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DEVELOPMENT OF CASTOR-OIL-RESISTANT POLYURETHANE SONAR
ENCAPSULANTS(U) NAVAL RESEARCH LAB WASHINGTON DC
W T MAURICE ET AL. 30 JUN 85 NRL-MR-5578

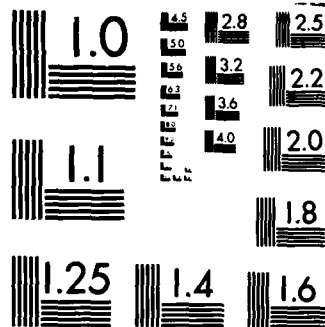
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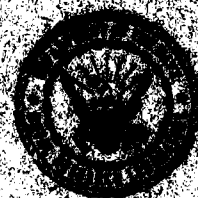
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Naval Research Laboratory

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Final Report

Contract No. N00019-67-1-0001



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SECURITY CLASSIFICATION OF THIS PAGE

AD A 156 576

REPORT DOCUMENTATION PAGE

1a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED			1b. RESTRICTIVE MARKINGS		
2a. SECURITY CLASSIFICATION AUTHORITY N/A			3. DISTRIBUTION / AVAILABILITY OF REPORT Approved for public release; distribution unlimited.		
2b. DECLASSIFICATION / DOWNGRADING SCHEDULE N/A					
4. PERFORMING ORGANIZATION REPORT NUMBER(S)			5. MONITORING ORGANIZATION REPORT NUMBER(S) NRL Memorandum Report No. 5578		
6a. NAME OF PERFORMING ORGANIZATION Products Research and Chemical Corporation		6b. OFFICE SYMBOL (If applicable)	7a. NAME OF MONITORING ORGANIZATION Naval Research Laboratory Underwater Sound Reference Detachment		
6c. ADDRESS (City, State, and ZIP Code) Box 1800 5426 San Fernando Road Glendale, CA 91203			7b. ADDRESS (City, State, and ZIP Code) P.O. Box 8337 Orlando, FL 32856-8337		
8a. NAME OF FUNDING / SPONSORING ORGANIZATION Naval Sea Systems Command		8b. OFFICE SYMBOL (If applicable) (SEA 63X5)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER Contract No. N00014-81-WR-2576		
8c. ADDRESS (City, State, and ZIP Code) Washington, DC 23062			10. SOURCE OF FUNDING NUMBERS		
			PROGRAM ELEMENT NO. 64503N	PROJECT NO. 1	TASK NO. S0219AS
			WORK UNIT ACCESSION NO. (59)0584		
11. TITLE (Include Security Classification) Development of Castor-Oil-Resistant Polyurethane Sonar Encapsulants					
12. PERSONAL AUTHOR(S) W.T. Maurice and C.M. Thompson					
13a. TYPE OF REPORT Final		13b. TIME COVERED FROM 9/81 TO 8/82		14. DATE OF REPORT (Year, Month, Day) JUNE 30, 1985	
15. PAGE COUNT 45					
16. SUPPLEMENTARY NOTATION This work was sponsored by the Sonar Transducer Reliability Improvement Program (STRIP) which is funded by the Naval Sea Systems Command.					
17. COSATI CODES			18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)		
FIELD 11	GROUP 07	SUB-GROUP -	Castor oil Polyurethane		
			Sonar transducer encapsulants. <i>Toluene Diisocyanate</i>		
19. ABSTRACT (Continue on reverse if necessary and identify by block number) Polyurethanes have been widely used as underwater acoustic encapsulants for over twenty years. This research was designed to produce a polyurethane encapsulant that is non-hazardous and non-proprietary for use in castor-oil-filled systems. Seven commercial diamines were evaluated as replacements for methylene bis(2-chloroaniline) (MBOCA) in the cure system. The polyurethanes produced were screened according to their handling properties, strength, modulus retention on water and castor oil exposure, electrical properties, and sound speed. The final candidates are commercial IDI/PIMG prepolymers cured with diamine/diol/catalyst system. The resulting polyurethanes exhibit better combined castor oil and water resistance than any commercially available material. <i>Originator supplied keywords include?</i> <i>(Polyoxytetramethylene glycol)</i>					
20. DISTRIBUTION / AVAILABILITY OF ABSTRACT ☐ UNCLASSIFIED/UNLIMITED ☒ SAME AS RPT. ☐ DTIC USERS			21. ABSTRACT SECURITY CLASSIFICATION UNCLASSIFIED		
22a. NAME OF RESPONSIBLE INDIVIDUAL C.M. Thompson			22b. TELEPHONE (Include Area Code) (305) 859-5120		22c. OFFICE SYMBOL NRL-USRD (Code 5975)

AUG 5 1985

DD FORM 1473, 84 MAR

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All other editions are obsolete.

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DEVELOPMENT OF CASTOR OIL RESISTANT POLYURETHANE SONAR ENCAPSULANTS

INTRODUCTION

A wide variety of elastomers such as neoprene, butyl, and polyurethanes are employed in the fabrication of underwater sonar devices. The sonar system must operate over wide ranges in temperature, pressure, and sound frequency in both salt and fresh water. In order to adequately perform this task, the sonar device must be encapsulated with some elastomeric material capable of protecting it not only from water ingress but also from potentially corrosive vapors and fluids, mechanical and vibrational shock, and abrasion.

Liquid polyurethanes which were originally developed as electrical potting and molding compounds have been successfully employed as sonar transducer encapsulants for over 20 years. In general, these materials have performed very well as the velocity of sound through the polyurethane is not too much greater than that through seawater. In some instances the sonar transducer may contain a fill fluid which is in long-term contact with the encapsulant. A potential problem arises when the encapsulant is a polyurethane and the fill fluid is castor oil. Most polyurethanes in service today do not exhibit the desired level of long-term castor oil resistance.

There has been concern over reported fleet failures of certain types of sonar transducer units. The polyurethanes exhibit several modes of failure. These are apparent encapsulant failures due to water permeation at high service temperatures and attack of fill fluids such as castor oil, on the encapsulant. These situations may also lead to adhesion loss between the encapsulant and other component substrates ultimately resulting in electrical and/or acoustical failure of the transducer.

Polyurethanes also are relatively hazardous materials. The isocyanate-containing prepolymers are very hazardous if they become airborne. Methylene (bis-orthochloroaniline) (MOCA or MBOCA) is not a particularly toxic material but is suspected to be a weak carcinogen. Consequently, polyurethanes are frequently reformulated to avoid legal problems with Government regulatory agencies.

There is also concern over the possibility of encapsulant suppliers changing their basic raw material composition without first informing the proper Government authority. The modified encapsulant may well meet all handling and performance criteria required by military specification, but somehow ultimately fail during service. This occurs, in part, because not all of the critical performance characteristics are fully understood; and, hence, they cannot be adequately defined in the military specification which covers the encapsulant. As a result, work was initiated to develop new polyurethane sonar encapsulants possessing improved castor oil resistance from readily

available commercial raw materials. The following compositional, handling, and performance goals for the urethane encapsulant were set forth at the inception of this work.

- Non-proprietary individual components.
- Free of all potentially carcinogenic and hazardous components.
- Thirty-minute minimum application life at 22°C.
- Two hundred P initial mixed viscosity at 25°C.
- Gel time of 45-90 minutes at temperatures between 25 and 90°C.
- Eight-hour full cure at temperatures between 25 and 90°C.
- Shore A hardness between 45 and 80, with 65 to 75 Shore A most desirable.
- A minimum tensile strength of 2000 psi.
- A minimum volume resistivity of 1.0×10^{12} ohm-cm.
- A minimum dielectric strength of 300 kV/cm.
- A glass transition temperature below -30°C.
- The cured elastomer should yield a cohesive failure to primed mild steel.
- The water vapor permeation constant should not exceed 100 ng-cm/cm²-hr-torr.
- The product of sound speed and density should be within 10% of 1.6×10^6 kg/M²-sec.
- Material transparent to visible light in 1 cm thickness.
- Shear or Young's modulus must not change by more than 25%, or volume resistivity by more than 90%, after a two-year equivalent of water immersion.
- Shear or Young's modulus must not change by more than 10% after a two-year equivalent of castor oil immersion.
- The formulation cost should not exceed \$11.00/kg (\$5.00/lb).

APPROACH

With such an extensive list of desirable properties, a reasonable approach was necessary to limit the number of material choices and tests. The approach was to successively screen the candidates based on one or more of the required properties. The initial set of screening tests was performed on several commercial prepolymer systems cured with proprietary diol/diamine curatives. These results permitted concentration of subsequent efforts on only four prepolymer systems. For these prepolymer systems, cure systems were developed based on seven commercial diamines. The choice of diamines was narrowed to three based on physical properties of the cured polyurethanes. These three cure systems were then modified to improve their handling properties. The next screening among the 12 candidates (4 prepolymers $\times$ 3 cure systems) was based on sound speed. The seven acceptable choices were then further screened by careful consideration of their detailed handling, electrical and engineering properties, and resistance to water and castor oil. Two excellent materials resulted.

INITIAL SYSTEM SCREENING

Initial screening tests were conducted with Adiprene L-100 and three similar commercial polyether urethane prepolymers from the Products Research and Chemical Corporation (PRC). These polymers were evaluated against four experimental liquid diamine/hydroxyl containing curing agents. The results of this work are summarized in Tables 1 and 2. The selection of optimum stoichiometry is important with a high-performance, castable polyurethane. Normally, if one is using a diamine to cure an isocyanate-terminated polymer, optimum properties will be obtained by employing a weight mixing ratio in which the equivalent ratio of isocyanate to diamine is between 1.05-1.10/1.0. If the curing agent is composed of only hydroxyl containing reactants the equivalent ratio of isocyanate to hydroxyl normally runs between 0.95-1.05/1.0. In this work, where the urethane curing agents were composed of both amino and hydroxyl functions, mixing ratios were chosen to give an equivalent ratio of isocyanate to diamine plus hydroxyl of 1.05/1.0. In all instances throughout the work, the urethane elastomers were cured 16 hours at 180°F unless otherwise stated.

Urethane elastomers derived from experimental curing agent 64 (see Table 1) proved to be of little interest because of low tensile strength and ultimate elongation. Elastomers prepared from experimental curing agents 78, 90, and 92 were too high in Shore A hardness for a suitable transducer encapsulant and were not investigated further.

The experimental curing agent 78/Adiprene L-100 combination gave a volume resistivity which was 0.89 decades lower than the same curing agent with Permapol U-82 polymer. The electrical insulation resistance of the Adiprene system was 37% lower than that of Permapol U-82. Consider experimental curing agent 92 with Adiprene L-100 and Permapol U-84; the Adiprene material afforded a 0.83 decade lower volume resistivity and a 22% lower electrical insulation resistance than the U-84 polymer. The Adiprene polymers are based upon polyoxytetramethylene glycol and the two Permapol U polymers on polyoxypropylene glycol. The significant difference in electrical properties

was attributed to the raw materials used in the manufacture of the polymers. We have found that similar polymers based upon PTMG from Uniroyal Chemical give higher initial resistivity properties than Adiprene polymers.

EVALUATION OF COMMERCIAL POLYMERS

Evaluation of commercially available urethane polymers included polyether urethanes derived from PPG (polyoxypropylene glycol) and PTMG (polyoxytetramethylene glycol). Polyether urethanes were selected because it was believed they would come the closest to providing the desired level of castor oil resistance. Urethane polymers based upon HPBD (hydroxy terminated polybutadiene) were not included in this study because they are not widely commercially available and it had been established earlier that urethane elastomers derived from HPBD do not possess the desired level of castor oil resistance.

One of the prime requisites of this work was to develop a material which affords significantly higher castor oil resistance than is provided by urethanes currently in use. Most of the sonar encapsulants currently used are either PPG or HPBD based. PTMG urethanes generally have been neglected for this application because they form crystalline solids at 65°F or below, they tend towards opaque products upon curing, and they form high Shore A hardness products when cured with MBOCA. If only castor oil resistance is considered, the PTMG systems theoretically should yield a higher level of resistance than either PPG or HPBD urethanes. Consequently, the basic emphasis was to investigate commercially available PTMG-based polymers and certain PPG-based polymers which were felt would give better castor oil resistance, provided suitable curing agent technology could be developed.

CURING AGENT DEVELOPMENT

One of the basic requirements of this work was to avoid the hazards associated with MBOCA [methylene bis(2-chloroaniline)]. Hence MBOCA substitutes were chosen from those commercially available in mid-1981. These were:

- Polycure 1000: methylene bis(methylantranilate) - PTM&W Industries Inc.
- Polycure 1001: a low melting derivative of Polycure 1000 - PTM&W Industries Inc.
- Polacure 740M: trimethylene glycol di-p-aminobenzoate - Polaroid Corporation.
- Cyanacure: 1,2-bis(2-aminophenylthio)ethane American Cyanamid
- Permapol D-562: A low melting derivative of methylene bis(methylantranilate) - PRC

- Apocure 601P: liquid version of Cyanacure - M&T Chemicals
- Baytec PU-1604: isobutyl 4-chloro-3,5-diaminobenzoate - Mobay Chemical Co.

One of the major goals was to develop an easily usable liquid castable urethane. This involves development of a liquid curing agent, or one which is easily converted into a suitably stable liquid composition. All of the currently known MBOCA substitutes are solids. Hence, combinations or solutions with isocyanate-reactive liquid components were employed to obtain a suitable viscosity liquid curing agent. Inert liquid plasticizers are not acceptable in high performance urethanes as they tend to migrate under service conditions and disrupt adhesion. Liquid components were selected which maintain a good balance between diamine solubility, low viscosity, and system compatibility, while providing reasonable handling and performance characteristics in the end products.

It was found very early in this work that all the proposed MBOCA substitutes under investigation lacked, to varying degrees, the desired solubility characteristics of MBOCA in liquid isocyanate-reactive curing agent components. Good diamine solubility in the other curing agent components is necessary to obtain optimum compatibility in the cured urethane elastomer. Experience in developing liquid castable polyurethanes has shown that general mechanical properties improve with increasing cured elastomer compatibility. Compatibility in the cured products is also necessary to attain an encapsulant which will allow visual inspection of the completed sonar device. Incompatible systems will result in cured urethane elastomers which are translucent or even opaque to visible light. They may also afford elastomers with less than optimum performance properties. Extensive effort was devoted to finding compositions which approached acceptable diamine solubility.

The first curing agent which showed promise is listed below:

<u>COMPONENT</u>	<u>SUPPLIER</u>	<u>PARTS BY WEIGHT (%)</u>
2-ethyl-1,3-hexanediol	Union Carbide	14.70
N,N-(2-hydroxypropyl)-aniline	Upjohn Co.	36.75
Diamine Component	(See pages 4 & 5)	22.05
N,N,N',N'-tetrakis-(2-hydroxypropyl)ethylenediamine or (Quadrol)	BASF Wyandotte	26.46
Ferric Acetyl Acetate	Harshaw Chemical	0.04
		<u>100.00</u>

This basic curing agent formula was selected so that the resulting cured urethane elastomers were within the desired hardness range, were flexible but not thermo-plastic, and were suitably compatible. This involved obtaining, through a combination of theoretical and experimental considerations, an optimum balance between the chemical equivalence of curing agent difunctional and polyfunctional components. Each of the seven diamine curatives were

substituted directly into the formula at 22.05 wt. %. All of the curing agent compositions remained homogeneous liquids for at least 8 to 11 hours at ambient temperature.

Catalyst selection played a key role in handling and appearance of castable urethanes. Organo tin compounds such as dibutyl tin dilaurate or the diacetate derivative afforded optically clear elastomers. They were not satisfactory in this curing agent as the elastomers were prone to gassing due to moisture or water sensitivity. Internal stress cracking also developed during elevated temperature cures. Tin catalysts are used in high-performance, castable urethanes, but each individual system must be evaluated as the tin derivatives may act to catalyze gas formation reactions yielding a foam as well as to catalyze reactions to form elastomers.

Lead naphthenate, oleate, and octoate eliminated water sensitivity and stress cracking but gave end products with poor compatibility, as evidenced by cured urethanes which were either translucent or opaque to visible light. Polymers based upon a PPG backbone were translucent, while those containing a PTMG backbone were opaque. Tertiary amine catalysts were not investigated because their activity is primarily directed towards the isocyanate-water reaction rather than the desired isocyanate-hydroxyl reactions. These same amines may also be electrically conductive and/or corrosive in the presence of water.

The best catalyst (in terms of activity, compatibility, and long term package stability) was found to be FeAA (ferric acetylacetonate). Other chelates such as cobalt AA or manganese AA had too low a level of activity to be practical. Iron carboxylates such as the naphthenate and tallate were found to be good overall catalysts, but final products tended to be opaque.

Sixteen-hour, 180°F cures were conducted using low and medium viscosity urethane polymers based upon TDI/PPG. The data is summarized in Tables 3 and 4. Additional work is also shown in Table 5 with PTMG-based polymers Adiprene L-100 and L-167.

All the materials shown in Tables 3 and 4 have desirable mixed viscosities, long application lives, and excellent volume and surface resistivities. The hardnesses, tear strengths, tensile strengths, and ultimate elongations are all low. In contrast, the materials shown in Table 5 have excellent electrical resistivities, tear and tensile strengths, but hardness is too high and elongations too low.

The PTMG-based elastomers shown in Table 5 are an improvement over the PPG-based elastomers shown in Tables 3 and 4, but the ultimate elongations remain low. One interesting note, the electrical resistivities of the Permapol U-47-based elastomers are much higher than those of the Adiprene L-167 systems.

Attempts were made to improve ultimate elongation by two means. The first was to increase the diamine content of the curing agent. This technique will generally raise elongation, tensile, tear, and hardness. This arises because the equivalent ratio of diamine to tri- and/or tetrafunctional hydroxyl is increased. In this instance, when the diamine level was raised from 22 to 36 wt. %, the resulting curing agents were no longer stable

liquids. Crystalline diamine rapidly came out of solution upon standing at ambient temperature. The worst case was Polacure 740M; it crystallized after two hours of room temperature conditioning. Lesser amounts of diamine did not suitably increase elongation. Based on experience, a practical, liquid-curing agent must be physically stable for a minimum of eight hours at ambient temperatures after liquefying from a solid or semi-solid state.

The second approach was to modify the curing agent by reducing the chemical equivalent ratio of polyfunctional hydroxyl to difunctional hydroxyl. This must be accomplished while retaining a reasonable level of diamine solubility. The initial attempts involved replacing a portion of the Quadrol with trifunctional castor oil, and replacing a portion of the ethylhexane diol with 1500 equivalent weight, TDI extended polyether diol. We were able to increase the ultimate elongation from 250-300% up to 380%. The curing agents remained liquids, but their individual viscosities were too high at 960-1000 P to be acceptable. Therefore, neither means attempted to improve ultimate elongation could be used.

During this period we were informed by M&T Chemicals that Apocure 601P was no longer a commercial product. It was deleted from further work. Of the remaining six diamine curatives, Polycure 1000, Polycure 1001, and Permapol D-562, all afforded nearly equivalent performance properties. This fact may be confirmed by a review of Tables 3 and 4. The most noteworthy feature among the three is the significantly higher curing agent solubility of the Permapol material. We were unable to find any difference between the two Polycures except that PC-1001 gave somewhat inferior elastomers compared to the PC-1000. Thus, Polycure 1000 and Permapol D-562 were retained for further study. We subsequently learned that Polycure 1001 is also no longer commercially available.

The low elastomer elongations and high curing agent viscosities were overcome by replacing the Quadrol and 1500-equivalent weight PTMG diol with castor oil and a very low viscosity 2000 molecular weight PPG diol. This had the overall effect of changing the chemical equivalent ratio of tetrol/diol from 0.655/1.0 down to a 0.244/1.0 triol/diol ratio. This modification amounted to a 62% reduction in curing agent hydroxyl functionality.

Subsequent work to further refine the curing agent composition afforded no improvement in properties. If castor oil was replaced with various molecular weight PPG triols, diamine solubilities were disproportionately reduced. If the castor oil content was increased much beyond 31 wt. %, diamine solubility decreased. Diamine solubility was found in the following order:

Baytec = Permapol > Polycure > Cyanacure > Polacure

Table 2 - First phase system evaluation electrical properties.

	RESISTIVITY AT 75°F		ELECTRICAL INSULATION RESISTANCE (MΩ)
	V (Ω CM)	SURFACE (Ω)	
PRC Permapol U-84 XCA*			
64	5.5 × 10 ¹²	9.3 × 10 ¹²	275,000
78	8.3 × 10 ¹²	1.9 × 10 ¹³	358,000
90	1.7 × 10 ¹³	6.0 × 10 ¹³	415,000
92	3.4 × 10 ¹³	5.1 × 10 ¹³	405,000
PRC Permapol U-82 XCA			
78	7.8 × 10 ¹²	2.2 × 10 ¹³	388,000
92	9.1 × 10 ¹²	4.7 × 10 ¹³	395,000
PRC Permapol U-56 XCA			
78	6.6 × 10 ¹²	1.7 × 10 ¹³	349,000
92	8.6 × 10 ¹²	2.2 × 10 ¹³	377,000
DuPont Adiprene L-100 XCA			
78	8.7 × 10 ¹¹	7.3 × 10 ¹²	243,000
92	5.6 × 10 ¹²	1.1 × 10 ¹³	317,000

*Experimental Curing Agent

Table 1 - First phase system evaluation mechanical properties.

	APPLICATION LIFE (TIME TO 2500 P)	HARDNESS* (SHORE A)	TEAR STRENGTH (LBS/IN.)	TENSILE STRENGTH (PSI)	ULTIMATE ELONGATION (%)
PRC Permapol U-84 (5% available isocyanate, 91 P) XCA **					
64	>3 hours	53	110	685	370
78	>4 hours	74	200	2500	700
90	>2 hours	66	150	850	500
92	>1.5 hours	68	190	1600	550
PRC Permapol U-82 (5% available isocyanate, 223 P) XCA					
78	2 hours	83	310	3500	575
92	>2 hours	79	245	2120	460
PRC Permapol U-56 (7.1% available isocyanate, 103 P) XCA					
78	2 hours	84	310	3700	520
92	2 hours	79	250	2750	425
DuPont Adiprene L-100 (4.1% available isocyanate, 241 P) XCA					
78	2 hours	78	250	3700	575
92	2 hours	77	260	4700	525

*Samples cured 16 hours at 180°F.

**Experimental curing agent.

castor oil as a fill fluid. These materials provide a level of castor oil resistance above that of commercially available polyurethane sonar encapsulants.

- One or two of these formulations should be specified for future sonar transducer encapsulation.

REFERENCES

1. Saunders, J.H. and Frisch, K.C. "Polyurethane Chemistry and Technology, Part I, Chemistry". Interscience, (1962), 129-215.
2. E.M. Zacharias, M.W. Zacharias, and R.A. Parnell, Jr., Modern Plastics, May, 1974.
3. Cassidy, P.E., T.M. Aminabhavi, and C.M. Thompson, Rubber Chem and Tech, 56 (3), p. 594 (1983).

ACKNOWLEDGMENT

This work was performed under the sponsorship of the Sonar Transducer Reliability Improvement Program (STRIP). STRIP is funded by the Naval Sea Systems Command (SEA 63X5) and managed by Dr. Robert W. Timme.

SONAR ENCAPSULANT COSTS

One of the program requirements specified was to develop expected costs of the potential encapsulant. Reported here are raw material costs (RMC) for the candidates. The component prices were quoted as of September 1982. It is not possible to supply actual encapsulant total costs, because manufacturing costs differ widely between firms, and chemical companies do not usually release their manufacturing costs.

The three curing agents range in RMC between \$1.58 to \$2.09/lb. The curing agents comprise 15 to 25 wt. % of the total encapsulant. The encapsulants themselves would cost between \$2.41/lb and \$2.55/lb RMC or \$5.31 to \$5.62/Kg.

CONCLUSIONS

- Two of the commercially available aromatic diamines referred to as potentially non-carcinogenic substitutes for MBOCA are acceptable in liquid castable urethanes. The two most common drawbacks encountered with other diamines were very limited solubility in liquid urethane curing agents and poor compatibility in the cured elastomer.
- Polycure 1000, Permapol D-562 and Cyanacure were the best general diamine curatives within the constraints of the program. The first two are the most attractive.
- Adiprene L-167 or Vibrathane B-601 are the polymers of choice for best castor oil resistance even though both crystallize at 65°F or below.
- Adiprene L-100-containing samples provide the best acoustic impedance match with water but at the cost of an increasing tendency to crystallize.
- Of the systems investigated, the Polycure 1000-or Permapol D-562-containing curing agent and Adiprene L-167 afforded the best combined castor oil and water resistance.
- General encapsulant electrical properties are somewhat lower than some commercial two-part castable urethanes, but are adequate for the intended application. Vibrathane TDI/PTMG urethane polymers may give slightly better values.
- Candidate encapsulant raw material costs run between \$2.41-2.55/lb or \$5.31-5.62/kg.
- The polyurethane sonar encapsulants developed during this program are designed for a very specific end use: the encapsulation of sonar devices which employ

Tables 17 and 18 summarize water and oil immersion data using precut tensile specimens dimensioned two ways. Only Polycure and Cyanacure curing agents were used with Adiprene L-100. The general agreement between these two approaches was surprisingly good. Comparative final elongations and tensile strengths are shown below.

<u>WATER IMMERSION</u>	<u>POLYCURE/L-100</u>		<u>CYANACURE/L-100</u>	
	<u>FRESH CUT</u>	<u>PRECUT</u>	<u>FRESH CUT</u>	<u>PRECUT</u>
Tensile (psi)	2671*	2355* (2369)**	2443*	2018 (2025)**
Ultimate Elongation, %	580	625	670	650
<u>CASTOR OIL IMMERSION</u>				
Tensile (psi)	1164	1310 (1693)	1663	1244 (1659)
Ultimate Elongation, %	400	500	550	550

*Calculated using pretest dimensions.

**Calculated using after exposure dimensions.

The freshly cut specimens which were measured after fluid exposure, but before tensile testing, tended to give higher moduli values. This approach better reflects potential service conditions than using precut and premeasured test samples. Table 19 carries the analysis a step further by showing percentage changes between initial and final moduli as well as standard deviation of individual modulus data points under the three sample criteria. The data support the proposal that specimens should not be precut.

FUNGUS RESISTANCE

Three of the candidate encapsulants were selected for resistance to fungus attack as per Mil-E-5272. Adiprene L-167 cured with Polycure, Permapol and Cyanacure was totally resistant to fungus attack. No growth whatsoever was observed. The test was conducted by Truesdail Laboratories for Los Angeles, an approved non-Government test facility.

WATER VAPOR PERMEABILITY

The water permeability of three of the candidate materials was measured by a cup weight-loss method. The permeability constant at 25°C of Adiprene L-100/Permapol was 470 ng cm/cm² hr torr of Adiprene L-100/Polycure 1000 was 490 ng cm/cm² hr torr, and of Adiprene L-167/Polycure 1000 was 350 ng cm/cm² hr torr. Reported values for proprietary polyurethanes range from 100 to 420 ng cm/cm² hr torr [3].

supported by the relative changes in hardness we observed during the 120-day, 158°F, 99%-RH exposure in which the three Cyanacure urethanes all remained elastomers, but lost a far higher percentage of their initial hardness.

- **Castor Oil Resistance:** The results of the 1000-hour, 122°F castor oil immersions are summarized in Table 16. The effect of castor oil immersion on moduli changes was greater than that observed with water. The TDI/PTMG-based polymers were initially selected for study as they offered the best chance for castor oil resistance over urethane polymers derived from polyoxypropylene glycol (PPG) or hydroxy-terminated polybutadiene (HPBD). We observed losses in ultimate elongations ranging from 9 to 46%. Of the seven formulas tested, all exhibited moduli increases over the 100 to 300% extension range. That is, the actual modulus change at any given exposure time went from a negative value to either a smaller negative value or a positive number as the material was extended from 100 up to 300%. This suggests that while castor oil immersion tended to reduce modulus at any given extension over time, the adverse effect on that modulus change became less pronounced at higher extensions.

The Polycure 1000/Adiprene L-100 and L-167, Permapol D-562/Adiprene L-100 and Cyanacure/Adiprene L-167 systems all meet the criterion of less than 25% moduli change. The Cyanacure/Adiprene L-100 and L-200 formulas gave the highest absolute moduli changes.

Our data shows that the castor oil resistance of the diamine curatives used with PTMG polymers decreases in the order:

Polycure 1000 > Permapol D-562 > Cyanacure

The fluid immersion studies were a very important aspect of the research. We have always believed that exposures of this nature should be conducted using sheets of cured elastomer from which tensile dumbbell specimens are freshly cut as required and dimensioned just prior to actual tensile-elongation determination. The rationale behind this philosophy is that either the aqueous or organic fluid exposure non-uniformly deforms the precut specimen thus making accurate measurement of sample crosssection and thickness very difficult. This expected irregularity in samples would also increase the probability of higher than normal statistical variances in the raw test data.

The fluid immersions were conducted using all freshly cut tensile specimens in which each sample dimension was determined just prior to tensile testing. A smaller sub-study was run in which the samples were precut before exposure. The necessary dimensions were determined in two ways: using initial dimensions before immersion, and dimensions after immersion.

CHANGES IN ELECTRICAL PROPERTIES UNDER WET OR HUMID CONDITIONS

Electrical insulation resistance and high potential resistance samples were exposed 28 days to 158°F and 99% RH. All formulas employing Adiprene L-167 performed very well (see Table 14). The Adiprene L-100 formulas did well, but fell half-a-decade below that of their L-167 analogues. These results are quite acceptable and are of the same order and magnitude that were observed previously with Adiprene and MBOCA-containing elastomers under the same conditions.

The high potential resistance after the same exposure was determined by applying 2000 V rms and observing any current leakage. Current leakage, if any, was less than the 2.0 μ A minimum detectable level of the test instrument.

The last electrical test conducted was volume resistivity of samples immersed 60 days in 122°F water. In all instances, the resistivity was essentially unchanged.

PHYSICAL RESISTANCE TO FLUID IMMERSION

The water- and castor-oil-resistance tests were the most extensive of the entire program. Stress/strain analysis, coupled with hardness and weight change were determined.

- Water Resistance: The seven candidate encapsulants were immersed in 122°F distilled water for a 60-day term. Samples were withdrawn periodically to determine hardness, modulus, tensile strength and ultimate elongation. As much statistical variation as possible was eliminated in the comparative data by using tensile samples of constant 75 to 85 mil thickness. At least five specimens were tested for each individual data point. The obvious high and low statistical outliers were eliminated from the final computation. The samples, once removed from the immersion media, were all tested within a five- to six-hour period. The results are summarized in Table 15.

It would be improper to compare modulus values over the entire range of elastomer extension because the cured elastomer will not experience in service conditions of high extension. A more practical approach from a field performance point of view would be to consider modulus changes up to and including 300% extension. Under these criteria, six of the seven urethanes shown in Table 15 exhibit modulus and ultimate elongation changes below 25%. The exception is the Permapol D-562/Adiprene L-167 combination which may fall within the 25% maximum change if the test was repeated because of similarities in overall chemical structure to the other systems.

The basic difference between the Polycure/Permapol systems and Cyanacures is the apparent higher hydrophobicity of the former two. This conclusion is

urethane elastomers derived from the four polymers and the Polycure 1000 liquid curing agent are given below:

	VIBRATHANE B-600	ADIPRENE L-100	VIBRATHANE B-601	ADIPRENE L-167
Hardness	68 A	67 A	71 A	70 A
Modulus at				
100% extension	271 psi	266	256	235
200	372	366	377	357
300	549	531	454	471
400	779	775	739	794
500	1591	1603	1606	1677
Tensile Strength	3378 psi	3304	3394	3475
Tear Strength	185 lbs/in.	188	208	185
Ultimate Elongation	630%	625	600	615

PHYSICAL HYDROLYTIC STABILITY

The results of the 120-day, 158°F/99% RH Hydrolytic Stability test are presented in Table 13. All elastomer formulas exhibited about the same degree of moisture absorption. The Polycure and Permapol urethanes lost the same amount of hardness over the term of exposure. The Cyanacure systems show significant inferiority in this respect, losing between 36 and 53% of their initial hardness. The 20 to 22% hardness losses observed for the Polycure and Permapol urethanes are not unusual if one considers the criterion set forth in MIL-M-24021. The specification allows a 20% maximum loss in hardness after the same exposure conditions, but covers a hardness range of 70 to 92 Shore A. In actuality, those urethanes listed on the Qualified Products List of MIL-M-24041C all exceed 80 Shore A in hardness. The lower the initial hardness of the product tested the more drastic the effect in the percentage of hardness lost. The Polycure- and Permapol-based encapsulants are lower at 60 to 62 Shore A and those of Cyanacure even more so at 55 to 64 Shore A.

The bis(phenylthio)ethane structure of the Cyanacure-based elastomers appears more susceptible to plasticization by moisture than do the orthoester substituted methylene bis(anthranilates) of the other curatives. The concept of moisture plasticization is defined accordingly: water permeates the polymer matrix and cleaves the intermolecular hydrogen bonds between urethane and urea sites on adjacent polymer chains. New hydrogen bonds are now formed between water and the urea/urethane linkages of the individual polymer chains. The result is that both hardness and modulus are reduced. This is exactly what is found in the stress/strain data of Table 15. As long as no irreversible polymer degradation occurs, such as actual destruction of backbone urea or urethane sites, a good portion of physical property loss will be regained as the material dries out. The general hydrolytic stability of these medium hardness urethanes is, nevertheless, good. The fact that all encapsulants remained elastomeric is sufficient evidence that no extensive polymer chain scission took place. In addition, had irreversible chain degradation occurred, it would have been reflected in the electrical performance data under wet or humid conditions of Table 14.

The primer was air dried 70 minutes at ambient temperature prior to encapsulating. The failure mode was all low level adhesive between the silver film and the ceramic surface at 5 to 6 lbs/in. The adhesion of the encapsulant to the primed silver appeared to be excellent. Failure at the silver/ceramic interface was doubtlessly due to the fact that the ceramic had been previously used.

GENERAL ELECTRICAL PERFORMANCE

Electrical test data are summarized in Table 12. Volume and surface resistivities, as well as insulation resistances, are a little more than one decade lower than the target profile, and between 2 and 3 decades lower in insulation resistance. As stated earlier, use of other PTMG based polymers might improve electrical properties. In support of this concept, the Permapol D-562/Adiprene L-100 system was re-examined where Uniroyal Vibrathane B-600 was used in place of the Adiprene. The respective resistivity and insulation resistances are compared below:

	<u>PERMAPOL D-562</u> <u>ADIPRENE L-100</u>	<u>PERMAPOL D-562</u> <u>VIBRATHANE B-600</u>
Volume Resistivity at 75°F, Ω -cm	2.2×10^{10}	4.7×10^{10}
Insulation Resistance at 75°F, M Ω	1.20×10^3	8.0×10^3

There is a noticeable difference electrically between two ostensibly equivalent PTMG-based polymers. The Vibrathane B-600 shows an insulation resistance 6.7 times higher and a volume resistivity over twice as high as the Adiprene. We would not expect to see similar large variances between dielectric constants or power factors for, in our experience, these properties have proven much more insensitive to variations in raw materials or manufacturing processes.

The values obtained for dielectric strength are somewhat lower than expected. Values in the range 300-315 V/mil for 125-mil-thick samples would be more typical. The sound velocities are all good save for the marginal Cyanacure/L-200 value of 1714 m/sec.

The dielectric constants and power factors are all somewhat higher than those normally found in liquid castable PTMG-based/MBOCA urethanes tested at frequencies of 1 kHz and 1 MHz. They are not, however, outside the range found in high performance, non-MBOCA liquid castable polyether urethanes currently available.

It appears that, except for the modest difference in electrical properties discussed above, the Vibrathane and Adiprene PTMG-based polymers are equivalent and either would be suitable for the intended application. In support of this conclusion, comparative mechanical properties of cured

GENERAL HANDLING AND PERFORMANCE PROPERTIES

The general handling, mechanical, and adhesion properties of the seven candidate urethane encapsulants are summarized in Table 11. The most outstanding features are the excellent compression sets, tear strengths, tensile strengths, and ultimate elongations; considering that these are all medium hardness-range materials.

System application lives are all long enough so that premixed and frozen packaging is feasible should the end user desire it. The initial mixed viscosities of the Adiprene L-100 systems are on the high side. The choice from a material handling point of view would be the Vibrathane B-601 or Adiprene L-167 formulas. The much lower viscosities in this instance make premixed and frozen packaging even more desirable. The Adiprene L-200 polymers offered no real advantage over that of L-167, and had only a marginally acceptable high sound speed. The Polycure and Permapol curing agents were essentially equivalent in overall performance, but hold a slight edge over the corresponding Cyanacure systems.

The evaluation of adhesion to various substrates was conducted using state-of-the-art surface preparation and primers as summarized below (data are in Table 11):

- PVC: The PVC corresponded to MIL-I-7444. The surface was tackified with clean, oil-free methyl ethyl ketone. The encapsulant was then applied. Samples were prepared and tested according to MIL-M-24041C. The test results of 25 lbs/in. width of peel or more are considered excellent.
- Neoprene: The rubber corresponded to MIL-R-3065. The surface was mechanically abraded with a clean, grease free wire wheel. A liberal coat of PR-1523-M primer was applied by brush to the abraded neoprene surface. The excess primer was wiped off with a lint-free cotton gauze pad after air drying 30 to 35 minutes. The encapsulant was then applied. Samples were prepared and tested as per MIL-M-24041C. The peel adhesion values are considered quite good.
- Stainless Steel: The metal corresponded to MIL-S-5059. The panels were cleaned with oil free methyl ethyl ketone and primed with PR-420 primer. The primer was allowed to dry 60 minutes at ambient temperature prior to encapsulating. The peel values all exceed 40 lbs/in.
- Transducer Ceramic: Samples of PZT-type transducer ceramic which had a silver surface finish. The silver surface was cleaned with methyl ethyl ketone prior to application of PR-1533. This is a primer especially developed for precious metal surfaces.

sing-around velocimeter at a frequency of 1.5 MHz. The measurement method is similar to that described by Zacharias, et al [2].

Several of the elastomers exhibit sound speeds that are unacceptably high. Thus, the following materials were excluded from the remainder of the work:

- All formulations derived from Permapol U-47.
- Formulations of Adiprene L-200 with Polycure 1000 and Permapol D-562 curing agents.

Seven potential encapsulant formulas remained for the last phase of the work. The final tests were:

- General Handling and Performance Properties (Table 11).
- General Electrical Properties (Table 12).
- Physical Hydrolytic Stability, 120 Days, 158°F/99% RH (Table 13).
- Resistance of Electrical Properties to Wet or Humid Environments (Table 14).
- Stress/Strain: Resistance to 60 Day Water Immersion at 122°F (Table 15).
- Stress/Strain: Resistance to 1000 Hours Castor Oil Immersion at 122°F (Table 16).
- Stress/Strain Specimen Analysis (Tables 17, 18, and 19).
- Encapsulant Raw Material Costs.
- Low Temperature Flexibility.
- Fungus Resistance.
- Water Vapor Permeability.

The mixing ratios, by weight, of the final candidates are shown below:

	POLYURE 1000		CYANACURE			PERMAPOL D-562	
	L-100	L-167	L-100	L-167	L-200	L-100	L-167
Curing Agent	19.0	29.6	17.8	27.3	32.5	18.8	28.8
Polymer	100.0	100.0	100.0	100.0	100.0	100.0	100.0

Two urethane polymers based on MDI and PTMG from Uniroyal Chemical were evaluated with the three diamine-containing curing agents. This was a practical approach as long as the FeAA catalyst level within the curing agent was the only item requiring adjustment.

The results of the study are summarized in Table 8. It was assumed, that because the 2500 P application lives of the TDI/PTMG- and PPG-based polymers all exceeded six hours, shorter but acceptable worklives would be obtained with the MDI polymers of similar available isocyanate content. This proved not to be the case. The time to reach a mixed viscosity of 2500 P ranged between 12 and 90 minutes, even when the FeAA catalyst was reduced to 15% of its initial concentration. Performance properties could not be determined for the Cyanacure system because the application life was only 12-18 minutes at 75°F.

If the FeAA catalyst was totally eliminated, unsuitable elastomers were obtained because there was now an imbalance between the hydroxyl/isocyanate and diamine/isocyanate reaction rates. This resulted in incompatibility and partial gelling during handling. The high activity level is due to the structural differences between 2,4-TDI and MDI. The rate constant for MDI at 86°F with diethylene glycol adipate is 0.34×10^4 liters/mole-sec. Under similar conditions, the rate constant for the isocyanate in the 2-position of TDI is 0.057×10^4 liters/mole-sec and 0.45×10^4 liters/mole-sec for the isocyanate in the 4 position of TDI [1]. It was assumed that when the initial prepolymer is manufactured, the isocyanate in the 4 position of TDI is consumed first. This leaves nearly a six-fold difference in activity between the isocyanate groups of an MDI prepolymer and the isocyanate group in the 2-position of TDI properties. MDI systems will require a new curing agent to be developed.

The use of MDI polymers becomes even less attractive when one considers polymer stability. A second set of materials had to be requested from the supplier because the first units were well below published specification values for both available isocyanate content and viscosity. The two MDI-containing polymers showed evidence of crystallizing after 24 to 48 hours storage at 71 to 77°F. During successive recrystallizations at 120°F, these polymers tended to lose available isocyanate and increase in viscosity. Partially full containers were most difficult to handle, and had to be well purged with dry nitrogen to ensure even a semblance of shelf stability. If this was not done, the polymer formed a medium off-white, opaque insoluble material. The insoluble material, probably dimerized MDI, could not be converted into active monomer by the application of mild heat. The polymer was no longer usable at this point.

SELECTION OF FINAL CANDIDATES

The three remaining diamine cure systems were to be evaluated with Permapol U-47 and Adiprenes L-100, L-167 and L-200. The Permapol U-47 is a non-crystallizing polymer while the latter three will crystallize at temperatures below 65°F.

The point had then been reached where a final selection criterion was necessary prior to starting the extensive environmental testing. The basis for the final selection was sound speed. These measurements were made in a

SCREENING OF CANDIDATE ELASTOMERS

The remaining four curing agents were employed as curatives for TDI/PTMG-, TDI/PPG-, and MDI/PTMG-based polymers. The resulting data are summarized in Tables 6, 7, 8 and 9.

Urethane elastomers based upon Adiprene L-200 exhibit very low initial mixed viscosities, long application lives, and quite acceptable hardnesses, tears, tensiles and ultimate elongations. The L-200 systems yield slightly higher electrical properties than either L-100 or L-167. In terms of mechanical and electrical properties, the Baytec-containing curing agents gave the best results followed by Polycure, Permapol, and Cyanacure.

Cyanacure and the now deleted Polacure 740M gave the best results electrically. This is evidenced by volume and surface resistivity data of Tables 4, 5 and 6, and the insulation resistance data of Table 6. The resistivities can be raised a full decade by changing to a PPG-based polymer from the TDI/PTMG types. Tear and tensile strengths are all lower with the PPG systems. This is substantiated by comparing the data of Table 6 to Table 7.

There is one very significant constraint on the candidate sonar encapsulant. It must be transparent to visible light. Previous experience with present day sonar encapsulants is that they are almost always a clear, or slightly translucent, lightly colored material. The transparency is necessary because the final component is subjected to visual inspection. This aspect is important as the transducer must be free from air bubbles, foreign matter, and other imperfections which, if present, will generally induce some form of signal attenuation.

At this point in the development, Baytec PU-1604 would have been the choice as a MBOCA substitute for sonar applications as well as general polyurethane potting and molding operations. Unfortunately, it has one serious drawback which cannot be overcome by curing agent modification. The Baytec-cured polyether urethanes are either exceptionally dark or opaque. Baytec PU-1604 was therefore eliminated from further consideration.

Initial efforts to match the mechanical properties of the TDI/PTMG polymers with those containing a PPG backbone were unsuccessful. However, Permapol U-47 is a non-crystallizing, high-strength TDI/PPG-based urethane polymer which meets and exceeds the basic performance properties of the three TDI/PTMG systems included in this work. This is confirmed by comparing the data of Table 9 to that of Table 6. The electrical properties are, in fact, a full decade higher.

Only TDI-terminated polyethers had been studied so far in the program. At this point, other types of isocyanate-terminated polyethers such as MDI (diphenylmethane diisocyanate) were investigated to determine if improvements in physical properties could be achieved. Urethane elastomers derived from MDI generally afford higher tear and tensile strengths, equal or slightly lower elongations, and have improved hydrolytic stability over equivalent TDI/polyether systems. The overall electrical properties are equivalent to or slightly better than TDI analogues.

The new curing agent composition, which ultimately was shown to be optimum is presented below:

<u>COMPONENT</u>	<u>SUPPLIER</u>	<u>PARTS BY WEIGHT (%)</u>
2-Ethyl-1,3-hexanediol	Union Carbide	6.02
N,N-(2-hydroxypropyl)aniline	Upjohn Co.	18.07
Diamine component	(See pages 4 & 5)	21.68
Castor Oil	Caschem	24.09
2000 PPG diol	Olin Corp.	30.11
Ferric acetyl acetate	Harshaw Chemical	0.03
		100.00

This basic curing agent composition affords viscosities in the 38-42-P range which is highly desirable. Most high-strength, castable polyurethanes which employ diamine containing curing agents will require some preprocessing of that curing agent. Consequently, the curing agent which, after decrystallization, remains liquid the longest is potentially the most trouble-free. All diamine curatives could be incorporated successfully save for Polacure 740M. The curing agent containing Polacure 740M readily crystallized upon cooling to 120°F, and did not yield a room-temperature-stable liquid. The Polacure 740M concentration had to be reduced from 21.6% down to an unacceptable 14.5% before a stable liquid curing agent could be obtained.

The elastomers resulting from the reduced Polacure concentration were inferior in cured physical properties to the remaining diamines used at the higher concentration. Polacure 740M was therefore dropped from further consideration. Long-term crystallization studies at 73-76°F were run on the remaining curing agents. The results are summarized as follows:

<u>CURING AGENT</u>	<u>AGING CHARACTERISTICS AT 73-76°F</u>
Polycure 1000	Crystallized after 16 hours
Cyanacure	Crystallized after 8 hours
Baytec PU-1604	Remained homogeneous liquid after 3 weeks exposure
Permapol D-562	Remained homogeneous liquid after 3 weeks exposure

As stated previously, a curing agent suitable for field use should remain in a liquid state for a minimum of eight hours at ambient temperatures. This minimum period allows for curing agent decrystallization and cool down prior to use. Under this criterion, the Cyanacure diamine formula would be only marginally acceptable but, because its basic performance properties were good, it was retained in the program.

Table 3 - Diamine containing curing agents with Permapol U-84 urethane polymer*.

DIAMINE COMPONENT	POLYURE 1000	POLYURE 1001	APOCURE 601P	CYANACURE	POLACURE 740M	BAYTEC PU-1604	PERMAPOL D-562
Mixed Viscosity (P at 75°F)	220	180	160	150	180	160	180
Application Life, minutes (time to 2500 P)	300	240	240	240	240	300	300
Hardness (Shore A)	44	45	44	45	44	45	44
Tensile Strength (psi)	340	355	320	475	218	370	350
Ultimate Elongation (%)	325	310	350	300	300	300	300
Tear Strength (lbs/in.)	53	62	46	55	38	51	59
Resistivity at 75°F Volume (G-cm) Surface (n)	6.3×10 ¹¹ 9.9×10 ¹²	5.8×10 ¹¹ 6.8×10 ¹²	3.1×10 ¹² 7.9×10 ¹³	9.9×10 ¹² 9.6×10 ¹³	9.4×10 ¹² 1.1×10 ¹⁴	3.0×10 ¹² 9.1×10 ¹³	8.6×10 ¹¹ 1.3×10 ¹³
Elastomer Color Clarity	Lt Amber, Clear	Lt Amber, Clear	Lt Amber, Clear	Lt Amber, Clear	Lt Amber, Clear	Very Dark, Clear	Lt Amber, Clear

4A 57 available isocyanate polymer based upon TDI and PPG polyethers (206 P viscosity) - PRC Corporation.

2A 57 available isocyanate polymer based upon TDI and PPG polyethers (206 P viscosity) - PRC Corporation.

Table 4 - Diamine containing curing agents with Permapol U-47 urethane polymer.*

DIAMINE COMPONENT	POLYURE		POLYURE		APOCURE		CYANACURE		POLACURE		BAYTEC		PERMAPOL	
	1000		1001		601P				740M		PU-1604		D-562	
Mixed Viscosity (P at 75°F)	320		260		220		290		320		300		300	
Application Life, minutes (time to 2500 P)	85		70		30		45		40		60		65	
Hardness (Shore A)	97		98		96		98		99		98		98	
Tensile Strength (psi)	46.0		4030		4775		3950		4850		4875		4440	
Ultimate Elongation (%)	250		250		250		250		250		250		250	
Tear Strength (lbs./in.)	480		405		485		345		545		490		469	
Resistivity at 75°F														
Volume (n-cm)	8.2x10 ¹³		5.1x10 ¹³		2.2x10 ¹⁵		3.7x10 ¹⁴		>1x10 ¹⁶		4.6x10 ¹⁵		7.2x10 ¹³	
Surface (Ω)	1.4x10 ¹⁴		2.7x10 ¹⁴		3.1x10 ¹⁵		2.0x10 ¹⁵		>1x10 ¹⁶		4.8x10 ¹⁵		3.6x10 ¹⁴	
Elastomer Color	Med Amber		Med Amber		Med Amber		Med Amber		Med Amber		Very Dark		Med Amber	
Clarity	Clear		Clear		Clear		Clear		Clear		Opaque		Clear	

*A 8.8% available isocyanate polymer based upon TDI and PPG polyethers (91 P viscosity) - PRC Corporation.

Table 5 - Polacure 740M containing curing agent with Adiprene L-100 and L-167 urethane polymers.*

	<u>ADIPRENE L-100</u>	<u>ADIPRENE L-167</u>
Mixed Viscosity (P at 75°F)	288	202
Application Life, minutes (time to 2500 P)	194	62
Hardness (Shore A)	58	84
Tensile Strength (psi)	2715	4470
Ultimate Elongation (%)	315	275
Tear Strength (lbs/in.)	285	485
Resistivity at 75°F		
Volume (Ω -cm)	2.3×10^{12}	3.6×10^{12}
Surface (Ω)	4.8×10^{13}	4.1×10^{13}
Elastomer Color	Medium Amber	Medium Amber
Clarity	Clear	Clear

*L-100, 4.08% available isocyanate (233 P viscosity).

L-167, 6.18% available isocyanate (88 P viscosity).

Table 6 - Final screening test of diamines with TDI/PTMG polymers.

	POLYCURE 1000	CYANACURE	BAYTEC PU-1604	PERMAPOL D-562
Mixed Viscosity (P at 75°F)				
L-100	350	380	380	250
L-167	110	120	90	70
L-200	90	115	110	110
Application Life, Hrs. (time to 2500 P)				
L-100	7	7	7	7
L-167	7.5	7	7	7
L-200	7	7	7	7
Hardness (Shore A)				
L-100	67	53	73	64
L-167	70	55	82	68
L-200	78	65	87	74
Tensile Strength (psi)				
L-100	3280	1375	1805	2815
L-167	3255	1340	2130	2585
L-200	3123	2560	2695	2765
Ultimate Elongation (%)				
L-100	630	600	800	600
L-167	600	600	775	550
L-200	500	500	600	500
(lbs/in.)				
L-100	178	87	172	153
L-167	187	105	244	143
L-200	205	150	295	185
Resistivity at 75°F Volume/Surface (Ω-cm/Ω)				
L-100	1.7×10 ¹⁰ /	4.7×10 ¹⁰ /	4.3×10 ¹⁰ /	2.2×10 ¹⁰ /
	3.4×10 ¹¹	1.3×10 ¹²	5.1×10 ¹¹	4.2×10 ¹¹
L-167	3.1×10 ¹⁰ /	1.5×10 ¹¹ /	1.3×10 ¹¹ /	4.8×10 ¹⁰ /
	5.4×10 ¹¹	5.7×10 ¹²	3.7×10 ¹²	7.9×10 ¹¹
L-200	9.3×10 ¹⁰ /	3.8×10 ¹¹ /	2.3×10 ¹¹ /	1.1×10 ¹¹ /
	1.4×10 ¹²	1.2×10 ¹³	5.7×10 ¹²	2.1×10 ¹²
Insulation Resistance (MΩ at 75°F)				
L-100	1.6×10 ³	3.8×10 ³	7.2×10 ³	1.2×10 ³
L-167	3.8×10 ³	8.2×10 ³	9.8×10 ³	7.9×10 ³
L-200	1.3×10 ³	5.1×10 ⁴	3.3×10 ⁴	1.6×10 ⁴
Elastomer, Color, Clarity				
L-100	Med Amber, Clear	Lt Amber, Clear	Very Dark, Opaque	Med Amber, Clear
L-167	Med Amber, Clear	Lt Amber, Clear	Very Dark, Opaque	Med Amber, Clear
L-200	Med Amber, Clear	Lt Amber, Clear	Very Dark, Opaque	Med Amber, Clear

Table 7- Diamine containing curing agents with Permapol U-56.*

DIAMINE COMPONENT	POLYURE		CYANACURE		BAYTEC PERMAPOL	
	1000				PU-1604	D-562
Mixed Viscosity (P at 75°F)	100		90		110	99
Application Life, Hrs. (time to 2500 P)	7		7		7	7
Hardness (Shore A)	65		72		85	60
Tensile Strength (psi)	815		735		1060	1100
Ultimate Elongation (%)	500		600		900	450
Tear Strength (lbs/in.)	130		165		220	130
Resistivity, at 75°F						
Volume (Ω -cm)	1.5×10^{11}		6.0×10^{11}		9.6×10^{11}	9.0×10^{11}
Surface (Ω)	2.7×10^{12}		1.4×10^{12}		2.6×10^{13}	2.3×10^{13}
Elastomer Color	Med Amber		Lt Amber		Very Dark	Med Amber
Clarity	Clear		Clear		Opaque	Clear

*A 7.1% available isocyanate polymer (91 P viscosity) - PRC Corporation.

Table 8 - MDI/PTMG polymer evaluation.*

	POLYURE 1000		CYANACURE		PERMAPOL D-562	
	VB-625	VB-635	VB-625	VB-635	VB-625	VB-635
Polymer Viscosity (P at 75°F)	300	215	300	215	300	215
Mixed Viscosity (P at 75°F)	255	190	--	--	250	180
Application Life, Hrs. (time to 2500 P)	1.50	1.00	0.30	0.20	1.00	0.75
Hardness (Shore A)	65	74	--	--	65	74
Tensile Strength (psi)	2605	3205	--	--	2565	3235
Ultimate Elongation (%)	500	350	--	--	400	380
Tear Strength (lbs/in.)	220	205	--	--	210	230
Resistivity at 75°F Volume (Ω cm) Surface (Ω)	2.6x10 ¹⁰ 6.2x10 ¹¹	3.8x10 ¹¹ 9.1x10 ¹²	--	--	8.2x10 ¹⁰ 2.8x10 ¹¹	2.7x10 ¹¹ 3.1x10 ¹²
Elastomer Color Clarity	Med Amber Clear	Med Amber Clear	--	--	Med Amber Clear	Med Amber Clear

*Uniroyal Chemical, VB-625, 6.36% available isocyanate.
VB-635, 7.73% available isocyanate.

Table 9 - Diamine containing curing agents with Permapol U-47.*

	<u>POLYCURE 1000</u>	<u>CYANACURE</u>	<u>PERMAPOL D-562</u>
Mixed Viscosity (P at 75°F)	140	155	130
Application Life, Hrs. (time to 2500 P)	2.5	1.0	2.75
Hardness (Shore A)	85	89	84
Tensile Strength (psi)	4895	4181	4645
Ultimate Elongation (%) 470	510	480	
Tear Strength (lbs/in.)	311	355	271
Elastomer Color Clarity	Med Amber Clear	Med Amber Clear	Dark Amber Clear
Resistivity, At 75°F			
Volume (Ω -cm)	2.6×10^{11}	1.9×10^{12}	3.4×10^{11}
Surface (Ω)	4.0×10^{12}	4.3×10^{13}	6.2×10^{12}
Insulation Resistance (M Ω at 75°F)	4.9×10^4	1.7×10^5	9.4×10^4
Power factor at 75°F			
1 kHz	0.093	0.093	0.090
1 MHz	0.056	0.059	0.056
Dielectric Constant at 75°F			
1 kHz	5.8	5.9	5.8
1 MHz	4.2	4.2	4.2

*An 8.8% available isocyanate based upon TDI/PPG - PRC Corporation.

Table 10 - Sound speed at 25°C.

<u>CURATIVE/POLYMERS</u>	<u>SOUND SPEED (METERS/SECOND)</u>	<u>ISOCYANATE CONTENT (%)</u>	<u>HARDNESS (SHORE A)</u>
<u>Polycure 1000</u>			
L-100	1604	4.1	67
L-167	1659	6.6	70
L-200	1718	7.5	78
U-47	1801	8.8	85
<u>Cyanacure</u>			
L-100	1599	4.1	53
L-167	1647	6.6	55
L-200	1714	7.5	65
U-47	1799	8.8	89
<u>Permapol D-562</u>			
L-100	1605	4.1	64
L-167	1658	6.6	68
L-200	1730	7.5	74
U-47	1819	8.8	84

Table 11 - General handling and performance data of final candidate encapsulants.

CURATIVE/POLYMER	POLYURE 1000		CYANACURE		PERMAPOL D-562	
	L-100	L-167	L-100	L-167	L-100	L-167
Mixed Viscosity (P at 75°F)	300	110	380	120	250	70
Application Life, Hrs. (time to 2500 P)	7	7.5	7	7	7	7
Hardness (Shore A)	67	70	53	55	64	68
Modulus (psi) at						
100% extension	263	237	176	141	269	173
200% extension	364	336	238	192	383	234
400% extension	770	758	389	320	854	431
500% extension	1582	1659	560	408	1817	686
Tensile Strength (PSI)	3280	3255	2899	2765	2815	2585
Ultimate Elongation (%)	630	600	700	650	600	625
Tear Strength (lbs./in.)	178	187	93	105	153	143
Compression Set (22 hrs/158°F), % Set	32	20	18	12	21	8
Adhesion to (lbs./in. width)*						
PVC	26	26	25	28	26	25
Neoprene	23	23	23	24	23	23
Stainless Steel	43	40	41	41	41	40
Transducer Ceramic	6	-	-	5	-	5
Volume Shrinkage (% volume decrease)	3.7	3.2	3.9	4.4	3.8	3.8

*See text for discussion.

Table 12 - General electrical properties of final candidate encapsulants.

CURATIVE/POLYMER	POLYURE 1000		CYANACURE		PERMAPOL D-562	
	L-100	L-167	L-100	L-167	L-100	L-167
Resistivity						
Volume, Ω -cm						
at 75°F	1.7×10^{10}	3.1×10^{10}	4.7×10^{10}	1.5×10^{11}	3.8×10^{11}	2.2×10^{10}
at 150°F	4.3×10^8	5.9×10^8	1.1×10^9	8.3×10^9	9.7×10^9	5.4×10^8
Surface, ohms						
at 75°F	3.4×10^{11}	5.4×10^{11}	1.3×10^{12}	5.7×10^{12}	1.2×10^{13}	4.2×10^{11}
at 150°F	1.1×10^{10}	8.3×10^9	1.0×10^{11}	1.9×10^{11}	9.3×10^{11}	1.0×10^{10}
Electrical Insulation						
Resistance, M Ω						
at 75°F	1.6×10^3	3.8×10^3	3.8×10^3	8.2×10^3	5.1×10^4	1.2×10^3
at 150°F	1.1×10^3	1.6×10^2	1.6×10^2	2.4×10^2	6.9×10^2	2.0×10^2
Power factor at 75°F						
at 1 kHz	0.094	0.088	0.068	0.080	0.097	0.106
at 1 MHz	0.092	0.075	0.088	0.074	0.056	0.089
Dielectric Constant at 75°F						
at 1 kHz	8.2	7.5	8.0	7.6	6.2	7.9
at 1 MHz	5.3	4.8	5.3	4.8	4.3	5.2
Sound Velocity at 25°F						
(m/sec)	1604	1659	1599	1647	1714	1605
Dielectric Strength						
V/mil, 125 mil thick sample	270	270	290	290	300	280

Table 13 - Physical hydrolytic stability (158°F/99% RH).

SYSTEM	INITIAL	5 DAYS					97 DAYS			% LOSS HARDNESS	
		19 DAYS	40 DAYS	54 DAYS	120 DAYS	SAMPLE STATE					
<u>Polycure 1000</u>											
Adiprene L-100	66	62	60	58	54	51	-22.7%				
Hardness, Shore A											
% Weight Change		+2.01	+2.06	+2.11	+2.32	+2.41					
Adiprene L-167	66	57	57	57	53	50	-24.2%				
Hardness											
% Weight Change		+1.76	+2.08	+2.18	+2.37	+2.44					
<u>Permapol D-562</u>											
Adiprene L-100	63	60	59	56	52	50	-20.6%				
Hardness											
% Weight Change		+1.67	+2.04	+1.93	+2.47	+2.38					
Adiprene L-167	63	57	56	55	51	50	-20.6%				
Hardness											
% Weight Change		+1.92	+2.25	+1.96	+2.70	+2.44					
<u>Cyanacure</u>											
Adiprene L-100	55	52	50	47	40	35	-36.4%				
Hardness											
% Weight Change		+1.63	+1.98	+1.78	+1.99	+2.06					
Adiprene L-167	54	46	45	44	35	32	-40.7%				
Hardness											
% Weight Change		+1.66	+2.08	+1.96	+2.15	+2.21					
Adiprene L-200	64	52	48	45	33	30	-53.1%				
Hardness											
% Weight Change		+1.94	+2.28	+2.13	+2.47	+2.56					

Table 14 - Resistance of electrical properties to wet or humid environments.

HIGH POTENTIAL RESISTANCE*		
(158°F/99% RH) (leakage value in microamperes)		
SYSTEM	INITIAL	AFTER 28 DAYS EXPOSURE
<u>Cyanacure</u>		
Adiprene L-100	0	0
Adiprene L-167	0	0
Adiprene L-200	0	0
<u>Polycure</u>		
Adiprene L-100	0	0
Adiprene L-167	0	0
<u>Permapol D-562</u>		
Adiprene L-100	0	0
Adiprene L-167	0	0

DIRECT WATER IMMERSION AT 122°F (50°C)				
(values in ohm-cm at 75°F)				
SYSTEM	INITIAL	7 DAYS	29 DAYS	60 DAYS EXPOSURE**
<u>Polycure 1000</u>				
Adiprene L-100	2.1×10^{10}	2.8×10^{10}	2.9×10^{10}	1.4×10^{10}
Adiprene L-167	3.5×10^{10}	7.5×10^{10}	8.1×10^{10}	2.3×10^{10}
<u>Cyanacure</u>				
Adiprene L-100	1.8×10^{10}	2.0×10^{10}	1.3×10^{10}	9.9×10^9
Adiprene L-167	6.1×10^{10}	5.2×10^{10}	4.9×10^{10}	2.1×10^{10}
Adiprene L-200	3.1×10^{11}	1.8×10^{11}	1.2×10^{11}	4.9×10^{10}
<u>Permapol D-562</u>				
Adiprene L-100	2.9×10^{10}	5.0×10^{10}	6.0×10^{10}	4.7×10^{10}
Adiprene L-167	5.7×10^{10}	8.9×10^{10}	1.3×10^{11}	3.2×10^{10}

ELECTRICAL INSULATION RESISTANCE***					
(Values in Megohms)					
SYSTEM	INITIAL	7 DAYS	14 DAYS	21 DAYS	28 DAYS
<u>Permapol D-562</u>					
Adiprene L-100	1.21×10^3	4.0×10^2	3.8×10^2	4.1×10^2	5.0×10^2
Adiprene L-167	7.00×10^3	1.1×10^3	1.1×10^3	1.0×10^3	1.0×10^3
<u>Polycure 1000</u>					
Adiprene L-100	1.55×10^3	6.0×10^2	5.0×10^2	6.5×10^2	6.5×10^2
Adiprene L-167	4.00×10^3	7.0×10^2	7.0×10^2	8.5×10^2	1.0×10^3
<u>Cyanacure</u>					
Adiprene L-100	3.2×10^3	3.6×10^2	3.6×10^2	4.5×10^2	4.5×10^2
Adiprene L-167	8.5×10^3	1.0×10^3	1.0×10^3	1.2×10^3	1.2×10^3
Adiprene L-200	5.6×10^4	3.3×10^3	3.0×10^3	3.6×10^3	3.2×10^3

* Electrical measurements determined after samples stabilized 2 hours at 75°F, 50 ±5.0% RH. Samples prepared and tested as per MIL-M-24041C, Paragraphs 4.7.4.5 and 4.7.4.6. Applied voltage is 2000 V rms using a Slaughter High Potential Leakage Tester Model 22/125-2.5. Minimum detectable leakage rate of 2 µA.

** Test values obtained after sample aged 2 hours, 72-77°F/50 ±5.0% RH.

*** Electrical measurements determined after samples stabilized 2 hours at 75°F/50 ±5.0% RH. Samples prepared and tested as per MIL-M-24041C.

Table 15 - Resistance to 60-day water immersion at 122°F (50°C)

<u>POLYCURE 1000</u>	<u>INITIAL</u>	<u>7 DAYS</u>	<u>14 DAYS</u>	<u>21 DAYS</u>	<u>29 DAYS</u>	<u>42 DAYS</u>	<u>60 DAYS</u>	<u>% CHANGE</u> <u>(INITIAL/FINAL)</u>
<u>Adiprene L-100</u>								
Hardness	67 A	68 A	68 A	64 A	64 A	64 A	62 A	-7.5
Modulus at								
100% extension	263 psi	263 psi	260 psi	261 psi	256 psi	263 psi	255 psi	-3.0
200%	364	383	348	356	346	363	349	-4.1
300%	515	524	476	492	466	495	475	-7.8
400%	770	809	705	753	663	757	704	-8.6
500%	1582	1570	1321	1465	1178	1465	1308	-17.3
540%	2567	--	--	--	--	--	--	--
550%	--	2501	2260	2265	1874	2575	2121	--
560%	2800*	2886	2654	--	--	--	--	--
570%	--	--	--	--	--	3248*	--	--
580%	--	2980	3173	--	--	--	2671*	--
590%	--	--	3800*	--	--	--	--	--
600%	--	3307*	--	3156*	3455*	--	--	--
<u>Adiprene L-167</u>								
Hardness	70 A	66 A	66 A	64 A	62 A	61 A	62 A	-11.4
Modulus at								
100% extension	237	215	207	--	215	237	230	-3.0
200%	336	311	295	--	318	349	317	-5.7
300%	466	433	440	--	459	520	430	-7.7
400%	758	664	689	--	743	810	610	-19.5
450%	--	--	--	--	1011	--	--	--
500%	1659	1210	1511	--	1805*	2217	1020	-38.5
510%	--	--	--	--	--	2486*	--	--
520%	--	--	1754*	--	--	--	--	--
540%	2636	2209	--	--	--	--	--	--
550%	--	--	--	--	--	--	1453	--
560%	2938*	--	--	--	--	--	2091*	--
600%	--	2407*	--	--	--	--	--	--
<u>Adiprene L-100</u>								
Hardness	68 A	68 A	68 A	65 A	65 A	64 A	65 A	-4.4
Modulus at								
100% extension	269 psi	274 psi	277 psi	281 psi	227 psi	244 psi	219 psi	-18.6
200%	383	395	382	391	306	329	299	-21.9
300%	534	564	553	587	420	445	418	-21.7
400%	854	985	876	965	637	680	605	-29.6
450%	--	--	--	1535	--	--	--	--
500%	1817	2349	2258	2958*	1348	1432	1138	-37.4
520%	--	3328*	3100*	--	--	--	--	--
550%	3742*	--	--	--	2333	2872	1879	-49.8
560%	--	--	--	--	--	3265*	--	--
570%	--	--	--	--	2928*	--	--	--
580%	--	--	--	--	--	--	2874*	--
<u>Adiprene L-167</u>								
Hardness	63 A	61 A	60 A	58 A	58 A	57 A	57 A	-9.5
Modulus at								
100% extension	173	210	208	211	208	206	217	+25.4
200%	234	304	290	315	290	292	320	+36.8
300%	314	454	438	433	440	440	481	-53.2
400%	431	756	811	695	778	755	872	+102.3
450%	--	--	--	--	1683	--	1394	--
500%	686	2099	2749*	1871	2572*	2226	2570*	+274.0
510%	--	--	--	--	--	2504*	--	--
520%	--	213*	--	2332*	--	--	--	--
600%	1621	--	--	--	--	--	--	--
650%	2293*	--	--	--	--	--	--	--

(continued)

Table 15 (continued)

CYANACURE	INITIAL	7 DAYS	14 DAYS	21 DAYS	29 DAYS	42 DAYS	60 DAYS	% CHANGE (INITIAL/FINAL)
Adiprene L-100								
Hardness	55 A	55 A	55 A	53 A	52 A	51 A	50 A	9.1
Modulus at								
100% extension	176 psi	163 psi	155 psi	158 psi	158 psi	155 psi	153 psi	-13.1
200%	238	216	207	209	214	209	203	-14.7
300%	303	267	265	265	270	263	253	-16.5
400%	389	369	348	357	365	344	318	-18.3
500%	560	549	521	511	571	498	451	-19.5
600%	1295	1436	1454	1554	1546	1236	939	-27.5
620%	2120	--	2180	2173*	2321*	--	--	
640%	--	2691*	2190*			2693*	--	
650%	--						1994	
670%	--						2443*	
700%	2899*							
Adiprene L-167								
Hardness	60 A	57 A	57 A	55 A	54 A	53 A	52 A	-13.3
Modulus at								
100% extension	141	204	200	218	152	145	171	+21.3
200%	192	284	270	317	216	195	224	+16.7
300%	241	381	380	463	288	260	285	+18.3
400%	320	639	592	762	405	354	388	+21.3
500%	408	989	1453	1700*	664	533	592	+45.1
530%	--	--	1878*		--	--	--	--
550%	--	1659			988	736	864	--
580%	--	1984*			--	--	--	--
600%	788				1830	1204	1440	+82.7
620%	--				2285*	1898*	--	
625%	--						2470*	
680%	2765*							
Adiprene L-200								
Hardness	62 A	60 A	57 A		54 A	53 A	52 A	-16.1
Modulus at								
100% extension	185 psi	205 psi	198 psi		192 psi	170 psi	181 psi	-2.2
200%	264	296	272		254	243	247	-6.4
300%	371	587	377		355	338	329	-11.3
400%	574	745	547		561	525	479	-16.6
500%	1385	1959*	1315		1299	1260	909	-34.4
530%	--		2232*		--	--	--	--
550%	2715				2522*	2488	1782	-34.4
560%	--					2742*	--	
570%	--						2341*	
600%	3311*							

*Ultimate elongation.

Table 16 - Resistance to 1000 hours castor oil immersion at 122°F (50°C).

							% CHANGE (INITIAL/FINAL)
<u>POLYCURE 1000</u>	<u>INITIAL</u>	<u>7 DAYS</u>	<u>14 DAYS</u>	<u>21 DAYS</u>	<u>29 DAYS</u>	<u>42 DAYS</u>	
<u>Adiprene L-100</u>							
Hardness	67 A	67 A	64 A	62 A	59 A	59 A	-11.90
Modulus at							
100% extension	263 psi	250 psi	254 psi	238 psi	199 psi	226 psi	-14.10
200%	364	371	372	356	309	367	+0.82
300%	515	559	543	547	460	589	+14.40
350%	--	--	--	--	--	772	--
400%	770	845	940	983	804	1164*	+51.20
450%	--	1396*	1381	1150*	1117	--	--
460%	--	--	--	--	1265*	--	--
480%	--	--	1829*	--	--	--	--
500%	1582	--	--	--	--	--	--
540%	2567	--	--	--	--	--	--
560%	2800*	--	--	--	--	--	--
<u>Adiprene L-167</u>							
Hardness	70 A	65 A	62 A	58 A	58 A	57 A	-13.60
Modulus at							
100% extension	237	194	157	--	179	179	-24.56
200%	336	290	299	--	283	290	-13.70
300%	466	422	447	--	413	426	-8.60
400%	758	647	702	--	671	735	-3.00
450%	--	--	1200	--	--	--	--
460%	--	--	1292*	--	--	--	--
500%	1659	1206	--	--	1299*	1538	-7.30
510%	--	--	--	--	--	1843*	--
550%	--	1919*	--	--	2098*	--	--
560%	2938*	--	--	--	--	--	--
<u>Adiprene L-100</u>							
Hardness	68 A	63 A	60 A		58 A	58 A	-14.7
Modulus at							
100% extension	269 psi	254 psi	250 psi	231 psi	211 psi	241 psi	-10.7
200%	383	390	370	347	317	402	+4.9
300%	534	567	579	550	402	586	+9.7
350%	--	--	--	--	667	867*	--
400%	854	926	1036	902	938*	--	+9.8
450%	--	1379*	1267*	1173*	--	--	--
500%	1817	--	--	--	--	--	--
550%	3742*	--	--	--	--	--	--
<u>Adiprene L-167</u>							
Hardness	63 A	60 A	55 A	54 A	50 A	50 A	-20.6
Modulus at							
100% extension	173	175	176	167	174	177	+2.3
200%	234	270	274	273	279	295	+26.1
300%	314	415	436	415	464	490	+56.1
350%	--	--	--	--	615	767*	--
400%	431	671	848	736	821*	--	--
450%	--	927	1131	1155*	--	--	--
480%	--	1093*	1587*	--	--	--	--
500%	686	--	--	--	--	--	--
600%	1621	--	--	--	--	--	--
650%	2293*	--	--	--	--	--	--

(continued)

Table 16 (continued)

	% CHANGE						
<u>CYANACURE</u>	<u>INITIAL</u>	<u>7 DAYS</u>	<u>14 DAYS</u>	<u>21 DAYS</u>	<u>29 DAYS</u>	<u>42 DAYS</u>	<u>(INITIAL/FINAL)</u>
<u>Adiprene L-100</u>							
Hardness	55 A	55 A	--	--	40 A	40 A	-27.3
Modulus at							
100% extension	176 psi	121 psi	120 psi	98 psi	90 psi	73 psi	-58.5
200%	283	177	166	151	138	120	-57.6
300%	303	221	242	228	218	185	-38.9
400%	389	317	330	333	339	283	-27.2
450%	--	--	--	--	495	--	--
500%	560	413	574	627	1353*	559	0.0
550%	--	--	--	1262*	--	1663*	--
570%	--	--	1343*	--	--	--	--
600%	1295	1048	--	--	--	--	--
640%	--	1470*	--	--	--	--	--
700%	2899*	--	--	--	--	--	--
<u>Adiprene L-167</u>							
Hardness	60 A	51 A	48 A	45 A	44 A	41 A	-31.7
Modulus at							
100% extension	141	221	207	203	110	110	-22.0
200%	192	323	319	318	183	180	-6.3
300%	241	514	508	465	261	280	+16.2
375%	--	--	--	743*	--	--	--
400%	320	842	980	--	419	451	+40.9
450%	--	1306	1415*	--	--	--	--
470%	--	1422*	--	--	--	--	--
500%	408	--	--	--	928	1149	+181.6
520%	--	--	--	--	--	1687*	--
530%	--	--	--	--	1350*	--	--
600%	788	--	--	--	--	--	--
680%	2765*	--	--	--	--	--	--
<u>Adiprene L-200</u>							
Hardness	62 A	60 A	57 A		52 A	50 A	-19.4
Modulus at							
100% extension	185 psi	155 psi	155 psi		129 psi	117 psi	-36.8
200%	264	223	224		191	158	-40.2
300%	371	331	324		280	288	-22.4
400%	574	503	485		462	497	-13.4
500%	1385	1050	1047		1073	1650	-19.1
510%	--	--	--		--	1845*	--
530%	--	--	1735*		1840*	--	--
550%	--	2125*	--		--	--	--
600%	3311*	--	--		--	--	--

*Ultimate elongation.

Table 17 - Resistance to 60-day water immersion at 122°F (50°C) (precut tensile samples).

		INITIAL	14 DAYS	29 DAYS	60 DAYS	% CHANGE (INITIAL/FINAL)		
ADIPRENE L-100								
Polycure 1000								
Modulus at								
100% extension (psi)	263	252*	(260)**	206	(215)	236	(239)	-10.2 (-9.1)
200%	364	340	(352)	295	(309)	318	(322)	-12.6 (-11.5)
300%	515	458	(474)	406	(426)	428	(433)	-16.9 (-15.9)
400%	770	647	(670)	602	(632)	606	(613)	-21.3 (-20.4)
500%	1582	1004	(1039)	984	(1030)	987	(997)	-37.6 (-37.2)
550%	---	1472	(1539)	1531	(1603)	1385	---	---
560%	2800†	---	---	---	---	---	---	---
600%	---	2468†	(2571)	2256	(2358)	1983	(2148)	---
620%	---	---	---	2911†	(3042)	---	---	---
625%	---	---	---	---	---	2355†	(2369)	---
Cyanacure								
Modulus at								
100% extension	176	160*	(169)**	136	(142)	152	(158)	-13.6 (-10.2)
200%	238	210	(223)	194	(202)	201	(209)	-15.5 (-12.2)
300%	303	280	(297)	254	(264)	250	(259)	-17.5 (-14.5)
400%	389	376	(399)	335	(348)	321	(333)	-17.5 (-14.4)
500%	560	576	(610)	513	(533)	450	(467)	-19.6 (-16.6)
600%	1295	1738	(1840)	1260	(1310)	1016	(1060)	-21.5 (-18.1)
620%	---	2079†	(2532)	---	---	---	---	---
650%	---	---	---	1898†	(1965)	2018†	(2025)	---
700%	2899†	---	---	---	---	---	---	---

* Sample dimensions determined after test exposure.

** Sample dimensions determined prior to test exposure.

† Ultimate elongation.

Table 18 - Resistance to 1000 hours castor oil immersion at 122°F (50°C).
(Precut Tensile Samples)

<u>ADIPRENE L-100</u>		<u>INITIAL</u>	<u>21 DAYS</u>		<u>42 DAYS</u>		<u>% CHANGE (INITIAL/FINAL)</u>
<u>POLYURE 1000</u>							
Modulus at							
100% extension (psi)		263	194* (216)**	176 (233)			-33.1 (-11.4)
200%		364	326 (364)	282 (373)			-22.5 (+ 2.5)
300%		515	475 (526)	418 (546)			-18.8 (+ 6.0)
400%		770	892 (965)	713 (947)			-7.4 (+23.0)
450%		--	1480† (1577)	--			
500%		1582†		1310† (1693)			
560%		2800†					
<u>Cyanacure</u>							
Modulus at							
100% extension		176	109* (124)**	76 (98)			-56.8 (-44.3)
200%		238	163 (185)	126 (162)			-47.1 (-31.9)
300%		303	240 (272)	195 (251)			-35.6 (-17.2)
400%		389	368 (415)	313 (403)			-19.5 (+ 3.6)
500%		560	654 (742)	668 (857)			+19.3 (+53.0)
550%		--	--	1244† (1659)			
600%		1295	1327† (1557)				
650%		--					
700%		2899†					

* Sample dimensions determined after test exposure.

** Sample dimensions determined prior to test exposure.

† Ultimate elongation.

Table 19 - Comparative test data precut vs fresh cut tensile specimens

	<u>% CHANGE BETWEEN INITIAL AND FINAL VALUES</u>			<u>STANDARD DEVIATION OF MODULI DATA</u>		
	<u>FRESH CUT</u>	<u>PRECUT</u>		<u>FRESH CUT</u>	<u>PRECUT</u>	
<u>POLYCURE 1000/ADIPRENE</u>						
<u>60-Day Water Immersion</u>						
Modulus at						
100% extension (psi)	-3.0*	-10.2*	(- 9.1)**	6.3	12.3*	(10.0)**
200%	-4.1	-12.6	(-11.5)	7.6	8.0	(4.7)
300%	-7.8	-16.9	(-15.9)	15.7	6.9	(6.9)
400%	-8.6	-21.3	(-20.4)	31.2	29.8	(23.5)
500%	-17.3	-37.6	(-37.2)	105.5	71.9	(59.3)
<u>1000-Hour Castor Oil Immersion</u>						
Modulus at						
100% extension	-14.10	-33.1	(-11.4)	7.1	79.6	(35.6)
200%	+ 0.82	-22.5	(+ 2.5)	21.3	42.1	(65.1)
300%	+14.40	-18.8	(+ 6.0)	47.9	21.4	(40.7)
400%	+51.20	-7.4	(+23.0)	75.0	46.1	(42.3)
<u>CYANACURE/ADIPRENE L-100</u>						
<u>60-Day Water Immersion</u>						
Modulus at						
100% extension	-13.1*	-13.6*	(-10.2)**	3.0	4.3*	(5.9)**
200%	-14.7	-15.5	(-12.2)	4.0	7.0	(5.0)
300%	-16.5	-17.5	(-14.5)	4.8	10.6	(12.3)
400%	-18.3	-17.5	(-14.4)	11.8	10.5	(15.3)
500%	-19.5	-19.6	(-16.6)	23.4	22.3	(30.7)
600%	-27.5	-21.5	(-18.1)	167.4	93.6	(80.0)
<u>1000-Hour Castor Oil Immersion</u>						
Modulus at						
100% extension	-58.5	-56.8	(-44.3)	6.8	4.0	(2.2)
200%	-57.6	-47.1	(-31.9)	4.6	8.0	(10.4)
300%	-38.9	-35.6	(-17.2)	11.0	19.1	(23.5)
400%	-27.2	-19.5	(+ 3.6)	30.4	37.3	(45.9)
500%	0.0	+19.3	(+53.0)	101.7	143.5	(169.7)

* Sample dimensions determined after test exposure.

** Sample dimensions determined prior to test exposure.

APPENDIX A

Trade Names/Trademarks and Their Affiliated Companies

Many of the raw materials used in this program carry a tradename or trademark which is registered with the United States Patent Office. These items are listed below:

- **ADIPRENE:**
E. I. du Pont de Nemours & Company, Elastomer Chemical Department
- **APOCURE:**
M & T Chemicals Incorporated
- **BAYTEC, DESMODUR W:**
Mobay Chemical Company, Polyurethane Division
- **CYANACURE:**
American Cyanamid Company, Elastomers Department
- **DB OIL:**
Caschem Company
- **ISONOL:**
The Upjohn Company, Polymer Chemicals Division
- **POLACURE:**
Polaroid Corporation, Chemical Commercial Development Department
- **POLYCURE:**
PTM&W Industries Incorporated
- **PERMAPOL:**
Products Research & Chemical Corporation
- **VIBRATHANE:**
Uniroyal Chemical Company
- **POLY bd:**
ARCO Chemical Company

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