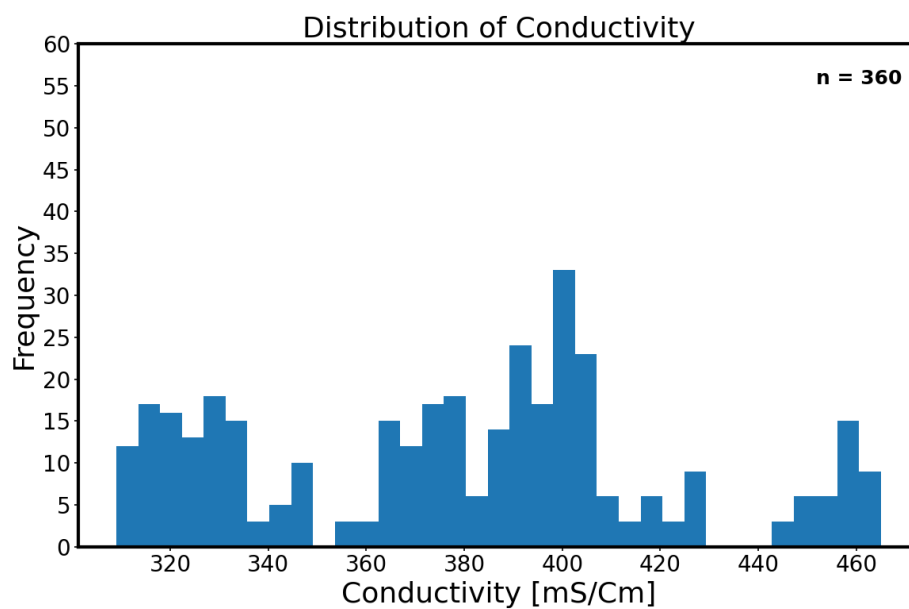


Figure 7.5 Distribution of electrical conductivity for all electrolyte baths in the dataset.

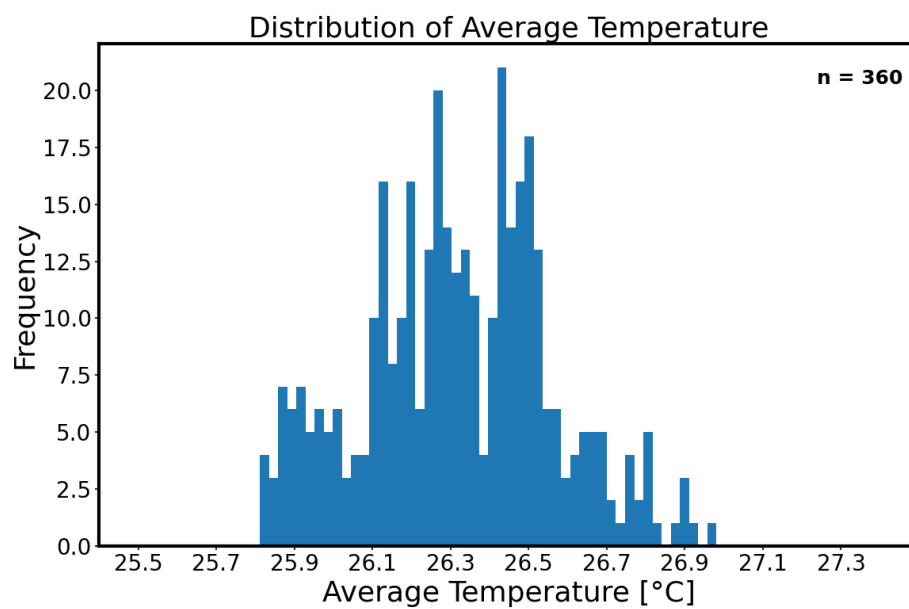


Figure 7.6 Distribution of average bath temperature during electroplating.

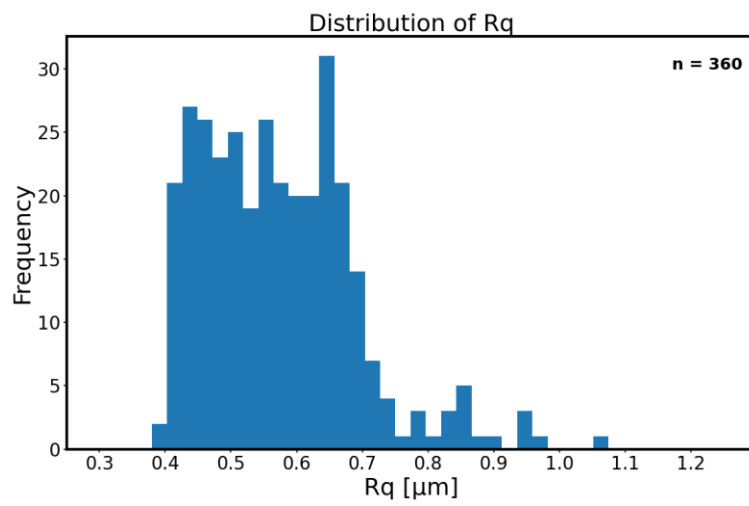
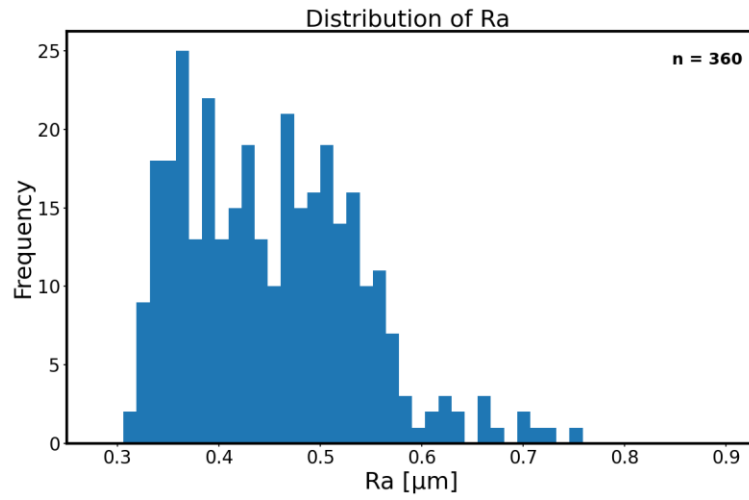
The data reveal a well-spread distribution of specific gravity values, ranging approximately from 1.075 to 1.18, which corresponds to the four baseline CuSO₄ concentrations and their subsequent usage stages. Conductivity values are distributed between 310 and 460 mS/cm, indicating that the dataset covers both highly conductive, acid-dominant baths and lower-conductivity, Cu-rich electrolytes. Together, these two variables provide a balanced representation of bath states, confirming that no single composition or aging condition dominates the dataset.

Bath temperature remained effectively constant throughout the experiments, as shown by the narrow Gaussian-like distribution centered around $26.3\text{ }^{\circ}\text{C} \pm 0.4\text{ }^{\circ}\text{C}$. This controlled temperature range ensured that any observed variations in surface properties primarily reflect differences in electrolyte condition and electrochemical dynamics, rather than thermal fluctuations.

7.5.2 Surface roughness and gloss distributions

The roughness presented in Figure 7.7 summarizes the topographical characteristics of the 360 samples. The Ra values are distributed between 0.3 and 0.8 μm , with an average of 0.55 μm , while the Rq follows a similar pattern, distributed between 0.3 and 1.1 μm , averaging 0.72 μm . The Rz spans a broader range (2–7.5 μm), averaging 4.44 μm , reflecting the higher sensitivity of this metric to isolated surface features and localized nodular growth.

The roughly right-skewed distributions observed for all roughness parameters indicate that most deposits achieved relatively smooth finishes, with fewer occurrences of coarser or defective morphologies. This statistical profile suggests stable process control across the experimental campaign.



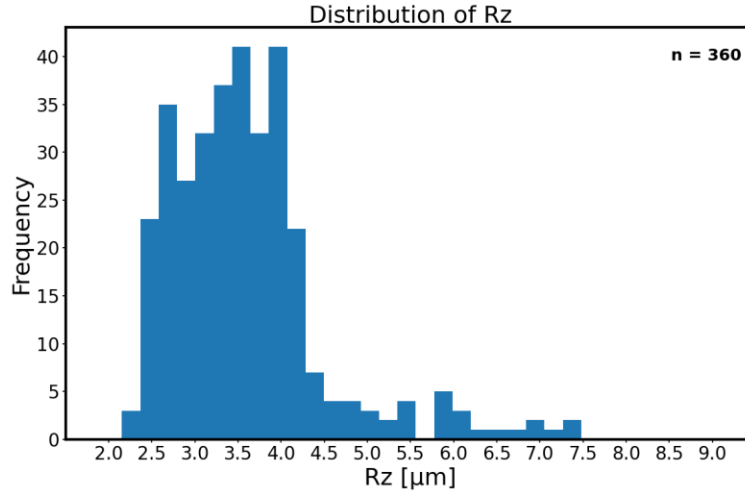


Figure 7.7 Distributions of surface roughness parameters (Ra, Rq, Rz) across all 360 electroplated samples.

The gloss measurements, shown in Figure 7.8, summarize the optical response of the electroplated surfaces under incident angles of 20° and 60° (G20 and G60, respectively). The dataset shows an average G20 value of 2.3 GU and an average G60 of 22.75 GU, both exhibiting right-skewed distributions. Most samples display moderate reflectivity with a limited number of high-gloss outliers, consistent with the visual appearance of semi-bright to bright copper deposits produced in additive-free electrolytes.

The G20 distribution (more sensitive to micro-level surface irregularities) shows a tighter clustering around low gloss values, whereas G60, which reflects broader angular scattering, exhibits a wider spread. This divergence highlights the complementary nature of these two measurements: G20 captures fine topographical uniformity, while G60 reflects overall specular behavior. Together with roughness descriptors, these optical data provide a quantitative link between the morphological and visual quality of the electroplated layers.

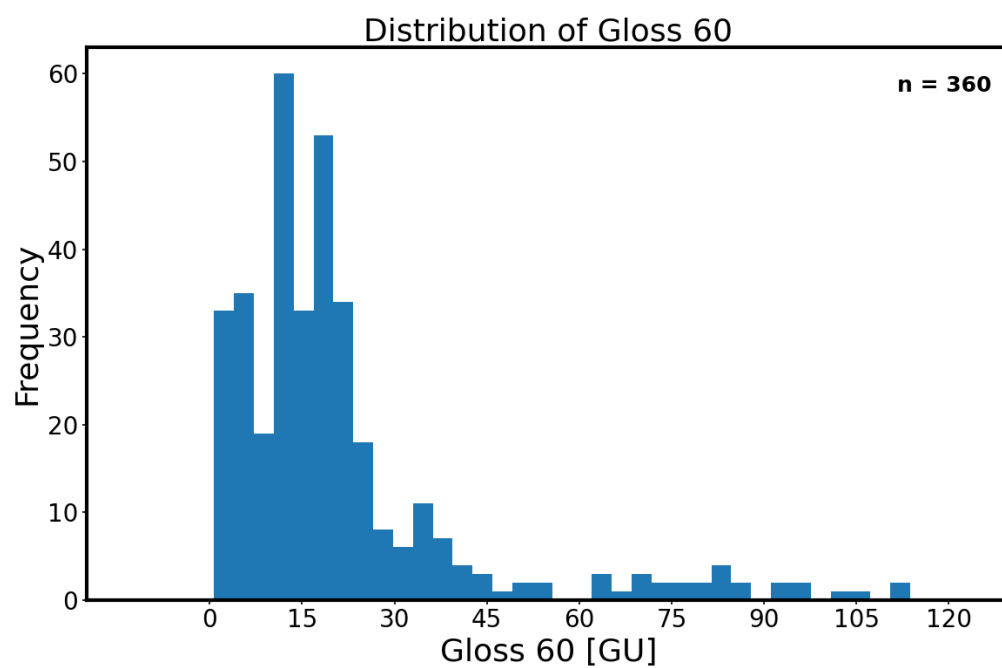
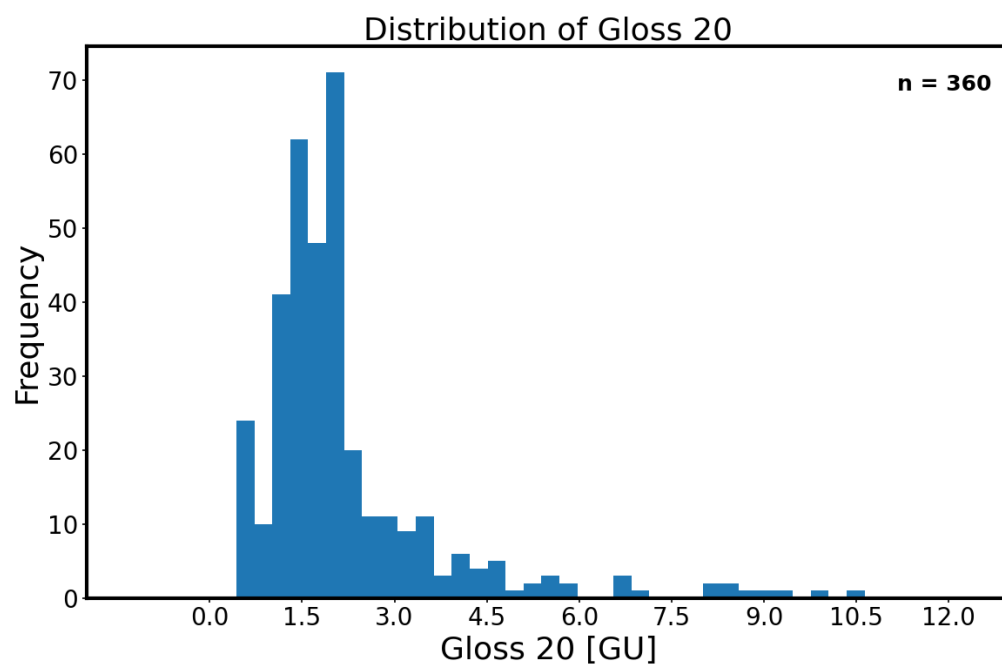


Figure 7.8 Distribution of surface gloss measured at 20° (G20) and 60° (G60) reflection angles across all 360 electroplated samples.

A clear inverse relationship exists between the surface roughness and gloss distributions presented in Figures 7.4 – 7.8. Samples with lower Ra and Rq values tend to exhibit higher gloss readings at both 20° and 60°, confirming the optical–topographical coupling typical of metallic deposits. This trend arises because smoother surfaces reflect light more specularly, increasing apparent brightness, whereas rougher morphologies promote diffuse scattering and reduce gloss intensity. The consistency of the right-skewed distributions for both roughness and gloss metrics supports this interpretation, indicating that most electroplated samples achieved moderate smoothness and corresponding brightness, with fewer extreme cases representing the roughest and dullest deposits.

The complementary Table 7.5 summarizes surface brightness and roughness of the deposits yielded four fresh electrolytes of increasing CuSO₄ concentrations under the same electroplating parameters ($E_{\text{dep}} = -150$ mV, $Q_{\text{dep}} = 0.1$ C/cm², $Q_{\text{pol}} = 0.01$ C/cm²). This highlights the clear influence of bath composition on deposit morphology. Increasing CuSO₄ concentration systematically enhanced surface brightness, from 1.5 and 12.6 GU at 0.1 M to 3.13 and 33.7 GU at 0.7 M, and simultaneously reduced roughness parameters. This improvement is consistent with enhanced metal-ion availability at higher Cu²⁺ concentrations, promoting more uniform nucleation and smoother growth during the cathodic phase.

Electrolyte	Composition (mol/L)		Surface Roughness (μm)	Surface Roughness Standard Deviation (μm)	Surface Brightness (GU)	Surface Brightness Standard Deviation (μm)
A	CuSO ₄ : 0.1 H ₂ SO ₄ : 1	Ra	0.450	0.018	G20:1.50	0.03
		Rq	0.569	0.029		
		Rz	3.516	0.310	G60:12.64	0.19
B	CuSO ₄ : 0.3 H ₂ SO ₄ : 1	Ra	0.408	0.011	G20: 1.86	0.03
		Rq	0.507	0.022		
		Rz	3.110	0.126	G60: 16.92	0.55
C	CuSO ₄ : 0.5 H ₂ SO ₄ : 1	Ra	0.424	0.014	G20: 2.09	0.03
		Rq	0.510	0.021		
		Rz	3.163	0.198	G60: 20.69	0.35
D	CuSO ₄ : 0.7 H ₂ SO ₄ : 1	Ra	0.361	0.011	G20:3.13	0.03
		Rq	0.452	0.016		
		Rz	2.745	0.174	G60:33.75	0.32

Table 7.5 Surface brightness and roughness of deposits obtained from the four fresh electrolyte baths (0.1– 0.7 M CuSO₄) under identical electroplating process parameters.

Together, these distributions confirm that the constructed open dataset captures a diverse yet statistically balanced range of bath and surface conditions. The narrow temperature variation, wide spread in electrolyte parameters, and continuous distribution of surface metrics make the dataset well-suited for both statistical correlation analysis and machine-learning-based predictive modeling. By covering the full spectrum from fresh to heavily used baths, the data provide the foundation for linking physicochemical state, electrochemical pulse behavior, and resulting surface quality in subsequent analyses.

7.6 Correlation between process parameters and surface properties

The relationship between the electroplating process parameters and the resulting surface characteristics was examined to identify the governing trends linking electrochemical conditions with surface morphology and brightness. The figures presented in this section summarize the variation of Ra, G20, and G60 as functions of E_{dep} , Q_{dep} , and Q_{pol} across four investigated CuSO_4 concentrations (0.1, 0.3, 0.5, and 0.7 M). The trends reflect the average behavior observed across multiple samples within each bath, providing an integrated view of how process control influences deposit quality.

7.6.1 Effect of deposition voltage

As Figure 7.9 shows, at constant deposition and polishing charge, the surface gloss exhibited a nonlinear dependence on cathodic potential, while surface roughness increased gradually once the voltage exceeded a bath-specific threshold. As the potential became more negative, both G20 and G60 rose to a maximum before declining, forming a characteristic dome-shaped response. This intermediate potential range corresponds to a regime of uniform nucleation and compact grain growth, where the balance between ionic transport and charge transfer remains optimal.

The width of this optimal potential window depends strongly on bath concentration. In the most concentrated electrolyte (0.7 M CuSO_4), the stable and bright-deposition region extended from approximately -300 to -600 mV, whereas in the 0.5 M bath, it spanned -300 to -500 mV. The 0.3 M and 0.1 M baths exhibited narrower ranges, limited to about -300 to -400 mV and -300 to -350 mV, respectively. These differences indicate that electrolytes with higher Cu^{2+} concentrations sustain stable deposition across a broader potential range, as improved ion availability reduces concentration polarization and delays the onset of transport limitations.

Beyond this optimal region, however, mass-transport limitation and hydrogen evolution begin to dominate the cathodic process. These effects induce local pH increases and micro-pitting on the deposit surface, leading to a rise in Ra and a decline in specular reflection. The transition point varies slightly between baths: the 0.7 M solution maintained smooth morphology down to approximately -600 mV without roughening, while the 0.1 M bath deteriorated near -380 mV. The 0.3 M and 0.5 M baths remained stable only up to about -450 mV. Such bath-specific limits arise from variations in ionic mobility and copper-ion replenishment, where higher Cu^{2+} content allows more uniform current distribution and suppresses localized gas evolution at high overpotentials.

Moreover, the gloss maximum shifts toward more negative potentials and higher absolute values as CuSO_4 concentration increases. This trend demonstrates that concentrated electrolytes can sustain higher current densities before mass-transport constraints emerge, allowing smoother, denser metal growth over a wider potential interval. Enhanced Cu^{2+} availability promotes faster

yet uniform nucleation and growth kinetics, resulting in greater surface brightness and a delayed onset of roughening compared with more dilute baths.

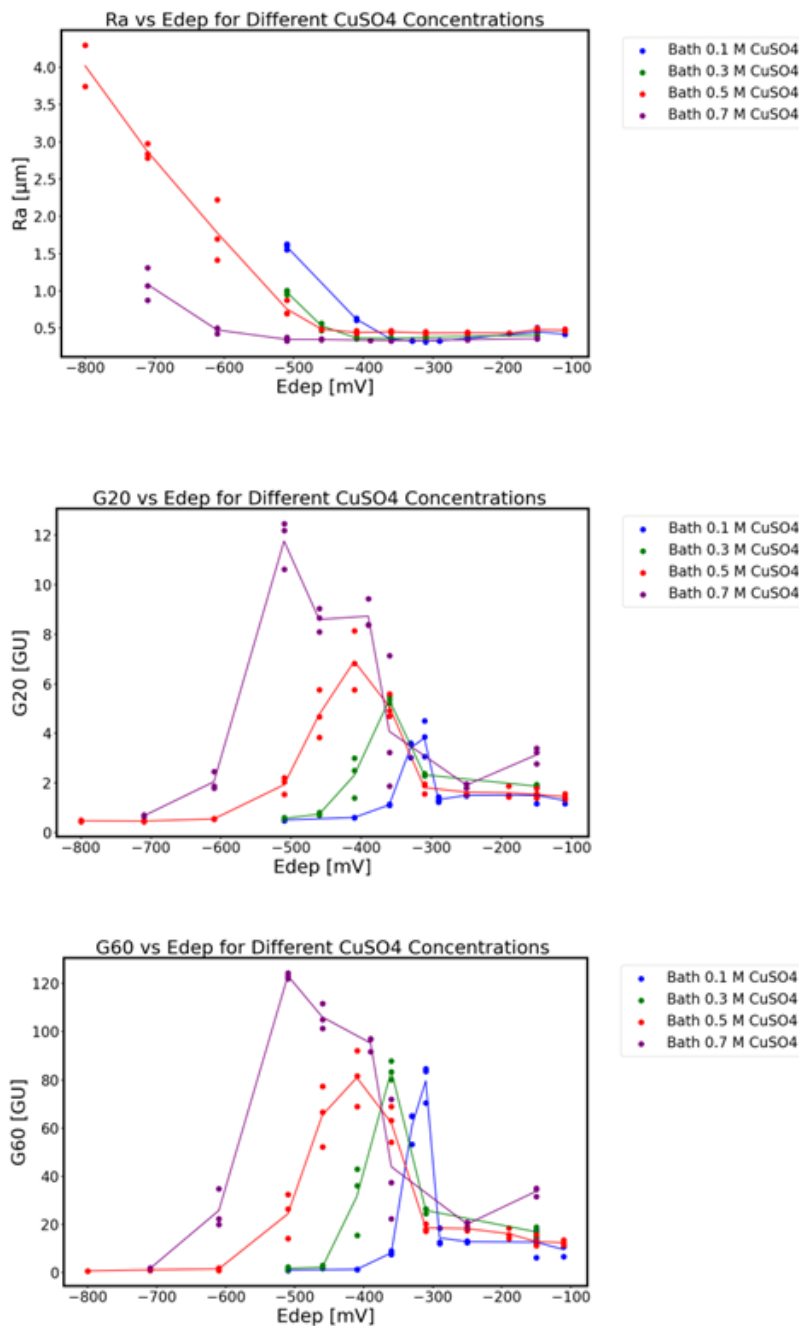


Figure 7.9 Effect of deposition voltage on Ra, G20, and G60 for baths with four initial CuSO4 concentrations

7.6.2 Effect of deposition charge

The variation of deposition charge from -1 to -26 C cm^{-2} , as shown in Figure 7.10, indicates that surface brightness decreased slightly with increasing $|Q_{\text{dep}}|$ before reaching a steady level, while surface roughness remained nearly constant. This behavior can be attributed to the uniform final thickness applied to all samples, meaning that changes in deposition charge primarily adjusted the current–time profile rather than the total deposited layer. Consequently, variations in $|Q_{\text{dep}}|$ did not significantly alter the overall surface morphology.

The influence of bath composition was again evident: the bath containing 0.7 M CuSO_4 produced smoother and glossier surfaces across the entire Q_{dep} range compared with the lower-concentration baths, consistent with enhanced ionic strength and faster charge replenishment during the cathodic phase.

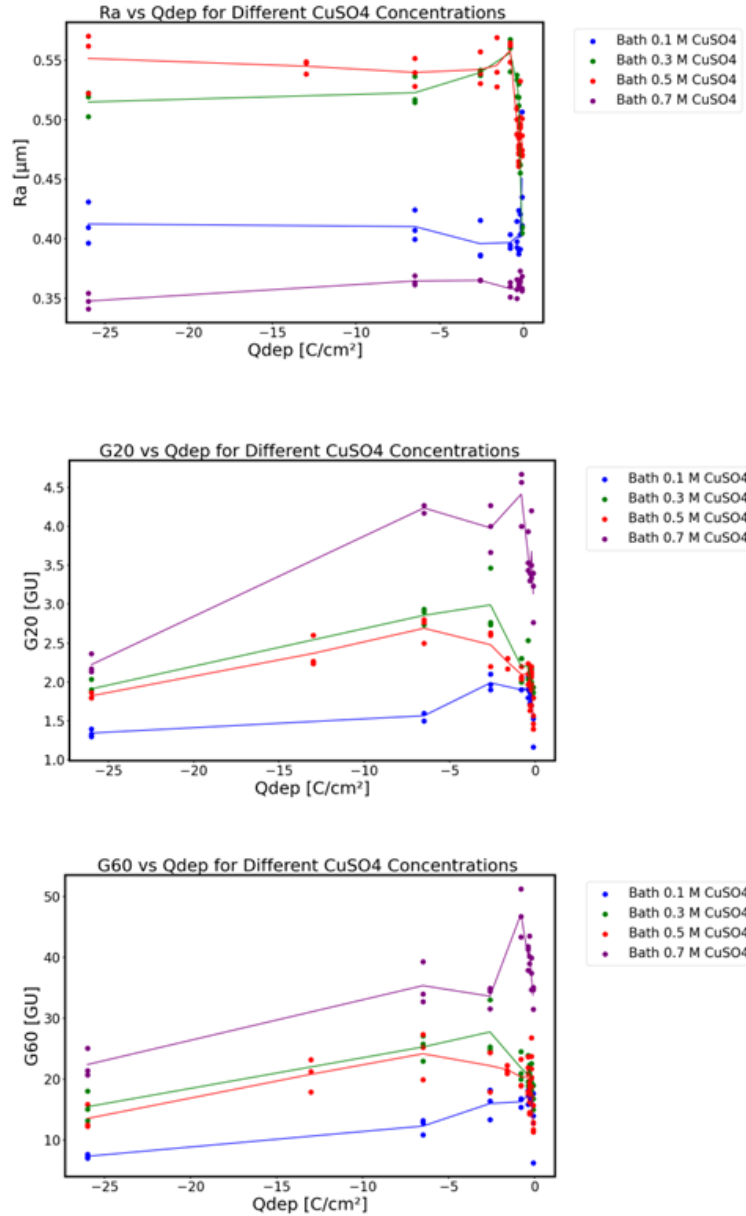


Figure 7.10 Effect of deposition charge on Ra, G20, and G60 for baths with four initial CuSO4 concentrations

7.6.3 Effect of polishing charge

The role of the anodic (polishing) phase was evaluated under two deposition-charge regimes: a $Q_{dep} = -0.1 C/cm^2$ and $Q_{dep} = -6.5 C/cm^2$ to represent both low and high deposition-charge conditions (Figure 7.11). In both cases, the surface properties followed an optimum-window behavior. At low to moderate Q_{pol} , the reverse (anodic) pulse selectively dissolved microscopic protrusions and surface irregularities, thereby reducing Ra and producing a significant increase in

gloss. Beyond this optimum range, however, excessive anodic charge reversed the trend: over-etching at grain boundaries and step edges introduced fine-scale roughness and reduced surface reflectivity.

Bath concentration again played a decisive role. Bath with higher Cu^{2+} content (0.7 M) produced higher gloss peaks at the optimal Q_{pol} values compared to the 0.1 M, 0.3 M, and 0.5 M baths. This behavior arises from the greater ionic availability and buffering capacity of concentrated electrolytes, which sustain smoother anodic dissolution and minimize surface pitting during the reverse pulse. In contrast, dilute baths experience faster local ion depletion and stronger polarization, causing premature surface dulling once the anodic charge exceeds the optimal range. As a result, the gloss enhancement is more pronounced in high-concentration baths, reflecting their superior electrochemical stability and leveling efficiency.

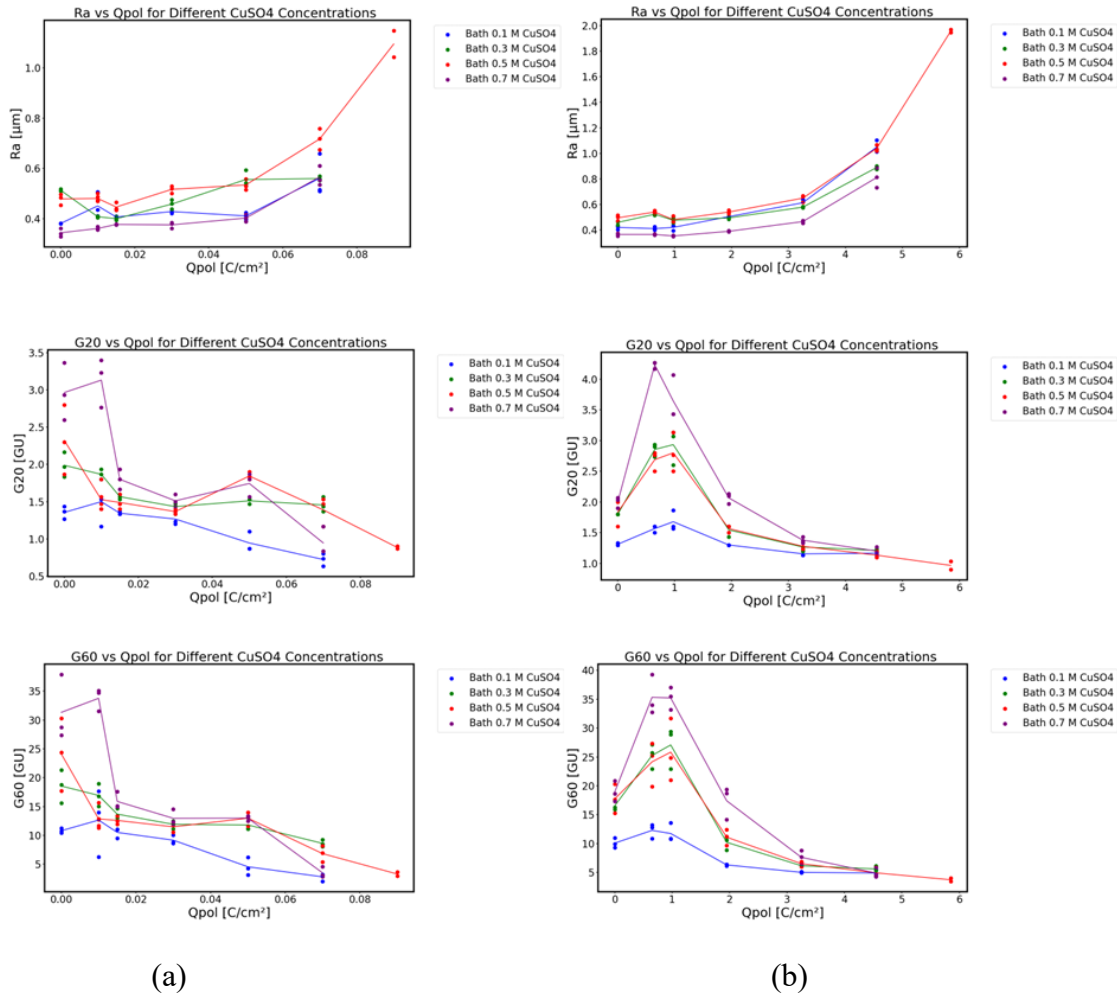


Figure 7.11 Effect of polishing charge on Ra, G20, and G60 baths with four initial CuSO_4 concentrations under two deposition-charge regimes: (a) $Q_{\text{dep}} = -0.1 \text{ C}/\text{cm}^2$ and (b) $Q_{\text{dep}} = -6.5 \text{ C}/\text{cm}^2$

7.7 The evolution of the electrolyte bath state

The long-term stability of the electrolyte plays a decisive role in determining both process reproducibility and the resulting surface quality of electroplated parts. To characterize this evolution, the relationship between specific gravity and electrical conductivity was analyzed for all recorded baths, as presented in Figure 7.12. Each point in this plot represents an individual bath condition at a given operational stage, from the initial (fresh) state to the heavily used condition reached after repeated deposition cycles.

In freshly prepared electrolytes, conductivity and specific gravity exhibit distinct values associated with the CuSO_4 concentration. However, as the baths are repeatedly used, both parameters evolve due to gradual ion consumption, by-product accumulation, and changes in the H_2SO_4 -to- Cu^{2+} ratio. Conductivity increases as the active Cu^{2+} content rises relative to the acid fraction. Simultaneously, specific gravity increases because of the higher total solute content and accumulation of reaction products, producing a characteristic trajectory that moves from low-specific gravity/low-conductivity (fresh) to high-specific gravity /high-conductivity (aged) regions in the plot.

The dataset reveals that bath evolution does not follow a single, universal trajectory. To illustrate this, a de-aging experiment was conducted using the 0.3 M CuSO_4 bath as a representative case. Portions of the aged electrolyte were mixed with fresh solution in 0%, 25%, 50%, and 100% (v/v) ratios to reproduce the common industrial practice of rejuvenating degraded baths. As shown by the cross symbols in Figure 7.12, the resulting recovery trajectory followed a markedly different slope from the original aging path. This observation demonstrates that bath evolution is not strictly reversible; each intervention, such as the partial addition of fresh electrolyte, effectively creates a new chemical environment with distinct ionic ratios and transport properties. Consequently, the bath continues to evolve along a new trajectory rather than retracing its previous path.

This nonlinearity highlights the path-dependent nature of electrolyte behavior, governed by its cumulative operational history rather than its instantaneous composition. Industrial practices that rely on periodic replenishment, therefore, yield baths with unique properties and aging kinetics, even when nominal concentrations are restored. In practical terms, bath aging does not proceed with a constant slope or predictable pattern but instead reflects the interplay of prior usage, replenishment strategy, and evolving ion balance within the electrolyte.

These findings have important implications for industrial electroplating and electroforming operations. In practice, operators may monitor conductivity or specific gravity to decide when to refresh or partially replace the electrolyte. However, as seen in Figure 7.12, baths of different concentrations can exhibit similar conductivity values at different stages of their lifetime, making either parameter alone an unreliable indicator of bath health. Instead, their combined evolution provides a more meaningful fingerprint of electrolyte condition. The observed nonlinearity emphasizes the need for multivariate monitoring strategies, where conductivity, specific gravity, and possibly other derived indicators (e.g., pulse-reverse characteristics and $\text{H}_2\text{SO}_4/\text{CuSO}_4$ ratio) are jointly analyzed to track bath aging and regeneration more accurately.

Together, these results confirm that electrolyte aging is a path-dependent process, where each intervention can alter the aging slope and create a new equilibrium state. Freshness or degradation

cannot be captured by a single variable but must be inferred from correlated trends in measurable properties. The constructed dataset, therefore, provides a robust experimental foundation for modeling bath state evolution and developing data-driven tools capable of predicting when and how an electrolyte will deviate from its optimal condition.

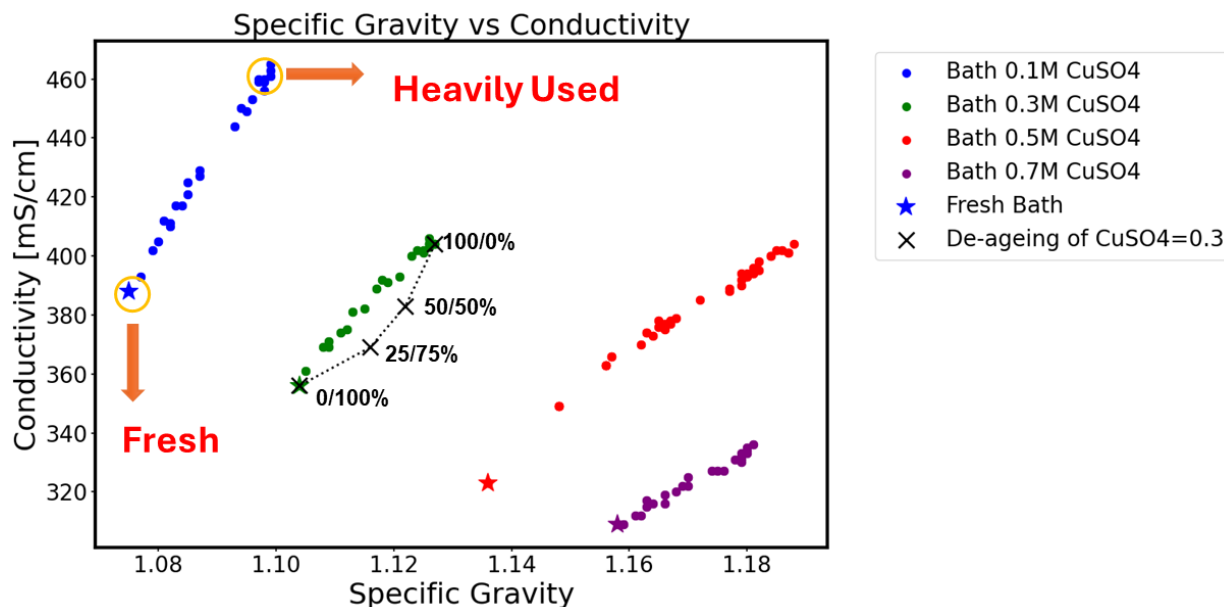


Figure 7.12 Relationship between specific gravity and conductivity for all captured $\text{CuSO}_4\text{--H}_2\text{SO}_4$ electrolytes in the open dataset, showing the transition from fresh to heavily used conditions. The “de-aging” trajectory of the 0.3 M bath demonstrates that rejuvenation through fresh-solution addition follows a distinct path, reflecting the nonlinearity of bath evolution and partial recovery behavior.

7.8 Effects of bath states (Aged vs Fresh) on the deposition properties under identical process parameters

To evaluate the influence of electrolyte aging on surface characteristics, a controlled comparison was conducted between fresh and aged 0.5 M CuSO_4 baths under identical pulse–reverse conditions ($E_{\text{dep}} = \text{variable}$, $Q_{\text{dep}} = -0.1 \text{ C/cm}^2$, $Q_{\text{pol}} = 0.01 \text{ C/cm}^2$). The results, summarized in Figure 7.13, highlight the pronounced impact of bath state on both surface roughness and optical brightness. For the aged bath, each condition was repeated up to three times to ensure statistical consistency in the gathered dataset, as greater variability was observed between runs due to the bath’s unstable electrochemical state. In contrast, for the fresh bath, a single repetition per condition was sufficient since the results remained consistent across trials, indicating stable behavior and excellent reproducibility in the unaged electrolyte.

In the fresh bath, increasing the cathodic potential from -100 mV to -600 mV produced a marked enhancement in surface brightness (reaching approximately 17.5 GU at G20 and 160 GU at G60) together with a reduction in Ra, achieving optimal smoothness and gloss near -300 mV. This behavior reflects the performance of an active, ion-rich electrolyte capable of sustaining uniform nucleation and compact grain growth throughout the potential range.

In contrast, the aged bath, characterized by a higher specific gravity (1.187) and lower conductivity (404 mS/cm) relative to the fresh state (1.136 and 323 mS/cm, respectively), exhibited substantially lower gloss and higher roughness across all voltages. In this case, Ra began to increase beyond -500 mV, and the gloss–potential curves retained the same dome-shaped profile but with reduced amplitude (approximately 60% for G20 and 50% for G60 compared to fresh bath) and shifted peak intensity, occurring at a higher overpotential around -400 mV within a narrower effective range (-300 mV to -500 mV). This attenuation indicates a decline in ionic mobility and a growing influence of diffusion-limited deposition in the aged electrolyte. As Cu^{2+} ions are progressively depleted and reaction by-products accumulate, mass transport toward the cathode becomes less uniform, favoring coarse-grained or nodular growth and thereby reducing specular reflection.

The increase in surface roughness at equivalent process conditions confirms that aging alters the electrochemical balance between deposition and dissolution during each pulse cycle. In the aged state, diminished proton activity and reduced Cu^{2+} availability (near the cathode) weaken the anodic smoothing efficiency, allowing persistent microasperities to remain after each reversal. This explains the observed reduction in both G20 and G60, as diffuse scattering increases with topographical irregularities. The magnitude of this degradation further supports the interpretation that electrolyte aging directly modulates the effective kinetics and transport regime of copper deposition, even when macroscopic parameters (voltage, charge, and temperature) are held constant.

The comparison clearly shows that bath condition exerts a stronger influence on final surface quality than moderate changes in operating potential within a given process window. Consequently, rather than relying solely on periodic regeneration or replacement, it becomes more critical to develop a reliable method for estimating the instantaneous state of the electrolyte. Such predictive evaluation would enable a clearer understanding of the expected surface outcome under specific bath conditions at any given time. Since aging does not follow a fixed or repeatable trajectory, as demonstrated in the previous section, even small shifts in bath chemistry can markedly alter deposition behavior. Accurate state estimation, therefore, represents the most effective strategy for achieving consistent plating performance and ensuring process predictability across varying operational histories.

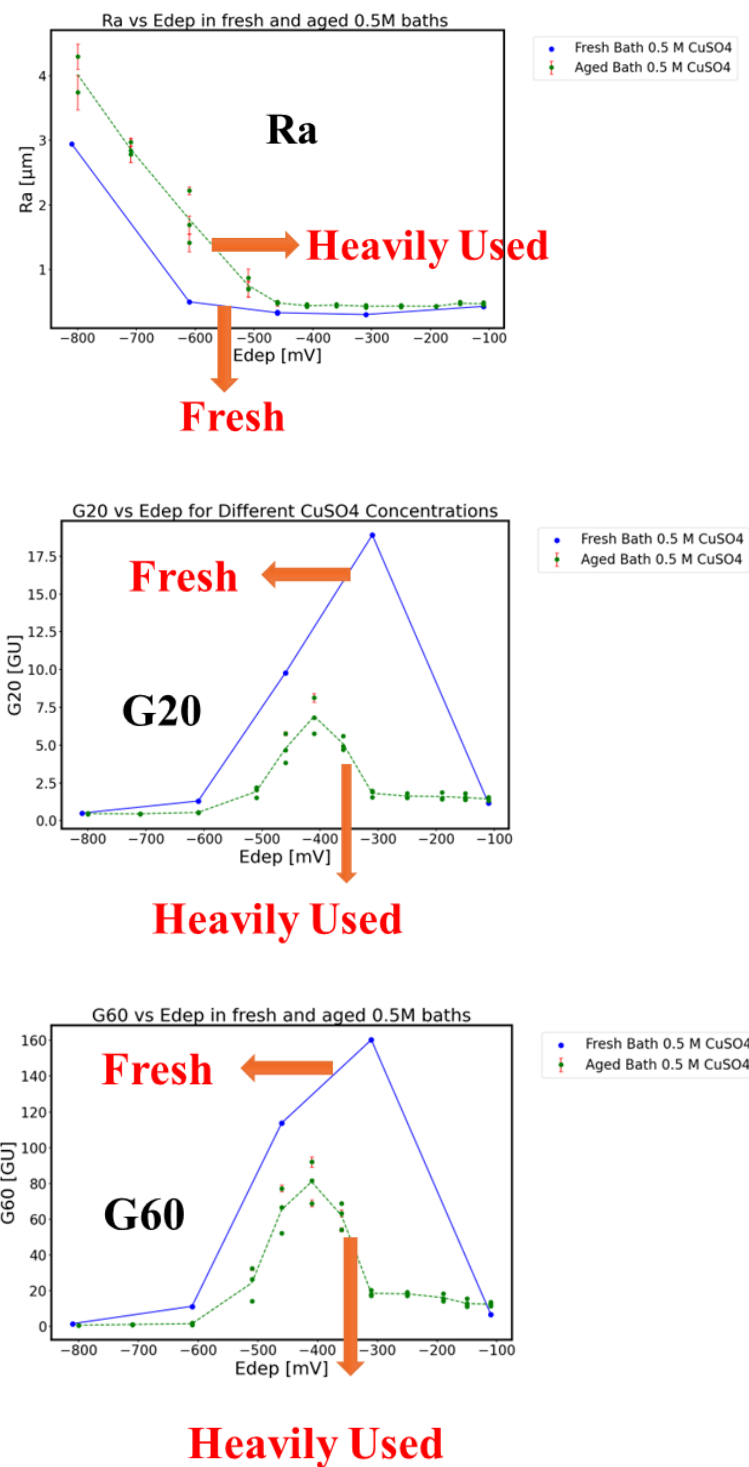
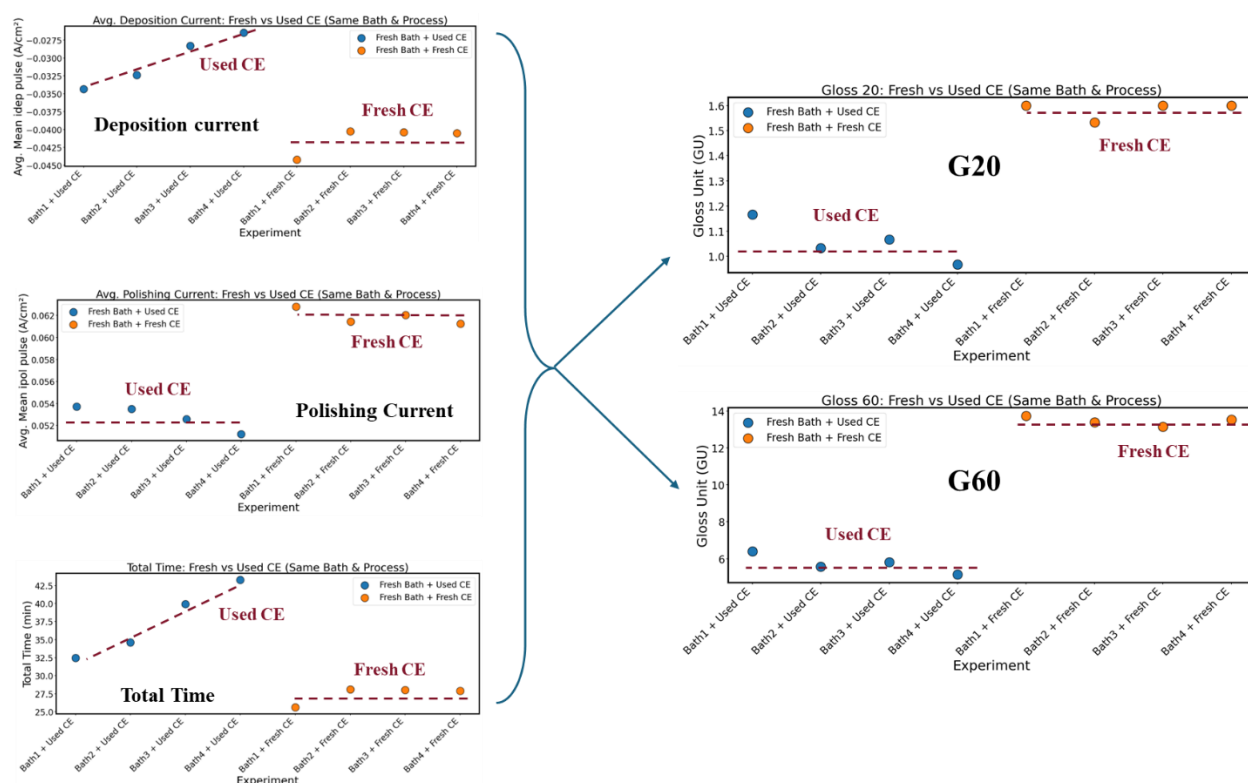


Figure 7.13 Comparison of surface roughness (Ra) and gloss (G20, G60) for fresh and aged 0.5 M CuSO₄ baths under identical pulse–reverse conditions (E_{dep} = variable, Q_{dep} = -0.1 C/cm², Q_{pol} = 0.01 C/cm²).

7.9 Counter electrode conditions effects

During the construction of the dataset, it was realized that the condition of the counter electrode (CE) exerts a significant influence on the electrochemical balance and the resulting optical properties of electroplated surfaces. To isolate this effect, a comparative study was conducted between fresh and used copper counter electrodes under identical pulse–reverse electroplating parameters ($E_{\text{dep}} = -150 \text{ mV}$, $Q_{\text{dep}} = -0.1 \text{ C cm}^{-2}$, $Q_{\text{pol}} = 0.01 \text{ C cm}^{-2}$). The used electrodes were those that had undergone multiple deposition runs with various process conditions, showing visible oxidation and discoloration, whereas the fresh electrodes were newly prepared and polished before testing. Importantly, each fresh CE and aged CE was tested four times in eight fresh baths (2 series of Bath1, Bath2, Bath3, Bath4) with the identical physicochemical properties to ensure that any observed variations originated solely from the electrode state.



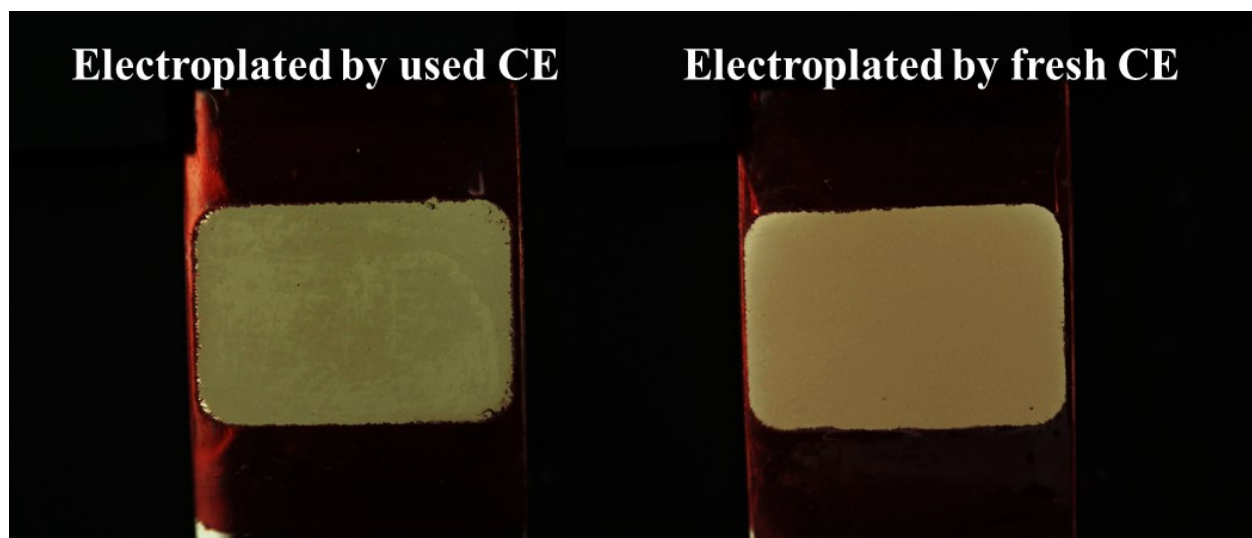


Figure 7.14 Comparison of deposition and polishing currents, total process time, and surface gloss (G20 and G60) between fresh and used counter electrodes under identical electroplating conditions. The electroplated samples with both used and fresh CE are shown here.

As shown in Figure 7.14, a clear distinction emerged between the two conditions. When a used counter electrode was employed, both the average deposition and polishing currents decreased compared with the fresh-CE setup. This reduction in electrochemical activity reflects the buildup of oxide layers and surface irregularities on the anode, which impede uniform copper dissolution and increase cell resistance. Consequently, the total time required to deliver the same deposition charge increased, indicating a loss of efficiency within the electrochemical system.

The diminished electrochemical performance of the used electrode translated directly to optical degradation of the plated surfaces. Both gloss measurements (G20 and G60) showed a substantial decrease under the used-CE condition, even though all other process parameters and bath compositions were identical. This decline in brightness suggests a reduction in the uniformity of ion flux and local current distribution at the cathode, leading to more scattered light reflection despite maintaining comparable roughness.

These results demonstrate that counter-electrode degradation introduces an additional dimension of system variability, distinct from electrolyte aging but with comparable consequences. Over multiple cycles, the progressive oxidation and roughening of the anode alter its dissolution behavior, shifting the effective current–time response and thereby modifying the deposition dynamics. Such effects can accumulate unnoticed in long-term electroplating or electroforming operations, ultimately influencing surface gloss and coating uniformity even when bath monitoring indicates a stable state.

Furthermore, the pulse-current data themselves (specifically the deposition and polishing current densities) can serve as valuable indicators of CE condition. Because changes in electrode oxidation, roughness, or conductivity directly influence current-time behavior, these pulse-related

parameters can be learned and modeled as diagnostic features for detecting counter-electrode aging. This approach would enable understanding performance shifts caused by electrolyte degradation and those arising from electrode wear, thereby providing a more complete and data-driven representation of the electrochemical system's state.

In this context, maintaining consistent plating quality requires not only monitoring the electrolyte condition but also tracking the evolving state of the counter electrode. The findings highlight the importance of integrating the pulse-current data into predictive frameworks. Together with bath-state descriptors such as conductivity and specific gravity, they can form a comprehensive basis for predicting surface outcomes with higher reliability based on electrolyte and CE state.

7.10 Conclusion

This chapter presented the development and analysis of a comprehensive open dataset for electroplating, constructed to capture the interdependencies between electrolyte conditions, pulse parameters, and resulting surface properties. The dataset systematically links physicochemical measurements, pulse-level current responses, and surface outcomes, providing an experimental foundation for understanding and modeling the electrochemical variability inherent in copper electroplating.

The results demonstrated that the electrolyte bath evolves dynamically over time, even under fixed process conditions. Each bath follows a distinct trajectory determined by its initial composition, operating history, and replenishment events. This evolution manifests as coupled variations in specific gravity and conductivity (moving from low-specific gravity/high-conductivity (fresh) to high-specific gravity /low-conductivity (aged) states) and is further complicated by nonlinear, path-dependent behavior. The de-aging experiments confirmed that industrial interventions such as partial bath replenishment do not restore the system to its original state but rather create new equilibrium pathways, emphasizing that electrolyte aging is neither reversible nor uniform.

These bath-state transformations directly affect the surface morphology and optical properties of the deposited copper. Aging results in diminished gloss and increased roughness, even when all process parameters remain constant. This degradation stems from altered ionic mobility, reduced proton activity, and the accumulation of by-products, which collectively disrupt the balance between deposition and dissolution within each pulse cycle. Comparisons between fresh and aged baths revealed that electrolyte condition exerts a stronger influence on surface brightness and uniformity than moderate adjustments in potential or charge.

In addition to bath aging, the condition of the counter electrode emerged as a critical factor influencing deposition stability. Used electrodes, exhibiting oxide buildup and surface roughening, caused lower deposition and polishing current densities, longer process times, and decreased gloss levels. These findings underscore that process reproducibility depends not only on electrolyte health but also on the evolving electrochemical behavior of the electrode surfaces. The pulse–reverse current data captured within the dataset thus hold diagnostic potential for identifying electrode degradation, as current–time responses inherently encode information about the state of both electrodes and the electrolyte.

Taken together, these findings highlight the limitations of traditional monitoring based solely on physical or chemical bath measurements. Parameters such as conductivity and specific gravity, while informative, fail to capture the complex electrochemical interactions that govern surface outcomes. Instead, the combined analysis of bath-state descriptors and pulse-level features offers a more complete and dynamic picture of process health. This integrated perspective enables the differentiation between performance shifts caused by electrolyte aging and those induced by electrode wear.

The chapter, therefore, establishes the foundation for a quantitative, data-driven framework to monitor and predict electroplating behavior. By leveraging multivariate indicators (including specific gravity, conductivity, CuSO_4 concentration, and pulse current metrics), the dataset enables the creation of predictive models capable of estimating both bath and system state in real time. Such models would allow proactive adjustments to maintain consistent deposit quality and advance the transition of electroforming toward a self-correcting, intelligent manufacturing process.

Ultimately, this open dataset bridges empirical experimentation and machine learning, transforming electroplating knowledge from a collection of isolated observations into a structured, reusable, and generalizable foundation for adaptive process control. It marks a crucial step toward making additive-free copper electroforming not only more predictable but also scalable for mass personalization and Industry 4.0 manufacturing ecosystems.

8. The search for bath state

8.1 Introduction

The preceding chapter established a comprehensive open dataset for additive-free copper electroplating in $\text{CuSO}_4/\text{H}_2\text{SO}_4$ electrolyte, capturing the interplay between process parameters, bath measurements, pulse-current electrochemical responses, and the resulting surface properties. That analysis revealed a critical insight: even under identical nominal deposition settings, substantial variability persisted in surface gloss and roughness. This inconsistency stemmed from the gradual, path-dependent evolution of the electrolyte bath and the coupled degradation of electrode surfaces, neither of which could be fully described by direct physicochemical measurements such as conductivity or specific gravity.

The open dataset thus highlighted a fundamental challenge: the electrolyte behaves as a dynamic system whose internal condition, or *state*, evolves with use and directly influences deposition outcomes. However, this bath state is not directly measurable. Traditional indicators capture only isolated aspects of the electrolyte, failing to account for the complex interdependence among ionic composition, transport phenomena, and electrochemical kinetics. To move beyond descriptive analysis toward predictive understanding, a new data-driven approach is required, one capable of inferring this hidden bath state from observable variables.

This chapter builds on that foundation to develop a quantitative framework for identifying and modeling the electrolyte bath state using machine learning. The goal is to determine whether the bath's evolving condition can be inferred from measurable process and electrochemical parameters and, in turn, used to predict the resulting surface characteristics (Ra, Rq, Rz, G20, G60). In essence, this chapter seeks to transform the qualitative notion of “bath aging” into a learnable, predictive variable embedded within a structured modeling workflow.

To achieve this, all available experimental features were first categorized into process parameters (E_{dep} , Q_{dep} , Q_{pol}) and bath-state-related features, including both physicochemical descriptors (conductivity, density, CuSO_4 concentration) and dynamic pulse-derived indicators (current and time statistics for deposition and polishing). A comprehensive feature-selection framework was then applied, combining eight complementary methods, including correlation analysis, Recursive Feature Elimination (RFE), Random Forest importance, mutual information, Lasso regularization, permutation importance, ANOVA F-test, and variance ranking, to identify the most informative predictors. The resulting feature subsets were grouped into six configurations, ranging from process-only inputs to hybrid combinations integrating bath and current parameters.

Each feature set was used to train multiple regression models, such as Linear Regression, K-Nearest Neighbors (KNN), Support Vector Regression (SVR), Random Forest, and Gaussian Process Regression (GPR), to predict surface outcomes and quantify their sensitivity to bath-related variables. Model performance was evaluated through 1,000 randomized 80/20 splits using relative mean absolute error (MAE/mean of target) as a robust metric for cross-model comparison.

Through this systematic exploration, the chapter seeks to identify which input structures and modeling strategies most effectively capture the latent condition of the electrolyte. Beyond identifying the best-performing model, this analysis establishes a foundation for defining and quantifying the bath state itself, a crucial step toward real-time monitoring and adaptive control in intelligent electroplating.

In this work, the term ‘*bath state*’ does not refer to a single directly measurable physicochemical quantity, nor to an assumed hidden physical constant. Instead, it is defined as an operational, low-dimensional representation of the electrolyte’s effective electrochemical condition, inferred from a combination of measurable bath properties and dynamic electrochemical response features.

Specifically, the bath state aggregates information from static physicochemical descriptors (e.g., CuSO_4 concentration, electrical conductivity, specific gravity) and dynamic pulse-response characteristics (e.g., deposition and polishing current statistics and timing). Together, these variables encode the cumulative effects of electrolyte aging, ion depletion, impurity accumulation, and electrode–electrolyte interactions that influence deposition behavior but cannot be fully captured by any single measurement.

The bath state is therefore treated as an engineering abstraction: it is not intended to represent a unique physical variable, but rather a predictive construct that captures the electrolyte’s capacity to support stable and repeatable deposition under given process conditions. Its validity is assessed not by direct physical observability, but by its ability to consistently predict surface-quality outcomes across different baths, aging stages, and operating scenarios.

8.2 Dataset preprocessing

Before developing the predictive framework for estimating the bath state, the open electroplating dataset was systematically refined and structured to ensure consistency, reproducibility, and analytical relevance. As discussed in the previous chapter, the dataset contains a broad collection of process parameters, electrochemical pulse descriptors, physicochemical bath properties, and surface-quality outcomes. However, not all recorded variables equally contribute to model performance or represent independent information. Therefore, a comprehensive preprocessing and feature-selection stage was implemented to isolate the most informative predictors for subsequent modeling.

8.2.1 Data cleaning

Ahead of feature engineering and model development, the dataset underwent a detailed cleaning to guarantee data integrity. Because measurements originated from multiple experimental runs, occasional missing entries or outliers were identified through inspection and, where possible, re-measured using the original raw data. This verification step eliminated inconsistencies and prevented biased training results.

For each electroplated sample, average R_a , R_q , R_z , and G_{20} , G_{60} values were calculated and used as representative outputs. These averaged quantities reduced measurement noise and provided

consistent targets for regression modeling. The resulting dataset formed the finalized analytical foundation for the subsequent model-architecture design and training procedures.

8.2.2 Feature categorization

To organize the available data into meaningful analytical groups, all measurable parameters were first classified into two overarching categories: process parameters and bath-state-related features.

a. **Process parameters** included the fundamental electroplating control variables:

- E_{dep}
- Q_{dep}
- Q_{pol}

These parameters define the direct electrochemical operating conditions governing copper deposition and dissolution during pulse–reverse cycles.

b. **Bath-state-related features** were subdivided into two subgroups:

- **Physicochemical properties**, describing the initial and evolving electrolyte condition through measurable quantities such as initial CuSO_4 concentration, electrical conductivity, and density.
- **Pulse-derived information**, representing the electrochemical response of the system during operation, including deposition and polishing currents and their respective durations, which encode dynamic feedback from the electrolyte and electrode surfaces.

This categorization (Figure 8.1) provided a structured basis for assessing which feature types most effectively capture the latent “bath state” influencing surface quality.

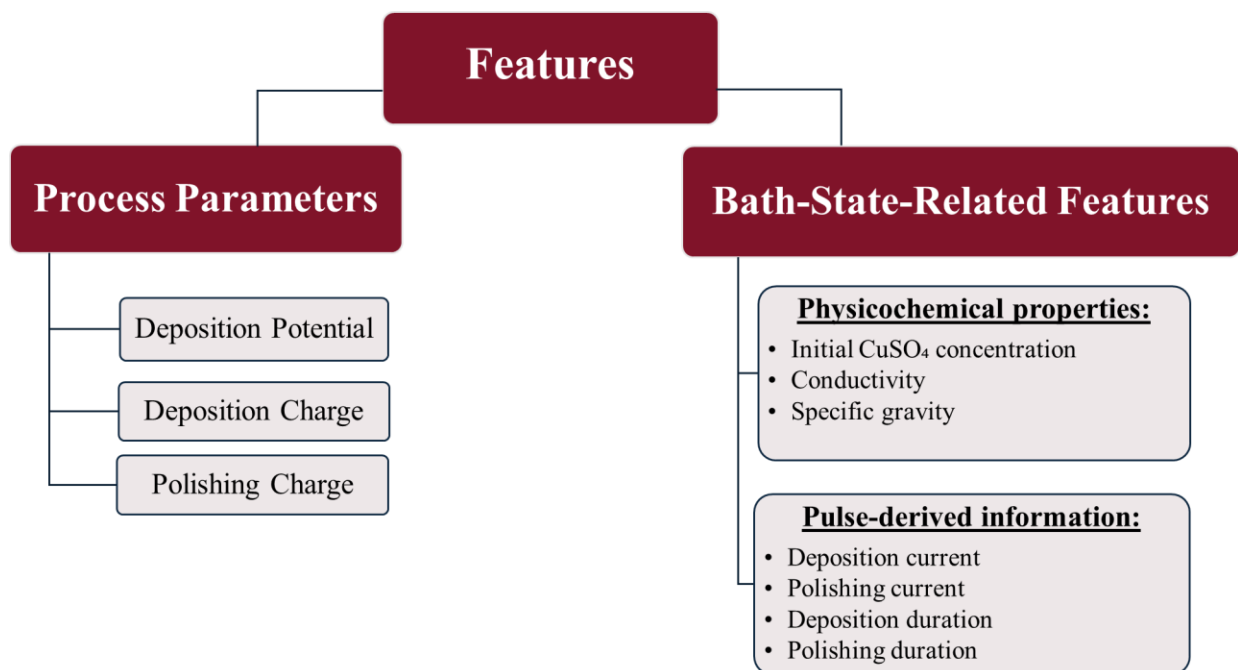


Figure 8.1 Feature categorization

8.2.3 Feature-selection workflow

Given the large number of potentially correlated or redundant variables in the open electroplating dataset, eight complementary feature-selection techniques were implemented to identify the most informative predictors for model development. Each method captures a distinct aspect of the data's statistical or structural relationship with the output variables, ensuring a balanced evaluation of both linear and nonlinear dependencies.

1. Correlation Analysis

This method quantifies the degree of linear association between each input variable and the target outputs (Ra, Rq, Rz, G20, G60). Highly correlated features were prioritized, as strong correlation often implies direct or proportional influence on surface properties. However, since correlation cannot capture nonlinear effects, it served primarily as an initial screening criterion.

2. Recursive Feature Elimination (RFE)

RFE employs an iterative process in which a predictive model (e.g., linear or tree-based) is trained repeatedly, and the least important feature at each iteration is removed. This continues until a specified number of features remain. The approach identifies a minimal yet effective

subset of variables that contribute most strongly to model accuracy, reducing dimensionality without significant loss of information.

3. Tree-Based Feature Importance (Random Forest)

Random Forest models naturally provide importance scores based on how much each feature reduces impurity (measured via Gini index or entropy) across decision-tree splits. This method captures complex nonlinear and interaction effects between variables. Features with higher average importance across all trees were considered key drivers of variability in surface outcomes.

4. Mutual Information (MI)

MI measures how much information one variable provides about another, regardless of whether the relationship is linear or nonlinear. It quantifies the reduction in uncertainty of the target variable when a given feature is known. This makes it particularly effective for detecting nonlinear dependencies that correlation analysis might overlook.

5. Lasso Regularization (L1 Penalty)

The Lasso method introduces an L1 penalty to regression coefficients, effectively shrinking less influential ones to zero. Features with nonzero coefficients after optimization are retained as the most relevant. This technique enforces sparsity in the model and helps identify variables with the strongest predictive impact, while naturally filtering out redundant or noisy features.

6. Permutation Importance

This approach measures the change in model performance after shuffling the values of a single feature while keeping all others constant. A large decrease in predictive accuracy indicates that the permuted feature was important for the model's decision process. Unlike built-in model importances, permutation importance is model-agnostic and evaluates the contribution of each feature in the final predictive context.

7. ANOVA F-Test

The Analysis of Variance (ANOVA) F-test compares the variance between groups (high vs. low target values) relative to the variance within groups. Features producing high F-scores are those whose mean values differ significantly across output categories, suggesting that they explain a meaningful portion of target variability. This statistical method complements model-based techniques with a purely inferential perspective.

8. Feature-Variance Sorting

Features with extremely low variance contain limited information and tend to have minimal predictive value. Sorting by variance allows the exclusion of near-constant variables that contribute little to model differentiation. This step ensures that only features exhibiting meaningful variability across samples are retained for modeling.

By integrating these eight techniques, the analysis captured both the linear and nonlinear relationships governing the electroplating process. The diversity of selection methods reduced bias from any single approach, leading to a robust and well-balanced identification of influential predictors.

8.2.4 Construction of input feature sets

The results obtained from the eight feature-selection methods were consolidated into six distinct input configurations. Each configuration represented a different hypothesis about which subset of variables best encapsulates the information necessary to predict surface outcomes and infer the underlying bath state. This structured approach also enabled systematic evaluation of how model performance changes with increasing data richness and complexity.

1) Top 5 Highly Correlated Features (Top 5 Corr)

This configuration targeted the most dominant predictors by selecting the five variables showing the highest absolute correlation with each surface-quality metric. The intent was to evaluate whether the strongest linear contributors alone could reproduce the observed deposition trends. Because this subset captures only the most pronounced direct dependencies, it serves as a baseline for assessing the lower bound of predictive performance, highlighting how much of the surface variation can be explained through the simplest linear relationships.

2) Top 10 Highly Correlated Features (Top 10 Corr)

Expanding beyond the strict top-five limit, this configuration incorporated the ten variables with the highest correlation coefficients. It was designed to test whether including moderately correlated predictors enhances generalization and reduces residual error without introducing excessive redundancy. By broadening the input space, the model can capture secondary relationships, those that may not appear dominant individually but collectively improve predictive stability and resilience to measurement noise.

3) Features Appearing in ≥ 4 of the 8 Selection Methods (Most Freq)

This intersection-based configuration aimed to isolate universally influential predictors, features that repeatedly emerged as important across at least half of the applied selection methods. Because each technique emphasizes different statistical or structural relationships, consistent appearance in four or more methods implies that the feature's relevance is robust and not dependent on a particular algorithmic bias. This subset then represents a method-independent consensus on the most reliable indicators of bath condition and surface response.

4) Process Parameters Only (Process Params)

This configuration included only the controllable electroplating settings: E_{dep} , Q_{dep} , and Q_{pol} . Excluding all bath or pulse-derived variables, it provided a benchmark for determining how much of the observed surface variation originates purely from the operator-controlled process

window. Evaluating model accuracy with this reduced input space quantified the predictive power of the process itself, independent of electrolyte condition or system feedback.

5) Process + Bath Parameters (Proc+Bath)

Here, the process parameters were augmented with measurable physicochemical descriptors of the electrolyte—conductivity, density, and CuSO_4 concentration. This hybrid configuration enabled the examination of how bath composition and ionic properties influence deposition quality when considered alongside electrochemical control variables. It also provided a bridge between experimental observables and the inferred bath state, allowing the model to learn interactions between process conditions and evolving electrolyte chemistry.

6) Process + Current Parameters (Proc+current)

This configuration integrates the fundamental process parameters (E_{dep} , E_{pol} , Q_{dep} , Q_{pol}) with pulse-level electrochemical responses, specifically the average deposition and polishing currents and their associated pulse durations. Unlike static bath descriptors such as conductivity or density, these current-based features reflect how the electrolyte behaves during the plating process itself. Because the current–time response reacts immediately to changes in ionic mobility, mass-transport limitations, local depletion, and counter-electrode condition, these features carry dynamic information that cannot be captured by physicochemical measurements alone.

The purpose of this configuration was to evaluate whether incorporating these real-time electrochemical signals improves prediction accuracy relative to using process parameters combined only with bath measurements (Proc + Bath). In other words, it tests the hypothesis that dynamic pulse behavior provides a more sensitive and informative window into the effective bath state than static measurements taken before plating. If predictive performance increases under this configuration, it would indicate that bath evolution, electrode condition, and ion-transport constraints are more effectively captured through current responses than through conventional bath characterization alone.

Thus, the Process + Current feature set serves as the most diagnostic and mechanistically meaningful configuration, designed not only to enrich the input space but to determine whether electrochemical feedback during plating can outperform traditional bath metrics in predicting surface quality and inferring bath state.

Together, these six configurations provided a hierarchical framework for model development. By progressively incorporating additional layers of process, bath, and electrochemical information, the subsequent analysis was able to evaluate how each class of features contributes to predicting surface quality and, ultimately, to inferring the electrolyte bath state.

8.2.5 Data splitting and normalization

For model development, the dataset was divided into training and testing subsets using an 80/20 split. In each iteration, 80% of the data was used to train the model, while the remaining 20%, kept completely unseen during training, served as the test set to evaluate predictive performance. This approach allowed the models to be assessed on genuinely new conditions drawn from across the full dataset rather than on data they had already learned from. Using randomized stratified sampling ensured that the distribution of surface outcomes remained consistent between the training and testing sets, preventing over-representation of any particular bath condition or process regime.

To avoid chance-based or biased conclusions tied to a single random split, this entire procedure was repeated 1,000 times. Each repetition generated a different combination of training and testing subsets, allowing the calculation of stable error distributions rather than a single-point estimate. This repetition strategy made it possible to observe how the model performs across many unseen subsets of data, capturing the natural variability present in the dataset and providing a more rigorous assessment of model reliability and generalization capability.

All input variables were standardized before training to eliminate scale disparities among parameters such as conductivity, current, density, and pulse durations. This normalization prevented high-magnitude features from dominating the optimization process and ensured that each variable contributed proportionally during learning. Together, these steps produced a robust, unbiased evaluation framework that accurately reflects how well each model can predict surface properties under previously unseen electroplating conditions.

8.3 Model Architecture Design

This section explains the full modeling workflow used to evaluate how different feature sets influence predictive accuracy for surface-quality outputs. Building on the feature-selection analysis from Section 2, six distinct input configurations were used to train and compare multiple regression models. The objective was to determine which models and feature subsets most effectively capture the relationships between electroplating conditions and resulting surface morphology and brightness, and by extension, which features best encode the evolving bath state.

8.3.1 Output parameter definition

The target variables for prediction were the averaged surface-quality measurements obtained for each sample: Ra, Rq, Rz, G20, and G60. As established in the Methodology chapter, each surface metric was measured multiple times on each electroplated sample, and the mean of these repeated measurements was used as the representative output value.

This averaging procedure reduces noise arising from localized surface irregularities or measurement variability and yields a consistent, statistically reliable output for model training.

Importantly, the modeling strategy followed a single-output regression approach. Each surface parameter (e.g., R_a) was predicted independently using the selected input features. No multi-output or joint-learning framework was applied. This allowed a focused evaluation of how each feature subset and model architecture performs for each specific mechanical or optical descriptor and ensured that the trained models learned only the relationships relevant to one target at a time.

The goal of this structured approach was to:

- Quantify how each model and feature configuration influences prediction accuracy,
- Determine which model–feature combinations carry the most predictive information for each physical property.

8.3.2 Feature importance analysis

To better understand the relationships between input variables and surface responses, two complementary feature-importance perspectives were shown for every target output (Figure 8.2): (1) the Top 10 highly correlated features (Top 10 Corr) and (2) the features appearing in at least four of the eight feature selection methods (Most Freq). These two perspectives represent, respectively, the strongest linear associations and the most consistently important predictors across linear, nonlinear, and model-based frameworks.



Figure 8.2 Two complementary feature-importance perspectives for each target output: 1) the Top 10 highly correlated features and 2) the features appearing in at least four of the eight feature selection methods.

From these analyses, it can be understood that:

For the roughness outputs (Ra, Rq, and Rz):

- The Top 10 correlated features were dominated by pulse-level current, particularly $\text{max_i}_{\text{dep_pulse}}$ and $\text{min_i}_{\text{pol_pulse}}$. These variables directly reflect the instantaneous electrochemical conditions during each cycle and therefore strongly influence the final texture of the deposit. Process parameters did not contribute equally to the roughness outputs: the E_{dep} showed a strong linear association with roughness and consistently appeared among the most influential predictors, whereas parameters such as Q_{pol} exhibited much weaker linear correlation in the Top-10 lists. This is consistent with the electrochemistry of the system. The cathodic potential directly controls nucleation rate, grain growth, and the onset of hydrogen evolution, all of which strongly influence the formation of surface asperities and therefore Ra, Rq, and Rz ¹⁰⁹.
- The polishing charge was explored over a narrower range, and its effect is more indirect: it primarily refines or slightly erodes features that have already formed during deposition and may act through more nonlinear or interaction-type relationships, which are not fully captured by simple correlation ranking.
- Bath properties, such as specific gravity or conductivity, also tended to appear with lower linear importance for roughness. However, their influence is still reflected in the pulse behaviour itself and can emerge more clearly in the multivariate, nonlinear feature-selection methods.
- The “Most Frequent” feature sets similarly highlighted a blend of process parameters and pulse-derived features. Variables such as E_{dep} , Q_{pol} , $\text{max_i}_{\text{dep_pulse}}$, $\text{max_i}_{\text{pol_pulse}}$, and $\text{min_i}_{\text{pol_pulse}}$ appeared repeatedly across several of the nonlinear and model-based selection methods.
- Bath properties such as conductivity also emerged consistently, even though they showed weaker linear correlations. This pattern reinforces the earlier observation that certain aspects of the bath condition are not captured well through simple linear correlation, yet become clearly detectable when nonlinear interactions and multivariate dependencies are considered. Their presence is fully consistent with electrochemical principles: copper deposition uniformity is shaped not only by the externally imposed potentials and charges but also by the evolving ionic environment of the electrolyte ¹²²⁻¹³⁰, which manifests through the real-time pulse-current response.

For G20:

- The Top-10 correlation plot shows that $\text{min_i}_{\text{dep_pulse}}$ has the highest linear association with gloss, followed closely by conductivity and the $\text{avg_i}_{\text{dep_pulse}}$. CuSO_4 concentration and

avg_i_{pol_pulse} also exhibit correlations, while specific gravity and E_{dep} appear with slightly lower, but still non-negligible values.

- The “Most Frequent” list for **G20** reinforces this interpretation. Conductivity is the most frequently selected variable across all feature-selection methods, and deposition-pulse statistics appear almost as consistently. Polishing current descriptors and pulse-duration features are also present, but with slightly lower counts.
- These results suggest that **G20** is particularly sensitive to how an ion-rich bath translates into smooth, well-controlled deposition pulses: the bath sets the electrochemical capacity through conductivity and CuSO₄ concentration, while the current waveform determines how effectively that capacity is used to level the surface.

For G60:

- The Top-10 correlation results emphasize deposition-pulse parameters even more strongly. The min_i_{dep_pulse} and avg_i_{dep_pulse} again dominate, with the avg_i_{pol_pulse} and CuSO₄ concentration following, and conductivity, E_{dep}, polishing-current extremes, and Q_{pol} contributing at a more moderate level. Compared with G20, G60 appears slightly more driven by the overall deposition behaviour and somewhat less by fine differences in bath composition, which is reasonable because G60 captures broader specular reflection and is therefore more influenced by large-scale surface leveling than by the finest micro-features.
- The “Most Frequent” diagram confirms this trend: the avg_i_{dep_pulse} and conductivity are the most recurrent features, closely followed by E_{dep}. In combination, these patterns indicate that high G60 gloss arises when the bath maintains good conductivity but, crucially, when the deposition phase delivers a stable, appropriately sized current over the entire plating time.

Collectively, these observations show that pulse-level features consistently appeared across both feature-importance analyses for all roughness outputs, while bath properties gained prominence as predictors of gloss. This justified the construction of the Proc + Current feature set using only pulse features that appeared in the “Most Frequent” category, ensuring that the included current descriptors were genuinely among the most influential predictors for each output. Similarly, the strong role of bath chemistry in gloss prediction supported the creation of the Proc + Bath configuration as a meaningful comparative baseline.

8.3.3 Model training

Five regression models were trained and evaluated across all six feature sets to determine how effectively each modeling approach could learn the nonlinear electrochemical relationships governing surface-quality outcomes. These models (Linear Regression, KNN, SVR, Random

Forest, and GPR) cover a wide methodological range, from simple parametric models to flexible nonparametric and probabilistic frameworks. This diversity allowed a systematic assessment of how different algorithmic assumptions interact with the structure of the electroplating data.

Linear Regression was included as a baseline. It assumes that the relationship between inputs and outputs can be represented as a weighted sum of features, making it highly interpretable. Its primary value lies in its simplicity and its ability to reveal whether a target exhibits any strong linear signal at all. However, there is a lack of ability to capture the nonlinearities inherent in electrochemical systems.

K-Nearest Neighbors (KNN) represents the opposite modeling philosophy. It makes no assumptions about functional form and instead relies entirely on the similarity of data points in the feature space. In principle, this allows KNN to model highly nonlinear electrochemical behavior by simply "copying" patterns from similar historical observations. In practice, however, KNN suffers from two major limitations: (1) its performance degrades in high-dimensional spaces where distance metrics become less meaningful, and (2) prediction requires scanning the entire dataset, making it computationally slow and unsuitable for real-time monitoring. For electroplating applications (where rapid decisions or classifications may be needed), KNN is therefore less practical despite its conceptual flexibility.

Support Vector Regression (SVR) provides a more structured approach to nonlinearity. By using kernel functions such as the radial basis function (RBF), SVR maps inputs into a higher-dimensional space where nonlinear relationships become approximately linear. This enables SVR to capture subtle interactions between bath properties, process parameters, and pulse-current behaviors. SVR is also robust to outliers due to the margin-based optimization. The trade-offs are its relatively high computational cost during training and the need for careful hyperparameter tuning (C , ϵ , and kernel width). While SVR is highly effective for batch predictions or offline optimization tasks, it is less ideal for deployment in fast-feedback control systems requiring high-frequency predictions.

Gaussian Process Regression (GPR) models the target function as a distribution over possible functions, producing not only predictions but also uncertainty estimates. This is particularly valuable for electroplating, where predictions may need to be accompanied by confidence intervals to guide process monitoring or adaptive control strategies. GPR is capable of fitting smooth trends and capturing nonlinearities through kernel design. However, its computational cost scales poorly with dataset size, making it impractical for large-scale or real-time applications. It is therefore best suited for offline analysis, model validation, or uncertainty-aware process optimization.

Random Forest demonstrated the most advantageous combination of characteristics for electroplating. Composed of many decision trees trained on bootstrapped samples, it learns a diverse set of nonlinear relationships without assuming any specific functional form. The ensemble averaging process naturally reduces overfitting, and the model remains effective even when faced with noisy, partially correlated, or partially redundant features (conditions common in pulse-electroplating data). Random Forest provides fast inference, making it suitable for real-time or near-real-time monitoring. Additionally, its built-in feature-importance metrics make it especially

valuable for interpreting which variables encode the bath state and how they influence surface outcomes. These strengths make Random Forest not only accurate but also operationally practical.

Each of these models was trained using a robust 1,000-iteration evaluation strategy. In every iteration, the dataset was partitioned into an 80% training set and a 20% testing set, with the test set kept entirely unseen during training. This repeated random sub-sampling ensured that performance metrics reflected true generalization and were not biased by any single split or particular distribution of bath conditions. Predictive accuracy was quantified using the relative mean absolute error (MAE divided by the mean of the target), which normalized errors across outputs of different magnitudes and enabled fair comparison across models and feature sets.

8.3.4 Hyperparameter tuning

To ensure that each model performed under its optimal configuration, a dedicated hyperparameter-tuning stage was carried out before comparing the predictive accuracy of the feature sets. The algorithms deployed in this study depend strongly on hyperparameters that govern model flexibility, smoothness, and generalization capacity. Selecting these parameters appropriately is essential, as sub-optimal values can either cause the model to underfit (failing to capture the complexity of the electrochemical system) or overfit (memorizing noise in the data and losing predictive power on unseen conditions).

Hyperparameter tuning was completed using a repeated K-fold cross-validation strategy applied to the training portion of the data. In this procedure, the training dataset was partitioned into k folds; the model was trained k times, each time leaving out one fold for validation. This entire k-fold process was then repeated multiple times with different random shuffles, producing a robust estimate of the model's true cross-validated performance for each hyperparameter candidate. This approach is well-suited for electroplating data, where bath evolution introduces inherent variability, and a single train/validation split may not be sufficient to estimate a model's true generalization ability.

During tuning, each hyperparameter value was scored based on the average validation error across all repeated folds. The hyperparameter setting with the lowest validation error was then selected for use in the final 1,000-iteration training–testing evaluation described in Section 8.3.3.

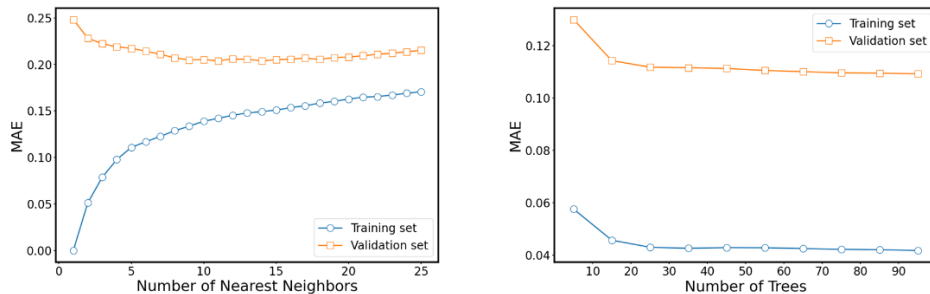
The behavior of the models during tuning followed the expected theoretical patterns. For KNN, the number of neighbors (K) acted as the primary control on model flexibility. Very small values of K led to extremely low training error because the model effectively memorized the training instances; however, the validation error was high because such a model generalizes poorly and is overly sensitive to noise. As K increased, the validation error decreased initially, as the model gained smoothing power, and then eventually plateaued or began to rise again once excessive averaging caused underfitting. This progression created a clear, easily identifiable region where the validation error reached its minimum, enabling the selection of the optimal K value that balanced stability with sufficient flexibility to capture the underlying electrochemical relationships.

For Random Forest, the number of trees played a similar role. When the forest contained only a few trees, both training and validation errors were high, reflecting insufficient model complexity. As the number of trees increased, the validation error consistently decreased and stabilized, while the training error quickly reached a low plateau. This behavior is expected because larger forests reduce variance through bagging and generally improve predictive stability. Importantly, after a certain number of trees, the validation error no longer improved significantly, indicating convergence of ensemble performance and helping identify the point beyond which additional trees yield diminishing returns while increasing computational cost.

For SVR, hyperparameters such as the regularization constant (C), the insensitive margin (ϵ), and the kernel width (γ for RBF kernels) were tuned jointly. Smaller values of C produced smoother, more regularized models that tended to underfit, whereas very large C values risked overfitting the training data. Similarly, the kernel width governed how locally or globally the model responded to variations in the input features. Validation curves revealed the parameter ranges where the balance between smoothness and flexibility produced the lowest cross-validated error.

Gaussian Process Regression requires selecting the kernel and tuning the kernel hyperparameters that determine the characteristic length scales of the model. Short length scales allow the model to track small variations closely (risking overfitting) while long length scales enforce smoother functions.

Figure 8.3 presents the results of the repeated K-fold cross-validation used for hyperparameter tuning of the KNN, Random Forest, and SVR models for predicting Ra. These plots demonstrate that each model exhibits the expected tuning behaviour, with performance changing systematically as the hyperparameters are varied and converging toward the ranges that yield the most accurate predictions for this output.



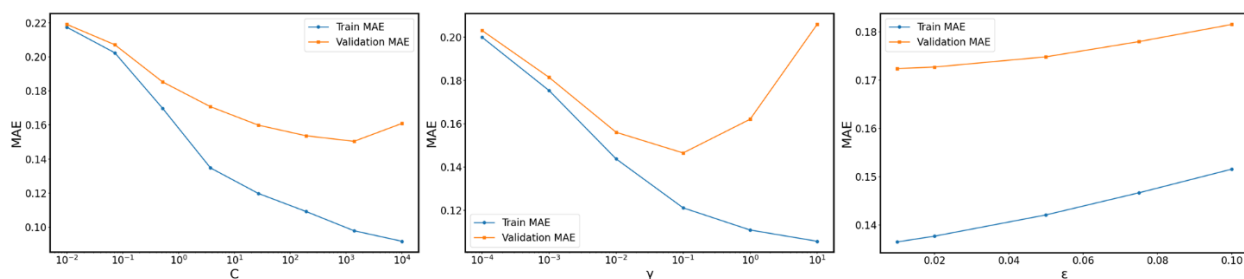


Figure 8.3 Results of repeated K-fold cross-validation for hyperparameter tuning of the KNN (top left), Random Forest (top right), and SVR (bottom)

8.4 Model Selection

To compare the predictive capability of the different machine-learning models and feature configurations, the relative MAE distributions obtained from the 1,000 randomized train–test splits were first visualized (figures 8.4–8.8) for each output variable (Ra, Rq, Rz, G20, and G60). For every combination of model and feature set, a box-and-whisker plot was generated to show the complete distribution of relative MAE values obtained across the 1,000 randomized train–test splits. These visualizations report not only the median accuracy of each method but also the interquartile range, the overall spread of errors, and the presence of outliers, enabling a clear evaluation of both prediction stability and central tendency. A horizontal red dotted line was added to each plot to indicate the minimum practically achievable error for that output, calculated from the intrinsic measurement limitations of the instruments: for gloss (G20 and G60), this line corresponds to the device’s stated measurement accuracy, whereas for roughness parameters (Ra, Rq, Rz) it reflects the standard deviation of repeated profilometry measurements. This reference line provides context for how close each model–feature combination approaches the physical lower limit of prediction accuracy.

For each target output, five separate graphs are presented, one for each regression model (Linear Regression, KNN, SVR, Random Forest, and GPR). Within every graph, six boxplots appear, corresponding to the six feature configurations evaluated in this study (Process Parameters, Process + Bath, Process + Current, Top-5 Correlated, Top-10 Correlated, and Most Frequent). This structure allows direct visual comparison of model behaviour under different informational inputs, making it possible to determine both which feature sets carry the most predictive value and which model architectures exhibit the most consistent and reliable performance across varying electrochemical conditions.

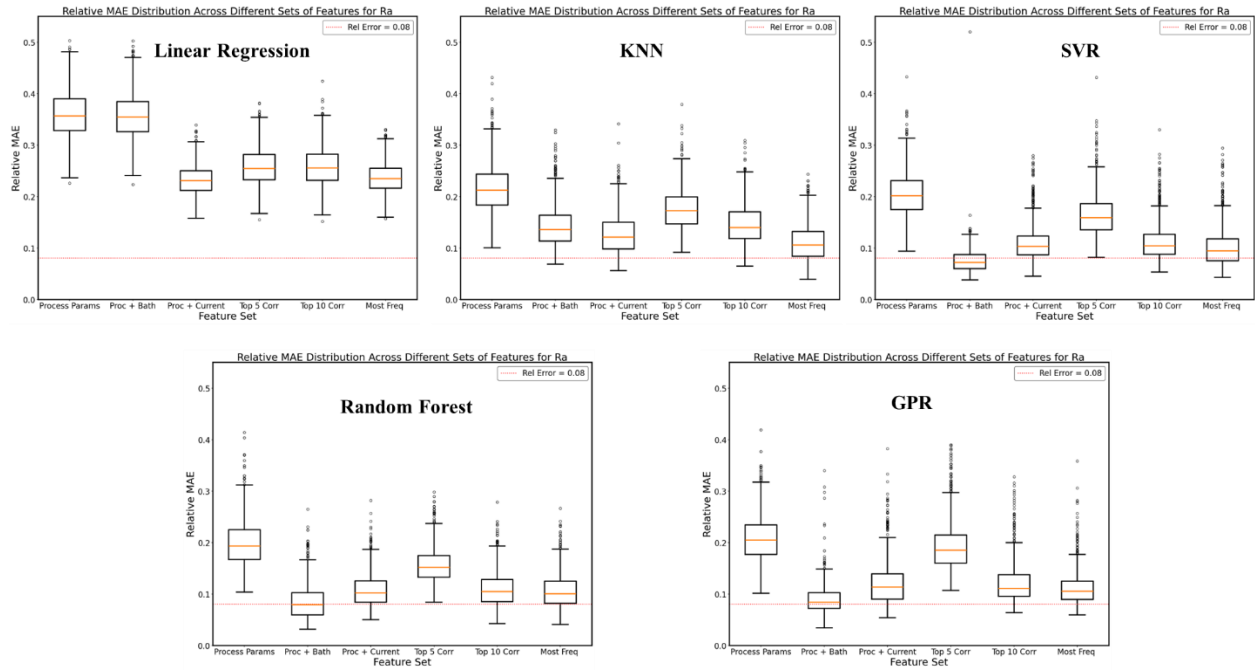


Figure 8.4 The relative MAE distributions obtained from the 1,000 randomized train–test splits for five models with six feature categorizations in the prediction of Ra

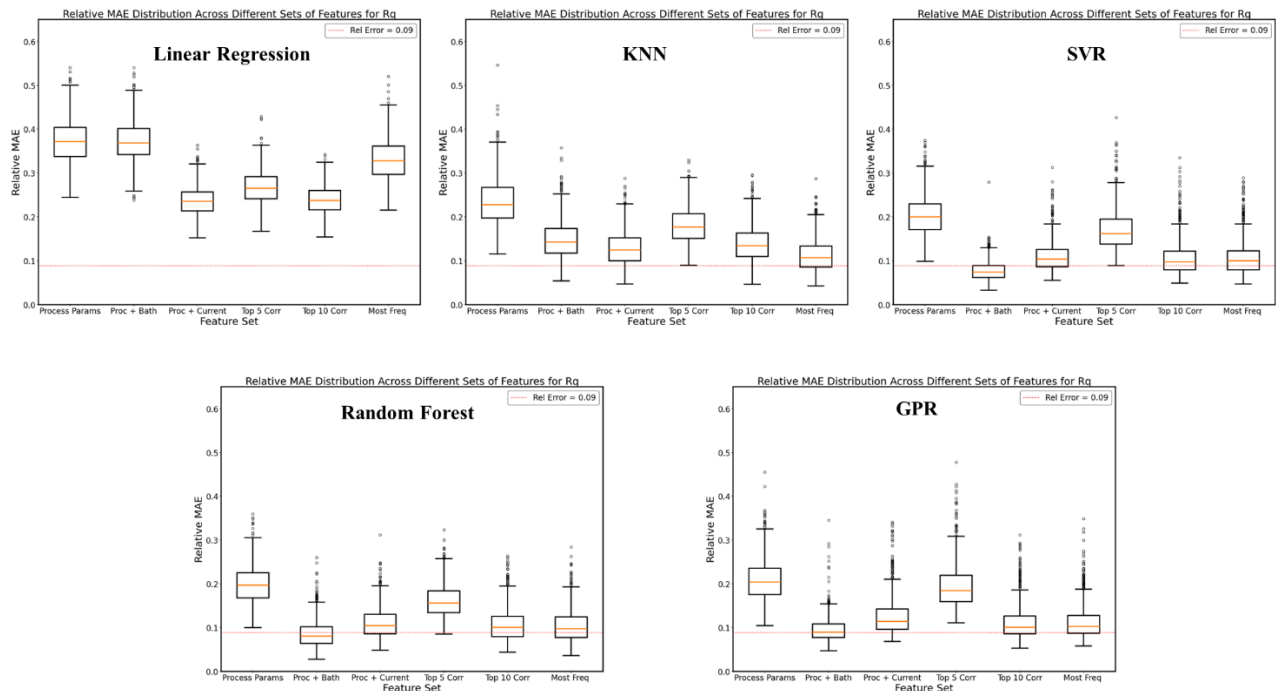


Figure 8.5 The relative MAE distributions obtained from the 1,000 randomized train–test splits for five models with six feature categorizations in the prediction of Rq

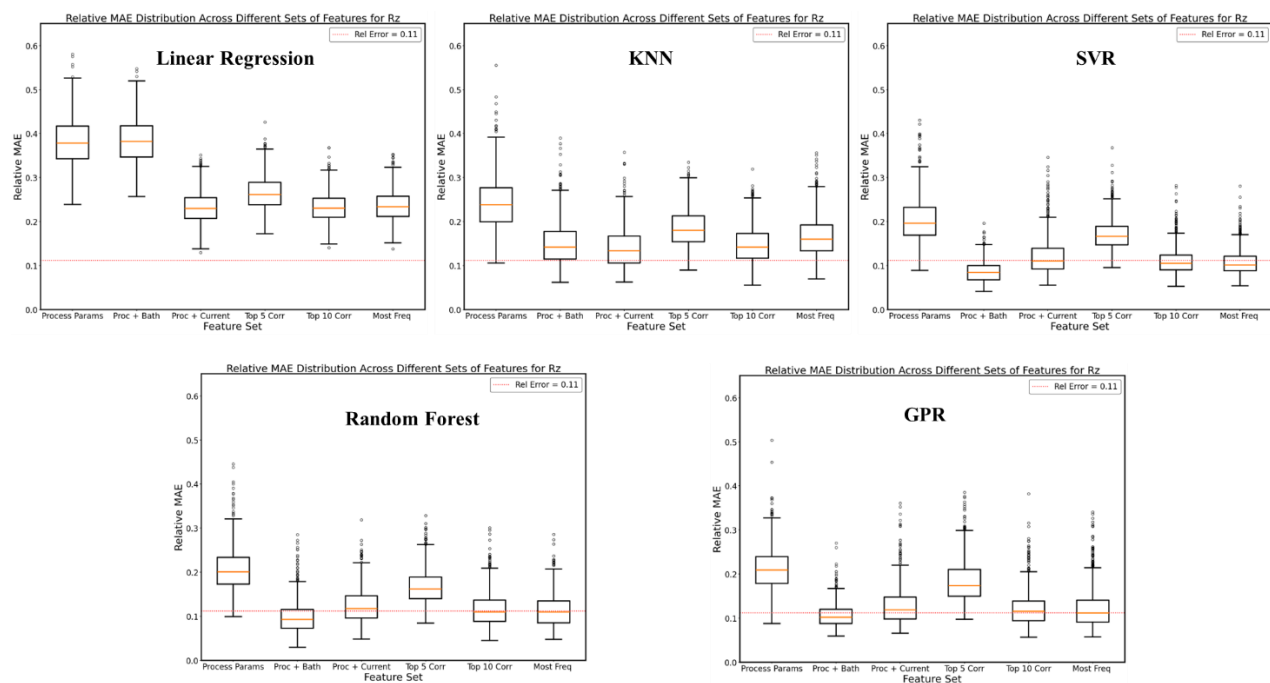


Figure 8.6 The relative MAE distributions obtained from the 1,000 randomized train–test splits for five models with six feature categorizations in the prediction of Rz

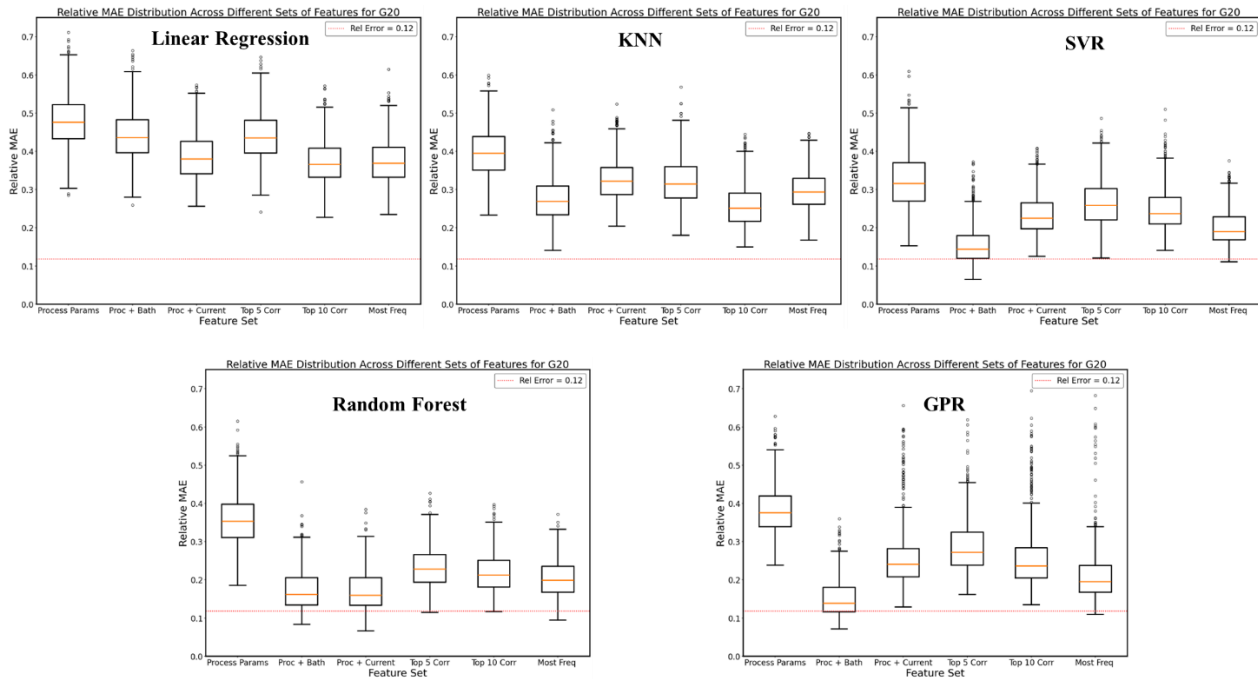


Figure 8.7 The relative MAE distributions obtained from the 1,000 randomized train–test splits for five models with six feature categorizations in the prediction of G20

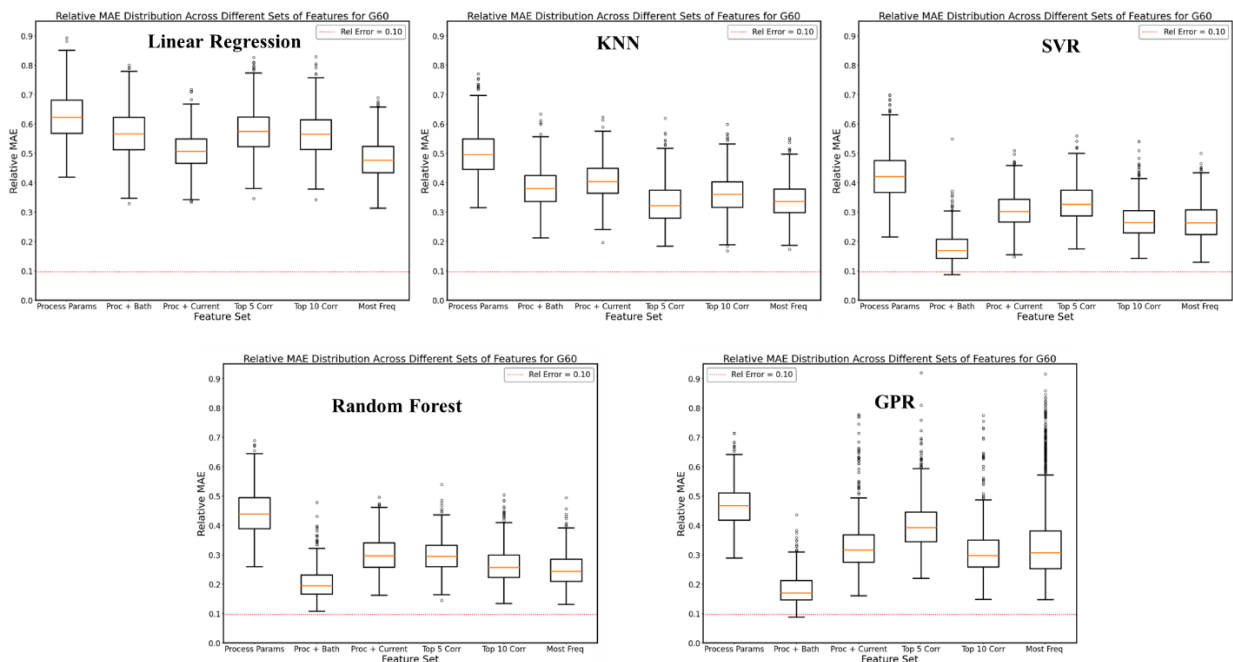


Figure 8.8 The relative MAE distributions obtained from the 1,000 randomized train–test splits for five models with six feature categorizations in the prediction of G60

- *Model-Comparison Analysis for Ra, Rq, and Rz:*

The model comparison results for Ra, Rq, and Rz reveal several consistent patterns in terms of predictive accuracy, stability, and sensitivity to feature selection. Across all three roughness outputs, Linear Regression and KNN exhibit visibly larger relative MAE values and wider spreads in their boxplots, indicating limited ability to capture the nonlinear and interaction-driven behavior that governs surface-height variability in pulse electroplating. Linear Regression produces the broadest distributions, especially for the simpler feature sets, confirming that roughness formation cannot be adequately represented through purely linear combinations of process variables or bath descriptors. Even so, Linear Regression shows noticeable improvement when trained on richer inputs such as Process + Current, Most Frequent, and the Top-10 Correlated feature sets; however, its median relative error remains substantially higher than that of the nonlinear models, demonstrating that improved feature sets cannot compensate for its inability to model complex electrochemical relationships.

KNN achieves somewhat lower median errors than Linear Regression, but still shows substantial scatter, particularly when using only the Process Parameters feature set. Its sensitivity to noise and to the structure of the feature space becomes evident in the wide variability observed in its predictions. Nonetheless, the addition of more informative variables significantly enhances its performance. When trained on Process + Bath, KNN's accuracy improves by approximately 25%, and when trained on Process + Current, Most Frequent, or Top-10 Correlated feature sets, its accuracy increases by roughly 30%. Despite these gains, the error levels remain well above the minimum practically achievable threshold defined by measurement uncertainty, indicating that KNN (even with optimized feature configurations) cannot fully capture the nuances of bath-dependent roughness evolution.

In contrast, SVR, Random Forest, and GPR show a clear and consistent advantage over the linear models. Across all three roughness outputs, these nonlinear approaches exhibit substantially lower median errors and visibly tighter distributions across nearly all feature configurations. One of the most important trends emerging from the graphs is the strong dependence of model accuracy on the informational richness of the feature set. When only the Process Parameters are used, all three nonlinear models still outperform Linear Regression and KNN, but their accuracy remains modest and far from the experimental error limit. However, once pulse-derived current features are introduced (Process + Current), the predictive accuracy improves dramatically (by roughly 50–60%) compared with using Process Parameters alone. This confirms that roughness formation is strongly influenced by the real-time electrochemical dynamics encoded in current behavior.

An even greater improvement is observed when process parameters are combined with bath properties (Process + Bath). For Ra, Rq, and Rz, SVR, Random Forest, and GPR all reach their lowest median errors under this configuration, frequently approaching the minimum achievable error indicated by the red dotted line. This suggests that the physicochemical state of the bath, particularly conductivity, density, and CuSO₄ concentration, adds meaningful, complementary information that enhances the model's ability to infer the underlying deposition conditions. The Most Frequent and Top-10 Correlated feature categories produce performance levels nearly identical to those obtained with Process + Current, further reinforcing the idea that roughness depends on a combination of dynamically responsive and statistically consistent predictors.

Random Forest and GPR maintain highly stable behavior across all enriched feature sets, with tight boxplots and minimal sensitivity to outliers. SVR performs comparably well, though with a slightly stronger dependence on feature selection and tuning. Overall, the results demonstrate that nonlinear models systematically outperform linear ones for roughness prediction, and that among the six feature configurations, Process + Bath provides the highest accuracy, followed by Process + Current, Top-10 Correlated, Most Frequent, Top-5 Correlated, and finally Process Parameters. This hierarchy confirms that roughness is governed by both the imposed electrochemical inputs and the evolving physicochemical state of the bath, and that only nonlinear models are capable of faithfully capturing these interactions.

- *Model-Comparison Analysis for G20 and G60*

The predictive performance trends observed for the gloss outputs (G20 and G60) show the same overall structure as the roughness results, but with several distinctions that highlight the different physical sensitivities of optical surface response. For both gloss outputs, Linear Regression consistently produces the highest relative MAE values and the widest variability among the models, particularly when trained on the simplest feature sets such as Process Parameters or even Correlated features. The widespread occurrence of errors in these cases confirms that gloss cannot be reliably predicted from linear combinations of input features alone. Nonetheless, Linear Regression demonstrates modest improvement when trained on richer inputs. The Process + Current, Process + Bath, Most Frequent, and Top-10 Correlated feature sets noticeably lower their median error, although the performance remains well above the measurement accuracy threshold and significantly inferior to that of the nonlinear models.

KNN follows a similar pattern. When trained only on Process Parameters, its performance is poor and exhibits large dispersion in prediction error. However, as more informative features are introduced, KNN's predictive accuracy improves substantially. With Process + Current, the error decreases by approximately 25% for G20 (20% for G60), and with Process + Bath, Most Frequent, or Top-10 Correlated feature sets, the improvement becomes even more pronounced. Despite these gains, the KNN predictions never reach the minimum practically achievable error, indicating that the local-distance-based structure of KNN is not ideally suited for capturing the optical response governed by complex interactions between surface microgeometry, mass transport, and localized current distribution.

The nonlinear models (SVR, Random Forest, and GPR) again show a much stronger capability to model gloss behavior. SVR exhibits the most dramatic improvements when current and bath inputs are used compared to when the process parameters are used alone. For both G20 and G60, the Process + Bath configuration yields the lowest median errors and compressed distributions, placing the predictions closest to the measurable lower bound indicated by the red dotted line. This strong performance aligns with the fact that gloss is sensitive not only to instantaneous pulse dynamics but also to the bulk ionic environment, which directly influences deposit compactness and reflectivity. SVR's ability to model nonlinear and multiparametric relationships enables it to exploit these mixed influences effectively. For the rest of the feature sets, the behaviour is the same for both G20 and G60.

Random Forest also delivers robust, stable performance across all feature sets. Its accuracy improves sharply when pulse features are included, and it reaches its best performance under the Process + Bath feature set. It achieves this reliability without requiring extensive hyperparameter tuning and trains significantly faster than SVR or GPR. This combination of stable performance, low computational cost, and minimal configuration effort makes Random Forest the most practical choice for fast, repeatable gloss prediction. For G20, the Process + Current model configuration performs nearly as well as Process + Bath, suggesting that the dynamic electrical response carries significant information about the evolving reflective quality of the surface.

GPR shows strong performance as well, producing low median errors and benefiting substantially from the richer feature configurations. As with SVR and Random Forest, the Process + Bath feature set consistently results in the smallest prediction errors. However, the broader spread in the GPR boxplots (especially for G60) indicates higher sensitivity to kernel tuning and potential numerical instability when operating near the gloss measurement boundaries. This is especially noticeable for the simpler feature sets, where the increased variance reflects insufficient regularization for describing the complex optical response.

A consistent hierarchy emerges from the G20 and G60 results: Process + Bath is the most informative feature configuration, followed by Process + Current, Top-10 Correlated, Most Frequent, Top-5 Correlated, and finally Process Parameters alone. This structure reinforces the physical understanding that gloss depends on both the macroscopic ionic environment of the electrolyte and the instantaneous current waveforms that modulate surface quality at micro- and submicron scales. Moreover, the results demonstrate that nonlinear models are essential for capturing these relationships; the linear models remain far from the minimum achievable gloss error even under the richest feature configurations.

Together, these findings confirm that gloss prediction, like roughness prediction, is dominated by nonlinear, multivariate relationships. SVR, Random Forest, and GPR consistently outperform Linear Regression and KNN, with Random Forest providing the most stable and interpretable performance across varying feature sets and test splits. This makes Random Forest the most balanced and practically advantageous model for predicting gloss in real or near-real-time electroplating applications.

8.5 Conclusion

- This chapter establishes a complete framework for data-driven bath-state estimation. By integrating rigorous feature selection, nonlinear modeling, and systematic cross-validation, the workflow developed here provides a scalable methodology for predicting surface quality and inferring hidden electrolyte conditions.
- Across all outputs, the analysis shows that roughness is governed primarily by pulse-current behaviour, with E_{dep} and Q_{pol} emerging as the only process parameters showing consistently strong influence. Bath properties such as conductivity contribute more subtly, becoming important only when nonlinear, multivariate interactions are considered. Gloss

outputs also depend heavily on pulse-derived features, but with conductivity and E_{dep} playing a stronger role in shaping deposition uniformity and optical response.

- Nonlinear models are essential for capturing the electrochemical complexity of copper electroplating. Support Vector Regression, Random Forest, and Gaussian Process Regression achieved substantially lower errors and narrower distributions than Linear Regression or KNN across all surface outputs. These nonlinear approaches handled the multiparametric dependencies among bath chemistry, process settings, and electrochemical dynamics, whereas the linear models systematically failed to reach the minimum practically achievable error.
- The Process + Bath feature configuration delivered the highest predictive accuracy for both roughness and gloss. Among the six feature sets, combining process parameters with physicochemical bath descriptors consistently produced the smallest relative MAE values, often approaching the experimental measurement limit. This confirms that static bath properties carry essential global information that complements the dynamic behavior captured in pulse responses.
- Process + Current and statistically derived feature sets provided similar high performance, confirming their diagnostic power. Models trained on pulse-current features (Process + Current), Top-10 Correlated variables, and Most-Frequent features achieved accuracy nearly equivalent to the Process + Bath configuration. This demonstrates that the evolving bath state is encoded not only in measurable parameters such as conductivity and CuSO_4 concentration but also in the instantaneous electrochemical response recorded during deposition.
- Random Forest, SVR, and GPR often achieved comparable accuracy. Random Forest delivers this level of performance without heavy hyperparameter tuning and with far shorter training times than SVR or GPR. This efficiency, together with its dependable accuracy, makes Random Forest the most practical model for rapid and repeatable gloss prediction. Its built-in feature-importance measures further make it ideal for interpreting which variables encode the bath state and how they influence surface outcomes.
- The hierarchical performance trend confirms that surface quality is governed by nonlinear, multivariate interactions. For all outputs, the ordering of predictive power, Process + Bath > Process + Current $\approx$ Top-10 Corr $\approx$ Most Freq > Top-5 Corr > Process Parameters, demonstrates that richer and more physically informative feature sets improve model accuracy. This hierarchy provides a quantitative basis for defining which measurements should be prioritized in future electroplating monitoring systems.

9. The search for process parameters

9.1 Introduction

This chapter examines how electroforming process parameters can be adjusted in response to bath aging using the predictive models developed in the previous chapter. The aim is to establish practical, data-driven relationships between controllable process parameters and resulting surface properties, relationships that remain usable even as the electrolyte chemistry evolves.

For this purpose, three bath states (fresh, semi-aged, and aged) were defined for each of the four CuSO_4 concentrations. Within each bath and aging condition, three representative process scenarios were explored: varying E_{dep} at fixed Q_{dep} and Q_{pol} , varying Q_{dep} at fixed E_{dep} , and varying Q_{pol} at fixed deposition conditions. These scenarios were chosen to demonstrate how a trained ML model can be used to adjust process settings based on desired surface properties as the bath drifts away from its original state.

The Random Forest models from Chapter 6 were applied to predict Ra, G20, and G60 across all combinations of bath state, bath concentration, and scenario. Spline smoothing was used to visualize the predicted trends and derive interpretable parameter–response curves. Additional robustness tests were performed by examining the effect of incorrect CuSO_4 inputs and by evaluating the model’s ability to generalize to unseen sulfuric acid concentrations (0.5–3 M), despite being trained only on 1 M.

Overall, this chapter demonstrates how the ML framework can assist in tuning process parameters under varying bath conditions, providing a practical foundation for adaptive, data-informed electroforming control within the chemically and operationally bounded domain determined in this study.

9.2 Scenario-driven prediction of surface properties across bath states

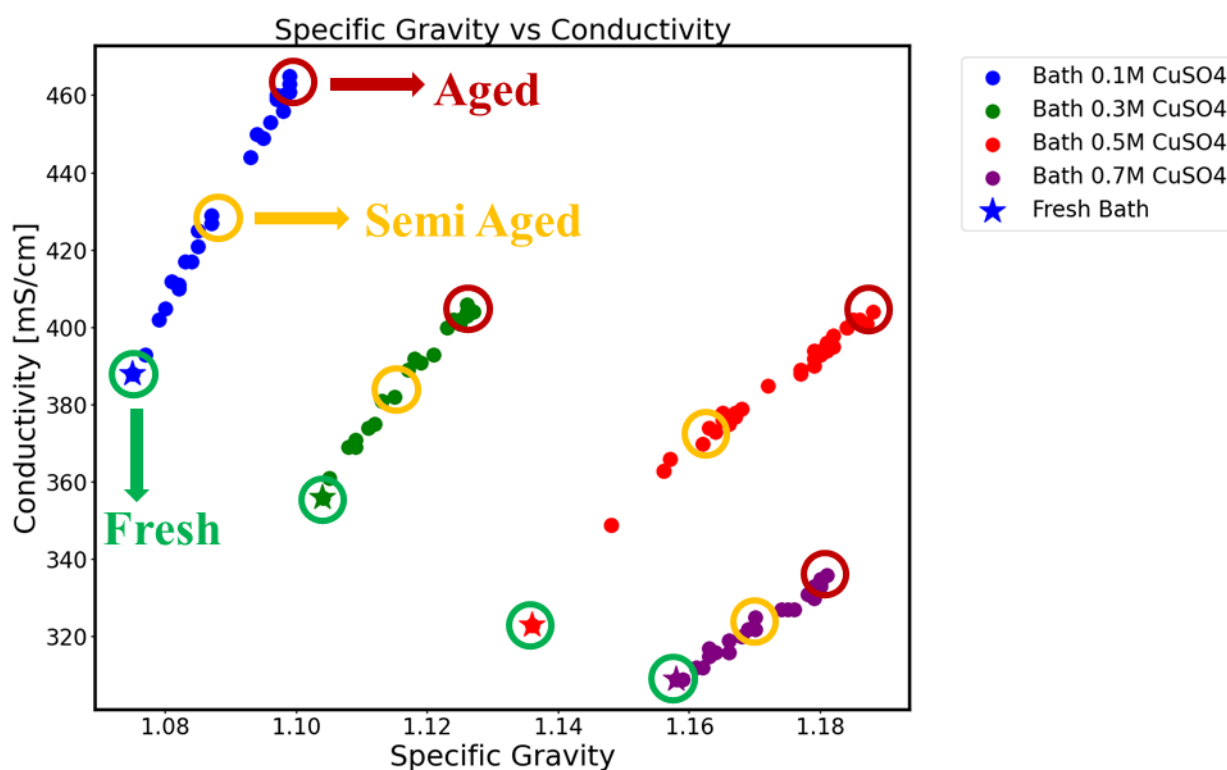
9.2.1 Defining bath aging states across all baths

To evaluate how process parameters should be adjusted as the electrolyte evolves, three representative aging states were defined for each of the four CuSO_4 concentrations used in this study:

- Fresh
- Semi-Aged
- Aged

These states were defined directly from the dataset collected over months of production-scale electroforming. For each CuSO_4 concentration, conductivity and specific gravity were monitored throughout the bath's operational lifetime and plotted to visualize the natural evolution of the solution. Points representing the early, mid-range, and late stages of this trajectory were selected as the Fresh, Semi-Aged, and Aged conditions, respectively (Figure 9.1). A corresponding summary table of CuSO_4 concentration, conductivity, and specific gravity for these selected points provides the quantitative basis for these classifications (Table 9.1).

This approach ensures that the “aging states” reflect the real physical and chemical drift of the solutions, rather than arbitrary labels, and that they remain consistent across all four bath concentrations.



(a)

Figure 9.1 The evolution of conductivity versus specific gravity for baths of different CuSO_4 concentrations. Markers indicate the selected trajectory points corresponding to the fresh, semi-aged, and aged bath states used in this chapter. These selections are based on the natural drift observed in the dataset and provide a representative sampling of the electrolyte's aging trajectory.

State	Solution	CuSO ₄ Concentration	Electric Conductivity	Specific Gravity
		Error: ±0.0002 mol/L	Error: ±0.4 mS/cm	Error: ±0.001 g/L
Fresh	1	0.1	388	1.075
	2	0.3	356	1.104
	3	0.5	323	1.136
	4	0.7	309	1.158
Semi Aged	1	0.1	434	1.089
	2	0.3	385	1.117
	3	0.5	373	1.165
	4	0.7	323	1.171
Aged	1	0.1	465	1.099
	2	0.3	406	1.127
	3	0.5	404	1.188
	4	0.7	336	1.181

(b)

Table 9.1 Evolution of electric conductivity and specific gravity during ageing for baths with different CuSO₄ concentrations.

9.2.2 Process parameter scenarios

Three parameter-variation scenarios were designed to demonstrate how the trained model can guide process adjustments as the bath ages:

Scenario A: Varying E_{dep}

- $Q_{\text{dep}} = -0.1 \text{ C/cm}^2$
- $Q_{\text{pol}} = 10\% \text{ of } Q_{\text{dep}}$

Scenario B: Varying Q_{dep}

- $E_{\text{dep}} = -150 \text{ mV}$
- $Q_{\text{pol}} = 10\% \text{ of } Q_{\text{dep}}$

Scenario C: Varying Q_{pol}

- $E_{\text{dep}} = -150 \text{ mV}$
- $Q_{\text{dep}} = -0.1 \text{ C/cm}^2$

These scenarios were not chosen to study fundamental electrochemical mechanisms (such as nucleation or mass-transport limits). Instead, they were selected as practical examples to demonstrate how a trained machine-learning model can be used to tune process parameters once the bath state is known.

By sweeping one parameter at a time while fixing the others, these scenarios provide clear one-dimensional response curves that show how Ra, G20, and G60 shift as the bath transitions from Fresh $\rightarrow$ Semi-Aged $\rightarrow$ Aged.

This structure highlights the central idea: a model trained on the complete dataset can be used in real time to select deposition conditions that compensate for bath aging and maintain desired surface properties.

9.2.3 Predicting Ra, G20, and G60 using trained random forest models

Random Forest was selected as the primary predictive model based on the comparative analysis in the previous chapter. Although Random Forest, SVR, and GPR frequently achieved similar accuracy, Random Forest consistently delivered this performance with minimal hyperparameter tuning and significantly shorter training times. This combination of efficiency and stability made it the most practical choice for running large numbers of predictions across multiple bath states and parameter scenarios. This model uses the most informative features, derived from bath chemistry, and measured process properties, to predict:

- Ra (roughness)
- G20 (low-angle gloss)
- G60 (high-angle gloss)

For each of the four CuSO_4 concentrations and for all three aging stages (different conductivities and specific gravities), predictions were generated across the full sweep of E_{dep} , Q_{dep} , or Q_{pol} defined in the three scenarios.

Maintaining identical model configurations across all tests ensures that differences in the trends arise only from the bath state or process parameter changes, not from model biases or retraining differences. These predictions form the basis of the trend interpretation presented in this chapter.

9.2.4 Spline-based trend reconstruction

The raw predictions across each sweep were smoothed using cubic and variable-degree spline fitting, selected case-by-case depending on the behavior of the underlying trend:

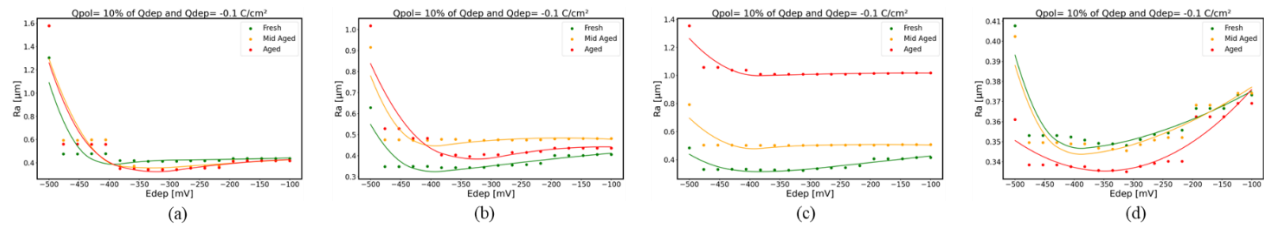
- Higher smoothing parameters were used for nearly linear or monotonic responses.
- Lower smoothing was used where sharper transitions or inflection points emerged.

This produces clean, continuous, interpretation-ready curves for all three scenarios and for all three surface properties.

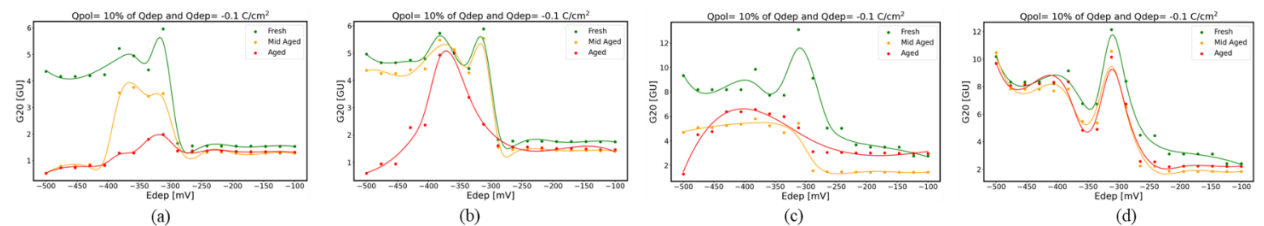
These graphs clearly show how the response of R_a , G_{20} , and G_{60} shifts as the bath transitions between Fresh, Semi-Aged, and Aged states, and illustrate how the model can be applied to support adaptive process control.

All resulting figures (Figures 9.2 to 9.4), covering the complete set of parameter sweeps for every bath state, are included here.

R_a



G_{20}



G60

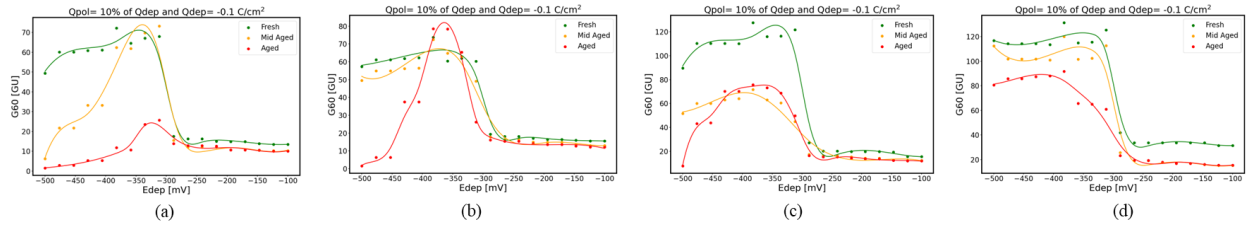
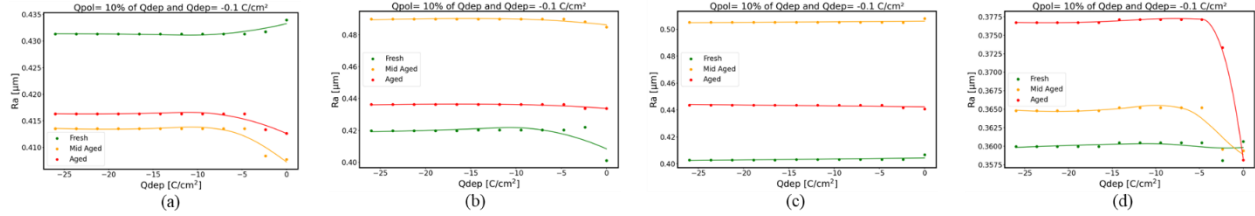
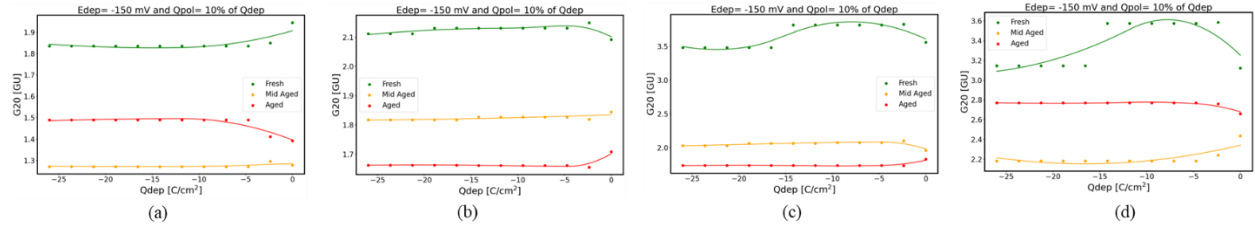


Figure 9.2 Scenario A: Predicted Ra, G20, and G60 vs. Edep across bath aging states (a- d represent the 0.1 M, 0.3 M, 0.5 M, and 0.7 M CuSO₄ concentrations).

Ra



G20



G60

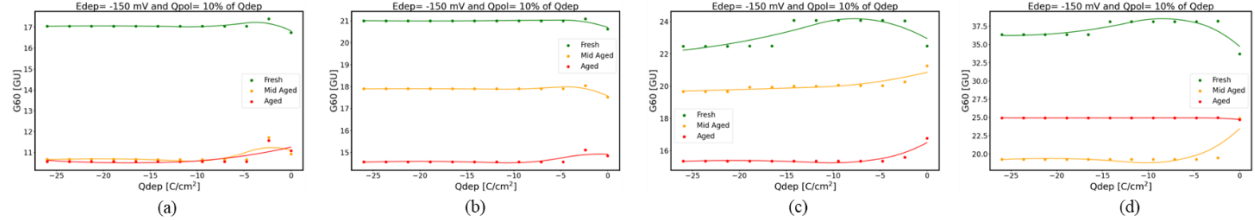
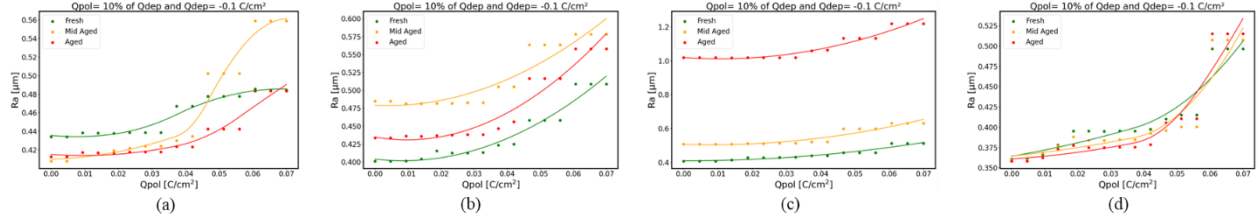
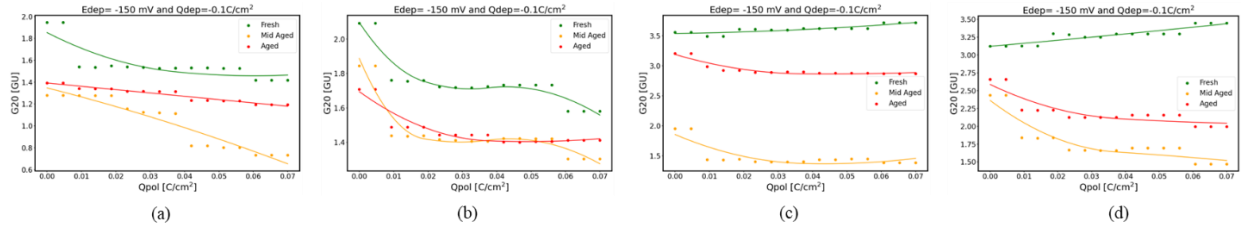


Figure 9.3 Scenario B: Predicted Ra, G20, and G60 vs. Qdep across bath aging states (a- d represent the 0.1 M, 0.3 M, 0.5 M, and 0.7 M CuSO₄ concentrations).

Ra



G20



G60

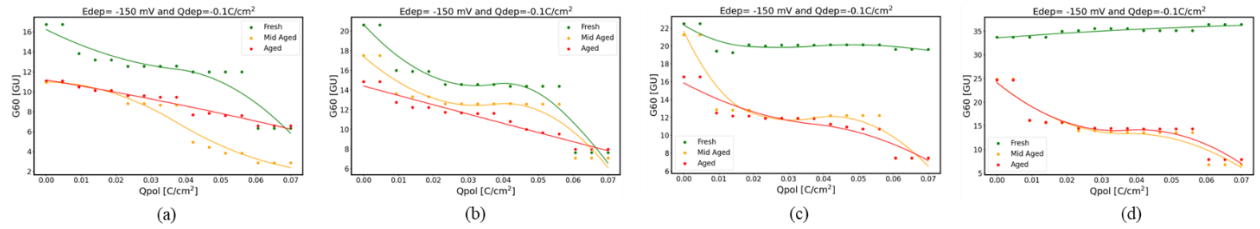


Figure 9.4 Scenario C: Predicted Ra, G20, and G60 vs. Qpol across bath aging states (a- d represent the 0.1 M, 0.3 M, 0.5 M, and 0.7 M CuSO₄ concentrations).

9.3 Sensitivity of predictions to incorrect CuSO₄ concentration

To evaluate how sensitive the predictive framework is to incorrect CuSO₄ inputs, the Random Forest models trained for the three outputs (Ra, G20, and G60) at the four original bath concentrations (0.1, 0.3, 0.5, and 0.7 M) were tested using a series of interpolated concentrations ranging from 0.1 to 0.9 M. The objective of this analysis is to determine the concentration range within which each model maintains error levels comparable to its trained condition, and to identify the point at which prediction accuracy degrades as the supplied CuSO₄ value deviates from the true concentration.

Figures 9.5–9.7 illustrate the relative error in Ra, G20, and G60 predictions under these mismatched-input conditions. For each trained model, predictions were generated across the full set of test concentrations, enabling a direct visualization of the robustness window and the region where errors increase noticeably.

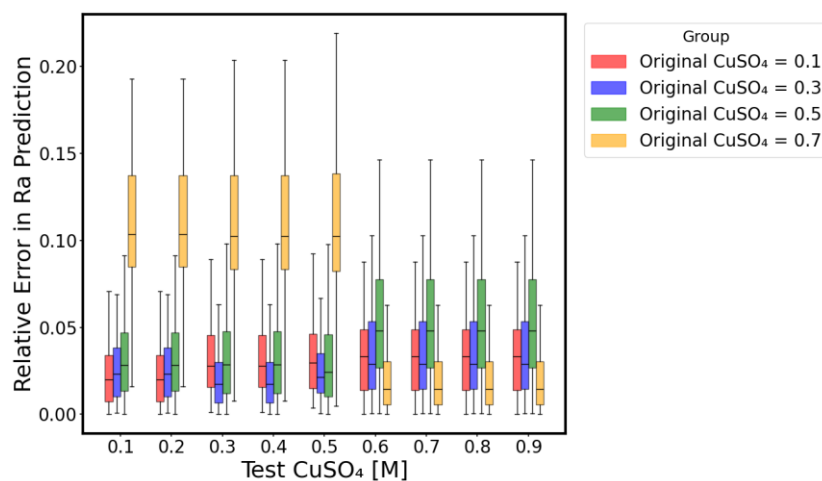


Figure 9.5 Effect of incorrect CuSO₄ input on Ra prediction

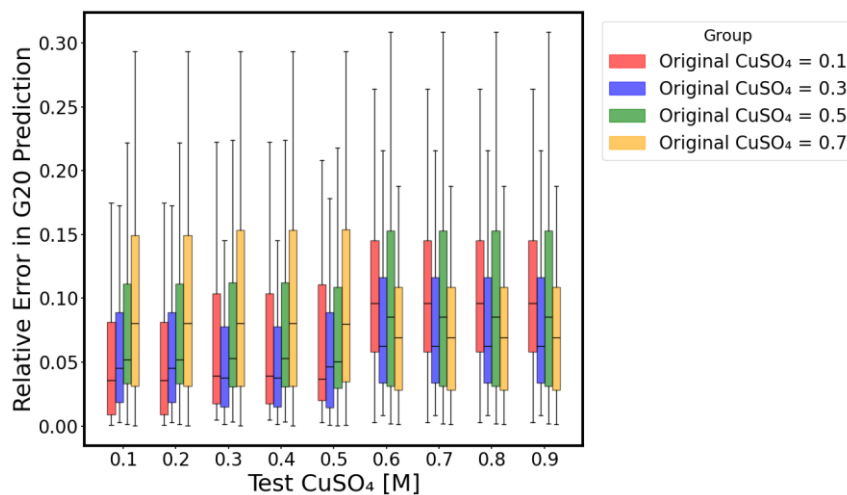


Figure 9.6 Effect of incorrect CuSO₄ input on G20 prediction

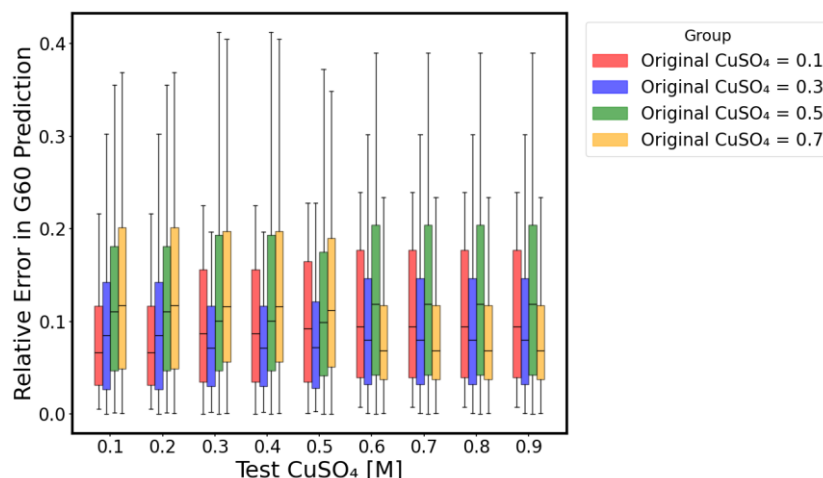


Figure 9.7 Effect of incorrect CuSO₄ input on G60 prediction

Ra prediction robustness

- 0.1 M and 0.3 M models: Exhibit the strongest robustness, maintaining low and stable relative error across almost the entire 0.1–0.9 M test range.
- 0.5 M model: Shows moderate robustness; its prediction accuracy begins to decline once the test concentration exceeds ~0.6 M.
- 0.7 M model: Displays the highest sensitivity to incorrect inputs, with substantially larger median errors and high variability whenever the test concentration deviates from its trained value.

G20 prediction robustness

- 0.1 M and 0.3 M models: Maintain the lowest and most stable G20 error
- 0.5 M model: Moderately robust, but error increases sharply when the assumed concentration exceeds ~ 0.6 M.
- 0.7 M model: Consistently the least robust; it shows higher median errors and greater variability across all test concentrations, with degradation especially severe when the input is below its trained concentration.

G60 prediction robustness

- 0.1 M and 0.3 M models: Again show the broadest robustness and comparatively low variability.
- 0.5 M model: Demonstrates moderate robustness but experiences a marked increase in both median and upper error bounds beyond ~ 0.6 M.
- 0.7 M model: Exhibits the largest sensitivity, with consistently high errors and the widest variability across the entire test range. Performance drops sharply when the test concentration falls below the training value.

Overall conclusions

Across all three surface outputs, a consistent pattern emerges:

- Models trained at lower CuSO_4 concentrations (0.1 and 0.3 M) are the most robust, maintaining stable and low errors even when the supplied concentration deviates substantially from its true value.
- The 0.5 M model provides moderate robustness, with acceptable accuracy until the mismatch becomes large.
- The 0.7 M model is the most sensitive (notably for roughness), showing high error growth and variability under any significant input mismatch.

These results demonstrate that reliable predictions can still be obtained despite imperfect concentration estimates, particularly for models trained on dilute and mid-range baths. However, as concentration increases, the system becomes progressively more nonlinear, and accurate CuSO_4 tracking becomes essential to avoid rapid degradation in prediction accuracy.

In practice, the model remains dependable as long as the estimated concentration lies within the robustness band indicated in Figures 9.5–9.7. Beyond this range, prediction reliability declines, underscoring the importance of consistent concentration monitoring in long-term electroforming operations.

9.4 Generalization to unseen bath condition and model robustness evaluation

9.4.1 Motivation for testing unseen acid concentrations

Electrolyte degradation in copper electroplating is strongly influenced by deviations in sulfuric acid content. As reported by *Grandos et al.*¹⁴⁵, several chemical indicators reflect poor bath health, including:

- Elevated sulfuric acid concentration
- Reduced or insufficient sulfuric acid levels
- Deficiency of copper ions in the electrolyte
- Contamination by foreign metal salts
- Accumulation of organic breakdown products

Among these factors, excess or insufficient sulfuric acid is both common in industrial operations and highly impactful on the electrochemical environment. Therefore, in this study, deviations in H_2SO_4 concentration were used as a controlled way to simulate bath aging and evaluate how well the trained models can generalize beyond their original chemical domain.

9.4.2 Testing generalization on unseen acid concentrations

All machine-learning models used in this chapter were trained exclusively on data collected at 1 M H_2SO_4 , representing the nominal and healthy bath condition. To assess generalization, each model was subsequently tested on a set of unseen and chemically shifted acid concentrations, specifically:

- 0.5 M
- 1.25 M
- 1.5 M
- 2.0 M
- 3.0 M

These concentrations represent both sub-optimal acidity (under-acidified bath) and severe over-acidification. Predictions were generated for all three parameter-sweep scenarios introduced earlier (varying E_{dep} , Q_{dep} , Q_{pol}), ensuring that generalization is evaluated across a broad operational envelope rather than a single condition.

9.4.3 Normalized error analysis

To quantify the deterioration in model accuracy as acid concentration deviates from 1 M, the following normalized error metric was used:

$$\text{Normalized Error} = \frac{\text{Absolute Error at Unseen Acid Concentration}}{\text{Mean of Measured Target in Dataset}}$$

A normalized error close to the value obtained at the trained condition indicates strong robustness, whereas values substantially above or below this baseline show that the model struggles to extrapolate under chemically altered conditions. This metric provides a consistent and interpretable measure of out-of-distribution performance across Ra, G20, and G60.

9.4.4 Process scenarios used for model comparison

To evaluate robustness under realistic deposition settings, three representative process conditions were selected:

Scenario 1:

- $E_{\text{dep}} = -150 \text{ mV}$
- $Q_{\text{dep}} = -0.1 \text{ C/cm}^2$
- $Q_{\text{pol}} = 0.01 \text{ C/cm}^2$
- $E_{\text{pol}} = +100 \text{ mV}$

Scenario 2:

- $E_{\text{dep}} = -190 \text{ mV}$
- $Q_{\text{dep}} = -0.1 \text{ C/cm}^2$
- $Q_{\text{pol}} = 0.015 \text{ C/cm}^2$
- $E_{\text{pol}} = +100 \text{ mV}$

Scenario 3:

- $E_{\text{dep}} = -240 \text{ mV}$
- $Q_{\text{dep}} = -0.1 \text{ C/cm}^2$
- $Q_{\text{pol}} = 0.02 \text{ C/cm}^2$
- $E_{\text{pol}} = +100 \text{ mV}$

These scenarios were selected to ensure that model robustness was evaluated across multiple, distinct process conditions, rather than being biased by a single operating point. By testing the models under different combinations of E_{dep} , Q_{dep} , and Q_{pol} , the analysis becomes more generalizable and provides a stronger validation of each model's ability to maintain predictive accuracy across a broader range of practical electroforming settings.

9.4.5 Comparative assessment of model robustness

The graphs shown in Figure 9.8 summarize and compare the performance of all models under the tested conditions.

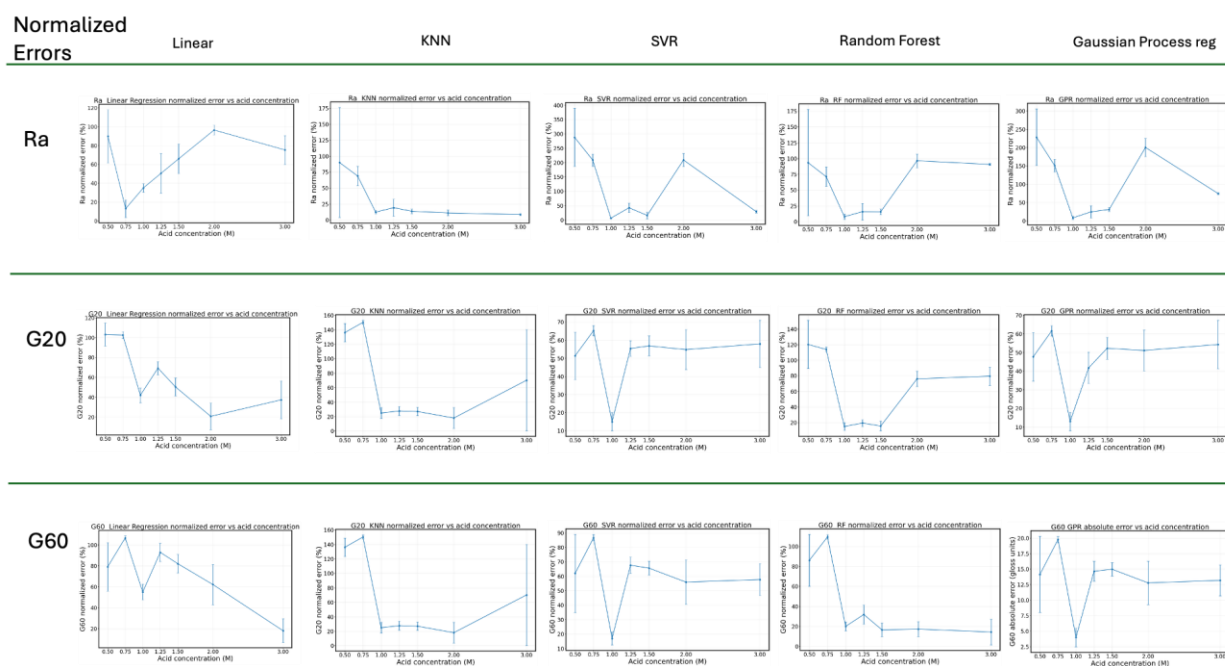


Figure 9.8 Comparison of the normalized error among the trained models in the prediction of Ra, G20, and G60 for unseen acid concentrations

Based on these graphs, the normalized-error plots reveal clear differences in robustness between the ML models:

K-Nearest Neighbors (KNN)

- Demonstrates good robustness from 1 M up to 2 M, maintaining normalized error close to trained error. This robustness can be extended to 3M for the roughness.

- Performance degrades rapidly for acidities below 1 M, indicating limited ability to extrapolate toward under-acidified baths.
- Confirms that KNN, being a local averaging method, struggles when the testing point falls outside the density of the training manifold.

Random Forest (RF)

- Shows very strong robustness across acid concentrations:
 - Ra and G20: robust up to 1.5 M
 - G60: robust up to 3.0 M, showing minimal deterioration even under significant over-acidification
- However, similar to KNN, RF accuracy drops sharply below 1 M, consistent with the increased chemical instability in under-acidified electrolytes.
- These results align with the fact that Random Forests handle nonlinear boundaries effectively, giving them stronger extrapolation behavior in chemically stable regimes.

Gaussian Process Regression (GPR)

- Exhibits moderate robustness for Ra, with acceptable performance up to ~1.5 M.
- For G20 and G60, robustness is weaker; the normalized error grows earlier and more sharply than for KNN or RF.
- The limited robustness of GPR reflects its sensitivity to out-of-distribution inputs, since kernel-based methods rely heavily on similarity to the training domain.

The pronounced sensitivity of Linear Regression and SVR to minor variations in acid concentration renders them unsuitable for real-world applications, where continuous bath degradation and chemical drift are unavoidable.

These results reveal a strong link between acid molarity and the stability of surface-property predictions. Prior studies have shown that both gloss and roughness are highly sensitive to acidity because shifts in sulfuric acid concentration directly affect conductivity, ion diffusion, hydrogen-evolution kinetics, and copper-ion transport^{80,99,100}.

The findings here carry important implications for practical electroforming. As the bath ages and the sulfuric acid level drifts from its nominal value, KNN and Random Forest consistently provide the most reliable predictions, maintaining stable accuracy across a wide range of chemically altered conditions. This robustness makes them particularly suitable for real production environments where acid concentration naturally changes over time. However, once the acidity moves outside this robustness range, especially below 1 M, the performance of most models declines sharply, highlighting the need for consistent bath monitoring in long-duration or high-throughput operations.

9.5 Conclusion

This chapter demonstrated how machine-learning models can be used to guide process optimization and maintain surface quality in evolving electroforming environments. By defining realistic bath-aging states across four CuSO_4 concentrations and evaluating three practical parameter-sweep scenarios, the trained models (particularly Random Forest) were shown to capture how Ra, G20, and G60 shift as both chemistry and process conditions change. The spline-smoothed trends revealed clear dependencies between deposition settings and surface outcomes at each aging stage, underscoring the value of data-driven tools for adaptive parameter selection.

The chapter also examined two critical robustness challenges: incorrect CuSO_4 inputs and deviations in H_2SO_4 concentration. The first analysis showed that models trained on dilute and mid-range CuSO_4 concentrations maintained stable predictions across a broad concentration span, whereas models trained at higher CuSO_4 became increasingly sensitive to mismatch. The second analysis (testing generalization to unseen acid concentrations) highlighted that both KNN and Random Forest provide the strongest resilience to chemical drift, reliably predicting Ra, G20, and G60 across moderate over-acidification while failing sharply below 1 M H_2SO_4 . Gaussian Process Regression showed partial robustness, whereas Linear Regression and SVR were unsuitable due to their strong sensitivity to even minor concentration deviations.

Together, these findings reinforce a central conclusion: machine-learning-based control is viable for electroforming, but only when grounded in models with demonstrated robustness to chemical evolution. KNN and Random Forest, in particular, exhibit the stability required for real-world deployment, where bath chemistry naturally shifts due to consumption, additives, contamination, and operational aging. The results also emphasize that maintaining accurate monitoring of sulfuric acid concentration remains essential, especially in low-acidity regimes where predictive performance deteriorates most rapidly.

Overall, this chapter establishes the feasibility of using ML models not only to predict surface properties but also to adapt process parameters as the bath ages, forming a critical foundation for the real-time, closed-loop electroforming control strategies proposed in the following chapter.

10. Outlook

This thesis addressed two central objectives:

- (1) the development of a hybrid manufacturing approach that integrates additive manufacturing with electroforming to enable cost-scalable mass personalization of micro-metal components, and
- (2) the establishment of a predictive modeling framework capable of capturing surface-quality evolution and adapting electroforming process parameters under evolving electrolyte conditions.

Together, these contributions open several high-impact avenues for future investigation. The following sections outline the most significant directions that build directly on the findings of this work.

10.1 Advancing hybrid AM–electroforming for personalized microfabrication

The hybrid AM–electroforming workflow demonstrated in this thesis shows that polymer-based molds produced through additive manufacturing can reliably support high-quality metal deposition and enable the fabrication of geometrically unique, personalized components. This represents a promising pathway toward high-precision, low-cost metal personalization, yet several scientific and practical questions remain unresolved.

1. Propagation of AM-induced surface texture through deposited metal layers

While AM templates are viable substrates for electroforming, the mechanisms by which mold texture, such as layer lines, surface anisotropy, or microscale waviness, propagates through the metal deposit remain insufficiently understood. Key open questions include:

- To what extent do mold roughness wavelength and amplitude influence the nucleation and early growth stages of copper deposition?
- How do waveform characteristics and current density affect texture replication or smoothing?
- Under which conditions does the electroforming process attenuate, preserve, or amplify mold-induced features?

Addressing these questions is essential for establishing predictable and repeatable surface-quality outcomes in personalized components.

2. *Automating mold generation for arbitrary user-defined geometries*

The current workflow still requires manual intervention for designing, preparing, and validating AM molds. Future work should focus on:

- translating arbitrary metal geometries into manufacturable mold designs,
- embedding surface-quality constraints (e.g., target Ra or gloss) into geometry generation,
- automating support-structure creation and release strategies, and
- integrating predictive models to recommend deposition parameters during mold creation.

Achieving this level of automation would mark a significant step toward true mass personalization, where design-to-fabrication occurs with minimal manual input.

Future Research Focus

Future efforts should establish predictive relationships between AM process variables (mold material, layer height, build orientation, surface roughness) and resulting electroformed surface quality. A digital-twin environment (Section 8.2) could enable virtual prototyping of molds and deposition conditions, reducing trial-and-error and improving consistency across personalized geometries.

Overall, deeper integration between AM and electroforming has the potential to enhance design freedom, reduce cost per part, and improve process repeatability. The lack of existing end-to-end workflows in the literature underscores the novelty and value of this research direction.

10.2 Toward a digital-twin framework for hybrid AM–electroforming

The methodologies developed in this thesis, structured dataset design, pulse-feature extraction, and cross-bath predictive modeling, form the technical foundation for a future digital twin of the electroforming process. Extending this concept to the hybrid AM–electroforming workflow would require:

- incorporating continuous sensing of bath-state properties,
- enabling real-time model updating as the electrolyte evolves,
- implementing closed-loop inverse prediction for target surface qualities,
- linking mold-design workflows to predictive models, and

- enabling automatic adjustment of process parameters during deposition.

A digital twin capable of coupling mold geometry, bath state, and deposition physics would allow geometry-specific optimization of waveform and process parameters. This capability would reduce manual tuning, lower cost per part at scale, and provide a unified framework for producing personalized metallic components with consistent quality.

1. Coupling geometry, chemistry, and deposition physics

Electroforming is highly sensitive to geometric variations that alter current distribution, mass-transport behavior, and local deposition rates. Personalized AM molds introduce unique geometries for each part, making traditional parameter-setting approaches insufficient. A digital twin could simulate:

- how curvature affects electric-field intensity,
- where thickness non-uniformities may occur,
- how bath aging affects deposition profiles, and
- how waveform parameters should be adapted for each geometry.

2. Real-time adaptation to electrolyte evolution

This thesis demonstrated that predictive accuracy deteriorates when bath chemistry shifts slightly. A digital twin equipped with real-time sensing (conductivity, optical measurements, current-response signatures, etc.) could update models continuously and correct deposition parameters automatically, forming a closed-loop adaptive system.

3. Automated parameter selection for unique geometries

For mass personalization, each part may require a different parameter set. A digital twin can:

- identify the waveform that achieves a target roughness or gloss,
- optimize deposition time versus uniformity,
- translate geometry into process parameters automatically.
- this would eliminate manual process development for individualized parts.

4. Integration with automatic mold-generation pipelines

By linking automated mold design (Section 8.1) with predictive control, the digital twin completes a closed manufacturing loop:

Input: a user-defined geometry

Output: a finished electroformed part with guaranteed quality

5. Industrial and research impact

A fully developed digital twin for AM–electroforming would:

- reduce cost per part,
- enable rapid prototyping of personalized metal structures,
- allow predictive quality assurance,
- support the large-scale deployment of individualized fabrication, and
- establish a benchmark modeling and data framework for future electroforming research.

This direction represents one of the most impactful continuations of the present thesis.

10.3 Generalization and strengthening of the predictive electroforming framework

While the experimental results presented in this thesis are specific to additive-free copper electroforming in $\text{CuSO}_4/\text{H}_2\text{SO}_4$ electrolytes, the underlying framework, comprising bath-state representation, pulse-response feature extraction, robustness mapping, and data-driven process-parameter adaptation, is not inherently tied to a specific chemistry. With appropriate retraining and domain-specific calibration, the same modeling and control strategy can be applied to other electrochemical deposition systems.

The findings further demonstrate that predictive performance is strongly governed by bath variability, waveform diversity, and the chemical operating environment. Consequently, extending the present framework toward a broadly deployable predictive platform will require targeted expansion of multi-bath datasets, incorporation of physics-informed modeling elements, and systematic validation across additional electrochemical systems.

1. Expansion of multi-bath and multi-aging datasets

The dataset developed in this thesis is extensive but remains limited in the number of baths, aging cycles, and acid concentrations. Future work should expand the dataset to include:

- wider CuSO_4 and H_2SO_4 ranges,
- intentional introduction of impurities to represent industrial drift,
- a broader set of pulse-reverse waveform conditions.

Such diversity will improve out-of-distribution robustness, enable stronger benchmarking, and enhance understanding of process–chemistry interactions across AM–electroforming workflows.

2. Development of multi-output, physics-informed predictive models

While this thesis demonstrated the value of data-driven modeling, it also showed that purely empirical models become unstable when bath conditions shift. Future models should incorporate:

- ionic-transport behavior,
- diffusion-limited current characteristics,
- nucleation and growth kinetics,
- hydrogen-evolution mechanisms,
- surface-leveling phenomena.

Embedding these principles into multi-output models (e.g., PINNs or mechanistic–ML hybrids) will improve extrapolation, interpretability, and reliability under evolving bath conditions.

3. Extension to other metals and hybrid electrochemical processes

The framework developed in this thesis is not limited to copper. It can be adapted to:

- nickel, cobalt, silver, gold, and alloy systems,
- ultrasonic-assisted, flow-assisted, or rotating-cathode deposition,
- hybrid or electroless–electroforming processes.

Investigating these systems would clarify the generality of bath-state modeling, waveform–surface relationships, and predictive performance across electrochemical environments. This would transform the current copper-specific study into a broadly applicable predictive platform for electrochemical manufacturing.

11. Conclusions

Throughout this thesis, a systematic investigation was conducted to advance the understanding of hybrid additive manufacturing–electroforming processes under mass-personalization conditions, with particular emphasis on the effects of electrolyte evolution on surface quality and process stability. The work provides quantitative insight into how bath condition, pulse-reverse process parameters, and electrochemical response jointly influence surface roughness and gloss in copper electroforming.

Most importantly, the objectives defined in the introduction chapter were addressed as follows:

- *Establishing a hybrid manufacturing architecture for cost-scalable mass personalization*

This thesis demonstrated that a hybrid process architecture combining additive manufacturing and electroforming provides a viable pathway toward cost-scalable personalization of metal components. Additive manufacturing enabled rapid, low-cost fabrication of geometrically unique polymer molds without dedicated tooling, while electroforming allowed parallel metal deposition with high dimensional fidelity and surface quality. Experimental results confirmed that this architecture restores a decreasing cost-per-part trend with increasing production volume, even when part geometries vary, thereby overcoming a fundamental limitation of conventional personalized manufacturing approaches.

- *Characterizing electroforming variability under personalized production conditions*

It was shown that electroforming behavior under mass personalization differs fundamentally from conventional mass production. Experimental observations revealed that electrolyte aging and geometry-dependent current distribution interact nonlinearly, leading to significant variability in surface roughness and gloss even under identical nominal process parameters. These results demonstrated that static parameter optimization and empirical tuning are insufficient for maintaining reproducible surface quality in personalized electroforming, motivating the need for a data-driven control framework.

- *Constructing a comprehensive open dataset for electroforming under evolving bath conditions*

A large, structured open dataset for additive-free copper electroplating was constructed, incorporating data from 114 distinct electrolyte baths across four CuSO_4 concentrations and multiple aging stages. The dataset includes physicochemical bath measurements, pulse-current response features, and detailed surface roughness and gloss measurements. Analysis of the dataset showed that conventional bath indicators such as conductivity and density alone cannot fully

explain surface-quality variability, highlighting the importance of combining static and dynamic process information.

- *Defining and modeling an operational electrolyte bath state*

To move beyond the ambiguous notion of bath “age,” this thesis introduced an operational definition of bath state as a low-dimensional representation of the electrolyte’s effective electrochemical condition. This bath state was inferred from a combination of physicochemical descriptors and pulse-response features. Through systematic feature selection and model comparison, it was demonstrated that this representation captures cumulative aging effects and enables stable prediction of surface properties. Among the evaluated models, Random Forest provided the most consistent accuracy and robustness without extensive hyperparameter tuning.

- *Developing a data-driven framework for adaptive process-parameter selection*

Building on the inferred bath state, this thesis demonstrated how machine-learning models can be used to adapt electroforming process parameters as the electrolyte evolves. Scenario-based studies showed that deposition potential, deposition charge, and polishing charge can be adjusted to compensate for bath aging and maintain target surface roughness and gloss within defined chemical and operational bounds. Robustness analyses further revealed that reliable predictions depend on bath chemistry, with Random Forest and K-Nearest Neighbors exhibiting the strongest resilience to concentration mismatch and moderate acid drift, while linear regression and SVR were unsuitable for evolving electrochemical environments.

To summarize, this thesis establishes a coherent pathway toward intelligent electroforming for mass-personalized production by integrating hybrid process architecture, structured experimentation, open dataset construction, bath-state inference, and data-driven process adaptation. Rather than relying on empirical tuning or static control, the proposed framework enables informed adjustment of process parameters under evolving electrolyte conditions, forming a practical foundation for adaptive and scalable electrochemical manufacturing.

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