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Improving the Durability of Silicone Sealants through Polyblending

Lesia Maria Beznaczuk

A Thesis

in

The Department

of

Building Engineering

Presented in Partial Fulfillment of the Requirements
for the Degree of Master of Engineering at
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Montreal, Quebec, Canada

July, 1985

ABSTRACT

Improving the Durability of Silicone Sealants through Polyblending

Lesia Maria Beznaczuk

A study has been conducted on the effect that polyblending, with vinyl polymers, has on the durability and mechanical properties of silicone (poly(dimethylsiloxane)) sealants. The types of polymers utilized as additives, in proportions by weight of five, ten, and fifteen per cent, were 1) vinyl polymers (PVC, VC/VAc), 2) three different nitrile elastomers (modified and unmodified), and 3) a natural polymer (Kraft lignin). Sealant beads were cast between wood, aluminum, and mortar substrates, and were then exposed to laboratory control, accelerated, and natural weathering conditions. Mechanical (tensile) testing was performed after weathering exposures were completed to determine stress-strain behaviour and toughness. Thermal testing, using a differential scanning calorimeter (DSC), determined glass transition temperatures and miscibility. Scanning electron microscopy (SEM), in conjunction with energy dipersive x-ray analysis (EDXA), related the morphology to durability and performance. Results indicate a definite improvement in mechanical properties and enhanced weathering resistance, through polyblending, for wood and aluminum substrates.

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TABLE OF CONTENTS

page

LIST OF FIGURES

page

LIST OF TABLES

LIST OF APPENDICES

LIST OF SYMBOLS

ACN	acrylonitrile
ASTM	American Society for Testing and Materials
A.W.	accelerated weathering
CGSB	Canadian Government Standards Board
CSA	Canadian Standards Association
DP	degree of polymerization
DSC	differential scanning calorimeter
DMA	dynamic mechanical analysis
EDXA	energy dispersive x-ray analysis
K211	nitrile rubber (Krynac 211, Polysar)
K1402	nitrile rubber (Krynac 1402 H83, Polysar)
K1411	nitrile rubber (Krynac 1411, Polysar)
N.W.	natural weathering
PDS	poly(dimethylsiloxane) silicone sealant
PVC	poly(vinyl chloride)
R.H.	relative humidity
RTV	room temperature vulcanizing
SEM	scanning electron microscope
Tg	glass transition temperature
U.V.	ultra-violet
VC/VAc	vinyl chloride - vinyl acetate copolymer

1. OBJECTIVES AND SCOPE OF RESEARCH

Sealants play a very important role in the overall efficiency of the building. Sealing a building properly is a critical step in assembling a building envelope. Thus the durability of any chosen sealant can be one of the factors affecting the service life of a building.

Improving the durability of sealants, silicone in particular, is the chosen topic of this research. Through mechanical polyblending, a relatively simple process that can be performed in any laboratory, selected additives were mixed (in various proportions by weight) with silicone sealant. These additives consisted of two vinyl polymers, three different nitrile rubbers, and Kraft lignin. Specimens were prepared on three different substrates: wood, aluminum, and mortar. The specimens were sized so that they would be best representative of a building joint in service. The size of the substrates was the standard one used by many researchers for sealants to be tested in tension. Tensile testing best represents the type of movement that causes failure in service.

To determine whether any of the polyblends are more durable than silicone, three series of specimens have been weathered. The first series were kept in laboratory conditions (24°C and approximately (~) 35% relative humidity) as control specimens. The second were exposed artificially in an accelerated weathering chamber, cycled four times daily between -30°C and +30°C. The third series were naturally weathered in a relatively polluted area of downtown Montreal. After exposure, tensile testing was performed to provide a basis for comparison of durability.

2. BUILDING SEALANTS AND THEIR PERFORMANCE

Building sealants evolved as a result of the use of larger (and lighter) module building materials, which consequently gave rise to large thermal differential movements. These movements are cyclic due to the daily and seasonal variations in the exterior thermal and moisture conditions of the building. Sealants which fill the joints between these exterior bulding materials must be formulated to withstand extension at low temperatures and compression at high temperatures. If the working capabilities of the sealant are exceeded, failure can occur and the sealant will be unable to fulfill its primary function of preventing the passage of air, moisture, heat, dust, etc. through the joint. Thus the proper performance of the sealant within the joint for a reasonable service life is critical to the efficiency of the building envelope. Some typical sealant applications are illustrated in figure 1.

The service life of a sealant is determined by many factors:

1) temperature
2) moisture
3) exposure to ultra-violet radiation
4) amount and severity of extension and compression cycles

One of the most important influences on service life is a combination of several of these factors - how well it

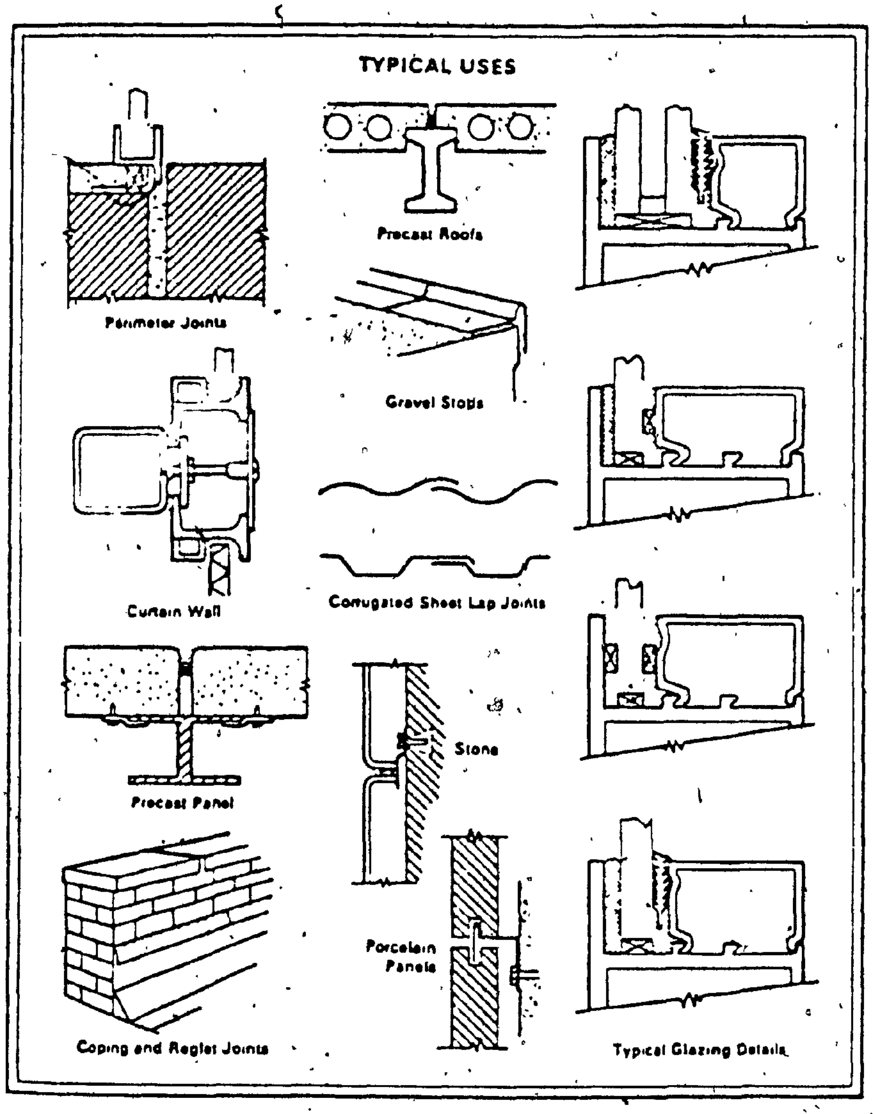


Figure 1 Typical sealant applications (4)

weathers. Weathering, a term which refers to how a material performs when exposed to environmental conditions, can affect a sealant in many ways and can lead to various degrees of degradation. Therefore improving the durability of a sealant against weathering will consequently extend service life and expand its capabilities. Durability, however, is not an inherent property of any material because it depends on the installation, use, and service conditions during its working life.

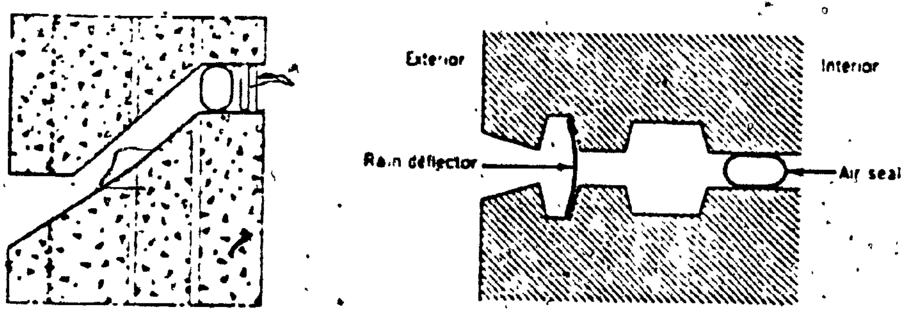
2.1 Classification

Sealants can be classified into four types:

1) viscous liquids
2) mastics
3) tapes
4) preformed gaskets.

Viscous liquids are pourable, mastics contain a thixotropic agent to control flow and prevent sagging, tapes are used as bedding compounds or in glazing work, and preformed gaskets are extruded or foamed into a multitude of shapes. Mastic sealants are available as one-component or two-component systems. One component are ready to install, and are cured by moisture absorption or solvent evapouration. Two-component sealants come in two packages which must be mixed on site prior to installation, and are chemically cured.

Exterior sealants can act as either one-stage or two-stage weatherproofing. One stage weatherproofing (fig. 2(a)) uses the seal, at the exterior face of the joint, as both a rain and air seal. Two stage weatherproofing (fig. 2(b)) uses the seal, at the interior face of the joint, as an air seal only. A non-airtight rain seal is used in an exterior location of the joint to protect the air seal from weathering effects.



(a) (b)

Figure 2 a) Horizontal joint with rain seal, b) vertical joint with rain seal and air seal (4)

2.2 Categories of Joints

The majority of joints fall into the following categories:

1) working, which change size and shape with relative movement of the substrates
2) non-working, whose movements are minimized or eliminated
3) butt (fig. 3(a)), which subject the sealant to alternating tensile and compressive stresses
3) lap (fig. 3(b)), where the sealant is primarily subjected to shear stresses.

The shape and dimension of the sealant cross-section is of primary importance on the the movement induced strains and

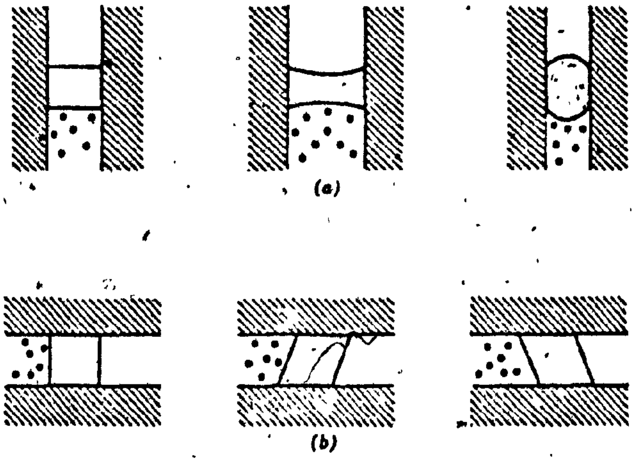


Figure 3 Deformation of sealant beads: a) butt joints, b) lap joints (4)

stresse (fig. 4). The optimal geometry of a seal is twice as wide as it is deep, but a ratio of one is used as a practical joint to reduce the frequency of failure (fig. 5). The bottom surface of the sealant must also be allowed to deform freely when movements are induced by the substrates bonded to the two opposite sides of the joint (fig. 6).

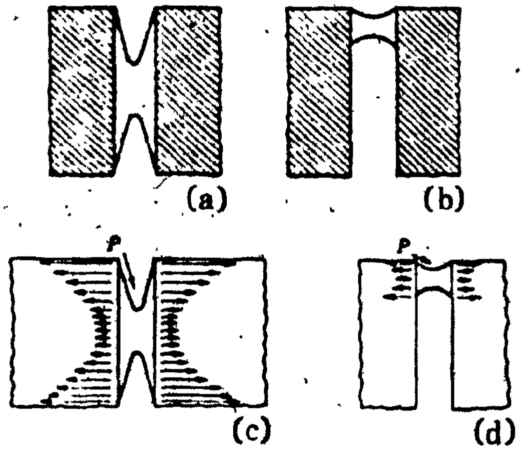


Figure 4 Comparison of maximum strains: a) for 50 mm deep seals - extension of joint 100%, sealant strain 550%; b) for 12,5 mm deep seals - extension of joint 100%, sealant strain 160%. Comparison of substrate stresses for: c) 50mm deep seals; d) 12,5 mm deep seals (4)

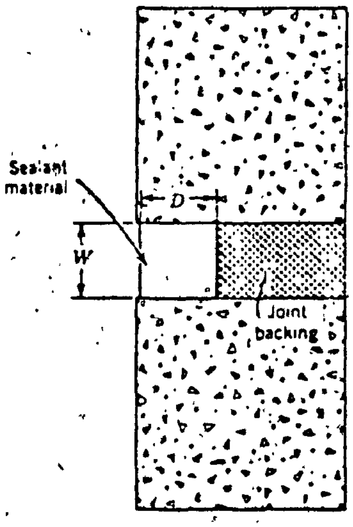
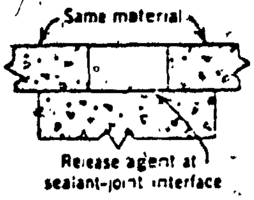


Figure 5 Joint with shape factor of one (W/D = 1) (4)



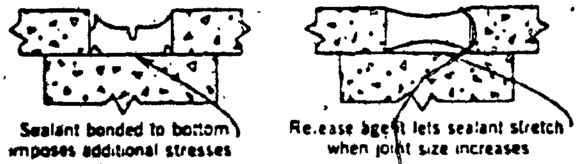


Figure 6 Effect of release agent on sealant (4)

2.3 Performance Classification

The height and mass of the building structure, wind loads, moisture absorption, amount of shade, compass orientation, ambient temperature, and colour of substrate dictate the movement that the building joints must endure. The greater the amount of movement, the more elastic and durable the sealant must be (fig. 7). The properties and performance of a sealant (table 1) are based on its constituents. High performance rubbery sealants that have high recovery, such as silicones, polyurethanes, and polysulfides, also have low tear resistance. Low and medium performance deformable sealants, such as oil-based and latex caulks, butyls, solvent-based acrylics, and polymercaptans, have instantaneous elasticity under short-term loads but will creep or flow under long-term loading. Representative costs (1970 prices) of various sealants are presented in table 2.

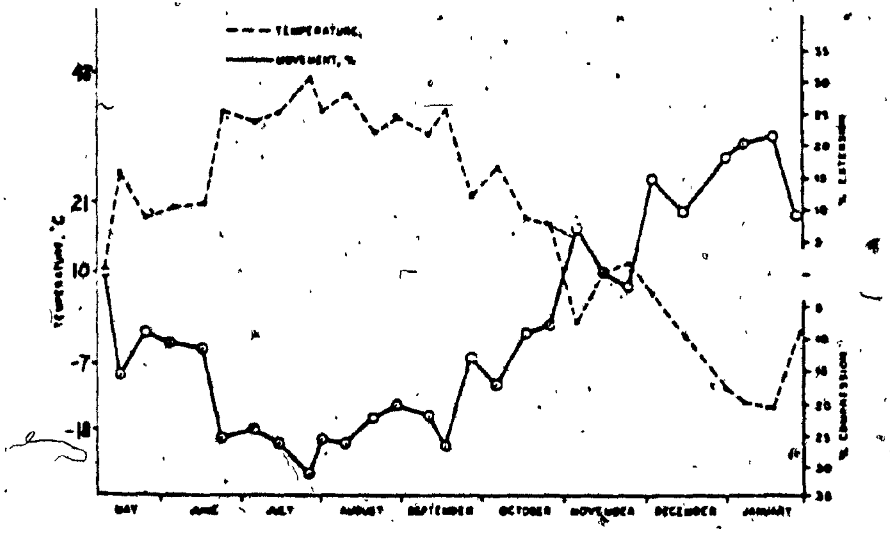


Figure 7 Expansion-contraction movement of a joint (7)

BUILDING SCIENCE INSIGHT '84

"The Performance of Materials in Use"

SUMMARY OF POPULAR SEALANT PROPERTIES

Type of Sealant	Low Performance		Medium Performance		High Performance				
	Oil-based 1-part	Latex (acrylic) 1-part	Butyl (skinning) 1-part	Acrylyc (solvent release) 1-part	Polysulfide 1-part	Polysulfide 2-part	Urethane 1-part	Urethane 2-part	Silicone 1-part
Movement capacity, percent	±3	±5	±7.5 to ±10	±7.5 to ±12½	±20	±25	±25	±25	±25 (±40)**
Maximum joint width, mm	12 - 18	12 - 18	12 - 18	18	12 - 25	18	12 - 36	50	30
Life expectancy, years	2 to 5	3 to 7	5 to 10	5 to 20	10 to 20	10 to 20	10 to 20	10 to 20	10 to 25
Service temperature range, C	-18 to 66	-17 to 71	-29 to 82	-29 to 82	-40 to 82	-40 to 82	-40 to 92	-40 to 82	-54 to 177
Recommended application temperature range (some may require heat at low end), C	4 to 49	4 to 49	4 to 49	-18*** to 49	4 to 49	4 to 49	4 to 49	4 to 49	-29 to 71
Cure time* to tack-free condition, hours	6 - 12	½ to 1	24	12 - 36	12 - 24	24 to 48	12 to 48	24 - 48	15 min - 1 h
Cure time* for specified performance, days	continuing	5	continuing	14	30 to 45	7	8 to 14	3 to 5	5 to 14
Shrinkage, percent	5	20	20	12 to 15	8 to 12	nil to 10	nil to 5	nil to 5	nil to 5
Hardness, new (1 to 6 months), A scale		15 to 40	10 to 40	25 to 50	20 to 45	20 to 45	20 to 45	20 to 85	15 to 45
Hardness, aged (5 years), A scale		30 to [illegible]	30 to 50	30 to 60	25 to 55	20 to 55	20 to 55	20 to 55	15 to 45
Resistance to extension at low temperature	very high	high	moderate to high	moderate to high	low to high	low to moderate	low to high	low to moderate	low
Primer required for full sealant bond to: Masonry Metal Glass	no no no	no no no	no no no	no no no	yes possibly possibly	yes possibly possibly	no no no	possibly possibly possibly	possibly possibly no
Clean up solvent before cure (try dry clean up first)	mineral spirits	soapy water	mineral spirits	xylol, toluol	xylol, toluol	xylol, toluol	xylol, toluol	xylol, toluol	xylol, toluol MEK
Applicable specifications, Canada	19-GP-6	19-GP-17	9-GP-14	19-GP-5	19-GP-13	19-GP-24	19-GP-11	19-GP-24	19-GP-13

*Affected by temperature and humidity

** Low modulus values in parenthesis, some sealants may have movement capacity beyond stated value

*** If application below 10°C, sealant is usually heated before use otherwise (4-49)

NOTE: It is recommended that joints should not be designed to move more than 25%

Table 1 Summary of various sealant properties (6)

Sealant Base	Cost Range (dollars per gal)	Uses
Nonhardening sealants		
Oleo-Resin	2–6	Sealing of concrete joints, masonry copings, glazing and sealing of cable pressure splices, electrical conduit, and glass-to-metal meter cases
Asphalt and bituminous	1–4	Sealing facing surface metal joints, silos, and air-conditioners. Caulking for expansion and contraction joints
Butyl	5–12	Caulking expansion and contraction joints. Metal-to-glass seals, and metal-to-metal sealing to separate dissimilar materials. Sealing electrical conduit.
Acrylic	5–12	Pipe joints, glazing masonry, and metal caulking. Special compounds used as liquid gaskets and pipe dope
Polybutene	3–7	General construction-type caulking and glazing. Seal between dissimilar metals
Hardening sealants—rigid types		
Epoxy	7–12	Potting electrical connectors, encapsulating miniature components, coating circuit boards, and cable splicing. Caulking and pipe sealing. Used as a sealer and abrasion resistant coating for concrete
Polyester	6–12	Potting, molding, and encapsulating. Gasket and pipe thread sealants.
Oleo-resin	2–6	Same general uses as nonhardening formulations. Hardening type materials should be used where pressure limits exceed the limitations of the nonhardening formulations.
Asphalt and bituminous	1–4	
Hardening sealants—nonrigid types		
Chemical reaction systems		
Polysulfide—two-part	10–22	Sealing integral fuel tanks and pressure tanks. Sealing faying surfaces and channels. Potting, molding, and sealing of plexiglass. General construction sealing and caulking of metal, wood and masonry joints.
Polysulfide—one-part	20–25	General construction type sealing, caulking, and glazing. Sealing between dissimilar metals. Deck caulking and sealing of refracting mirrors
Urethene—two-part	12–15	Potting and molding of electrical connectors, encapsulation of hydrophones, transducers and circuit boards. Caulking where compatibility with LOX is required
Urethane—one-part	12–15	General construction type sealing, caulking and glazing
Silicone—two-part	45–100	Potting and molding of electrical connectors. Potting firewall connectors and coating umbilical cables. Sealing of heat shields
Silicone—one-part	24–37	General construction type sealing, caulking, and glazing. Potting and molding
Polysulfide—modified	4–6	Sealing of expansion and contraction joints in runways and taxi strips where flame and fuel resistance are required
Bitumen—two-part		
Modified epoxy—two-part	7–12	Potting, molding, encapsulating. Sealing transformers. High voltage splicing, capacitor sealing. General construction caulking
Acrylic—one part	6–12	General construction type sealing, caulking and glazing
Viton—two-part	50–115	For high temperature service where fuel and oil resistance is required. Sealing fuel tanks, channels, and faying surfaces
Solvent release systems		
Neoprene	8–12	Sealing between dissimilar metals. Caulking and general sealing
Hypalon	8–12	Similar in uses to neoprene
Butyls	5–12	Glazing and caulking of metal, glass, and masonry type joints

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Table 2 Representative costs of sealants (4)

2.4 Sealant Failures

Failures of the sealant (fig. 8) depend on:

1) the type of sealant used
2) the installation
3) service conditions

Adhesive failure is the most common in mastic sealants, which is the loss of bond between the sealant and its substrate. Mastic sealants are also subject to cohesive failure within the material, and spalling failure when the adhesive strength of the sealant exceeds the cohesive strength of the substrate. Deformable sealants can fail with a change in sealant shape due to flow. Failure can also occur if there is a large joint movement before the sealant is fully cured. Sealants can fail due to aging and weather exposure. This failure is usually in the form of discolouration, crazing of the sealant surface, or a stiffening at the surface which reduces elastomeric properties. This can be a result of the individual or combined effects of solvent evapouration, ozone attack, migration of plasticizers, U.V. attack, water immersion, etc. (4,5).

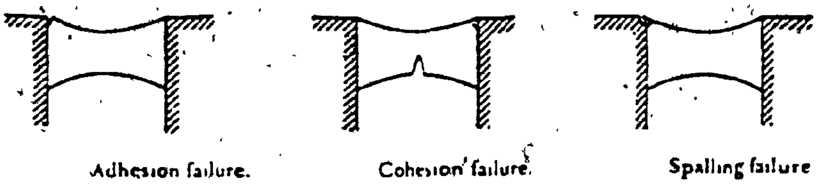


Figure 8 Types of sealant failure (4)

To prevent failure and promote certain performance characteristics, additives such as adhesion promoters, fillers, pigments, plasticizers, thixotropic agents, etc. can be introduced into the polymer. To promote better performance, external accessories used include primers, release agents, and backup materials. Elastomeric sealants usually consist of a base polymer, filler (to control consistency, improve physical properties, and lower cost), plasticizer (to modify hardness and modulus), and a curing agent.

3. SILICONE SEALANTS

Silicone sealants are high quality, high recovery, expensive elastomers which are stable in both cured and uncured states. They are easy to work with and provide a very neat joint. Their outstanding property is high recovery, and after one year of service the sealant may show as much as 98% recovery (4). The drawback of high recovery is the inherent low tear resistance, which is about half the value of the tear resistance of polysulfides or polymercaptans. Once the deformed sealant is cut or punctured, the existing internal stresses will extend a tear rapidly, leading to sealant failure. Silicones require no solvent for workability, thus curing shrinkage is negligible due to almost 100% solids. The advantages and disadvantages of silicone sealants are presented in table 3.

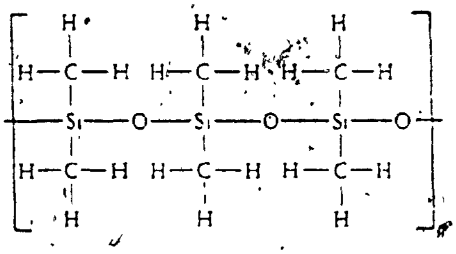


Figure 9 Poly(dimethylsiloxane)

Silicone elastomers consist of a poly(organosiloxane), usually poly(dimethylsiloxane) (fig. 9) due to its lower cost (in comparison with other poly(organosiloxanes)), better chain flexibility, and wide range of attainable properties; additives which provide strength and other desirable properties; and a crosslinking agent. These sealants are classified as room temperature vulcanizing (RTV), because they cure at room temperature and do not require heat to cure, as is the case for heat-cured rubbers. The polymer chain used in heat-cured rubber has a DP (degree of polymerization) of 7000, while the chain length in RTV elastomers has a DP of 1000 (3). The shorter chain length is necessary because lower viscosity is required for RTV applications, but the tensile strength and elongation is lower than heat-cured rubber. Varying the organic groups attached to the silicone atoms can expand the capabilities of silicone elastomers. Substituting methyl with phenyl groups, for example, provides greater low temperature flexibility, which is required for sealants used in the aerospace industry. Fillers can be used to enhance desirable properties, reduce cost, add colour, or to increase resistance to degradation.

One-component silicone sealants are condensation curing, which means that they cure by reacting with moisture present in the atmosphere. A trifunctional organosilane is the crosslinking agent, while a metal soap acts as a catalyst for the condensation reaction. The sealant is stable until it is exposed to the atmosphere, and when exposed it undergoes hydrolysis with atmospheric moisture. This reaction liberates an acid such as acetic acid, which gives off a slight odour. If the sealant is allowed to develop its entire crosslinked network through complete cure, all of its inherent characteristics and properties can be achieved.

Curing is accelerated by hot humid weather, and retarded by cold weather. If cured in cold weather, the cured sealant remains more flexible at low temperatures (although the sealant may harden and become more brittle than organic elastomers). The range of colour tints is virtually unlimited and colour stability is good, thus substrates are not stained. Silicone sealants exhibit excellent resistance to weathering, moisture, U.V. radiation, and are not affected by ozone. After 8000 hours of accelerated weathering, there was no discolouration, appreciable hardening, loss of flexibility, or staining of the substrate (5). This makes them prime candidates for exterior one-stage sealing. The tensile adhesive strength is low in relation to the tensile cohesive strength. This necessitates the use of a primer for working joints, which is one of the primary applications of silicone sealants. Installation requires a very clean joint, thus silicone is a more labour intensive sealant than others.

Advantages	Disadvantages
1. One-component systems	1. Too expensive
2. Available in the widest range of colors	2. Require critical surface preparation and priming (adhesion highly dependent on both)
3. Color stability	3. Poor adhesion to concrete
4. High temperature resistance	4. Inferior elongation relative to polyurethanes and polysulfides
5. Excellent handling characteristics at ambient temperatures of −35 F to +140 F	5. Inferior tensile strength relative to polyurethanes
6. Good UV light and ozone resistance	6. Poor tear resistance
7. Nonsagging on vertical walls	7. Joint width limitations
8. Good adhesion to metal and glass	8. Shelf life too short
9. Exhibit no shrinkage	9. Require contact with atmospheric moisture to cure
10. Become tack-free in a short period of time (fast cure)	10. Cohesion too great for their adhesion
11. Excellent flexibility; permanently flexible	11. Pick up dirt
12. Almost 100% recovery from elongation or compression (very low compression set)	12. Unpleasant odor (release of acetic acid)
13. Good heat and chemical resistance	
14. Excellent durability or long life (about 30 years)	
15. Longer pot life (when mixed) than two-part polysulfides	
16. Good resistance to digging and gouging	
17. Nonstaining to most materials	

Table 3 Advantages and disadvantages of silicone sealants (as reported by consumers) (4)

4. POLYBLENDS

Polyblending is a technique that can be used not only to improve polymer properties, but also to lower cost through the use of additives. Polyblends can be defined as physical mixtures of structurally different or similar polymers. The two polymers adhere to each other (to various degrees) through the action of secondary bond forces, with no covalent bonding between them (21). Most polymers are incompatible with each other, and when used in polyblending exhibit phase separation (the polymers co-exist in separate phases). The presence of these distinct phases gives the inherent properties of the polyblend. The level to which the two or more polymers adhere to each other can determine the mechanical properties. Polymers that adhere well to each other can exhibit greater mechanical properties than those that don't.

Polymers can also be compatible with each other (miscible). This occurs when polyblending results in a mixture which exhibits single phase behaviour. Unlike the incompatible polyblend, which has two glass transition temperatures due to phase separation, a miscible polyblend has only one glass transition temperature (Tg) (the temperature at which the polymer behaviour changes from glassy to rubbery) located somewhere between the glass transition temperatures of the constituent polymers (18).

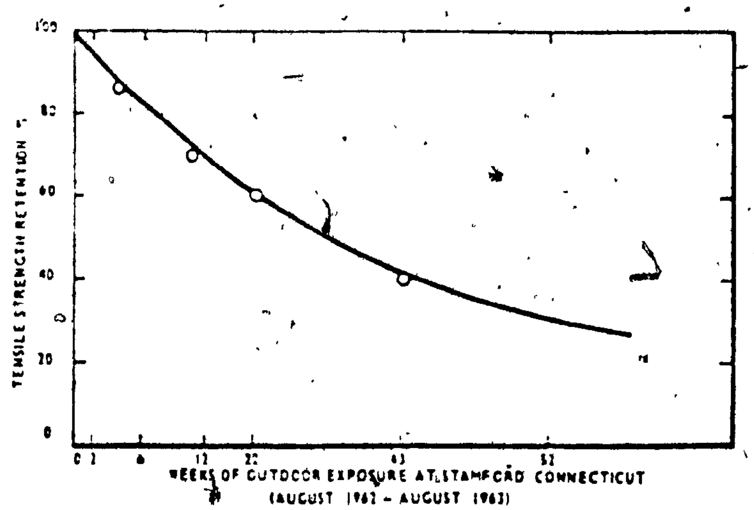
Determination of glass transition temperatures is a technique often used to find out whether a polyblend is miscible (although this method may be unreliable depending upon the nature of the polymers used). The term miscible is best defined as a polyblend that achieves a level of molecular mixing adequate to yield macroscopic properties expected of a single phase material (24). More precise techniques for determining the level of mixing are electron microscopy and equation-of-state thermodynamics.

A polyblend does not necessarily have to achieve miscibility in order to have useful properties. Many desirable properties are often a result of two-phase blends. The type of properties that can be obtained by polyblending are dependent on how well the two polymers adhere to each other, and the proportion of each polymer in the polyblend. Thus the morphology of the polyblend system is important in determining its properties. The polymer that constitutes the highest proportion in the polyblend is referred to as the continuous phase or the matrix, while the polymer that is present in smaller proportions is referred to as the dispersed phase, which exists as discrete domains. The ultimate mechanical properties, such as strength and toughness, of a phase-separated polyblend are determined by the interfacial adhesion between the phases. Poor adhesion can lead to flaws located at the phase interfaces, which can result in much lower ultimate properties than expected from the averaged properties of the two phases (24). Polymers that have similar dispersive and polar forces will exhibit better interfacial adhesion than those widely differing in molecular forces.

The extent to which phase separation occurs depends on the compositional dissimilarity, molecular weight, crystallizability (23), functional groups present and their polarity. These characteristics also determine the size and shape of the domains within the continuous phase. When the molecular weight of the dispersed phase is higher than that of the continuous phase, the domains have the shape of coarse droplets. When the molecular weight of the dispersed phase is lower, the domains are shaped like layers or ribbons (21). The proportion by weight of the discrete domains within the matrix also influences the polyblend morphology, and hence its properties. At low proportions, the domains are spherically shaped, and become rod-like at slightly higher proportions. At nearly equivalent proportions, co-continuous phases occur with lamellar domain shapes (23). Electron microscopy can be used to determine these different morphologies.

5. NATURAL AND ACCELERATED WEATHERING EXPOSURE

Weathering exposures were developed as a means of determining how a given material will perform in its service conditions (outdoor exposure). One major problem in analyzing the weathering of any polymer is the fact that weather is transient, and its components can act either in succession or simultaneously. The complexity of each situation is unique onto itself, thus any prior testing will not be entirely representative of the actual on-site conditions. The study of weathering performance can be done by actual outdoor exposure (natural weathering), which is time consuming, or by exposure in an accelerated weathering machine, whose results must be correlated with natural weathering (fig. 10). Both methods have their advantages and disadvantages.



Correlation of Weather-Ometer and outdoor exposure data on polystyrene using the exposure parameter method. (—) outdoor results predicted from Weather-Ometer tests; (O) observed outdoor results. The abscissa has been normalized for intervals of equal solar energy [illegible] during the test period (Aug 1962–Aug 1963 at Stamford, Conn.)

Figure 10 Correlation of A.W. and N.W. data (11).

The advantages of natural weathering are as follows:

1) the spectral distribution of normal sunlight is accounted for
2) factors other than moisture, temperature, and sunlight are represented
3) correlation of test results to actual outdoor exposure is not necessary
4) weather element values tend to average out over long exposures
5) the effects of non-continuous sunlight are accounted for
6) outdoor conditions may not provide the minimum required energy to initiate some reactions (as indicated by accelerated exposures)

The advantages of accelerated weathering methods are:

1) test results are gathered at a much faster rate
2) exposure conditions can be precisely controlled and reproduced
3) if the polymer deteriorates very quickly under accelerated conditions, its potential for outdoor use is very low, and no additional time is wasted for more tests
4) the spectral distribution, temperature, and moisture can be adjusted, within limits, to represent a large range of climates
5) the additional maintenance required for outdoor equipment is eliminated

Regardless of which of the two test methods is preferred, one important fact must be realized. Outdoor exposure tests may be no more representative of actual exposure conditions, for a given polymer application, than are tests carried out in a controlled laboratory environment. Whether the results are from outdoor exposure or accelerated weathering, the relative data useful for the on-site conditions must still be extrapolated from them.

Weather factors which tend to contribute to the degradation of polymers are:

1) the ultra-violet portion of solar radiation
2) temperature
3) water (solid, liquid, or vapour) in the presence of acids or alkalis

4) oxygen

5) ozone

6) micro-organisms

7) industrial gases

8) mechanical loadings which arise from exposure to wind and snow loadings

9) severity of cycling (fatigue), which can be caused by wet/dry, freeze/thaw, or compression/tension cycles.

Various combinations of these factors can initiate complex processes of degradation. These processes can include:

1) volatilization of plasticizers and solvents

2) chemical decomposition of additives and fillers

3) breakage of the polymer backbone

4) splitting off of side groups

5) reactions among new groups formed

6) breakage or formation of associated bonds

7) dispersion and/or breakage of crystallites

6. SILICONE SEALANT PERFORMANCE AND MECHANICAL TESTING

The performance of silicone sealants in service conditions can be influenced by stress, strain, time (rate of extension and/or compression), temperature, humidity, oxygen, light, the type and condition of substrate, and various other factors. Mechanical properties are usually defined by stress and strain, and are influenced by time and temperature. Silicone sealants posess the characteristic of having their mechanical properties independent of temperature within the building service condition temperature range (-30oC to +50oC). This reduces the parameters defining mechanical properties to stress, strain, and time.

Since tensile extension is the type of loading most likely to produce failure in joint sealants in service, tensile testing is primarily used to obtain information about mechanical properties. A tensile test consists of pulling a specimen (a sealant bead between two pieces of substrate) in a direction perpendicular to the attached surfaces, until adhesive or cohesive failure occurs. Tensile elongation causes parallel surfaces of the sealant bead to become concave and builds up stresses at these surfaces. If stresses build up to a critical level, stress concentration at the ends (where stress is greatest) can initiate failure of the sealant bead.

As the temperature of the sealant changes, there is a corresponding change in its adhesive properties. There is a transition from cohesive to adhesive failure with decreasing temperature and increasing loads. Silicone sealants also exhibit higher extensibility with decreasing temperature (fig. 11), which acts as a safety factor because joints are at their widest at low temperatures. This holds true only to

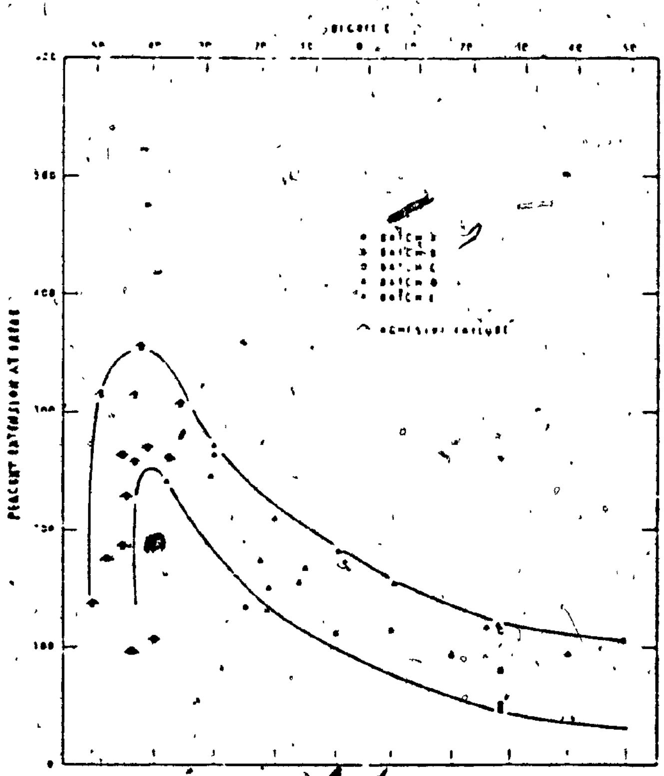


Figure 11 Extension at break as a function of temperature, primed aluminum subsrate (12)

a temperature of -40oC, below which extensibility is reduced considerably due to crystallization which changes material properties. In an unstrained state the crystallization temperature of silicone sealants is much lower than in a strained state. Although there is increasing extensibility with lower temperatures, as the elongation and load increase there is a hardening of the sealant.

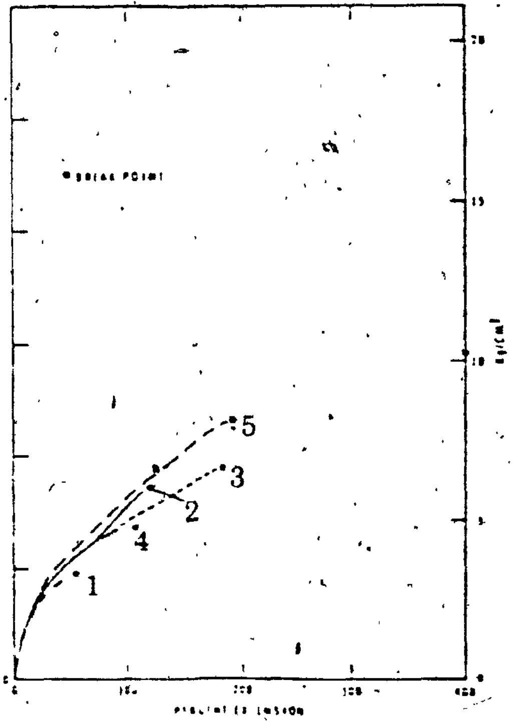


Figure 12 Tensile tests at various rates of extension, 22oC. Curve 1-0,012 mm/min; curve 2-1,3 mm/min; curve 3-5,08 mm/min; curve 4-13,0 mm/min; curve 5-130 mm/min (13)

6.1 Influence of Rate of Extension and Temperature

The rate of extension used in tensile testing affects the performance results of the sealant. The strength of silicone sealants decreases with decreasing rate of extension (fig. 12) (13), but it takes more time for failure to occur with slow movement than with fast movement. This is due to the fact that there is a more frequent reversal of load with fast movement, leading to fatigue failure much sooner. Failure occurs at increasingly longer times with decreasing strain rates.

Tensile curves can change not only with rate of extension but with temperature as well. Using a constant rate of elongation, tensile test curves can vary considerably with varying temperatures. When elongations are below 50% and loads are less than 0,34 N/mm^2, there is negligible difference in results at varying temperatures. If testing is done at room temperature according to standards, priming of the substrate has no influence on obtained results. Tensile curves would be identical at varying temperatures if silicone sealants did not crystallize at low temperatures (7).

Tensile test results can be converted to stress/strain curves using the specimen geometry. The area under the curve is a measure of sealant toughness. The greater the area, the higher the fracture resistance of the sealant.

7. RESEARCH PROGRAM

7.1 Materials and Specimen Preparation

A silicone sealant (PDS) was chosen to be used as the matrix for polyblending. Dow Corning Silicone Sealant 697 was selected because of its excellent properties and commercial availability. Seven different types of specimens were prepared for testing. Silicone sealant (unmodified), to be used as a basis of comparison, was the first set of specimens prepared. The following groups of polymers, in the powder state, were polyblended with the PDS:

1) vinylic polymers - poly(vinyl chloride) (PVC) (Geon Resin, B.F. Goodrich)
   - vinyl chloride - vinyl acetate copolymer (VC/VAc), which consists of 85% vinyl chloride and 15% vinyl acetate by weight (Hostaflex M131, Hoechst)

2) nitrilic elastomers - unmodified: acrylonitrile (ACN) - butadiene copolymer with medium (33%) ACN content (K1402) (Krynac 1402 H83, Polysar)

- unmodified: acrylonitrile - butadiene copolymer with high (41%) ACN content (K1411) (Krynac 1411, Polysar)

- modified: medium carboxylated (3%) acrylonitrile - butadiene copolymer with high (40%) ACN content (K211) (Krynac 211, Polysar)

3) natural polymers - Kraft lignin, a commercial mixture extracted from a mixture of hardwoods (maple, beech, elm), precipitated from the black liquor precipitated with sulfuric acid, and pH adjusted to a pH level between 6 and 7 with carbon dioxide (Domtar). Lignin is a by-product of the pulp and paper industry and is obtained in high quantities (and relatively low cost) in Canada. In 1980, 18,2 x 10^6 kg. of Kraft lignin were generated in the pulping process of the pulp and paper industry in the United States. Only 31,8 x 10^3 kg. of this lignin was consumed worldwide for various industrial uses.

The 1980 price of Kraft lignin was 0,30-0,50$ per kg (19). As the 1985 price of commercial PDS is approximately 17$ per kg, using lignin in polyblends with PDS would most definitely lower its cost.

Each of the polyblends were prepared in the following proportions of dispersed phases by weight: 5%, 10%, and 15%. The process of polyblending was executed by means of mechanical mixing only. Each polyblend was mixed 5-10 minutes to ensure a reasonable degree of dispersion through mechanical shearing.

The sealant beads were cast between each of the following three substrates: wood, aluminum, and Portland cement mortar. These substrates are representative of the majority of building components which require sealants in their joints. The wood (California redwood) substrates were cut lengthwise along the grain, thus subjecting the sealant only to the lengthwise grain. Aluminum substrates were prepared according to ASTM C719-79. This method consists of cleaning the substrate, prior to use, with a methyl ethyl ketone or similar solvent, followed by a thorough cleaning with a detergent solution and a final rinse with distilled or de-ionized water, then air dried. Mortar substrates were prepared according to CGSB CAN2-19.0-M77 Method 12.1-1978, which consists of the following: one part high earlystrength Portland cement (CSA A5) with two parts by mass of clean, well graded fine aggregate (ASTM C33). Sufficient water is used to produce a flow of 120 ± 5% when tested in accordance with the test for consistency of cement mortar (ASTM C109). After curing one day in moist air (95-100% R.H.) and 6 days in water at 23 ± 2°C, one face of each substrate bar is surfaced by wet grinding on an iron lap using a No. 60 silicone carbide or aluminum oxide abrasive grain or equivalent grinding surface. Bars are oven-dried to a constant mass at 104-110°C, cooled to 23 ± 2°C and 50% R.H. Any bar that has a surface cavity greater than 2 mm is rejected.

A total of at least fifteen specimens of every type of polyblend were cast with each different substrate.

To ensure proper correlation between test results and actual on-site performance of the sealant bead, the size and shape of specimens should duplicate service conditions as closely as possible. Since building sealants are subjected primarily to tension and compression, and most failures are tensile, a butt joint was chosen to be best representative of a working building joint. This type of joint can provide cohesive or adhesive failure of the sealant, as occurs in service conditions.

The sizes of the sealant bead and its substrate were the ones used in many standards and testing literature (7, 12-15). The sealant bead was cast as 12,5/12,5/50 mm. The butt joint was formed along the 12,5/50 mm opposite sides of the sealant bead. Two sizes of substrate were used (fig. 13): sealant beads of PDS, PDS-PVC, and PDS-VC/VAc were cast between substrates of 25/25/75 mm, while the other sealant beads were cast between substrates of 12,5/25/75 mm. In all cases the size of the sealant bead remained constant.

The size of the sealant bead was controlled with wooden spacers between the substrate pieces. Spacers were covered with either cellophane tape or waxed paper to ensure that no adhesion would occur between the sealant and the spacers. Substrate pieces and spacers were positioned firmly within a wooden mould which maintained them at right angles with the sealant bead (fig. 14). The mastic silicone sealant and polyblend mixtures were then placed between the substrates. A putty knife was used to work out some of the air bubbles, and the sealant bead was finished flush with the substrate. Specimens were cured for at least 7 days, as recommended by the PDS manufacturer, at room temperature (24°C and 35% RH) before they were removed from the mould.

Note that 10% and 15% PVC-PDS polyblend sealant specimens with aluminum substrates were not prepared due to the fact that they exhibit virtually no adhesive strength when slightly flexed by hand (as determined with some preliminary samples).

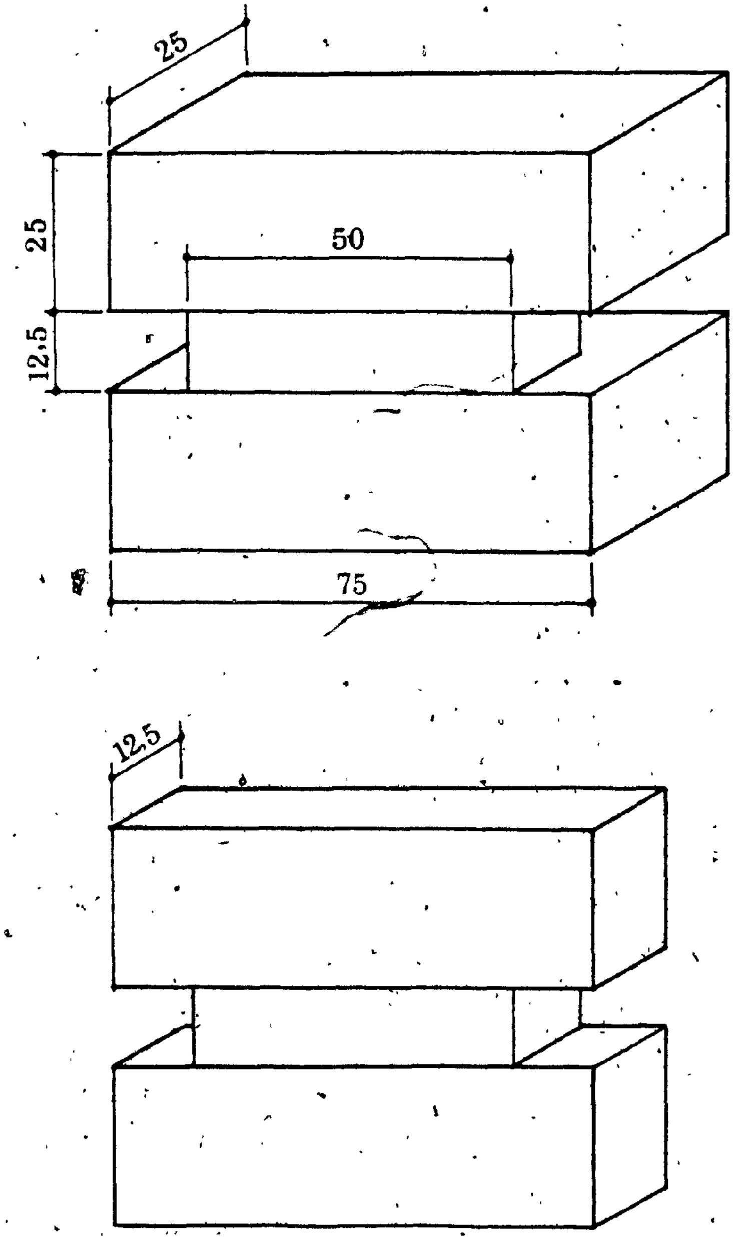


Figure 13 Specimen sizes (all values in mm)

Figure 14 Substrate setup within mould prior to casting of sealant specimens

7.1.1 Elevated Temperature Mixing

Investigations were conducted on the effect of elevated PDS temperatures prior to mixing of the polyblend. Mastic PDS was heated to +30°C for 30 minutes, then mixed with the additives. Mixing proved to be more difficult than with the unheated specimens, as the surface of the PDS had started to cure. Heating did not give any mechanical advantage over unheated specimens, thus only unheated specimens were prepared for exposure tests.

7.1.2 Grading of Particle Sizes

Investigations were also conducted on the effect of using only fine, graded particle sizes of the additives in the polyblend. Grading of particles was done as follows: 1) vinyl polymers through 0,15 and 0,30 mm sieves; 2) K1402 through 0,60 and 1,18 mm sieves; 3) K1411 through 0,15 and 0,30 mm sieves; 4) K211 through 0,30 and 0,60 mm sieves. Grading of lignin was not performed due to the fact that lignin particles were all finer than the smallest sieve available. Graded particles were then mixed with the unheated PDS. This improved the dispersion of the particles within the polyblend, but mechanical properties were not enhanced, therefore only ungraded particles (as provided by the manufacturer) were used to prepare specimens.

7.2 Procedure for Weathering Exposure

The fifteen fully cured specimens (each polyblend type with every different substrate) were sub-divided equally into groups in order that they could be subjected to the three following conditions:

1) laboratory conditions (referred to as control specimens) at 24^{o}C and approximately (~) 35% R.H.

2) accelerated weathering (A.W.) between -30ºC and +30ºC (~50% R.H.) at 4 cycles per day for a total of 400 cycles. Specimens were set vertically on a table in the chamber. After 36 cycles, the specimens were subjected to overhead ultra-violet radiation lamps (wavelength of 254 nm) for 12 continuous hours daily.

3) natural weathering (N.W.) exposure in a relatively polluted location of downtown Montreal for 40 weeks between July 3, 1984 and April 9, 1985 (figs. 15 & 16). Specimens were placed vertically in wooden racks which kept rows separated (fig.17). The highest temperature, occurring on August 4th, was +32,4ºC, while the lowest temperature of -26ºC occurred on February 7th.

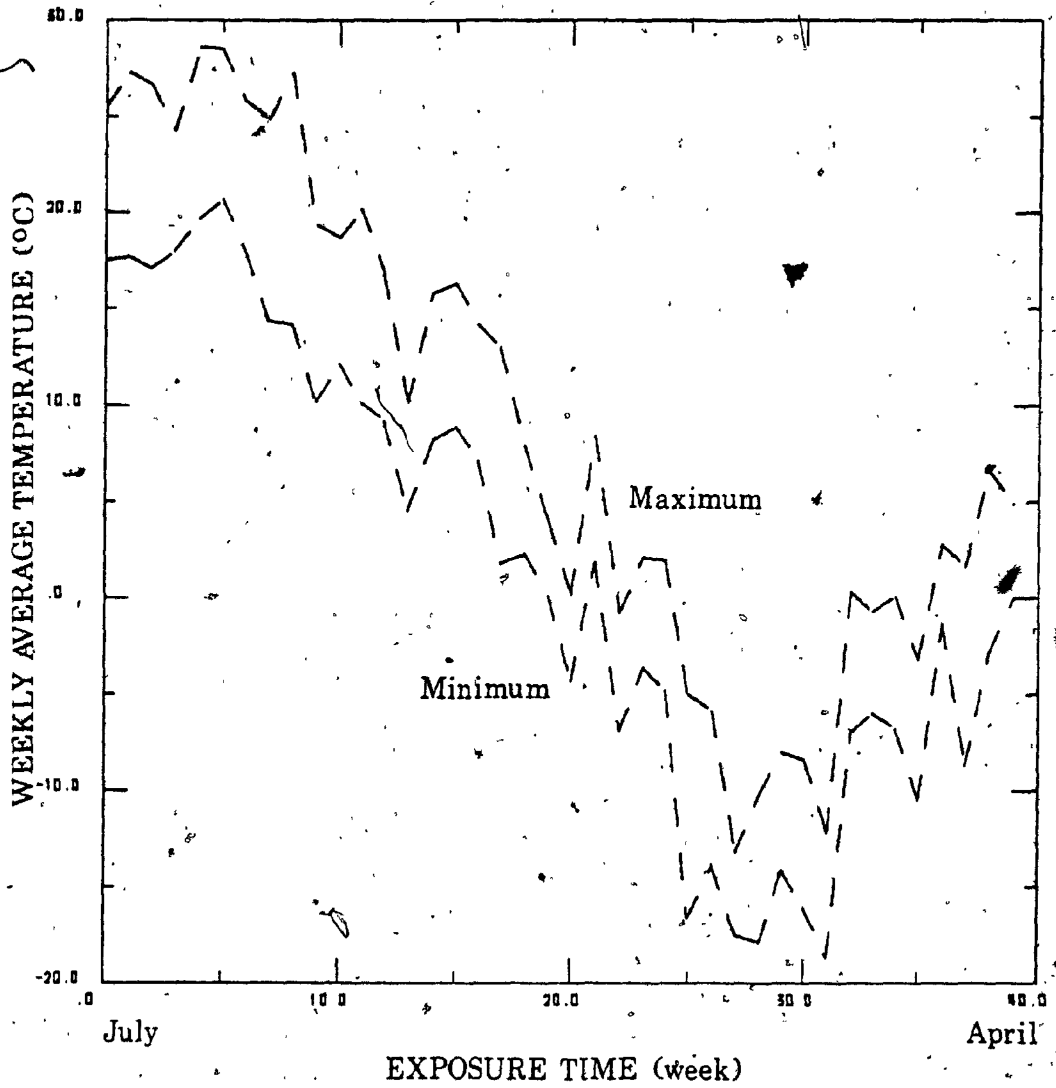


Figure 15 N.W. exposure conditions: average weekly maximum and minimum temperatures

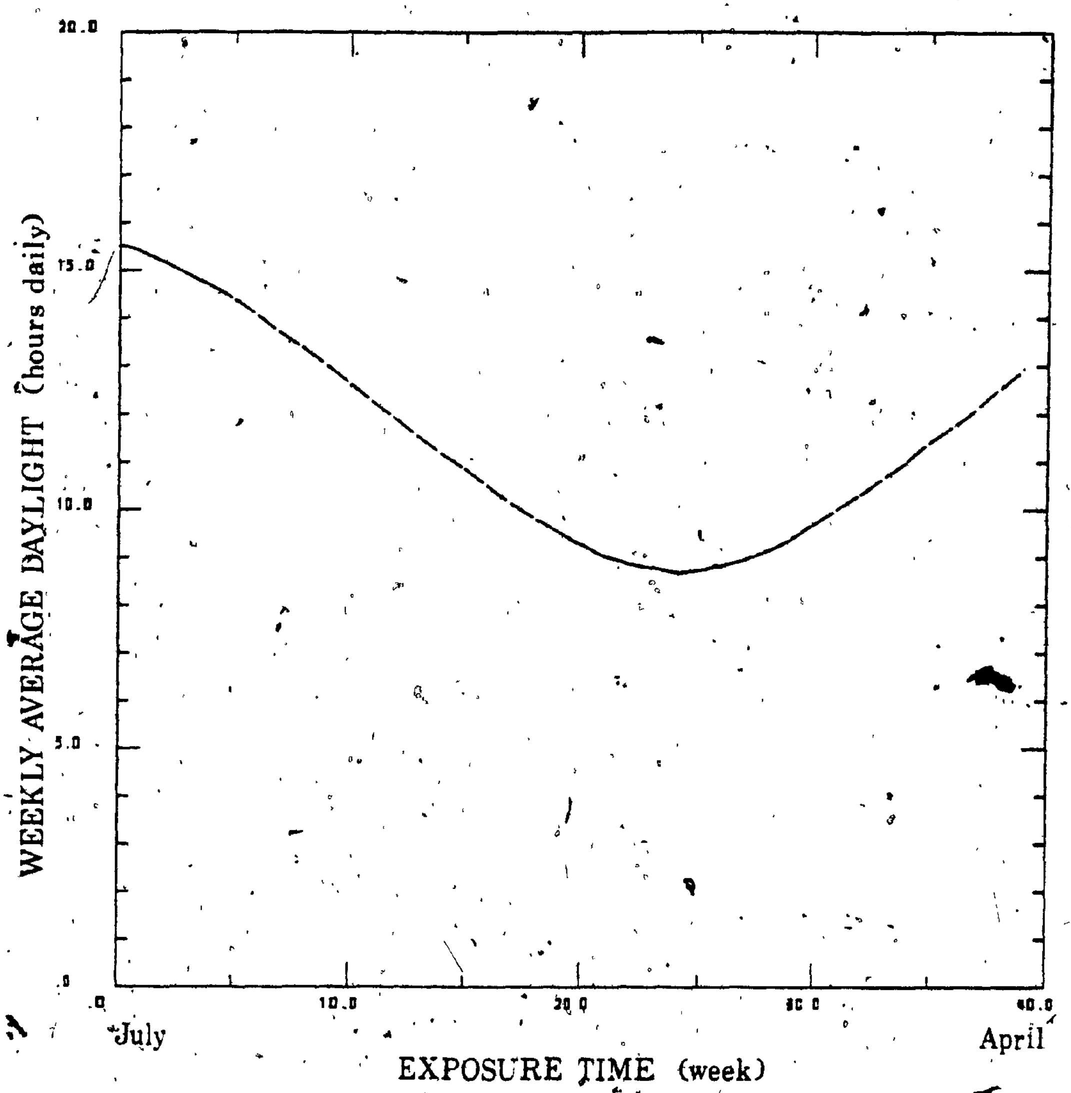


Figure 16 N.W. exposure conditions: average weekly hours of daylight

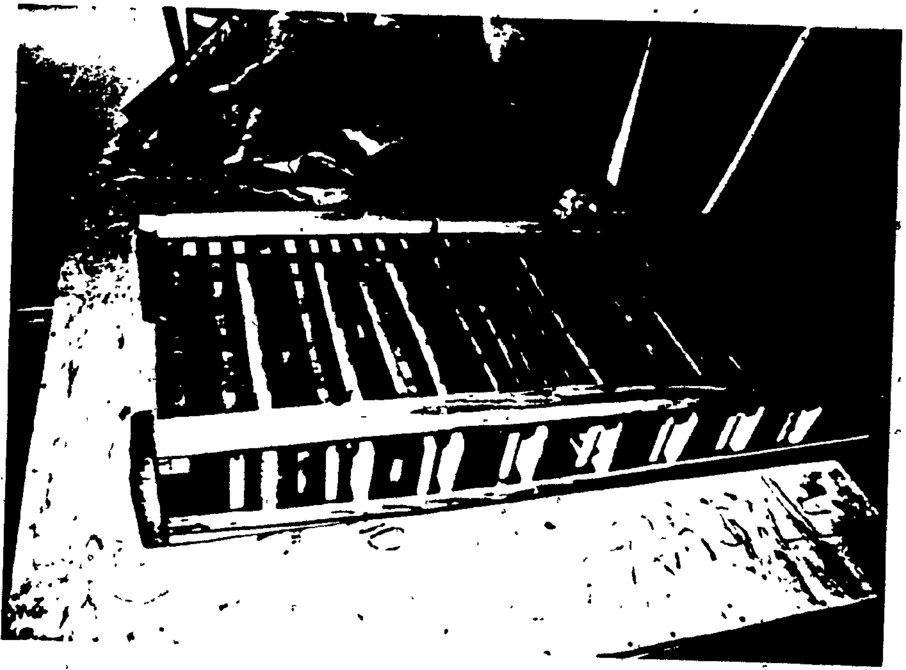


Figure 17. Racks used to hold specimens during N.W.

7.3 Specimen Testing

As a means of investigating the durability and associated properties of silicone sealants and its polyblends, the following tests were performed on the specimens:

1) mechanical - tensile testing to determine stress/strain behaviour of all specimens prepared.

- dynamic mechanical analysis (DMA) was attempted, but due to the highly flexible nature of PDS which made it impossible to secure a sample properly within the DMA, significant results could not be obtained.

2) thermal - differential scanning calorimetry (DSC) to determine glass transition temperatures.

3) spectroscopy - infrared spectroscopy using a KBr disk for mounting samples. This technique helps determine the possible interaction between side groups of the polymer chain and the additives used in the polyblends.

4) microscopy - scanning electron microscopy (SEM) was used to study the influence of morphology on the performance of the polyblends.

- energy dispersive x-ray analysis (EDXA) was performed in conjunction with SEM to determine the elements present within the polyblends and their possible interactions.

7.3.1 Tensile Testing

After completion of exposures, the specimens underwent tensile testing to determine changes in mechanical properties. All tensile tests were performed on the Instron 1125 universal testing machine at the Centre for Building Studies, using a crosshead with a maximum cell load of 500 kg. Tensile tests were performed under laboratory conditions of 24°C and 35% RH. Since building joints in service expose the sealant bead to a very slow rate of movement, the crosshead speed was chosen as a relatively slow 2 mm/min.

Readings from the tensile tests are transferred by the Instron machine onto a chart which moves at a rate of 20mm/min. The chart full scale was set at 50 kg. Testing was conducted until there was a visible adhesive or cohesive failure, or until the results recorded on the chart indicated a sharp decrease in load. Recorded results can be read off the chart to determine extension and load at that extension, which can then be converted to stress/strain data. The apparent stress is calculated by dividing the load by the perpendicular cross-sectional area of the sealant bead. In this case, the area is 12,5x50,0 mm = 625 mm^2. The strain is the recorded extension divided by the parallel (to the tension) length of the sealant bead (12,5 mm). The stress/strain data can then be plotted to obtain a characteristic stress/strain curve which gives an indication of engineering properties that can be used for comparison.

7.3.2 Thermal Testing

The glass transition temperatures of the PDS and polyblend sealants were determined by thermal analysis using a differential scanning calorimeter (DSC). The temperature range used for testing was -170°C to +50°C. This temperature range was chosen because pure poly(dimethylsiloxane), the building block of the silicone sealant polymer chain, exhibits a glass transition temperature of -123°C, and a crystalline melting temperature of -40°C (22). The rate of heating used was 20°C/min, which is a rate that was used by Ranganathan (26) to determine the glass transition temperature of room temperature vulcanizing silicones. The sample size was kept consistently between 10-14 mg.

The technique for testing the samples was as follows. A thin slice was cut from the center of a cured sealant bead, then was weighed and placed in a metal sample pan. This pan, along with an empty reference pan, was placed in the DSC sample holder. The sample holder was cooled with liquid nitrogen to below -170°C. Then both pans were heated at a constant rate in an inert atmosphere of nitrogen, and the difference in energy required to heat the two pans was recorded and analyzed. At a transition point, the sample requires either more or less ener[illegible] than the reference pan. An endothermic change occurs when a melting point is reached, thus the sample would require more energy than reference. An exothermic change requires less energy than reference as is the case when a sample is curing and energy is released. Thus DSC is a technique that measures the quantity of energy evolved or absorbed by a sample as its temperature is changed at a constant rate (28).

After testing all the sealant specimens, the glass transition temperature was observed to be between -115°C and -108°C, and the crystalline melting point was between -30°C and -35°C. These results agree (taking into account the fact that the silicone sealant used is not pure PDS but also consists of additives, fillers, etc.) with the corresponding temperatures for pure poly(dimethylsiloxane) mentioned above. DSC testing is also used to determine miscibility (presence or absence of a second glass transition temperature).

7.3.3 Infrared Spectroscopy

The KBr disc method was used to prepare specimens for infrared examination. This method consisted of grinding 15mg of the polymer with a few mg of coarse KBr until the polymer was very finely divided. This powder was then well mixed with 500mg of infrared grade KBr and 250mg of this mixture was loaded into a 13 mm die. The die was placed in a hydraulic press and a $7,2 \times 10^4$ N load was applied for 2 minutes. The KBr disc was then placed in a Beckman IR 4240 spectrophotometer and subsequently examined.

7.3.4 Microscopic Testing

Electron microscopy is one of the methods used in determining the morphology of polyblends. Extra samples of each of the polyblends were prepared so that their morphology could be studied under a scanning electron microscope (SEM). This technique consists of creating a fracture surface of the sealant bead. This is done by freezing the sealant bead in liquid nitrogen, then mechanically fracturing the sealant in the center of the bead. This creates a rough surface whose morphology can then be studied. The samples were then sputter coated with gold to prevent electrical charging during SEM analysis. Energy dispersive x-ray analysis was also conducted during SEM testing. This technique analyzes x-rays which are given off from the samples during SEM testing. As each chemical element emits x-rays of different wavelengths, a computer software package analyses wavelengths and identifies the associated elements. Elements that have an atomic mass number of less than four cannot be detected by the apparatus used, since the low energy x-rays are absorbed by the protective window surrounding the Si(Li) drift detector. SEM tests were conducted at the Division of Building Research at the National Research Council of Canada in Ottawa.

8. RESULTS: ANALYSIS AND DISCUSSION (2)

In all cases, the ultimate tensile strength of the specimens varied with the substrate used. The substrate which exhibited the best performance also varied with the material used for the sealant bead. Values used to plot the stress-strain curves presented in the figures are averages of five successful determinations. Note that curves of certain proportions of the polymers that are not presented in the figures indicate that specimens failed during exposure (prior to testing).

8.1 Accelerated Weathering

After 263 cycles in the accelerated weathering chamber, the condition of the specimens was checked. All of the sealant beads were intact, but several of the mortar substrates had either partially or completely disintegrated (fig. 18). The disintegrated substrates had broken up into small granules, having lost virtually all cohesion. The specimens that were positioned on the edges of the table (inside the chamber) were the worst cases (fig. 19), suggesting that the condensation, cycling, and U.V. light had active roles in the disintegration of the mortar substrates. Mortar specimens positioned in the center of the table, surrounded on all sides by other specimens, were

Figure 18 Disintegration of mortar substrates with A.W.

barely or not at all affected. Specimens with mortar substrates that did not exhibit cohesive integrity (not solid when hand inspected) were removed from the chamber. One sample of each sealant mixture and each substrate was set aside for testing. The remaining specimens were again subjected to the accelerated weathering cycles. Disintegrated specimens were discarded, and new sealant beads on mortar substrates were prepared. These specimens were placed in the weathering chamber only after the initial specimens were

Figure 19 Position of disintegrated mortar specimens on table within A.W. chamber

weathered for 400 cycles and removed.

The specimens set aside (after 263 cycles) were conditioned at room temperature for at least 48 hours. They were then subjected to tensile testing at a crosshead speed of 2 mm/min. The test results of the pure silicone specimens were then compared to each of the polyblend specimens.

After 400 cycles in the weathering chamber, specimens were removed and conditioned at room temperature for at least 48 hours. They were then subjected to tensile testing as outlined in the preceeding paragraph, and the results confirmed those obtained during testing of the 263 cycle specimens.

8.2 Morphology

The morphology of PDS and its blends with vinyl polymers is illustrated in SEM photomicrographs of Figures 20 to 24. SEM examination indicates that specimens of PDS-based blends containing 5, 10, and 15% of either vinyl polymer, prepared at 24°C, are heterogeneous. The blends consist of two distinct solid phases, the continuous phase (matrix) of PDS, and the well-distributed particles of vinyl polymer. All blends contained a relatively large number of voids, resulting from occluded air bubbles formed during mixing (figs. 21 and 23). SEM observations also indicate that the adhesion between particles is good, as evidenced by the absence of gaps in the boundary regions of the two phases (figs. 22 and 24).

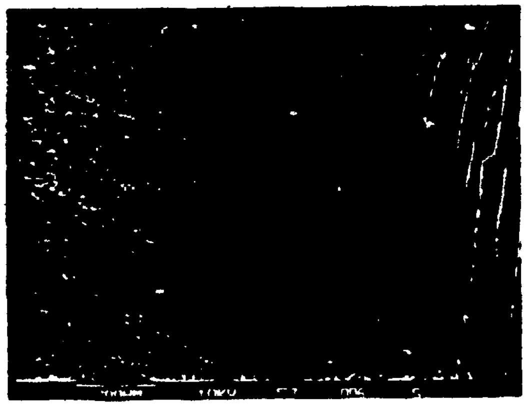


Figure 20 SEM photomicrograph of cross-section of PDS

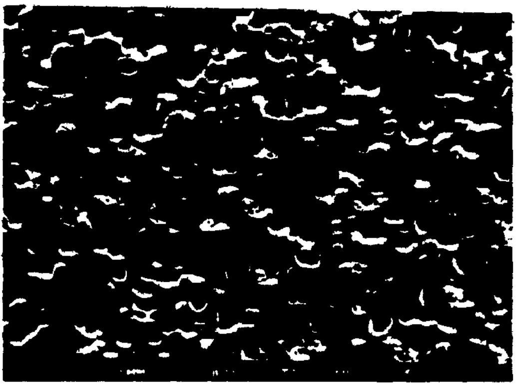


Figure 21 PDS-PVC blend (15%) photomicrograph 10x magnification

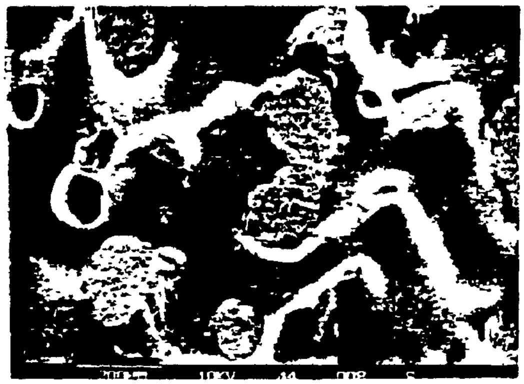


Figure 22 PDS-PVC blend (15%) photomicrograph 50x magnification

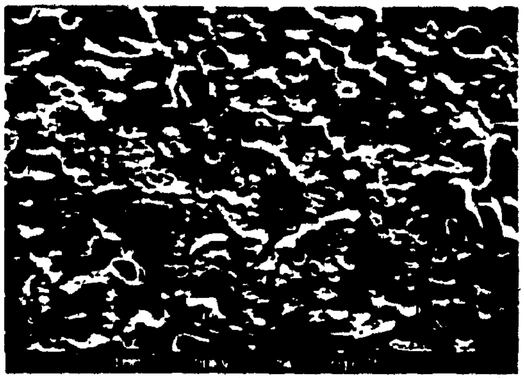


Figure 23 PDS-VC/VAc blend (15%) photomicrograph 10x magnification

Figure 24 PDS-VC/VAc blend (15%) photomicrograph 50x magnification

SEM analysis of weathered specimens indicated that voids (air bubbles) are reduced or eliminated, which may explain why the weathered specimens have higher toughness than the control specimens. This phenomenon could be due to the compacting of the specimen during expansion and contraction.

Measurements by energy dispersive X-ray analysis (EDXA) have indicated that PDS material is present in either the PVC or VC/VAc particles of all blends, as evidenced by the Si count (figs. 28-29). Conversely, no PVC or VC/VAc was detected by EDXA in the PDS matrix of the blends (figs. 30-31).

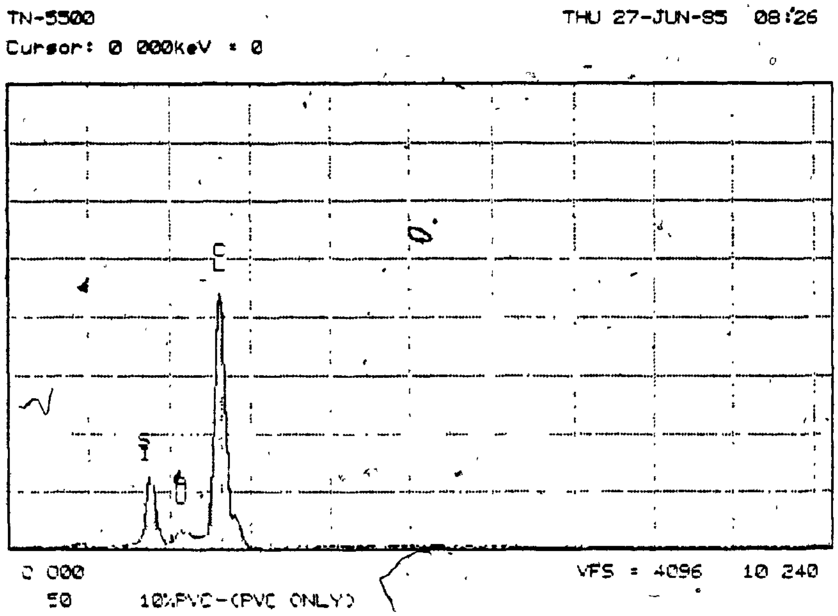


Figure 25. EDXA spectrum of dispersed phase (PDS-PVC)

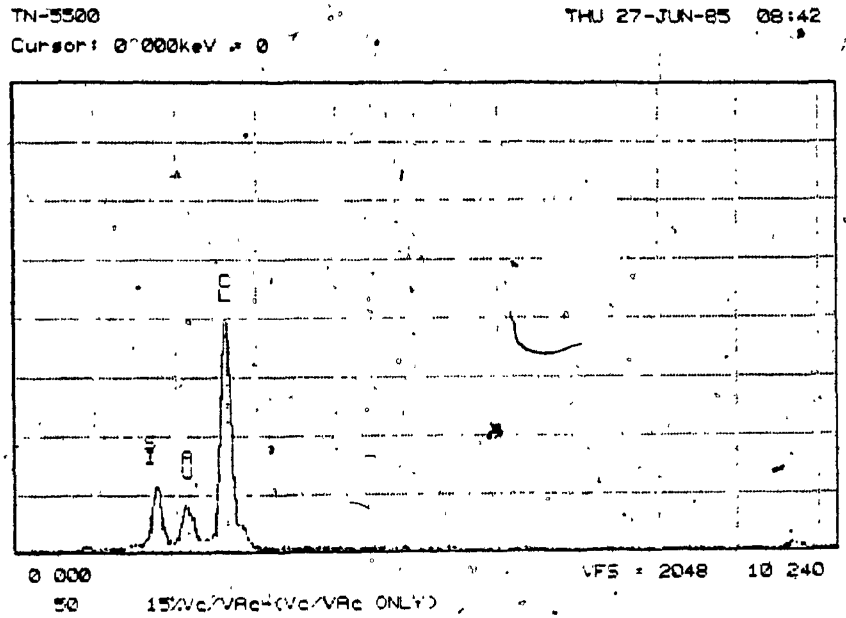


Figure 26 EDXA spectrum of dispersed phase (PDS-VC/VAc)

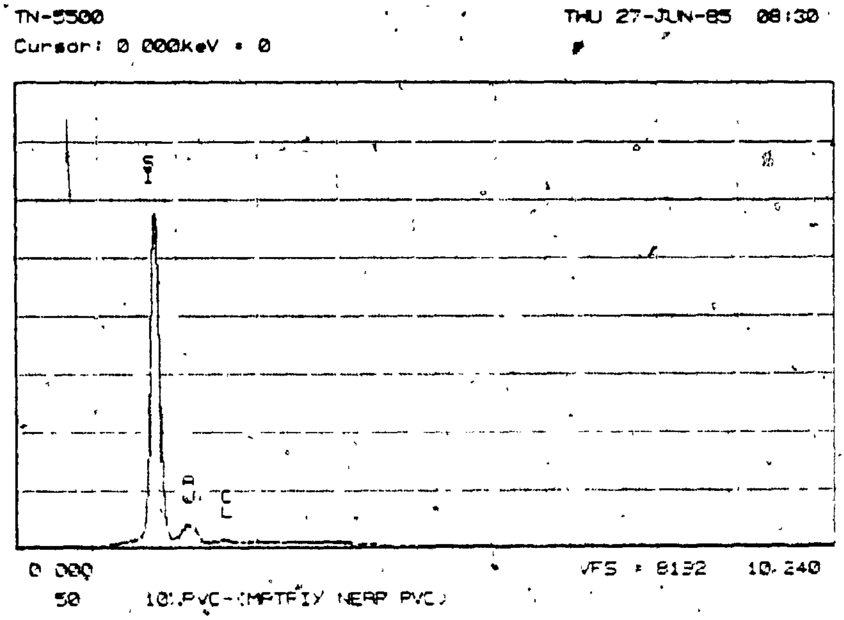


Figure 27 EDXA spectrum of matrix (PDS-PVC)

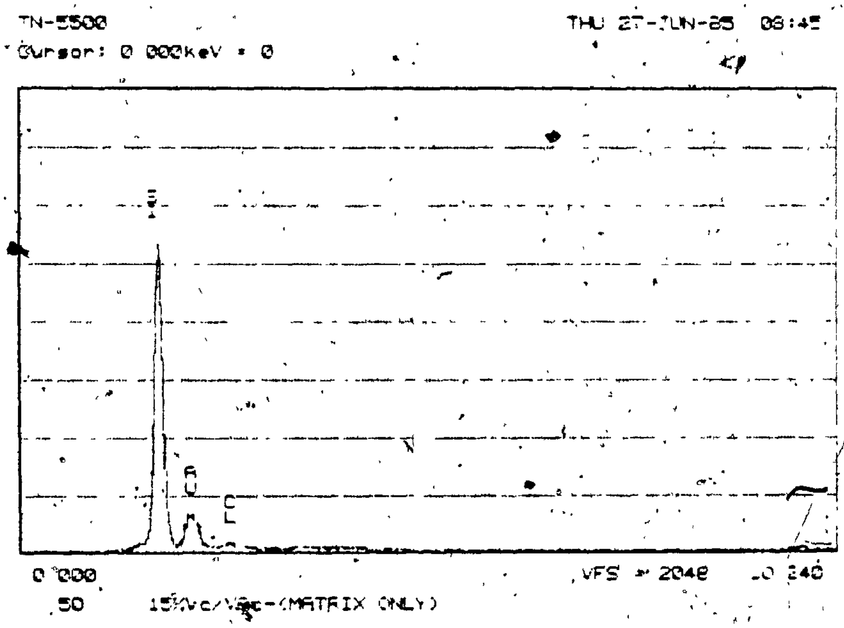


Figure 28 EDXA spectrum of matrix (PDS-VC/VAc)

8.3 Infrared Spectroscopy

The spectra of siloxane polymers, whether oils, rubbers, or rigid resins, have as their most prominent features broad bands of extremely high intensity in the 1100-1000 cm-1 region, often with two or more distinct maxima which arise from the Si-O-Si group.

Methyl groups directly attached to silicone also have a highly characteristic band. There are two characteristic bands for dimethylsiloxane: one sharp single peak at 1258 cm-1, and two peaks at 800-806 cm-1 and 854 cm-1 (this peak is smaller than the one at 800 cm-1).

All of these bands can be observed (±5 cm-1) in the spectra of polysiloxane (PDS) alone (fig. 29), or in its polyblends with PVC (fig. 30), VC/VAc (fig.31), K211 (fig. 32), and lignin (fig. 33). With PDS, PDS-PVC, or PDS-K211, the weaker band characteristic for the $Si(CH_3)_2$ group at 854 cm-1 is difficult to distinguish. This could be explained by an interaction between the CH_3 groups and a functional group of the additive polymers. Mixtures with 15% VC/VAc and 10% Lignin did not display this modification, even though all the spectra were run under the same conditions and the discs were prepared with the same quantities of material.

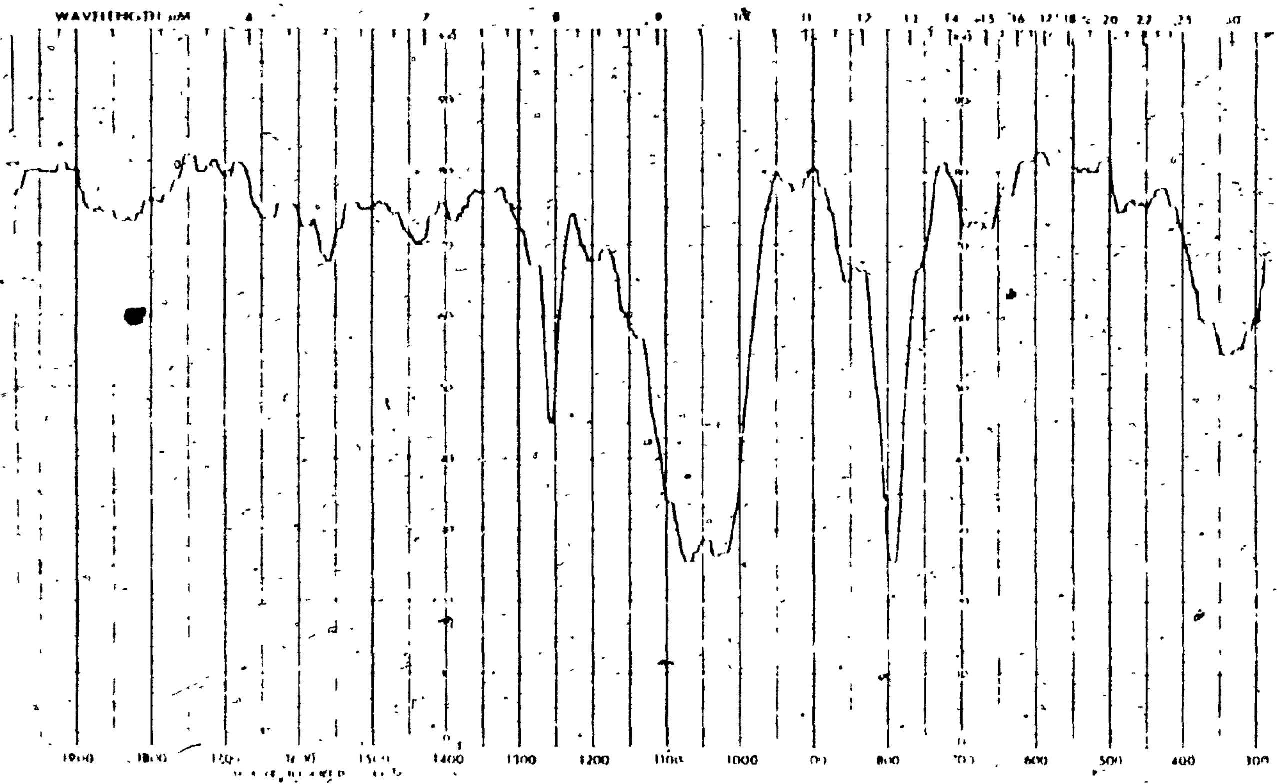


Figure 29. Infrared spectrum of PDS

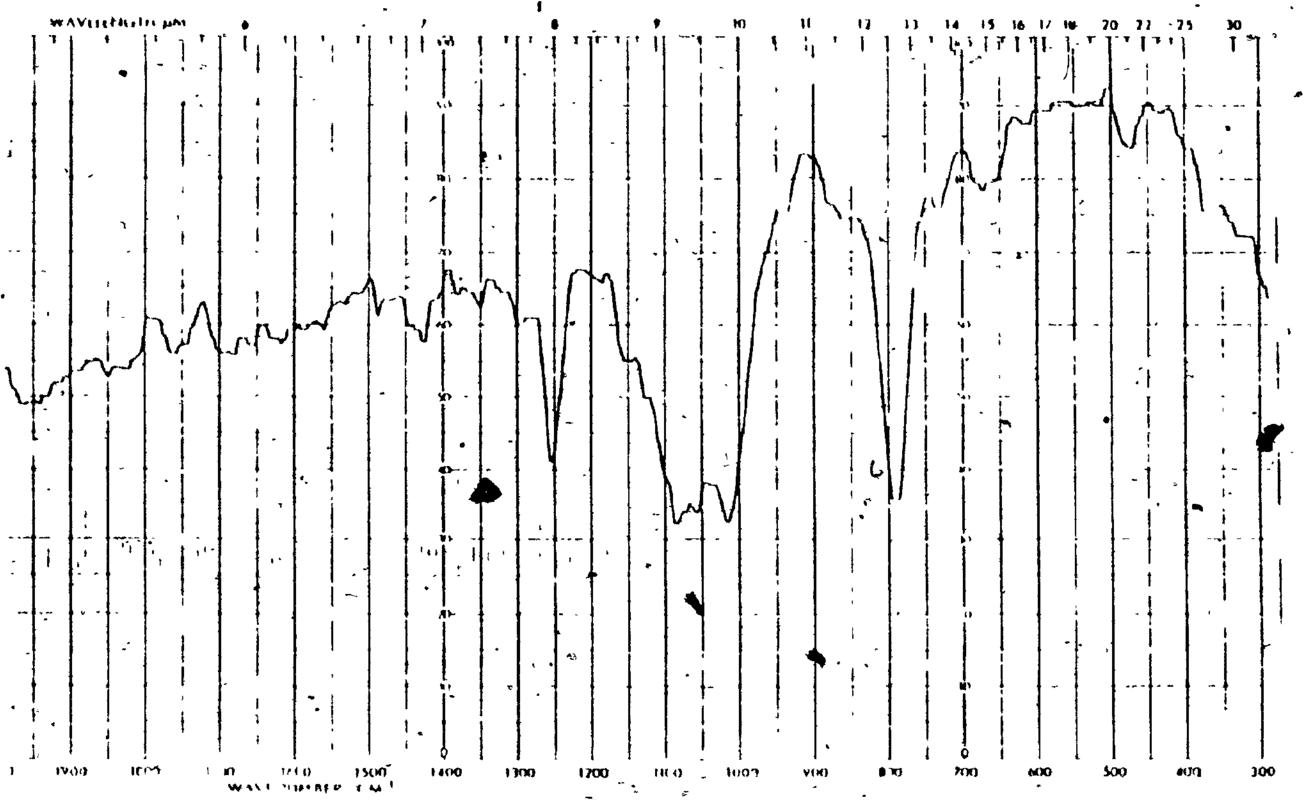


Figure 30 Infrared spectrum of PDS-PVC (10%)

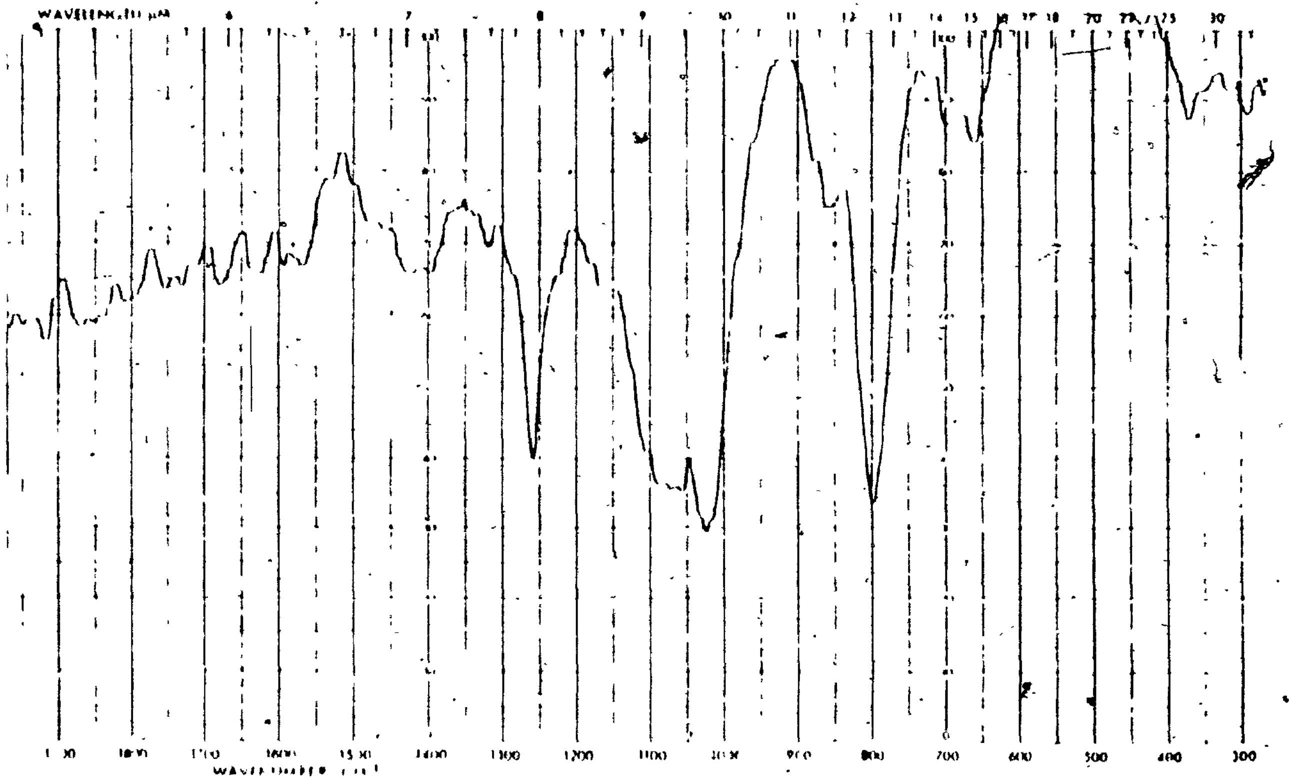


Figure 31 Infrared spectrum of PDS-VC/VAc (15%).

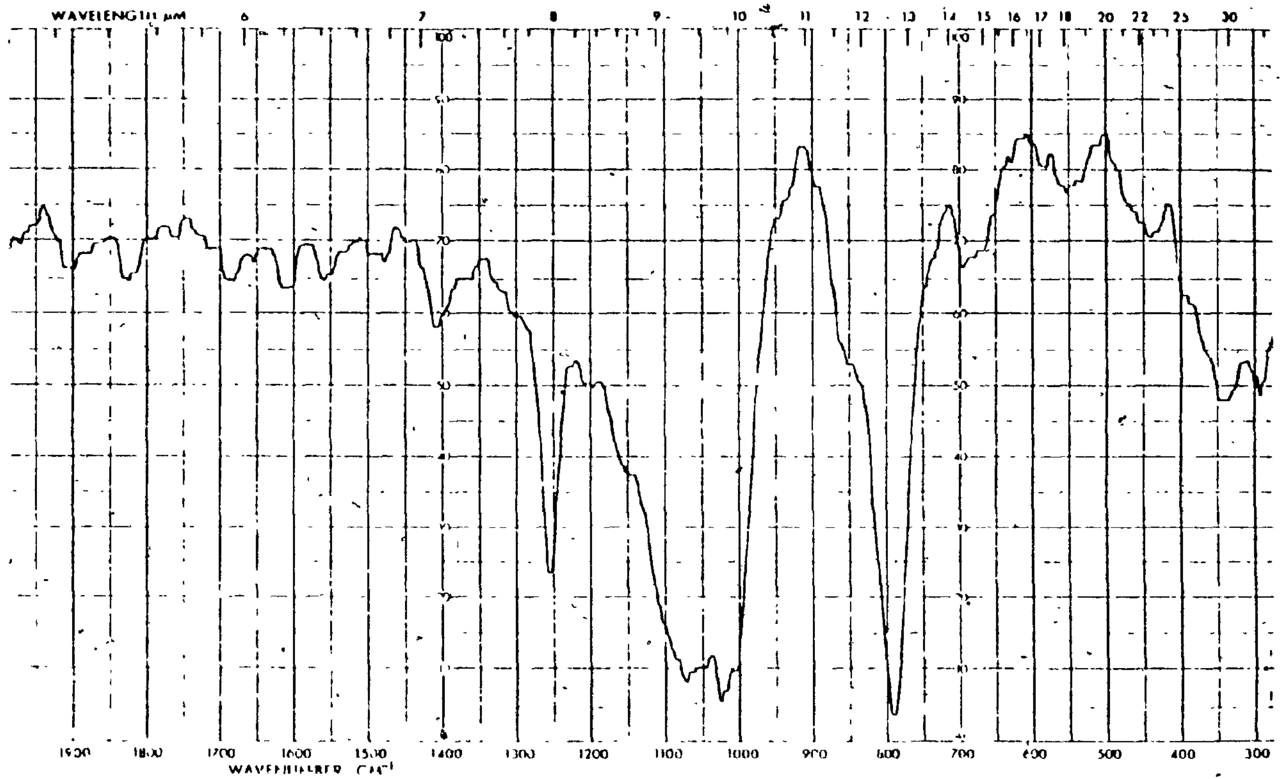


Figure 32 Infrared spectrum of PDS-K211 (10%)

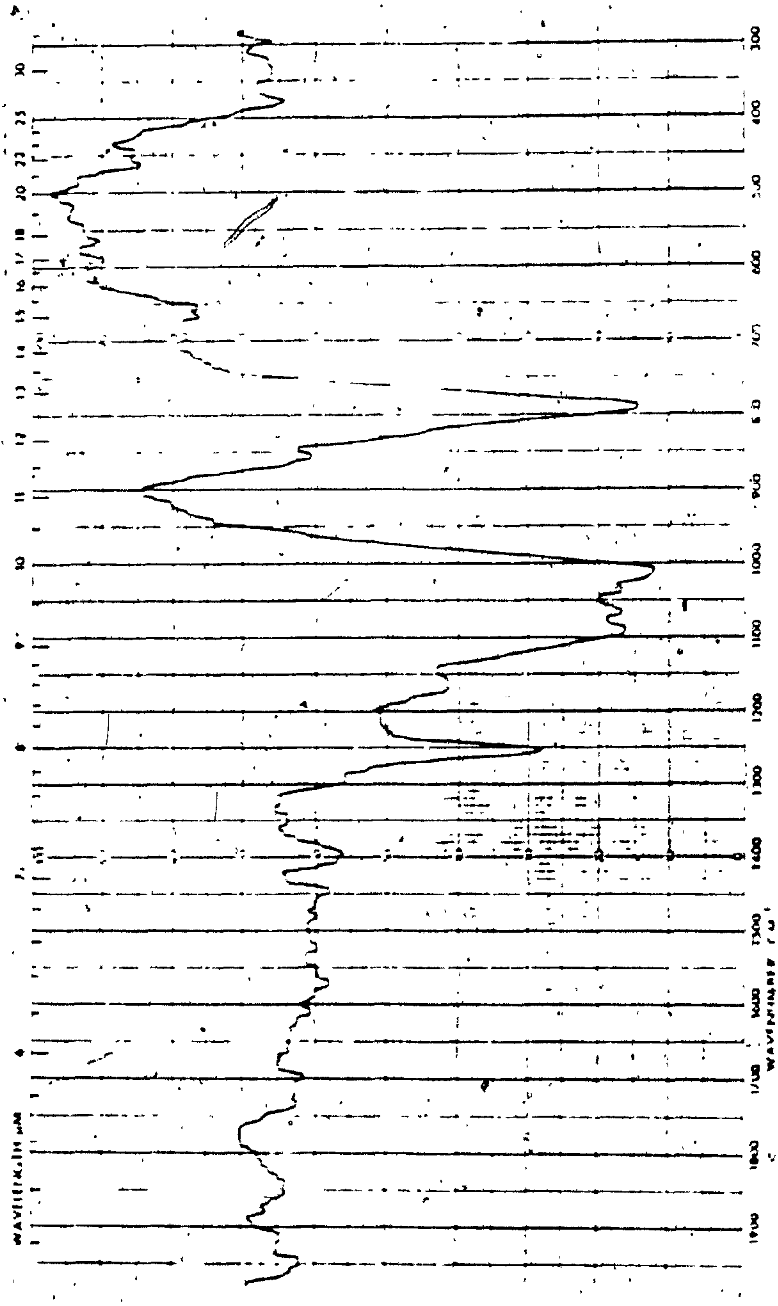


Figure 33 Infrared spectrum of PDS-lignin (10%)

8.4 Silicone Sealants

Testing of all silicone (PDS) specimens indicated that, in terms of toughness where Wood>Mortar>Aluminum, the best performance occurred with a wood substrate (figs. 34-35). The control sample was the toughest, with the naturally weathered specimens at 75% toughness of control, and accelerated weathering specimens at 60% of control toughness.

Control PDS specimens with mortar substrates had 50% of the toughness of those with wood substrates, while control PDS specimens with aluminum substrates had only 15% of the toughness with wood. After accelerated weathering, these percentages reduced to 38% for mortar, and 7,5% for aluminum. With naturally weathered specimens, the percentage for mortar further decreased to 30%, but increased to 45% for aluminum.

Control PDS specimens with mortar substrates were the toughest with respect to this substrate. Both accelerated and naturally weathered specimens had 46% of the toughness of the control specimens. Specimens with aluminum substrates behaved slightly differently, as the naturally weathered specimens performed the best, with control specimens at 50% toughness of the naturally weathered ones, and accelerated weathering specimens at 14%.

DSC testing resulted in a Tg for PDS specimens of -111°C (fig. 36).

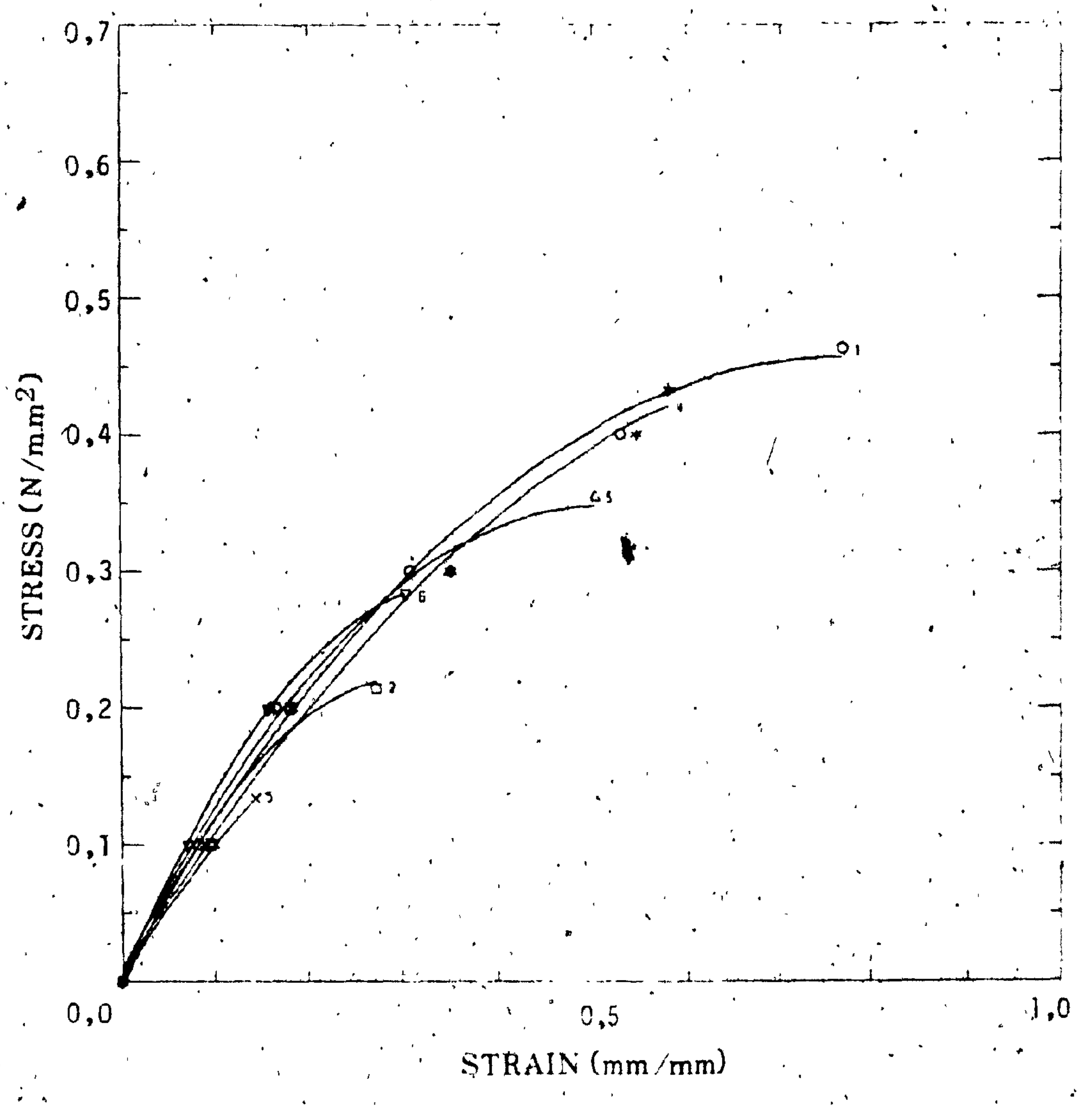


Figure 34 Stress-strain curves obtained with control and A.W. PDS specimens. Curve 1-control/wood substrate; curve 2-control/aluminum substrate; curve 3-control/mortar substrate; curve 4-A.W./wood substrate; curve 5-A.W./aluminum substrate; curve 6-A.W./mortar substrate

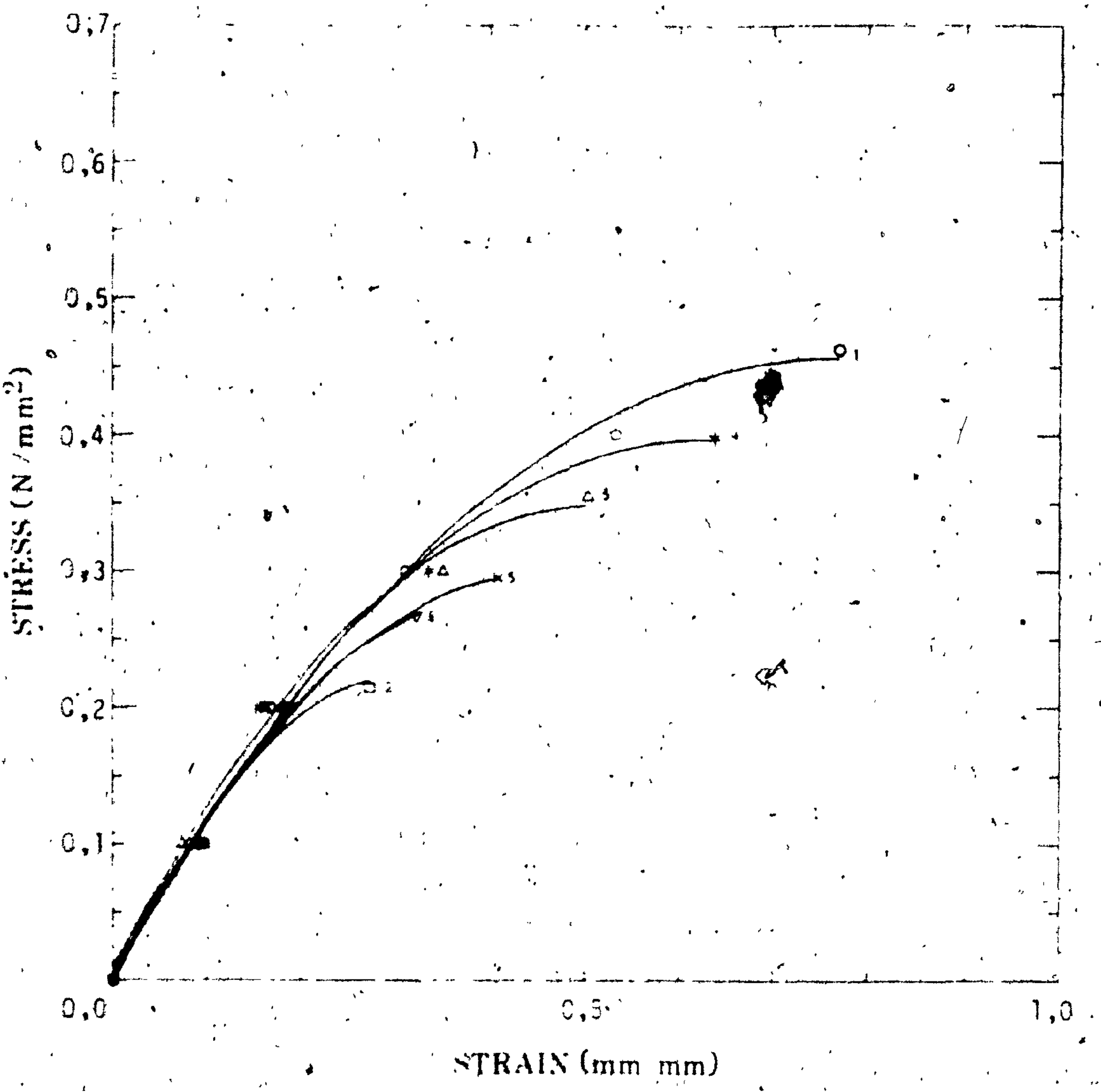


Figure 35 Stress-strain curves obtained with control and N.W. PDS specimens. Curve 1-control/wood substrate; curve 2-control/aluminum substrate; curve 3-control/mortar substrate; curve 4-N.W./wood substrate; curve 5-N.W./aluminum substrate; curve 6-N.W./mortar substrate

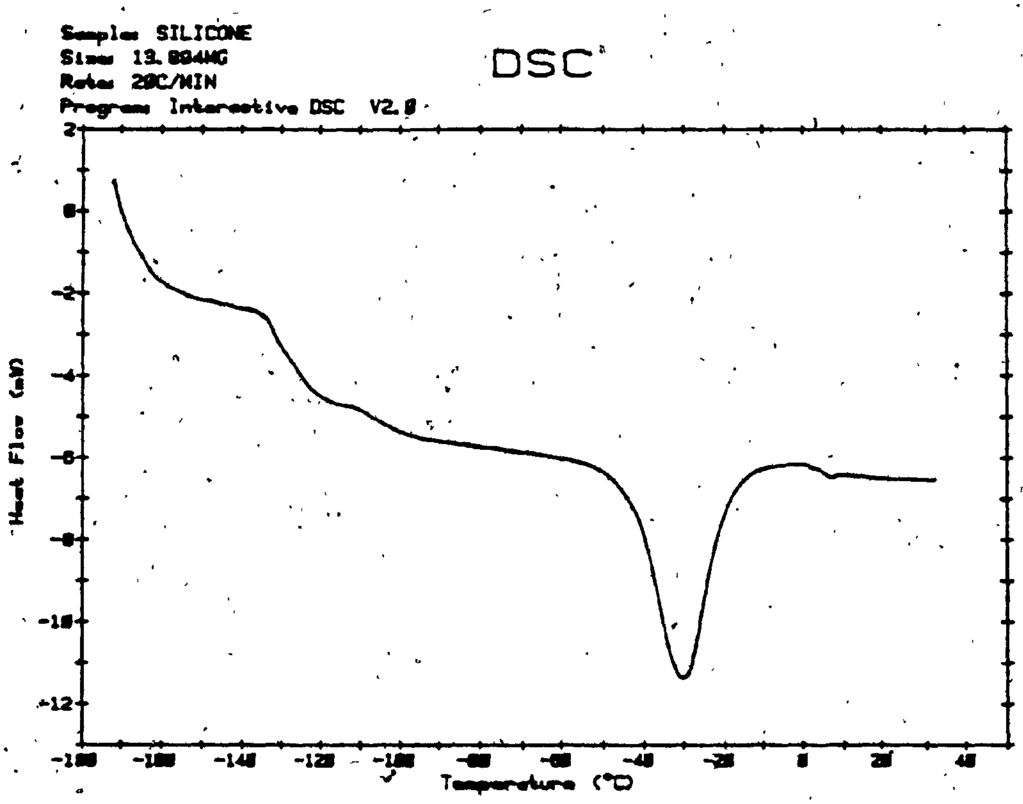


Figure 36 DSC scan (-170°C to +50°C) of PDS specimen

8.5 Silicone - Vinyl Polymer Polyblends

8.5.1 PDS-PVC Polyblends

PDS-PVC polyblends exhibited the best stress-strain performance with wood substrates. Failures were generally cohesive (fig. 37), indicating a strong adhesion between sealant and substrate. **The polyblends with 10% PVC consistently performed the best under mechanical testing, showing the greatest increase in toughness over PDS** (table 4) in all cases (control, accelerated, and natural weathering) (fig. 38). The accelerated weathering 10% PVC polyblend was also tougher than the control PDS specimens. 5% and 15% PVC specimens were consistently less tough than the PDS specimens.

Results with mortar substrates (fig. 39) were not as consistent as with wood. Only the 10% PVC control and accelerated weathering specimens performed better than their associated PDS specimens. The naturally weathered 10% PVC specimens had basically identical toughness as their associated PDS specimens.

Performance results with aluminum substrates (fig. 40), where only the 5% PVC polyblends were prepared, were generally poor. The only exception was the case of the naturally weathered which were tougher than the control and accelerated weathering specimens, and the control PDS specimens.

5% PVC polyblend specimens (naturally weathered) were tougher than control for all substrates, and tougher than accelerated weathering specimens with aluminum and mortar substrates only.

DSC testing of pure PVC indicates a Tg of 87°C. Testing of PDS-PVC polyblends shows two Tg's, one at -110,3°C (fig. 41), and a second at 92,3°C (fig. 42). These values correspond to the Tg's of the individual polymers, which indicates a two-phase polyblend (incompatibility).

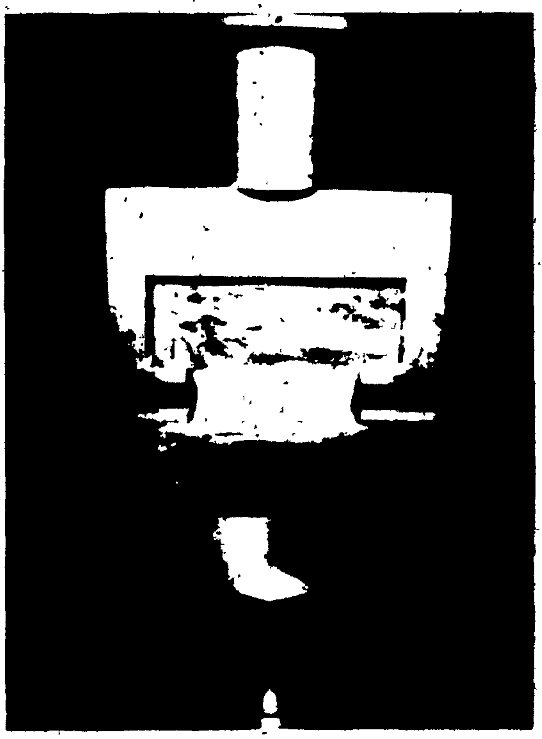


Figure 37 Cohesive failure of specimen

SPECIMEN	SUBSTRATE	TOUGHNESS ($N/mm^2.mm/mm$)		
		Exposure Conditions		
		Control	Accelerated	Natural
PDS	Wood	0,23	0,14	0,17
PDS	Aluminum	0,04	0,01	0,08
PDS	Mortar	0,12	0,05	0,05
PDS-PVC(5%)	Wood	0,04	0,11	0,08
PDS-PVC(5%)	Aluminum	0,01	0,02	0,09
PDS-PVC(5%)	Mortar	0,04	0,04	0,10
PDS-PVC(10%)	Wood	0,26	0,25	0,20
PDS-PVC(10%)	Aluminum	-	-	-
PDS-PVC(10%)	Mortar	0,23	0,06	0,05
PDS-PVC(15%)	Wood	0,12	1,17	0,12
PDS-PVC(15%)	Aluminum	-	-	-
PDS-PVC(15%)	Mortar	0,12	0,02	0,02

Table 4 Toughness of PDS-PVC polyblends

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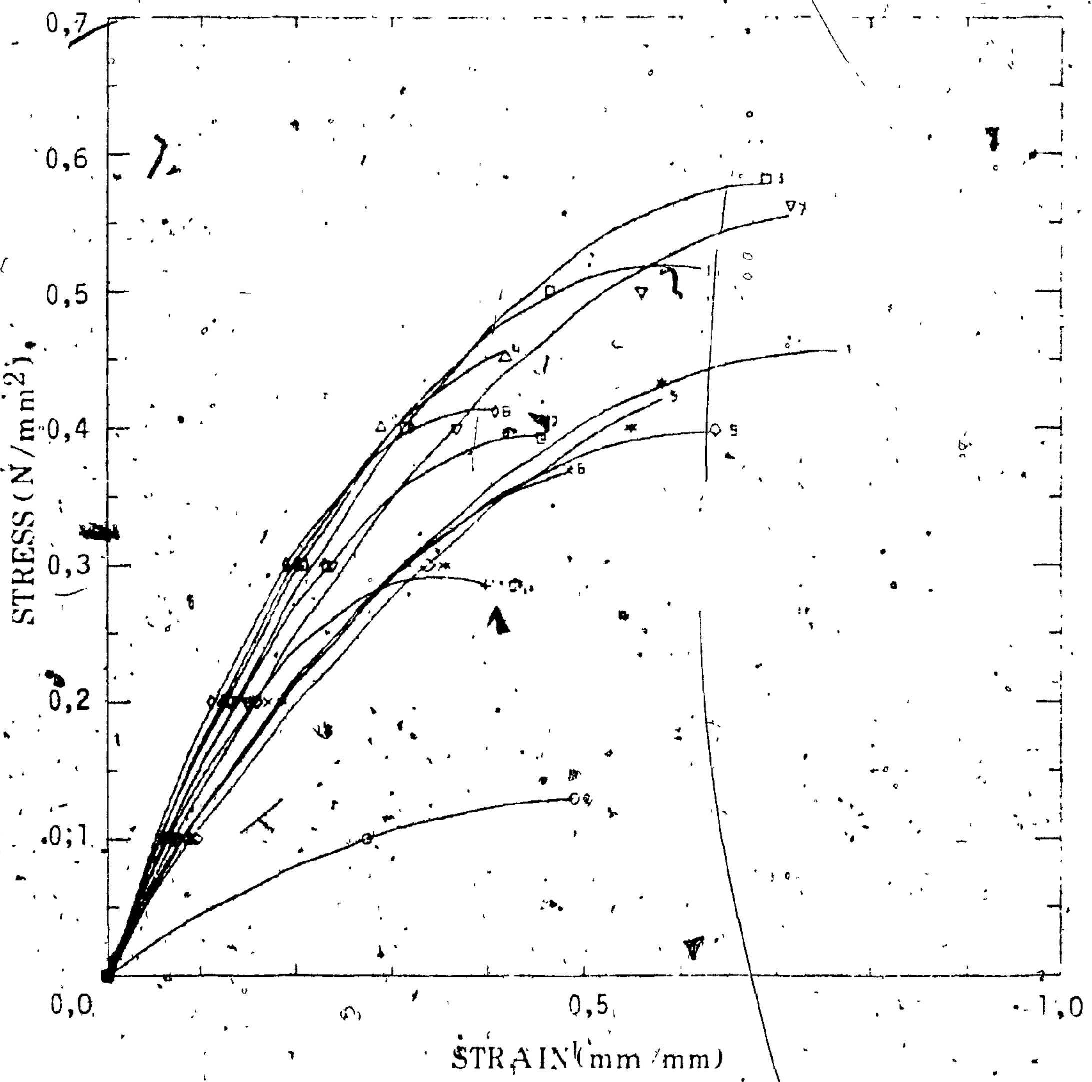

Figure 38 Stress-strain curves obtained with PDS-PVC polyblends on a wood substrate. Curve 1-control 0% PVC; curve 2-control 5% PVC; curve 3-control 10% PVC; curve 4-control 15% PVC; curve 5-A.W. 0% PVC; curve 6-A.W. 5% PVC; curve 7-A.W. 10% PVC; curve 8-A.W. 15% PVC; curve 9-N.W. 0% PVC; curve 10-N.W. 5% PVC; curve 11-N.W. 10% PVC; curve 12-N.W. 15% PVC

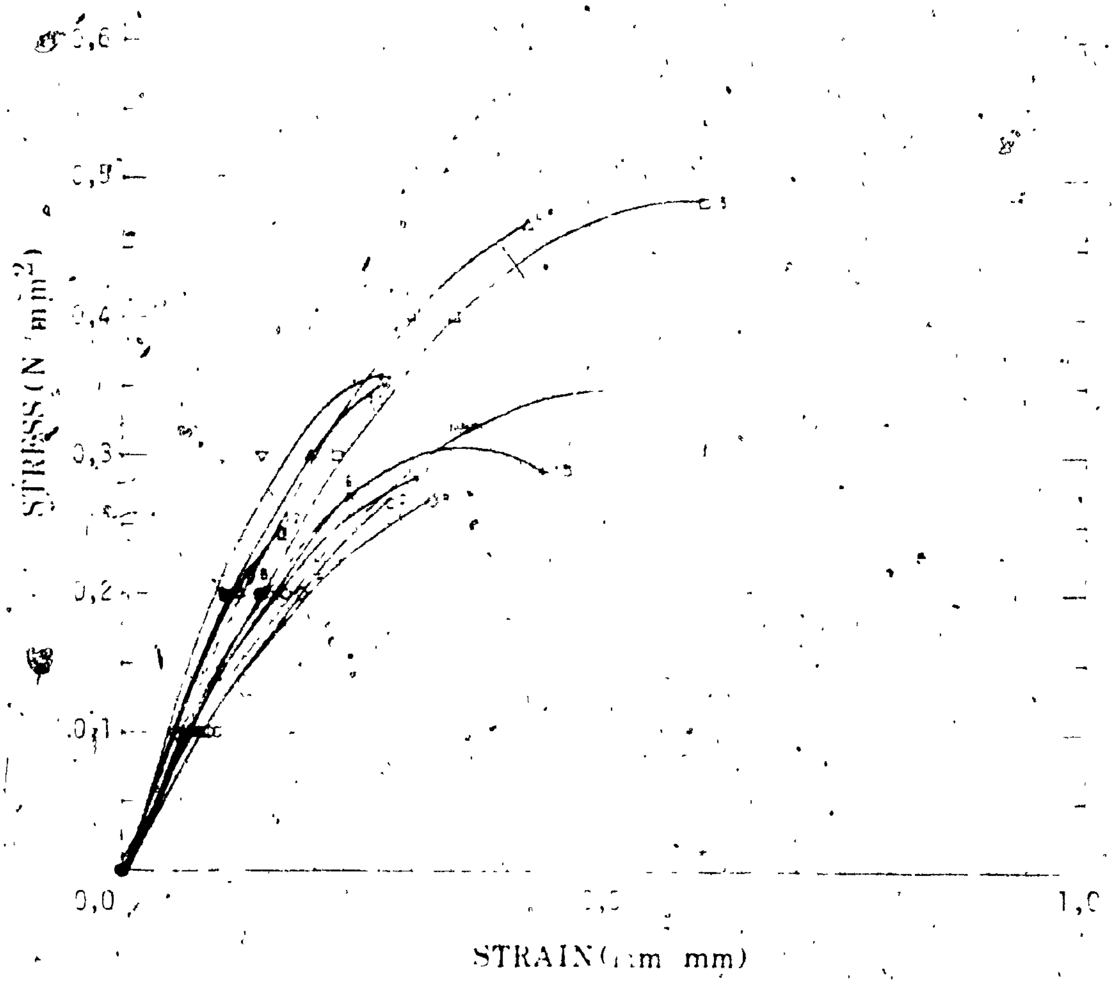

Figure 39 Stress-strain curves obtained with PDS-PVC polyblends on a mortar substrate. Curve 1-control 0% PVC; curve 2-control 5% PVC; curve 3-control 10% PVC; curve 4-control 15% PVC; curve 5-A.W. 0% PVC; curve 6-A.W. 5% PVC; curve 7-A.W. 10% PVC; curve 8-A.W. 15% PVC; curve 9-N.W. 0% PVC; curve 10-N.W. 5% PVC; curve 11-N.W. 10% PVC; curve 12-N.W. 15% PVC

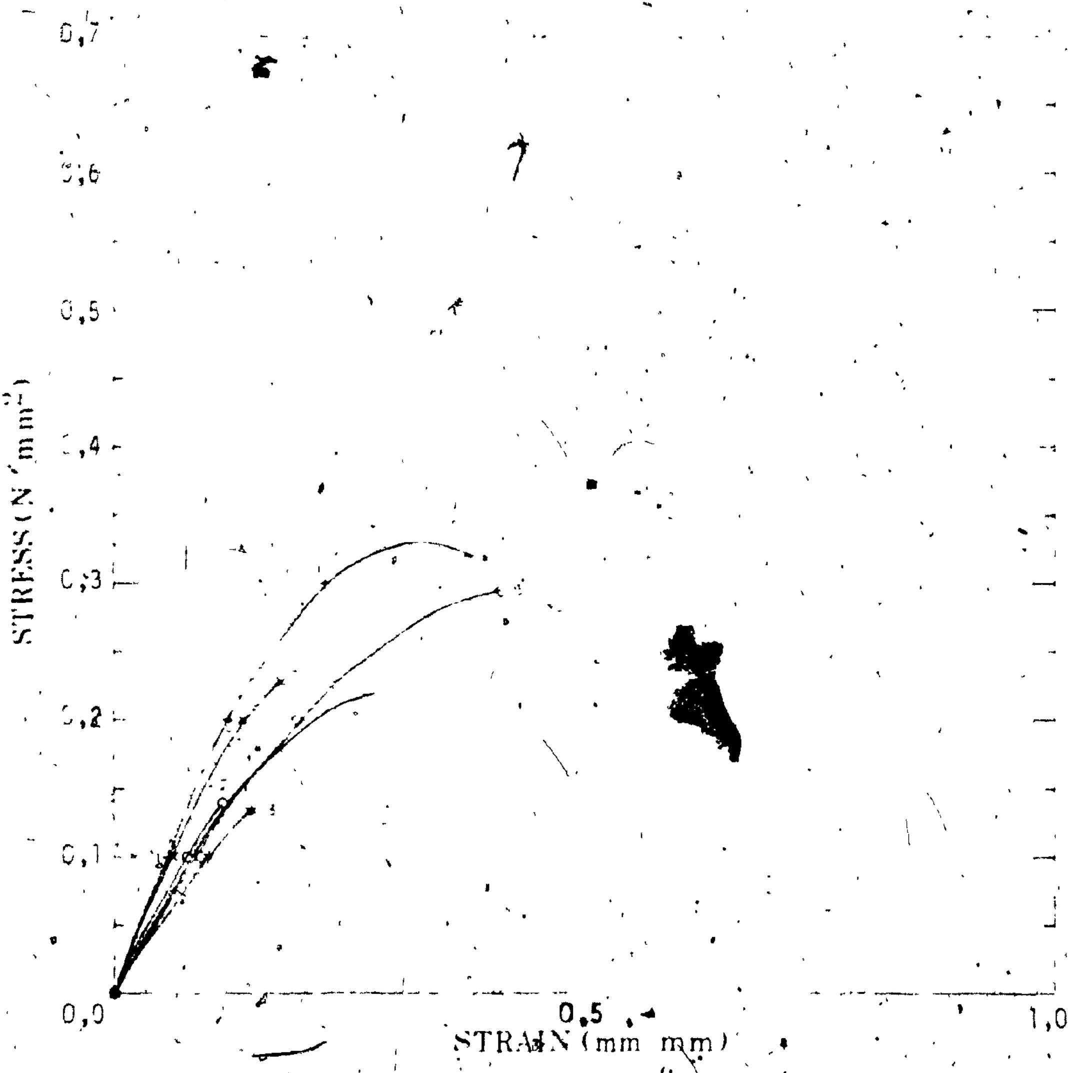

Figure 40 Stress-strain curves obtained with PDS-PVC polyblends on an aluminum substrate. Curve 1-control 0% PVC; curve 2-control 5% PVC; curve 3-A.W. 0% PVC; curve 4-A.W. 5% PVC; curve 5-N.W. 0% PVC; curve 6-N.W. 5% PVC

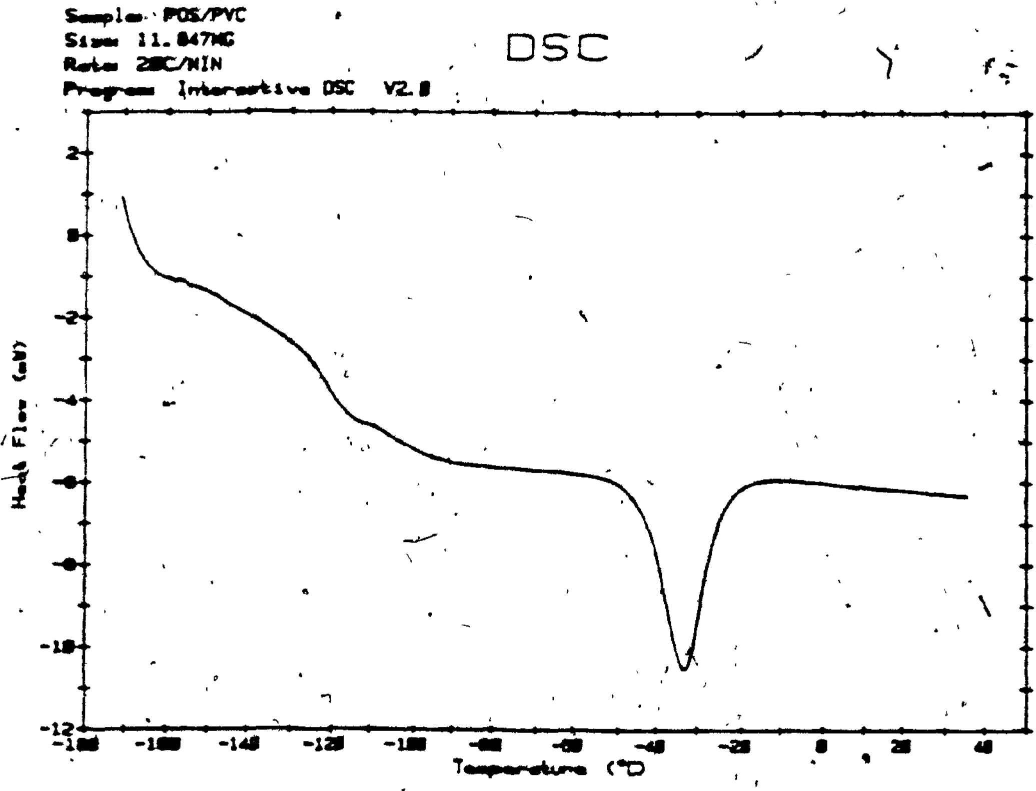

Figure 41 DSC scan (-170°C to +50°C) of PDS-PVC specimen

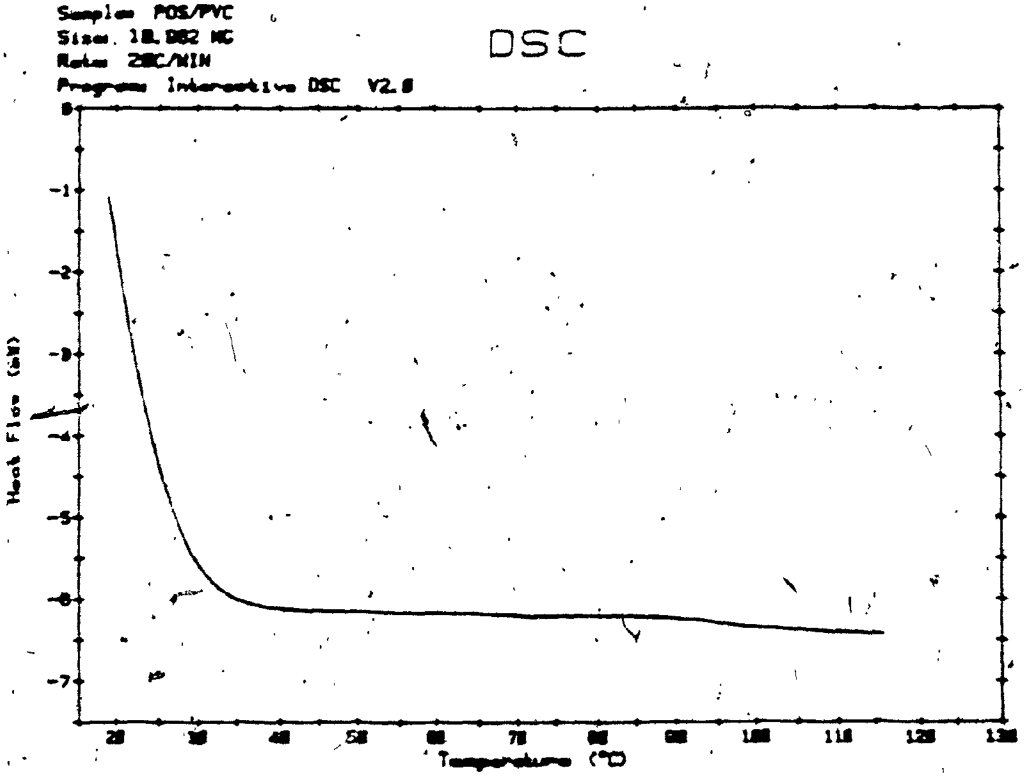

Figure 42 DSC scan (+20°C to +150°C) of PDS-PVC specimen

8.5.2 PDS-VC/VAc Polyblends

Failure of all PDS-VC/VAc specimens was adhesive (fig. 43), indicating good cohesion within the polyblend. PDS-VC/VAc polyblends exhibited the best stress-strain performance with aluminum substrates (fig. 44). As was the case with the 10% PVC-PDS polyblends on a wood substrate, **the 15% VC/VAc polyblends with aluminum substrates consistently performed the best under mechanical testing, and exhibited superior stress-strain behaviour over all the specimens tested** (including PVC and PDS). The only exception was the case of the control specimens where the 10% VC/VAc polyblends were slightly tougher than 15% (table 5), although the allowable stress of the 15% specimens was much higher than that of 10%. Failures of these specimens was adhesive, indicating strong cohesion within the polyblend. All the PDS-VC/VAc polyblend specimens with aluminum substrates were tougher than the control PDS specimens on this substrate.

Although the control PDS-VC/VAc specimens with mortar substrates (fig. 45) were all tougher than control PDS specimens with this substrate, the weathered specimens (accelerated and natural) were all much weaker than the PDS specimens.

All of the PDS-VC/VAc polyblend specimens with wood substrates (fig. 46) were much weaker than their associated PDS specimens.

DSC testing of the VC/VAc copolymer resulted in a Tg of [illegible]2°C, which is a value that is characteristic for copolymers with a high amount of of vinyl chloride. Testing of the PDS-VC/VAc polyblend specimens resulted in two Tg's, one at -11[illegible]°C (fig. 47), the second at [illegible]°C (fig. [illegible]). As was the case with the PDS-PVC polyblends, these Tg's correspond to the Tg's of the constituent polymers, which indicates a two-phase polyblend (incompatibility).

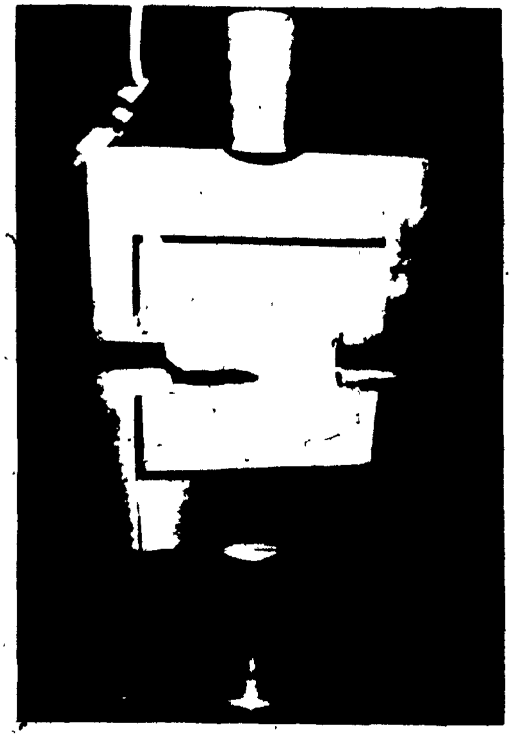

Figure 43 Adhesive failure of specimen

SPECIMEN	SUBSTRATE	TOUGHNESS ($N/mm^2.mm/mm$)		
		Exposure Conditions		
		Control	Accelerated	Natural
PDS	Wood	0,23	0,14	0,17
PDS	Aluminum	0,04	0,01	0,08
PDS	Mortar	0,12	0,05	0,05
PDS-VC/VAc(5%)	Wood	0,14	0,56	0,06
PDS-VC/VAc(5%)	Aluminum	0,08	0,06	0,20
PDS-VC/VAc(5%)	Mortar	0,14	0,03	0,03
PDS-VC/VAc(10%)	Wood	0,09	0,05	0,04
PDS-VC/VAc(10%)	Aluminum	0,19	0,07	0,18
PDS-VC/VAc(10%)	Mortar	0,18	0,03	0,03
PDS-VC/VAc(15%)	Wood	0,18	0,05	0,07
PDS-VC/VAc(15%)	Aluminum	0,18	0,14	0,27
PDS-VC/VAc(15%)	Mortar	0,18	0,03	0,01

Table 5 Toughness of PDS-VC/VAc polyblends

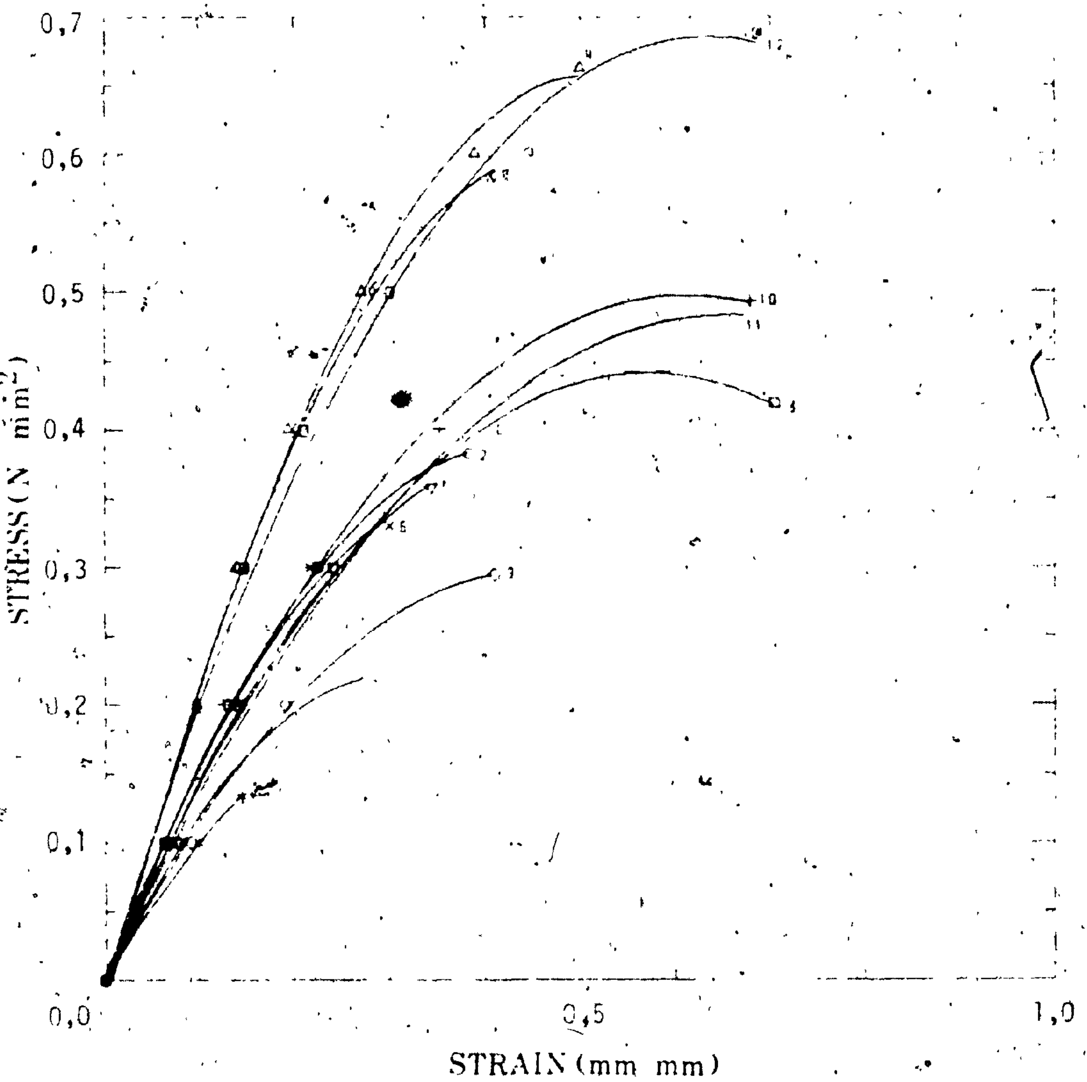

Figure 44. Stress-strain curves obtained with PDS-VC/VAc polyblends on an aluminum substrate. Curve 1-control 0% VC/VAc; curve 2-control 5% VC/VAc; curve 3-control 10% VC/VAc; curve 4-control 15% VC/VAc; curve 5-A.W. 0% VC/VAc; curve 6-A.W. 5% VC/VAc; curve 7-A.W. 10% VC/VAc; curve 8-A.W. 15% VC/VAc; curve 9-N.W. 0% VC/VAc; curve 10-N.W. 5% VC/VAc; curve 11-N.W. 10% VC/VAc; curve 12-N.W. 15% VC/VAc

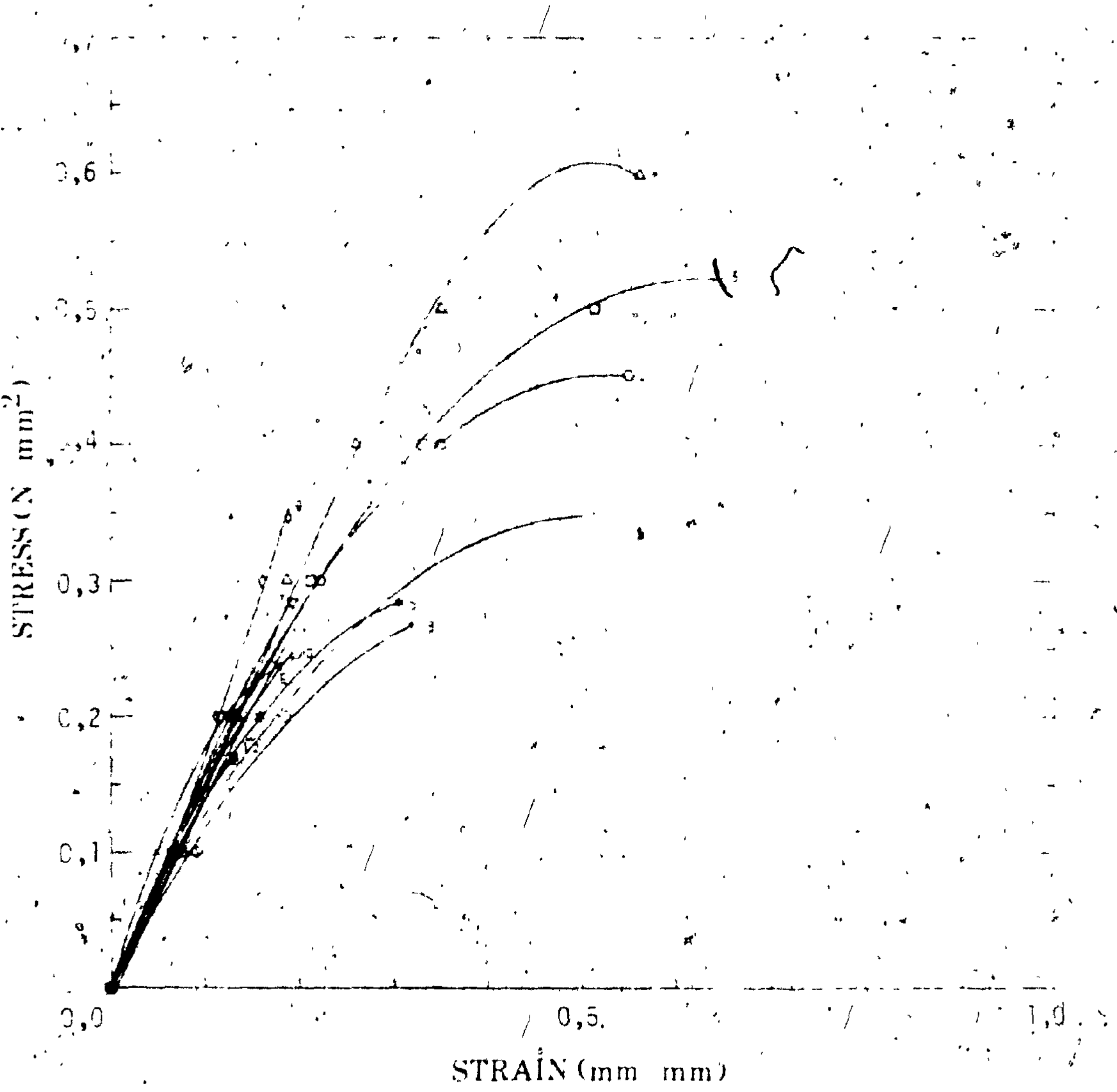

Figure 45 Stress-strain curves obtained with PDS-VC/VAc polyblends on a mortar substrate. Curve 1-control 0% VC/VAc; curve 2-control 5% VC/VAc; curve 3-control 10% VC/VAc; curve 4-control 15% VC/VAc; curve 5-A.W. 0% VC/VAc; curve 6-A.W. 5% VC/VAc; curve 7-A.W. 10% VC/VAc; curve 8-A.W. 15% VC/VAc; curve 9-N.W. 0% VC/VAc; curve 10-N.W. 5% VC/VAc; curve 11-N.W. 10% VC/VAc; curve 12-N.W. 15% VC/VAc

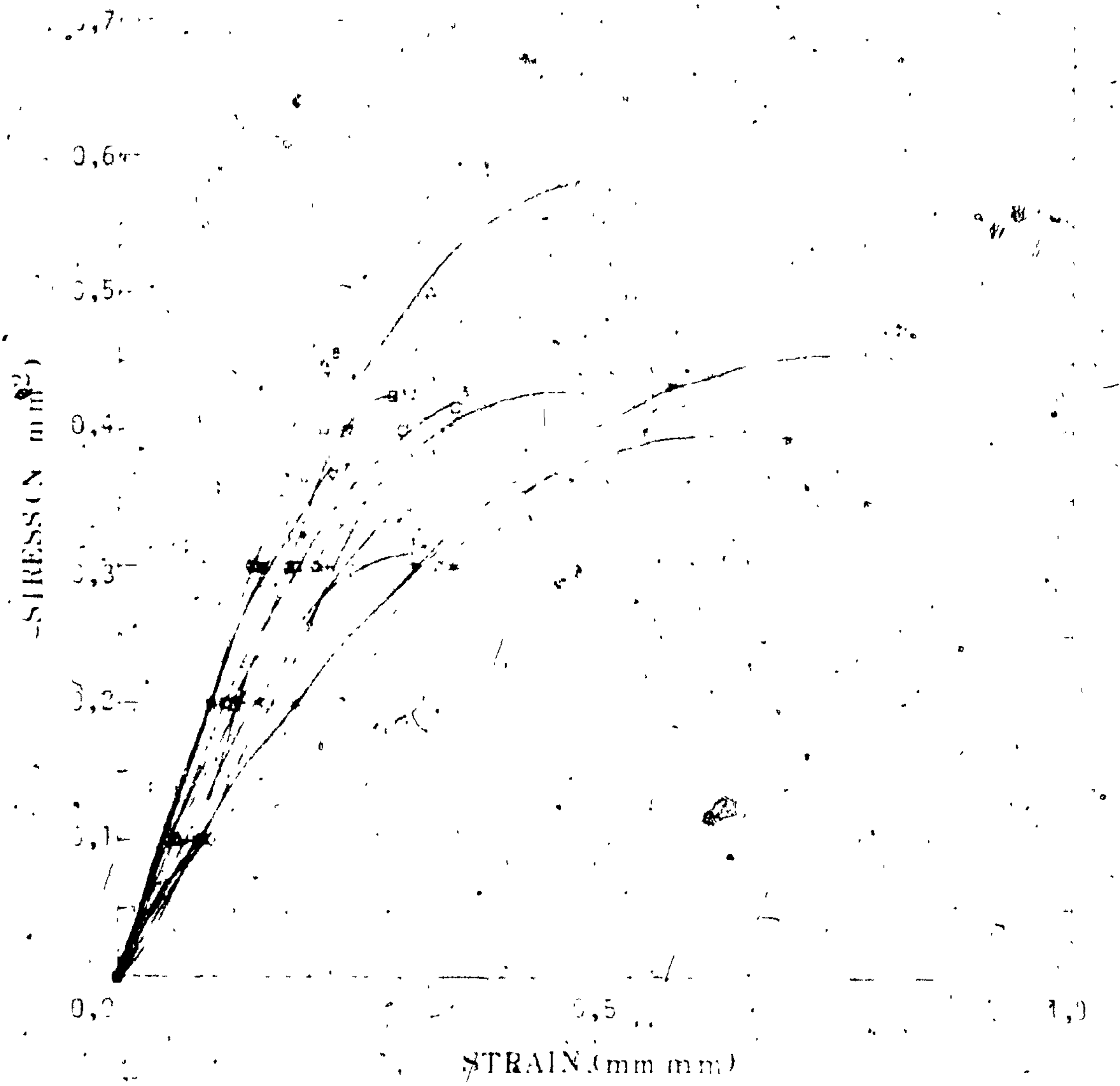

Figure 46 Stress-strain curves obtained with PDS-VC/VAc polyblends on a wood substrate. Curve 1-control 0% VC/VAc; curve 2-control 5% VC/VAc; curve 3-control 10% VC/VAc; curve 4-control 15% VC/VAc; curve 5-A.W. 0% VC/VAc; curve 6-A.W. 5% VC/VAc; curve 7-A.W. 10% VC/VAc; curve 8-A.W. 15% VC/VAc; curve 9-N.W. 0% VC/VAc; curve 10-N.W. 5% VC/VAc; curve 11-N.W. 10% VC/VAc; curve 12-N.W. 15% VC/VAc

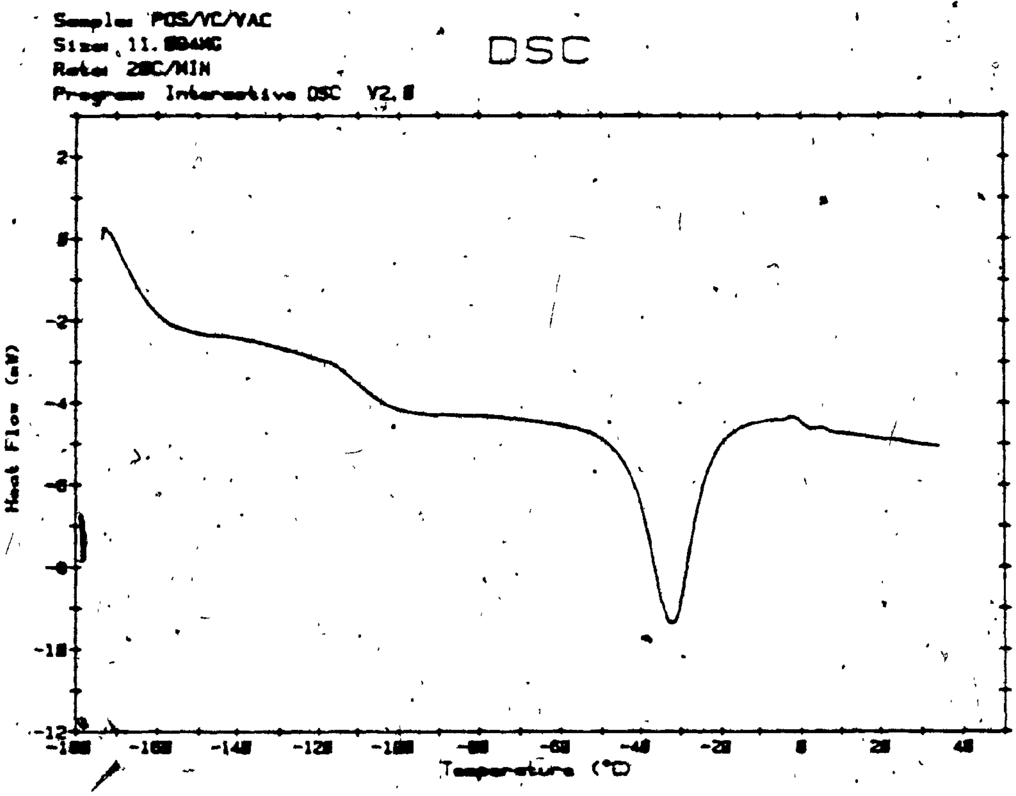

Figure 47 DSC scan (-170°C to +50°C) of PDS-VC/VAc specimen

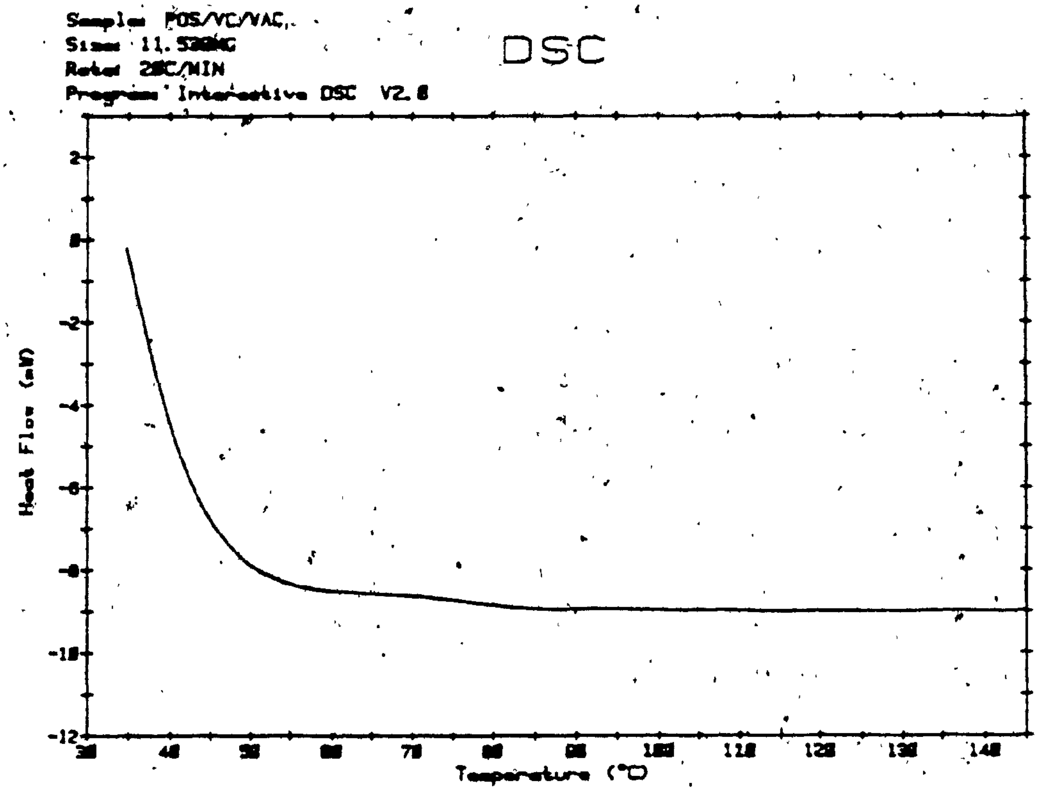

Figure 48 DSC scan (+20°C to +150°C) of PDS-VC/VAc specimen

8.5.3 Discussion and Conclusions

DSC and SEM results indicate that both vinyl polyblends, PDS-PVC and PDS-VC/VAc, are not miscible, thus their mechanical properties are influenced by the interfacial adhesion between the two co-existing phases. The existence of these two phases is not necessarily unfavourable since many useful properties, characteristic of a single phase, may be preserved in the blend while other properties may be averaged according to the blend composition (20).

Consistency of results in mechanical testing gives an indication of which of the polyblends has truly superior performance, therefore improved durability, over the PDS specimens. In the case of a wood substrate, the best results were obtained with the 10% PVC polyblend (as indicated by stress, strain, and toughness). Failures of specimens were cohesive, showing good adhesion between the sealant bead and the wood substrate. Improvements in adhesion with wood substrates might be attributed to some interactions between the polar groups of PVC and those of wood components.

With aluminum substrates, the 15% VC/VAc polyblend exhibited superior performance over all the experimented specimens in this study. The improved properties of this polyblend could be due to the favourable influence, on the adhesion towards all substrates, of the acetylic groups of VC-VAc. Failures of this polyblend were adhesive, indicating very strong cohesion within the polyblend.

8.6 Silicone - Nitrile Elastomer Polyblends

8.6.1 PDS-K1402 Polyblends

During mechanical testing, PDS-K1402 polyblends **consistently exhibited better stress-strain performance** (toughness) **than PDS with aluminum substrates only** (fig. 49). The A.W. and N.W. specimens on this substrate also performed better than the control PDS specimens. **The 5% K1402 polyblend specimens showed the greatest improvement in toughness over PDS** (table 6) **in all exposure cases** (control, A.W., and N.W.). The majority of failures with the 5% polyblend with an aluminum substrate were cohesive. Failures above the 5% dispersed phase were adhesive, and adhesive strengths at failure decreased with increasing percentage of K1402 within the polyblend. This serves as an indication that any addition of K1402 above the 5% level decreases the degree of adhesion between the sealant bead and the aluminum substrate.

Results with mortar substrates (fig. 50) were not as consistent as with aluminum. Control and A.W. specimen testing results with 5 and 10% polyblends were encouraging, but N.W. results indicated that the polyblend was substantially weaker than PDS. These failures were generally cohesive, but were all adhesive for the N.W. and 15% polyblend specimens. As was the case with the aluminum substrate, this indicates that the addition of an increasing amount of K1402 decreases the adhesive strength of the sealant. Natural weathering elements not accounted for within the accelerated weathering chamber also had a detrimental effect on the performance of the sealant.

Wood substrate results were inconsistent, as shown in figure 51. While the 10% polyblend specimens, exposed to control and A.W. conditions, exhibited superior toughness over PDS, N.W. specimens were weaker than PDS (although the decrease in toughness was not as dramatic as with mortar). 5 and 15% polyblend specimens were not as tough as PDS specimens, except in the case of the A.W. 5% polyblend specimens, where results indicated that they were substantially tougher than PDS. All failures with this substrate were adhesive, indicating a strong cohesive force within the substrate.

DSC testing of K1402 resulted in a Tg of -35oC. Testing of the PDS-K1402 specimens yielded only one observable Tg at -114,1oC (fig. 52), which corresponds to the Tg of PDS. As was mentioned previously, a miscible polyblend exhibits only one Tg, but it is located somewhere between the Tg's of the constituent polymers. As PDS is an unpolar polymer while K1402 is polar, it can be concluded from these DSC results that PDS-K1402 is not a miscible polymer (incompatible).

SPECIMEN	SUBSTRATE	TOUGHNESS (N/mm^2.mm/mm)		
		Exposure Conditions		
		Control	Accelerated	Natural
PDS	Wood	0,23	0,14	0,17
PDS	Aluminum	0,04	0,01	0,08
PDS	Mortar	0,12	0,05	0,05
PDS-K1402(5%)	Wood	0,15	3,39	0,07
PDS-K1402(5%)	Aluminum	0,19	0,19	0,20
PDS-K1402(5%)	Mortar	0,13	0,32	0,02
PDS-K1402(10%)	Wood	0,32	1,87	0,12
PDS-K1402(10%)	Aluminum	0,06	0,10	0,10
PDS-K1402(10%)	Mortar	0,39	0,26	0,01
PDS-K1402(15%)	Wood	0,15	0,08	0,05
PDS-K1402(15%)	Aluminum	0,05	0,08	0,09
PDS-K1402(15%)	Mortar	0,06	0,01	0,01

Table 6 Toughness of PDS-K1402 polyblends

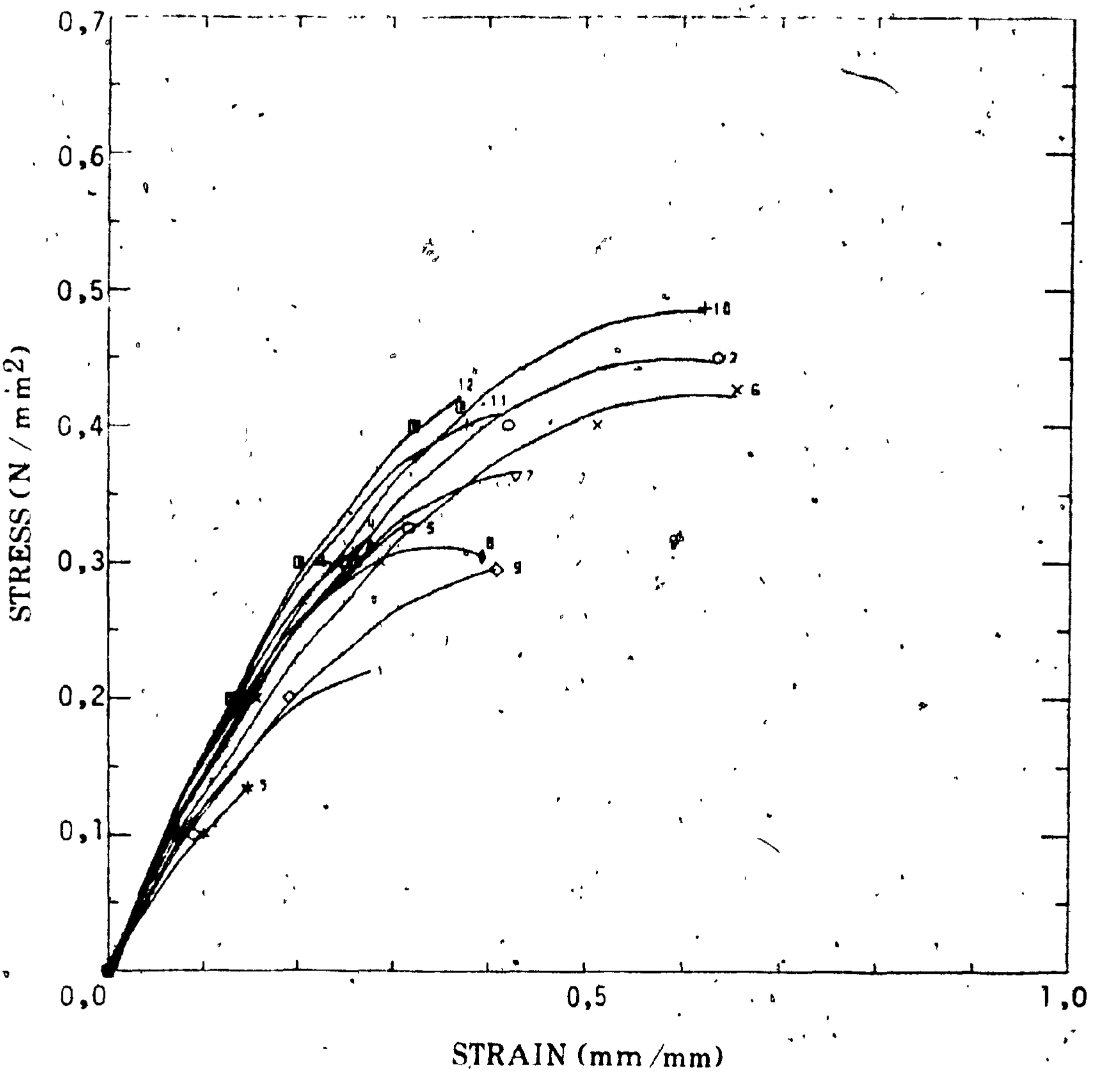

Figure 49 Stress-strain curves obtained with PDS-K1402 polyblends on an aluminum substrate. Curve 1-control 0% K1402; curve 2-control 5% K1402; curve 3-control 10% K1402; curve 4-control 15% K1402; curve 5-A.W. 0% K1402; curve 6-A.W. 5% K1402; curve 7-A.W. 10% K1402; curve 8-A.W. 15% K1402; curve 9-N.W. 0% K1402; curve 10-N.W. 5% K1402; curve 11-N.W. 10% K1402; curve 12-N.W. 15% K1402

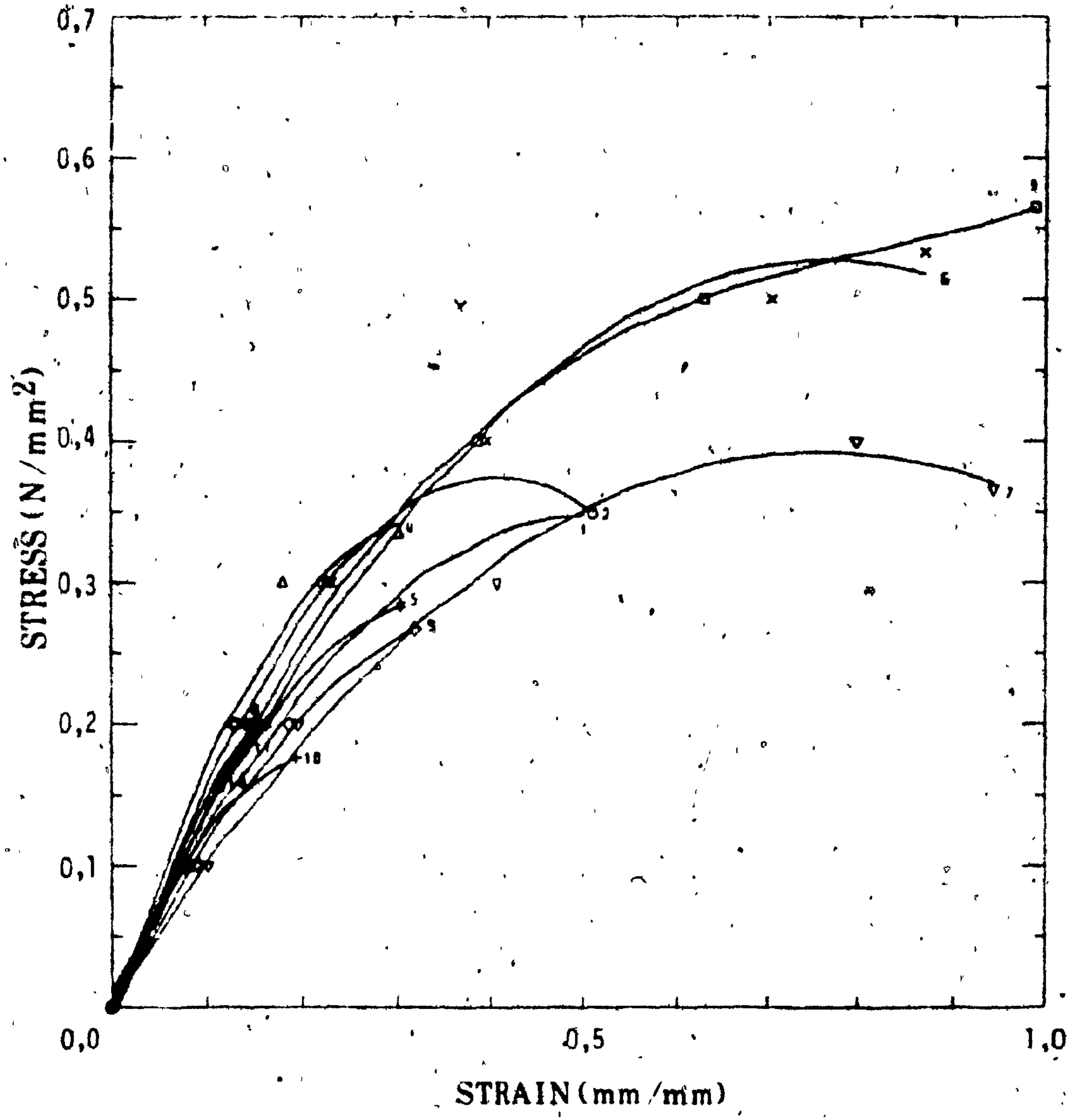

Figure 50 Stress-strain curves obtained with PDS-K1402 polyblends on a mortar substrate. Curve 1-control 0% K1402; curve 2-control 5% K1402; curve 3-control 10% K1402; curve 4-control 15% K1402; curve 5-A.W. 0% K1402; curve 6-A.W. 5% K1402; curve 7-A.W. 10% K1402; curve 8-A.W. 15% K1402; curve 9-N.W. 0% K1402; curve 10-N.W. 5% K1402; curve 11-N.W. 10% K1402; curve 12-N.W. 15% K1402

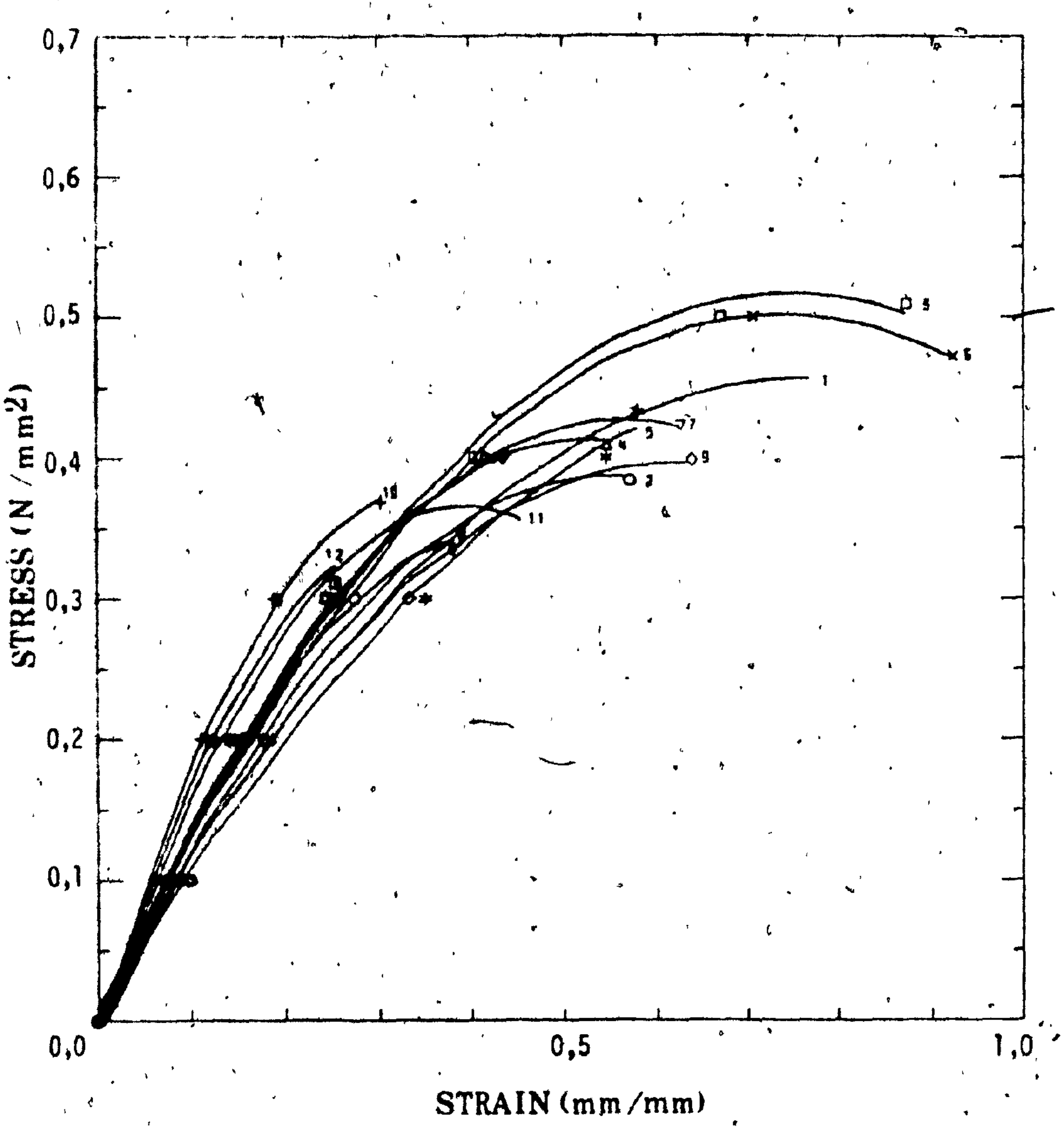

Figure 51 Stress-strain curves obtained with PDS-K1402 polyblends on a wood substrate. Curve 1-control 0% K1402; curve 2-control 5% K1402; curve 3-control 10% K1402; curve 4-control 15% K1402; curve 5-A.W. 0% K1402; curve 6-A.W. 5% K1402; curve 7-A.W. 10% K1402; curve 8-A.W. 15% K1402; curve 9-N.W. 0% K1402; curve 10-N.W. 5% K1402; curve 11-N.W. 10% K1402; curve 12-N.W. 15% K1402

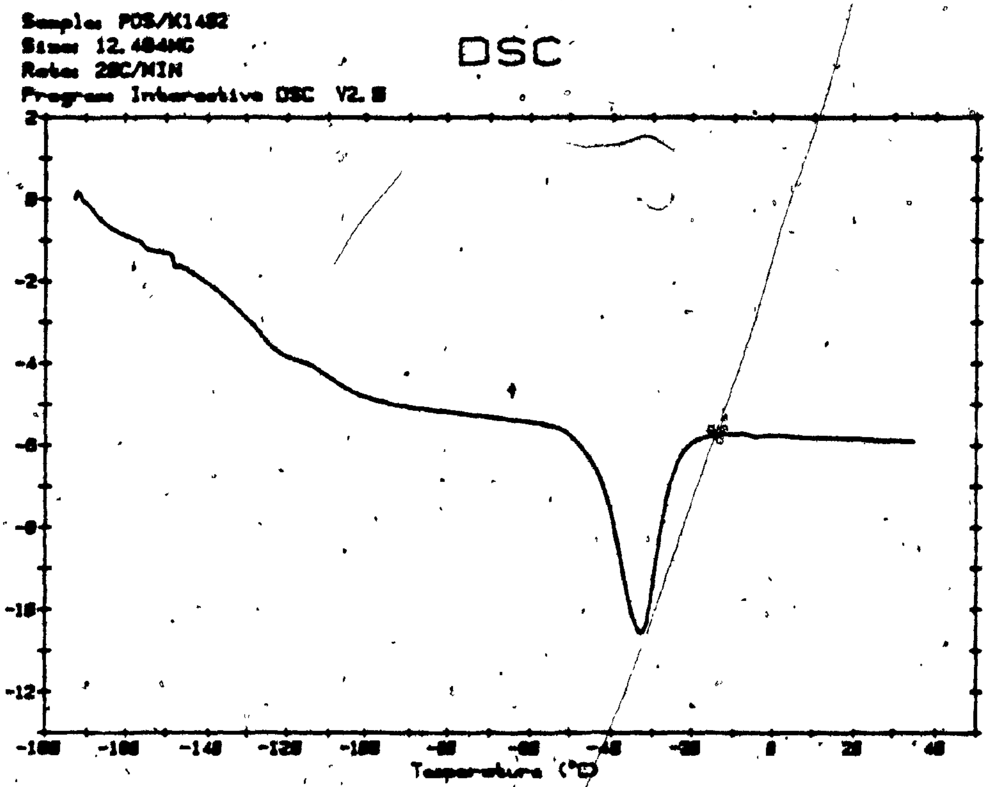

Figure 52 DSC scan (-170°C to +50°C) of PDS-K1402

8.6.2 PDS-K1411 Polyblends

Unlike the previous elastomer K1402, the elastomer K1411 has a high amount of ACN. Stress-strain results with the PDS-K1411 polyblend specimens were generally poor (Figs. 53-55), except in the case of the 5% polyblend on an aluminum substrate (fig. 55). Under control and A.W. exposure, mechanical testing results were significantly better than PDS, while the toughness of the N.W. specimens were essentially equal to that of the N.W. PDS specimens (table 7). The A.W. and N.W. specimens, with this substrate, were tougher than the control PDS specimens. Failures of all PDS-K1411 specimens tested were adhesive indicating a lack of adhesive strength between the sealant bead and substrates in this study.

In addition, there was a significant colour change of the sealant beads, from a translucent white to an opaque deep yellow, after exposure under all conditions (control, A.W., and N.W.). This indicates that the colour of the PDS-K1411 is light sensitive, probably due to the different composition of K1411.

DSC testing of K1411 resulted in a Tg of -20oC. As was the case with PDS-K1402, testing with PDS-K1411 resulted in only one Tg at -112,5oC (fig 56), which corresponds to the Tg of PDS. Since K1411 is a polar polymer, it can be concluded that PDS-KI411 is not a miscible polyblend. In comparing these results with those obtained with K1402, it can be concluded that the increase in the amount of ACN does not contribute to the improvement of PDS blend properties.

SPECIMEN	SUBSTRATE	TOUGHNESS ($N/mm^2.mm/mm$) Exposure Conditions Control	 Accelerated	 Natural
PDS	Wood	0,23	0,14	0,17
PDS	Aluminum	0,04	0,01	0,08
PDS	Mortar	0,12	0,05	0,05
PDS-K1411(5%)	Wood	0,04	0,01	0,04
PDS-K1411(5%)	Aluminum	0,06	0,04	0,07
PDS-K1411(5%)	Mortar	0,05	0,04	0,00
PDS-K1411(10%)	Wood	0,03	0,01	0,01
PDS-K1411(10%)	Aluminum	0,02	0,00	0,01
PDS-K1411(10%)	Mortar	0,02	0,02	0,00
PDS-K1411(15%)	Wood	0,01	0,01	0,00
PDS-K1411(15%)	Aluminum	0,01	0,00	0,00
PDS-K1411(15%)	Mortar	0,01	0,01	0,00

Table 7. Toughness of PDS-K1411 polyblends

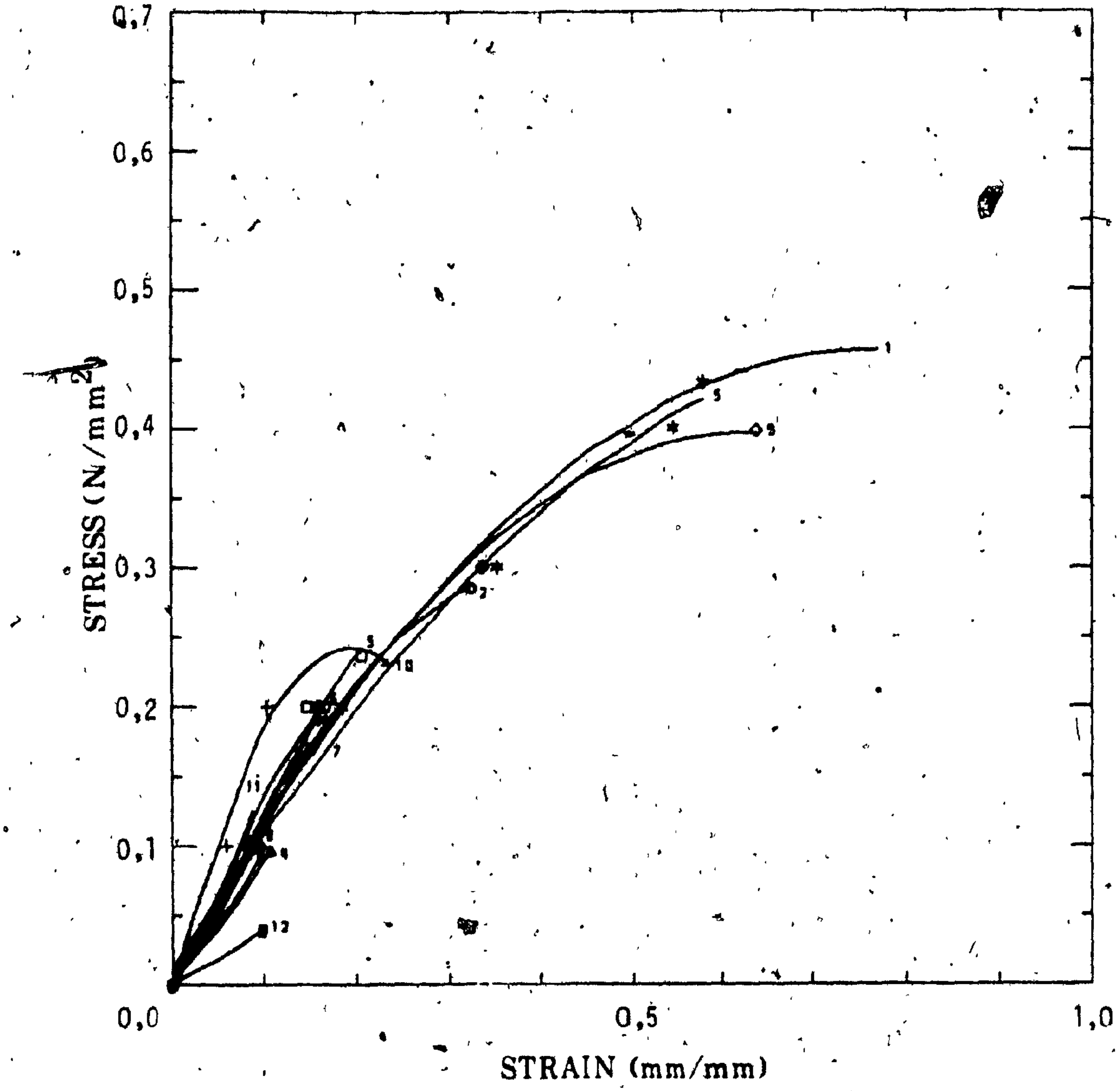

Figure 53 Stress-strain curves obtained with PDS-K1411 polyblends on a wood substrate. Curve 1-control 0% K1411; curve 2-control 5% K1411; curve 3-control 10% K1411; curve 4-control 15% K1411; curve 5-A.W. 0% K1411; curve 6-A.W. 5% K1411; curve 7-A.W. 10% K1411; curve 8-A.W. 15% K1411; curve 9-N.W. 0% K1411; curve 10-N.W. 5% K1411; curve 11-N.W. 10% K1411; curve 12-N.W. 15% K1411

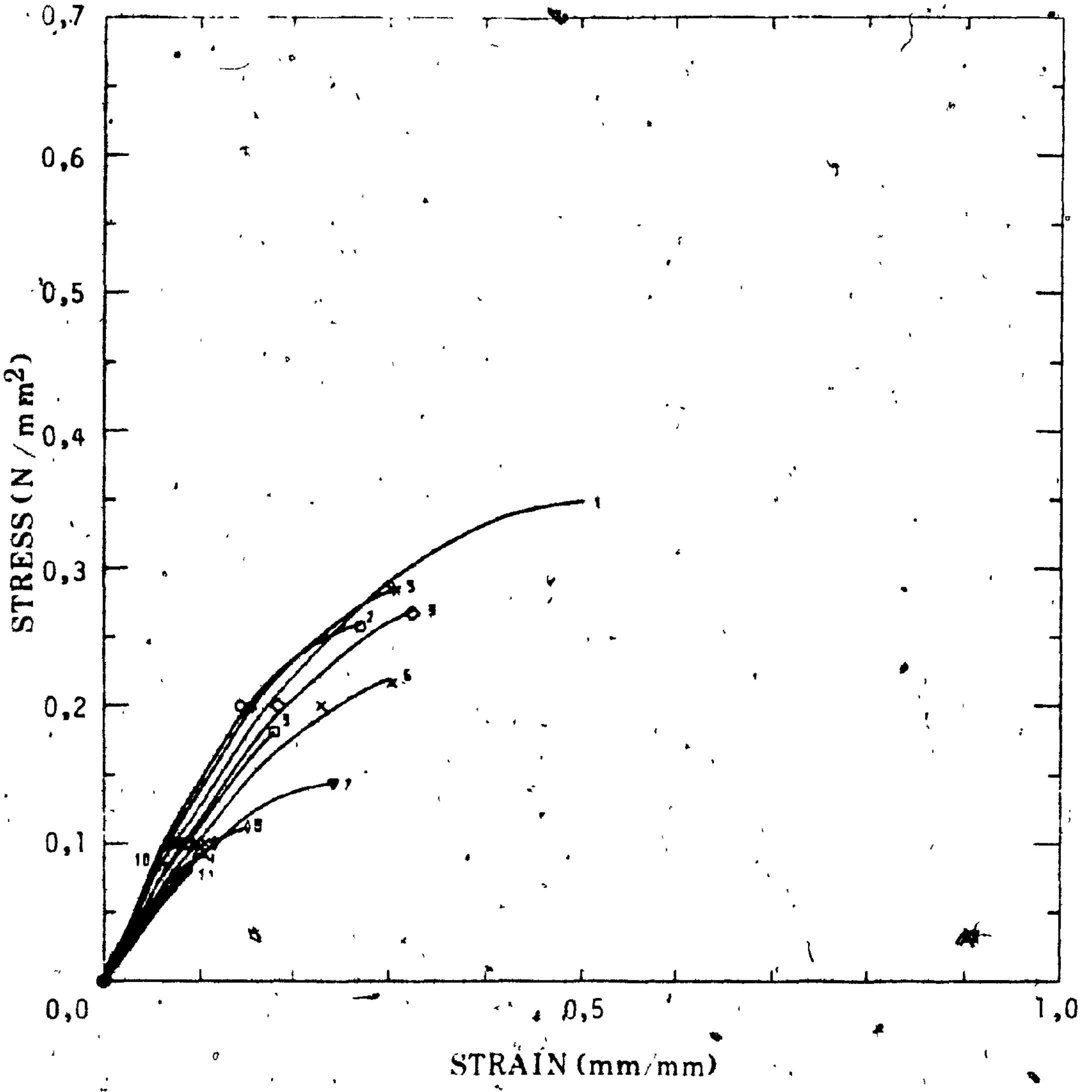

Figure 54 Stress-strain curves obtained with PDS-K1411 polyblends on a mortar substrate. Curve 1-control 0% K1411; curve 2-control 5% K1411; curve 3-control 10% K1411; curve 4-control 15% K1411; curve 5-A.W. 0% K1411; curve 6-A.W. 5% K1411; curve 7-A.W. 10% K1411; curve 8-A.W. 15% K1411; curve 9-N.W. 0% K1411; curve 10-N.W. 5% K1411; curve 11-N.W. 10% K1411

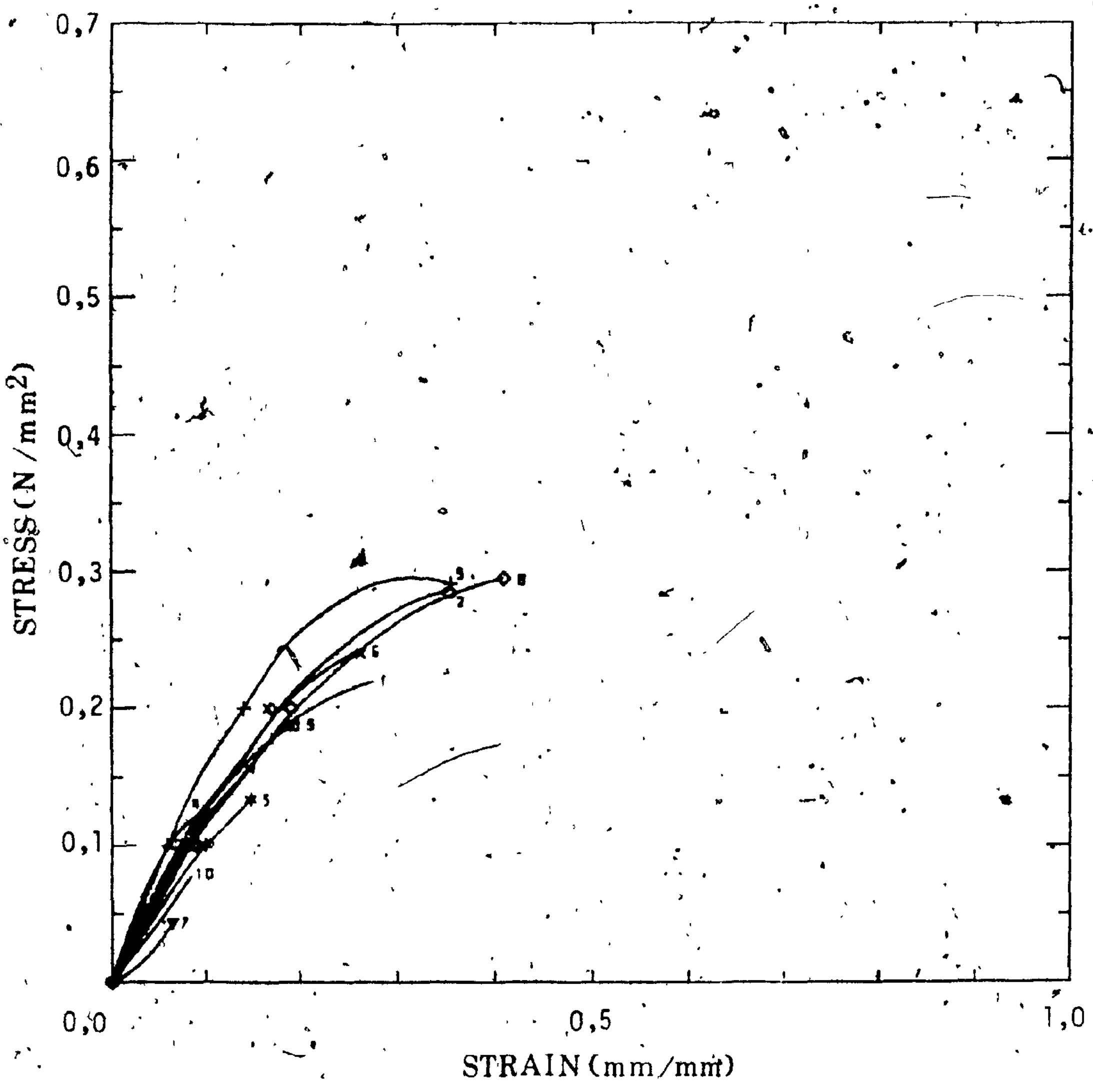

Figure 55 Stress-strain curves obtained with PDS-K1411 polyblends on an aluminum substrate. Curve 1-control 0% K1411; curve 2-control 5% K1411; curve 3-control 10% K1411; curve 4-control 15% K1411; curve 5-A.W. 0% K1411; curve 6-A.W. 5% K1411; curve 7-A.W. 10% K1411; curve 8-N.W. 0% K1411; curve 9-N.W. 5% K1411; curve 10-N.W. 10% K1411

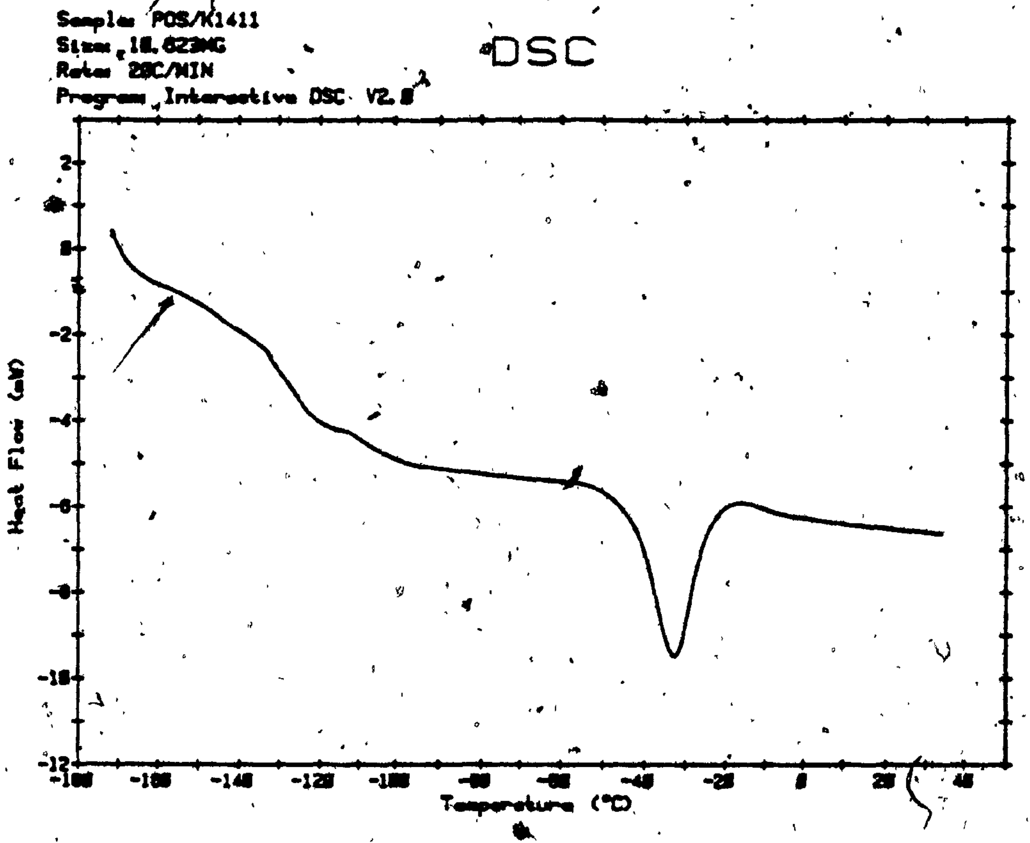

Figure 56 DSC scan (-170°C to +50°C) of PDS-K1411

8.6.3 PDS-K211 Polyblends

Unlike the two nitrile elastomers previously discussed, K211 contains approximately 3% carboxylic polar groups. Based on possible chemical interactions with wood or mortar substrates, important property modifications were expected with K211 (being a carboxylated copolymer), but results obtained with PDS-K211 polyblends were inconsistent through the three conditions of exposure. Several improvements were observed with the control and A.W. specimens only (figs. 57-59). The stress-strain characteristics of the N.W. specimens were poor, exhibiting no improvement (and in most cases a marked decrease) over PDS specimens.

The 5 and 10% polyblend specimens with aluminum substrates exposed to control and A.W. conditions (fig. 58) indicated an increase in toughness over PDS (table 8), as did the 5% polyblend specimens on a mortar substrate (fig. 59). Additionally, the control 15% polyblend specimens on an aluminum substrate showed an improvement in toughness over PDS specimens.

Since only the N.W. specimens were consistently weaker than the control and A.W. specimens, factors within the N.W. environment not accounted for with A.W., such as higher humidity variation, stronger U.V. irradiation, wind factors, pollution, etc., contributed to the decrease in mechanical properties, thus diminishing durability.

DSC testing of K211 resulted in a Tg of -32°C. Similar to the cases of the polyblends with K1402 and K1411, the PDS-K211 polyblends tested resulted in only one Tg at -113,2oC (fig. 60), which corresponds to the Tg of PDS. K211 is also a polar polymer, therefore it can be concluded that PDS-K211 is not a miscible polyblend.

SPECIMEN	SUBSTRATE	TOUGHNESS $(N/mm^2.mm/mm)$		
		Exposure Conditions		
		Control	Accelerated	Natural
PDS	Wood	0,23	0,14	0,17
PDS	Aluminum	0,04	0,01	0,08
PDS	Mortar	0,12	0,05	0,05
PDS-K211(5%)	Wood	0,03	0,02	0,00
PDS-K211(5%)	Aluminum	0,06	0,06	0,01
PDS-K211(5%)	Mortar	0,32	0,15	0,00
PDS-K211(10%)	Wood	0,03	0,01	0,00
PDS-K211(10%)	Aluminum	0,06	0,04	0,03
PDS-K211(10%)	Mortar	0,11	0,03	0,02
PDS-K211(15%)	Wood	0,06	0,05	0,05
PDS-K211(15%)	Aluminum	0,03	0,03	0,03
PDS-K211(15%)	Mortar	0,28	0,04	0,05

Table 8. Toughness of PDS-K211 polyblends

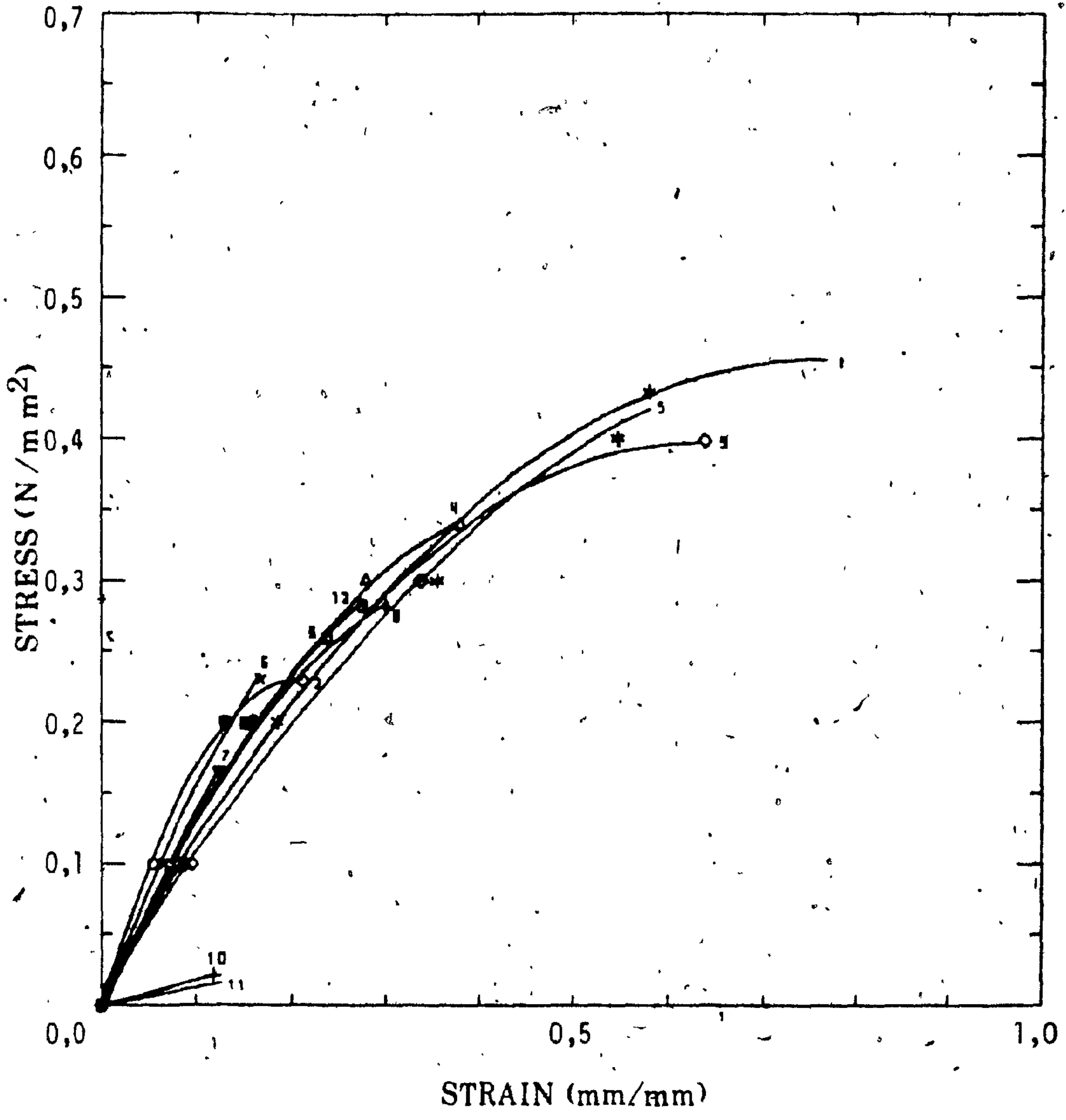

Figure 57 Stress-strain curves obtained with PDS-K211 polyblends on a wood substrate. Curve 1-control 0% K211; curve 2-control 5% K211; curve 3-control 10% K211; curve 4-control 15% K211; curve 5-A.W. 0% K211; curve 6-A.W. 5% K211; curve 7-A.W. 10% K211; curve 8-A.W. 15% K211; curve 9-N.W. 0% K211; curve 10-N.W. 5% K211; curve 11-N.W. 10% K211; curve 12-N.W. 15% K211 .

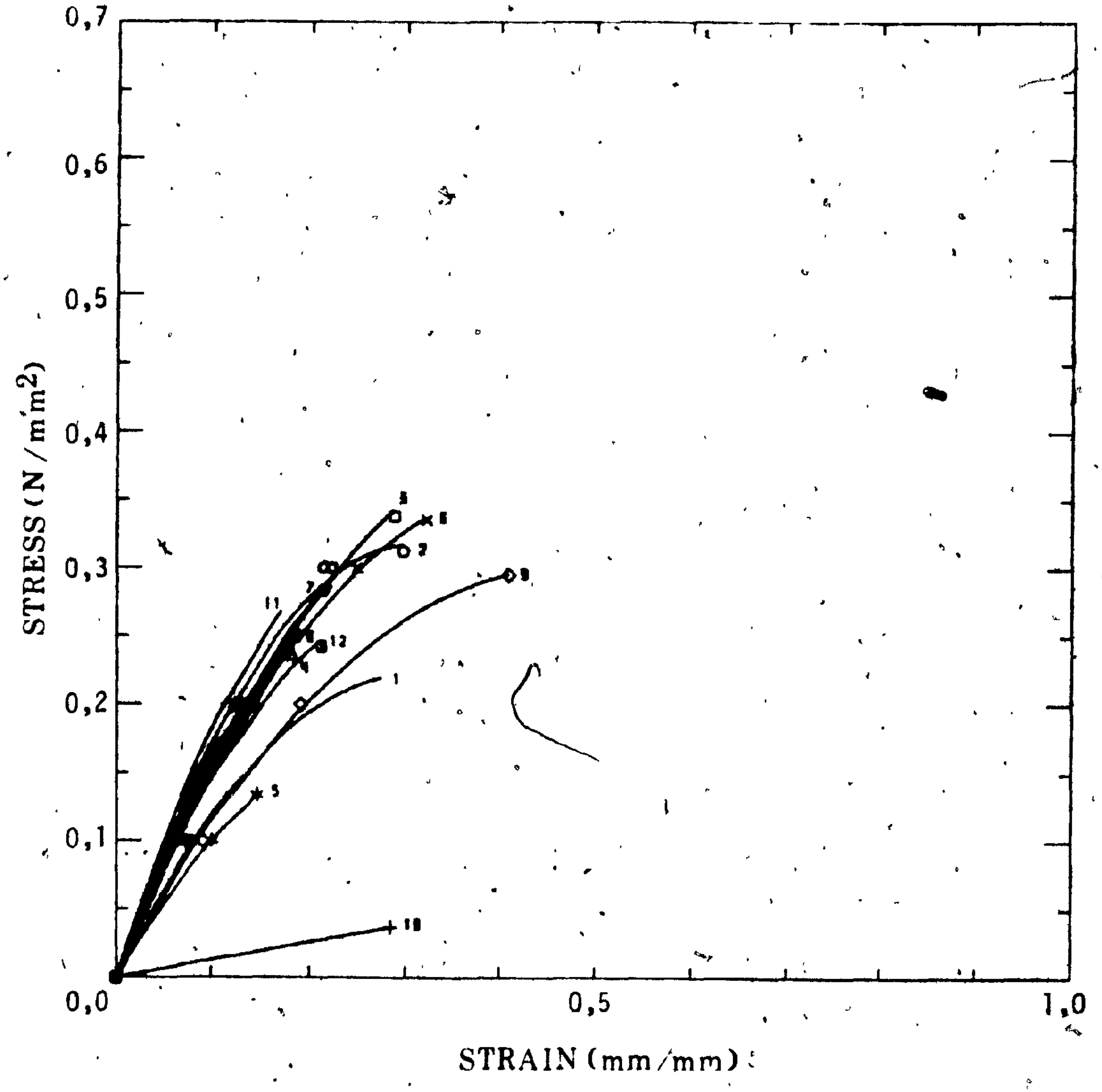

Figure 58 Stress-strain curves obtained with PDS-K211 polyblends on an aluminum substrate. Curve 1-control 0% K211; curve 2-control 5% K211; curve 3-control 10% K211; curve 4-control 15% K211; curve 5-A.W. 0% K211; curve 6-A.W. 5% K211; curve 7-A.W. 10% K211; curve 8-A.W. 15% K211; curve 9-N.W. 0% K211; curve 10-N.W. 5% K211; curve 11-N.W. 10% K211; curve 12-N.W. 15% K211

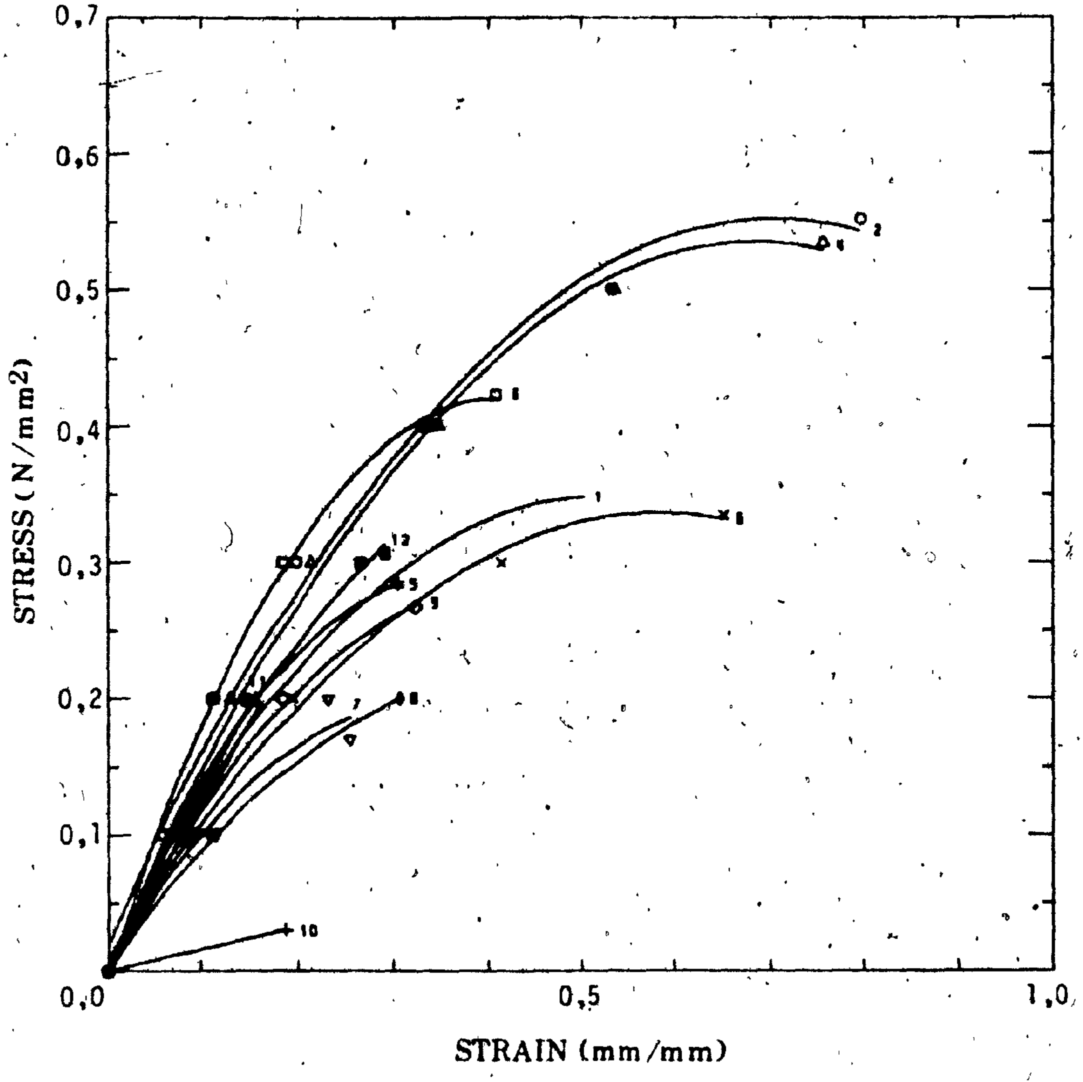

Figure 59 Stress-strain curves obtained with PDS-K211 polyblends on a mortar substrate. Curve 1-control 0% K211; curve 2-control 5% K211; curve 3-control 10% K211; curve 4-control 15% K211; curve 5-A.W. 0% K211; curve 6-A.W. 5% K211; curve 7-A.W. 10% K211; curve 8-A.W. 15% K211; curve 9-N.W. 0% K211; curve 10-N.W. 5% K211; curve 11-N.W. 10% K211; curve 12-N.W. 15% K211

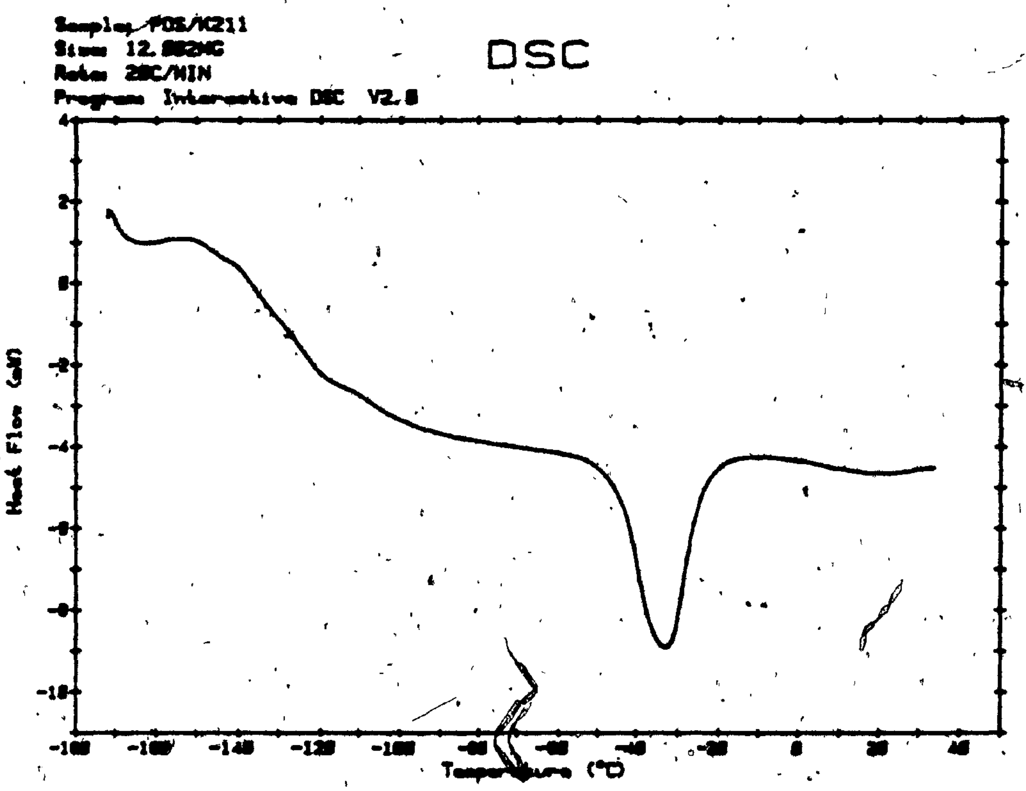

Figure 60 DSC scan (-170°C to +50°C) of PDS-K211

8.6.4 Discussion and Conclusions

DSC results indicate that the three polyblends, PDS-K1402, PDS-K1411, and PDS-K211, are not miscible. Thus their mechanical properties are influnced by the adhesion between the two phases co-existing within the polyblend.

The only silicone - nitrile elastomer polyblend that exhibited consistently better testing results than PDS was the non-carboxylated elastomer, PDS-K1402, with a medium ACN content, on an aluminum substrate. The polyblend with the 5% proportion of K1402 gave the biggest increase in toughness over PDS on the same substrate.

The increase in the amount of ACN in the nitrile elastomers was detrimental to the properties of the experimented polyblends, as was the presence of carboxylated comonomers.

Some better results in the case of the carboxylated copolymer (K211) with a mortar substrate might be attributed to chemical interactions between K211 and alkaline components of Portland cement mortar.

The majority of failures of the 5% nitrile elastomer polyblends with aluminum substrates were cohesive, which suggests that the use of a primer would not improve their performance. Since failures of the rest of these polyblends were generally adhesive, the use of a primer could prove beneficial in improving their durability (a measure of material performance within its service conditions).

8.7 Silicone - Lignin Polyblends

8.7.1 PDS-Lignin Polyblends

All PDS-lignin polyblend specimens were less tough than the corresponding PDS specimens (fig. 61-63), with two exceptions. The 5% lignin polyblend on a mortar substrate (fig. 63) was almost twice as tough as the PDS specimen on the same substrate (table 9). After A.W., the toughness of the 5% lignin polyblend decreased to two thirds the toughness of PDS, and to less than one half the PDS toughness after N.W.

The second exception was that of the 5% lignin polyblend on an aluminum substrate (fig. 62). Both the control and A.W. specimens were very slightly tougher than their corresponding PDS specimens. After N.W., however, the toughness value for the 5% lignin polyblend decreased to 20% of the PDS toughness.

DSC testing of lignin resulted in a Tg of 150°C. Testing of the PDS-lignin polyblends resulted in a Tg of -116°C (fig. 64), a slightly depressed Tg from that of PDS, but there was no observed Tg corresponding to the Tg of lignin (fig. 65). Since there was no single Tg between the Tg's of the constituent polymers, and only one which corresponded to the Tg of PDS, it can be concluded that PDS-lignin is a two-phase polyblend.

SPECIMEN	SUBSTRATE	TOUGHNESS $(N/mm^2.mm/mm)$		
		Exposure Conditions		
		Control	Accelerated	Natural
PDS	Wood	0,23	0,14	0,17
PDS	Aluminum	0,04	0,01	0,08
PDS	Mortar	0,12	0,05	0,05
PDS-lignin(5%)	Wood	0,04	0,01	0,01
PDS-lignin(5%)	Aluminum	0,04	0,01	0,01
PDS-lignin(5%)	Mortar	0,22	0,04	0,02
PDS-lignin(10%)	Wood	0,02	0,01	0,03
PDS-lignin(10%)	Aluminum	0,01	0,01	0,01
PDS-lignin(10%)	Mortar	0,09	0,04	0,02
PDS-lignin(15%)	Wood	0,02	0,02	0,01
PDS-lignin(15%)	Aluminum	0,01	0,01	0,02
PDS-lignin(15%)	Mortar	0,07	0,03	0,00

Table 9 Toughness of PDS-lignin polyblends

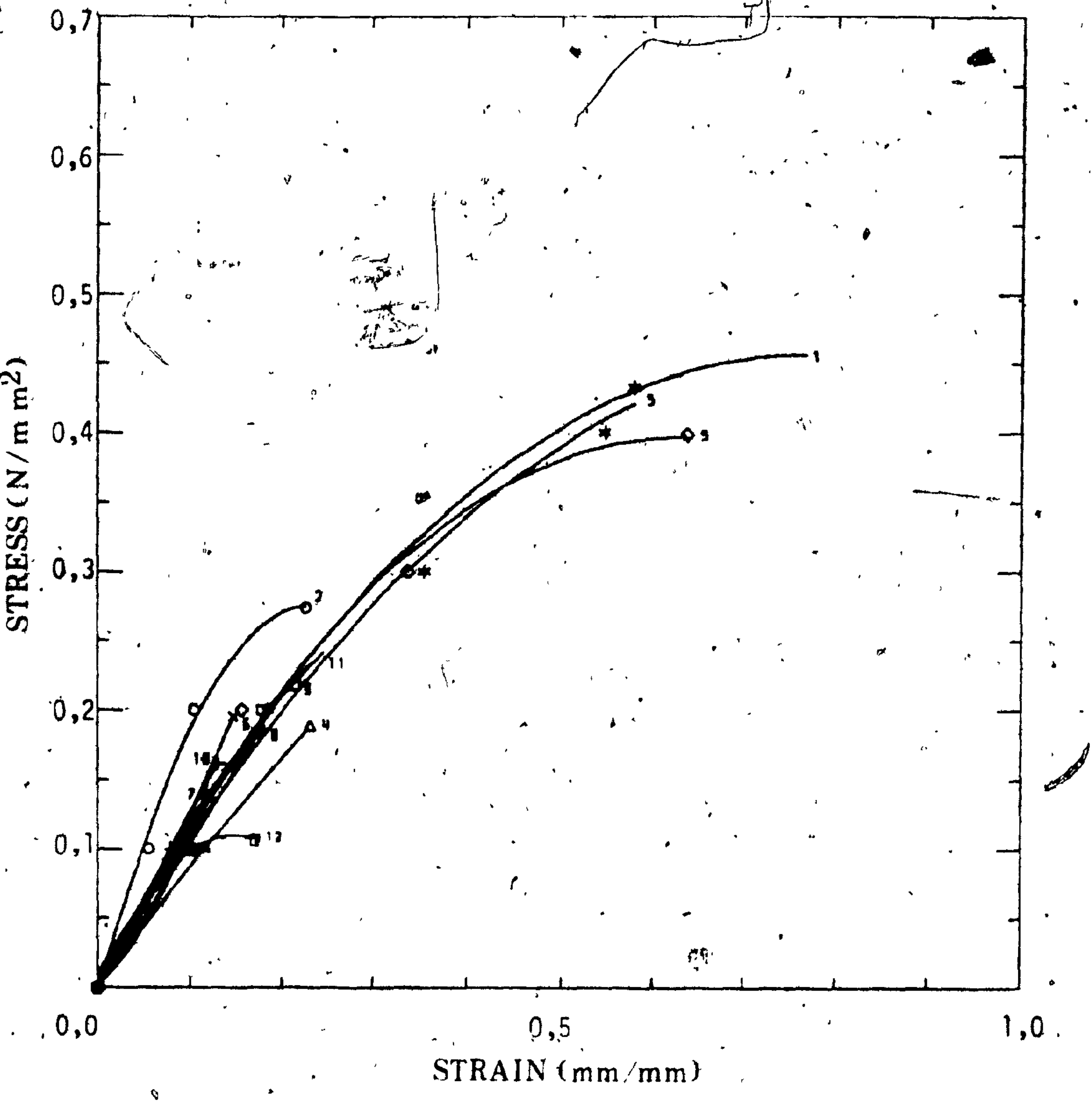

Figure 61 Stress-strain curves obtained with PDS-lignin polyblends on a wood substrate. Curve 1-control 0% lignin; curve 2-control 5% lignin; curve 3-control 10% lignin; curve 4-control 15% lignin; curve 5-A.W. 0% lignin; curve 6-A.W. 5% lignin; curve 7-A.W. 10% lignin; curve 8-A.W. 15% lignin; curve 9-N.W. 0% lignin; curve 10-N.W. 5% lignin; curve 11-N.W. 10% lignin; curve 12-N.W. 15% lignin

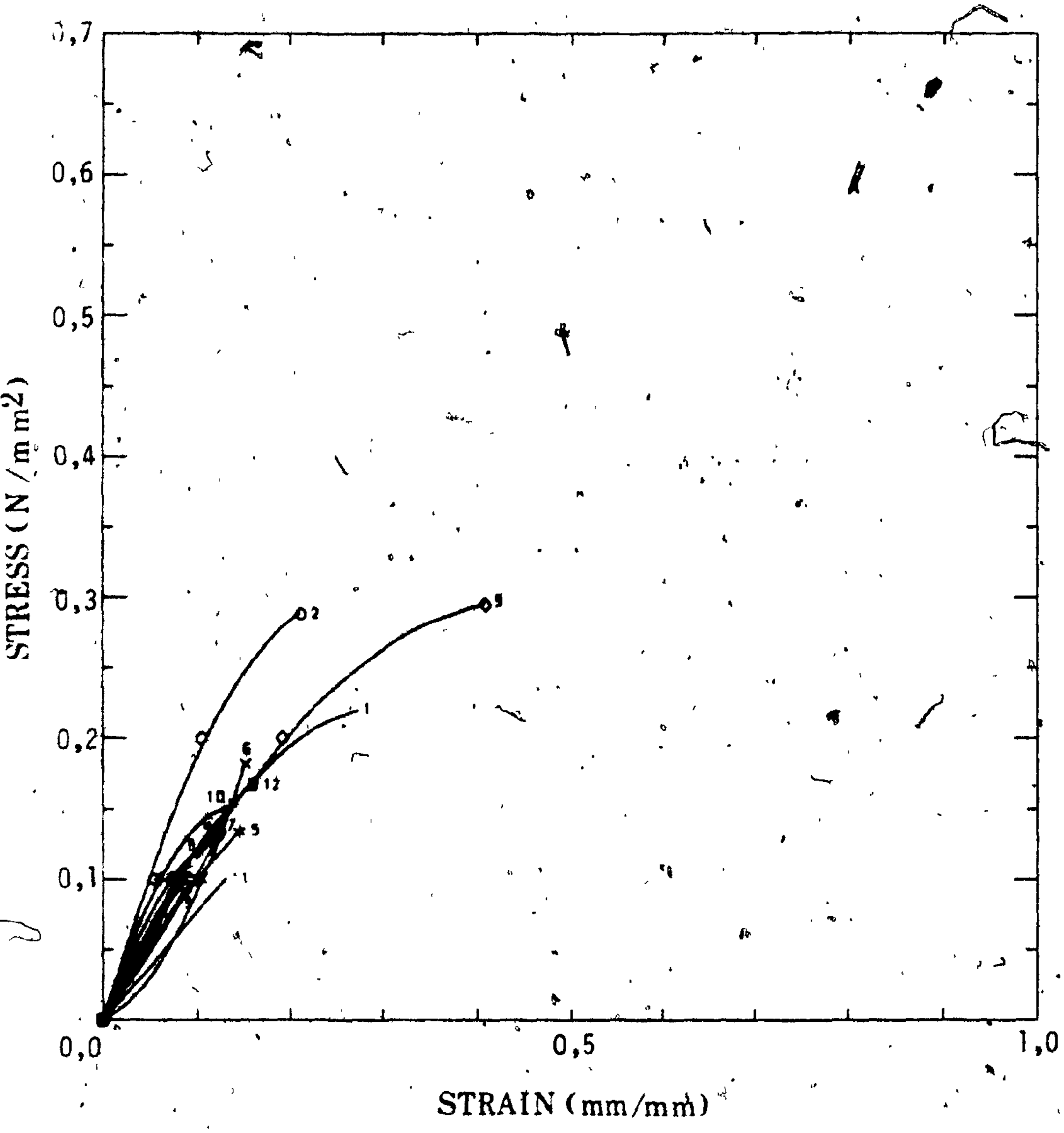

Figure 62 Stress-strain curves obtained with PDS-lignin polyblends on an aluminum substrate. Curve 1-control 0% lignin; curve 2-control 5% lignin; curve 3-control 10% lignin; curve 4-control 15% lignin; curve 5-A.W. 0% lignin; curve 6-A.W. 5% lignin; curve 7-A.W. 10% lignin; curve 8-A.W. 15% lignin; curve 9-N.W. 0% lignin; curve 10-N.W. 5% lignin; curve 11-N.W. 10% lignin; curve 12-N.W. 15% lignin

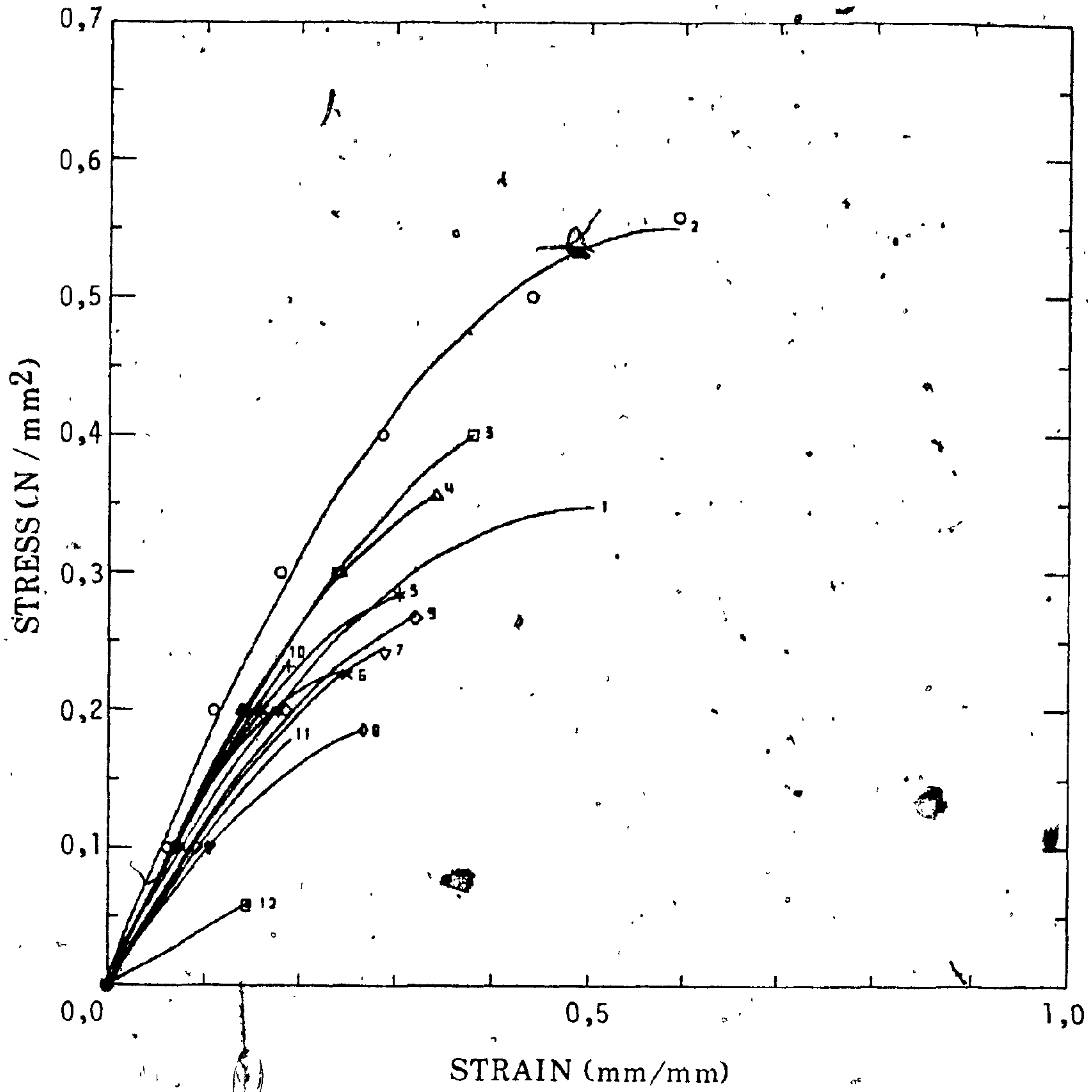

Figure 63 Stress-strain curves obtained with PDS-lignin polyblends on a mortar substrate. Curve 1-control 0% lignin; curve 2-control 5% lignin; curve 3-control 10% lignin; curve 4-control 15% lignin; curve 5-A.W. 0% lignin; curve 6-A.W. 5% lignin; curve 7-A.W. 10% lignin; curve 8-A.W. 15% lignin; curve 9-N.W. 0% lignin; curve 10-N.W. 5% lignin; curve 11-N.W. 10% lignin; curve 12-N.W. 15% lignin

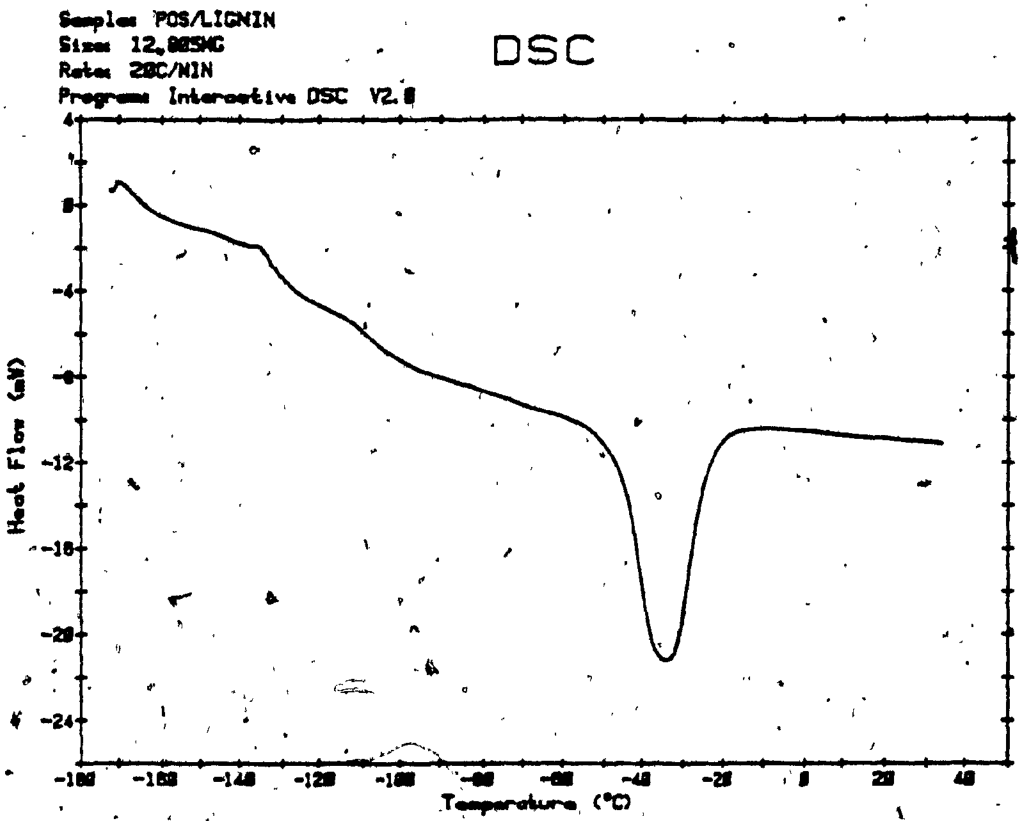

Figure 64 DSC scan (-170°C to +50°C) of PDS-lignin

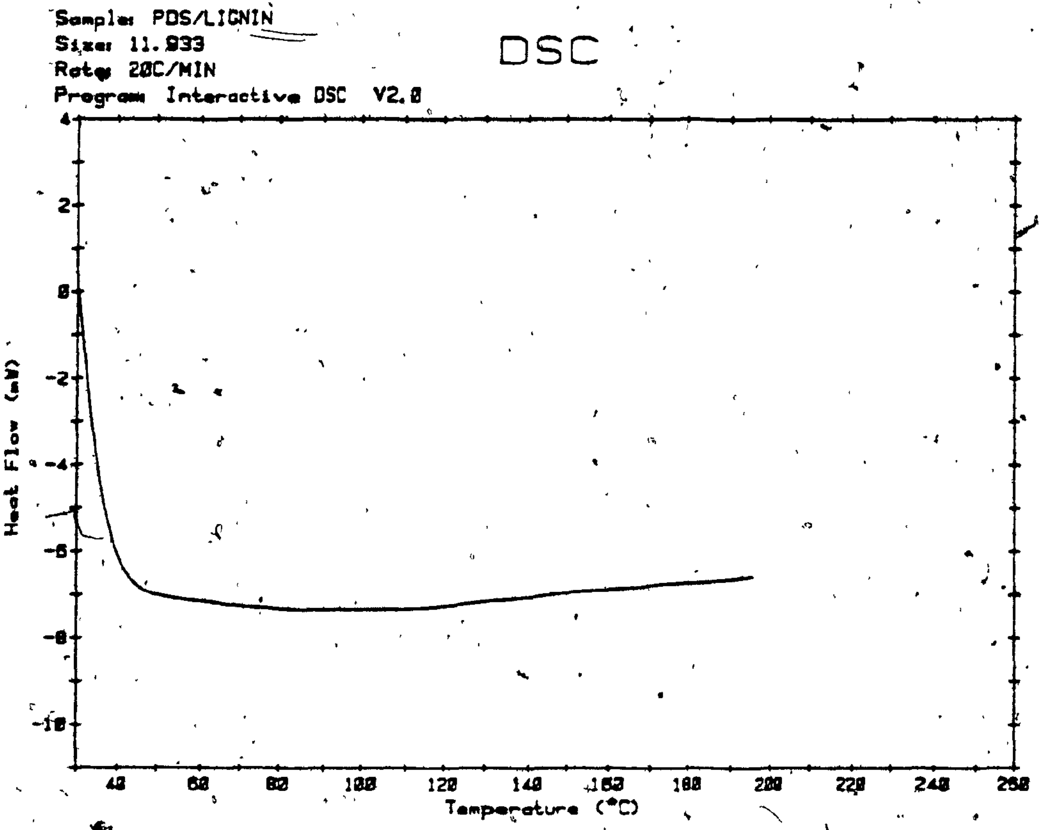

Figure 65 DSC scan (+30°C to +200°C) of PDS-lignin

8.7.2 Discussion and Conclusions

DSC results indicate that PDS-lignin polyblends are not miscible, and therefore their mechanical properties are highly influenced by the degree of interfacial adhesion between the two phases.

Mechanical testing results and toughness determinations were inconsistent through the three differently conditioned specimens. All failures were adhesive, thus the exposure factors also affected the adhesion of the sealant bead to the substrate. We can therefore conclude that the addition of lignin in the percentages used in this study (5, 10, and 15) does not improve the properties of silicone sealants. Lignin can not be recommended as a polymer to be polyblended with PDS, as after weathering the mechanical properties (and thus durability) of the polyblend are reduced to insignificance.

9. CONCLUSIONS AND RECOMMENDATIONS

All of the polyblends prepared for this study were heterogeneous (two-phase). This was confirmed by SEM determinations of morphological structure and DSC measurements of glass transition temperatures. The presence of these two distinct phases (matrix and dispersed) indicated that mechanical properties and durability of the polyblends were influenced by the interfacial adhesion between the phases. Although all of the studied polyblends were incompatible, no significant phase separation was observed in the specimens.

SEM observations of morphology showed that the PDS polyblends that exhibited superior performance (stress-strain and toughness) over PDS also had good adhesion between the phases. The lack of flaws or gaps at the phase interfaces served as evidence of these observations.

Mechanical testing results of the accelerated and naturally weathered PDS-vinyl polyblends were quite similar, while results with the weathered nitrile elastomers indicated that these polyblends were not as durable under natural weathering conditions. This can be attributed to the fact that exposure conditions in the weathering chamber do not exactly duplicate those of a natural environment. Factors such as pollution, rate of cycling, moisture conditions, variation in U.V. radiation, etc., cannot be accounted for within the chamber conditions. These factors can attack and decrease the durability of polymers, as must have been the case with the nitrile elastomers.

Of all polyblend groups studied, the PDS blends with vinyl polymers had the best test results. PDS-PVC polyblends (10% PVC by weight, specifically) with a wood substrate improved the mechanical performance and durability of PDS considerably. As failures of these polyblends were cohesive, any future investigations should concentrate on the enhancement of cohesion within the polyblend.

The PDS-VC/VAc (15%) polyblends specimens with aluminum substrates substantially increased the toughness of PDS. This type of polyblend exhibited the greatest toughness over all the specimens prepared and tested in this study. Failures were adhesive, thus additional improvement of durability may be possible with the use of an effective primer. Future research should investigate this aspect as this polyblend is potentially a viable alternative for silicone sealants within the construction industry.

The PDS-nitrile elastomer polyblends were promising under control and accelerated weathering conditions, but the inconsistency of results with naturally weathered specimens indicates that further research should be done to determine which natural weathering factors are the most detrimental. Once this is known, solutions to this problem can be further investigated through modification of the polyblend.

PDS-lignin polyblends are not recommended for any sealing applications, as their adhesive properties are much lower than that of PDS. The use of a primer could possibly be beneficial to the adhesion of this type of polyblend, and may warrant further study as lignin is a plentiful by-product in the Canadian pulp and paper industry.

10. REFERENCES

1- Aggarwal, S.L. (Ed.) 'Block Polymers', Plenum Press - 1970

2- Blaga, A., Beznaczuk, L.M., & Feldman, D. Poly(dimethylsiloxane) Sealant with Improved Properties by Blending with Vinyl Polymers, Journal of Applied Polymer Science, in print

3- Bruins, Paul F. (Ed.) 'Silicone Technology', Applied Polymer Symposia No. 14, John Wiley & Sons Inc. - 1970

4- Cook, John Philip 'Construction Sealants & Adhesives', John Wiley & Sons Inc. - 1970

5- Damusis, Adolfas (Ed.) 'Sealants', Reinhold Publishing Corp. - 1967

6- Division of Building Research, 'Building Science Insight '84', National Research Council of Canada Conference Proceedings, 1985, in print

7- Evans, Robert M. Evolution of Sealant Durability Testing, Adhesives Age, Vol. 23, No. 12, Nov. 1980, p. 31

8- Feldman, D. Natural and Accelerated Weathering of Some Polyblends, Journal of Applied Polymer Science, Vol. 26, 1981, p. 3493

9- Feldman, D. Modifications of the Properties of Polyurethane by Blending, Reinforcing, or Plasticizing, Journal of Applied Polymer Science, Vol. 27, 1982, p. 1933

10- Grassie, N. (Ed.) 'Developments in Polymer Degradation - 1', Applied Science Publishers Ltd. - 1977

11- Kamal, Musa R. (Ed.) 'Weatherability of Plastic Materials', Journal of Applied Polymer Science Applied Polymer Symposia No. 4, John Wiley & Sons Inc. - 1967

12- Karpati, K. K. Mechanical Properties of Sealants: I. Behaviour of Silicone Sealants as a Function of Temperature, Division of Building Research Research Paper No. 515, National Research Council of Canada, February 1972

13- Karpati, K. K. Mechanical Properties of Sealants: II. Behaviour of Silicone Sealants as a Function of Rate of Movement, Division of Building Research Research Paper No. 524, National Research Council of Canada, June 1972

14- Karpati, K. K. Mechanical Properties of Sealants: III. Performance Testing of Silicone Sealants, Division of Building Research Research Paper No. 533, National Research Council of Canada, September 1972

15- Karpati, K. K. New Testing Methods for Sealants, Durability of Building Materials and Components, ASTM STP 691, P.J. Sereda and G.G. Litvan (Eds.), American Society for Testing and Materials, 1980 p. 658

16- Kinloch, A. J. 'Durability of Sructural Adhesives', Applied Science Publishers Ltd. - 1983

17- Klempner, Daniel & Frisch, Kurt C. (Ed.) 'Polymer Alloys III: Blends, Blocks, Grafts & Interpenetrating Networks', Polymer Science & Technology Vol. 20, Plenum Press - 1983

18- Krause, Sonja Polymer-Polymer Compatibility, edited by D.R. Paul, 'Polymer Blends' Vol. 1, Academic Press - 1978

19- Lin, Stephen Y. Lignin Utilization: Potential & Challenge, Progress in Biomass Conversion, Vol. 4, edited by David A. Tillman & Edwin C. Jahn, Academic Press - 1983

20- Maglio, G. & Palumbo, R. The Role of Interfacial Agents in Polymer Blends, Polymer Blends, 1984, p. 41

21- Manson, John A. & Sperling, Leslie H. 'Polymer Blends and Composites', Plenum Press - 1976

22- Murayama, Takayuki 'Dynamic Mechanical Analysis of Polymeric Material', Materials Science Monographs 1, Elsevier Scientific Publishing Co. (Netherlands) - 1978

23- Noshay, Allen & McGrath, James E. 'Block Copolymers: Overview and Critical Survey', Academic Press Inc. - 1977

24- Olabisi, Olagoke , Robeson, Lloyd M. & Shaw, Montgomery 'Polymer-Polymer Miscibility', Academic Press Inc. - 1979

25- Perkins, R. B. & Glarum, S. N. 'Adhesives Sealants and Gaskets - A Survey', Office of Technology Utilization, NASA - 1967

26- Ranganathan, B. N. Differential Scanning Calorimetry of Room Temperature Vulcanizing Silicones, SAMPE Quarterly, Vol. 15, No. 2, January 1984

27- Sereda, P. J. & Litvan, G. G. (Ed.) 'Durability of Building Materials and Components', ASTM STP 691, American Society for Testing and Materials - 1980

28- Shah, Vishu 'Handbook of Plastics Testing Technology', John Wiley & Sons Inc. - 1984

29- Sharman, W. R., Fry, J. I. & Whitney, R. S. Six Years Natural Weathering of Sealants, Durability of Building Materials, 1983, No. 2, p. 79

30- Siegmann, A. Crystalline/Crystalline Polymer Blends: Some Structure-Property Relationships, Journal of Applied Polymer Science, Vol. 24, 1979, p. 2333

31- Skeist, Irving (Ed.) 'Plastics in Building', Reinhold Publishing Corp. - 1966

32- Van der Maas, J.H. 'Basic Infrared Spectroscopy', Heyden & Son Ltd. - 1969

11. APPENDIX

11.1 STRESS-STRAIN DATA AT FAILURE

Tension testing results of control samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm2)	Strain (mm/mm)	Toughness (N/mm2.mm/mm)
PDS			
wood substrate	0,45	0,768	0,23
Aluminum substrate	0,27 *	0,195 *	0,04
Mortar substrate	0,35	0,413 *	0,12
5% PVC/PDS polyblend			
Wood substrate	0,13	0,472 *	0,04
Aluminum substrate	0,09 *	0,176 *	0,01
Mortar substrate	0,32 *	0,200 *	0,04
10% PVC/PDS polyblend			
Wood substrate	0,57	0,686	0,26
Mortar substrate	0,47	0,600	0,23
15% PVC/PDS polyblend			
Wood substrate	0,44	0,387 *	0,12
Mortar substrate	0,46	0,413 *	0,12

STRESS-STRAIN DATA AT FAILURE

Tension testing results of control samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
5% (VC/VAc)/PDS polyblend			
Wood substrate	0,42	0,613 *	0,14
Aluminum substrate	0,37	0,355 *	0,08
Mortar substrate	0,44	0,517 *	0,14
10% (VC/VAc)/PDS polyblend			
Wood substrate	0,41	0,316 *	0,09
Aluminum substrate	0,48	0,800 *	0,19
Mortar substrate	0,52	0,685 *	0,18
15% (VC/VAc)/PDS polyblend			
Wood substrate	0,57	0,512 *	0,18
Aluminum substrate	0,67	0,498	0,18
Mortar substrate	0,59	0,622 *	0,18

STRESS-STRAIN DATA AT FAILURE

Tensile test results of control samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
5% K1402/PDS polyblend			
Wood substrate	0,38	0,345 *	0,15
Aluminum substrate	0,4[illegible]	0,797 *	0,19
Mortar substrate	0,36	0,379 *	0,13
10% K1402/PDS polyblend			
Wood substrate	0,50	1,010 *	0,32
Aluminum substrate	0,32	0,346 *	0,06
Mortar substrate	0,55	0,986	0,39
15% K1402/PDS polyblend			
Wood substrate	0,40 *	0,412 *	0,15
Aluminum substrate	0,30	0,274	0,05
Mortar substrate	0,33	0,302	0,06

STRESS-STRAIN DATA AT FAILURE

Tension testing results of control samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
5% K1411/PDS polyblend			
Wood substrate	0,25 *	0,316 *	0,04
Aluminum substrate	0,28	0,350	0,06
Mortar substrate	0,25	0,269	0,05
10% K1411/PDS polyblend			
Wood substrate	0,24 *	0,160 *	0,03
Aluminum substrate	0,18	0,192	0,02
Mortar substrate	0,16 *	0,162 *	0,02
15% K1411/PDS polyblend			
Wood substrate	0,10 *	0,107	0,01
Aluminum substrate	0,10 *	0,090 *	0,01
Mortar substrate	0,10 *	0,088 *	0,01

STRESS-STRAIN DATA AT FAILURE

Tension testing results of control samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
5% K211/PDS polyblend			
Wood substrate	0,16 *	0,133 *	0,03
Aluminum substrate	0,33 *	0,262 *	0,06
Mortar substrate	0,54	0,794	0,32
10% K211/PDS polyblend			
Wood substrate	0,25	0,235	0,03
Aluminum substrate	0,33	0,286	0,06
Mortar substrate	0,41	0,340 *	0,11
15% K211/PDS polyblend			
Wood substrate	0,33	0,378	0,06
Aluminum substrate	0,23	0,181	0,03
Mortar substrate	0,52	0,754	0,28

STRESS-STRAIN DATA AT FAILURE

Tension testing results of control samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
5% Lignin/PDS polyblend			
Wood substrate	0,27	0,226	0,04
Aluminum substrate	0,28	0,211	0,04
Mortar substrate	0,55	0,534 *	0,22
10% Lignin/PDS polyblend			
Wood substrate	0,21	0,214	0,02
Aluminum substrate	0,08 *	0,088	0,01
Mortar substrate	0,40	0,376	0,09
15% Lignin/PDS polyblend			
Wood substrate	0,20 *	0,200 *	0,02
Aluminum substrate	0,13	0,115	0,01
Mortar substrate	0,35	0,342	0,07

11.2 STRESS-STRAIN DATA AT FAILURE

Tension testing results after 400 cycles A.W.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness ($N/mm^2.mm/mm$)
PDS			
Wood substrate	0,36	0,572	0,14
Aluminum substrate	0,11 *	0,128 *	0,01
Mortar substrate	0,28	0,316	0,05
5% PVC/PDS polyblend			
Wood substrate	0,36	0,588	0,11
Aluminum substrate	0,21	0,180	0,02
Mortar substrate	0,22	0,165 *	0,04
10% PVC/PDS polyblend			
Wood substrate	0,54	0,712	0,25
Mortar substrate	0,34	0,213 *	0,06
15% PVC/PDS polyblend			
Wood substrate	0,46 *	0,603 *	0,11
Mortar substrate	0,21	0,148	0,02

STRESS-STRAIN DATA AT FAILURE

Tension testing results after 400 cycles A.W.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm²)	Strain (mm/mm)	Toughness (N/mm².mm/mm)
5% (VC/VAc)/PDS polyblend			
Wood substrate	0,30 *	0,245 *	0,05
Aluminum substrate	0,21 *	0,205 *	0,06
Mortar substrate	0,23	0,180	0,03
10% (VC/VAc)/PDS polyblend			
Wood substrate	0,33 *	0,181 *	0,05
Aluminum substrate	0,35	0,348	0,07
Mortar substrate	0,28	0,208	0,03
15% (VC/VAc)/PDS polyblend			
Wood substrate	0,50 *	0,291 *	0,05
Aluminum substrate	0,57	0,412	0,14
Mortar substrate	0,31 *	0,172	0,03

STRESS-STRAIN DATA AT FAILURE

Tension testing results after 400 cycles A.W.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
5% K1402/PDS polyblend			
Wood substrate	0,31 *	0,347 *	0,33
Aluminum substrate	0,42	0,682 *	0,19
Mortar substrate	0,52	0,924	0,32
10% K1402/PDS polyblend			
Wood substrate	0,42	0,664	0,18
Aluminum substrate	0,36	0,435 *	0,10
Mortar substrate	0,44 *	1,173 *	0,26
15% K1402/PDS polyblend			
Wood substrate	0,33	0,293 *	0,08
Aluminum substrate	0,30	0,294	0,08
Mortar substrate	0,20	0,152	0,01

STRESS STRAIN-DATA AT FAILURE

Tension testing results after 400 cycles A.W.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm2)	Strain (mm/mm)	Toughness (N/mm2.mm/mm)
5% K1411/PDS polyblend			
Wood substrate	0,19	0,184	0,01
Aluminum substrate	0,27 *	0,395 *	0,04
Mortar substrate	0,21	0,302	0,04
10% K1411/PDS polyblend			
Wood substrate	0,17	0,182	0,01
Aluminum substrate	0,07 *	0,118 *	0,00
Mortar substrate	0,17 *	0,240	0,02
15% K1411/PDS polyblend			
Wood substrate	0,08 *	0,104 *	0,01
Aluminum substrate	0,00	0,000	0,00
Mortar substrate	0,13 *	0,152	0,01

STRESS-STRAIN DATA AT FAILURE

Tension testing results after 400 cycles A.W.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
5% K211/PDS polyblend			
Wood substrate	0,23	0,184	0,02
Aluminum substrate	0,32	0,334	0,06
Mortar substrate	0,33	0,432 *	0,15
10% K211/PDS polyblend			
Wood substrate	0,14 *	0,120 *	0,01
Aluminum substrate	0,16	0,224	0,04
Mortar substrate	0,19 *	0,144 *	0,03
15% K211/PDS polyblend			
Wood substrate	0,28	0,389 *	0,05
Aluminum substrate	0,25	0,204	0,03
Mortar substrate	0,20	0,339 *	0,04

STRESS-STRAIN DATA AT FAILURE

Tension testing results after 400 cycles A.W.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
5% Lignin/PDS polyblend			
Wood substrate	0,19	0,158	0,01
Aluminum substrate	0,20 *	0,170	0,01
Mortar substrate	0,16 *	0,123 *	0,04
10% Lignin/PDS polyblend			
Wood substrate	0,13	0,152	0,01
Aluminum substrate	0,13	0,168	0,01
Mortar substrate	0,12 *	0,190 *	0,04
15% Lignin/PDS polyblend			
wood substrate	0,11 *	0,168 *	0,02
Aluminum substrate	0,09	0,130	0,01
Mortar Substrate	0,18 *	0,235 *	0,03

11.3 STRESS-STRAIN DATA AT FAILURE

Tension testing results of naturally weathered samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
PDS			
wood substrate	0,39	0,637	0,17
Aluminum substrate	0,33 *	0,533 *	0,08
Mortar substrate	0,26	0,322	0,05
5% PVC/PDS polyblend			
wood substrate	0,24 *	0,334 *	0,08
Aluminum substrate	0,31	0,352 *	0,09
Mortar substrate	0,31 *	0,312 *	0,10
10% PVC/PDS polyblend			
Wood substrate	0,51	0,621	0,20
Mortar substrate	0,34	0,230 *	0,05
15% PVC/PDS polyblend			
Wood substrate	0,47 *	0,608 *	0,12
Mortar substrate	0,24	0,136 *	0,02

STRESS-STRAIN DATA AT FAILURE

Tension testing results of naturally weathered samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
5% (VC/VAc)/PDS polyblend			
Wood substrate	0,30	0,227 *	0,06
Aluminum substrate	0,48	0,648 *	0,20
Mortar substrate	0,24	0,192	0,03
10% (VC/VAc)/PDS polyblend			
Wood substrate	0,33	0,259	0,04
Aluminum substrate	0,48	0,776 *	0,18
Mortar substrate	0,27 *	0,204 *	0,03
15% (VC/VAc)/PDS polyblend			
Wood substrate	0,42	0,286	0,07
Aluminum substrate	0,67	0,685	0,27
Mortar substrate	0,21 *	0,131	0,01

STRESS-STRAIN DATA AT FAILURE

Tension testing results of naturally weathered samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm²)	Strain (mm/mm)	Toughness (N/mm².mm/mm)
5% K1402/PDS polyblend			
Wood substrate	0,36	0,302	0,07
Aluminum substrate	0,48	0,622	0,20
Mortar substrate	0,18 *	0,165 *	0,02
10% K1402/PDS polyblend			
Wood substrate	0,39 *	0,595 *	0,12
Aluminum substrate	0,40	0,410	0,10
Mortar substrate	0,19	0,144	0,01
15% K1402/PDS polyblend			
Wood substrate	0,31	0,315 *	0,05
Aluminum substrate	0,41	0,366	0,09
Mortar substrate	0,15	0,102 *	0,01

STRESS-STRAIN DATA AT FAILURE

Tension testing results of naturally weathered samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm2)	Strain (mm/mm)	Toughness (N/mm2.mm/mm)
5% K1411/PDS polyblend			
Wood substrate	0,16 *	0,152 *	0,04
Aluminum substrate	0,29	0,352	0,07
Mortar substrate	0,10 *	0,051 *	0,00
10% K1411/PDS polyblend			
Wood substrate	0,11 *	0,093 *	0,01
Aluminum substrate	0,08	0,086	0,01
Mortar substrate	0,08	0,093	0,00
15% K1411/PDS polyblend			
Wood substrate	0,02 *	0,107 *	0,00
Aluminum substrate	0,00	0,000	0,00
Mortar substrate	0,00	0,000	0,00

STRESS-STRAIN DATA AT FAILURE

Tension testing results of naturally weathered samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm2)	Strain (mm/mm)	Toughness (N/mm2.mm/mm)
5% K211/PDS polyblend			
Wood substrate	0,02	0,117	0,00
Aluminum substrate	0,04	0,283	0,01
Mortar substrate	0,03	0,221 *	0,00
10% K211/PDS polyblend			
Wood substrate	0,02	0,125	0,00
Aluminum substrate	0,26	0,170	0,03
Mortar substrate	0,19	0,165	0,02
15% K211/PDS polyblend			
Wood substrate	0,28	0,238 *	0,05
Aluminum substrate	0,24	0,210	0,03
Mortar substrate	0,30	0,291	0,05

STRESS-STRAIN DATA AT FAILURE

Tension testing results of naturally weathered samples.

Note: Asterisk (*) indicates that less than 5 (never less than 3) samples were used to calculate the mean.

Sealant	Stress (N/mm^2)	Strain (mm/mm)	Toughness (N/mm^2.mm/mm)
5% Lignin/PDS polyblend			
Wood substrate	0,16	0,130	0,01
Aluminum substrate	0,15	0,134	0,01
Mortar substrate	0,23	0,187	0,02
10% Lignin/PDS polyblend			
Wood substrate	0,23	0,276 *	0,03
Aluminum substrate	0,12 *	0,130	0,01
Mortar substrate	0,18	0,189	0,02
15% Lignin/PDS polyblend			
Wood substrate	0,09 *	0,148 *	0,01
Aluminum substrate	0,16	0,158	0,02
Mortar substrate	0,04 *	0,144	0,00