The Novel Reactor Design and Operating Conditions for Sustainable Electrokinetic Recovery of Oil and Water from Oily Sludge

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Abstract

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Setareh Taslimi Taleghani

Oily sludge has been an emerging concern as a waste product in the petroleum industry. It is generated in huge amounts in both upstream and downstream industries. Oily sludge is a very complex matrix with a wide range of constituents including light and heavy petroleum hydrocarbons (PHCs), solids, water and different kinds of heavy metals such as nickel and vanadium. Since oily sludge has proven to expose dangers to the public health and the environment, it has been considered as hazardous waste and its treatment before disposal has been essential. Oily sludge is a very stable water-in-oil suspension that is naturally emulsified by PHCs and small solid particles, and hence its treatment is a challenging issue. The objective of this study is to present the electrokinetic method as a sustainable approach to the successful treatment of oily sludge. To achieve this purpose, a novel design of the EK reactor was proposed to undergo further investigation. The optimization of the EK reactor design was conducted on a small scale. The new design was applied to larger scales with the required modifications. In order to enhance phase separation and obtain a higher quality of recovered material, different additives were introduced into the system. Ferric chloride proved to be the most effective one according to the qualitative and quantitative study. The large-scale EK reactor (20 L) achieved 35% of water recovery and 15% of oil recovery while it reduced the disposable volume by 35% after 150 hours of operation. The optimal conditions and configurations of the batch system were used to design an innovative continuous-flow reactor. The new reactor aimed to attain a higher recovery of oil and water and lower energy consumption during a 24-h retention period. A water recovery of 31% and an oil recovery of 11% was accomplished after 24 hours as a result of the successful operation of the novel system. The developed EK systems are ready to be applied at full scale to treat industrial and municipal sludge.

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Dedications

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List of abbreviations

Name	Definition
API	American Petroleum Institute
ASTM D4007	Standard Test Method for Water and Sediment in Crude Oil by the Centrifuge Method (Laboratory Procedure)
ASTM D95	Standard Test Method for Water in Petroleum Products and Bituminous Materials by Distillation
BS&W	Basic Sediment and Water
Cmix	A mixture of all oil sludge samples except samples C2 and C7
Cmix (C1-C8)	A mixture of all eight samples of oil sludge
EK	Electrokinetics
Heavy oil	Typical carbon range from C8 to C50
Light oil	Typical carbon range from C5 to C7
N/A	Not Applicable
ND	Not Detected
O/W	Oil-in-water emulsions
PAHs	Polycyclic Aromatic Hydrocarbons
PHCs	Petroleum Hydrocarbons

VOCs	Volatile Organic Compounds
W/O	Water-in-oil emulsions
Wt. %	Weight percentage

1 Introduction and research objectives

Petroleum sludge is generated in an enormous amount by petroleum industries through the production and processing of crude oil. In general, a higher capacity of crude oil production results in an increased generation of oily waste. According to an estimation, processing 500 tons of crude oil generates one ton of oily sludge (Hu et al., 2013).

Oily sludge consists of various amounts of waste oil and water, mineral material, and fine particles. It has been declared as hazardous waste by the Environmental Protection Act; Therefore, it cannot be landfilled due to the major exposure of health and environmental hazard (Islam, 2015).

Oily sludge has a high viscosity and a complex composition which mostly consists of alkanes, asphaltenes, and resin (Prakash et al., 2015). It is a very stable water-in-crude oil emulsion that is created by natural emulsifiers such as asphaltenes (Atta et al., 2018).

As one of the most important environmental concerns, management of oily sludge is a priority, however, many factors should be considered in order to choose an effective and adequate method of treatment. Some of these factors include feasibility, environmental impact, energy resources, cost estimation, and sustainability (Battikhi, 2014).

Mostly, methods for treatment of oily sludge focus on volume reduction by removal of moisture from the sludge. Centrifugation, solvent extraction, and thermal desorption are among these methods which do not present a cost-effective and sustainable approach towards oily sludge management. They are often associated with operational problems or require extra treatment procedures to meet the regulations (Elektorowicz et al., 2006).

On the other hand, electrokinetics has emerged as a successful treatment technique for the removal of organic and inorganic pollutants. Amongst the advantages of applying this method to the management of oily sludge are the recovery of oil and water from waste, possible reuse of the recovered material, elimination of the hazards of environmental pollution including contamination of water resources, financial gain through the recovery of fuel and generally the sustainability of the method (Elektorowicz et al., 2006; Taleghani et al., 2019; Taleghani, 2018).

The main objective of this research was to promote the electrokinetic (EK) method in order to achieve the best results concerning the quality and quantity of the separated phases (water and oil). The detailed objectives were to: i) optimize the configuration of the batch EK reactors; ii) find effects of different voltage gradients and various additives; iii) assess the impact of scaling up of EK reactors; iv) design and evaluate the efficiency of a continuous-flow EK reactor.

2 Literature review

2.1 Source of oily sludge

An amount of 30000 tons of oily sludge is being generated in each refinery, every year (U.S. EPA,1991) which is considered as hazardous waste by USEPA (U.S. EPA, 2012). This sludge is basically a stable water-in-oil emulsion that is generated in both upstream and downstream petroleum industries and consists of a huge amount of solids resulting in a high viscosity of sludge (Hu et al., 2013).

Upstream operations consist of exploration, drilling, and extraction. Drilling by-products such as mud residue, bottom sediments of the crude oil tank, and the slop oil at wells are among the significant sources of oil sludge in upstream industries (O'Rourke and Connolly, 2003).

Downstream industries contribute to oil sludge generation in various ways including the sludge from storage tanks, American Petroleum Institute (API) separator sludge, dissolved air flotation (DAF), biological sludge from treating the produced wastewater, sludge from flocculation and flotation, parallel and corrugated place inceptors and oil/water separator residues (Hu et al., 2013; Shie et al., 2000)

Among many types of oily sludge, there have been several studies on the bottom sediments of the crude oil storage tanks. While stored, crude oil tends to separate into heavier and lighter PHCs. Heavier PHCs then gather together with solids and water, based on their densities (Ayotamuno et al., 2007).

2.2 Characteristics of oil sludge

The constituents of the crude oil, type of operations and processes, and utilized equipment determine the various components of the oily sludge's complex matrix. This matrix contains a variety of petroleum hydrocarbons (PHCs), solids, water, and some heavy metal ions including nickel, cadmium, vanadium, zinc, and lead (Mazlova and Meshcheryakov, 1999). Many of these constituents are toxic and carcinogenic (Zhou et al., 2009).

Oily sludge is composed of 30-90% of water and 4-7% of sediments which mostly include halite, calcite, kaolinite, and quartz. The remaining 5-60% of the oil sludge contains petroleum hydrocarbons. A range of 40-60% saturated hydrocarbons, 25-40% aromatic hydrocarbons, 10-15% resins, and 10-15% asphaltenes compose the oily fraction of the sludge (da Silva et al., 2012). PHCs and small solid particles naturally emulsify the stable oily sludge suspension (Hu et al., 2013). The surface-active constituents including resins and asphaltenes and wettable fragments such as sands and clays are located at the water-oil interface and prevent coalescence of the water droplets by forming a barrier between them (Sztukowski and Yarranton, 2005).

Bhattacharyya and Shekdar (2003) indicated some common disposal approaches of oily waste including landfill, land farming, disposal to lagoons, and incineration. They assessed the risks and problems associated with improper disposal of this hazardous waste such as probable leakage, foul odor, and groundwater pollution.

2.3 Various methods for treatment of oil sludge

Various methods for treatment of oil sludge suspension have been studied. However, there are few of them which meet the regulations that are introduced regarding oil sludge management (Elektorowicz et al., 2006).

2.3.1 Centrifugation

A centrifuge is high-speed rotational equipment which employs the gravitational force to separate phases based on their densities. Reducing the viscosity of oily sludge through pretreatment would benefit the centrifugation process. For this purpose, the addition of different chemicals such as organic solvents and demulsifiers to the sludge or pre-heating would be effective (Hu et al., 2013; Zubaidy and Abouelnasr, 2010).

Centrifugation is the most common method which is used by the industry and is mostly based on decreasing the volume of the sludge by water extraction. Centrifugation, as well as other methods including solvent extraction, thermal desorption, and hydrothermal processing, does not provide the industries with a cost-effective and sustainable method of treatment (Elektorowicz et al., 2006). The high equipment cost restricts the feasibility of a large scale centrifugation equipment (Nii et al., 2009). This technique only requires a small space. However, its operation might lead to frequent noise and vibrations in case of improper installation. The centrifugation process involves high-energy consumption to reach enough centrifugation force to separate oil from petroleum sludge, heat sludge additionally, and pump viscous sludge. High-energy consumption limits the application of centrifugation method to the small-scale industries (Dia et al., 2017). It is a relatively expensive process, especially when extra heat and chemicals are supplied to make sludge more treatable (Alshammari et al., 2008; Elektorowicz and Habibi, 2005).

2.3.2 Solvent extraction

Al-zahrani and Putra (2013) conducted studies on the regeneration of lubricating oil by different solvent extraction methods. They described their approach as one of the simplest and most cost-effective methods of treatment. A proportional amount of solvent was combined with the oil, based on the miscibility of the oil in the chosen solvent. Carbonaceous contaminants were extracted by the solvent and settled due to gravity. The solvent was finally recycled through the distillation process.

High temperature increases the extraction rate in the solvent extraction process. However, the evaporation of some hydrocarbons and the solvent itself would take place. On the other hand, the low temperature would be more cost-effective but less efficient with regards to the recovery of oil. Overall, the low efficiency of solvent extraction limits its use in oil recovery from oil sludge especially in the large scale process (Hu et al., 2013; Jafarinejad, 2016).

2.3.3 Pyrolysis

Pyrolysis is a thermal disposal method that is effective mostly in waste reduction and reutilization of the energy (Shen et al., 2016). A carrier gas is used in this process which is nitrogen in the case of pyrolysis of the oil sludge. This method mostly focuses on water removal from the oil to enhance its heating value. The process is conducted at 380 to 1073K (Shie et al., 2000).

Pyrolysis proves to be a more effective thermal method than incineration. It recovers oil that is readily applicable to diesel-fueled engines. Moreover, leaching of metals that remain in solid chars is less probable in comparison with that of incineration. However, there are various disadvantages to this method including high operational cost due to the high use of energy, restrictions in large scale applications due to the complexity of equipment, possible need for dewatering prior to the process and presence of a large amount of carcinogenic PAHs (Hu et al., 2013; Jafarinejad, 2016).

2.3.4 Freeze/Thaw

Due to the studies conducted by Chen and He (2003), the freeze/thaw method would be effective in the demulsification of water-in-oil emulsions by achieving a 90% removal of water. The proper freezing temperature was reported to be -40 ° C. During the freezing part, water droplets were frozen and some of the surfactant molecules were driven out of the water interface into the oil phase. During the thawing process, the temperature increased, and more surfactant molecules were removed from the ice as it melted. The absence of the surfactant in the interface facilitated the coalescence of water droplets and water removal from the emulsion. The rate of water removal depends highly on the rate and temperature of the freezing and thawing process as well as the initial water content (Rajakovic and Skala, 2006).

Generally, the freeze/thaw process proved to be effective for oil and water recovery from oily sludge. However, the industrial application of this method should be further investigated due to the long freezing time and the high costs and energy consumption associated with it. Therefore, this process would be more beneficial for cold regions with a probable natural freezing process (Hu et al., 2013; Jean and Lee, 1999).

2.3.5 Microwave irradiation

Microwave, which has been described as clean energy with high efficiency, is an electromagnetic wave with a 300 MHz to 300 GHz frequency range. Microwave radiation increases the temperature when it is applied for the treatment of a water-oil emulsion. The rise in temperature results in a reduction of viscosity. Microwave radiation is different from conventional heating since it generates a uniform distribution of heat throughout the material rather than resulting in heat convection, conduction, and radiation from the surface of the substance. In water-in-oil emulsions, more heating energy of the microwave is absorbed by water compared to oil. Thus, the expansion of water would lead to a decrease in the thickness of the interfacial film. Finally, water droplets will attach together, and the separation of phases will take place (Tan et al., 2007).

Microwave radiation provides the emulsions with a high energy source that could increase the temperature to 300℃ and above. Water behaves as the acceptor of this energy and transfers it to the organic contaminants in the emulsion (Robinson et al., 2008).

This method of treatment could achieve a water removal percentage of above 90%. However, the associated operational costs and requirement of special equipment restricts its utilization in the industrial-scale management of the oily sludge (Hu et al., 2013).

2.3.6 Ultrasonic irradiation

A periodic sound pressure with a frequency of more than 20 kHz is called the ultrasound. The penetration of ultrasonic waves into the sludge texture generates compression and rarefaction cycles. Liquid molecules undergo positive pressure during the compression cycle that pushes them together. On the other hand, these molecules are pulled from each other under the exposure of negative pressure during the rarefaction cycle. A large amount of negative pressure leads to the generation and successful growth of microbubbles in rarefaction areas. Microbubbles finally collapse violently as a result of their unstable diameter. The shock waves are generated at a very high temperature and pressure for a few microseconds during this violent collapse. This is described as the cavitation process which strongly affects the sludge demulsification process (Pilli et al., 2011).

The ultrasonic energy displaces the fluid particles and changes their velocity. It also results in the cavitation process, instability of the liquid interface, and radiation pressure. the movement of the liquid through the medium depends mostly on the cavitation and capillary forces. In general, acoustic energy enhances the liquid conductivity and efficiency of contaminant removal (Chung and Kamon, 2005).

Ultrasonic radiation is a very efficient method of treatment. However, its application to the industrial scale is not very effective. Large ultrasonic cleaning tanks could facilitate the treatment of large-scale oily sludge but the low intensity of ultrasound discourages the rate of oil recovery.

High equipment and maintenance cost also builds upon this scenario (Canselier et al., 2002; Hu et al., 2013).

2.3.7 Froth flotation

Froth flotation is a method based on the surface chemistry for the removal of fine solids from other particles in an aqueous suspension. In this method, air bubbles catch fine particles. These particles are then collected in a layer of froth. Froth flotation basically depends on the interface interactions between the solid, gas, and the continuous liquid phases. Chemical reagents must be added to introduce a balance to the interactions between solids and water and also solids and air bubbles (Urbina, 2003).

In an oil-water emulsion, when gas bubbles are added into the water phase, they attach to immiscible oil droplets. The oil droplets then rise to the surface of the flotation unit as a result of an increase in the difference between oil and water density. The oil phase then gets trapped in a foam that will be removed later.

A reduction in the viscosity of the continuous phase by increasing the temperature would be beneficial. Moreover, flotation best works with a drop diameter range of 20-150 µm (Moosai and Dawe, 2003).

Froth flotation has a vast application for different purposes including minerals industry, coal cleaning, plastics separation, wastewater treatment, oily sewage treatment, pulp and paper production, and food and chemical processing (Urbina, 2003).

Research conducted on froth flotation exhibited successful utilization of this method in the treatment of oily wastewater. However, its application to oil sludge treatment has recently come to attention. Studies showed that up to 55% of oil removal from oily sludge could be achieved with this method (Ramaswamy et al., 2007).

There are some disadvantages associated with using froth flotation for oily sludge treatment including the high concentration of moisture in the recovered oil and necessity for pre-treatment to decrease the viscosity of oily sludge and removal of the coarse solids. Furthermore, the addition of a large amount of water for treating highly viscous oily sludge might generate an oily wastewater by-product (Hu et al., 2013).

2.3.8 Surfactant enhanced oil recovery (EOR)

Surfactants are successfully applied to different systems for fast and economical removal of organic contaminants. The chemical structure of a surfactant provides its molecule with hydrophilic and hydrophobic ends. The hydrophilic tail enhances the solubility of PHCs by dissolving the surfactant molecule in water. Hydrophobic tail, on the other hand, makes the molecules accumulate at the interfaces. Therefore, the surface tension decreases and results in the increased mobility of PHCs (Hu et al., 2013; Mulligan, 2009).

Choosing an appropriate surfactant relies mostly on its effectiveness, the associated costs, biodegradability, products of degradation, and toxicity. Industrial application of bio-surfactants is restricted due to their costly production process which could be decreased by using wastes and enhancement of recovery (Calvo et al., 2009; Hu et al., 2013).

2.3.9 Electrokinetics

The emerging amount of hazardous waste threatens the environmental and human health; Therefore, a technology that promotes reuse and recycling of the oily sludge within the frameworks of sustainable management must be considered (da Silva et al., 2012).

Recycling oil and water from oily sludge results in decreasing the volume of pollutants, minimizing the contaminants in the waste, recovery of energy, and saving natural resources. Electrokinetics is a remediation technique that meets these criteria by a high recovery of hydrocarbons, reducing the cost and time associated with the treatment and disposal of oil waste and reduction of the waste volume by extraction of water from oil sludge. Moreover, refineries use rectangular pools for oily sludge collection before its treatment and disposal. A significant amount of money would be saved if these pools are utilized as the electrokinetic treatment units (Elektorowicz et al., 2006). Besides, the electrokinetic technology helps decrease the emission of main greenhouse gases including CO_2, CH_4, and N_2O by 40,026,000, 1057, and 566 kg per refinery per year, respectively (Elektorowicz and Habibi, 2005).

There are particular advantages to the electrokinetic method that make it worthwhile. It could be an ex-situ or in-situ approach to remediate a wide range of contaminated matrices including low-permeable soils. It is suited for the treatment of saturated or unsaturated soils or any mixture of different contaminants such as heavy metals, organic contaminants, and radionuclides. Moreover, the EK could be combined with other treatment methods including the barrier systems (Reddy and Cameselle, 2009).

In electrokinetic treatment, at least two electrodes are inserted into the matrix and are exposed to a low direct current. Several processes including the transport and transfer of the contaminants to the electrodes take place under the exposure of the electric field. These contaminants could then be removed at the respective electrodes (Reddy and Cameselle, 2009).

Electrokinetics relies on three different phenomena: I) electro-osmosis or the transport of the liquid phase from the anode to the cathode, II) electro-migration of the ions towards the electrode with an opposite charge, and III) electro-phoresis or the movement of the colloidal particles to the anode (Virkutyte and Sillanpaa, 2002).

Mhatre et al. (2015) showed that exposure to an electric field would induce the polarization of each drop in an emulsion. The polar drops with positive and negative ends attract each other as a result of the electrostatic force between opposite charges. The electrostatic and molecular forces between the two drops gradually reduce the thickness of the thin film of the medium liquid that exists between them. Thus, the film finally breaks and the drops attach together. This defines the electro-coalescence of two drops.

Pamukcu et al. (2016) investigated the transport of non-polar oil particles in the oil-polluted clay-rich sediment under the exposure of the direct current electric field. They proved that the electroosmotic flow of the polar and conductive water phase and the oil phase would take place in opposite directions. This kind of movement was facilitated by the reduction in surface tension of the water phase. The batch experiment that was conducted on the oil and water phase proved the possibility of phase separation and recovery of light crude oil from the polar water phase under the exposure of the DC field.

Yang et al. (2005) showed that water removal from oil sludge was effective. They concluded that an optimal electrical intensity and a proper configuration of electrodes would benefit the efficiency of water removal. The vertical placement of the electrodes proved to be effective in the bench-scale experiments. The ideal electrical intensity was achieved through consideration of a balance between the water recovery percentage and energy consumption. They also proved that the horizontal placement of electrodes with an upward anodic electroosmotic flow would benefit the concurrent removal of oil and water. Thus, a three-phase separation would be achieved.

Karaminezhad and Elektorowicz (2017) investigated the effect of different direct current (DC) electrical regimes on the demulsification of oil sludge. The oil sludge matrix was exposed to constant direct current (CDC), pulsed DC (PDC), incremental DC (IDC), and decremental DC (DDC). It was concluded that the wettability of solids altered to water-wet in all cases and enhanced the movement of solids toward the anode. However, the DDC and IDC regimes resulted in a higher degree of solids' wettability alternation. Hence, an enhanced destabilization of the water-in-oil emulsion and improved phase separation was achieved.

2.3.9.1 Electrokinetic treatment of oil sludge with the implementation of various additives

Badawieh (2016) and Tang et al. (2017) conducted studies on the removal of heavy metals from sludge using electrokinetics. The last study showed that a combination of a biodegradable complexing agent and a biosurfactant would increase the efficiency of heavy metals removal from the sludge. The enhancement of efficiency is determined by the characteristics of sludge and the interaction between the heavy metals and the combined agents. The agents proved to be effective in increasing the conductivity of the system under the exposure of a constant voltage.

Gidudu and Chirwa (2019) performed studies on different configurations of electrodes that were placed inside the matrix of petrochemical contaminated soil. Electrodes were installed at fixed positions with different distances of 185, 260, and 335 mm. Two different types of biosurfactants were added to the system. The study showed that the current increased with the decrease in distance between electrodes. The increase in current enhanced the electroosmotic flow inside the reactor and facilitated the separation of soil and oil. Besides, the biodegradation of petrochemical contaminants was enhanced due to the notable bacterial growth within the first 6 days of the experiment.

Valero et al. (2011) introduced electrocoagulation as a promising technology for the treatment of the almond industry's wastewater. They proved that using the smallest amount of coagulant agents containing Fe^{3+} or Al^{3+} and placing iron or aluminum sacrificial anodes into the system would highly benefit the process. Ibeid et al. (2015) also reached the same conclusions. They reported that conditioning under the exposure of an electric field would lead to more significant water extraction from the flocs than the one observed where chemical coagulation took place in the absence of the electric field.

Elektorowicz and Oleszkiewicz (2009) conducted studies on electrokinetic remediation for the removal of mixed organic and inorganic contaminants. Results of the studies proved electrokinetic to be effective in the removal of such contaminants (e.g. lead and phenanthrene) depending on the voltage gradient, configuration of the electrodes, and the type of additives introduced to the matrix. Ammami et al. (2015) also suggested that the addition of a non-ionic surfactant and applying a periodic voltage gradient would significantly enhance the removal of PAHs and metals.

Karaminezhad and Elektorowicz (2019) investigated the impacts of titanium dioxide (TiO_2) nanoparticles and a synthesized catalyst on the electrokinetic treatment of oil sediments. They observed that the presence of the TiO_2 or the synthesized catalyst would increase the wettability of the solids (water-wet solids), and hence improve the phase separation and water removal.

Yang (2007) showed that the use of iron electrodes would enhance the demulsification of the oil-in-water emulsion. Ferric ions are generated due to the dissolution of anodic electrodes. These ions attract the surfactant molecules which are responsible for stabilizing the emulsion and result in the coalescence of the oil drops. Finally, the introduced coagulant will adsorb the coalesced drops and the destabilization of the emulsion will take place. Sodium chloride as an additive would be effective in increasing the conductivity of the system.

Fellah Jahromi and Elektorowicz (2018) conducted a study on the four-phase separation of the oily sludge (US patent 8,329,042) with implementing a controller system. They concluded that water removal mainly depended on the applied voltage gradient and time of exposure. In this case, a constant voltage gradient would allow for a continuous increase in water removal. Using the controller resulted in a higher O:W ratio and a high recovery rate of light oil with lower energy consumption.

Research by Kariminezhad and Elektorowicz (2018a) on the oily sediments, demonstrated that using electrokinetic phenomena and application of a direct current mode would benefit the liquid and solids phase separation. The separated phases showed high purity by employing the DC mode. Their study also proved to be a more effective treatment approach than the common methods including centrifugation and solvent extraction (Kariminezhad and Elektorowicz, 2018b).

Studies on the phase separation of the water-in-crude oil exhibited a major improvement in the destabilization of the emulsion in case of introducing a demulsifier into the system. This was especially observed when applying a sole electric field was not effective in demulsification of the stable emulsion (Less et al., 2008).

Mhatre et al. (2018) investigated the effects of demulsifiers on water-in-crude oil emulsion. Their research showed that demulsifiers would decrease the interface tension between water and oil and enhance the rate of electro-coalescence between two drops. The demulsifiers diffuse into the water-oil interface to encounter asphaltene molecules and other natural emulsifiers which stabilize the emulsion. The type and concentration of the demulsifiers determine the rate of demulsification.

3 Experimental methodology

Results from previous studies (Elektorowicz and Habibi, 2005; Fellah Jahromi and Elektorowicz, 2018; Habibi, 2004) on bottom tank sludge and extracted crude oil waste treatment showed their positive response to electrokinetic (EK) phenomena. Therefore, sludge showing properties of water-in-oil emulsion was used in this study. Figure 3-1 illustrates different phases of EK treatment of the oil sludge.

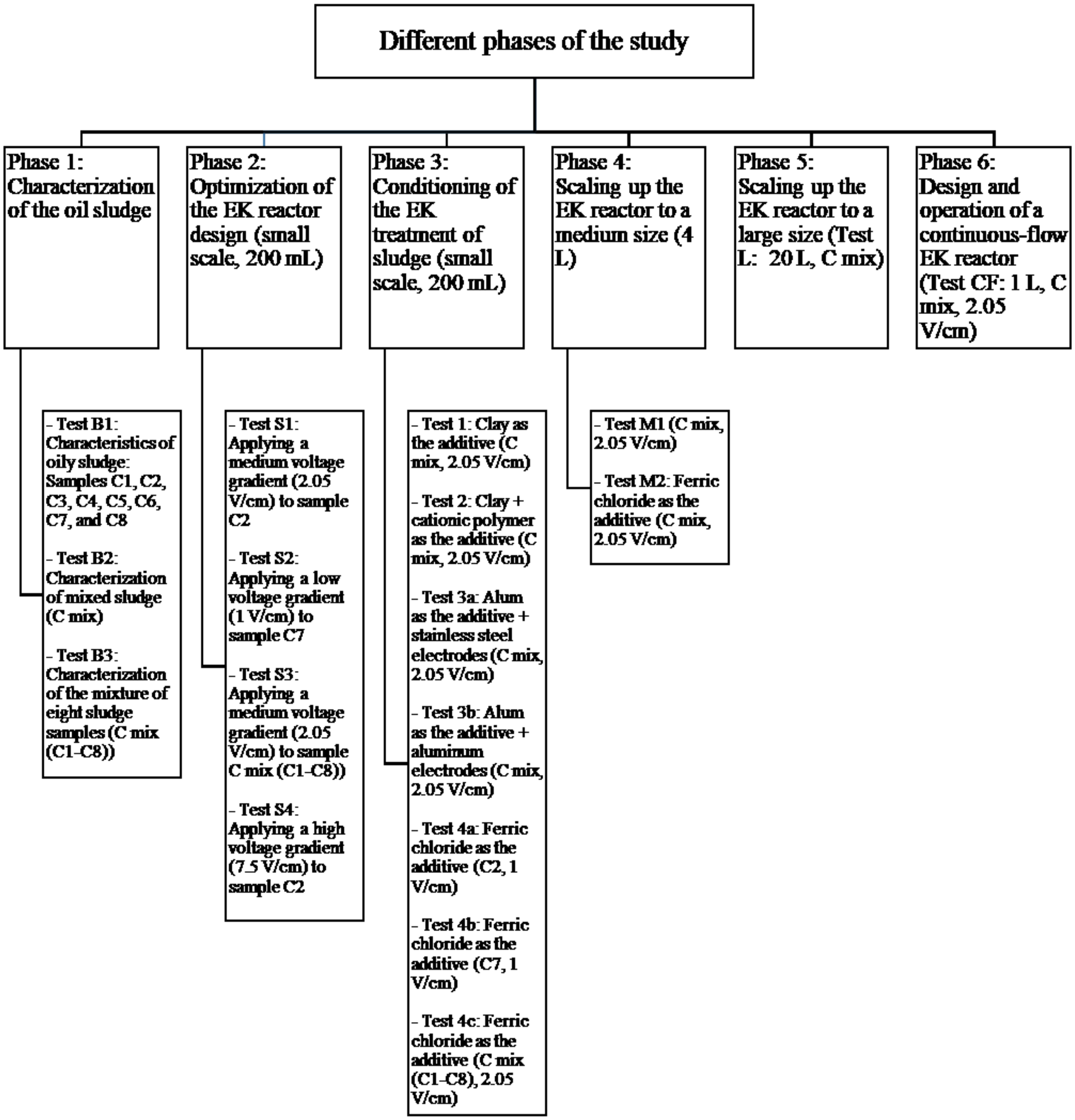

Figure 3-1 Overview of various phases of the study on EK treatment of the oil sludge

3.1 Phase 1: Characterization of the oil sludge

A petroleum company provided oil sludge in 8 buckets. Characterization of each bucket was conducted in order to keep the uniformity of all tests. Oil sludge samples taken from each of eight buckets were called: C1, C2, C3, C4, C5, C6, C7, and C8, respectively. Water, light hydrocarbons, solids, and heavy hydrocarbons contents were studied in each sample. Then, the characterization of a composite sample Cmix, (C1 to C8) and also Cmix, (C1, C3, C4, C5, C6, C8), consisting of sludge originated from different buckets were operated.

a) Water content (wt. %):

The water content that was present in oil samples, was measured according to ASTM D95-05 (American Society for Testing and Materials) (El Naggar et al., 2010). An amount of 20-25g of different samples was measured and moved into a glass vessel with a short neck and suitable joint. Then, an amount of 100 ml of toluene (solvent) was added, and the components of the device were put together, as shown in Figure 3-2. To ensure free drainage of water into the bottom of the trap, the condenser tube and trap should be clean. After adjusting the heater, the sample material was heated under reflux with a water-immiscible solvent. Condensed solvent and water (in the oily sludge sample) were continuously separated in a trap after co-distillation. The solvent returned to the still, and the volume of trap water that was settled in the graduated cylinders was read. Finally, the water content that was present in the samples was calculated by the volume percentage as follows (Taiwo and Otolorin, 2009):

$$Water\ content\ \% = \frac{(Volume\ in\ water\ trap, ml) - (Water\ in\ solvent\ blank, ml)}{Volume\ in\ test\ sample, ml} \times 100 \qquad (1)$$

Figure 3-2 Water content measurement by solvent extraction method (ASTM D95-05,2005) in the lab

b) Light oil (wt. %):

A weighed amount of an oily sludge sample was heated in an oven at 105 ˚C for 24 hours in order to find out the amount of light hydrocarbons content. The weight loss of the sample presented the water and light hydrocarbon content in the oily sludge. The light hydrocarbon content (wt. %) was calculated based on the water content that had been measured previously, as follows:

$$Light\ hydrocarbon\ content\ \% = \frac{Reduced\ mass\ in\ g}{Mass\ of\ tested\ sample\ in\ g} \times 100 - Water\ content\ in\ wt\% \qquad (2)$$

Figure 3-3 shows the oil sludge samples before and after being dried in the oven at 105 ˚C.

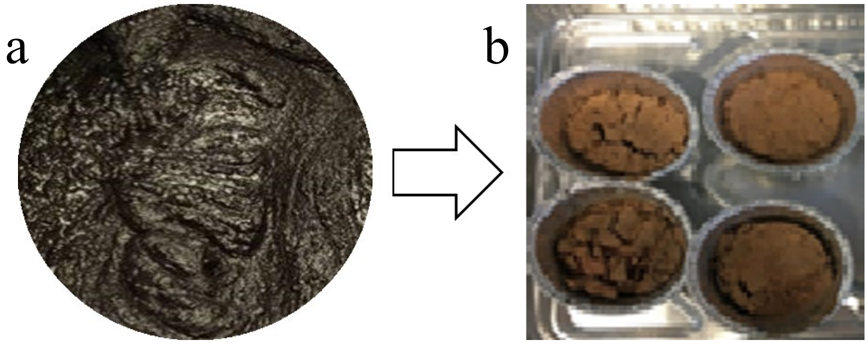

Figure 3-3 Oil sludge samples: a) raw oil sludge; b) dried samples, taken out of the oven at 105 °C

c) *Solids content (wt. %):*

Solids content is the solid materials including sediment, ash, and organics which were measured by a method described by Taiwo and Otolorin (2009) with some modifications. They put the dried sample in a furnace at 550 °C for 120 minutes. The residue indicated the weight percentage of solids content of the sludge (El Naggar et al., 2010):

$$Solids\ content\ \% = \frac{Residue\ remaining\ after\ burning\ in\ g}{Mass\ of\ tested\ sample\ in\ g} \times 100 \qquad (3)$$

d) *Heavy oil content (wt. %):*

The nonvolatile hydrocarbons content which is known as the heavy hydrocarbons content in the sludge can be calculated in weight percentage after the water content, light hydrocarbons content and solids content are determined:

$$Heavy\ hydrocarbon\ content\ \% = 100 - (Light\ hydrocarbons\ content + Solids\ content + Water\ content) \qquad (4)$$

3.1.1 Characteristics of oily sludge: Samples C1, C2, C3, C4, C5, C6, C7, and C8 (Test B1)

The characterization of each sample was conducted based on the previously described methods. The results of this characterization are presented in section 4.1.

3.1.2 Characterization of mixed sludge (C_{mix}) (Test B2)

The term "C_{mix}" indicates a mixture of sludge that consists of all samples except samples C2 and C7, which were employed mostly at the small scale tests. The characterization of sample C_{mix} was conducted three times based on the previously described methods (section 3.1), and the average of results was presented.

3.1.3 Characterization of the mixture of eight sludge samples (C_{mix} (C1-C8)) (Test B3)

The term "C_{mix} (C1-C8)" indicates the mixture of all eight sludge samples which consists of samples C1 to C8. All eight samples were mixed with the same ratio, and the same methods as described in section 3.1 were used.

3.2 Phase 2: Optimization of the EK reactor design (small scale)

The objective of phase 2 is to find the best conditions for oil sludge EK treatment; while water-in-oil sludge (in this case bottom tank) presents the most difficult petroleum waste to treat. Despite the variability of sample characteristics, it is necessary to find trends that would be applied to all such kinds of sludge. The most important parameters influencing a successful oil and water recovery are electrical parameters including voltage gradient, electrode configuration, and electrode materials. Such optimization has been required for the EK scale-up process.

Small scale tests were conducted in EK reactors that contained 193.5 g (197.4 mL) of an oily sludge sample. Flat stainless steel electrodes were placed inside the reactor at the same distance from each other and connected to a power distributor and finally to a power supply (Abra Electronique AB-5300-1) (Figure 3-4). Various voltage gradients were applied to the EK reactors in different tests. As a result, conductivity and energy consumption varied. The investigations of results are presented in Section 4.2. The characteristics of the studied raw oily sludge samples are described in Table 4-1.

The first series of tests were conducted on samples C2 and C7, the tests were continued on a mixture of samples C1 to C8. Due to the variability of the operating conditions as well as sludge characteristics, the operation conditions are defined in the same table as results (Chapter 4).

Figure 3-4 Top view of the small scale EK reactor at the beginning of a test

3.2.1 Applying a medium voltage gradient to sample C2 (Test S1)

Test S1 was conducted in a small EK reactor containing 193.5 g (197.4 mL) of oily sludge sample C2. The voltage gradient of 2.05 V/cm was applied.

3.2.2 Applying a low voltage gradient to sample C7 (Test S2)

Test S2 was conducted in a small EK reactor containing 193.5 g (197.4 mL) of oily sludge sample C7. The voltage gradient of 1 V/cm was applied.

3.2.3 Applying a medium voltage gradient to sample C_{mix} (C1-C8) (Test S3)

Test S3 was conducted in a small EK reactor containing 193.5 g (197.4 mL) of C_{mix} (C1-C8). A constant voltage gradient of 2.05 V/cm was applied.

3.2.4 Applying a high voltage gradient to sample C2 (Test S4)

Test S4 was conducted in a small accelerated EK reactor containing 193.5 g (197.4 mL) of oily sludge sample C2. A voltage gradient of 7.5 V/cm was applied, while the exposure time was 65 minutes. This Test S1imed to investigate the feasibility of reducing exposure time to 1 hour while increasing the voltage gradient with a high order of magnitude.

3.3 Phase 3: Conditioning of the EK treatment of sludge (small scale)

The objective of phase 3 was to study the effect of conditioners on the efficiency of the oil and water recovery from the sludge. Common additives in the petroleum industry were used as conditioners, namely ferric chloride, alum, a cationic polymer, and clay as well as a mixture of clay and cationic polymer. Investigations were conducted on different types of sludge and different voltage gradients. The study was performed on a small scale. Then, conclusions from these tests were applied to the medium and large scales (phases 4 and 5).

For small-scale tests, about 193.5 grams of sludge were used. This sludge was either of one type (among C1 to C8 samples) or a mixture of different types of samples. Additives were added to the system according to a specific ratio with respect to the amount of oily sludge. The application of each additive was defined by a number (e.g. Test 1, Test 2, etc.). Stainless steel or aluminum electrodes were installed in the EK reactors. The overall design of these reactors was similar to those described in phase 2. Each EK reactor was connected to a power supply, where an adequate constant voltage gradient was provided. Various technological conditions were defined by alphabets (e.g. a, b, etc.). At the end of each test, the electrodes were replaced with new ones. The characteristics of oily sludge which was used in these tests are described in Tables 4-1, 4-2, and 4-3 of the results section.

3.3.1 Clay as the additive (Test 1)

Test 1 was conducted in a small EK reactor containing 193.5 g (197.4 mL) of a mixture of oily sludge samples C1 to C8 (without C2 and C7) (C_{mix}) (Table 4-2). An amount of 5.91 g of clay was added as a conditioner. A voltage gradient of 2.05 V/cm was applied.

3.3.2 Clay + cationic polymer as the additive (Test 2)

Test 2 was conducted in a small EK reactor containing 200 grams of mixed oily sludge sample (C_{mix}) (Table 4-2). An amount of 6 g of clay (bentonite) and 1.92 g of cationic polymer was added as the additive. A voltage gradient of 2.05 V/cm was applied.

3.3.3 Introducing alum as the additive and applying stainless steel electrodes (Test 3a)

Test 3a was conducted in a small EK reactor containing 193.5 g (197.4 mL) of mixed oily sludge sample (C_{mix}) (Table 4-2). An amount of 0.67 mL of alum was added as the additive. A voltage gradient of 2.05 V/cm was applied. Stainless steel electrodes were placed in the reactor at an even distance.

3.3.4 Introducing alum as the additive and applying aluminum electrodes (Test 3b)

Test 3b was conducted in a small EK reactor containing 193.5 g (197.4 mL) of oily sludge sample (C_{mix}) (Table 4-2). A volume of 0.67 mL of alum was added. A voltage gradient of 2.05 V/cm was applied. Aluminum electrodes were set instead of stainless steel ones (as in Test 3a).

3.3.5 Ferric chloride as the additive (Test 4a)

Test 4a was conducted in a small EK reactor containing 193.5 g (197.4 mL) of the oily sludge sample C2. A volume of 0.66 mL of ferric chloride was added (1 mL of ferric chloride for each 300 mL of the oily sludge). A voltage gradient of 1 V/cm was applied.

3.3.6 Ferric chloride as the additive (Test 4b)

Test 4b was conducted in a small EK reactor containing 192.25 g (196.2 mL) of oily sludge sample C7. A volume of 0.57 mL of ferric chloride was added. A voltage gradient of 1 V/cm was applied.

3.3.7 Ferric chloride as the additive (Test 4c)

Test 4c was conducted in a small EK reactor containing 193.5 grams of a mixture of oily sludge samples C1 to C8 (C_{mix} (C1-C8)). The mixture was composed of 40.16% water, 4.88% light oil, 31.31% heavy oil, and 23.63% solids (Table 4-3). A volume of 0.68 mL of ferric chloride was added. A voltage gradient of 2.05 V/cm was applied.

3.4 Phase 4: Scaling up the EK reactor to a medium size

In order to investigate the EK reactor scale up to medium scale, Tests M1 and M2 were conducted. Analysis of the results of the previous stages of this study with respect to the upscaling led to achieving the optimal electrodes configuration and reactor design for the current stage.

The scope of phase 4 was investigating the scaling up of an EK reactor for oil recovery from oily sludge. In fact, scaling up to the medium size was conducted on 4000 mL (3920 g) of a mixture of oily sludge samples C1 to C8 except for samples C2 and C7 (C_{mix}).

3.4.1 Conducting the medium-scale (4 L) experiment on the mixture of raw oily sludge samples (Test M1)

The scaled-up EK reactor (4 L) followed a majority of the design parameters for the small-scale reactors. Based on the results from phase 2, the applied voltage gradient was set to 2.05 V/cm. The EK reactor consisted of three middle compartments, and each compartment contained approximately 1.33 L of raw oily sludge. In order to facilitate the collection of extracted liquids, the reactor was equipped with two side banks for separate anode and cathode extractions. Four electrodes permitted on sludge demulsification, phase separation and vertical and horizontal transport of separated liquids to the appropriate electrodes (Figure 3-5). Separated liquids were drained through four orifices located at the extremities of the EK reactor (Figure 3-6). Figure 3-7 presents the top view of the testing system. The characteristics of the raw oily sludge that was used in this test are described in Table 4-2. It is noteworthy to mention that Samples C2 and C7 were excluded from the mixture of oily sludge due to a shortage of available sludge samples for conducting the medium-scale experiments.

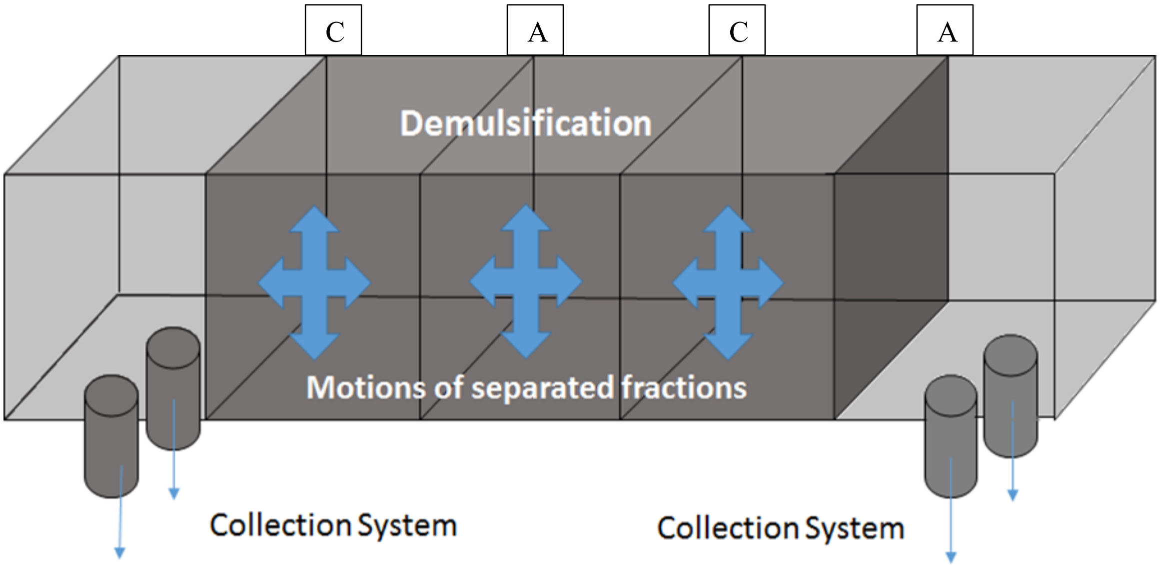

Figure 3-5 Vertical and horizontal separation of the oil sludge phases

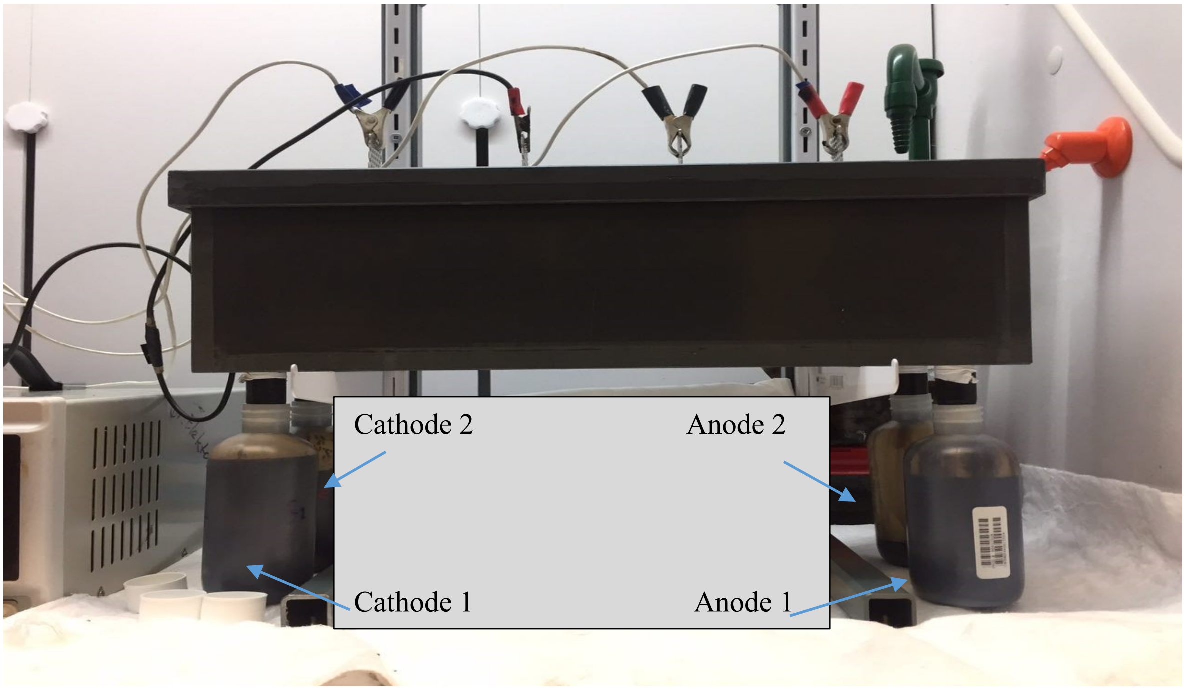

Figure 3-6 Side view of the 4 L EK reactor with bottom collection system (Test M1)

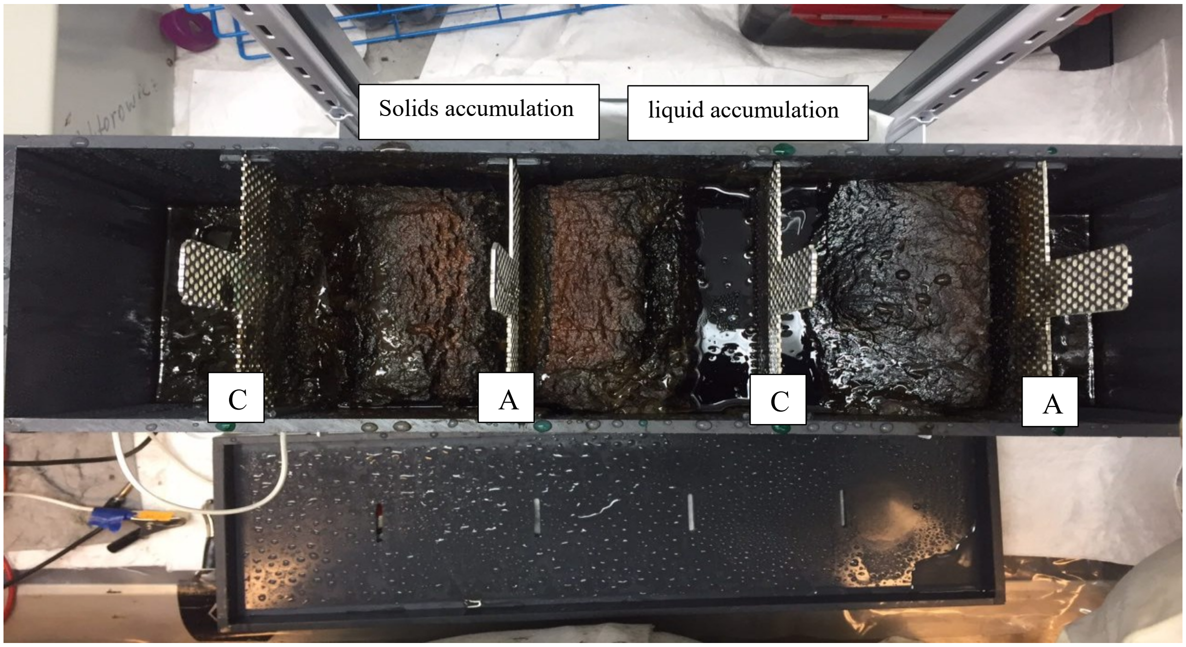

Figure 3-7 Top view of the separated phases in the 4 L reactor after EK treatment (Test M1)

3.4.2 Conducting the medium-scale (4 L) experiment on the mixture of raw oily sludge samples with the implementation of ferric chloride as the additive (Test M2)

Test M2 was conducted in a 4 L EK reactor similar to the one illustrated in section 3.4.1. the reactor contained a mixture of oily sludge samples (C $_{mix}$) (Table 4-2). Based on results from the small scale tests with additives (phase 3), a 1:300 volumetric ratio (a total amount of 13.4 mL) of ferric chloride was added to the sludge as a conditioner for the EK sludge treatment. A voltage gradient of 2.05 V/cm was applied. The exposure time was equal to 95 h, and mixing of the sludge during the treatment process took place twice.

3.5 Phase 5: Scaling up the EK reactor to a large size (Test L)

The objectives of phase 5 were: a) to scale up the EK system to a five times bigger size than the medium scale for enhanced oil recovery from the oil sludge, b) to verify the effectiveness of the EK process at a large scale, c) to verify the response of sludge to the EK system without and with a conditioner at a large scale.

The large scale 20 000 mL- EK system was prepared based on the results from scaling up the small scale reactor (around 200 mL) to the medium-scale reactor (4000 mL). Thus, the best configurations, operation parameters, and materials were used to set up the large-scale EK reactor.

A schematic view of the 20 L reactor is shown in Figure 3-8, where the potential vertical and horizontal motions of the separated fractions and the collection of liquids are also indicated. Once an electrical field is generated, demulsification of the sludge, a stable water-in-oil emulsion, takes place. Then, the separation of phases starts. Separated sludge fractions firstly undergo the electrokinetic phenomena, then, mostly electrokinetic and electrophoretic horizontal flows. Once a substantial amount of the matrix is under such flow, vertical motion of fractions is observed; a bigger mass pronounces more vertical motion. Therefore, a combination of all phenomena accelerates the phase separation process, which seems to be more efficient in a large scale than in a small one.

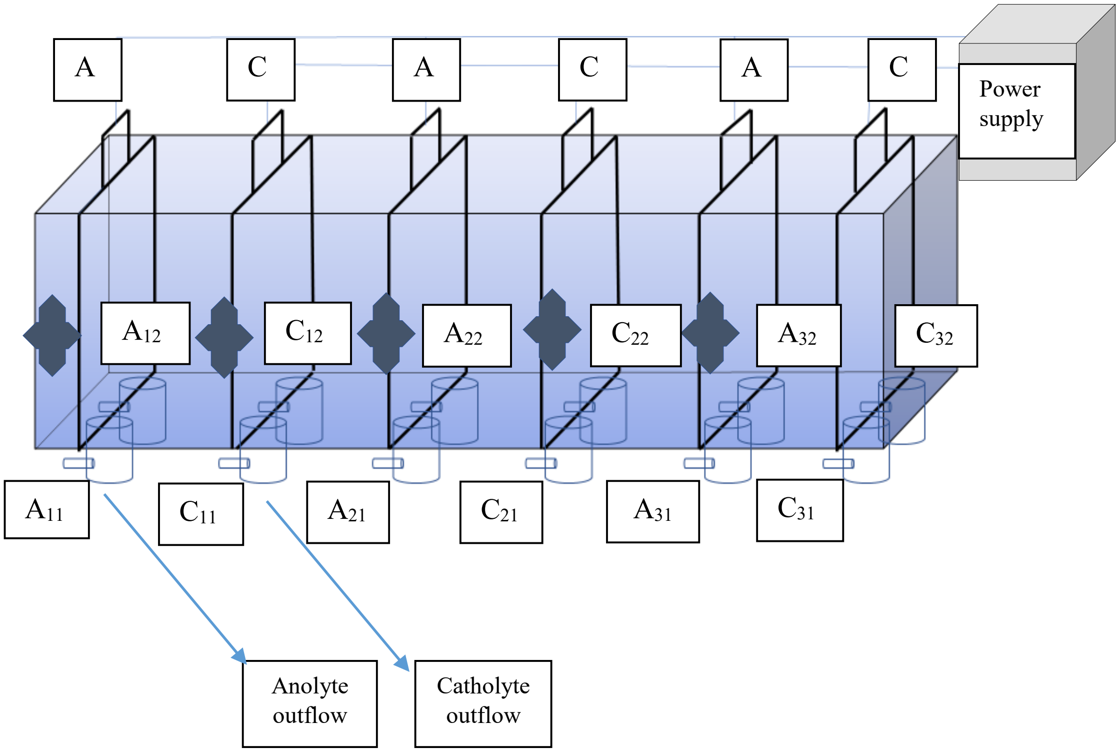

Figure 3-8 Schematic view of the 20 L reactor for the large-scale EK process

For the scale-up process, a mixture of oily sludge samples (C_{mix}) was used (Table 4-2*)*. The 20-liter EK reactor (61x21x26.5cm = LxWxH) was made of PVC. The EK reactor consisted of five compartments, divided with perforated stainless steel electrodes located at an even distance (Figure 3-9). The reactor was filled with an effective volume of 17690.4 mL of sludge mixture (Figure 3-10). Verification of the impact of a conditioner on the EK process in a large scale was done on the same oil sludge, while the conditioner (ferric chloride) was added after 5 days of the operation (without an additive).

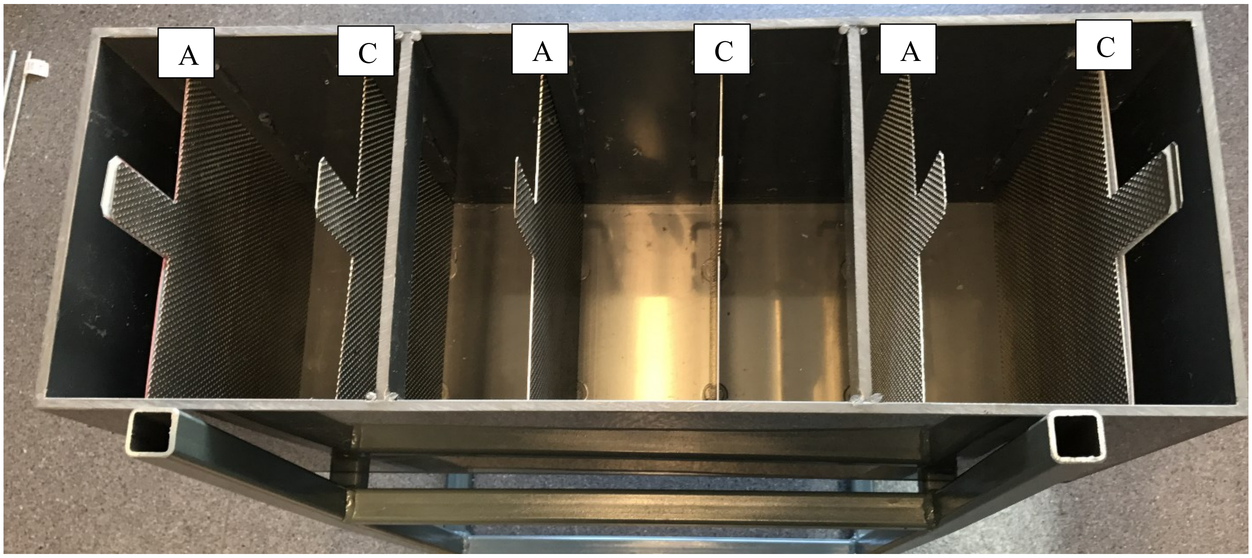

Figure 3-9 Configuration of electrodes in the 20 L EK reactor before filling

Figure 3-10 EK reactor after filling with the raw oily sludge

The system might mimic the pilot-scale design where 3 anodes (A1, A2, A3) and 3 cathodes (C1, C2, C3) were installed. The electrodes were connected to the power supply (BK precision 1902) to provide a constant voltage.

The EK reactor was equipped with 8 bottom outlets inside of the reactor and two pairs of outlets on both banks (the side bank close to A1 and the one close to C3) (Figure 3-11*)*. In the extraction process, the stainless steel filters were used to prevent solids from entering the collected catholyte and anolyte.

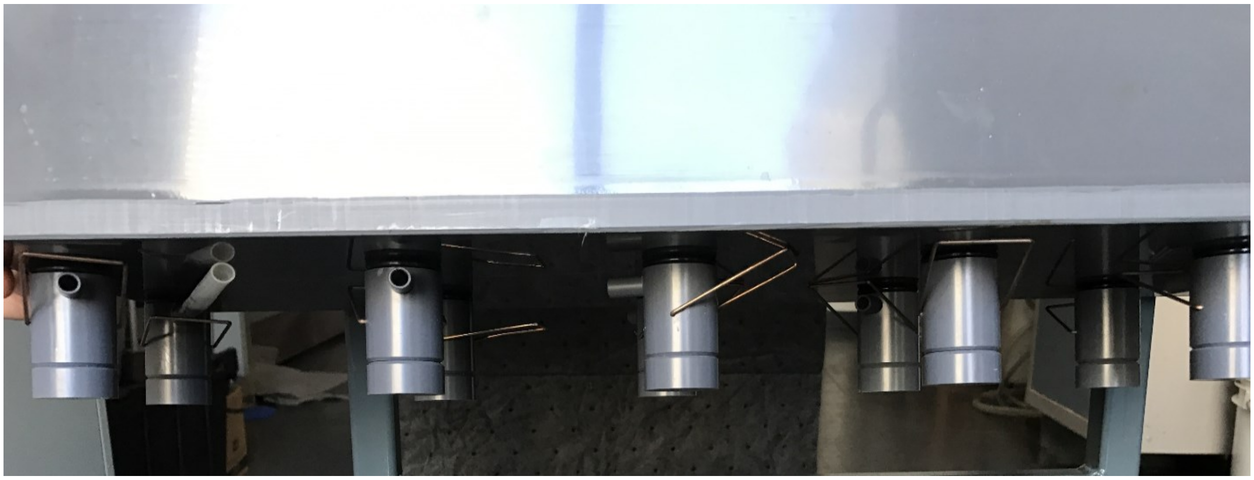

Figure 3-11 Bottom outlets of the 20 L reactor

3.6 Phase 6: Design and operation of a continuous-flow EK reactor (Test CF)

The objective of a continuous-flow EK reactor was to achieve an efficient recovery of water and oil from oil sludge within a 24-hours retention time. This kind of system would result in less energy and power consumption than the batch supply system. The in-situ application of the continuous supply system could help reduce the labor costs for manual handling of the sludge and the costs of transportation. After the sludge EK treatment, the recovered materials could be redirected to special operation units for further treatment and reuse.

A novel EK continuous system was designed based on the results of the batch experiments. The best configuration of electrodes, operating parameters, and materials were applied.

For the continuous EK process, a mixture of oily sludge samples (C_{mix}) was used (Table 4-2). The PVC reactor consisted of two perforated stainless steel electrodes that were placed horizontally and at a distance of 5 cm from each other. Figure 3-12 and Figure 3-13 exhibit different components of the electrical system and the EK continuous-flow reactor.

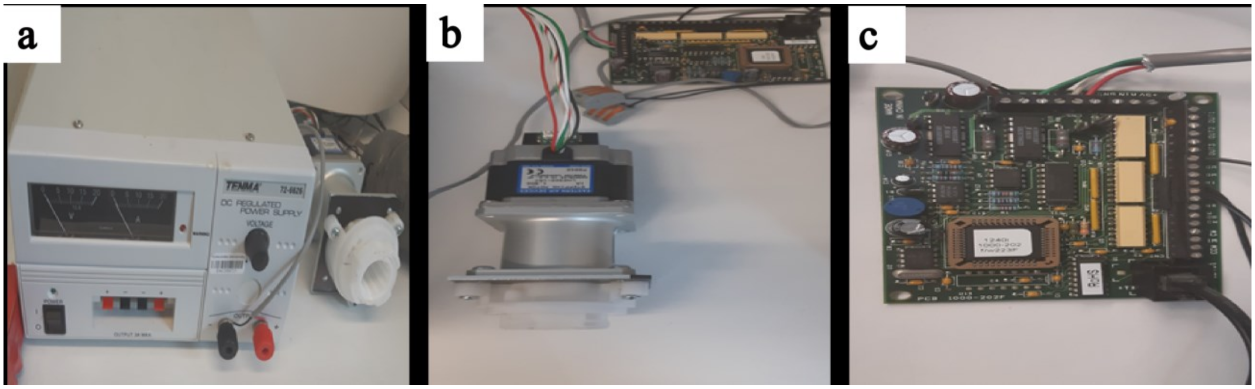

Figure 3-12 Electrical compartments for Test CF: a) power supply; b) step motor; c) circuit

The effective volume of the reactor was 1018 mL that was exposed to the electrical field while the electrodes were connected to a power supply, providing a constant voltage of 10.2 V (voltage gradient of 2.05 V/cm). A volume of 998 mL of sample Cmix was fed to the reactor over the first 6 hours of the experiment.

The Si Programmer was installed on the laptop to modify parameters and data that control the step motor through the circuit. The motor was adjusted to function at 46 revolutions per second and 2000 steps per revolution. The sludge was gradually put through the inlet of the reactor and led towards the screw, connected to the motor. An increase in the current was observed once the sludge reached the electrodes zone.

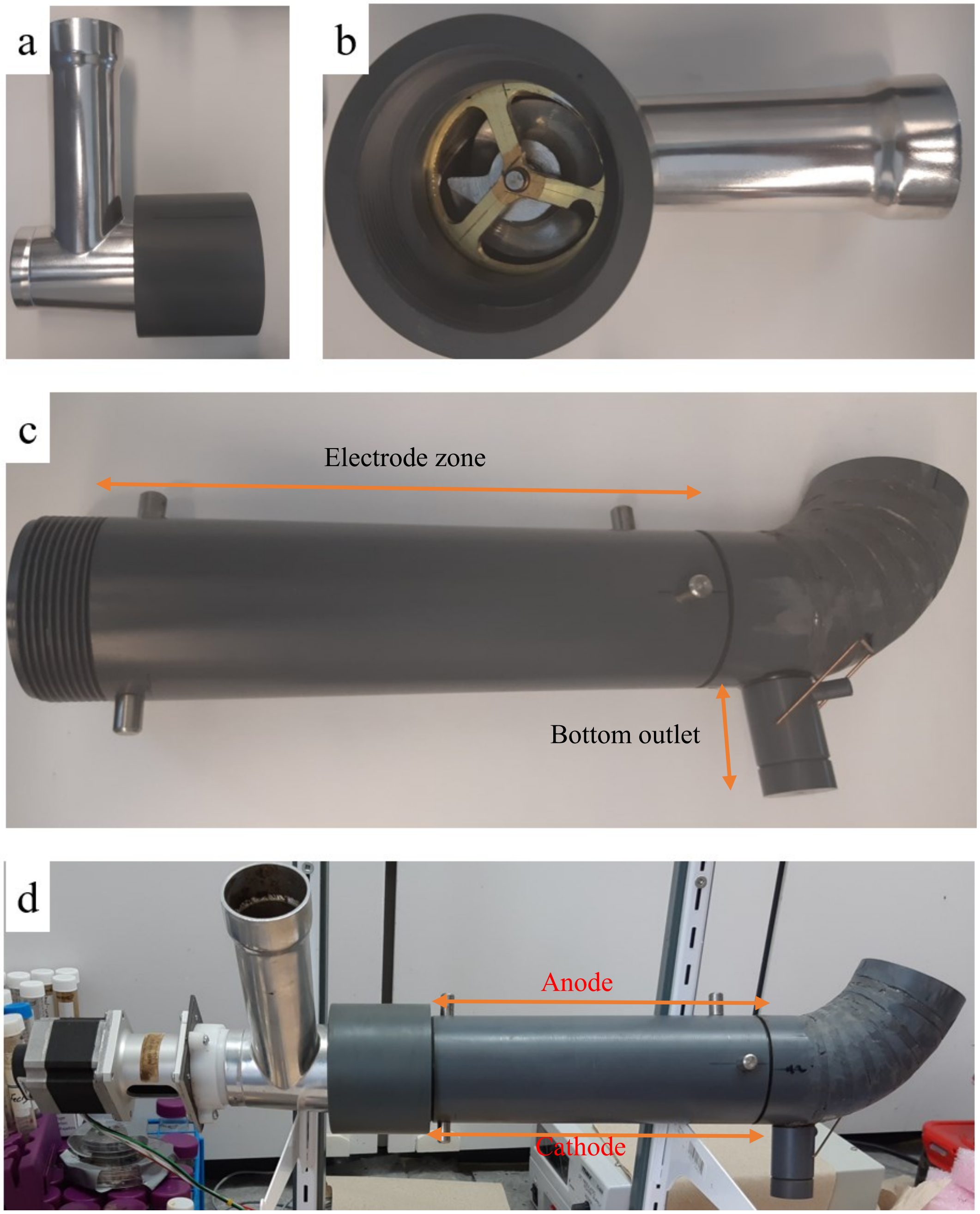

Figure 3-13 Different compartments of the reactor for Test CF: a) inlet; b) screw inside the reactor for moving the sludge forwards; c) electrode zone and the outlet section of the reactor; d) setup of the continuous EK reactor for Test CF

The reactor was equipped with one bottom outlet for the catholyte extraction. The stainless steel filters were used to prevent the solids from entering the collected catholyte. Figure 3-14 shows all elements of the operating system, prepared to initiate the experiment.

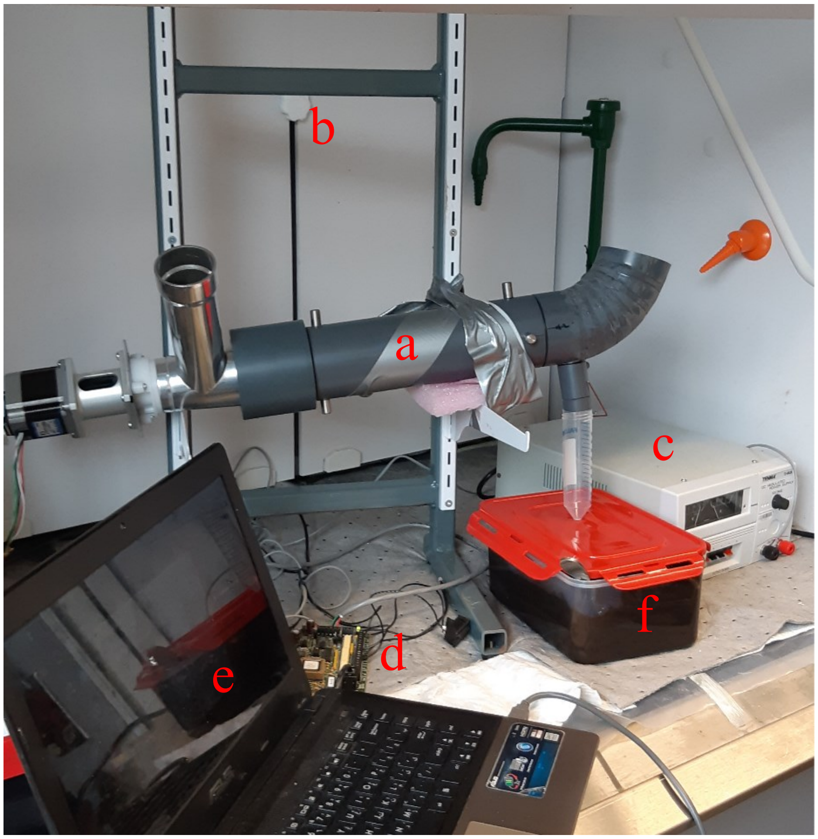

Figure 3-14 Operating elements for Test CF: a) EK reactor; b) support stand; c) power supply; d) circuit; e) Si Programmer running on PC; f) raw oily sludge sample

4 Results and discussion

4.1 Phase 1: Characterization of the oil sludge

4.1.1 Characteristics of oily sludge: Samples C1, C2, C3, C4, C5, C6, C7, and C8 (Test B1)

The results (Table 4-1) show the variation of water content between 46% to 35%; solids content between 33% to 24%, light oil content between 15% to 2% and heavy oil content between 30% to 16%. It was concluded that provided oil sludge was not uniform and varied from one bucket to another.

Table 4-1 Properties of eight oily sludge samples

Samples	Water content%	Light oil content%	Heavy oil content%	Solids content%
C1	46.11	1.91	26.64	25.34
C2	37.34	9.74	24.94	27.98
C3	42.77	3.49	29.87	23.87
C4	39.64	6.70	26.01	27.65
C5	44.87	8.23	16.79	30.11
C6	40.35	5.71	25.07	28.87
C7	43.26	7.53	16.03	33.18
C8	34.88	14.91	21.61	28.60

Due to the non-uniformity of the sludge in each bucket, the investigations on a small scale concentrated on the sludge from the same bucket to eliminate the impact of non-uniformity as much as possible. For bigger scale tests, sludge from several buckets was mixed. In fact, the provided amount of sludge permitted to proceed with one test on a large scale (20L) only.

4.1.2 Characterization of mixed sludge (C $_{mix}$) (Test B2)

Table 4-2 *s*hows the characteristics of C $_{mix}$. Sample C $_{mix}$ approximately comprises of 32.22% of water, 23.57% of solids, 13.92% of light oil, and 30.28% of heavy oil.

Table 4-2 Properties of mixed sludge (C mix)

Sample	Water content %	Light oil content %	Heavy oil content %	Solids content %
Cmix	32.22	13.92	30.28	23.57

4.1.3 Characterization of the mixture of eight sludge samples (C $_{mix}$ (C1-C8)) (Test B3)

Table 4-3 show the characteristics of sample C $_{mix}$ (C1-C8). Sample C $_{mix}$ (C1-C8) approximately comprises of 40.16 % of water, 23.63 % of solids, 4.88 % of light oil, and 31.31% of heavy oil.

Table 4-3 Properties of mixed eight sludge (C mix (C1-C8))

Sample	Water content %	Light oil content %	Heavy oil content %	Solids content %
C $_{mix}$ (C1-C8)	40.16	4.88	31.31	23.63

4.2 Phase 2: Optimization of the EK reactor design (small scale)

4.2.1 Applying a medium voltage gradient to sample C2 (Test S1)

The results are shown in Table 4-4 and 4-5.

Table 4-4 Conditions and results for Test S1

Time [h]	**Voltage [V]**	**Current [A]**	**Current after mixing [A]**	**Light oil [mL]**	**Catholyte water [mL]**	**Heavy oil [mL]**	**Catholyte* [mL]**	**Anolyte ** [mL]**
0	8.0	0.11	N/A	0.00	0.00	0.00	0.00	0.00
10	7.9	< 0.003	0.20	1.00	2.50	7.00	3.50	7.00
24	7.9	< 0.003	0.08	6.50	4.00	11.25	10.50	11.25
33	7.9	< 0.003	0.07	7.00	5.50	12.25	12.50	12.25
52	7.9	< 0.003	0.04	7.50	5.75	12.75	13.25	12.75
74	7.9	< 0.003	N/A	7.75	5.75	13.00	13.50	13.00

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side.

Table 4-5 Recovery percentages of oil and water for Test S1

Time [h]	Light oil [%]	Water [%]	Heavy oil [%]
0	0.00	0.00	0.00
10	4.72	3.46	13.20
24	30.69	5.54	21.21
33	33.05	7.61	23.10
52	35.41	7.96	24.04
74	36.59	7.96	24.51

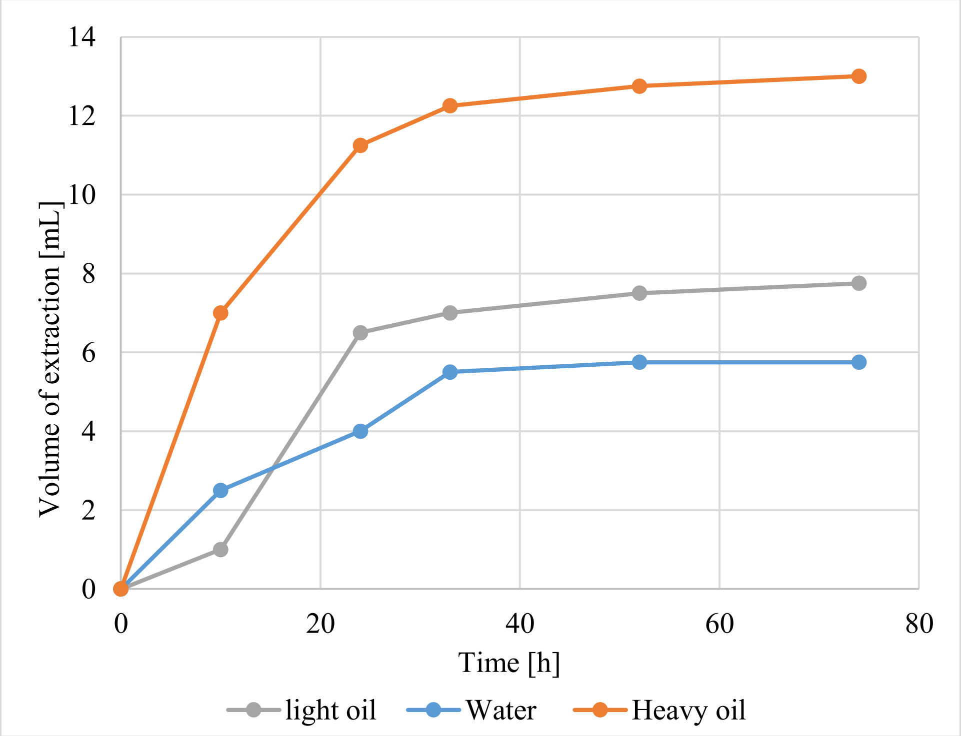

Figure 4-1 Volume of extracted light oil, heavy oil and catholyte water versus time for Test S1

Results (Figure 4-1) stated that most of the extraction took place in the first 24 hours. Light oil and water were extracted from the cathode side at a slower pace after 24 hours. The same behavior was observed for the heavy oil extraction from the anode side. No water extraction was detected from the anode side.

The total recovery percentages of light oil, water, and heavy oil after 74 hours of exposure were around 37 %, 8%, and 25 %, respectively (Table 4-5). No significant extraction was observed after 1 day of treatment.

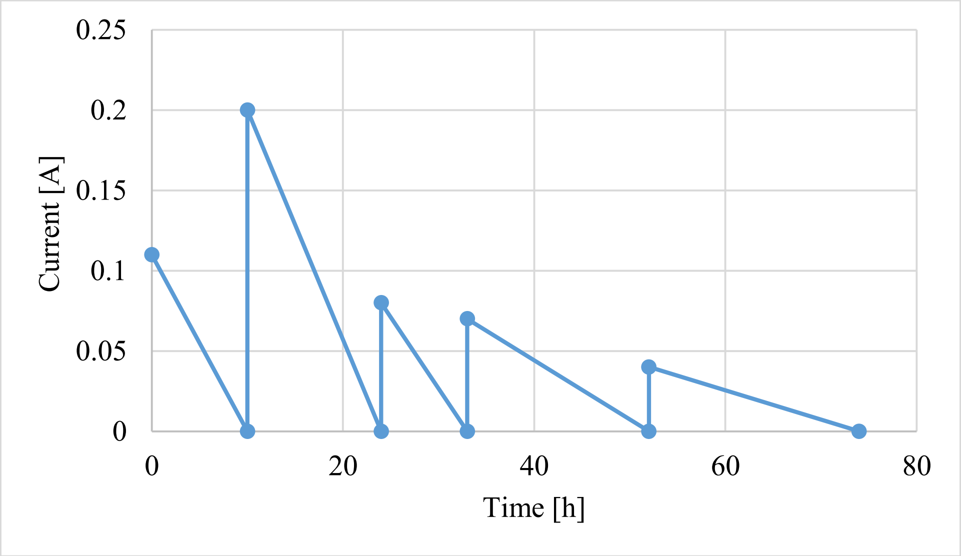

Figure 4-2 Current changes versus time for Test S1

Current variations (Figure 4-2) show that mixing the sludge inside of the EK reactor significantly influences the current value. A sharp decrease of current after 10 h, shows that the conductivity of the matrix decreased due to the transport of water to the cathode side. After mixing at 10 h, the current increased sharply to 0.2A comparing to the initial current of 0.11A in the EK cell. Current maintained in the system for the following 14 hour-period (10 h to 24 h) which showed the effectiveness of the mixing process once the voltage gradient of 2.05 V/cm was employed. In addition, the same trend was observed after each mixing (24 h and 33 h). However, the evolution of current after each mixing gave a lower value compared to the initial current (0.11 A). The final 22-hour time interval (from 52 h to 72 h), which started after mixing sludge at 52 h, gave the lowest initial current value among all periods. It shows that the conductivity of the system was significantly decreased throughout the last assessed period (from 52 h to 72 h).

4.2.2 Applying a low voltage gradient to sample C7 (Test S2)

The results are shown in Table 4-6 and 4-7.

Table 4-6 Conditions and results for Test S2

Time [h]	Voltage [V]	Current [A]	Current after mixing [A]	Light oil [mL]	Catholyte water [mL]	Heavy oil [mL]	Catholyte* [mL]	Anolyte** [mL]
0.0	3.9	0.17	N/A	0.00	0	0.00	0.00	0.00
16.0	3.9	< 0.003	0.06	6.00	0	1.00	6.00	1.00
26.0	3.9	< 0.003	0.06	7.50	3	1.75	10.50	1.75
39.0	3.9	< 0.003	0.05	8.00	3	2.50	11.00	2.50
49.5	3.9	< 0.003	0.03	8.25	3	2.50	11.25	2.50
65.5	3.9	< 0.003	N/A	9.00	3	2.50	12.00	2.50

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

Table 4-7 Recovery percentages of oil and water for Test S2

Time [h]	Light oil [%]	water [%]	Heavy oil [%]
0	0.00	0.00	0.00
16	36.65	0.00	2.93
26	45.82	3.58	5.13
39	48.87	3.58	7.33
49.5	50.40	3.58	7.33
65.5	54.98	3.58	7.33

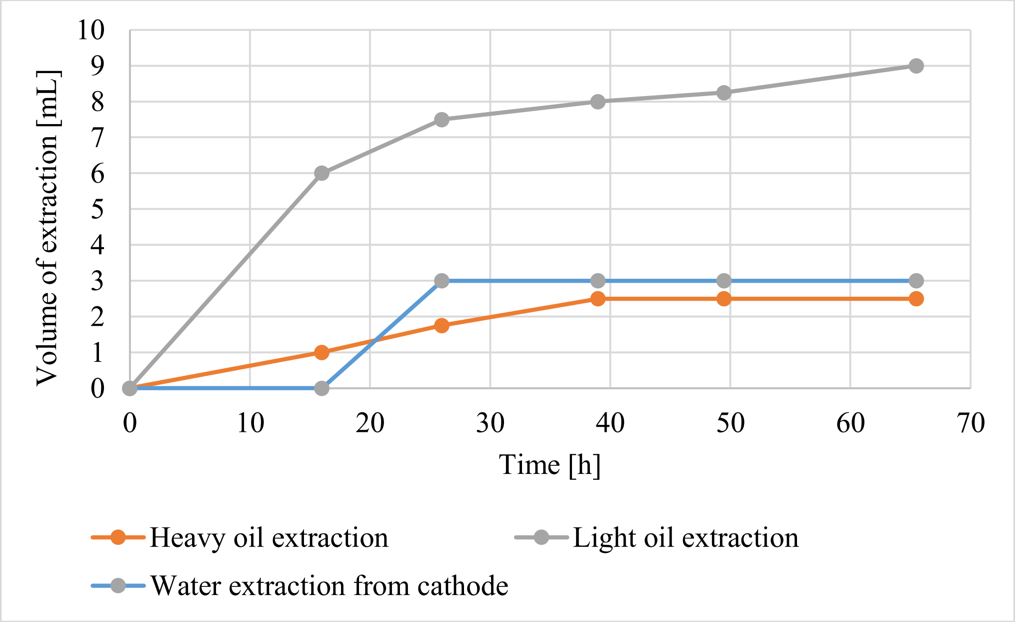

Figure 4-3 Volume of extracted light oil, heavy oil and catholyte water versus time for Test S2

The results (Figure 4-3) stated that most of the light oil extraction from the cathode side took place during the first 26 hours of the EK test. The production of heavy oil was observed at the anode side. There was no extraction from the anode side after about 39 hours. Water extraction from the cathode side started after 16 hours and stopped after 26 hours. There was no extraction of water from the anode side.

The recovery percentages of light oil and heavy oil after 65.5 hours of exposure were around 55%, and 7%, respectively (Table 4-7). In fact, no heavy oil extraction and light oil recovery of around 6% only, was observed between the second and the third days. It can be concluded that the sludge exposure time for these technological parameters can be shortened.

Due to the use of different types of sludge in Test S1 and Test S2, it is expected to have various results concerning the volume of extraction. The initial water and light oil content of sludge that was employed in Test S1 were higher by 5.92% and 2.21% respectively. However, the initial heavy oil content of the sludge in Test S1 was lower by 5.2%.

In detail, the anolyte extraction (heavy oil) of Test S1 (2.05 V/cm without additive) was significantly higher than the relevant result of Test S2 (1 V/cm, no additive). For instance, the highest cumulative volume of the extracted heavy oil was 13 mL for Test S1; while 2.5 mL of heavy oil was extracted during Test S2. Besides, the volume of extracted catholyte water was bigger by 2.75 mL for Test S1. However, results of the light oil extraction showed that applying 1 V/cm without introducing an additive led to a bigger extracted volume (9 mL) in comparison with employing 2.05 V/cm without using an additive (7.75 mL). The total volume of extracted light and heavy oil for Test S1 (2.05 V/cm, no additive) and Test S2 (1 V/cm, no additive), was 20.75 mL and 11.5 mL, respectively. With respect to the recovery of light and heavy oil (Figure 4-4), applying a voltage gradient of 2.05 V/cm resulted in the efficient involvement of horizontal flows of separated fractions towards both electrodes. Figure 4-4 compares the two tests concerning the quality of separated fractions.

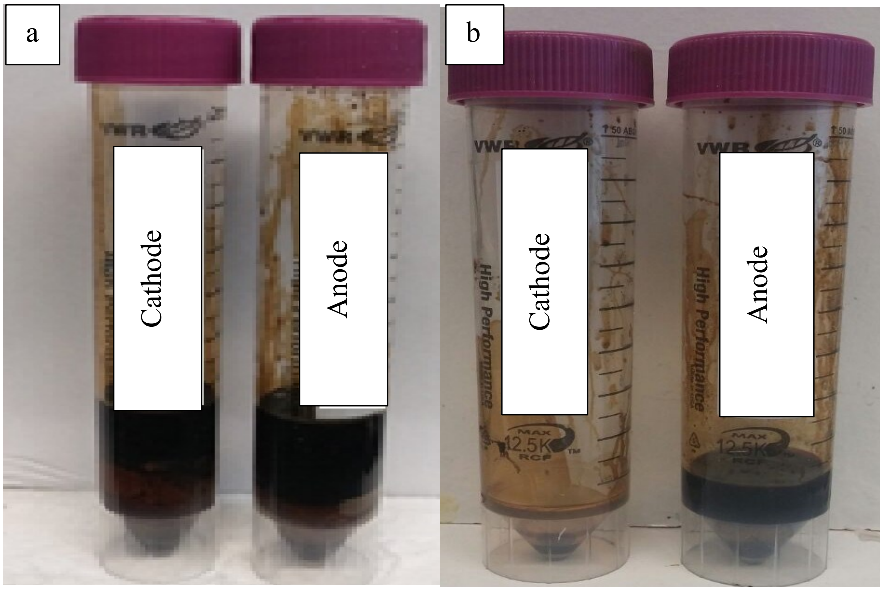

Figure 4-4 Qualitative assessment of extracted anolyte and catholyte: a) Test S1 conducted on sample C2 with the voltage gradient of 2.05 V/cm; b) Test S2 conducted on sample C7 with the voltage gradient of 1 V/cm

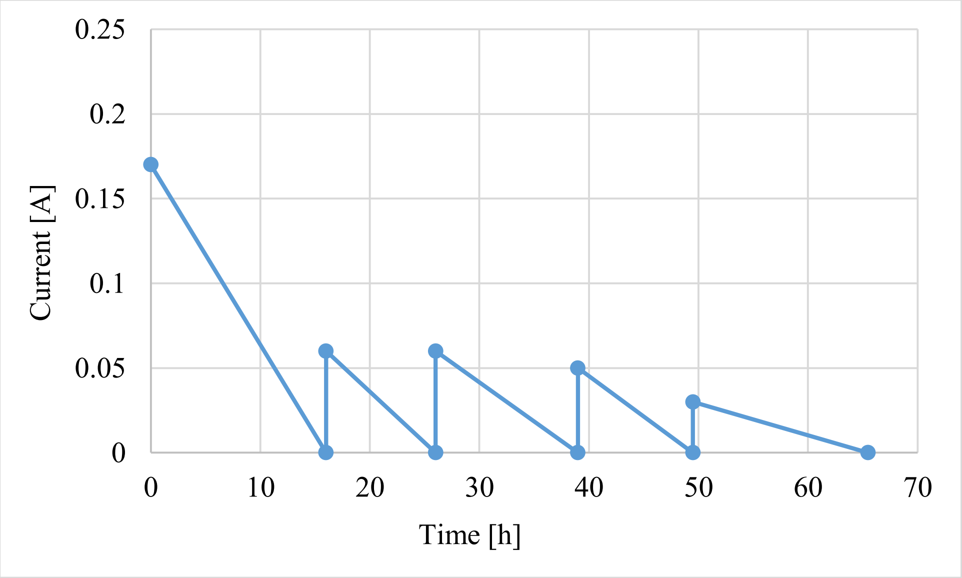

Figure 4-5 Current changes versus time for Test S2

Current variations in Test S2 (Figure 4-5) show a sharp drop during the first 16-hour period. Although the initial current was 0.17 A, it only increased to 0.06 A after the first mixing at 16 hours. It kept decreasing to a very low value from 16 h to 26 h. The same behavior was observed after each mixing until current after the last mixing reached the lowest value of 0.03 A. The current variations in Tests S1 and S2, showed a similar range of numbers. It can be concluded that in addition to a constant voltage, the behavior of electrochemical transport of oil (light and heavy oil) and water was governed by current (i.e. trend of its variation). The current could not exceed 0.2 A once the raw oily sludge samples C2 and C7 were the targets for treatment.

4.2.3 Applying a medium voltage gradient to sample C $_{mix}$ (C1-C8)) (Test S3)

The results are presented in Table 4-8 and 4-9. A constant voltage of 8 V was applied to the system.

Table 4-8 Conditions and results for Test S3

Time [h]	Current [A]	Light oil [mL]	Catholyte water [mL]	Heavy oil [mL]	Anolyte water [mL]	Catholyte* [mL]	Anolyte ** [mL]
0.0	0.28	0.00	0.00	0.00	0.00	0.00	0.00
15.0	0.00	1.50	1.50	0.25	2.75	3.00	3.00
19.5	0.02	1.50	1.75	1.00	2.75	3.25	3.75
35.5	0.00	1.75	2.00	2.50	4.00	3.75	6.50
40.0	0.01	2.0	2.00	2.50	4.00	4.00	6.50
56.0	0.00	2.0	2.00	2.75	4.00	4.00	6.75

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

Table 4-9 Recovery percentages of oil and water for Test S3

Time [h]	Light oil [%]	Water [%]	Heavy oil [%]
0.0	0.00	0.00	0.00
15.0	14.14	5.47	0.38
19.5	14.14	5.79	1.50
35.5	16.49	7.72	3.76
40.0	18.85	7.72	3.76
56.0	18.85	7.72	4.13

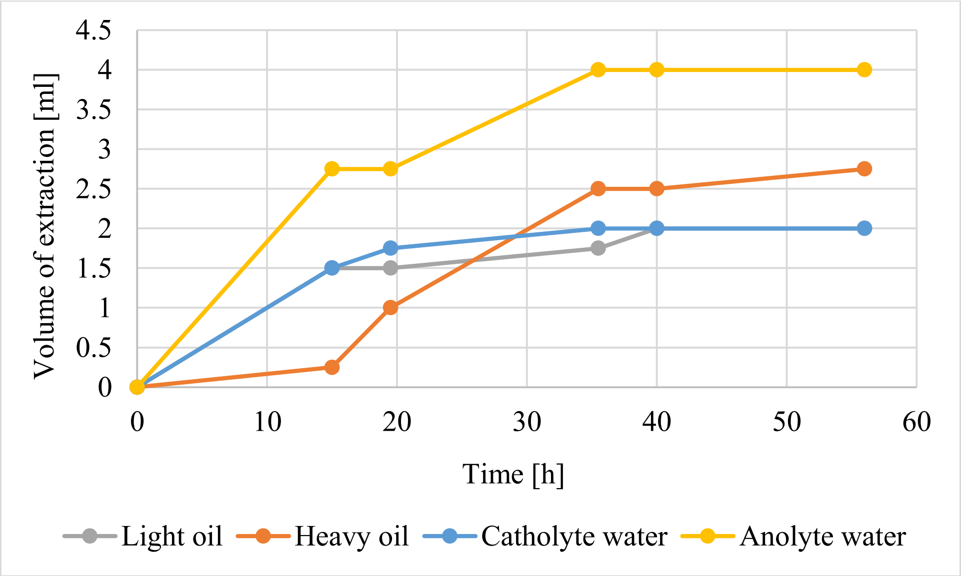

Figure 4-6 Volume of extracted light oil, heavy oil, catholyte water and anolyte water versus time for Test S3

According to the results (Figure 4-6 and Table 4-9) and observation, extraction from the cathode side mostly took place in the first 15 hours. Water was extracted from both cathode and anode sides but the volume of the extracted anolyte was twice the catholyte. Most of the anolyte extraction took place in the first 35 hours, and the overall extracted volume was more than the volume of extracted catholyte. The extraction from the cathode took a slower turn after 15 hours while the anolyte extraction continued at a slower rate after 35 hours. Recovery percentages of light oil, heavy oil, and water after 56 hours of exposure to the EK treatment were 18.85 %, 4.13 %, and 7.72 %, respectively. The qualitative assessment of Test S3 with respect to the catholyte and anolyte extraction is illustrated in Figure 4-7.

Figure 4-7 Qualitative assessment of Test S3 (voltage gradient of 2.05 V/cm, sample C mix (C1-C8)))

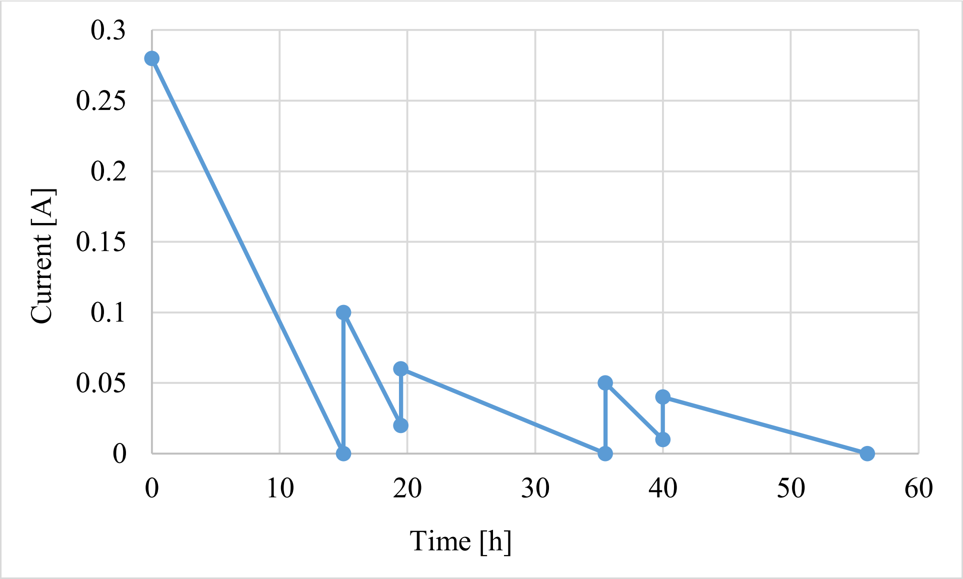

Figure 4-8 Current changes versus time for Test S3

Current changes (Figure 4-8*)* presented the highest initial value of 0.28 A. After 15 hours, the current decreased to a very low value. In that case, a mixing process was considered, which extended the period of extraction to 35.5 h. The study showed that the distribution of conductivity was mainly influenced by the type of matrix and distribution of water and conductive minerals. Water extraction from both anode and cathode sides intensified a decrease in conductivity. Comparing the trend of current variation in Test S3 (2.05 V/cm; mixed sample) with Test S1 (2.05 V/cm; sample C2) permits one to conclude that however, the initial current of Test S1 was lower than Test S3, a longer exposure time was maintained in the test.

4.2.4 Applying a high voltage gradient to sample C2 (Test S4)

Tables 4-10 and 4-11 exhibit the results and conditions of Test S4. A constant voltage of 29.1 V was applied.

Table 4-10 Conditions and results for Test S4

Time [min]	Time [h]	Current [A]	Light oil [mL]	Catholyte water [mL]	Heavy oil [mL]	Catholyte* [mL]	Anolyte ** [mL]
0	0.00	2.4	0.00	0.0	0.00	0.00	0.00
15	0.25	1.5	0.00	0.0	0.00	0.00	0.00
25	0.42	0.8	0.05	0.3	0.50	0.35	0.50
35	0.58	0.4	0.10	0.3	1.00	0.40	1.00
45	0.75	0.3	0.10	0.3	4.25	0.40	4.25
55	0.92	0.1	0.15	0.3	4.50	0.45	4.50
65	1.08	0.1	0.15	0.3	4.75	0.45	4.75

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

Table 4-11 Recovery percentages of oil and water for Test S4

Time [min]	Time [h]	Light oil [%]	Water [%]	Heavy oil [%]
0	0.00	0.00	0.00	0.00
15	0.25	0.00	0.00	0.00
25	0.42	0.24	0.42	0.94
35	0.58	0.47	0.42	1.89
45	0.75	0.47	0.42	8.01
55	0.92	0.71	0.42	8.49
65	1.08	0.71	0.42	8.96

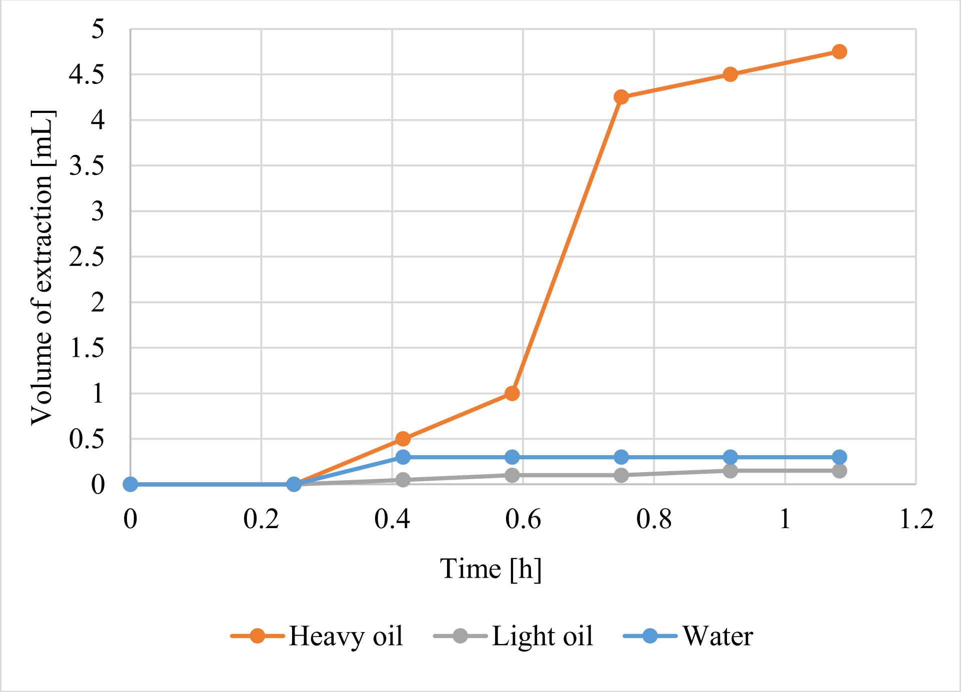

Figure 4-9 Volume of extracted light oil, heavy oil and catholyte water versus time for Test S4

The results (Figure 4-9 and Table 4-11) showed that there was no extraction, neither from the cathode nor the anode side, within the first 15 minutes. All water extraction was collected from the cathode side in a 10-minutes duration after 15 minutes of exposure to the EK treatment. However, light oil was extracted at almost a constant rate over the entire exposure time. A significant anolyte extraction was observed; heavy oil extraction exceeded its average pace after 35 minutes and resumed with a higher rate for 10 minutes before getting back to its normal pace. A Comparison between Test S1 (2.05 V/cm; sample C2) and Test S4 (7.5 V/cm; sample C2) demonstrated that the extracted volume of heavy oil in Test S4 was 2.25 mL lower than that of a 10-hour monitoring period in Test S1. On the contrary, the volume of extracted light oil and catholyte water was not significant. Recovery percentages of light oil, heavy oil, and water after 65 minutes of exposure to a high voltage were 0.71 %, 8.96 %, and 0.42 %, respectively. The qualitative assessment of anolyte and catholyte extraction (Figure 4-10) showed a remarkable separation of the heavy oil and the anolyte water phases. After comparing Test S1 (Figure 4-4 a) with Test S4, it was concluded that increasing the voltage gradient would enhance the clarity of the anolyte water.

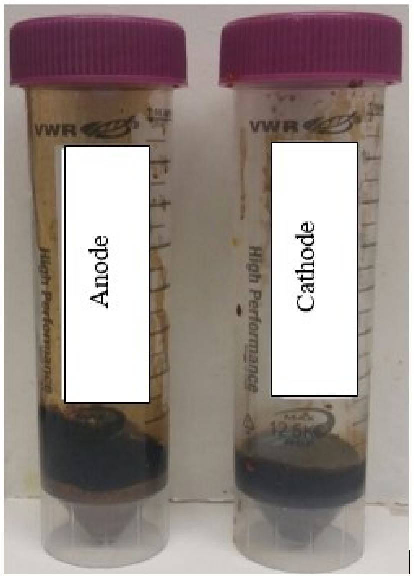

Figure 4-10 Qualitative assessment of Test S4 (voltage gradient of 7.5 V/cm, sample C2)

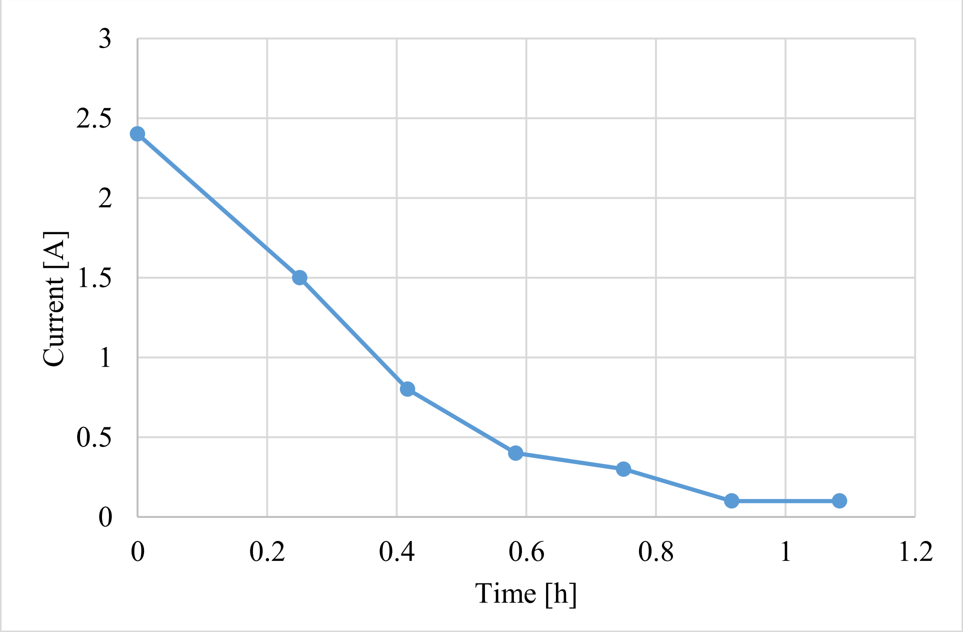

Figure 4-11 Current changes versus time for the Test S4

A fast decrease of current during the exposure time was observed (Figure 4-11*)*; however, it did not reach the zero value. This decrease was due to the corrosion that took place on stainless steel electrodes and the separation of phases inside the EK reactor. Subsequently, the material of electrodes was changed to a type that was more resistant to corrosion.

General conclusion of phase 2

The experiments of section 4.2, conducted on a small scale, reveal that the applied voltage gradient has an impact on the volume of extracted catholyte and anolyte. In addition, the clarity of anolyte water can be significantly improved once the applied voltage gradient is increased. Furthermore, two-sides recovery was enhanced once the voltage gradient was increased from 1 V/cm to 2.05 V/cm. Therefore, applying the voltage gradient of 2.05 V/cm is recommended. It is feasible to increase the voltage gradient to a value that is higher than 2.05 V/cm. However, it is important to wait at least 10 hours to have a remarkable phase separation and extraction. A significant extraction is expected to be achieved after 10 hours but it is suggested to extend the exposure time to 24 hours to achieve an enhanced oil recovery.

4.3 Phase 3: Conditioning of the EK treatment of sludge

4.3.1 Clay as the additive (Test 1)

Conditions and results are presented in Table 4-12, 4-13, and 4-14.

Table 4-12 Conditions for Test 1

Time [h]	Voltage [V]	Current [A]	Current after mixing [A]
0.0	8	0.45	N/A
2.0	8	0.16	N/A
4.0	8	0.01	0.27
18.5	8	< 0.003	0.14
26.5	8	0.01	0.10
42.0	8	< 0.003	0.03
45.0	8	0.01	N/A
48.0	8	< 0.003	N/A

Table 4-13 Results for Test 1

Time [h]	Light oil [mL]	Catholyte water [mL]	Catholyte sediments [mL]	Heavy oil [mL]	Anolyte water [mL]	Anolyte sediments [mL]	Catholyte* [mL]	Anolyte** [mL]
0.0	0.00	0.00	0.00	0.00	0.00	0.000	0.00	0.00
2.0	0.00	0.00	0.00	0.00	1.50	0.000	0.00	1.50
4.0	0.00	0.00	0.00	0.00	1.75	< 0.025	0.00	1.75
18.5	1.50	3.25	0.00	0.00	1.75	< 0.025	4.75	1.75
26.5	3.50	3.50	< 0.05	0.25	1.75	< 0.025	7.00	2.00
42.0	3.75	4.00	< 0.05	0.25	1.75	< 0.025	7.75	2.00
45.0	3.75	4.00	< 0.05	0.25	1.75	< 0.025	7.75	2.00
48.0	3.75	4.25	< 0.05	0.25	1.75	< 0.025	8.00	2.00

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

Table 4-14 Recovery percentages of oil and water for Test 1

Time [h]	Light oil [%]	Water [%]	Heavy oil [%]
0.0	0.00	0.00	0.00
2.0	0.00	2.41	0.00
4.0	0.00	2.81	0.00
18.5	4.96	8.02	0.00
26.5	11.56	8.42	0.39
42.0	12.39	9.22	0.39
45.0	12.39	9.22	0.39
48.0	12.39	9.62	0.39

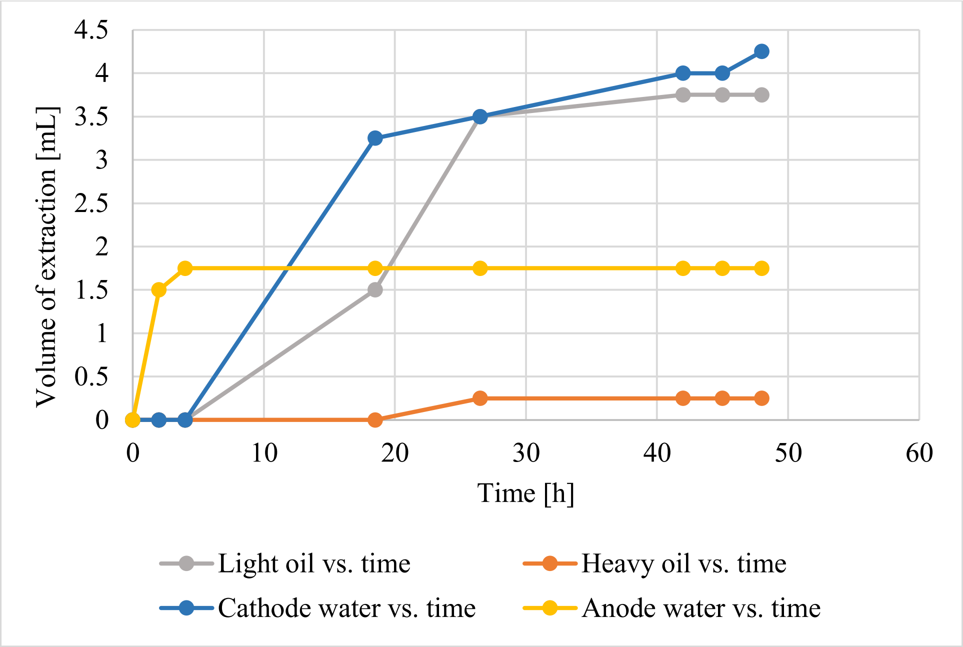

Figure 4-12 Volume of extracted light oil, heavy oil, and water versus time for Test 1

The results (Figure 4-12) illustrated a significant extraction, especially from the cathode side. There was also an extraction from the anode side which was mostly composed of water. Most of the anolyte extraction took place in the first 4 hours, while there was no extraction from the cathode side in the same period. After 4 hours, a significant extraction from the cathode side took place. This fast pace continued for the next 22 hours, so most of the cathode extraction took place in the first 26 hours. The results demonstrated a large volume of extracted catholyte water and light oil which indicated a successful development of the electroosmotic flow in the system. The positively charged ions dragged the oil along and caused the fluid to move towards the cathode. Comparing the anolyte and catholyte extraction in Test 1 with Test S3 in section 4.2 revealed that the volume of extracted anolyte is significantly lower in Test 1, while the volume of collected catholyte water is remarkably higher than the relevant value (2.00 mL) in Test S3 (section 4.2). It can be concluded that a majority of the water content moved towards the cathode side once clay was introduced to the system. The reason was the present negative surface charge of clay particles which contributed to the development of electroosmotic flow.

Recovery percentages of the light oil, heavy oil, and water after 48 hours of exposure to the EK treatment were 12.39 %, 0.39 %, and 9.62 %, respectively (Table 4-14).

The qualitative assessment of collected anolyte and catholyte (Figure 4-13) showed that the clarity of anolyte water is remarkable. Besides, significant clarity of catholyte water was achieved. Thus, if the extraction of water is a major objective of the sludge treatment, clay material might be suggested as a conditioner in EK technology.

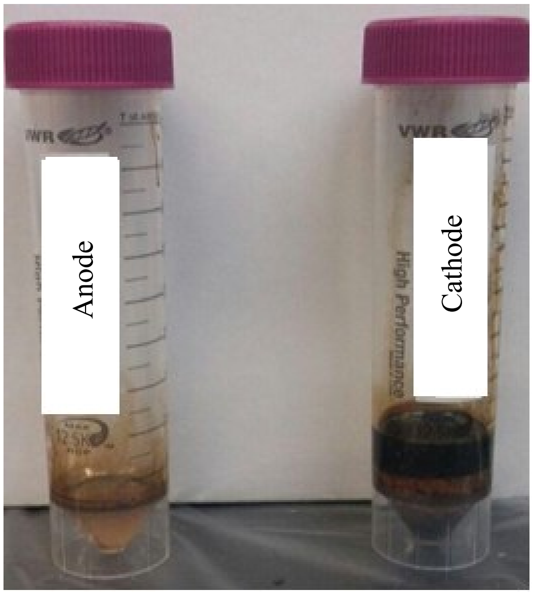

Figure 4-13 Qualitative assessment of part of anolyte and catholyte extraction for Test 1

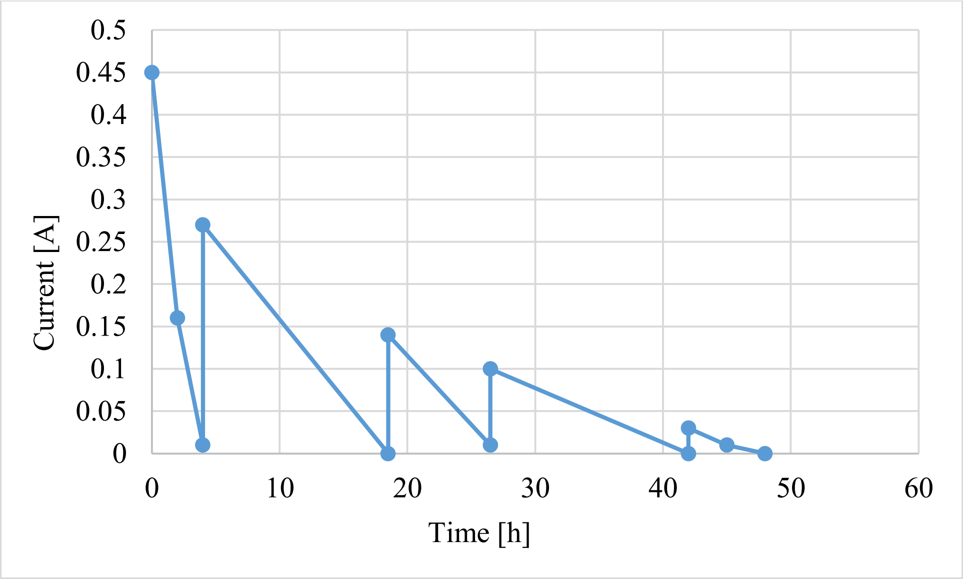

Figure 4-14 Current changes versus time for Test 1

According to Figure 4-14, The initial value of current (0.45 A) was significantly higher than the current value of Test S3 (section 4.2), with a value of 0.28 A (Figure 4-8). It showed that employing clay significantly increased the conductivity of the system. Also, the recovered current after the first mixing was equal to 0.27 A which proved the effectiveness of using clay in improving conductivity. Based on the relatively low value of current after the last mixing, it was implied that the conductivity of the system was affected by the volume of the remaining water in the reactor.

4.3.2 Clay + cationic polymer as the additive (Test 2)

The results are shown in Table 4-15 and 4-16.

Table 4-15 Conditions and results for Test 2

Time [h]	Voltage [V]	Current [A]	Current after mixing [A]	Light oil [mL]	Heavy oil [mL]	Water [mL]	Catholyte* [mL]	Anolyte** [mL]
0.0	8	0.31	N/A	0.00	0	0.0	0.00	0
2.0	8	0.20	N/A	2.50	0	0.0	2.50	0
17.0	8	< 0.003	0.25	2.50	0	2.5	5.00	0
21.0	8	0.02	N/A	2.50	0	2.5	5.00	0
23.5	8	0.01	0.21	2.50	0	2.5	5.00	0
37.5	8	< 0.003	0.13	2.50	0	3.0	5.50	0
41.0	8	0.02	0.10	2.75	0	3.0	5.75	0
55.0	8	< 0.003	N/A	2.75	0	3.0	5.75	0

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

Table 4-16 Recovery percentages of oil and water for Test 2

Time [h]	Light oil [%]	Water [%]	Heavy oil [%]
0.0	0.00	0.00	0
2.0	7.99	0.00	0
17.0	7.99	3.88	0
21.0	7.99	3.88	0
23.5	7.99	3.88	0
37.5	7.99	4.66	0
41.0	8.79	4.66	0
55.0	8.79	4.66	0

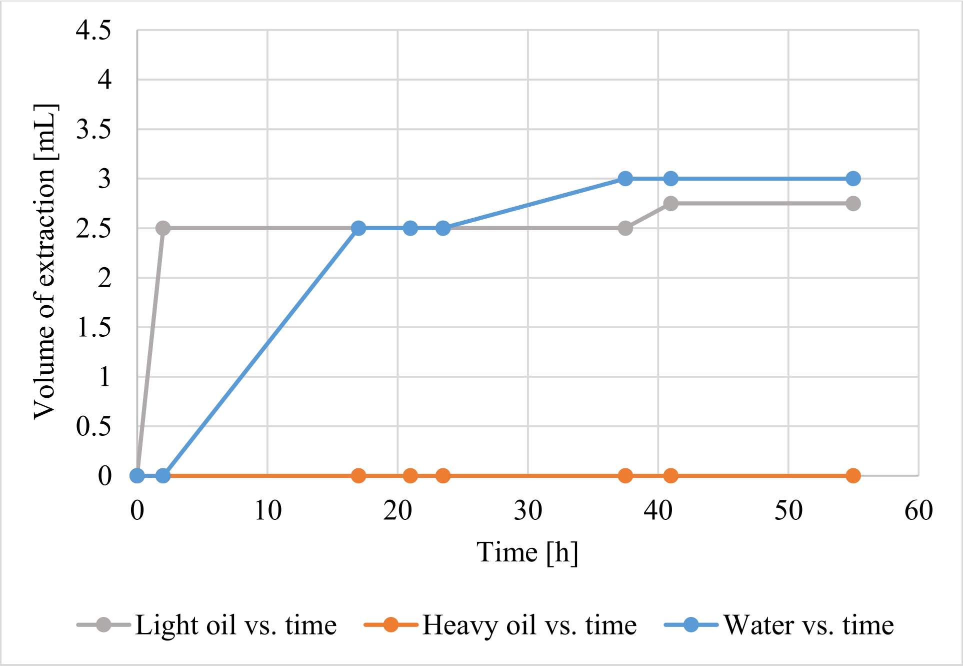

Figure 4-15 Volume of extracted light oil, heavy oil, and water versus time for Test 2

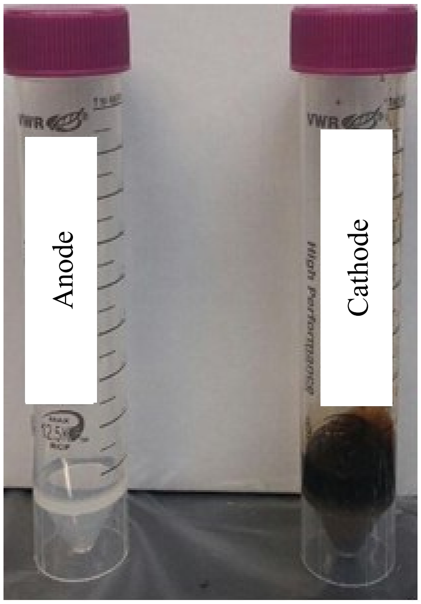

Figure 4-16 Qualitative assessment of a portion of anolyte and catholyte extraction for Test 2 (2.05 V/cm, clay + cationic polymer)

The results (Figure 4-15 and Table 4-16) showed that applying a mixture of clay and a cationic polymer prevented extraction from the anode side; however, oil and water were extracted from the cathode side. Most of the water extraction took place in the first 17 hours of the test. On the other hand, light oil was extracted at a faster pace in the first 2 hours. It was concluded that after the first 2 hours, mostly, extraction of the catholyte water took place. Due to the properties of the additives, electrokinetic motions were mostly towards the cathode. It was concluded that the viscosity of the matrix increased significantly which limited the mobility of oil and water in both anode and cathode sides. Recovery percentages of the light oil and water after 55 hours of exposure were 8.79 % and 4.66 %, respectively. No heavy oil was recovered in this period. Most of the light oil recovery took place within the first two hours; while a significant volume of water was collected within 15 hours. The qualitative assessment of Test 1 and Test 2 (Figure 4-16) demonstrated a smaller volume of water in Test 2. It determined that the combination of a cationic polymer and clay would not be effective in demulsifying water-in-oil sludge.

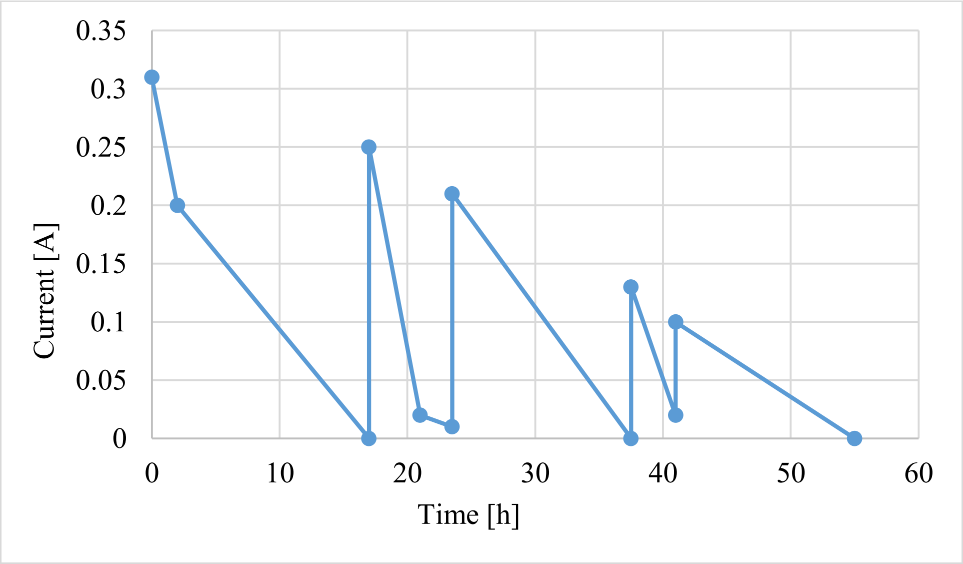

Figure 4-17 Current changes versus time for Test 2

Current variations (Figure 4-17) showed that the initial system had a conductivity of 0.31 A which was much lower than the initial current value of Test 1 (0.45 A). It indicated that the initial conductivity of the system was mainly reduced by the presence of the cationic polymer. However, the recovery of current value after each mixing was notable. Reaching a value of 0.25 A and 0.21 A after the first and second mixings, proved the efficiency of cationic polymer in maintaining current in the medium. After the final mixing, the value of current was equal to 0.1 A. It implied that the remaining water in the matrix provided conditions to maintain a high current level in the system. Although the viscosity of the matrix increased significantly after introducing the cationic polymer along with clay, the remaining fraction of water moved to the interconnected pores and contributed to the enhancement of the system's conductivity.

4.3.3 Introducing alum as the additive and applying stainless steel electrodes (Test 3a)

The results of this test are presented in Table 4-17 and 4-18. A constant voltage of 8 V was applied to the system.

Table 4-17 Conditions and results for Test 3a

Time [h]	Current [A]	Current after mixing [A]	Light oil [mL]	Water [mL]	Heavy oil [mL]	Catholyte* [mL]	Anolyte** [mL]
0	0.42	N/A	0.00	0.0	ND	0.00	ND
1	0.41	N/A	0.00	0.0	ND	0.00	ND
3	0.05	0.30	0.00	2.5	ND	2.50	ND
18	< 0.003	0.16	0.25	4.0	ND	4.25	ND
22	0.03	0.10	0.50	4.0	ND	4.50	ND
27	0.01	N/A	0.50	4.0	ND	4.50	ND
42	< 0.003	0.07	0.50	4.0	ND	4.50	ND
62	< 0.003	N/A	0.75	4.0	ND	4.75	ND

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

Table 4-18 Recovery percentages of oil and water for Test 3a

Time [h]	Light oil [%]	Water [%]	Heavy oil [%]
0	0.00	0.00	ND
1	0.00	0.00	ND
3	0.00	4.01	ND
18	0.83	6.42	ND
22	1.65	6.42	ND
27	1.65	6.42	ND
42	1.65	6.42	ND
62	2.48	6.42	ND

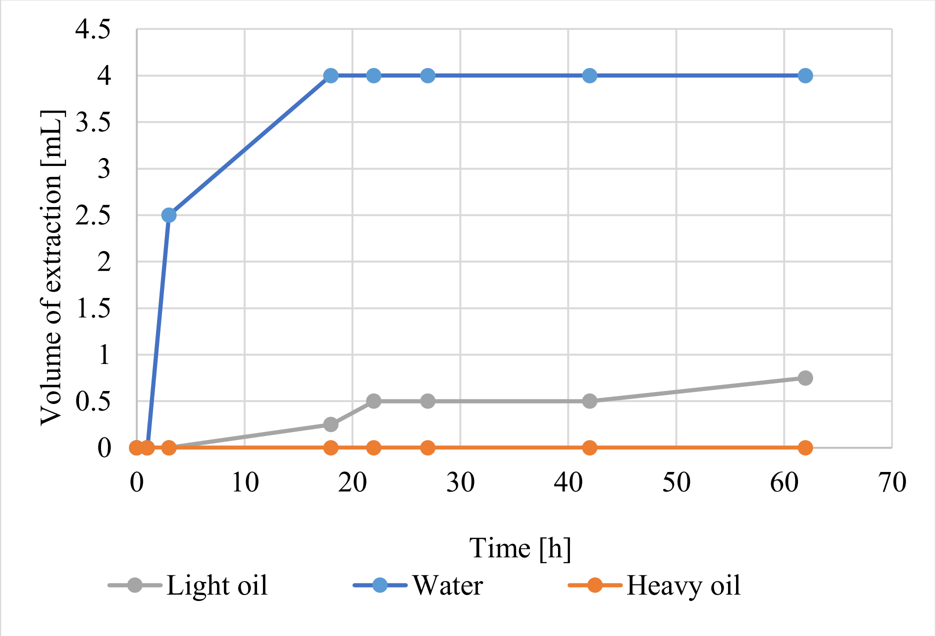

Figure 4-18 Volume of extracted light oil, heavy oil, and water versus time for Test 3a

The results (Figure 4-18 and Table 4-18) stated that the EK system with alum responded mostly to water removal; however, no anolyte extraction was observed. A small extracted volume (mostly water) was observed at the cathode (4.75 mL after 62 hours). The clarity of the water in this test was not as noticeable as the clarity of extracted water once ferric chloride was implemented (Test 4). The volume of extraction was also smaller than the time either clay or ferric chloride was introduced to the system. After the first 18 hours, no more extraction of catholyte water was observed which implied that the addition of alum in case of employing stainless steel electrodes would not be a practical approach with respect to oil extraction. Finally, the recovery percentages of the light oil and water after 62 hours of exposure were 2.48 % and 6.42 %, respectively. No heavy oil extraction was detected.

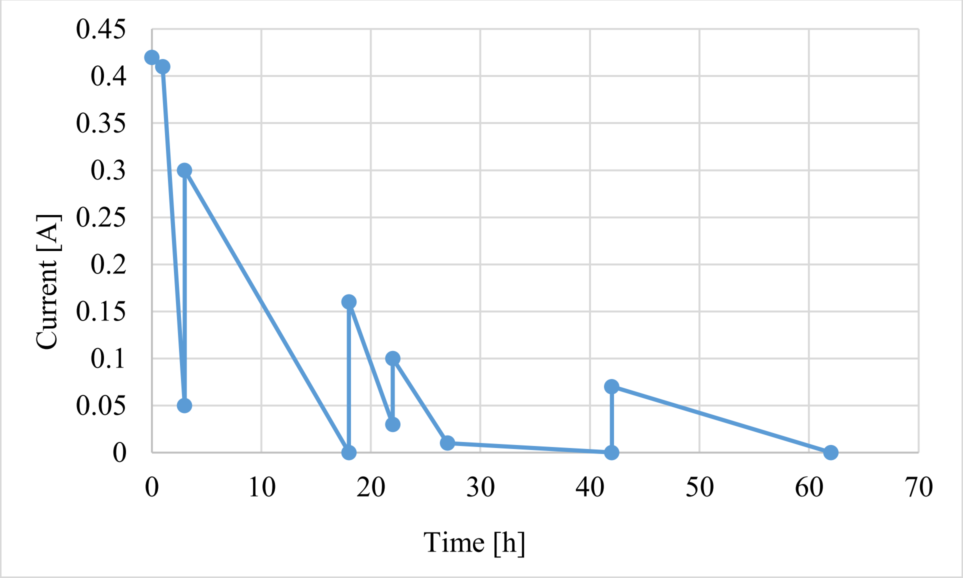

Figure 4-19 Current changes versus time for Test 3a

The initial current value (Figure 4-19) was the highest among all small-scale experiments with a voltage gradient of 2.05 V/cm. After 3 h, the value of current decreased to 0.05 A. After mixing at 3 h, the current reached the value of 0.3 A which was a relatively high magnitude. The overall trend of the current variation showed that using alum resulted in shorter effective exposure time, and the conductivity of the system met significant fluctuations.

4.3.4 Introducing alum as the additive and applying aluminum electrodes (Test 3b)

Table 4-19 and 4-20 present the results. A constant voltage of 8 V was applied to the system.

Table 4-19 Conditions and results for Test 3b

Time [h]	Current [A]	Current after mixing [A]	Light oil [ml]	Water [mL]	Heavy oil [mL]	Catholyte* [mL]	Anolyte** [mL]
0	0.36	N/A	0.00	0.0	0	0.0	0
1	0.35	N/A	0.00	0.0	0	0.0	0
3	0.16	N/A	0.00	0.0	0	0.0	0
18	< 0.003	0.22	< 0.25	3.5	0	3.5	0
22	0.03	0.14	< 0.25	3.5	0	3.5	0
27	0.02	N/A	< 0.25	3.5	0	3.5	0
42	< 0.003	0.11	< 0.25	3.5	0	3.5	0
62	< 0.003	N/A	< 0.25	3.5	0	3.5	0

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

Table 4-20 Recovery percentages of oil and water for Test 3b

Time [h]	Light oil [%]	Water [%]	Heavy oil [%]
0	0	0.00	0
1	0	0.00	0
3	0	0.00	0
18	0	5.61	0
22	0	5.61	0
27	0	5.61	0
42	0	5.61	0
62	0	5.61	0

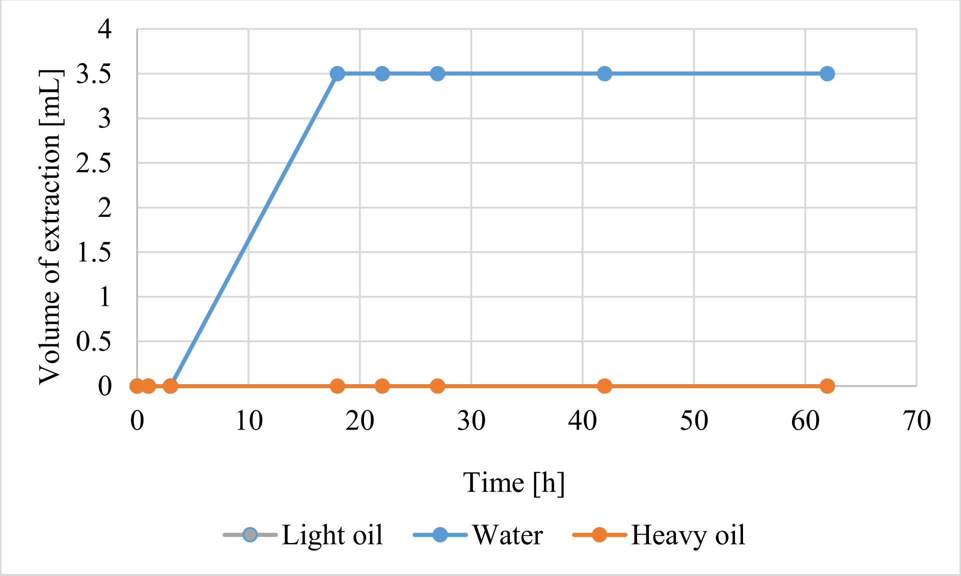

Figure 4-20 Volume of extracted light oil, heavy oil, and water versus time for Test 3b

Setting aluminum electrodes instead of stainless steel ones, where alum was applied, resulted in a small volume of extracted liquids, even smaller than that of Test 3a (Figure 4-20). In fact, no oil extraction from the system was observed. There was only a small amount of catholyte water that was extracted in the first 18 hours of the experiment. The results showed that implementing the combination of aluminum electrodes and alum would not provide the system with proper conditions for oil recovery, and no electroosmotic flow was developed in the system. The recovery percentage of water after 18 hours of exposure to EK treatment was 5.61%, and it never changed in spite of the exposure to EK treatment for over 60 hours (Table 4-20). The qualitative assessment of extracted anolyte and catholyte for both Test 3a and Test 3b are provided in Figure 4-21. Water quality in both tests seemed to be adequate, however, the overall quantity was insufficient. Considering no anolyte extraction in both tests, it could be concluded that using alum did not permit an adequate separation of phases and their two-sides transport. It was found that alum would not be applicable to oil recovery from oil sludge.

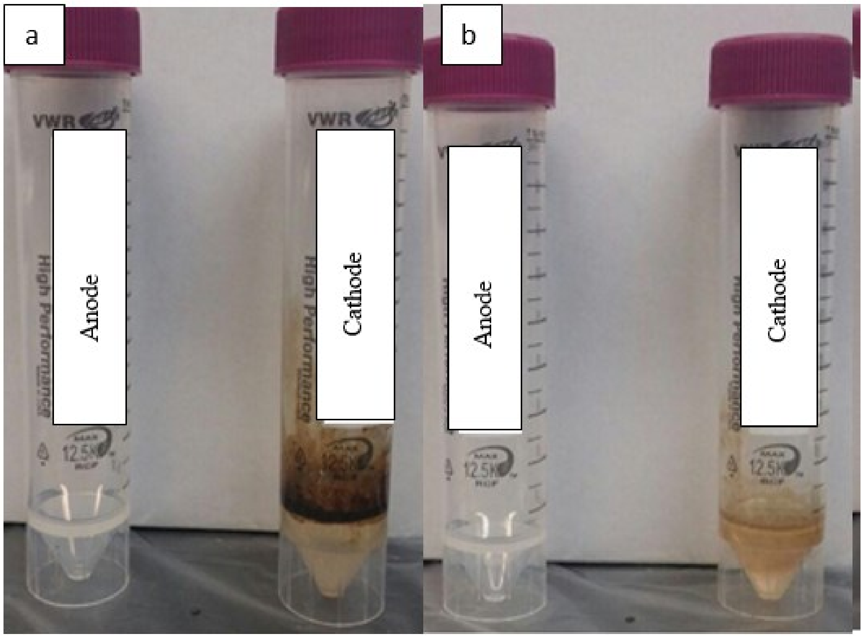

Figure 4-21 Qualitative assessment of extracted anolyte and catholyte: a) using stainless steel electrodes, alum, voltage gradient of 2.05 V/cm; b) using aluminum electrodes, alum, voltage gradient of 2.05 V/cm (same sludge was used in both cases)

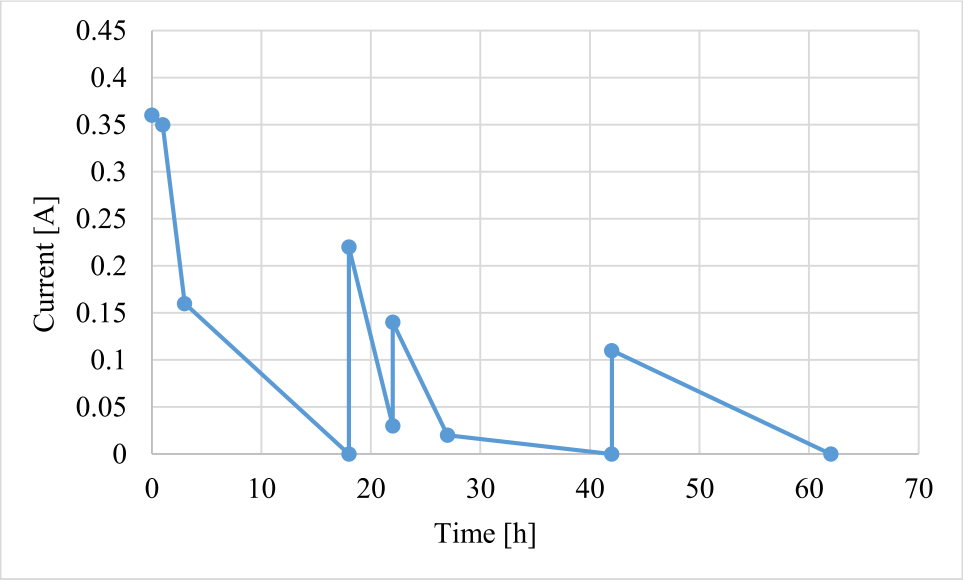

Figure 4-22 Current changes versus time for Test 3b

The initial current value of 0.36 A (Figure 4-22) was observed which was around 0.1 A lower than the initial current value of Test 3a. Therefore, using the stainless steel electrodes resulted in the generation of a bigger current flux in the system. Using the aluminum electrodes helped maintain the current in the system for longer exposure time, so the first mixing was only required after 18 hours. The trend of variation in this test was identical to the trend in Test 3a. In addition, the current value after the final mixing was higher than that of Test 3a. Once more, it was related to the prevention of water extraction from the matrix.

4.3.5 Ferric chloride as the additive (Test 4a)

The results are shown in Table 4-21 and 4-22. This test was conducted on sample C2 (small scale) and under the exposure of a 1 V/cm voltage gradient (a constant voltage of 3.9 V).

Table 4-21 Conditions and results for Test 4a

Time [h]	Current [A]	Current after mixing [A]	Light oil [mL]	Catholyte water [mL]	Heavy oil [mL]	Catholyte* [mL]	Anolyte** [mL]
0	0.16	N/A	0.00	0.00	0.00	0.00	0.00
14	< 0.003	0.12	2.25	6.25	0.25	8.50	0.25
32	< 0.003	0.06	5.00	6.25	3.00	11.25	3.00
50	< 0.003	0.02	5.75	6.25	3.25	12.00	3.25
70	< 0.003	N/A	6.00	6.50	3.25	12.50	3.25

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

Table 4-22 Recovery percentages of oil and water for Test 4a

Time [h]	Light oil [%]	Water [%]	Heavy oil [%]
0	0.00	0.00	0.00
14	10.62	8.65	0.47
32	23.61	8.65	5.66
50	27.15	8.65	6.13
70	28.33	9.00	6.13

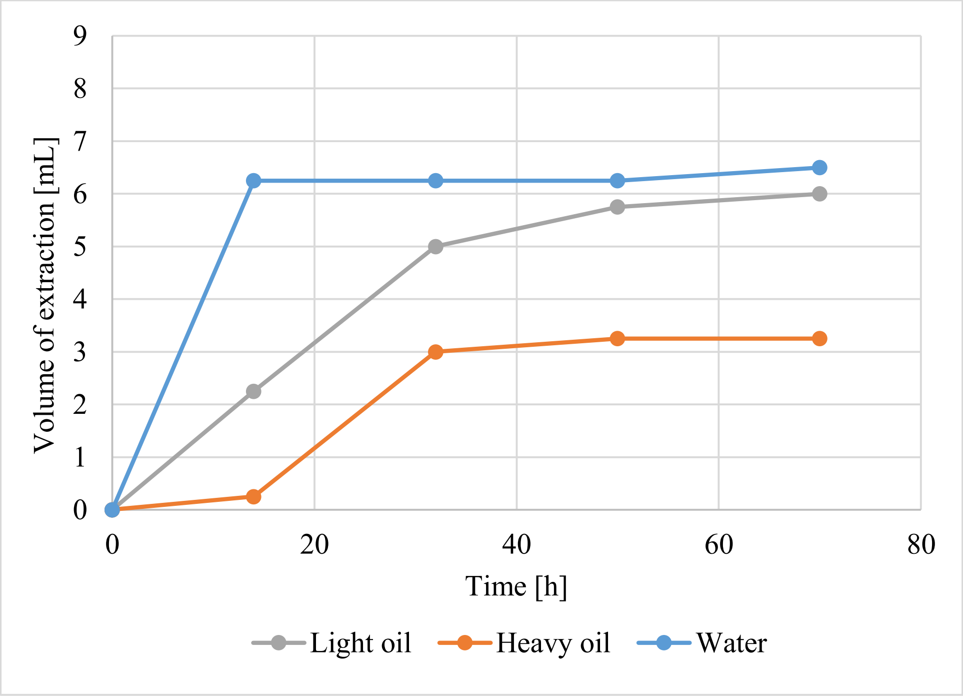

Figure 4-23 Volume of extracted light oil, heavy oil, and water versus time for Test 4a

The results (Figure 4-23) showed a fast oil extraction from both cathode and anode sides within the first 32 hours of the experiment; after this period, the oil extraction took place at a much slower pace. Furthermore, most of the water extraction was achieved within the first 14 hours. There was no water extraction from the anode side. The resemblance of the evolution of light oil and catholyte proved that using ferric chloride as the additive led to successful demulsification of water-in-oil sludge. In addition, after a few hours, no extraction of water was justified by a reduction in surface tension and proved the wettability alteration of the system from oil-wet to water-wet. The reason might be a very strong affinity of water to solids, while oil extraction was being enhanced in the absence of viscous forces of water. The previous works (Elektorowicz et al., 2006) demonstrated that lower voltage and longer exposure time might be more effective in breaking the water affinity forces.

The recovery percentages of light and heavy oils and water after 70 hours of exposure to EK treatment were over 28%, 6%, and 9%, respectively. No substantial extraction of oils was observed after 32 hours, while extraction of water started to be insignificant only after a few first hours of the experiment (Table 4-22). It can be concluded that a shorter retention time can be considered for this combination of technological parameters.

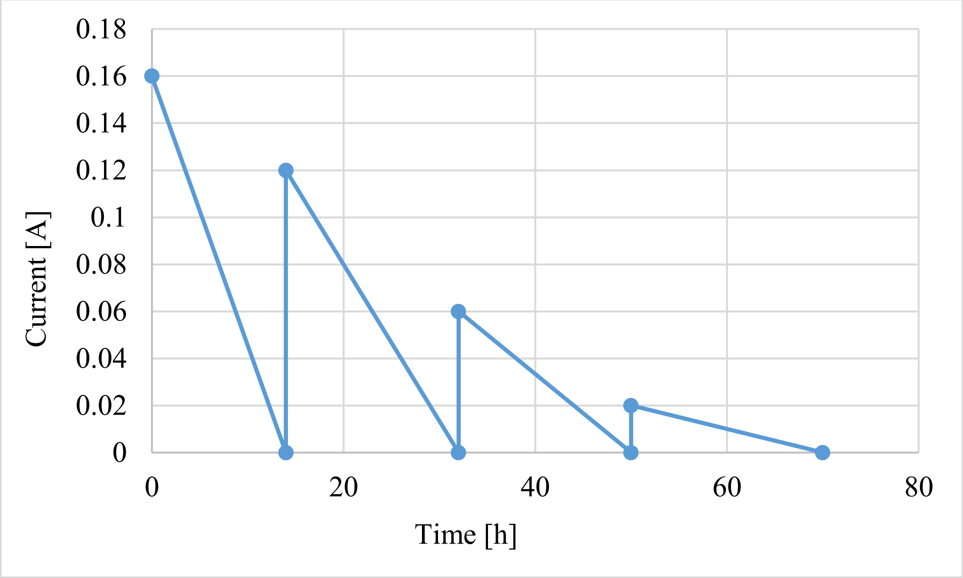

Figure 4-24 Current changes versus time for Test 4a

Although the initial current (0.16 A) was lower than that of Test 3b, the current was maintained in the system for a significant part of the exposure time. Averagely, a mixing was required after every 16 hours; meaning the current was maintained in the system for another 2 hours before each mixing, comparing to the relevant value in Test 3b. After the first mixing, 75% of the initial current value was recovered, while only 62.8% of the current value was recovered after the first mixing in Test 3b. A much faster decrease in the current value was observed during the first hours of the experiment; therefore, more power was probably required for demulsification in that period. Most of the oily sludge demulsification took place in the first 24 to 32 hours of the experiment. Separate phases were visible in the reactor after this period (Figure 4-24).

4.3.6 Ferric chloride as the additive (Test 4b)

The results are exhibited in Table 4-23 and 4-24. The test was conducted on sample C7 in the small EK reactor and under the exposure of a 1 V/cm voltage gradient.

Table 4-23 Conditions and results for Test 4b

Time [h]	**Current [A]**	**Current after mixing [A]**	**Voltage [V]**	**Heavy oil [mL]**	**Light oil [mL]**	**Water [mL]**	**Catholyte* [mL]**	**Anolyte** [mL]**
0.0	0.15	N/A	3.9	0.00	0.00	0.0	0.00	0.00
12.0	< 0.003	0.07	3.9	1.75	7.00	3.0	10.00	1.75
23.0	< 0.003	0.05	3.9	4.00	7.50	6.0	13.50	4.00
36.5	< 0.003	0.04	3.9	4.00	8.00	6.5	14.50	4.00
48.0	< 0.003	0.02	3.9	4.25	8.00	6.5	14.50	4.25
64.0	< 0.003	N/A	3.9	4.50	8.25	6.5	14.75	4.50

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

Table 4-24 Recovery percentages of oil and water for Test 4b

Time [h]	**Light oil [%]**	**Water [%]**	**Heavy oil [%]**
0.0	0.00	0.00	0.00
12.0	43.02	3.61	5.17
23.0	46.10	7.21	11.81
36.5	49.17	7.82	11.81
48.0	49.17	7.82	12.55
64.0	50.71	7.82	13.29

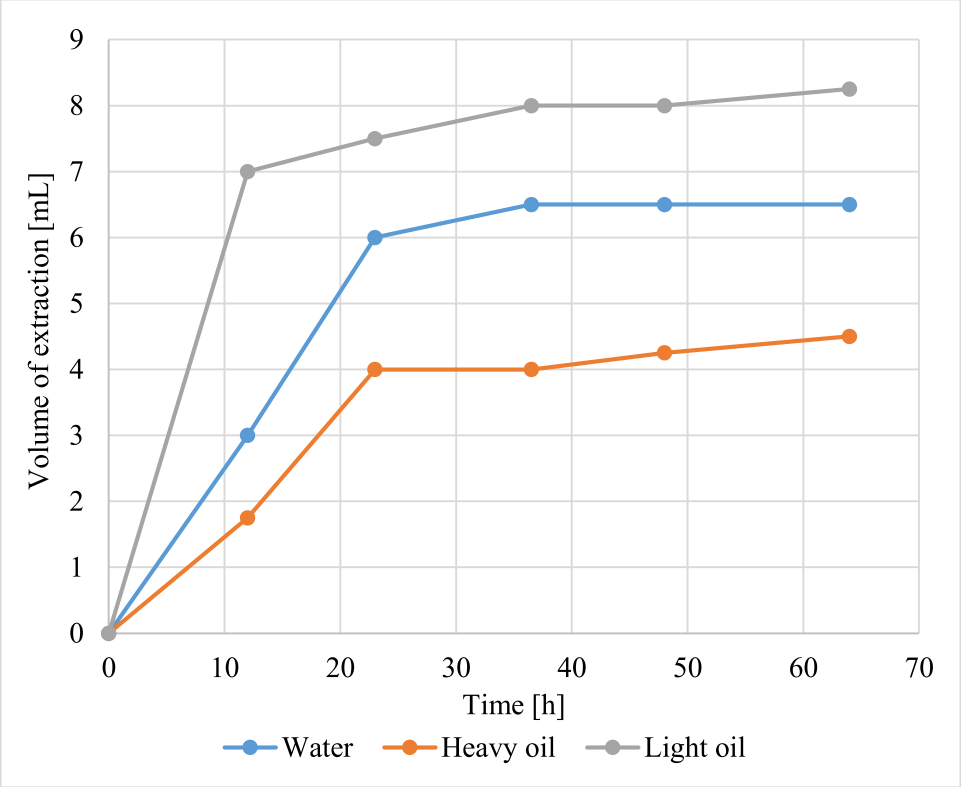

Figure 4-25 Volume of extracted light oil, heavy oil, and water versus time for Test 4b

According to the results (Figure 4-25), most of the extraction took place within the first 24 hours of the experiment. Water was only extracted from the cathode side. Moreover, there was much more extraction from the cathode than the anode side. Comparing with Test 4a, Test 4b needed a longer exposure time (36.5 h) to achieve the plateau state with respect to water extraction. An insignificant amount of water was extracted afterward. Due to the similarity of experimental conditions, the only parameter which was responsible for a difference between the results of Test 4a and Test 4b was the type of the matrix. It was concluded that the wettability alteration of the system to water-wet was mainly influenced by the type of oily sludge. Concerning the extracted volume of catholyte water and light oil, a similar trend was observed for Test 4b in which both values were significant. The volume of extracted heavy oil in Test 4b was 1.25 mL higher than that of Test 4a. It proved that the two-side transport developed better in the matrix of Test 4b (sample C7).

The recovery percentages of light oil, heavy oil, and water after 64 hours of exposure to the EK treatment were 50.71%, 13.29%, and 7.82%, respectively (Table 4-24). In fact, most of the light oil extraction was achieved during the first few hours.

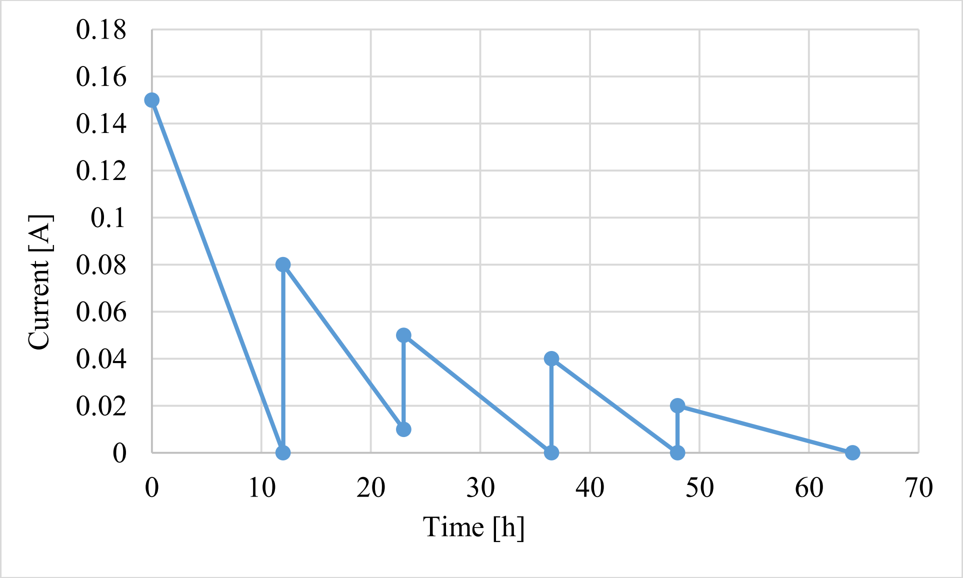

Figure 4-26 Current changes versus time for Test 4b

The initial current value of 0.15 A was detected for Test 4b (Figure 4-26) which was slightly lower than the initial current value of Test 4a (0.16 A). After each mixing, the current value remarkably increased, although it was still lower than the initial value after each mixing. The current value after the final mixing was 0.02 A.

4.3.7 Ferric chloride as the additive (Test 4c)

The results are presented in Table 4-25 and 4-26. A voltage gradient of 2.05 V/cm (a constant voltage of 8 V) was applied to a small-scale reactor filled with sample C $_{mix}$ (C1-C8).

Table 4-25 Conditions and results for Test 4c

Time [h]	Current [A]	Current after mixing [A]	Light oil [ml]	Catholyte water [mL]	Heavy oil [mL]	Anolyte water [mL]	Catholyte* [mL]	Anolyte** [mL]
0.0	0.36	N/A	0.00	0.00	0.0	0.0	0.00	0.0
15.0	< 0.003	0.15	2.00	4.50	0.5	3.5	6.50	4.0
19.5	0.04	0.12	4.50	4.75	0.5	3.5	9.25	4.0
35.5	< 0.003	0.06	5.00	5.50	1.0	3.5	10.50	4.5
40.0	0.01	0.04	5.25	5.50	1.0	3.5	10.75	4.5
56.0	< 0.003	N/A	5.50	5.50	1.0	3.5	11.00	4.5

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

Table 4-26 Recovery percentages of oil and water for Test 4c

Time [h]	Light oil [%]	Water [%]	Heavy oil [%]
0.0	0.00	0.00	0.00
15.0	18.85	10.29	0.75
19.5	42.41	10.62	0.75
35.5	47.13	11.58	1.50
40.0	49.48	11.58	1.50
56.0	51.84	11.58	1.50

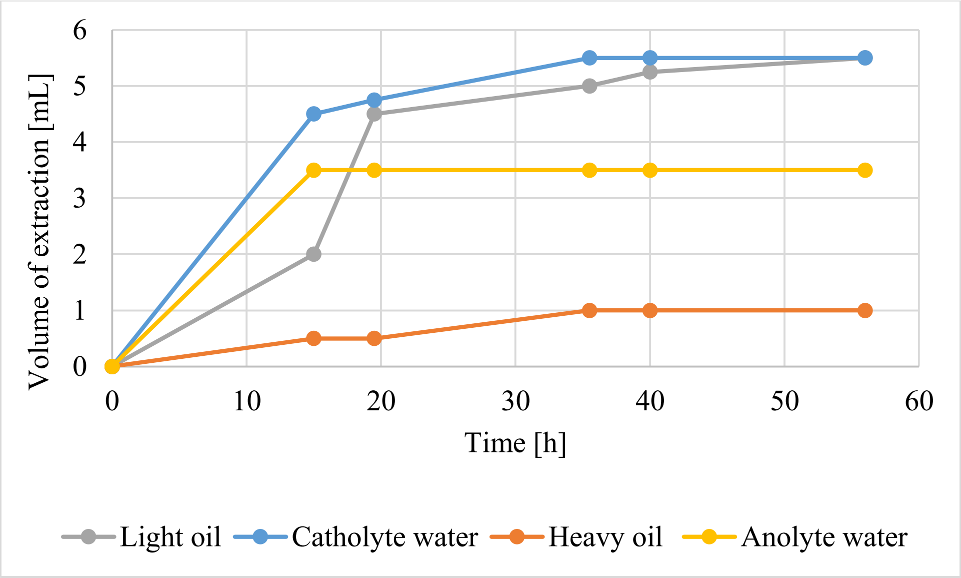

Figure 4-27 Volume of extracted light oil, heavy oil, cathode water and anode water versus time for Test 4c

The results (Figure 4-27 and Table 4-26) stated that catholyte extraction had a much faster pace in the first 20 hours of the experiment. Anolyte water extraction also took place but stopped after 15 hours. Catholyte extraction was twice more than anolyte extraction in Test 4c. Comparing to section 4.2, Test S3 (2.05 V/cm, the mixture of C1 to C8, no additive), the total extracted light oil significantly increased to a volume of 5.5 mL. In addition, extraction of catholyte water remarkably improved once ferric chloride was introduced to the matrix. It showed that the electroosmotic flow developed effectively in the system. In contrast, the anolyte volume reduced to 4.5 mL comparing to section 4.2, Test S3 (6.75 mL). However, the overall catholyte extraction increased by a volume of 7 mL which proved the effectiveness of using additives on catholyte extraction. The recovery percentages of the light oil, heavy oil, and water after 56 hours of exposure to the treatment were 51.84%, 1.5%, and 11.58%, respectively. The qualitative assessment of catholyte extraction revealed that the clarity of catholyte water significantly improved comparing to Test S3. It confirmed the effectiveness of adding ferric chloride on obtaining an enhanced demulsification of oil sludge (Figure 4-28).

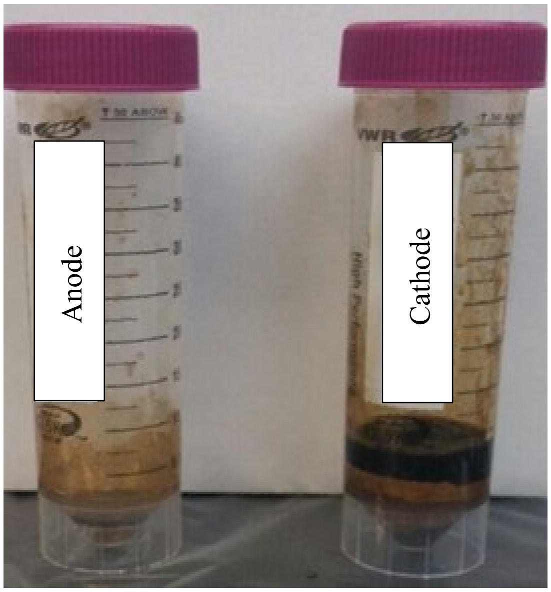

Figure 4-28 Qualitative assessment of anolyte and catholyte extraction for Test 4c (FeCl3 as the additive, mixture of C1 to C8, 2.05 V/cm)

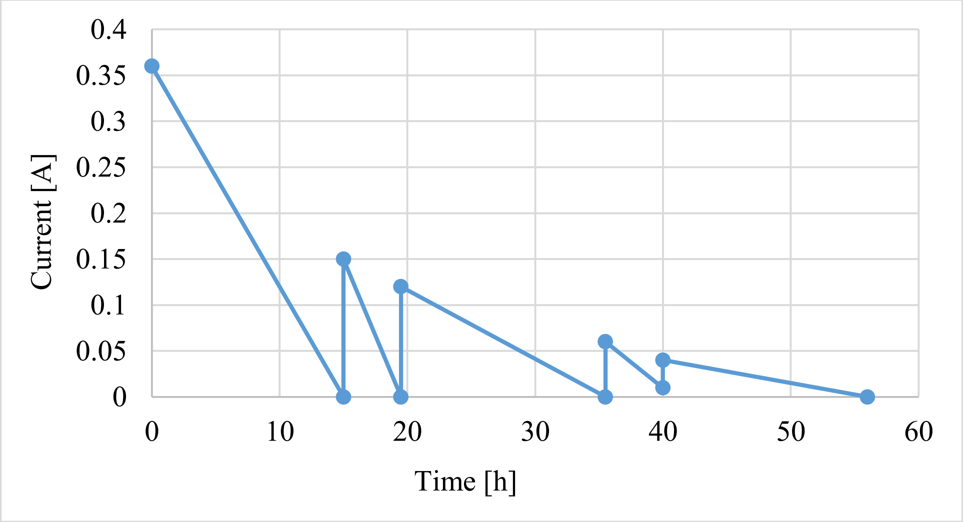

Figure 4-29 Current changes versus time for Test 4c

Introducing ferric chloride as the additive significantly increased the initial current value in comparison with the initial value of Test S3 (0.28 A). It showed that the conductivity of the system would meaningfully increase once ferric chloride is used. The trend of variations was similar to that of Test S3; however, the recovered current value after the first and second mixing was significantly higher (Figure 4-29).

General conclusion of phase 3

After comparing the impacts of all additives applied to the EK system, it was concluded that the biggest volume was extracted in the presence of ferric chloride, and the order of influence was as follows:

ferric chloride > clay > alum > clay + cationic polymer

It seemed that ferric chloride was the best additive for improving the electrokinetic extraction of light oil, heavy oil, and water. Furthermore, the quality of the extracted anolyte and catholyte water was also much higher than in the case of other additives. As a result, ferric chloride was the recommended additive for the scale-up process.

4.4 Phase 4: Scaling up the EK reactor to a medium size

4.4.1 Conducting the medium-scale (4 L) experiment on a mixture of raw oily sludge (Test M1)

A significant volume of heavy oil was collected at the anodes and light oil at the cathodes (Figure 4-30). Unfortunately, the design of the reactor did not permit the evacuation of liquids that were separated inside the EK reactor. Subsequently, such design was improved and applied to the large scale reactor (20 L). The collected fractions (water and light and heavy oils) are shown in Figure 4-31. Calculations for removal percentage were done based on the initial weights of 381.8 g, 1463.7 g, and 977.7 g for light oil, water, and heavy oil, respectively. The percentage of water removal was 43%, and due to the symmetrical design of the reactor, the total water recovery could be doubled. The same assumption could be made about the recovery of light and heavy oils.

A continuous water removal was observed over the test period. However, after around 24 hours, lower oil recovery was noticed (Figure 4-31). Remaining in the reactor, solids were transported toward the anode due to the electrokinetic phenomena and compacted (Figure 3-7). It was estimated that the remaining volume for disposal has been decreased by around 50%. It could be noticed, that the remaining solids might present a value-add product.

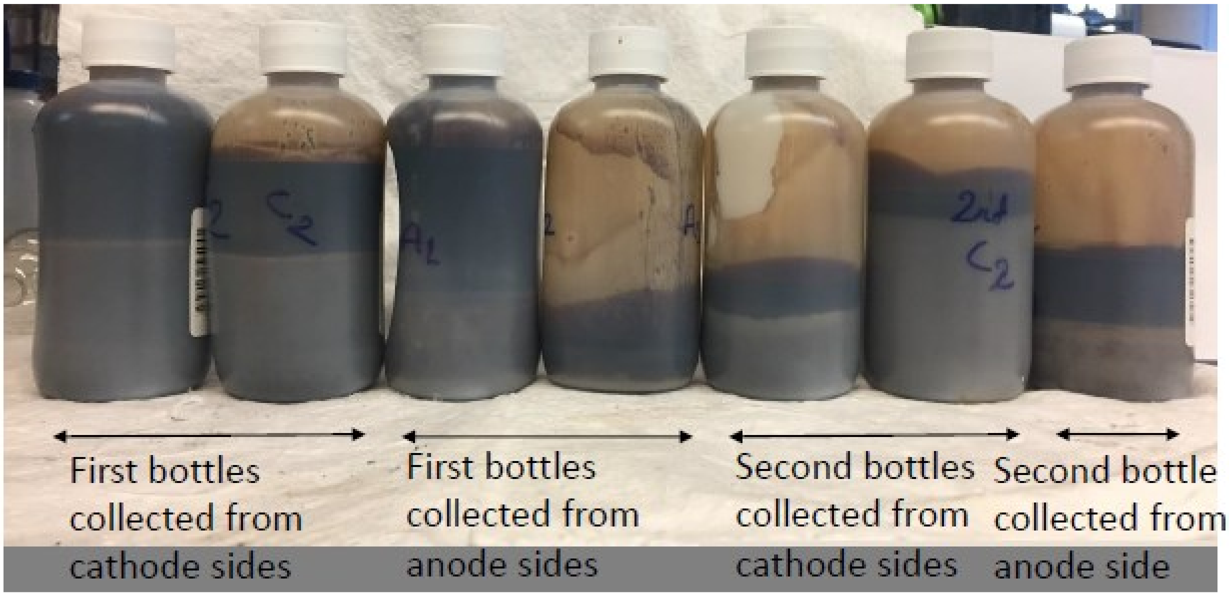

Figure 4-30 Containers with EK-separated oil sludge fractions at the anode and cathode for Test M1

The collected fractions underwent physicochemical analyses. The viscosity value of liquid collected at the anode area was 104 mPa.s @25ºC, which was higher than the viscosity of liquid from the cathode (77 mPa.s @25ºC), however, much lower than the viscosity of oil sludge that was 1400 mPa.s. This showed the changes in rheological properties of the tested matrix, which led to easier transportation of the separated phases and emphasized the high quality of the extracted oil.

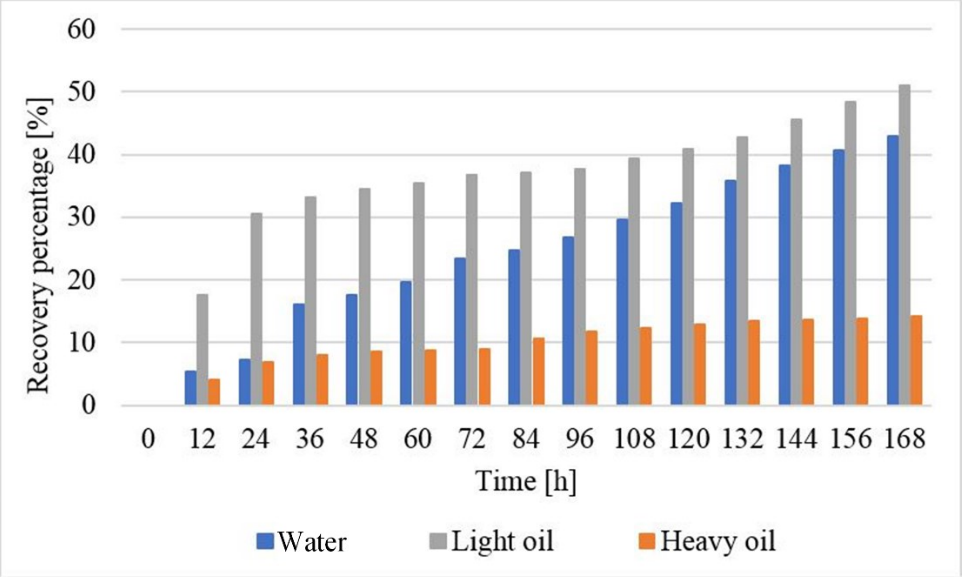

Figure 4-31 Recovery percentages of water and light and heavy oils after the EK treatment in the 4 L reactor (Test M1)

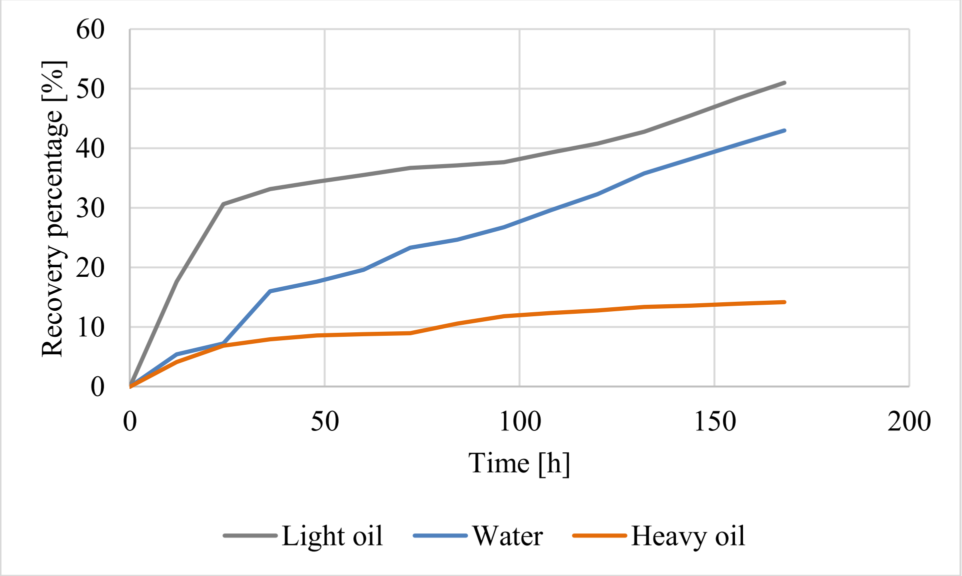

Figure 4-32 Trends of light and heavy oil recovery as well as water production in the 4 L EK reactor (Test M1)

Since the viscosity of oil was higher than water, the relative mobility of water and oil was essential to assess. The changes in water and oil removal were related to the mobility ratio of W:O over 160 hours (Figure 4-32). This controlled the recovery of oil fractions, and it was an important factor for designing the EK reactor. According to Figure 4-32, the heavy oil production tendency was strongly upwards during the first and second 12 hours. The heavy oil extraction kept a gradual increase up to 144 hours (the sixth day) while the volume of the extracted fraction was slightly stabilized during the ninth and tenth 12-hours period. Therefore, it would be a fair justification to imply that due to the high dynamic viscosity of heavy oil, the volumetric rate of heavy oil extraction would be lower than light oil with lower dynamic viscosity.

The final recovered fractions were 42.98%, 50.99%, and 14.16% for water, light oil, and heavy oil respectively. Based on Figure 4-31, the extracted fraction of light oil was notably higher than water, which demonstrated the low water to oil mobility ratio.

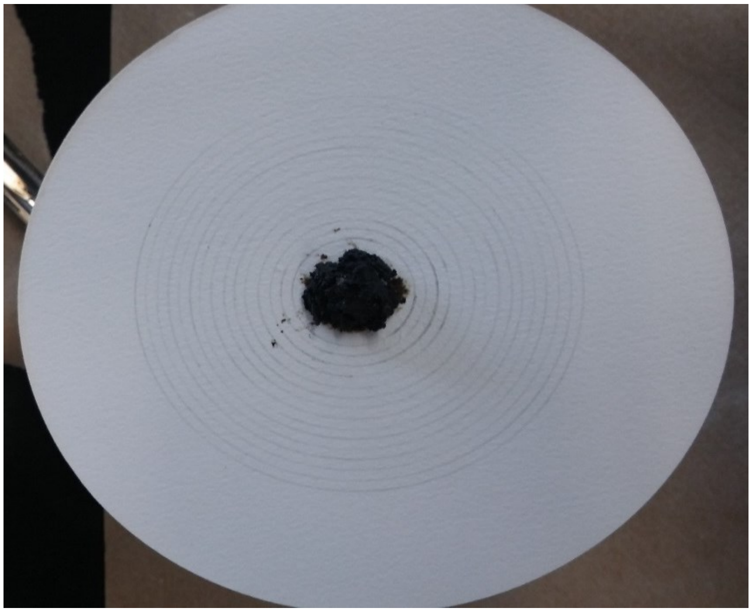

Figure 4-33 Solids after the EK treatment; initial part of the experiment for detection of the moisture content

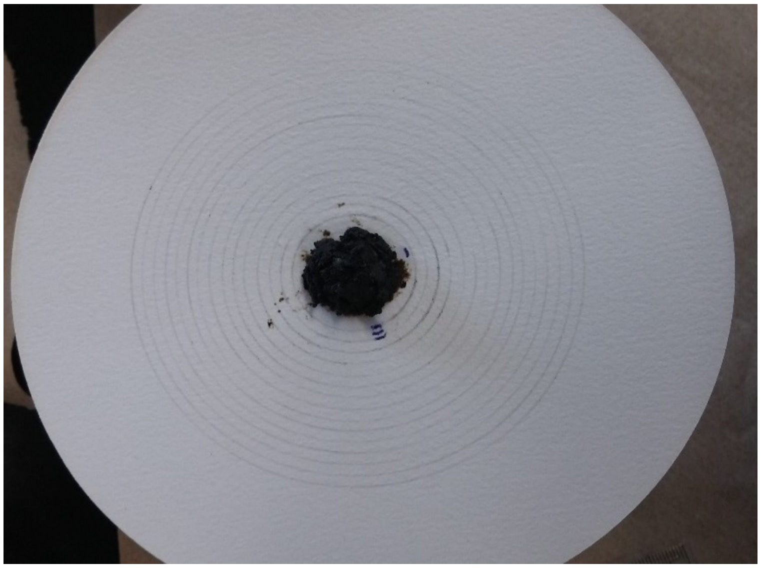

Figure 4-34 Solids after the EK treatment; time elapsed: 22 min of the experiment for detection of the moisture content

Additionally, the characterization of remaining sludge in the reactor after the liquid extraction was conducted. The objective was to assess the remaining water content in wasted solids for its final disposal. An amount of 1.954 g of solids was located in the center of a filter paper, then, the moisture propagation was monitored (Figure 4-33 and 4-34). The duration of this test was 22 minutes, although practical propagation of the moisture was not observed after a few minutes. The rate of propagation was defined in cm/(min.g) of sludge (Table 4-27).

Table 4-27 Verification of the moisture content in oil sludge after the EK treatment for Test M1

Elapsed time [min]	**The radius of moisture in the filter [cm]**	**Rate of moisture propagation [cm/(min.g)]**
0	1.25 cm: radius of the sample	N /A
5	1.50	0.0250
14	1.65	0.0072
22	1.75	0.0061

For comparison, the moisture propagation in the filter paper before the EK treatment of oil sludge is shown in Table 4-28. The procedure of this test was the same as the one that was conducted on sludge after the EK treatment; an amount of 1.817 g of non-treated sludge was placed in the center of the paper. The results demonstrated that twice as much time was required to reach a similarly low rate of the moisture propagation that was shown in Table 4-27 (sludge after the EK treatment). It seemed that the remaining sludge after the EK treatment had an insignificant amount of moisture as well as a much-decreased volume; in fact, no oil was leaking. Furthermore, sludge was solidified; thus, it was easy to be manipulated and transported, and it did not require a special pumping system.

Table 4-28 Verification of the moisture content in oil sludge before the EK treatment for Test M1

Elapsed time [min]	**The radius of moisture in the filter [cm]**	**Rate of moisture propagation [cm/(min.g)]**
0	1.4 cm: radius of the sample	N/A
1	1.65	0.1370
5	2.15	0.0870
11	2.60	0.0420
41	2.90	0.0055

The results of BS&W analysis (ASTM D4007-11, 2016) on samples extracted from the 4 L EK reactor are shown in Table 4-29. According to the regulations, the percentage of water in the extracted oil should be less than 3 %. The EK treatment proved to be efficient by achieving a water percentage of less than 3%, and the sediments were either not detected or less than 3%.

Table 4-29 Results of BS&W analysis on samples collected from the cathode and anode outlets after the treatment in the 4 L EK reactor

Sample	Oil [%]	Water [%]	Sediments [%]
Anode 1	> 95.0	< 2.0	< 3
Cathode 1	> 99.4	< 0.6	ND

The results regarding the calorific value of oily sludge before and after the medium-scale EK treatment are stated in Table 4-30. They showed that the electrokinetic treatment generated end-products with calorific values which were 4 times higher than that of the initial sludge sample. Since calorific values between 42,499 and 44,020 kJ/kg were similar to the calorific value of crude oil (45,668 kJ/kg), the end-products presented a high selling value in the petroleum market. It is noteworthy to mention that due to the simultaneous electrokinetic removal of various impurities (e.g. heavy metals) and the minimal content of sediments and water, the end-products presented much higher quality than crude oil.

Table 4-30 Calorific values of samples collected from the cathode and anode outlets after the treatment in the 4 L EK reactor

Sample	Calorific value [kJ/kg]
Raw oily sludge (before treatment)	10835
Anode 1	42499
Cathode 1	44020

Table 4-31 Cumulative energy consumption and cost of energy for Test M1

Time [h]	**Current [A]**	**Cumulative energy consumption [kWh]**	**Cost [CAD]**	**Cost [USD]**
0	4.15	0.00	0.000	0.000
12	0.25	1.54	0.054	0.139
24	0.07	1.66	0.058	0.149
36	0.08	1.71	0.060	0.154
48	0.07	1.76	0.062	0.159
60	0.06	1.81	0.063	0.163
72	0.04	1.84	0.064	0.166
84	0.03	1.87	0.065	0.168
96	0.02	1.88	0.066	0.170
96	0.75	1.88	0.066	0.170
108	0.19	2.21	0.078	0.199
120	0.13	2.33	0.081	0.209
132	0.09	2.40	0.084	0.216
144	0.05	2.45	0.086	0.221
156	0.03	2.48	0.087	0.223
168	0.005	2.49	0.087	0.224

Table 4-31 shows the cumulative energy consumption and the cost of energy for Test M1. According to the results, more than 50% of the total energy was consumed during the first 12 hours of the experiment while the current fluctuated between 4.15 and 0.25 A. During this period the highest value of the current was observed. After 12 hours, energy consumption increased at a slower pace due to the decrease in the system’s conductivity. The cost analysis was done for the industrial Quebec market with a cost of 0.035 CAD/kWh of energy as well as for the USA market with a cost of 0.09 USD/kWh.

General conclusion of section 4.4.1

The results of Test M1 proved that the compartmental distance and the configuration of electrodes are important parameters for the upscaling process.

The EK treatment of oil sludge at the medium-scale (4 L reactor) confirmed outcomes of the small-scale investigations.

EK successfully demulsified oil sludge, recovered water and light and heavy oils. The volume of the remaining solids after the EK treatment decreased by almost 50%. The remaining solids practically did not leak, so it was ready for manipulating and transportation. Such a decrease in volume might also decrease the cost of sludge disposal.

Tests on a medium scale attained the objective of high water removal, more than it was accomplished in a small-scale EK system. Therefore, one of the main objectives of this study was to achieve the highest possible water removal in a scaled-up 20 L EK reactor.

4.4.2 Conducting the medium-scale (4 L) experiment on the mixture of raw oily sludge samples with the implementation of ferric chloride as the additive (Test M2)

Table 4-32 and 4-33 present the results. Test M2 was conducted in a 4 L EK reactor and on sample C $_{mix}$. The voltage gradient of 2.05 V/cm (a constant voltage of 19.6 V) was applied.

The sludge contained a significant amount of solids, which settled during the vertical separation of phases. They were able to reach the collecting bottles through the outlets. Therefore, an additional screening was considered in the large-scale (20 L) design.

Table 4-32 Results for Test M2

Time [h]	**A2 water [mL]**	**A2 oil [mL]**	**Ct1 solids [mL]**	**Ct1 oil [mL]**	**Ct2 solids [mL]**	**Ct2 water [mL]**	**Ct2 oil [mL]**	**DA oil [mL]**	**DCt solids [mL]**	**DCt water [mL]**	**DCt oil [mL]**
0.0	0	0	0	0	0	0	0	N/A	N/A	N/A	N/A
14.5	165	75	0	0	95	35	20	N/A	N/A	N/A	N/A
19.5	165	75	0	0	100	40	20	N/A	N/A	N/A	N/A
24.0	170	80	0	0	100	40	20	N/A	N/A	N/A	N/A
41.0	185	105	50	30	160	60	80	N/A	N/A	N/A	N/A
48.0	190	110	50	30	160	60	80	N/A	N/A	N/A	N/A
63.0	190	120	55	30	195	105	90	0	0	0	0
66.5	190	120	55	30	195	105	90	0	50	40	0
73.0	190	120	55	35	200	110	90	0	90	70	10
86.0	190	130	60	40	200	115	100	20	90	90	30
95.0	190	130	60	40	200	115	100	40	90	100	30

Note: A1 extraction was less than 5 mL.

Table 4-33 Recovery percentages of oil and water for Test M2

Time [h]	Light oil [%]	Water [%]	Heavy oil [%]
0.0	0.00	0.00	0.00
14.5	3.26	15.84	5.75
19.5	3.26	16.23	5.75
24.0	3.26	16.63	6.13
41.0	17.94	19.40	8.05
48.0	17.94	19.79	8.43
63.0	19.57	23.36	9.20
66.5	19.57	26.52	9.20
73.0	22.02	29.29	9.20
86.0	27.73	31.27	11.50
95.0	27.73	32.07	13.03

After 24 hours, the successful separation of phases was observed in Test M2 (Figure 4-35). Solids were accumulated and compacted in the anode side while water was transferring to the cathode side.

Figure 4-35 Top view of the 4 L EK reactor in the middle of the experiment

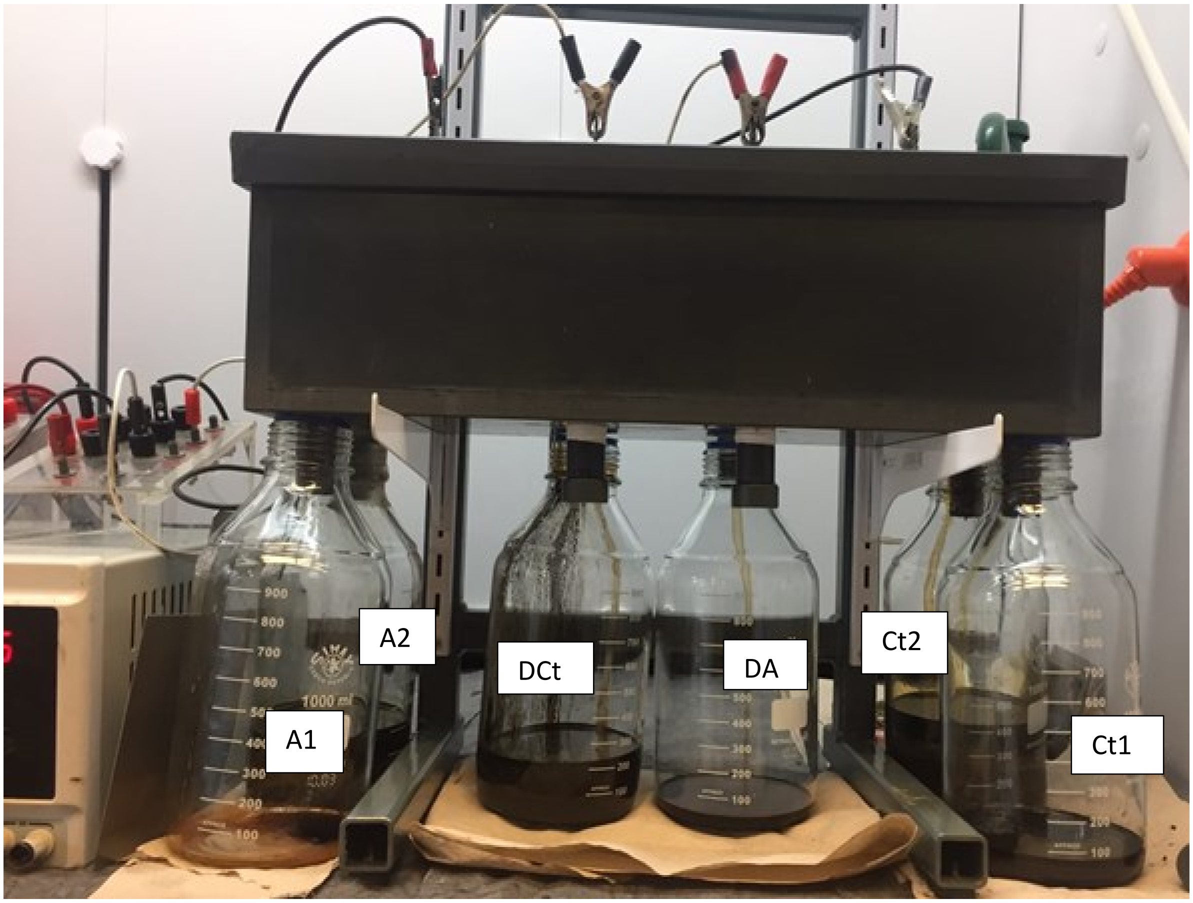

Figure 4-36 Side view of the 4 L EK reactor with extracted oil, water, and solids at the end of the experiment

Extracted liquids were accumulated in 1 L bottles installed at the bottom extensions and located close to electrodes and side banks (Figure 4-36). A1 and A2 referred to bottles located under two outlets on the side bank close to Anode 1. Labels Ct1 and Ct2 referred to the bottles that were collecting liquids from two outlets installed on the side bank close to Cathode 2. To collect the extraction from the middle compartments, bottom outlets were installed close to Cathode 1 and Anode 2, where called bottles were labeled DCt and DA, respectively.

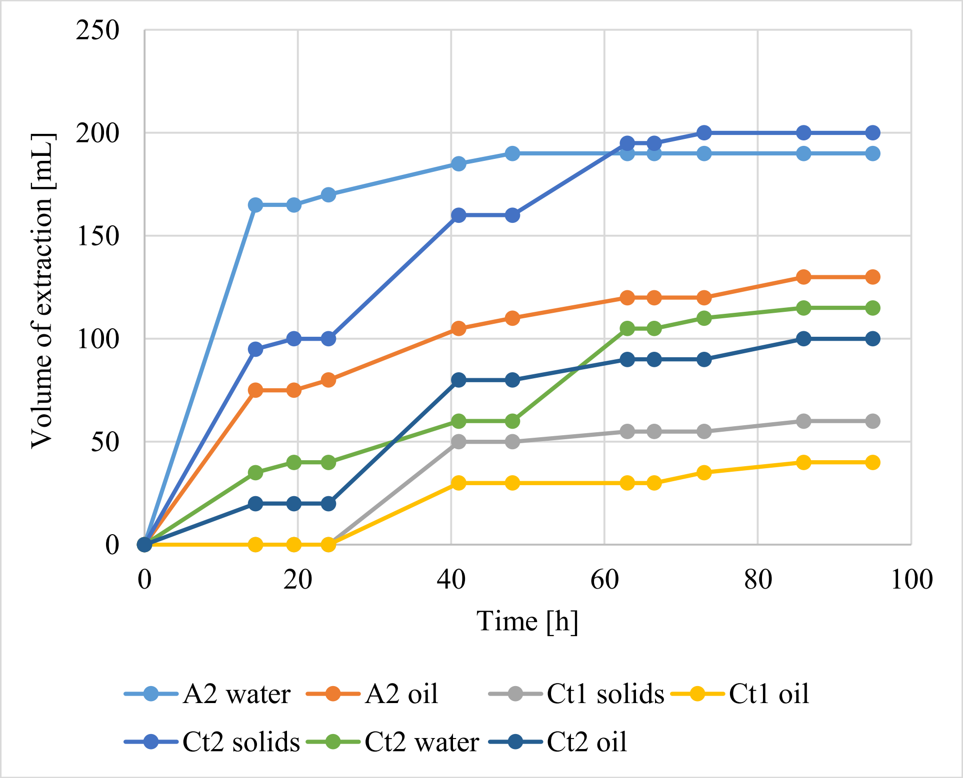

Figure 4-37 Volume of extracted oil, water, and solids in the side bottles versus time for Test M2

An amount of 140 mL of oil was extracted from the side cathode (Ct1 and Ct2) by the end of the exposure time while 115 mL of water was extracted from outlet Ct2, and no water extraction from outlet Ct1 was observed (Figure 4-37). Therefore, the overall extraction of water from outlets Ct1 and Ct2 was 115 mL. On the other hand, an amount of 190 mL of water was extracted from the side anode. In addition, 130 mL of oil was extracted from outlet A2. No extraction from A1 (one of the outlets placed under the side anode) was detected, showing that a certain non-uniformity of phase separation might be expected at a bigger scale. Overall, the liquid extraction from the side bank close to the anode was 320 mL while the total volume of the liquids extracted from the side cathode was 255 mL.

The results (Figure 4-37) stated that most of the extraction took place in the first 40 hours of the experiment; however, the removal of water and solids from the cathode area continued at a significant rate for the next 23 hours. Most of the separated water and oil and all solids were extracted from the outlets close to the cathodes.

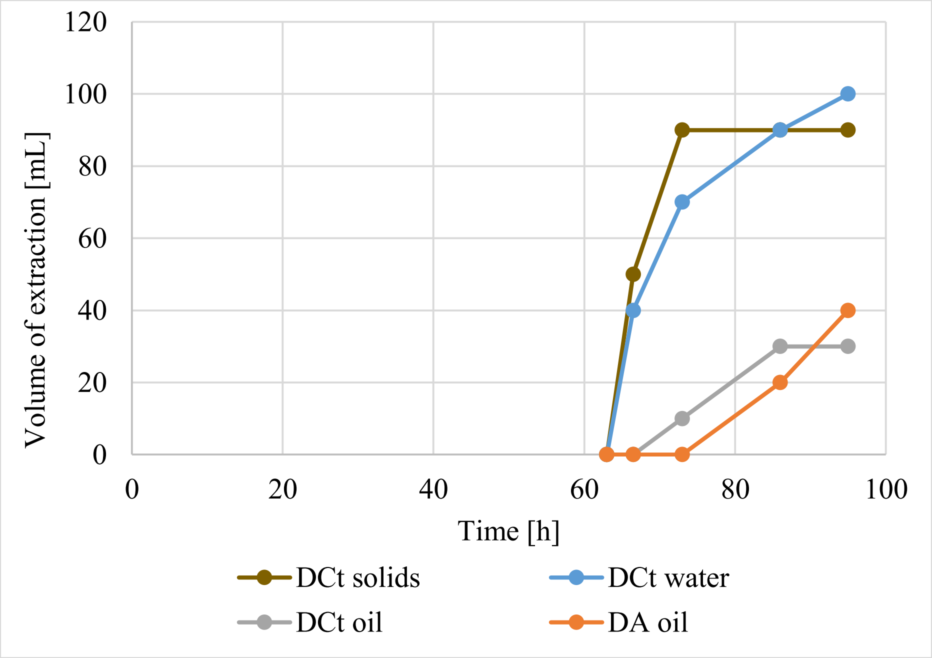

Figure 4-38 Volume of extracted oil, water, and solids from the middle compartments versus time for Test M2

The middle outlets were not open during the first 63 hours of the experiment. The elapsed time was given for longer exposure time in order to achieve better separation of the phases in the middle part of the EK reactor. After opening two outlets in the middle, an insignificant anolyte extraction (DA) was noticed. Furthermore, most of the catholyte extraction (DCt) was water and solids (Figure 4-38).

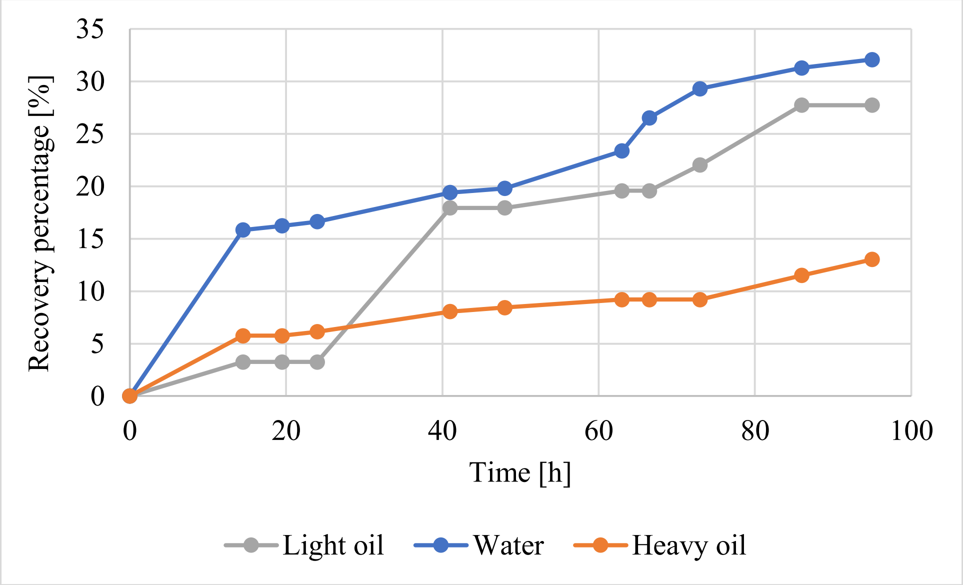

Figure 4-39 Recovery percentages of oil and water versus time for Test M2

Recovery percentages of the light oil, heavy oil, and water after 95 hours of exposure to the treatment were 27.73%, 13.03%, and 32.07%, respectively (Figure 4-39). The recovery percentage increased continuously.

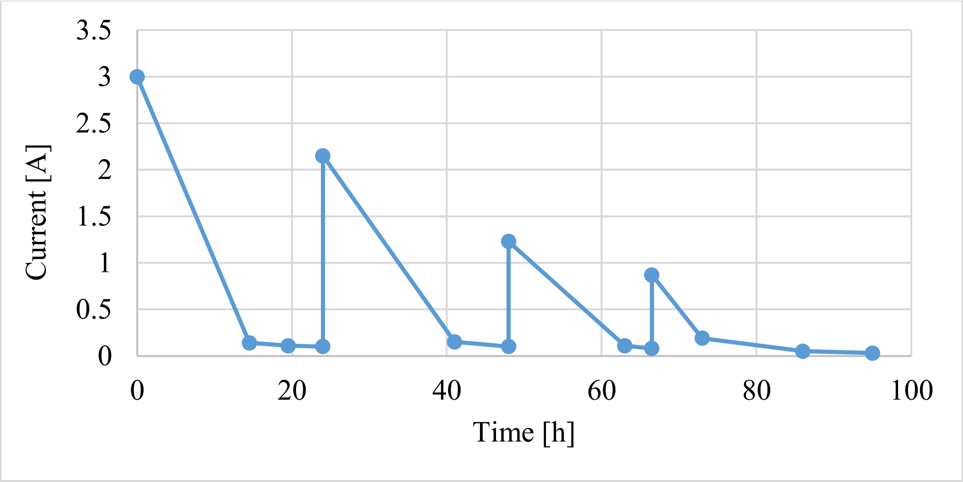

Figure 4-40 Current changes versus time for Test M2

The initial value of the current was 3 A which showed the effect of adding ferric chloride on increasing the conductivity (Figure 4-40). After 24 hours, the current value reduced to 0.1 A, and the first mixing took place. The current value after the first mixing was 2.15 A (about 72% of the initial current value). The last mixing at 66.5 h recovered 29% of the initial current value.

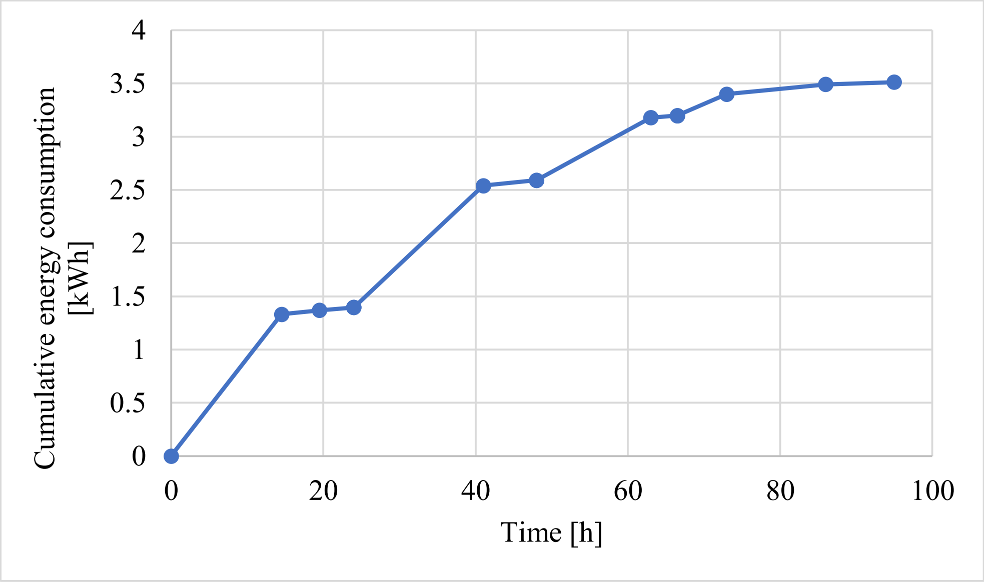

Figure 4-41 Current changes versus time for Test M2

According to the results (Figure 4-41), most of the energy consumption took place in the first 63 hours of the experiment. An amount of 1.4 kWh of energy was consumed meanwhile the current was 3 A. During the time interval between 34 h and 63 h, current fluctuated between 0.1 A and 2.15 A and resulted in 1.78 kWh of energy consumption. During the last 22 hours, 0.11 kWh of energy was consumed. Overall, additives would increase energy consumption more than longer exposure time would do.

The results of BS&W analysis (ASTM D4007-11, 2016) on the extracted catholyte and anolyte are presented in Table 4-34. According to the results, the EK technology proved once again to be an efficient method for treating oily sludge, since there was no detected water or sediments in the samples after the BS&W test.

Table 4-34 Results of BS&W analysis on samples collected from cathode and anode outlets after the treatment in the 4 L EK reactor

Sample	Oil [%]	Water [%]	Sediments [%]
Anode 1 (A2)	100	0	~0
Cathode 2 (Ct2)	100	0	~0

The results with respect to the calorific values of the samples before and after the medium-scale EK treatment are stated in Table 4-35.

Table 4-35 Calorific values of samples collected from cathode and anode outlets after treatment in the 4 L EK reactor

Sample	**Calorific value [kJ/kg]**
Raw oily sludge (before treatment)	10835
Anode 1 (A2)	42857
Cathode 2 (Ct2)	43716

General conclusion of section 4.4.2

The results from the medium-scale EK test confirmed the adequate choice of optimal conditions made during the small-scale EK tests. The best recoveries of water, light oil, and heavy oil (32%, 28%, and 13%, respectively) were achieved while applying the voltage gradient of 2.05 V/cm to the system with stainless steel electrodes. An improvement in the recovery of water and oil was observed when ferric chloride was added as a conditioner to the oily sludge with a volumetric ratio of 1 to 300. The EK treatment proved to be effective in water recovery since the remaining solids in the reactor after the EK treatment presented a very low amount of water distributed on the filter. The quality of the recovered oil was assessed by BS&W analysis and the calorific value. Less than 1% of water and sediments were detected in the oil, and the calorific value was above 41,000 kJ/kg.

4.5 Phase 5: Scaling up the EK reactor to a large size (Test L)

Different voltage gradients were applied to the EK reactor in different periods to optimize the extraction conditions. These voltage gradients and total anolyte and catholyte extractions after different periods are shown in Table 4-36 and Figure 4-42.

Table 4-36 Conditions and results for Test L

Time [h]	**Voltage gradient [V/cm]**	**Current [A]**	**Current after extraction [A]**	**Catholyte* [mL]**	**Anolyte** [mL]**
0.00	1.00	6.6	N/A	N/A	N/A
20.00	0.79	1.7	2.1	N/A	45.0
43.28	1.30	0.8	2.0	1703.00	694.5
55.95	1.30	0.6	0.8	1959.00	694.5
84.07	1.30	0.4	0.6	2171.50	787.5
103.95	1.30	0.4	1.7	2309.00	787.5
107.57	2.05	1.7	2.6	2490.00	787.5
127.68	2.05	0.3	0.5	2623.00	787.5
150.12	2.05	0.4	N/A	2715.75	799.5

* Catholyte: total volume of extraction from the cathode side; ** Anolyte: total volume of extraction from the anode side

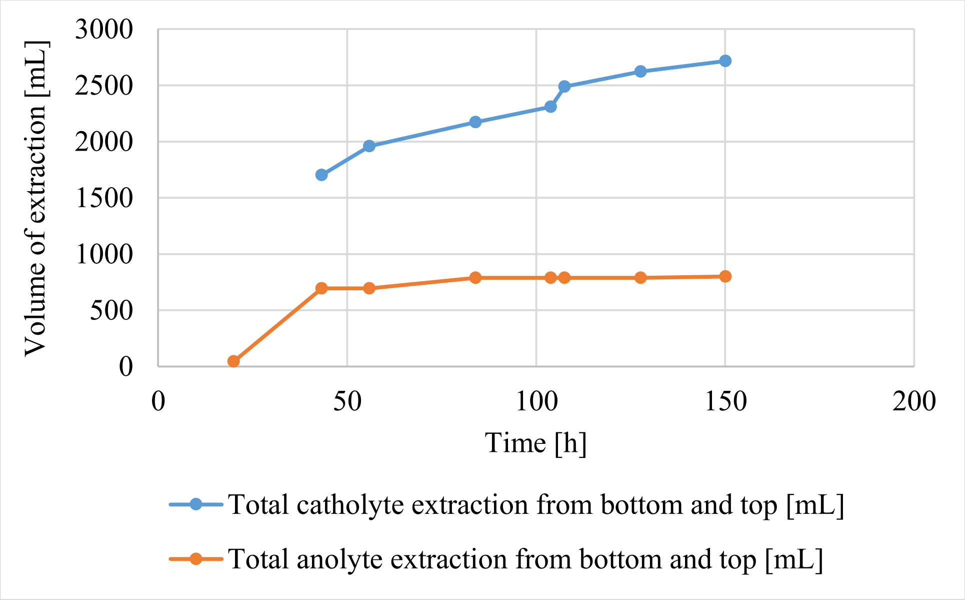

Figure 4-42 Total catholyte and total anolyte extraction versus time for Test L

Based on the previous tests at lower scales, it was assumed that 24 hours should be provided for the phase separation. To confirm this assumption, the first extraction at the anode A1 took place at 20 hours. It was found that at the time electro-demulsification of sludge had already taken place in the large EK reactor. Therefore, at larger scales, the retention time might decrease without a negative impact on the performance of the EK reactor. The collection of all anolyte and catholyte extractions was conducted after 43 hours (Figure 4-42). During the entire test, an excellent separation of oil, solids, and water was observed (Figure 4-43). Subsequently, the collection of liquids was conducted from the bottom and the top of the EK reactor (Table 4-37 and 4-38, Figure 4-44 and 4-45). No mixing process took place since an assessment of the behavior and quality of the remaining solids was required. The mixing process would have increased the amount of the extracted liquids, as it was demonstrated in the tests at the lower scales.

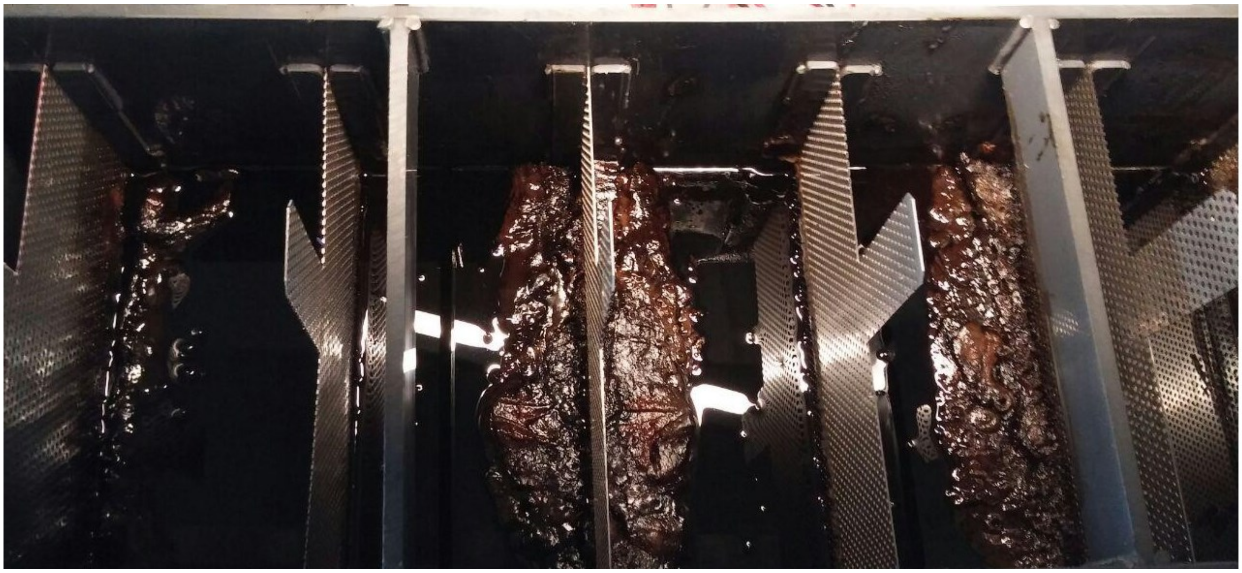

Figure 4-43 Significant separation of the phases inside the reactor after 43.28 hours (Test L)

Table 4-37 Conditions and volume of catholyte and anolyte, extracted from the top of the reactor, for Test L

Electrical parameters		**Catholyte top extraction [mL]**					**Anolyte top extraction [mL]**
Time [h]	**Voltage gradient [V/cm]**	**C1-L**	**C2-R**	**C2-L**	**C3-R**	**C3-L**	**A1-L**
0.00	1.00	N/A	N/A	N/A	N/A	N/A	N/A
20.00	0.79	N/A	N/A	N/A	N/A	N/A	N/A
43.28	1.30	515.0	ND	200.0	ND	414.00	N/A
55.95	1.30	85.0	18.0	ND	15.00	138.00	N/A
84.07	1.30	75.0	ND	15.0	12.5	110.00	93
103.95	1.30	57.0	ND	1.5	15.00	64.00	ND
107.57	2.05	34.5	22.5	8.0	42.25	73.75	ND
127.68	2.05	63.5	20.5	3.0	24.00	22.00	ND
150.12	2.05	44.0	28.5	ND	16.00	4.25	12

Table 4-38 Conditions and volume of catholyte and anolyte, extracted from the bottom of the reactor, for Test L

Electrical parameters		**Catholyte bottom extraction [mL]**				**Anolyte bottom extraction [mL]**	
Time [h]	**Voltage gradient [V/cm]**	**C11**	**C21**	**C22**	**C31**	**A11**	**A12**
0.00	1.00	N/A	N/A	N/A	N/A	N/A	N/A
20.00	0.79	N/A	N/A	N/A	N/A	45.0	N/A
43.28	1.30	50	9	5	510	584.5	65
55.95	1.30	N/A	N/A	N/A	N/A	N/A	N/A
84.07	1.30	N/A	N/A	N/A	N/A	N/A	N/A
103.95	1.30	N/A	N/A	N/A	N/A	N/A	N/A
107.57	2.05	N/A	N/A	N/A	N/A	N/A	N/A
127.68	2.05	N/A	N/A	N/A	N/A	N/A	N/A
150.12	2.05	N/A	N/A	N/A	N/A	N/A	N/A

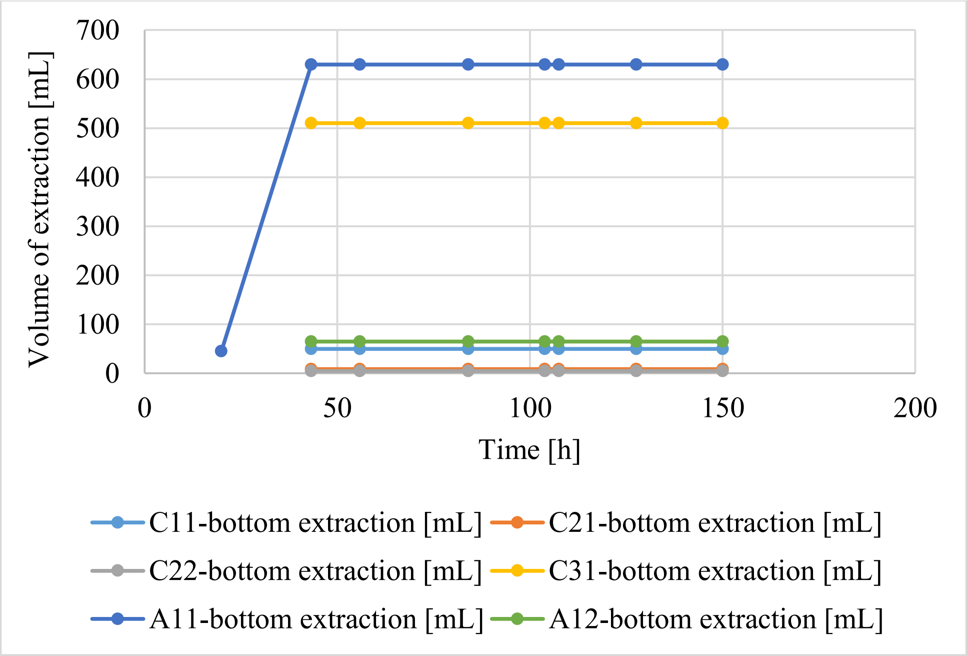

Figure 4-44 Volume of anolyte and catholyte, extracted from the bottom of the reactor during the period of Test L

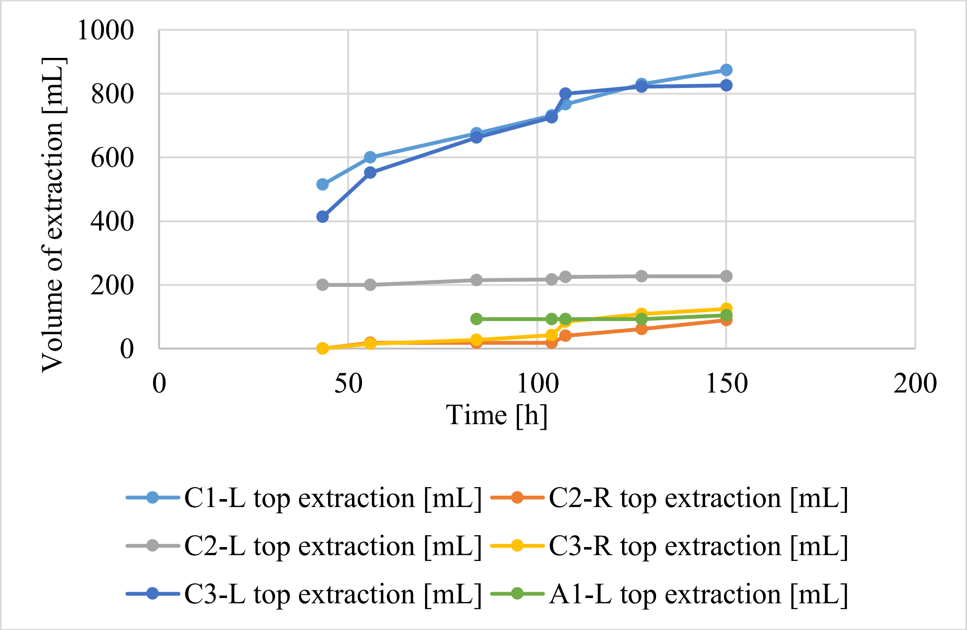

Figure 4-45 Volume of anolyte and catholyte, extracted from the top of the reactor during the period of Test L

Results regarding the recovery percentages of oil and water are exhibited in Table 4-39.

Table 4-39 Recovery percentages of oil and water for Test L

Time [h]	Oil [%]	Water [%]
0.00	0.00	0.00
20.00	0.00	0.71
43.28	10.61	21.86
55.95	12.12	23.62
84.07	13.22	26.79
103.95	13.51	28.52
107.57	13.98	30.68
127.68	14.14	32.53
150.12	14.45	33.73

Note: 2 % of the extracted water was occupied by the suspended fine fractions (porosity of 0.98 was assumed.).

During the first hours of the experiment, the rate of the extraction was higher than the next hours with the voltage gradient of around 1 V/cm (Table 4-40). After 104 hours, the voltage gradient was set to a recommended level of 2.05 V/cm, leading to a sudden increase in the current value and the catholyte extraction rate.

Table 4-40 Total anolyte and catholyte extraction and volume reduction inside the 20 L reactor

Time [h]	**Total catholyte extraction [mL/h]**	**Total anolyte extraction [mL/h]**	**Volume reduction inside the reactor [mL]**	**Percentage of volume reduction [%]**
0.00	N/A	N/A	N/A	N/A
20.00	N/A	2.25	N/A	N/A
43.28	39.35	16.05	3326.40	18.80
55.95	35.01	12.41	4392.36	24.83
84.07	25.83	9.37	5223.96	29.53
103.95	22.21	7.58	5462.44	30.88
107.57	23.15	7.32	5549.04	31.37
127.68	20.54	6.17	5628.42	31.82
150.12	18.09	5.33	6112.26	34.55

After 127.68 hours, ferric chloride was added to the system to study the impact of electrocoagulation on the EK process. Adding the coagulant resulted in maintaining almost the same rate of catholyte and anolyte extraction as it was calculated during the previous 20 hours. Generally, an additive changes the properties of sludge by neutralizing the colloidal charges and influencing the diffuse double layer. Subsequently, the flow of the water phase is promoted. An additive might also increase the conductivity of the system which leads to higher amperage and higher extraction rate. Considering the initial content of the sludge, 34% of the water was removed from the matrix and over 14% of the oil was recovered. The remaining oil seemed to have a high

affinity to the solid phase since the jelly initial sludge was not observed at the end of the test. In this case, a subsequent step of the EK treatment, similar to the EK treatment of contaminated soil, might be applied.

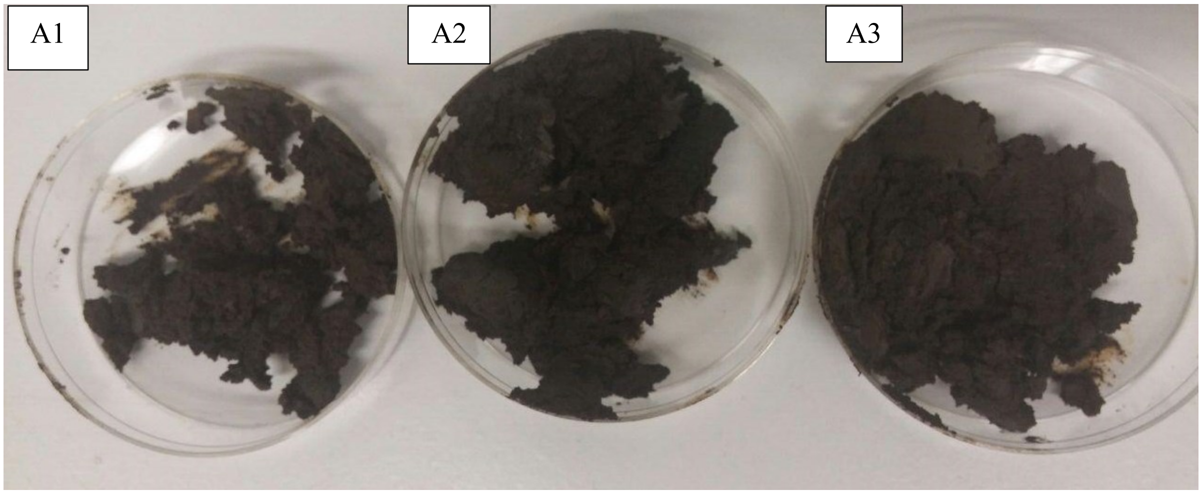

Figure 4-46 Samples of solids accumulated next to the anodes A1, A2, and A3 in Test L

Furthermore, another horizontal motion with regards to the organic solids took place at this Phase. Fractions were transported toward the anodes, mostly due to the electrophoresis phenomenon. They were compacted due to the continuous removal of water hidden in their conglomerate/flocs. Subsequently, the volume reduction of the solids took place not only through the removal of liquids but also due to the simultaneous compaction of solids within the EK reactor (Figure 4-46). A volume reduction of around 35% was achieved (Table 4-40). It was concluded that the EK treatment could also decrease the costs of the solids' final disposal. Moreover, solidified sludge facilitates solids manipulation and decreases labor and transportation costs. Nevertheless, such high-quality solids present an added-value product with improved rheological parameters and low emission of VOCs. Such products can generate revenue if applied to construction works.

The results showed that a bigger volume of liquid was extracted from the cathode side than the anode side (Figure 4-42). It seemed that the applied current generated a big horizontal electroosmotic flow toward cathodes. Both water and oil were collected on that side, however, an excellent vertical separation was observed (Figure 4-47). A significant amount of water was collected at the beginning of Test L. A part of the extracted water was occupied by fine solids, which escaped the screen filters. These solids that had colloidal properties created a suspension in the extracted water. These solids were most likely originated from the geological strata and were transported through the extraction wells. In this case, the extracted water should undergo further treatment in a wastewater treatment plant if the reuse of such water is considered. Sludge should be combined with a coagulant (e.g. ferric chloride) before the EK process in order to produce water of excellent quality without the necessity of further treatment measures (section 4.3).

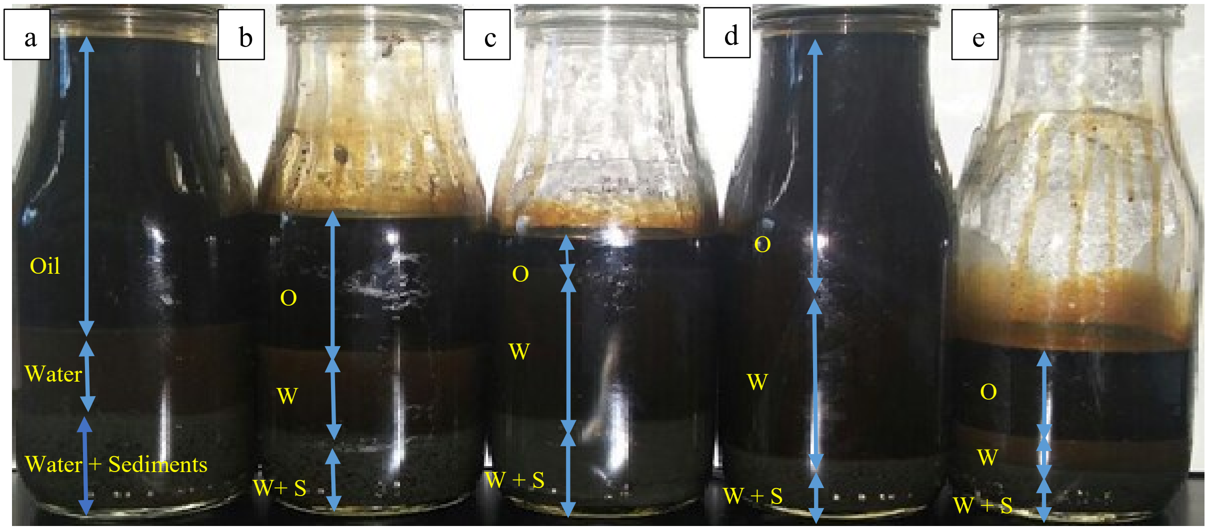

Figure 4-47 Catholyte collection from the top after 43.28 hours: a) from C1, b) from C1, c) from C2, d) from C3, e) from C3

Subsequently, after 5 days of EK treatment without a conditioner, the remaining sludge in the large EK reactor was mixed with ferric chloride for further investigation of the results. An increase in the rate of extraction was observed.

In order to assess the quality of the collected liquids, the representative samples were subjected to the following analyses: viscosity, calorific value and standard BS&W (Table 4-41 and 4-42).

Table 4-41 Calorific value and viscosity for samples collected after different time periods

Sample	**Calorific value [kJ/kg]**	**Viscosity [mPa.s]**
Raw oily sludge before treatment	10835	1400.0
C31 bottom after 43.28 h	42744	93.0
A11 bottom after 43.28 h	41095	98.3
C3 top after 43.28 h	40415	142.0
C3 top (left side) after 127.68 h	37163	N/A
A1 top (left side) after 127.68 h	41592	N/A
C1 top (left side) after 150.12 h	39246	N/A
A1 top (left side) after 150.12 h	41436	N/A

Note: C and A refer to cathode and anode, respectively; top and bottom refer to the top and bottom extraction, respectively.

The results (Table 4-41) showed that the raw oily sludge, before EK treatment, had a high viscosity (1400 mPa.s) and a low calorific value (10,835 kJ/kg). Manipulation of such material was difficult, and the cost of pumping and transporting was very high. The use of such sludge as fuel was not cost-effective due to a low calorific value. After EK treatment, the samples represented the viscosity of a fuel (e.g. 93 mPa.s). Furthermore, the calorific value increased to a value of around 41000 kJ/kg which was similar to the value of crude oil, even after the addition of a coagulant. It was noteworthy that the anolyte oil had a higher calorific value than the catholyte oil which was accompanied by the extracted water.

BS&W analysis was conducted following ASTM D4007-11 (reapproved 2016). The dedicated samples from bottom and top liquid collections were subjected to the analysis. The results showed that all oil samples collected from the top had water fraction that was ranged between 0 to 5% (an average of 2.8%), while the solids were undetectable (Table 4-42 and Figure 4-48).

Table 4-42 Results of BS&W analysis on samples collected after different periods for Test L

Sample	**Oil [%]**	**Water [%]**	**Sediments [%]**
Raw oily sludge before treatment	60	30	10
Anode 11 bottom after 43.28 h	98	2	~0
Cathode 31 bottom after 43.28 h (before further phase separation by gravity)	79	21	~0
Cathode 2 top after 43.28 h	96	4	~0
Cathode 3 top after 43.28 h	97	3	~0
Cathode 3 (left side) after 127.68 h	95	5	~0
Anode 1 top (left side) after 127.68 h (pure oil)	100	0	~0
Cathode 1 top (left side) after 150.12 h	98	2	~0

Note: C and A refer to cathode and anode, respectively; top and bottom refer to the top and bottom extraction, respectively.

The cumulative energy consumption in a large-scale EK reactor containing oil sludge is shown in Table 4-43. The results showed the energy consumption of 3.92 kWh after phase separation, 6.11 kWh after the first liquid extraction, and 11.62 kWh at the end of the test. The fluctuation in energy consumption was related to the change of matrix properties as well as the variability of the applied voltage (Figure 4-49).

The cost analysis was done for the industrial Quebec market with a cost of 0.035 CAD for 1 kWh of energy as well as for the USA market with a cost of 0.09 USD/kWh. In both cases, the energy cost was insignificant for a large laboratory reactor. It was around half a CAD and 1 USD for 150 hours of treatment, respectively (Table 4-43).

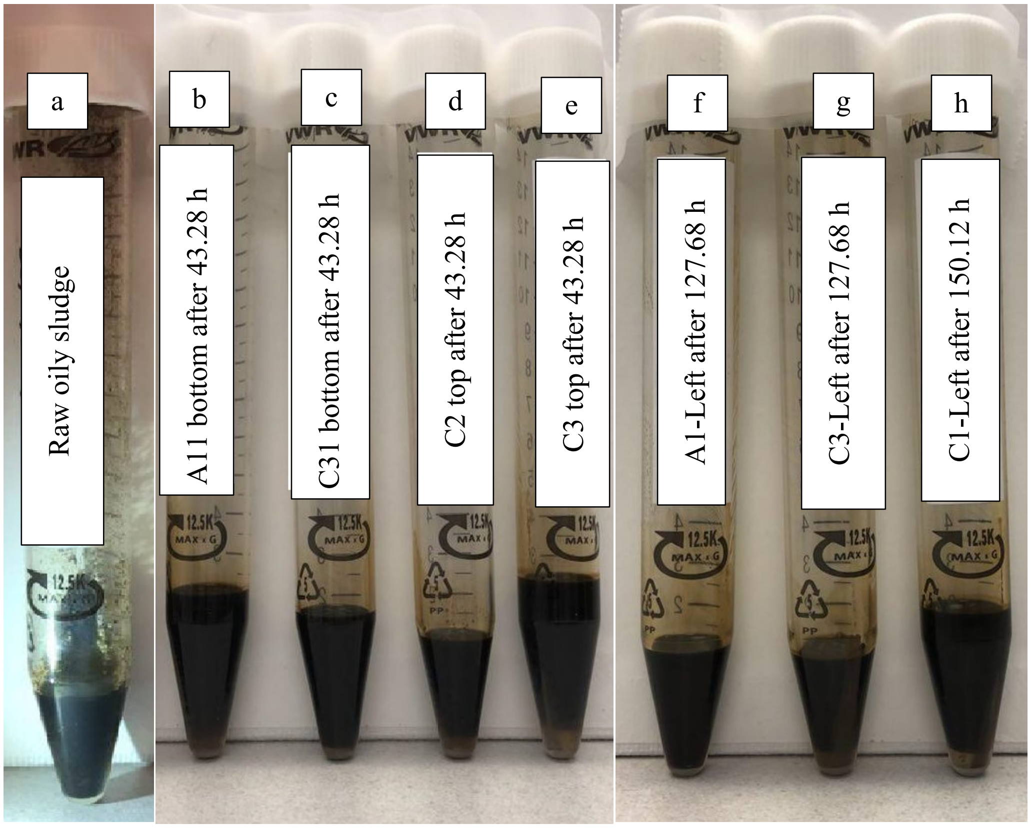

Figure 4-48 Samples after BS&W test: a) raw oily sludge; b) bottom extraction from A11 after 43.28 h; c) C31 bottom, 43.28 h; d) C2 top, 43.28 h; e) C3 top, 43.28 h; f) A1-Left top, 127.68 h; g) C3-Left top, 127.68 h; h) C1-Left top, 150.12 h

Table 4-43 Cumulative energy consumption and cost of energy for Test L

Time [h]	**Voltage gradient [V]**	**Cumulative energy consumption [kWh]**	**Cumulative cost [CAD]**	**Cumulative cost [USD]**
0.00	1.00	0.00	0.00	0.00
20.00	0.79	3.92	0.14	0.35
43.28	1.30	6.11	0.21	0.55
55.95	1.30	6.81	0.24	0.61
84.07	1.30	7.66	0.27	0.69
103.95	1.30	8.50	0.30	0.77
107.57	2.05	9.46	0.33	0.85
127.68	2.05	10.61	0.37	0.95
150.12	2.05	11.62	0.41	1.05

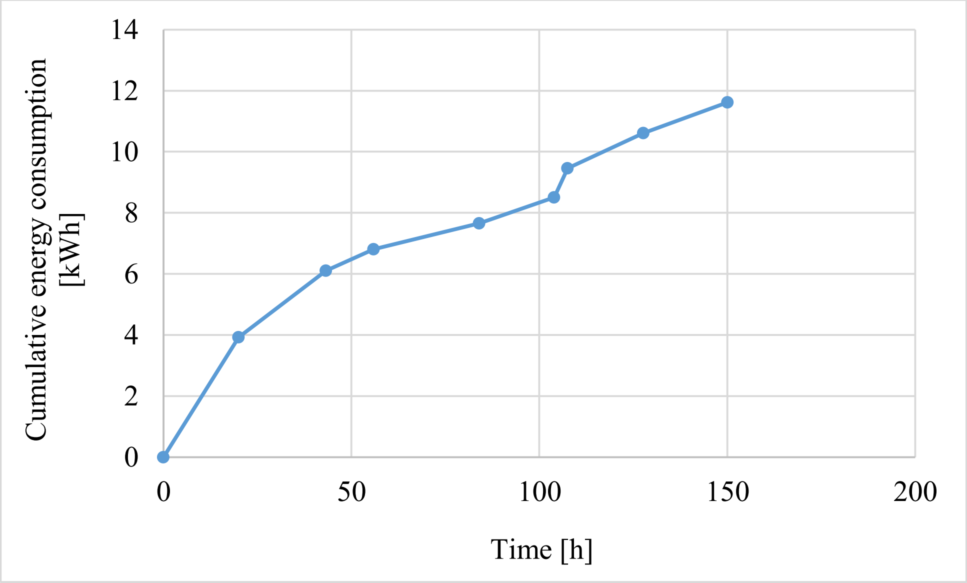

Figure 4-49 Cumulative energy consumption versus time for Test L

General conclusion of phase 5

Tests at the large scale of 20 L confirmed the feasibility of oil recovery from oil sludge using EK extraction; over 30% of water and over 14% of oil were recovered during this test. The remaining material in the reactor also underwent demulsification, but its extraction rate decreased to a very low level due to the lower current value. In such a case, a subsequent step of EK treatment, similar to EK treatment of contaminated soil, might be applied. EK treatment decreased the volume of disposable material by 35%, which might facilitate solids manipulation and decrease labor and transportation costs. Since the high-quality solids present an add-value product, they generate revenue when sold to the road construction industry.

The quality of the recovered oil was comparable to the crude oil considering its calorific value of over 41000 kJ/kg.

BS&W analysis showed that the recovered oil had acceptable average sediment and water content of less than 3%.

Based on the observation conducted in Test L, it was recommended to gradually increase the voltage gradient in the EK system. Keeping a low voltage gradient permitted to decrease the operation costs, decrease the impact of physicochemical phenomena at the electrodes, and slow down the changes within the reactor matrix. Such an approach might increase the exposure time but it also increases the quality of the extracted fluids. Therefore, a voltage gradient of around 2 V/cm was suggested. Furthermore, the application of a conditioner could improve the extraction process.

Overall, EK treatment of oil sludge, a water-in-oil suspension, has met the defined objectives and could proceed to pilot feasibility studies.

4.6 Phase 6: Design and operation of a continuous-flow EK reactor (Test CF)

A volume of 998 mL of oil sludge (sample C_{mix}) was exposed to a constant voltage of 10.2 V (voltage gradient of 2.05 V/cm) during Test CF. Table 4-44 and 4-45 show the volume of extracted liquids and recovery percentages of oil and water for Test CF, respectively.

Table 4-44 Volume of extracted water and oil for Test CF

Time [h]	Water [mL]	Oil [mL]	Total [mL]
0	0.0	0.0	0.0
6	38.0	29.5	67.5
13	76.5	42.0	118.5
20	85.5	46.5	132.0
24	97.0	53.0	150.0
35	106.0	56.0	162.0

Table 4-45 Recovery percentages of water and oil for Test CF

Time [h]	Water [%]	Oil [%]
0	0.00	0.00
6	12.06	6.18
13	24.28	8.79
20	27.13	9.74
24	30.78	11.10
35	33.64	11.72

Note: 2 % of the extracted water was occupied by the suspended fine fractions (porosity of 0.98 was assumed.).

Based on the results from the previous phases, a retention time of 24 hours was expected to be sufficient for proper phase separation. The filling process took 6 hours, in which oil sludge was gradually fed to the reactor with an average flow rate of 166.3 mL per hour.

All extractions were collected from the bottom outlet of the reactor. Mixing the sludge was impossible during the test due to the specific design of the reactor, nevertheless, it would have enhanced the extraction.

The first collection of the separated oil and water took place after 6 hours, and excellent water recovery of 12% and oil recovery of 6% were achieved (Table 4-45). The extraction of water continued with almost the same rate for another 7 hours and the water recovery was doubled after the second extraction. The elimination of water from the system led to a decrease in conductivity. Therefore, water extraction continued at a slower pace for the rest of the experiment (Figure 4-50).

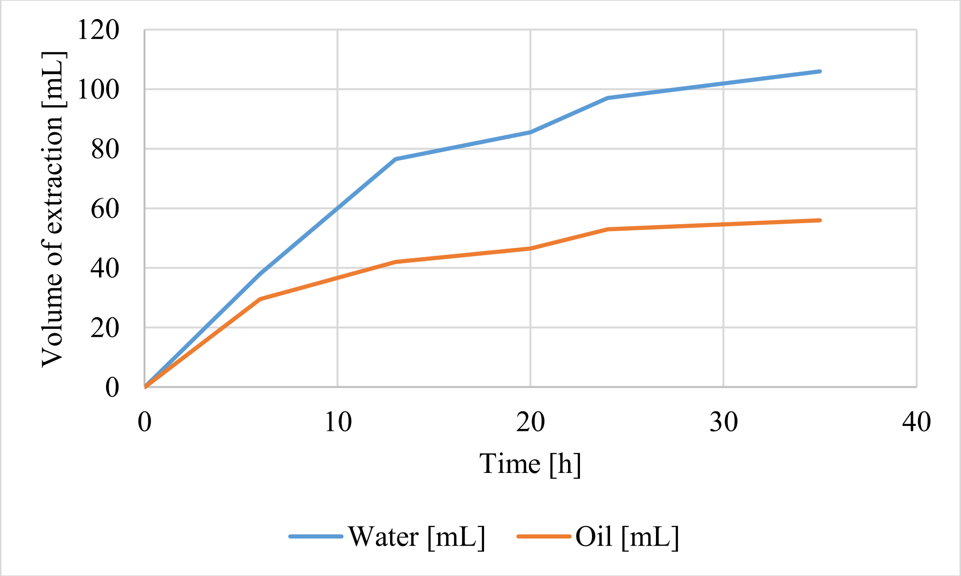

Figure 4-50 Volume of extracted oil and water versus time for Test CF

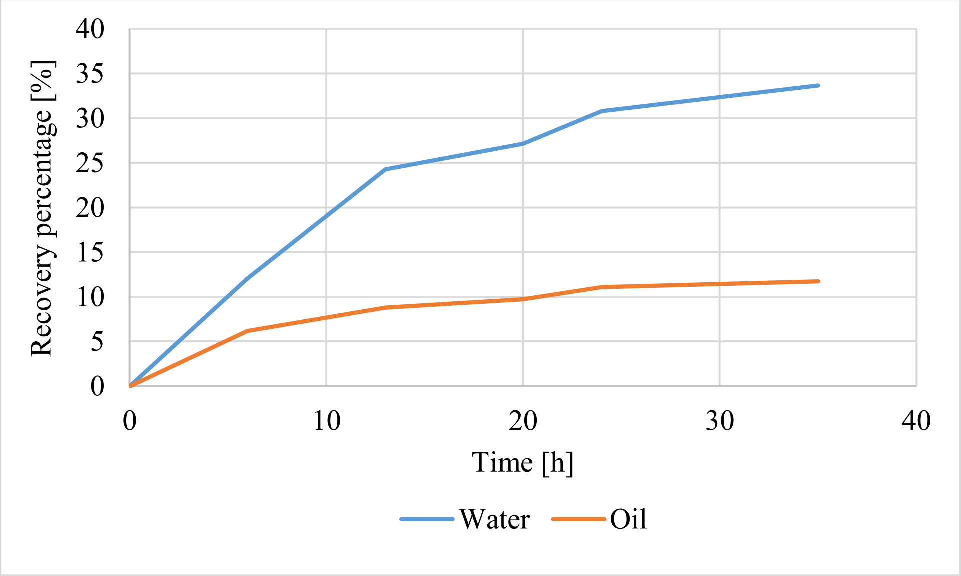

Figure 4-51 Recovery percentages of oil and water versus time for Test CF

On the other hand, the recovered oil was about half of the recovered water after 6 hours, and the rate of oil recovery continued to decrease after the first extraction took place. After 24 hours, the recovery percentages of oil and water were 11% and 31%, respectively. Water was recovered more than twice as much, compared to the oil. This was due to the higher mobility of water (Figure 4-51).

The results of the large-scale batch experiment (Test L) showed that a water recovery of 31% and an oil recovery of 11% could be achieved after 108 h and 47 h, respectively (Table 4-39), while the same recovery percentages would be achieved by the continuous-flow system within a 24-hours retention time (Table 4-45). This comparison proved the significant operating efficiency of the designed continuous-flow reactor. Figure 4-52 illustrates the excellent separation of water and oil after different periods.

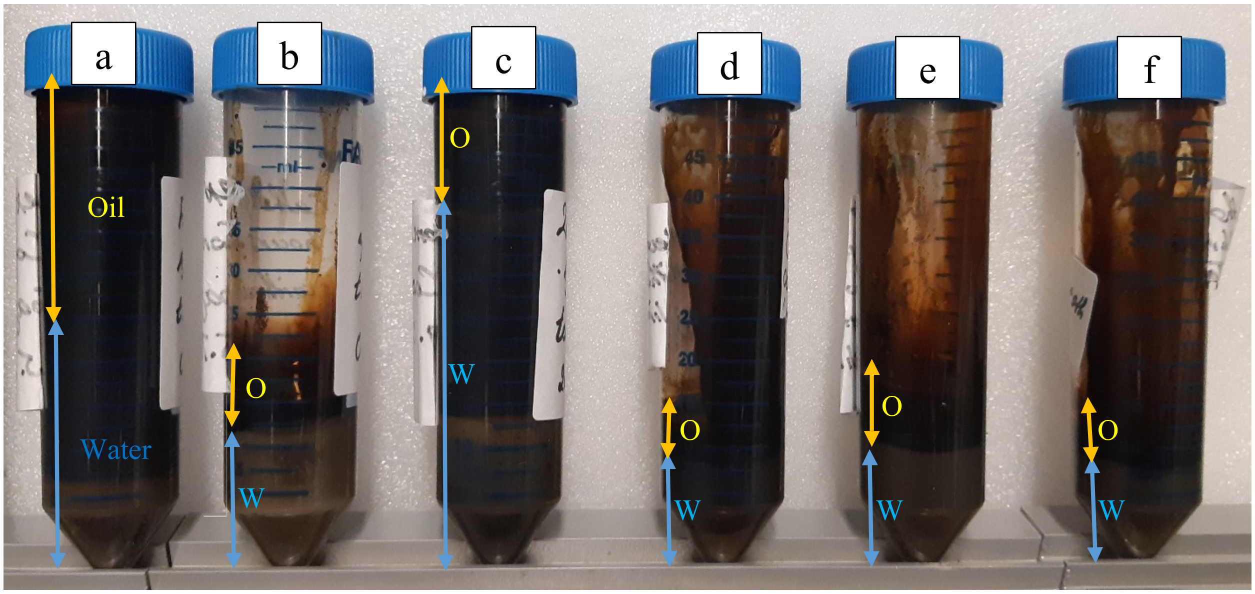

Figure 4-52 Extracted water and oil after a,b) 6 h, C) 13 h, d) 20 h, e) 24 h, f) 35 h

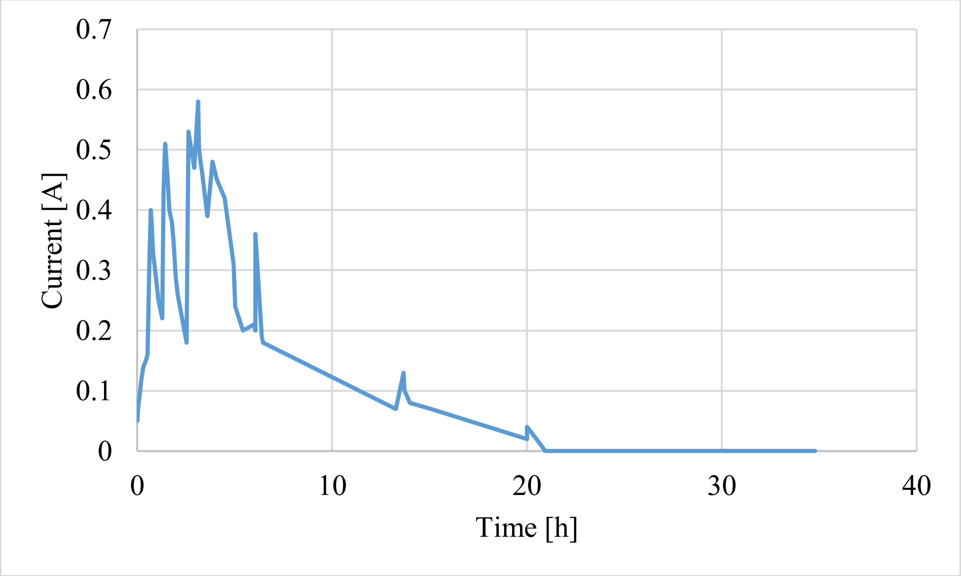

Figure 4-53 Current changes versus time for Test CF

According to Figure 4-53, the initial value of the current was 0.05 A, which represented the time when sludge reached the electrodes for the first time. The current value increased as more volume of the sludge was exposed to the electrical field. Therefore, the current increased after each addition of the sludge to the system during the first 6 hours of the experiment (filling process). Then, it decreased until the next portion of sludge was added. The highest value of the current during this period was 0.58 A.

After the first 6 hours, the current continued to decrease as a result of the elimination of water and the reduced conductivity of the system, however, it increased slightly after each extraction due to an increase in the electrical potential.

After 21 hours, the current value decreased to zero (without the addition of new material). At that time, about 136 mL of oil and water was extracted which presented a 14% reduction in the volume of the sludge (Table 4-46). This value was calculated without considering a certain simultaneous decrease of the volume due to the compaction of solids. Since the electrodes were placed horizontally, the compacted sludge could not contact the anode, and the conductivity of the system was most probably interrupted by this division.

Table 4-46 Reduction in the volume of sludge inside the reactor for Test CF

Time [h]	**Volume reduction [mL]**	**Volume reduction [%]**
0	0.0	0.00
6	67.5	6.76
13	118.5	11.87
20	132.0	13.23
24	150.0	15.03
35	162.0	16.23

According to Table 4-46, a volume reduction of 15% was achieved for the oil sludge only due to the extraction of liquids. About half of this value was obtained only after 6 hours. The promising results with regards to the reduced volume of the remaining solids suggested an outstanding function of the introduced continuous-flow system.

Table 4-47 presents the cumulative energy consumption during the experiment and the cost of energy in both Quebec and the USA markets. The results indicate that most of the energy was consumed during the first 13 hours of the experiment. Due to the higher conductivity of the system and the higher value of the current, about 50% of the total energy consumption through the electrodes happened in the first 6 hours of the experiment. An amount of 0.033 kWh of energy was consumed by the step motor during the first 6 hours of the test (filling process). Overall, the total energy consumption and the cost of energy were insignificant for a 1 L laboratory reactor.

Table 4-47 Cumulative energy consumption and cost of energy for Test CF

Time [h]	**Cumulative energy consumption [kWh]**	**Total cumulative energy consumption* [kWh]**	**Cumulative cost [CAD]**	**Cumulative cost [USD]**
0	0.000	0.000	0.0000	0.0000
6	0.021	0.054	0.0019	0.0049
13	0.037	0.070	0.0025	0.0063
20	0.042	0.075	0.0026	0.0068
24	0.043	0.076	0.0027	0.0068
35	0.043	0.076	0.0027	0.0068

*Total cumulative energy consumption: the sum of electrical energy that was applied to the electrodes and energy consumed by the step motor

Table 4-48 Cumulative energy consumption for 1 mL of oil and water extraction after 24 hours for different tests

Test	Oil [%]	Water [%]	Oil [mL]	Water [mL]	Cumulative energy consumption [kWh]	Energy per 1 mL of oil recovery [kWh/mL]	Energy per 1 mL of water recovery [kwh/mL]
Test S3	3.86	6.33	2.99	4.9	0.060	0.0201	0.0122
Test M1	13.50	7.25	204.75	106.1	1.660	0.0081	0.0156
Test L	5.88	12.12	500.63	677.0	4.300	0.0086	0.0064
Test CF	11.10	30.78	53.00	97.0	0.076	0.0014	0.0008

Table 4-48 exhibits a comparative study on different tests and different scales concerning energy consumption for 1 mL of recovered oil and water after the first 24 hours of the experiment. Test S3, Test M1, Test L, and Test CF represented the small-scale (200 mL), the medium-scale (4 L), the large-scale (20 L), and the 1 L continuous-flow system, respectively. According to the results, energy consumption did not necessarily increase with the same ratio as the volume of the reactor and the matrix. For example, the energy consumption of Test L was not 100 times bigger than the energy consumption of Test S3, nevertheless, the recovery percentages of oil and water were higher for the first 24 hours of the test. The results proved that scaling up the EK system would reduce the required energy for the recovery of 1 mL of oil and water. The energy consumption per 1 mL of water and oil recovery for the small EK reactor (Test S3) was 0.012 and 0.02 kWh/mL, respectively when the same parameter for the large EK reactor (Test L) was 0.006 and 0.009 kWh/mL, respectively.

On the other hand, more than twice as much water was recovered in Test CF as it was in Test L while the consumed energy for the recovery of each mL of water was one-eighth of the consumed energy for Test L. A similar trend was observed for the recovery of oil and the energy consumption per 1 mL of oil recovery. It was concluded that by scaling up the continuous-flow EK reactor, the recovery percentages of water and oil would increase even more.

General conclusion of phase 6

The continuous-flow system operates based on 3 steps; filling, exposure to EK treatment, and evacuation. The duration of Test CF was 24 hours to investigate the efficiency of the design and operating conditions in an optimal retention period. The continuous supply of the sludge took place over the first 6 hours and with an average flow rate of 166.3 mL per hour.

It was concluded that a voltage gradient of 2.05 V/cm was effective. Also, the horizontal installment of the electrodes on the inner surface of a tubular reactor and the movement of the sludge with the assistance of a screw were efficient.

Based on the previous results, the addition of a coagulant such as ferric chloride would help maintain the rate of extraction and the conductivity of the system during the treatment process. It would also enhance the clarity of the recovered water.

Overall, significant water recovery of 31% and oil recovery of 11% were achieved after only 24 hours. The results indicated the advantages of the application of the continuous supply system for oil sludge treatment.

5 Conclusions and future work

5.1 Conclusions

Considering all results, the EK treatment method proved to be a successful approach towards sustainable management of oil sludge, hence the pilot-scale studies should be performed.

The BS&W analysis revealed that the recovered oil contained an acceptable value of less than 1% and 3% of sediments and water in medium-scale and large-scale tests, respectively.

The results of the calorific value tests demonstrated that the recovered oil in both medium and large-scale tests is comparable to the crude oil with a calorific value of above 41000 kJ/kg.

The optimal conditions for enhanced EK treatment were investigated. It was concluded that a voltage gradient of 2.05 V/cm would improve the two-sides recovery (anolyte and catholyte) and the clarity of anolyte water.

A higher voltage gradient would reduce exposure time. On the other hand, a low voltage gradient would decrease the operating costs and the physicochemical phenomena at the electrodes and increase the quality of the extracted material.

Conditioning the EK treatment proved to be effective. Introducing ferric chloride to the system with a volumetric ratio of 1 to 300, demonstrated an enhancement in the light oil, heavy oil, and water recovery as well as the quality of the extracted water.

Batch experiments were conducted on a small scale of 200 mL, a medium scale of 4 L and a large scale of 20 L. The medium and large-scale tests confirmed the results of the small-scale investigations. Besides, the performance of the continuous-flow system was examined. Significant recovery of water and oil was observed over 24 hours of exposure to the EK treatment. The results indicated that the continuous-flow system had higher efficiency than the batch system.

It was concluded that the energy requirement for 1 mL of oil and water recovery would decrease with scaling up the EK reactor; therefore, performing the electrokinetic remediation at a higher scale would be more cost-effective. It would also be more efficient with respect to the recovery of oil and water.

The EK treatment proved to achieve a high water recovery since the remaining solids presented a very low amount of water distributed on a paper filter. The volume of disposable material reduced by almost 50 % and 35% in the medium-scale and large-scale experiments, respectively. This would result in a significant decrease in disposal costs and easier transportation and manipulation of the solids. Moreover, the high-quality solids present an add-value product that generates revenue if it is sold to the road construction industry.

The developed EK reactors are ready to scale up to full industrial level belonging to upstream and downstream petroleum facilities. The developed EK systems can be also applied to sludge produced by other types of industrial and municipal plants.

5.2. Novelty

The novelty of this work is based on the following reasons:

i) developing adequate operation conditions for a 4-phase separation (water, light oil, heavy oil, and solids) of the oily sludge (rather than a 3-phase separation);

ii) designing advanced configurations of batch electrokinetic reactors for oil sludge treatment;

iii) developing a novel continuous-flow reactor and testing it successfully for more advanced treatment of oil sludge;

iv) produced added-value products based on the new design of electrokinetic reactors.

5.3 Future work

The medium and large-scale studies confirmed the optimal design and conditions that were obtained from the small-scale tests. Therefore, the pilot-scale experiments could be initiated in order to study the results of the EK treatment of oil sludge at the industrial scale.

The following suggestions are made to enhance the future results of the experiments in this field:

Increasing the extraction rate of water and oil by providing an evacuation system that permits a consistent flow of the liquids. In such a system, solids would not be able to block the flow of oil and water.

Examining the quality of water by studying its composition and probable contaminants in order to determine the feasibility of its recycle and reuse after adequate treatment.

Studying the composition of the remaining solids after the treatment in order to investigate its application in the road construction or other industries.

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