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DIELECTRIC BEHAVIOUR OF POLYURETHANE COATINGS DERIVED FROM PALM OIL OLEIC ACID-BASED POLYOLS – A RENEWABLE RESOURCE

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ABSTRACT

Dielectric relaxation spectroscopy (frequency range from 100 Hz to 40 MHz) was used to examine molecular motion in polyurethane (PUR) derived from palm oil oleic acid-based polyols at room temperature. PUR was prepared by varying the oleic acid content in the polyol (28%, 40% and 65%), while the NCO/OH ratio of PUR was varied to 1.2, 1.4 and 1.6. The effect of chemical contribution on dielectric behaviour of the samples was described. Dielectric parameters were in the range of 2.0 to 3.0 for real permittivity ε' *and 0.02 to 0.08 for imaginary permittivity,* ε''*. A Cole-Cole plot was fitted using the Havriliak-Negami model, and curve-fitting simulations were performed using Origin software program. A* γ*-relaxation process was proposed as the probable mechanism for the dielectric behaviour of PUR. The dielectric constant and loss mechanism of the material were dependent on the NCO/OH ratio and the percentage of oleic acid content in the polyols used in synthesising PUR. The frequency-dependent conductivity of PUR materials was analysed using a Jonscher's power-law expression, and the plot exhibited a DC plateau and a frequency-dependent region.*

Keywords: polyurethane coatings, oleic acid, polyols, dielectric properties, renewable resource.

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INTRODUCTION

Polymers exhibit a variety of electrical properties reflecting their structure and molecular motion. Most polymers are highly insulating material because of the saturated chemical bonding which consumes all electrons. Probing the dielectric response of polymers provides valuable information with respect to morphology and phase changes, molecular structure, polarisation, dipole motions and their mutual influence on the properties of the material (Tsangaris and Psarras, 1999; Riande and Diaz-Calleja, 2004; Psarras *et al*., 2006; Kontos *et al*., 2007). Generally, dielectric properties are studied as a function of frequency, temperature, pressure, plasticiser concentration and degree of polymerisation (*e.g*., Sasabe and Saito, 1968). This technique can be applied to polar materials and seems to be ideal for studies of the linear and branched polyurethane (PUR) systems (Gowri Krishna *et al*., 1982; Frubing *et al*., 2002).

PUR have been intensively studied for many years and have wide applications (Oertel, 1985). Almost all current commercial PUR are obtained from petroleum derivatives. However, the scientific community has lately become very interested in developing materials that originate from renewable resources such as vegetable oils. The vegetable oils usually used to manufacture PUR are castor, soya, sunflower, linseed, canola and safflower oils (Oertel, 1985; Robertson *et al*., 1988; Bhunia *et al*., 1999; Andrew *et al*., 2002; Sharif *et al*., 2002; Dutta *et al*., 2004; Hamid and Mohammad, 2004). The novel PUR reported in this article was derived from palm

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oil-based oleic acid polyols. In a previous article, we have discussed the synthesis as well as the mechanical and physico-mechanical properties of the novel polyurethane (Velayutham *et al.*, 2009a, b). This article will concentrate on the complementary dielectric properties of the material. Oleic acid content in the polyols and NCO/OH ratio of PUR are the two variables described here. The main aim of this article was to reveal the dielectric response of PUR by varying these variables and to study the variables contribution to the molecular motion of the polymer. Thus, the findings will enable us to have an in-depth knowledge of the molecular structure, polarisation and dipole motions of the novel PUR.

MATERIALS AND METHODS

Materials

Oleic acid (99.5% purity) and glycerol (99.5% purity) were obtained from Cognis Oleochemical (M) Sdn Bhd (Malaysia). Phthalic anhydride, PA (P T Petrowidada, Indonesia) and toluene diisocyanate (TDI) from Aldrich, USA (80:20; 2,4-toluene diisocyanate: 2,6-toluene diisocyanate), a mixture of the two isomers, were used as received. Toluene (JT Baker, USA) was used as a solvent; it was dried over an activated molecular sieve overnight before use. The silicone surfactant used was L6900 (Air Products, USA) and the defoamer BYK-088 was from BYK Chemie, Germany.

Synthesis of Polyester Polyol and Polyurethane

The synthesis of polyols from oleic acid and PUR has been described in detail in literature (Velayutham *et al.*, 2009a, b). The formulations of polyols were based on the method described by Patton (1962). Pre-weighed PA, glycerol and oleic acid were charged into a 2-litre four-neck round-bottomed reactor flask, stirred and heated to a temperature of 120°C-130°C, while a slow stream of nitrogen gas was bubbled through the mixture for 30 min. The temperature was then increased to 180°C-200°C so that the reaction could proceed readily with the evolution of water, which was collected at the decanter arm. The reaction was considered complete after the amount of water of reaction collected was as predicted in the formulation. Three different formulations of polyols were synthesised by varying the amount of oleic acid to 28%, 40% and 65%. The samples were named Alk28, Alk40 and Alk65, respectively, in accordance to the percentage of oleic acid in each sample. The composition and characteristics of the polyols have been summarised in *Tables 1* and *2* in Velayutham *et al.* (2009a).

As for the synthesis of PUR, the polyol and additives were dried under reduced pressure at 80°C for 2 hr before being poured into a three-neck flat-bottomed flask equipped with a thermometer, dropping funnel and magnetic stirrer. The polyol was allowed to react with a calculated amount of TDI in the presence of the solvent toluene, surfactant and defoamer. TDI was added dropwise into the reaction mixture at 80°C over 3 hr with constant stirring. The partially reacted sample formed a viscous solution, which was then poured into a 12 cm × 12 cm mould to cure under ambient temperature overnight to form a film of 0.5-1.0 mm thickness. This film was then left in the oven at 60°C for two days to drive off all the solvent. The NCO/OH ratios of the samples were varied to 1.2, 1.4 and 1.6. For the measurement of dielectric properties, circular discs of diameter 38 mm and thickness of about 0.5-1.0 mm were used.

Measurement of Dielectric Properties

Dielectric measurements were carried out according to ASTM D150 on PUR in order to determine their real (ε') and imaginary (ε'') permittivity as a function of frequency. These measurements were conducted over the frequency range 100 Hz to 40 MHz, using a Hewlett Packard 4194A impedance/gain-phase analyser equipped with a HP 16451B dielectric material test fixture. A micrometer electrode was used and this electrode system eliminated any error caused by series inductance, resistance of the connecting leads and of the measuring capacitor at high frequencies.

RESULTS AND DISCUSSION

Impedance Analysis

The structure of the main chain of PUR consists of a urethane backbone, with oleic acid as a flexible side chain. As the oleic acid content increased, so did the percentage of flexible side chain. The NCO/OH ratio of PUR varied from 1.2 to 1.6. Hence, more three-dimensional allophanate and isocyanurates crosslinks (Hepburn, 1982) would have been formed in the system for the highest ratio. The increase in chemical crosslinking leads to a change in morphology and an increase in intermolecular attraction between hard segments (Spathis *et al.*, 1994).

The variations in real (ε') and imaginary (ε'') permittivity as a function of frequency for all the samples (except PU with Alk65; NCO/OH ratio 1.2) at room temperature are shown in *Figure 1*. The reason for exception of the sample was due to adversity in preparing it; the samples were too soft to form films.

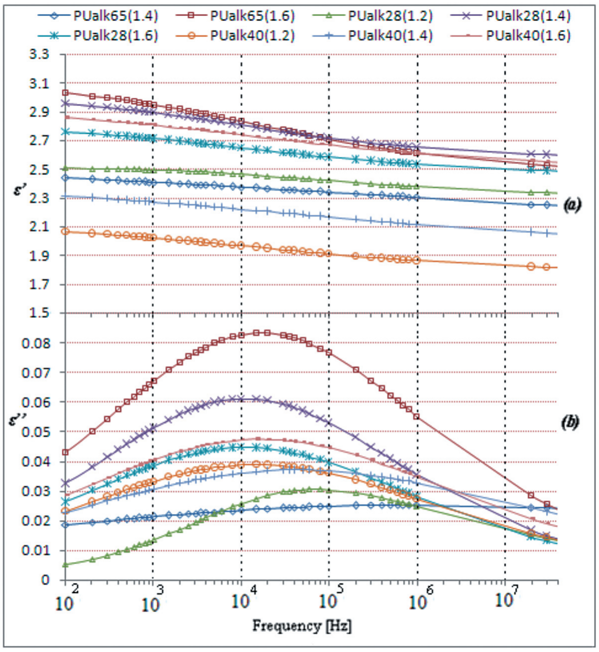

Figure 1. Variation of (a) real permittivity (ε′) and (b) imaginary permittivity (ε″) for PUalk28, PUalk40 and PUalk65 with frequency measured at room temperature and NCO/OH ratios of 1.2, 1.4 and 1.6.

From *Figure 1a*, it can be seen that the value of ε' increased as the NCO/OH ratio increased at any given oleic acid percentage in the polyols of PUR. A corresponding change in the behaviour of ε'' could be observed where distinct loss peaks at the frequency range of 10 to 20 kHz were obtained for all the samples at room temperature. A γ-relaxation was usually observed at a frequency of 10 kHz, and was proposed as the probable mechanism for the dielectric behaviour of PUR in this research. A γ-relaxation mechanism is attributed to the local motion of the chain segments located between the hydrogen bonds (ILLERS, 1960; Hedvig, 1977). These chain segments consisted mainly of polar $-CH_2$ and -C-O groups where the transition could be observed dielectrically. The existence of CH_2 and C-O groups in PUR can be confirmed from the FTIR spectrum as shown by Hamid and Mohammad (2004). In the spectra, the characteristic carbonyl stretching was observed at 1734-1745 cm^{-1}, and the band observed at 2930-2940 cm^{-1} was due to -CH-stretching. The main factor in determining the transition is the hydrogen-bond chain structure and not the structure of the main polymer chain (Hedvig, 1977; Pissis *et al.*, 2002).

Figure 2 shows the effect of oleic acid content of the polyols on the dielectric permittivity of PUR. For a NCO/OH ratio of 1.6, PUalk65 had the highest real permittivity ε' and imaginary permittivity ε'' spectra, followed by PUalk40 and PUalk28. It is presumed that the protons originating from the oleic acid content of the polyols dominated the dielectric relaxation mechanism of PURs.

The experimental dielectric spectrum was fitted by the Havriliak-Negami (HN) expression as shown below:

$$\varepsilon'(\omega) = \varepsilon_\infty + \frac{(\varepsilon_s - \varepsilon_\infty)\cos(\beta\varphi)}{[1 + 2(\omega\tau)^{1-\alpha}\sin(\frac{\alpha\pi}{2}) + (\omega\tau)^{2(1-\alpha)}]^{\frac{\beta}{2}}} \quad (1a)$$

$$\varepsilon''(\omega) = (\varepsilon_s - \varepsilon_\infty) + \frac{\sin(\beta\varphi)}{[1 + 2(\omega\tau)^{1-\alpha}\sin(\frac{\alpha\pi}{2}) + (\omega\tau)^{2(1-\alpha)}]^{\frac{\beta}{2}}} \quad (1b)$$

where

$$\varphi = arctg\left(\frac{(\omega\tau)^{1-\alpha}\cos\frac{\alpha\pi}{2}}{1 + (\omega\tau)^{1-\alpha} + \sin(\frac{\alpha\pi}{2})}\right)$$

The ε_∞ and ε_s are high frequency limits of the permittivity and the vacuum permittivity, respectively. The curve fitting was performed using the software OriginPro 8.1.

HN relaxation is an empirical modification of the Debye relaxation model, accounting for the asymmetry and broadness of the dielectric dispersion curve. The model was first used to

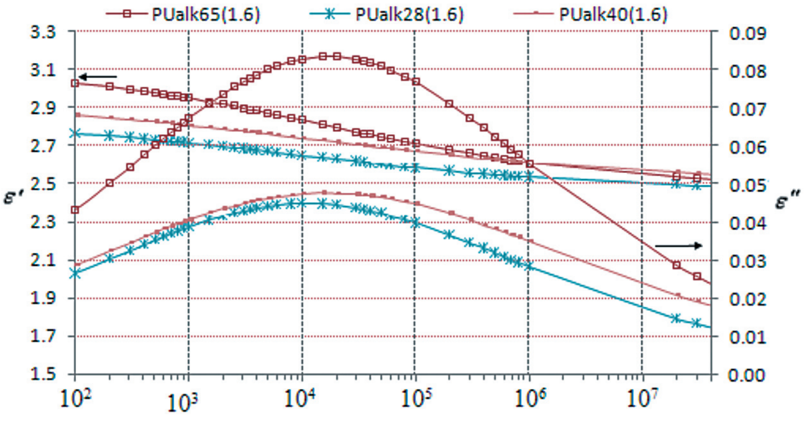

Figure 2. Variation of real permittivity (ε′) and imaginary permittivity (ε″) for PUalk28, PUalk40 and PUalk65 with frequency measured at room temperature and NCO/OH ratio of 1.6.

describe the dielectric relaxation of some polymers (Havriliak and Negami, 1967). The principal use of this function has been for polymers, and it can fit results for many amorphous polymers over a considerable range of frequencies (Cole, 1980).

This strategy of analysis has been used for many PUR systems (Pissis *et al.*, 1996; 2002; Georgoussis *et al.*, 2000; Korzhenko *et al.*, 2000). The exponents α and β are the shape parameters that describe the asymmetry and broadness of the corresponding shape of the plots, while τ is the HN relaxation time. The HN fit for the relaxation phenomenon measured at room temperature is illustrated in *Figure 3*.

Parameters such as ε_∞, ε_s, $\Delta\varepsilon = (\varepsilon_s - \varepsilon_\infty)$ and τ were obtained from the experimental HN plots. The shape parameters such as α and β were derived from the HN relaxation model. These parameters are tabulated in *Table 1*. *Figure 3* shows that, except for PUalk28 (at NCO/OH ratios of 1.4 and 1.6), the HN plot of PUR shifted to the right of the graph as the NCO/OH ratio increased. The dielectric strength ($\Delta\varepsilon \pm 0.02$) increased as the NCO/OH ratio increased for both PUalk40 and PUalk65. However, for PUalk28, the dielectric strength increased when the ratio increased from 1.2 to 1.4, and then decreased as the ratio was increased further to 1.6. The relaxation time could be related to the flexibility of the particular PUR sample. In the case of the orientation of molecular segments or segmental groups, the presence of crosslinks should increase the relaxation time overall because mobility is reduced (Gowri Krishna *et al.*, 1982). This shows clearly that the dielectric loss observed was due to the orientational polarisation.

The internal degrees of freedom of the molecules which led to smaller inter-molecular interactions can be related to the parameter α. Increased α values indicate a decrease in the internal degrees of freedom or less molecular interaction in the dielectric. From *Table 1*, the α value for PUalk28 increased as the NCO/OH ratio increased. Theoretically, as the NCO/OH ratio increases, the crosslinking of the materials increases as well, and,

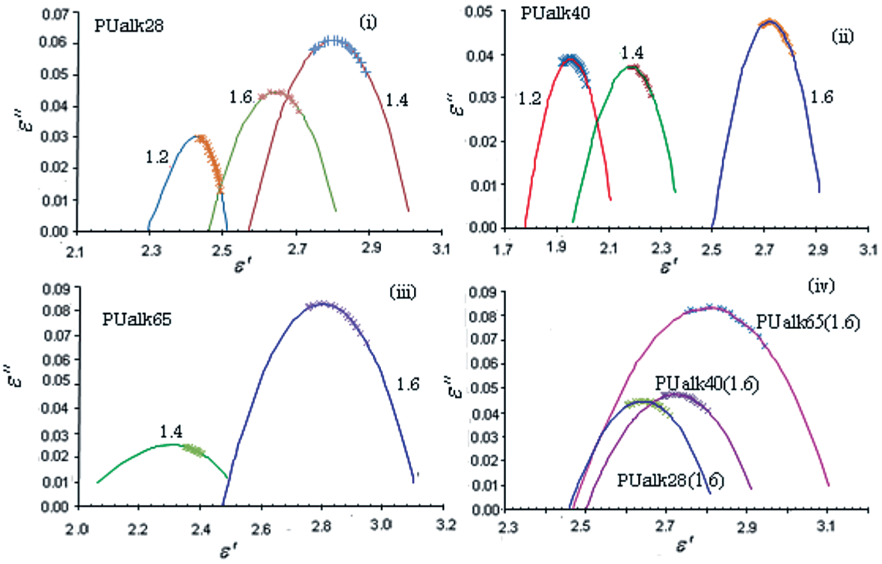

Figure 3. Assymetrical Havriliak-Negami plots of the dispersion curves for (i) PUalk28, (ii) PUalk40, (iii) PUalk65 and (iv) PUR with NCO/OH ratio of 1.6 measured at room temperature. Dots are experimental points and continuous lines correspond to simulated fit.

TABLE 1. STATIC PERMITTIVITY ε_s, INDUCED PERMITTIVITY ε_∞, DIELECTRIC STRENGTH $\Delta\varepsilon$, α AND β PARAMETERS AT ROOM TEMPERATURE DERIVED FROM HN PLOTS

Parameters	PUalk28			PUalk40			PUalk65	
	1.2	1.4	1.6	1.2	1.4	1.6	1.4	1.6
$\alpha \pm 0.01$	0.50	0.65	0.68	0.70	0.75	0.71	0.86	0.67
$\beta \pm 0.001$	0.390	0.890	0.900	0.860	0.710	0.860	0.550	0.901
$\varepsilon_\infty \pm 0.01$	2.29	2.57	2.46	1.78	1.96	2.50	2.00	2.47
$\varepsilon_s \pm 0.01$	2.51	3.02	2.82	2.12	2.38	2.93	2.55	3.12
$\Delta\varepsilon \pm 0.02$	0.22	0.45	0.36	0.34	0.42	0.43	0.55	0.65
τ (s) $\times 10^{-5}$ ($\pm 0.01 \times 10^{-5}$)	1.30	1.98	1.98	1.77	1.45	1.45	1.77	1.27

hence, it can be assumed that the internal degrees of freedom of the molecules decrease. Therefore the α parameter should increase as the ratio of the formulation increases, which was certainly true for the case of PUalk28. However, PUalk40 and PUalk65 contradicted this stated assumption due to the higher oleic acid content in the polyol which presumably produced a higher amount of flexible side chains, and thus made the polymer less compact.

The frequency dependence of the total AC conductivity is depicted in *Figure 4*. The conductivity was found to be frequency-independent in the low frequency region, and DC conductivity, $\sigma(0)$, had been extracted from the region. Each curve shows the frequency behaviour described through Jonscher's universal power feature (Jonscher, 1977), which is

$$\sigma(\omega) = \sigma(0) + A\omega^n \qquad (2)$$

where $\sigma(0)$ is the DC conductivity of the material, A is the dispersion parameter, and $0 < n < 1$ is the dimensionless frequency exponent. Equation 2 has been used to fit the AC conductivity data where A and n values were varied simultaneously to get the best fit.

The fitted curves are shown in *Figure 4*. It was found that the fit was (visually) satisfactory in all cases. Extrapolation of the fitted results to zero frequency gave DC conductivity, $\sigma_{dc,}$ the values of which are tabulated in *Table 2*.

The DC conductivity values were measured experimentally by using a Keithley Source Measurement Unit (SMU 236) at room temperature. The validity of the $\sigma_{dc,}$ values which were obtained experimentally was high as the values were in good agreement with the fitted σ_{dc}. The switchover from the frequency-independent region to frequency-dependent region is a signal of the onset of conductivity relaxation (Anantha and Hariharan, 2005). The plot exhibits the typical behaviour of ionic materials, *i.e.* having a DC plateau and a frequency-dependent region (Murugaraj *et al.*, 2003). The observed behaviour is in general agreement with the prediction of the jump relaxation model (Funke, 1997; Kulkarni *et al.*, 1998; Kanapitsas *et al.*, 1999; Dyre and Schrøder, 2002). According to this model, at low frequencies an ion jumps successfully from one site to its neighbouring vacant site, thus contributing to DC conductivity.

TABLE 2. EXPERIMENTAL AND FITTED RESULTS OF DC CONDUCTIVITY AT ROOM TEMPERATURE DERIVED FROM POWER LAW PLOTS

	NCO/OH ratio	**Experimental $\sigma_{dc} \times 10^{-11}$ (± 0.1 Sm^{-1})**	**Fitted $\sigma_{dc} \times 10^{-11}$ (± 0.02 Sm^{-1})**
Pualk28	1.2	31.6	33.90
	1.4	36.6	35.30
	1.6	37.8	36.80
PUalk40	1.2	3.6	3.31
	1.4	3.3	2.94
	1.6	3.9	3.32
PUalk65	1.4	4.0	3.85
	1.6	7.4	7.15

CONCLUSION

The ac conductivity and dielectric behaviour of PUR were studied as a function of frequency at room temperature. The dielectric parameters were in the range of 2.0 to 3.0 for real permittivity ε' and 0.02 to 0.08 for imaginary permittivity, ε''. In general, most PUR had ε' values in the range

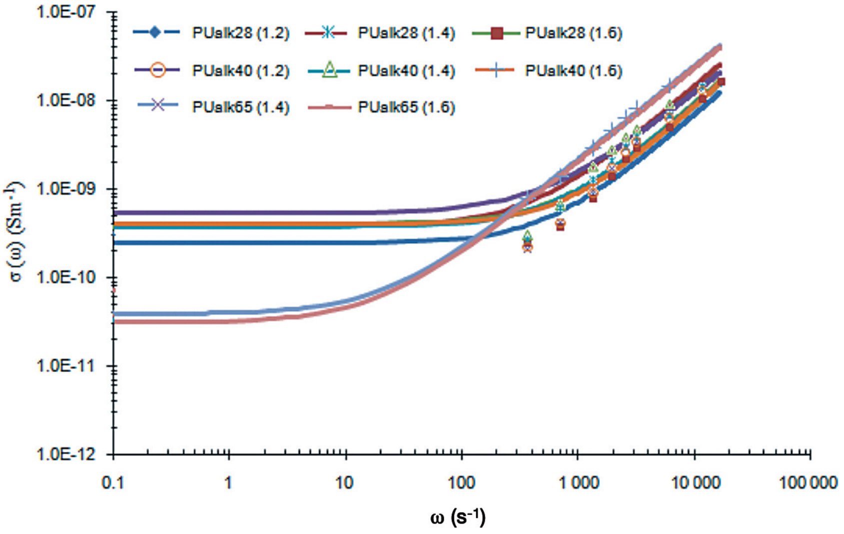

Figure 4. Frequency-dependent conductivity, σ *(ac)* vs. *frequency for PUR at room temperature. Dots are experimental points and continuous lines correspond to simulated fit.*

of 1.5 to 7.0 and ε'' in the range of 0.001 to 1.0 (Kanapitsas *et al.*, 1999; Korzhenko *et al.*, 1999; 2000; Chwang *et al.*, 2004) which indicates that the dielectric values obtained for the oleic acid-based PUR were comparable to those mentioned above. Cole-Cole plots were fitted successfully using the Havriliak-Negami model. The γ-relaxation was proposed as the probable mechanism for the dielectric behaviour of PUR. The dielectric constant and loss mechanism of the material were found to be dependent on the frequency variation, NCO/OH ratio and polyol used in synthesising PUR. The frequency-dependent conductivity of PUR materials was analysed using a Jonscher's power law expression to compare the validity of the $\sigma_{dc,}$ values which were obtained experimentally. The frequency-dependent permittivity showed dielectric dispersion, and the variation of the dielectric constant with frequency was ascribed to the contribution of the charge carriers.

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