

# Elastic Compositions Based on Polyurethane/ Aliphatic Polyesters

By

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## **Abstract:**

Aliphatic polyesters were prepared by melt condensation reaction of a dicarboxylic acid such as adipic and sebacic with different types of diol compounds in 1: 1.1 molar ratio. Ethylene glycol, di-, tri-, tetraethylene glycol and poly(ethylene glycol) with different molecular weights 1000, 4000, 6000 as well as the prepared hydroxy natural rubber were used as diol compounds. Polyurethane, with NCO/OH ratio equal 4, was synthesized from the reaction of toluene diisocyanate with poly(ethylene glycol)1000. The prepared polyurethane was mixed with different weight percentages of the prepared aliphatic polyesters. The film samples were tested mechanically, electrically and chemically. The results show that the weight percentage 10% of the added polyadipate or polysebacate glycols improves flexibility, electrical volume resistivity, dielectric constant and dielectric loss of unmodified rigid polyurethane film sample as well as reduces its swelling by aromatic solvents. All the above mentioned properties improve by increasing the hydrocarbon chain length of the glycol portion in the glycol used and the number of methylene in the aliphatic dicarboxylic acid. Compositions based on hydroxy natural rubber impart better properties than those based on ethylene glycols.

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**Key words:**

Adipic acid, sebacic acid, aliphatic polyesters, ethylene glycol, di-, tri-, tetra-ethylene glycol, poly(ethylene glycol), polyurethane, polyurethane/polyesters, elastic compositions.

**INTRODUCTION :**

The group of polyester polymers is one of the most versatile. They contain widely different materials with a large spectrum of characteristics and applications which are produced by a variety of manufacturing techniques<sup>(1-4)</sup>. Many linear diacids are available in order to provide varying size aliphatic chains for the polyester structure. Adipic acid is one of the most frequently used linear diacids since it gives an excellent compromise in terms of technical performance and price<sup>(5)</sup>. Succinic acid was used to manufacture aliphatic polyesters with biodegradability, useful for bottles of food and films<sup>(6)</sup>. Diacids with much longer chains are significantly more expensive, thus they are limited to specific applications. They are used for their extremely good mechanical qualities such as their resistance towards gritting or elongation<sup>(7)</sup>. Aliphatic polyesters are at present the usual materials used as flexible segments in the synthesis of polyurethane elastomers. Aliphatic polyesters have glass transition temperatures below room temperature and are low melting point solid or liquids. They are amorphous, and the molecular weight range used is from about 600 to about 3000<sup>(8)</sup>. The main objective of this work is the preparation of elastic compositions based on polyurethane/aliphatic polyesters to be applied as engineering sheets in construction field. Also the modification of some epoxidized polyurethane and polyester resins or its derivatives based on linseed oil was used as petroleum pipeline coatings<sup>(9, 10)</sup>

- **Experimental:**

- **Materials used:**

- **Adipic Acid**, produced by Fluka Chemicals Co.,  $C_6H_{10}O_4$  content 99%, melting point 151-154 °C, molecular weight 146.14.
- **Sebacic Acid**, produced by Fluka Chemicals Co.,  $C_{10}H_{18}O_4$  content 97%, melting point 131-133 °C, molecular weight 202.25.
- **Ethylene Glycol EG**, produced by Prolabo Co., average molecular weight of 62.07, density 1.112 g. cm<sup>-3</sup>, melting point -13 °C.
- **Natural Rubber NR**, Malizian rubber in the form of rubber smoked sheets RSS-1, density  $0.913 \pm 0.005$  g. cm<sup>-3</sup> and moony viscosity ML(1+4) at 100 °C, 60-90.
- **Poly(ethylene Glycol) PEG**, produced by Fluka Chemicals Co., with different molecular weight 1000, 4000, 6000, and melting point 37-40, 59-61 or 60-63 °C respectively.
- **Toluene diisocyanate TDI**, produced by FOB Sigma Co., is a mixture of 80 % 2,4 and 20 % 2,6 toluene diisocyanate,  $C_9H_6N_2O_2$ , with average molecular weight of 174, boiling point 129-133 °C, freezing point 110 °C, density 1.225 g. cm<sup>-3</sup>.
- All solvents used were pure grade.

- **Methods:**

- **Preparation of hydroxylated natural rubber<sup>(11)</sup>:**

A solution of natural rubber (5 g in 200 ml carbon tetrachloride) was introduced into a three necked round bottom flask 500 ml fitted with a dropping funnel, thermometer and mechanical stirrer. Bromine 4 ml (12 g) was charged into the dropping funnel and added slowly over a period of about 1h with continuous stirring. The temperature was maintained at -5 °C in an ice bath for 90 min till orange colored solution appeared. The resinous layer was separated from the solution using separating funnel, washed three times with distilled water

dried in an oven at 100 °C to evaporate the solvent giving brominated natural rubber.

Potassium hydroxide (4 g in 13 ml methanol) was added slowly over a period of about 1h into a three necked round bottom flask 500 ml contained the prepared brominated natural rubber solution (5 g in 200 ml cyclohexanone), with continuous stirring at 150-160 °C in an oil bath for 8 h. The liquid layer was separated from the solution, washed with distilled water and dried in an oven at 100 °C to evaporate the solvent giving hydroxylated natural rubber. Characteristics of the prepared hydroxylated natural rubber are as follows: specific viscosity in acetone at 20°C:0.1570<sup>(12)</sup>, total iodine value:26.33 according to ASTM D1541-86, hydroxyl group value:530<sup>(13)</sup> according to ASTM D4274-94 and absorption band at 3300-3500 cm<sup>-1</sup> for hydroxyl groups<sup>(14)</sup>.

• **Preparation of different aliphatic polyester types<sup>(5, 13)</sup> :**

Polyesters were prepared using dicarboxylic acid (adipic as well as sebacic) and different types of diol compounds in 1:1.1 molar ratio. Adipic acid 0.25 mol (36.5 g) or sebacic acid 0.25 mol (50.6 g), ethylene glycol 0.275 mol (17.1 g), and p-toluenesulfonic acid (0.012 g) as a catalyst were introduced into a three necked round bottom flask 250 ml fitted with a Dean-Stark tube, reflux condenser, mechanical stirrer, thermometer and nitrogen inlet tube. The flask was heated slowly at 150 °C under a gentle nitrogen stream with stirring for 30 min. The temperature was raised to 180-200°C and the reaction was carried out at 6-8h with continuous stirring, to ensure that water was completely eliminated during this period. By the same method diethylene glycol DEG, triethylene glycol TEG, tetraethylene glycol TTEG, poly(ethylene glycol) PEG with different molecular weight (1000, 4000, 6000) and the prepared hydroxylated natural rubber were also used as diol compounds. The producing compounds

were recrystallized from methanol giving a series of aliphatic polyesters.

Characteristics of the prepared polyethylene glycols and hydroxy natural rubber adipate as well as those of sebacate, such as color, melting point, specific viscosity<sup>(12)</sup>, acid number<sup>(13)</sup>, hydroxyl values<sup>(13)</sup> and molecular weight<sup>(15)</sup> are given in Tables 1,2, respectively.

• **Infrared spectra**<sup>(14)</sup>:

IR-spectra of the prepared polyester samples were recorded using ATI Mattson-Genesis Series FTIR<sup>TM</sup> infrared spectroscopy. Absorption band of IR-spectra of the prepared adipate and sebacate polyesters with different types of diol compounds are shown in Tables 1,2, respectively.

**Table 1. Characteristics of the prepared adipate polyesters:**

Sample Symbol	Polyester Compounds	Color	Mp, °C	Specific Viscosity, $\eta_{sp}$ In Acetone, at 20 °C <sup>(12)</sup>	Acid number value, mg KOH/1g <sup>(13)</sup>	Hydroxyl Group Value, mg KOH/1g <sup>(13)</sup>	Molecular Weight, Mn <sup>(15)</sup>	IR-absorption for (-COO ester group), cm <sup>-1</sup> <sup>(14)</sup>
P <sub>1</sub>	Polyethylene glycol adipate	White	60	0.1570	18.1	27.0	2487	1736
P <sub>2</sub>	Polydiethylene glycol adipate	Yellow	58	0.2103	8.6	32.3	2743	1732
P <sub>3</sub>	Polytriethylene glycol adipate	Yellow	52	0.2832	5.8	38.3	2542	1732
P <sub>4</sub>	Polytetraethylene glycol adipate	Yellow	54	0.2937	4.0	35.2	2861	1732
P <sub>5</sub>	Polyethylene glycol(1000) adipate	Yellow	50	0.3295	2.1	33.3	3165	1732
P <sub>6</sub>	Polyethylene glycol(4000) adipate	Brown	66	0.3493	3.7	30.6	3265	1736
P <sub>7</sub>	Polyethylene glycol(6000) adipate	Yellow	84	0.3600	5.9	25.5	3564	1729
P <sub>8</sub>	Polyhydroxy natural rubber adipate	Black	120	0.0360	20.7	13.9	3240	1729

**Table 2. Characteristics of the prepared sebacate polyesters:**

Sample Symbol	Polyester Compounds	Color	Mp, °C	Specific Viscosity, $\eta_{sp}$ In Acetone, at 20 °C <sup>(12)</sup>	Acid number value, mg KOH/1g <sup>(13)</sup>	Hydroxyl Group Value, mg KOH/1g <sup>(13)</sup>	Molecular Weight, M <sub>n</sub> <sup>(15)</sup>	IR-absorption for (-COO ester group), cm <sup>-1</sup> <sup>(14)</sup>
PE <sub>9</sub>	Polyethylene glycol sebacate	White	78	0.1907	4.8	48.0	2124	1739
PE <sub>10</sub>	Polydiethylene glycol sebacate	White	76	0.2059	5.9	41.5	2364	1728
PE <sub>11</sub>	Polytriethylene glycol sebacate	Yellow	74	0.2467	3.1	39.1	2657	1735
PE <sub>12</sub>	Polytetraethylene glycol sebacate	White	69	0.2661	7.6	32.1	2825	1732
PE <sub>13</sub>	Polyethylene glycol(1000) sebacate	White	60	0.2911	9.8	27.1	3035	1731
PE <sub>14</sub>	Polyethylene glycol(4000) sebacate	Brown	76	0.3323	4.6	29.9	3247	1732
PE <sub>15</sub>	Polyethylene glycol(6000) sebacate	Yellow	82	0.3961	3.1	30.7	3315	1732
PE <sub>16</sub>	Polyhydroxy natural rubber sebacate	BLack	115	0.0716	17.9	16.7	3235	1737

• **Preparation of polyurethane PU<sup>(16)</sup>:**

Polyurethane with molar ratio NCO/OH equal 4 was prepared by addition of toluene diisocyanate 0.08 mol (14 g) to a solution of poly(ethylene glycol) PEG with molecular weight 1000 (20 g in 100 ml carbon tetrachloride) into a three necked round bottom flask 250 ml fitted with a dropping funnel, thermometer and mechanical stirrer. The addition was dropwise in a period of 30 min with continuous stirring for 2h at 60 °C. The obtained homogenous viscous solution was then poured into a petridish and placed in a rotatory evaporator under reduced pressure to evaporate the solvent giving rigid unmodified polyurethane film.

- **Preparation of PU/PE samples<sup>(17, 18)</sup>:**

Toluene diisocyanate 0.08 mol (14 g) was added dropwise to a solution of poly(ethylene glycol) with molecular weight 1000 (20 g in 100 ml carbon tetrachloride) into a three necked round bottom flask 250 ml fitted with a dropping funnel, thermometer and mechanical stirrer. The addition was dropwise in a period of 30 min with continuous stirring for 2h at 60 °C. Then the prepared adipate or sebacate polyester samples in the following percentage (2, 4, 6, 8, 10 or 12 % with respect to the reactant polyurethane weight) were added dropwise to the prepared homogenous polyurethane solution. The obtained homogenous viscous solutions were then poured into petridishes and placed in a rotatory evaporator oven under reduced pressure to evaporate the solvent giving polyurethane/polyester film. Mechanical, dielectric properties and chemical resistances of different PU/PE compositious film samples (with approximately thickness 3-4 mm measured by micrometer) were determined and compared with those of the unmodified PU film sample as tabulated in Tables 3, 4, 5, 6 and 7, respectively.

- **Tests and evaluation of the prepared PU/PE film samples:**

- **Tensile strength and Elongation at break:**

Tensile strength values and elongation percentages of the prepared film samples were carried out according to ASTM D2240-81 using Instron testing machine (Model 1026) at  $23\pm 2^{\circ}\text{C}$  with cross head speed of 100 mm/min., chart speed of 200 mm/min and a load cell range of 0-500 Newton. Tensile strength values and elongation percentages data of the prepared PU/PE 2, 4, 6, 8, 10 or 12% film samples were measured and compared with those of the unmodified PU with NCO/OH=4 (c.f. Tables 3, 4).

• **Dielectric Properties** <sup>(19-21)</sup>:

Dielectric constant, dielectric loss, alternating current AC conductivity as well as volume resistivity of the prepared film samples were measured at 20 °C and different frequencies (100, 120, 1000 or 10000 Hz) by using LCR-meter bridge type SR (Stanford Research System) 720.

- **Dielectric Constant (specific inductive capacity):**

Dielectric constant ( $\epsilon'$ ) is obtained by measuring the capacity (C) of the sample from the equation:

$$\epsilon' = C / C_0$$

Where:

C : is the measured capacity of the sample, and

C<sub>0</sub> : is the capacity of air, which calculated from the equation:

$$C_0 = \epsilon_0 \cdot A / d$$

Where:

$\epsilon_0$  : is constant of free space dielectric permittivity =  $8.58 \times 10^{-12}$

f/m,

d : is the thickness of the sample in mm, and

A : is the cross-sectional area of the electrode in mm<sup>2</sup>.

-**Dielectric loss:**

Tangent of the loss angle ( $\tan \delta$ ) was measured directly using the above mentioned bridge, in different frequencies.

- **Alternating current AC Conductivity:**

The electrical conductivity ( $\sigma$ ) was calculated from the equation:

$$\sigma = \epsilon_0 \cdot \epsilon' \cdot \omega \cdot \tan \delta$$



Where:

$\omega$  : is the frequency  $f$  of the alternating current Applied in Hz, It was calculated from the following equation:

$$\omega = 2 \pi \cdot f$$

**-Electrical Resistivity:**

The electrical resistivity ( $\rho$ ) was calculated also from the equation:

$$\rho = 1 / \sigma$$

**N.B.:** in this case the sample in cylinder shape so it has circuit surface area calculated by means of its radius ( $r$ ) from the following equation:

$$A = \pi \cdot r^2$$

The dielectric properties of the prepared modified PU/PEs 10% film samples were measured and compared with those of the unmodified PUs with NCO/OH=4 (c.f. Tables 5, 6).

• **Water and chemical resistance<sup>(22)</sup>:**

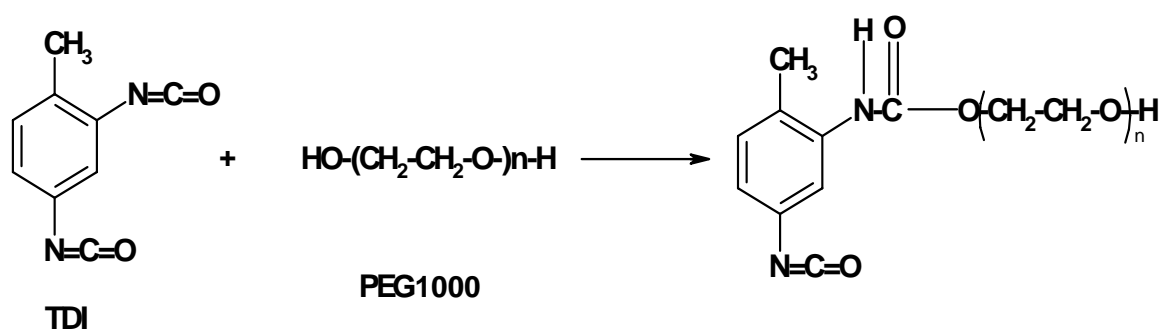
Water and chemical resistances of the prepared PU/PE 10% film samples was carried out at 28°C according to ANSI/ASTM D543-67 (c.f. Table 7).

• **Results and discussion:**

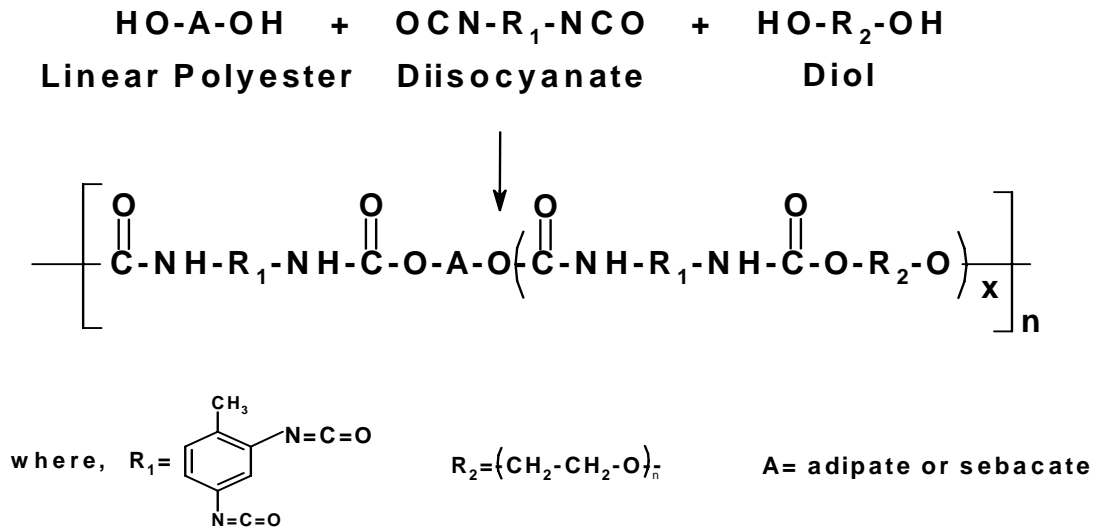
Polyurethane elastomers have unique combination of performance and application properties<sup>(23)</sup>. They provide excellent mechanical, adhesion properties and solvent resistance. This combination property is not possessed by any other commercial rubber or plastic<sup>(1, 3)</sup>. Thus, chemical modification process has led to opportunities for new compositions with very useful properties for many and varied end-use applications<sup>(22, 23)</sup>.

In the present work, the prepared aliphatic polyesters were mixed in different weight percentages with the prepared PUs by NCO/OH ratio of 4 (samples represented by different groups in tables 3,4). Unmodified PU samples were also prepared for the purpose of comparison (sample represented by PU).

The preparation of polyurethane NCO/OH= 4 involves the reaction of toluene diisocyanate TDI with poly(ethylene glycol) PEG with n=1000 as a polyol:



Throughout the mixing process of PUs with aliphatic polyester chains at 60 °C, the attacking species (e.g. isocyanate groups) contain sites for proton acceptance and electron acceptance in proper steric relationship<sup>(8)</sup>. This will permit the following reaction:



The structure of PU/PE compositions were elucidated and confirmed by infrared spectra<sup>(10)</sup> as given in Figs. 1, 2 which show absorption bands at the following wave length ( $\text{cm}^{-1}$ ) 3357(NH), 1660(C=O), 1729-1736 (-COO ester group) and 1200-1300 for (-CO ester group). The intensity of these bands sharply increase with the increase in the hydrocarbon chain length in the glycol portion of polyethylene glycol 6000 adipate as illustrated in Fig.2. as an example. Samples were tested mechanically, electrically and chemically (as indicated in Tables 3-7) to be evaluated as engineering sheets in construction field:

### **Tensile strength and elongation at break percentage:**

Tensile strength and elongation at break percentage of the tested samples were investigated. The data in Tables 3, 4 indicate that:

PU samples provide the greatest tensile strength values and the least flexibility comparatively. This may be attributed to the very strong physical chemical bonds, which can be formed through the crosslinking formation of PUs<sup>(8)</sup>. Also the chains sliding over each other is restricted by the presence of polar groups under an applied stress leading to the least elongation

comparatively. Therefore at NCO/OH ratio equal to 4, there is an incremental increase in tensile strength values with a sharp decrease in flexibility<sup>(8)</sup>.

The prepared aliphatic PEs improve PUs stiffness properties based on following facts.

The intrinsic chain flexibility is determined by the nature of the chain backbone and by the nature of groups directly attached to it<sup>(25)</sup>. This means that film samples based on aliphatic C-C bond are quite flexible (group samples PE<sub>1</sub>- PE<sub>8</sub>, PE<sub>9</sub>- PE<sub>16</sub>).

Flexibility is also affected by the number of -CH<sub>2</sub>- units in aliphatic ester chain, increasing the number of -CH<sub>2</sub>- units in the ester chain can improve the flexibility of the film samples. Thus, one can conclude that film samples of PU/PE based on sebacate impart greater flexibility than those based on adipate ester<sup>(7)</sup>.

All glycol compounds contribute flexibility to the system. Flexibility increases with increasing chain length of the glycol used. Thus the PU/PE group samples can be arranged in the following order of elongation percentages:

PU/PE<sub>7</sub>> PU/PE<sub>6</sub>> PU/PE<sub>5</sub>> PU/PE<sub>4</sub>> PU/PE<sub>3</sub>> PU/PE<sub>2</sub>> PU/PE<sub>1</sub>> PU (for polyglycol adipate)

also,

PU/PE<sub>15</sub>> PU/PE<sub>14</sub>> PU/PE<sub>13</sub>> PU/PE<sub>12</sub>> PU/PE<sub>11</sub>> PU/PE<sub>10</sub>> PU/PE<sub>9</sub>> PU (for polyglycol sebacate )

Polyester samples based on hydroxy natural rubber (PE<sub>8</sub>, PE<sub>16</sub>) for both adipic and sebacic, respectively impart the greatest flexibility as compared with those based on glycol chains.

The aliphatic polyester weight percentages added to PU, through mixing process, also play a significant role in determining the flexibility of the film samples. It was found that polyester in 10% is better than that in 2% in this respect, imparts the greatest flexibility as compared with 2, 4, 6 and 8%.

On the other hand, beyond 10% (e.g. 12%), the samples begin to deteriorate leading to a destruction in their mechanical properties. Thus we can say that 10% is the optimum weight percentage of a polyester through mixing process with polyurethane of NCO/OH=4.

### **Electrical properties:**

The electrical properties of PU/PE 10% at 20°C and different frequencies were studied and compared with those of PUs. The obtained results in Tables 5, 6 indicate the following:

PU films have little volume resistivity as compared with modified PU/PE samples. Since their composition based on the hydrocarbon chain of the glycol portion of poly(ethylene glycol) 1000 as a polyol decreases their polar nature and some hydrophilic character leading to some insulation resistance<sup>(8)</sup>.

Prepared PE films with different hydrocarbon chain length of the glycol portion of both polyglycol adipate and sebacate improve the electrical volume resistivity of PUs with different degrees. This can be attributed to the fact that the chemical composition of the polyol used can affect electrical properties due to its water absorption properties.

It was found that the longer the hydrocarbon chain of the glycol portion of a polyglycol adipate or sebacate imparts excellent insulation resistance, of PUs composition, due to its hydrolytic stability<sup>(8)</sup>.

Prepared PE films based on hydroxy natural rubber improve the insulation resistance of PUs composition with the highest degree. This may be due to chemical structure of NR being based on all carbon-carbon linked atoms<sup>(1)</sup>.

For all tested samples values of resistivity or conductivity are influenced by frequency used in Hz. In general, the values of resistivity decrease and values of conductivity increase with increasing frequency of applied current  $10^4$  on the sample. This can be attributed to the fact that, increasing frequency of current applied increases the mobility of intermolecular segment chains, and fragments (molecules and /or ions)<sup>(19)</sup>.

### **Dielectric constant ( $\epsilon$ ):**

PU film samples, with little volume resistivity, have high dielectric constants as compared with PU/PE tested samples with higher glycol chains as well as with hydroxy natural rubber. This can be attributed to the fact that the polar nature of PUs leads to a higher dielectric constant and this limits their use as high frequency electrical insulators<sup>(25)</sup>. This result is in agreement with the fact that materials to be employed as insulators in electrical applications should have low dielectric constants, whereas those to be employed as conductors or semiconductors should exhibit high dielectric constants<sup>(12)</sup>.

For all tested samples, the dielectric constants are independent of the frequency at low to moderate frequencies but are dependent on the frequency at high frequencies<sup>(12)</sup>.

**Dielectric loss ( $\tan \delta$ ):**

Dielectric loss in polymers  $-\tan \delta-$  depends on the chemical constitution of the repeating unit in the chain.

For PU/PE film samples, the value of dielectric loss decreases with increasing the hydrocarbon chain length of the glycol portion and also with using hydroxy natural rubber, as compared with dielectric loss values of PU samples. This is can be attributed to the fact that indroduction of very long hydrocarbon segment in the main chain always sharply reduces molecular mobility as compared with PU sample<sup>(19)</sup>.

The applied current frequency also affects the value of dielectric loss ( $\tan \delta$ )<sup>(19)</sup>, it was found that for all tested samples, the values of dielectric loss decrease with increasing frequency of applied current on the sample (e.g.  $10^4$  Hz). This decrease was sharp for PU/PE samples as compared with PU tested samples.

**Water resistance:**

Water resistance of PU and PU/PE 10% samples was studied after immersion in water at 28°C for 4h. The data given in Table 7 indicate that:

PUs show good water resistance since its composition based on the hydrocarbon chain of the glycol portion of poly(ethylene glycol)1000 as a polyol which in turn decreases its polar nature<sup>(8)</sup>. Also increasing isocyanate index  $\text{NCO/OH}=4$  increases their hydrolytic stability<sup>(22)</sup>. Thus, all modified aliphatic polyester samples show excellent water stability at 28°C for 4h.

**Chemical resistance:**

Samples were also subjected to chemical resistance test, at 28°C for 6 months, by direct immersion in alkaline or acidic media as well as organic solvents. The obtained data in Table 7 indicate that:

The resistance of PUs to immersion in dilute solutions of NaOH, KOH, H<sub>2</sub>SO<sub>4</sub> and HCl is relatively poor. This may be attributed to the fact that, alkaline or acidic media degrade PUs<sup>(8)</sup>.

Increasing the hydrocarbon chain length of the glycol portion in the glycol used as well as using hydroxy natural rubber as a diol improve the resistance of PU/PE samples to alkaline or acidic media at 28°C for 6 months as compared with PU samples.

PU with NCO/OH=4 show solvent stability, since increasing NCO/OH reduces their swelling by a solvent toluene<sup>(20)</sup> and increases their stability in other solvents used.

PU/PE film samples show solvent stability.

**• Conclusion:**

Tensile strength and elongation percentage are a function of polyester composition, its structure and its molecular weight.

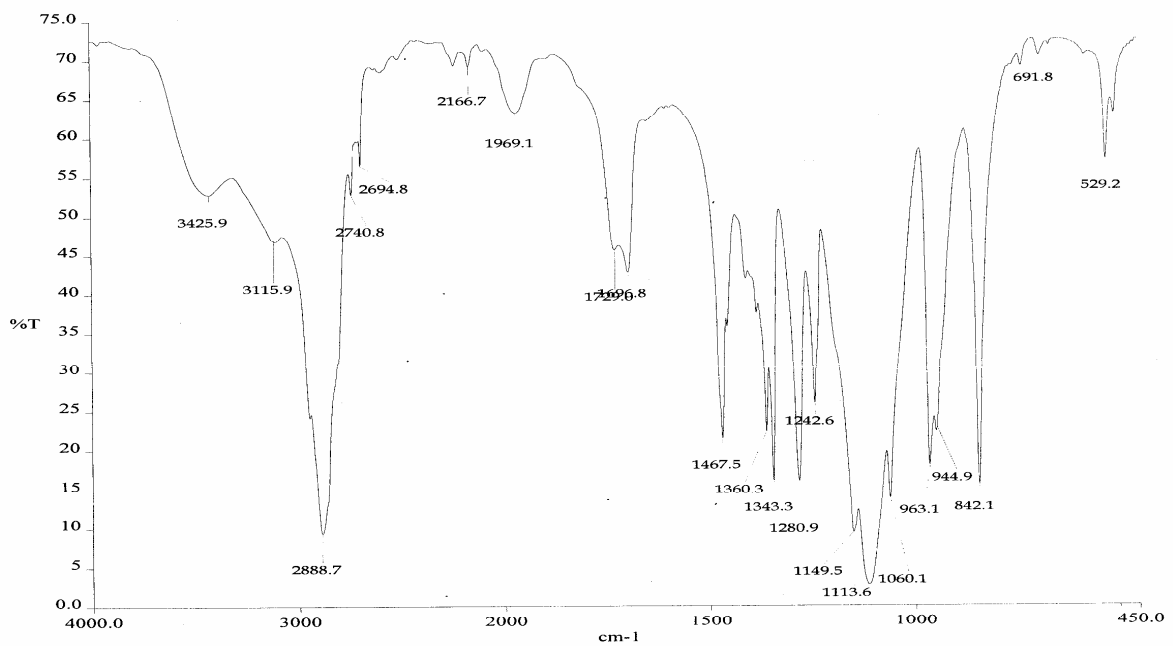
The aliphatic polyester weight percentage 10% added to PU through mixing process, is the optimum one in the mechanical and electrical properties.

Increasing the hydrocarbon chain length of the glycol portion as well as the number of methylene in the aliphatic dicarboxylic acid improve flexibility, electrical volume resistivity of rigid polyurethane as well as reducing the swelling by aromatic solvents.

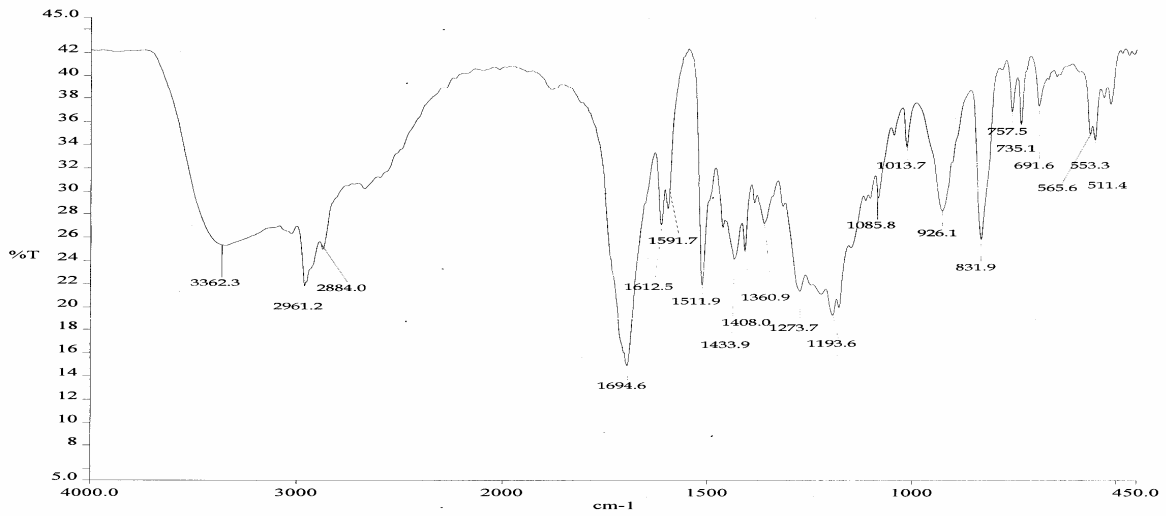


Composition based on PU/Poly(ethylene glycol)6000 of sebacic acid with NCO/OH=4 are characterized with higher flexibility, excellent insulation properties, hydrolytic stability and chemical resistance as compared with unmodified PU samples.

Composition based on hydroxy natural rubber impart better mechanical and electrical properties than those based on ethylene glycol.



**Fig.1. IR-spectra of unmodified PU film sample.**



**Fig.2. IR-spectra of PU/Polyethylene glycol 6000 adipate 10% film sample.**

**Table 3. Tensile strength and elongation at break percentage of the prepared PU/PE adipate samples with film thickness, 3-4mm:**

Group	Polyurethane, NCO/OH=4, %	Adipate polyester, %	Mechanical properties	
			Tensile Strength, Kg/Cm <sup>2</sup>	Elongation at Break, %
PU	100	---	84.24	85
PU/PE <sub>1</sub>	98	2	91.46	95
	96	4	102.56	110
	94	6	109.37	130
	92	8	154.51	145
	90	10	166.67	155
	88	12	185.68	100
PU/PE <sub>2</sub>	98	2	103.12	100
	96	4	109.37	115
	94	6	128.12	135
	92	8	156.25	155
	90	10	174.37	165
	88	12	196.14	105
PU/PE <sub>3</sub>	98	2	106.25	105
	96	4	118.75	125
	94	6	140.26	140
	92	8	166.67	165

	90	10	188.14	175
	88	12	205.36	115
<b>PU/PE<sub>4</sub></b>	98	2	109.37	110
	96	4	125.00	125
	94	6	150.00	140
	92	8	184.37	160
	90	10	202.50	180
	88	12	218.45	120
<b>PU/PE<sub>5</sub></b>	98	2	120.26	110
	96	4	132.25	115
	94	6	164.67	125
	92	8	205.25	130
	90	10	226.55	135
	88	12	241.26	115
<b>PU/PE<sub>6</sub></b>	98	2	130.78	105
	96	4	151.12	110
	94	6	178.37	120
	92	8	210.45	125
	90	10	245.24	130
	88	12	268.48	110
<b>PU/PE<sub>7</sub></b>	98	2	142.45	100
	96	4	165.11	105
	94	6	187.54	110
	92	8	229.41	115
	90	10	258.14	120
	88	12	269.35	105
<b>PU/PE<sub>8</sub></b>	98	2	107.15	105
	96	4	112.75	110
	94	6	125.26	125
	92	8	134.67	130
	90	10	151.24	135
	88	12	174.34	110

**Table 4. Tensile strength and elongation at break percentage of the prepared PU/PE sebacate samples with film thickness, 3-4mm:**

Group	Polyurethane, NCO/OH=4, %	Sebacate polyester, %	Mechanical properties	
			Tensile Strength, Kg/Cm2	Elongation at Break, %
<b>PU</b>	100	---	84.24	85
<b>PU/PE<sub>9</sub></b>	98	2	98.65	100
	96	4	103.86	115
	94	6	111.37	135
	92	8	162.61	160
	90	10	172.62	175
	88	12	184.75	120
<b>PU/PE<sub>10</sub></b>	98	2	105.18	105
	96	4	112.47	120
	94	6	133.62	130
	92	8	164.24	170
	90	10	183.31	180
	88	12	201.12	110
<b>PU/PE<sub>11</sub></b>	98	2	107.23	110
	96	4	123.45	130
	94	6	149.46	140
	92	8	173.87	175
	90	10	195.34	190
	88	12	212.35	120
<b>PU/PE<sub>12</sub></b>	98	2	114.57	130
	96	4	128.32	150
	94	6	153.14	175
	92	8	188.34	190
	90	10	209.62	205
	88	12	223.38	135
<b>PU/PE<sub>13</sub></b>	98	2	128.41	105
	96	4	137.63	110
	94	6	170.85	115
	92	8	212.74	120
	90	10	231.51	125
	88	12	252.46	110
<b>PU/PE<sub>14</sub></b>	98	2	138.42	100
	96	4	159.32	105
	94	6	185.18	110
	92	8	217.96	115
	90	10	253.41	120
	88	12	272.68	105
<b>PU/PE<sub>15</sub></b>	98	2	148.61	95
	96	4	170.12	100
	94	6	193.63	105
	92	8	234.52	110

	90	10	262.41	115
	88	12	289.23	100
<b>PU/PE<sub>16</sub></b>	98	2	109.15	120
	96	4	118.76	130
	94	6	129.94	145
	92	8	138.83	150
	90	10	157.81	170
	88	12	170.35	125

**Table 5 : Dielectrical Properties of the prepared PU/PE adipate 10% film samples, at 20°C and different frequencies:**

<b>PU/PE Type (NCO/OH =4)</b>	<b>Frequency, Hz</b>	<b>Resistivity (<math>\rho \times 10^{-8}</math>) <math>\Omega.m</math></b>	<b>Conductivity (<math>\sigma \times 10^8</math>) <math>\Omega^{-1}.m^{-1}</math></b>	<b>Dielectric Constant (<math>\epsilon</math>)</b>	<b>Dielectric Loss (<math>\tan \delta</math>)</b>
<b>PU</b>	<b>10 0</b>	0.473	2.1155	0.2816	14.11
	<b>12 0</b>	0.467	2.1428	0.2782	11.75
	<b>10<sup>3</sup></b>	0.450	2.2243	0.2455	1.68
	<b>10<sup>4</sup></b>	0.251	3.9882	0.2175	0.34
<b>PU/PE<sub>1</sub></b>	<b>10 0</b>	0.478	2.092	0.2988	13.45
	<b>12 0</b>	0.468	2.137	0.2908	11.18
	<b>10<sup>3</sup></b>	0.442	2.262	0.2612	1.63
	<b>10<sup>4</sup></b>	0.235	4.255	0.2334	0.38
<b>PU/PE<sub>2</sub></b>	<b>10 0</b>	1.425	0.702	0.3638	3.69
	<b>12 0</b>	1.387	0.721	0.3509	3.17
	<b>10<sup>3</sup></b>	0.974	1.027	0.2587	0.77
	<b>10<sup>4</sup></b>	0.345	2.899	0.1996	0.28
<b>PU/PE<sub>3</sub></b>	<b>10 0</b>	1.459	0.6854	0.2324	5.67
	<b>12 0</b>	1.407	0.7106	0.2244	4.72
	<b>10<sup>3</sup></b>	1.123	0.8904	0.1603	1.03
	<b>10<sup>4</sup></b>	0.381	2.6254	0.1202	0.41
<b>PU/PE<sub>4</sub></b>	<b>10 0</b>	1.524	0.656	0.2321	5.73
	<b>12 0</b>	1.462	0.684	0.2214	4.86
	<b>10<sup>3</sup></b>	1.174	0.852	0.1589	1.74

	$10^4$	0.416	2.404	0.1188	0.56
<b>PU/P<sub>5</sub></b>	$\frac{10}{0}$	2.109	0.474	0.2008	4.41
	$\frac{12}{0}$	2.064	0.484	0.1968	3.86
	$10^3$	1.654	0.605	0.1664	0.74
	$10^4$	0.472	2.119	0.1475	0.32
<b>PU/PE<sub>6</sub></b>	$\frac{10}{0}$	4.321	0.231	0.1874	3.74
	$\frac{12}{0}$	3.621	0.276	0.1825	3.18
	$10^3$	3.114	0.321	0.1604	0.57
	$10^4$	0.485	2.062	0.1335	0.26
<b>PU/PE<sub>7</sub></b>	$\frac{10}{0}$	7.352	0.136	0.1867	2.54
	$\frac{12}{0}$	6.354	0.157	0.1774	2.11
	$10^3$	4.568	0.219	0.1541	0.49
	$10^4$	0.508	1.969	0.1325	0.19
<b>PU/PE<sub>8</sub></b>	$\frac{10}{0}$	9.346	0.1070	0.1601	1.24
	$\frac{12}{0}$	8.921	0.1121	0.1561	1.11
	$10^3$	5.216	0.1917	0.1222	0.29
	$10^4$	0.561	1.7826	0.1168	0.21

**Table 6: Dielectrical Properties of the prepared PU/PE sebacate 10% film samples, at 20°C and different frequencies:**

PU/PE Type (NCO/OH =4)	Frequency, Hz	Resistivity ( $\rho \times 10^{-8}$ ) $\Omega.m$	Conductivity ( $\sigma \times 10^8$ ) $\Omega^{-1}.m^{-1}$	Dielectric Constant ( $\epsilon$ )	Dielectric Loss ( $\tan \delta$ )
PU	10 0	0.473	2.1155	0.2816	14.11
	12 0	0.467	2.1428	0.2782	11.75
	10 <sup>3</sup>	0.450	2.2243	0.2455	1.68
	10 <sup>4</sup>	0.251	3.9882	0.2175	0.34
PU/PE <sub>9</sub>	10 0	0.484	2.0652	0.2946	13.39
	12 0	0.470	2.1274	0.2862	11.15
	10 <sup>3</sup>	0.456	2.1950	0.2576	1.58
	10 <sup>4</sup>	0.240	4.1729	0.2296	0.32
PU/PE <sub>10</sub>	10 0	1.436	0.6966	0.3588	3.60
	12 0	1.398	0.7155	0.3499	3.16
	10 <sup>3</sup>	0.985	1.0148	0.2476	0.76
	10 <sup>4</sup>	0.359	2.7848	0.1986	0.26
PU/PE <sub>11</sub>	10 0	1.485	0.673	0.2451	5.71
	12 0	1.405	0.712	0.2368	4.76
	10 <sup>3</sup>	1.122	0.891	0.16158	1.11
	10 <sup>4</sup>	0.374	2.674	0.12168	0.48
PU/PE <sub>12</sub>	10 0	1.568	0.638	0.2265	5.68
	12 0	1.475	0.678	0.2108	4.84
	10 <sup>3</sup>	1.196	0.836	0.1564	1.68
	10 <sup>4</sup>	0.425	2.353	0.1168	0.54
PU/PE <sub>13</sub>	10 0	2.116	0.4725	0.1996	4.39
	12 0	2.081	0.4805	0.1954	3.80
	10 <sup>3</sup>	1.675	0.5969	0.1616	0.69
	10 <sup>4</sup>	0.471	2.1245	0.1443	0.28
PU/PE <sub>14</sub>	10 0	4.325	0.231	0.1868	3.68

	120	3.657	0.273	0.1812	3.12
	10 <sup>3</sup>	2.154	0.464	0.1584	0.55
	10 <sup>4</sup>	0.498	2.008	0.1324	0.23
PU/PE <sub>15</sub>	100	7.658	0.131	0.1784	2.45
	120	6.487	0.154	0.1728	2.02
	10 <sup>3</sup>	4.854	0.206	0.1425	0.45
	10 <sup>4</sup>	0.515	1.942	0.1296	0.17
PU/PE <sub>16</sub>	100	9.489	0.1015	0.1618	1.247
	120	8.996	0.1034	0.1578	1.123
	10 <sup>3</sup>	5.351	0.1789	0.1236	0.297
	10 <sup>4</sup>	0.5784	1.7714	0.1177	0.224

**Table 7. Water and chemical resistances of the prepared PU/PE adipate and sebacate 10% film samples:**

PU/PE Type (NCO/OH=4)	Water resistance at 28°C for 4h	Chemical resistance at 28°C for 6 months											
		Alkalis		Acids		Solvents							
		NaOH 1%	KOH 1%	H <sub>2</sub> SO <sub>4</sub> 3%	HCl 3%	Toluene	Xylene	Acetone	Butanone	Methanol	Ethanol	Chloroform	Petroleum ether
PU	G	P	P	P	P	F	F	G	G	G	G	G	G
PU/PE <sub>1</sub>	E	P	P	P	P	G	G	G	G	G	G	G	G
PU/PE <sub>2</sub>	E	P	P	P	P	G	G	G	G	G	G	G	G
PU/PE <sub>3</sub>	E	P	P	P	P	G	G	G	G	G	G	G	G
PU/PE <sub>4</sub>	E	P	F	F	P	G	G	E	G	G	G	E	G
PU/PE <sub>5</sub>	E	F	F	F	P	G	E	E	E	E	E	E	G
PU/PE <sub>6</sub>	E	F	F	F	F	G	E	E	E	E	E	E	G
PU/PE <sub>7</sub>	E	F	F	G	G	E	E	E	E	E	E	E	E
PU/PE <sub>8</sub>	E	G	G	G	G	E	E	E	E	E	E	E	E
PU/PE <sub>9</sub>	E	P	P	P	P	G	G	G	G	G	G	G	G
PU/PE <sub>10</sub>	E	P	P	P	P	G	G	G	G	G	G	G	G
PU/PE <sub>11</sub>	E	P	P	P	P	G	G	E	G	G	G	G	G
PU/PE <sub>12</sub>	E	F	P	P	F	E	G	E	E	E	E	E	E
PU/PE <sub>13</sub>	E	F	F	F	F	E	E	E	E	E	E	E	E
PU/PE <sub>14</sub>	E	F	F	F	F	E	E	E	E	E	E	E	E
PU/PE <sub>15</sub>	E	G	G	G	G	E	E	E	E	E	E	E	E
PU/PE <sub>16</sub>	E	G	G	G	G	E	E	E	E	E	E	E	E

E : Excellent , G : Good , F : Fair, P : Poor



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