

# Polyurethane Ionomers. I. Structure-Properties Relationships of Polyurethane Ionomers

S. L. HSU, H. X. XIAO, H. H. SZMANT, and K. C. FRISCH, *Polymer Institute, University of Detroit and Chemistry Department, University of Detroit, Detroit, Michigan 48221*

## Synopsis

The influence of chemical structure on mechanical properties of polyurethane ionomers (PU ionomers) has been examined. NCO-terminated prepolymers prepared from primarily 4,4-methylene bis(phenyl isocyanate) (MDI) and poly(oxytetramethylene) glycol (PTMO) were chain extended with tertiary amine-containing diols and the ionomers obtained by quaternization of the prepolymers. The *N*-methyldiethanolamine chain extender gave the best physical properties. The mechanical properties of the PU ionomers were improved with decreasing chain length of PTMO and with increasing concentration of quaternary ammonium centers (or NCO/OH ratio of PU prepolymers). A lower degree of quaternization resulted in a decrease in the mechanical properties of the resulting PU ionomers, but their properties could be improved by post-quaternization. The adhesion of the PU ionomers to aluminum and the glass transition temperature increased with increasing concentration of quaternizing centers.

## INTRODUCTION

Polyurethane (PU) ionomers are a relatively new class of special polyurethanes which contain associated ionic centers at different chain intervals. Both thermoplastic and thermoset PU ionomers have excellent mechanical properties due to the presence of coulombic forces and hydrogen bonding interaction between the chains.<sup>1-13</sup> The materials are of considerable scientific and commercial interest due to their unique structures and the fact that they could be utilized in the form of water dispersions for coatings and adhesives (e.g., as leather and textile finishing agents).

In spite of many studies of PU ionomers, relatively few references deal with a systematic investigation of the effect of chemical structure on the properties of PU ionomers.<sup>1-20</sup> This paper examines the effects of polyols, diisocyanates, tertiary amine-containing diols, and the chain lengths of the polyol on their mechanical properties. In addition, the influence of the degree of quaternization, post-quaternization, as well as the plasticization of ionomers by water, has also been investigated.

The effect of different quaternizing agents on the properties of the thermoplastic and thermoset PU ionomers will be reported in another paper.

## EXPERIMENTAL

**Raw Materials.** The raw materials are described in Table I. All of the polyols were dried and degassed overnight at 80°C under a vacuum of 3-5 mm Hg before use. The solvents (dioxane and acetone) were dried over 4-Å molecular sieves before use. Other materials were used as received.

TABLE I  
Raw Materials

Abbreviation	Description	Source
PTMO 650	Polyoxytetramethylene glycol MW = 658, OH = 170.6	Quaker Oats Co.
PTMO 1000	Polyoxytetramethylene glycol MW = 1013, OH = 110.8	Quaker Oats Co.
PTMO 2000	Polyoxytetramethylene glycol MW = 2036, OH = 55.4	Quaker Oats Co.
PPG 1000	Polyoxypropylene glycol MW = 1010, OH = 111.1	BASF Wyandotte Corp.
PCL	Polycaprolactone glycol MW = 1250, OH = 89.8	Union Carbide Corp.
MDI	4,4'-methylene bis(phenyl isocyanate)	Mobay Chemical Co.
TDI	Toluene diisocyanate (100 % 2,4 isomer)	Mobay Chemical Co.
HMDI	Methylene bis(4-cyclohexylisocyanate)	Mobay Chemical Co.
<i>N</i> -MDEA	<i>N</i> -Methyldiethanolamine	Aldrich Chemical Co.
BHBA	Bis(2-hydroxyethyl) benzylamine	Aldrich Chemical Co.
BHPA	Bis(2-hydroxypropyl)aniline	Upjohn Co.
P-DCX	$\alpha,\alpha'$ -Dichloro- <i>p</i> -xylene	Aldrich Chemical Co.
O-DCX	$\alpha,\alpha'$ -Dichloro- <i>o</i> -xylene	Aldrich Chemical Co.
BC	Benzyl chloride	Aldrich Chemical Co.
OA	Oxalic acid	Aldrich Chemical Co.
T-9	Stannous octoate	M&T Chemical Co.
—	Dioxane	Aldrich Chemical Co.
—	Acetone	Aldrich Chemical Co.

### Preparation of Tertiary Amine-Containing Linear Polyurethanes.

A 1000-mL four-necked reaction kettle equipped with a stirrer, nitrogen inlet, and thermometer was charged with a diisocyanate (MDI, TDI, or HMDI). The respective polyol (PTMO, PPG, or PCL) was then added slowly with stirring to yield prepolymers of different NCO/OH ratios. The NCO-terminated prepolymers were then chain-extended with tertiary amine-containing diols to obtain the linear polyurethanes.

The reactions were carried out under dry nitrogen at 70–75°C in the case of MDI, or at 80–85°C in the case of TDI and HMDI. When HMDI was used, 0.02% T-9 catalyst based on the prepolymer was added. After the theoretical NCO content was reached, as determined by di-*n*-butylamine titration, a mixture (3/1 by volume) of acetone and dioxane was added to lower the exothermic heat of reaction and to avoid a substantial increase in viscosity by the chain extending reaction. Stoichiometric amounts of tertiary amine-containing extenders were added to the diluted (about 50% concentration by weight) prepolymer solutions with high speed stirring. After the initial exothermic reaction had subsided, accompanied by an increase in viscosity, a small amount of T-9 catalyst (0.03% based on the weight of prepolymer) was added to insure the completion of the chain-extending reaction. The reaction was continued until the NCO content reached zero as evidenced by titration or when the IR peak for NCO had disappeared. These tertiary amine-containing linear polyurethane solutions were stored in glass bottles in preparation for quaternization.

**Quaternization.** A stoichiometric amount of quaternizing agent was added after determining the concentration of the tertiary amine-containing linear PU solution by removal of the solvent. The reaction was carried out at room temperature for 2–3 h until no further increase in viscosity was observed. The homogeneous solution was then degassed at room temperature under 2–3 mm Hg vacuum in order to remove any air bubbles. The degassed quaternized PU ionomer solution was cast in an aluminum mold which was previously treated with a release agent (RTV silicone rubber). After standing overnight at room temperature, the resulting sample film was heated in an oven first at 60°C for 4 h and then at 80°C for 16 h to complete the quaternization. The PU ionomer films were then heated overnight in an oven at 80°C and 2–3 mm Hg vacuum in order to remove completely any solvent before testing.

**Testing Methods.** The tensile strength, modulus and elongation at break were measured by means of an Instron Tensile Tester at a crosshead speed of 5 in./min, and the hardness by means of Shore A Durometer. (ASTM 2240-75).

The lap shear strength was obtained by means of an Instron Tensile Tester using aluminum plates 3 in. × 0.5 in. lapped 0.5 in. from their edges. (ASTM D-3163-73).

A TMS-2 Thermomechanical Analyzer (Perkin-Elmer) was used to determine the glass transition temperature of the PU ionomers at temperatures ranging from –100°C to +100°C and 0.01 mm of penetrating range, 100 g of penetrating weight, and 10°C/min of heating rate.

The water absorption was measured by placing the samples in water at room temperature for 1 week in order to examine the absorption and plasticization of the PU ionomers.

Post-quaternization was carried out on partially quaternized samples by immersion in 5 wt % of oxalic acid solution in water. The resulting samples were placed in an oven at 70°C for 3 h to remove the water before testing.

## RESULTS AND DISCUSSION

**Effect of Diisocyanate Structure.** PU ionomers made from MDI exhibited slightly better properties than those based on HMDI and TDI (Table II). MDI was, therefore, selected as the diisocyanate for the preparation of the subsequent PU ionomers.

**Effect of Polyol Structure.** As shown in Table III, three types of polyols

TABLE II  
Effect of Different Diisocyanates on Mechanical Properties of PU Ionomers<sup>a</sup>

Polyol <sup>b</sup>	Diisocyanate <sup>b</sup>	Quaternizer <sup>b</sup>	$T_g$ (psi)	$M_{100}$ (psi)	$M_{300}$ (psi)	$E$ (%)	$H$ (A)
PTMO 1000	MDI	p-DCX	2267	1591	—	150	83
PTMO 1000	TDI	p-DCX	2048	1517	—	130	83
PTMO 1000	HMDI	p-DCX	2023	1583	—	140	82

<sup>a</sup> Made from Polyol + MDI (NCO/OH=2/1), chain extended by tertiary amine-containing diol (NCO/OH=1/1).

<sup>b</sup> As described in Table I.

TABLE III  
Effect of Different Polyols on Mechanical Properties of PU Ionomers<sup>a</sup>

Type of polyol	MW of polyols	Quaternizer <sup>b</sup>	$T_s$ (psi)	$M_{100}$ (psi)	$M_{300}$ (psi)	$E$ (%)	$H$ (A)
PTMO	1013	p-DCX	2621	1547	—	157	80
PPG	1010	p-DCX	2067	1258	—	167	82
PCL	1250	p-DCX	4129	2529	—	192	86

<sup>a</sup> As described in Table II.

were used to prepare the PU ionomers. Polycaprolactone glycol (PCL) exhibited the best mechanical properties, presumably because of the hydrogen bonding by the ester groups and the high coulombic forces of the PU ionomers.<sup>5</sup> PTMO was chosen as the polyol for further studies due to its excellent resistance to hydrolysis.

A decrease in the molecular weight of PTMO enhances the concentration of tertiary amine groups in the PU backbone. The ionic crosslinking density produced by quaternization, the coulombic forces resulting from the ionic centers, and hydrogen bonding strengthened by the coulombic forces are all increased by a decrease in the molecular weight of PTMO since more tertiary amine-containing extender is present in the resulting PU backbone. Thus, the mechanical properties were enhanced as shown in Table IV and Figure 1.

**Effect of Tertiary Amine-Containing Chain Extender Structure.** In order to examine the effect of different tertiary amine-containing chain extenders on mechanical properties of PU ionomers, PTMO 1000, MDI, and NCO/OH of 2/1 were chosen to prepare the PU prepolymer and *p*-dichloroethylene (*p*-DCX) as the quaternizer. Bis(2-hydroxypropyl) aniline (BHPA), in which the tertiary amine is attached directly to the benzene ring, and which is a weaker base as compared to the other two types of tertiary amine-containing diols (*N*-MDEA and BHBA, see Table I), gave incomplete quaternization and very low crosslinking density. This resulted in increased elongation and decreased tensile strength, modulus, and hardness (Table V). The best properties were observed with *N*-methyldiethanolamine (*N*-MDEA) due to the strong basic properties of the tertiary amine bonded to a methyl group. Hence, *N*-MDEA was chosen as the chain extender for the introduction of tertiary amine groups into the PU backbone.

**Effect of Quaternary Ammonium Concentration.** When the NCO/OH ratio of the prepolymer prepared from PTMO 1000 and MDI increased from

TABLE IV  
Effect of Molecular Weight of Polyoxytetramethylene Glycols on Mechanical Properties of PU Ionomers<sup>a</sup>

MW	Quaternizer <sup>b</sup>	$T_s$ (psi)	$M_{100}$ (psi)	$M_{300}$ (psi)	$E$ (%)	$H$ (A)
658	p-DCX	5101	3339	—	159	89
1013	p-DCX	2057	1245	—	183	80
2025	p-DCX	1154	415	—	1188	72

<sup>a</sup> As described in Table II.

<sup>b</sup> As described in Table I.

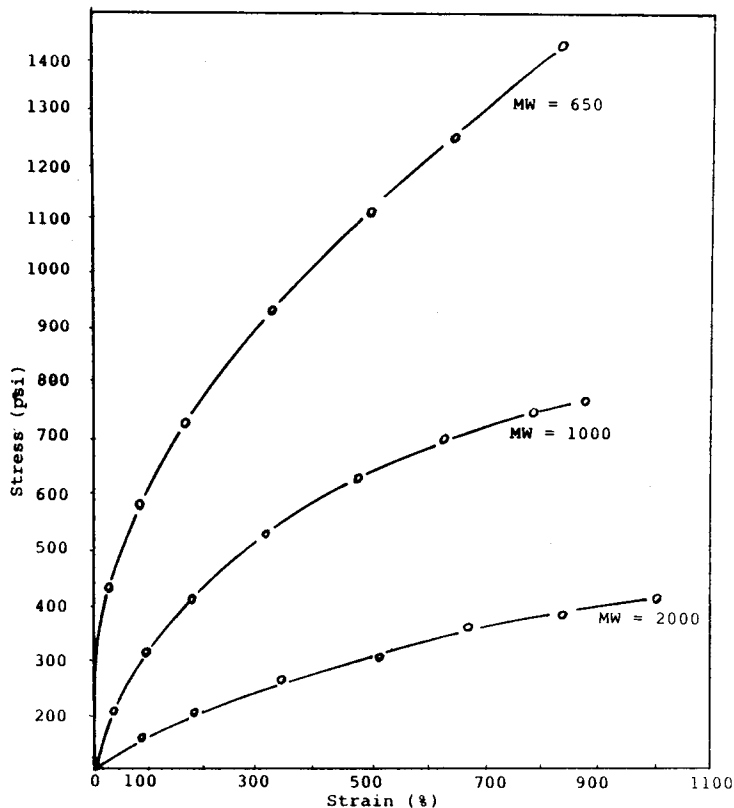


Fig. 1. Effect of molecular weight of PTMO on stress-strain properties of PU ionomers.

2.0 to 5.0, the concentration of quaternary ammonium increased from 0.85% to 2.05% due to a greater amount of *N*-MDEA being incorporated into the NCO-terminated prepolymer or quasi prepolymer. In consequence, the concentration of tertiary amine groups in the PU backbone and, of course, the concentration of quaternary ammonium centers after quaternization of the tertiary amines were increased with an increase of the NCO/OH ratios. As explained above, the mechanical properties were obviously improved as shown in Table VI and Figure 2.

**Effect of Degree of Quaternization (DQ).** As shown in Table VII and Figure 3, if the amount of quaternizer is smaller than the stoichiometric amount calculated from the concentration of tertiary amine groups in the

TABLE V  
Effect of Different Tertiary Amine-Containing Diols on Mechanical Properties of PU Ionomers<sup>a</sup>

Polyol	Tertiary amine <sup>b</sup> -containing diol	Quaternizer <sup>b</sup>	$T_g$ (psi)	$M_{100}$ (psi)	$M_{300}$ (psi)	$E$ (%)	$H$ (Å)
PTMO 1000	<i>N</i> -MDEA	p-DCX	2375	1791	—	169	80
PTMO 1000	BHBA	p-DCX	420	101	259	1650	50
PTMO 1000	BHPA	p-DCX	130	100	107	1500	40

<sup>a</sup> As described in Table II.

<sup>b</sup> As described in Table I.

TABLE VI  
Effect of Quaternary Ammonium Ion Concentrations on Mechanical Properties of PU Ionomers<sup>a</sup>

Polyol	NCO/OH	$-\overset{+}{N}^{\ominus}$ (%)	Tertiary amine- containing diol	Quaternizer	$T_g$ (psi)	$M_{100}$ (psi)	$M_{500}$ (psi)	$E$ (%)	$H$ (Å)
PTMO 1000	2.0	0.85	N-MDEA	p-DCX	2057	1245	—	184	80
PTMO 1000	3.0	1.41	N-MDEA	p-DCX	4935	4085	—	155	86
PTMO 1000	4.0	1.78	N-MDEA	p-DCX	5787	5543	—	117	88
PTMO 1000	5.0	2.05	N-MDEA	p-DCX	5863	—	—	94	95

<sup>a</sup> As described in Table II.

<sup>b</sup> % Quaternary ammonium ion concentration based on PU ionomers (by weight) was calculated as follows:

$$\frac{\text{MW of quaternary ammonium in backbone}}{\text{theoretical MW of PU ionomer}} = -\overset{+}{N}^{\ominus}\%$$

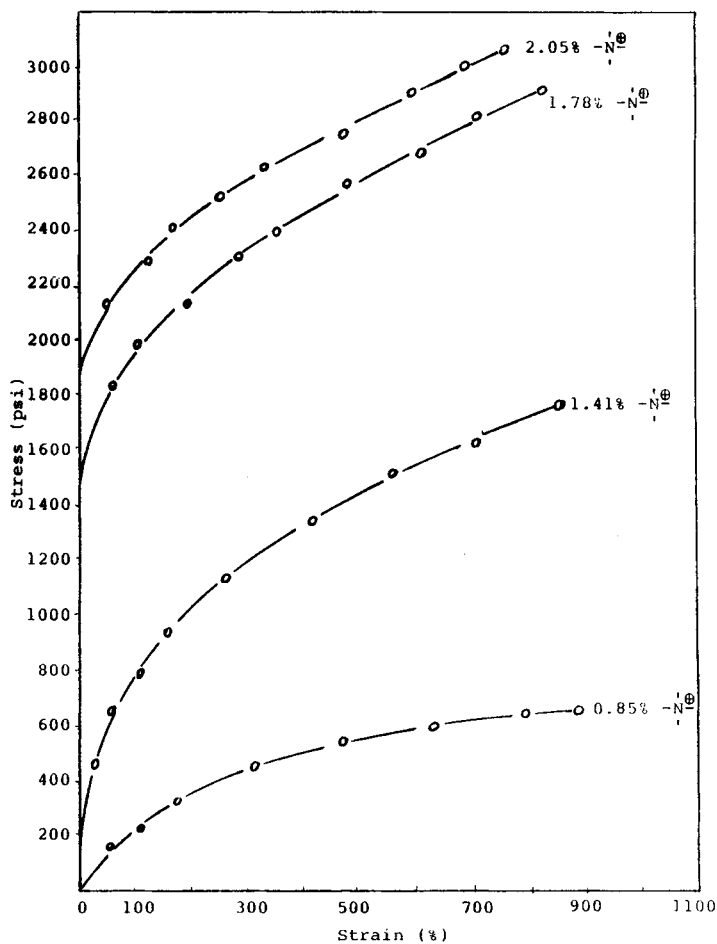


Fig. 2. Effect of different quaternary ammonium ion concentrations on stress-strain properties of PU ionomers.

PU backbone, the mechanical properties of the PU ionomers are weaker due to a decrease in the ionic crosslinking density, coulombic forces and hydrogen bonding. The theoretical degree of quaternization (DQ) in Table VII is calculated from the concentration of tertiary amine groups in the PU backbone.

**Effect of Post-Quaternization.** Samples with low DQ of 10% were post-quaternized by a 5% aqueous solution of oxalic acid. This resulted (Table VIII) in improvement of the mechanical properties due to additional quaternization. The mechanical properties approached those of the sample with 100% of DQ after a post-quaternizing time of about 30 h.

**Plasticization of PU Ionomers by Water.** PU ionomers can absorb water because of the hydrophilic ionic centers in the backbone. The weakened mechanical properties of samples immersed in water at room temperature for 1 week, were recovered after heating at 70°C for 3 h (Table IX). This implies that a decrease of mechanical properties is due to plasticization by water and the breaking and reforming of hydrogen bonds.

TABLE VII  
Effect of Degree of Quaternization on Mechanical Properties of PU Ionomers<sup>a</sup>

Polyol	NCO/OH	$\left  \text{N}^{\oplus} \right $ (%)	DQ <sup>c</sup> (%)	Quaternizer	$T_g$ (psi)	$M_{100}$ (psi)	$M_{300}$	$E$ (%)	$H$ (A)
PTMO 1000	3/1	1.41	5	BC	578	210	273	1037	72
PTMO 1000	3/1	1.41	10	BC	946	409	538	725	75
PTMO 1000	3/1	1.41	20	BC	2296	1071	1384	608	80
PTMO 1000	3/1	1.41	40	BC	3367	1900	2497	480	82
PTMO 1000	3/1	1.41	40	BC	3367	1900	2497	480	82
PTMO 1000	3/1	1.41	50	BC	4504	3012	3033	459	86
PTMO 1000	3/1	1.41	70	BC	6145	3842	5484	367	87
PTMO 1000	3/1	1.41	80	BC	5369	3187	4373	409	90
PTMO 1000	3/1	1.41	100	BC	4970	3473	4266	392	91
PTMO 1000	2/1	0.85	5	OA	89	89	71	1500	—
PTMO 1000	2/1	0.85	10	OA	128	106	117	1500	—
PTMO 1000	2/1	0.85	20	OA	858	185	267	1175	—
PTMO 1000	2/1	0.85	40	OA	2727	359	569	1167	—
PTMO 1000	2/1	0.85	60	OA	3453	579	956	959	—
PTMO 1000	2/1	0.85	80	OA	4115	748	1278	884	—
PTMO 1000	2/1	0.85	100	OA	4136	871	1416	817	—

<sup>a</sup> As described in Table II.

<sup>b</sup> As described in Table VI.

<sup>c</sup> DQ = degree of quaternization calculated from the concentration of  $\left| \text{N}^{\oplus} \right|$  in PU ionomers.



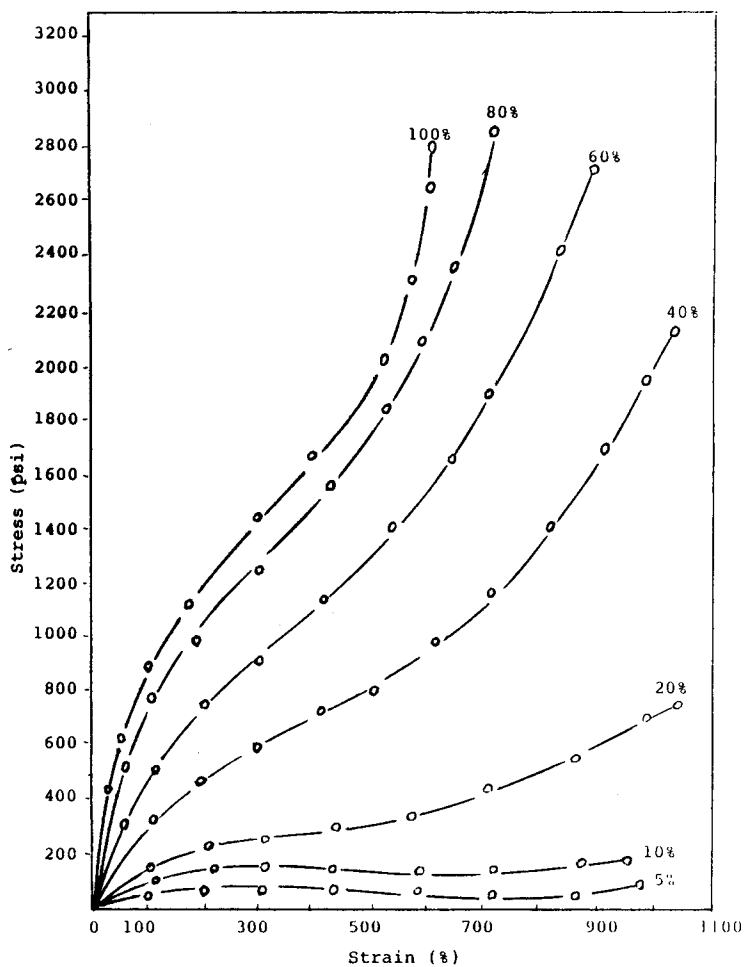


Fig. 3. Effect of different degree of quaternization on stress-strain properties of PU ionomers.

TABLE VIII  
Effect of Post-Quaternization on Mechanical Properties of PU Ionomers<sup>a</sup>

DQ % of sample <sup>b</sup>	Quaternizer <sup>c</sup>	Post-quaternization time (h) at RT	$T_g$ (psi)	$M_{100}$ (psi)	$M_{300}$ (psi)	$E$ (%)
10	OA	0	128	106	117	1500
10	OA	4	1910	380	598	680
10	OA	8	2393	499	745	715
10	OA	12	2778	565	874	705
10	OA	16	2778	595	893	745
10	OA	24	3630	642	1070	745
10	OA	30	3239	630	900	750
10	OA	36	3012	656	985	705

<sup>a</sup> As described in Table I.

<sup>b</sup> As described in Table VII.

<sup>c</sup> 5% aqueous solution (by weight) of oxalic acid.

TABLE IX  
Effect of Plasticization of PU Ionomers<sup>a</sup> by Water

Polyol	NCO/OH	DQ <sup>b</sup> (%)	Quaternizer	H <sub>2</sub> O, 1 week, RT			After immersion, 70°C for 3 h				
				$T_g$ (psi)	$M_{100}$ (psi)	$M_{300}$ (psi)	$T_g$ (psi)	$M_{100}$ (psi)	$M_{300}$ (psi)	$E$ (%)	
PTMO 650	2.0	100	p-DCX	1351	—	—	83	3865	3623	—	120
PTMO 1000	2.0	100	p-DCX	842	449	—	133	2267	1591	—	150
PTMO 2000	2.0	100	p-DCX	504	372	—	183	909	707	—	140

<sup>a</sup> As described in Table II.

<sup>b</sup> As described in Table VII.

**Glass Transition Temperature of PU Ionomers Having Different Quaternary Ammonium Concentrations.** PU ionomers of equal DQ exhibited an increase in the glass transition temperature ( $T_g$ 's) with an increase in the concentration of quaternary ammonium centers, as shown in Table X. The increase in the ionic crosslinking density and in coulombic forces decreases the mobility of the PU ionomer molecules at the glass transition temperature. In contrast to the above, at the same concentration of quaternary ammonium centers the  $T_g$ 's of PU ionomers decreased with decrease of DQ because the decrease in the ionic crosslinking density and coulombic forces allows an increase in the mobility of PU ionomers at the glass transition temperature.

**Effect of Degree of Quaternization on the Lap Shear Strength with Aluminum.** The lap shear strength to aluminium of PU ionomers with different DQ and concentrations of quaternary ammonium centers introduced by benzyl chloride were determined as shown in Table XI. The increase in the DQ and concentration of quaternary ammonium centers resulted in an increase in ionic crosslinking density, coulombic forces, and hydrogen bonding. All of these factors favor adhesion and result in an increase of lap shear strength.

### SUMMARY

The synthesis and some properties of PU ionomers were studied. The mechanical properties of PU ionomers were improved with decreasing molecular weight of the polyols and increase of the concentration of quaternary ammonium ions and degree of quaternization. The glass transition tem-

TABLE X  
Effect of DQ and Quaternary Ammonium Ion Concentrations on Glass Transition Temperatures of PU Ionomers<sup>a</sup>

Polyol	NCO/OH	$\text{—N}^{\oplus b}$ (%)	DQ <sup>b</sup> (%)	Quaternizer	Tertiary amine- containing diol	$T_g$ (°C)
PTMO 1000	2.0	0.85	100	BC	N-MDEA	-47
PTMO 1000	3.0	1.41	100	BC	N-MDEA	-42
PTMO 1000	4.0	1.78	100	BC	N-MDEA	-25
PTMO 1000	5.0	2.05	100	BC	N-MDEA	+23
PTMO 1000	4.0	1.78	5	BC	N-MDEA	-67
PTMO 1000	4.0	1.78	10	BC	N-MDEA	-60
PTMO 1000	4.0	1.78	20	BC	N-MDEA	-55
PTMO 1000	4.0	1.78	40	BC	N-MDEA	-53
PTMO 1000	4.0	1.78	50	BC	N-MDEA	-50
PTMO 1000	4.0	1.78	70	BC	N-MDEA	-48
PTMO 1000	4.0	1.78	80	BC	N-MDEA	-32
PTMO 1000	4.0	1.78	100	BC	N-MDEA	-25

<sup>a</sup> As described in Table II.

<sup>b</sup> As described in Table VII.

TABLE XI  
Effect of DQ and Quaternary Ammonium Ion Concentrations on Lap Shear Strength to Aluminum

Polyol	NCO/OH	$\begin{array}{c}   \\ -N^{\oplus} \\   \end{array}$ (%)	DQ (%)	Quaternizer	Tertiary amine- containing diol	Lap shear (psi)
PTMO 1000	2.0	0.84	100	BC	N-MDEA	402
PTMO 1000	3.0	1.41	100	BC	N-MDEA	623
PTMO 1000	4.0	1.78	100	BC	N-MDEA	812
PTMO 1000	5.0	2.05	100	BC	N-MDEA	827
PTMO 1000	4.0	1.78	5	BC	N-MDEA	174
PTMO 1000	4.0	1.78	10	BC	N-MDEA	305
PTMO 1000	4.0	1.78	20	BC	N-MDEA	353
PTMO 1000	4.0	1.78	40	BC	N-MDEA	495
PTMO 1000	4.0	1.78	50	BC	N-MDEA	628
PTMO 1000	4.0	1.78	70	BC	N-MDEA	664
PTMO 1000	4.0	1.78	80	BC	N-MDEA	796
PTMO 1000	4.0	1.78	100	BC	N-MDEA	812

\* As described in Table VII.

peratures of PU ionomers increased both with an increase in the quaternary ammonium ion concentration and with DQ. The properties of samples with low DQ could be improved by post-quaternization. The lap shear strength to aluminum increased with increasing DQ and quaternary ammonium ion concentration.

We would like to thank Mr. H. Hansen of the Department of Chemistry, University of Detroit, for his help in the instrumentation and Mr. E. F. Cassidy of the Department of Chemistry, State University of New York at Albany, for his assistance in the preparation of samples.

### References

1. Farbenfabriken Bayer A. G., Neth. Appl. 6,604,394 (1966); *Chem. Abstr.*, **66**, 65098 (1967).
2. Farbenfabriken Bayer A. G., Neth. Appl. 6,512,438 (1966); *Chem. Abstr.*, **65**, 9134 (1966).
3. H. Reiff and D. Dieterich, Ger. Offen. 2,019,324 (1971); *Chem. Abstr.*, **76**, 128117 (1972).
4. H. H. Neumaier, *Coated Fabrics Technol., Am. Assoc. Text. Chem Color. Symp.*, **62**, 70 (1973); *Chem. Abstr.*, **78**, 160793 (1973).
5. D. Dieterich, W. Keberle, and H. Witt, *Angew. Chem. Int. Ed.*, **9**(1),40 (1970).
6. D. Dieterich, W. Keberle, and R. Wuest, *J. Oil Col. Chem. Assoc.*, **63**, 363 (1970).
7. D. Dieterich, *Angew. Makromol. Chem.*, **26**, 85 (1972).
8. O. Lorenz, F. Havlena, and D. Kleborn, *Angew. Makromol. Chem.*, **33**, 159 (1973).
9. O. Lorenz and G. Rose, *Angew. Makromol. Chem.*, **45**(1), 85 (1975).
10. J. J. Kozak and P. Penczek, Pol. Pat. 84017 (1976).
11. O. Lorenz, H. J. August, H. Hick, and F. Triebs, *Angew. Makromol. Chem.*, **63**(1), 11 (1977).
12. O. Lorenz and H. Hugo, *Angew. Makromol. Chem.*, **72**, 115 (1978).
13. D. Dieterich, G. Balle, and H. Schmelzer, Ger. Offen. 2,735,013; *Chem. Abstr.*, **90**, 169568 (1979).
14. S. A. Sukhorukova, N. J. Levchenko, and A. P. Grekov, *Ukr. Khim. Zh. (Russ. Ed.)*, **47**(3), 286 (1981).
15. O. Lorenz, *FH-Texte*, **29**, 91 (1981); *Chem. Abstr.*, **96**, 70049 (1982).

16. O. Lorenz and K. H. Reinmoeller, *Kautsch. Gummi. Kunstst.*, **34**(10), 827 (1981); *Chem. Abstr.*, **96**, 7431 (1982).
17. Y. S. Lipatov, L. V. Karabarova, L. H. Sergeava, and E. Y. Gorichko, *Vysokomol. Soedin. Ser. A*, **24**(1), 110 (1982); *Chem. Abstr.*, **96**, 16360 (1982).
18. S. V. Laptil, V. N. Vatulev, Y. Y. Kercha, A. G. Kakovenk, V. V. Yoroshenko, A. P. Grekov, and I. Y. Litvin, *Vysokomol. Soedin. Ser.*, **23**(10), 760 (1981); *Chem. Abstr.*, **96**, 53111 (1982).
19. K. K. S. Hwang, C. W. Yang, and S. L. Cooper, *Polym. Eng. Sci.*, **21**(15), 1027 (1981).
20. D. Dieterich and H. Reiff, *FH-Texte*, **29**, 49 (1981); *Chem. Abstr.*, **96**, 36432 (1982).

Received September 27, 1983

Accepted January 4, 1984