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Polydimethyl Siloxane Toughened Epoxy Resins: Tensile Strength and Dynamic Mechanical Analysis

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ABSTRACT: Mechanical properties of the Interpenetrating Polymer Networks (IPN's) based on epoxy resin and polydimethy siloxane (PDMS) were investigated. The main objective of the present work was to use PDMS as impact modifier for epoxy resin. New IPN's between epoxy resin and different weight percentage of PDMS were prepared in the presence of tetraethoxysilane (TEOS) as crosslinking for PDMS and triethylene tetra amine (TETA) as hardener for epoxy resin. The tensile properties of the cured materials were studied, and from the result shown that the ultimate tensile strength (UTS) was decreased with increasing percentage of PDMS in the prepared IPN's, while the elongation was increased. On the other hand, the storage modulus E', loss modulus E'', and complex modulus E* of epoxy resin were decreased with increasing weight percentage of PDMS in the network structure.

Keywords: polydimethy siloxane, dynamic mechanical analysis, tensile strength, diqlycidyl ether, epoxy resin.

1.0 INTRODUCTION

Epoxy resins are among the best matrix materials for many fiber composites, and are widely used polymers for its many good properties such as coatings, adhesives, binders for abrasives, automotive and electrical components [1]. However it is rather brittle material [2-4]. Epoxies are generally synthesized by one of the two methods: from phenols and epichlorohydrin, or from alkenes and peroxy acids [5]. Epoxy resins can be cured via additions or homo polymerization. During the cure of epoxy resins, there is initially linear growth with a gain in molecular weight. Large number of new epoxy resins have been prepared and studied in the last two decades [6]. On the other hand silicone polymers are of great important [7,8] due to their resistance to high temperature, high flexibility, their good dielectric properties over wide frequency and temperature ranges, their chemical inertness and water repellency.

The toughening of thermoset resins like epoxies has received much attention in the area of fiber reinforced plastic (FRP) composites [9,10], and since the toughened resins improve the fracture toughness impact strength and delaminating resistance of the composites. There are several papers involves modification of epoxy resin with reactive liquid rubber [11,12], like use the carboxy-terminated poly butadiene co-acrylonitrile (CTBN) dispersed in the epoxy resin [13]. So the present work however is an attempt to use the silicone elastomer as impact modifier for epoxy resin in order to decrease the brittleness of epoxy resin.

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2.0 METHODS & MATERIALS

2.1 Materials and Sample Preparation

The epoxy resin diglycidyl ether of bisphenol-A-ARALDITE 2011A from CIBA with a number average molecular weight <700 was dried at 90C° under vacuum for two hours before use throughout this work.

PDMS elastomer was prepared by reaction of hydroxy terminated polydimethyl siloxane with (TEOS) as crosslinking agent in the presence of tin octaot as co-catalyst [6]. Araldite 2011B, triethylenetetramine from C1B17 was used as hardener for epoxy resin. FTIR spectra were recorded on Spectrum-2000 by JASCO.

Mechanical properties were studied by use universal testing machine INSTRON-5582 (100KN). Dynamic mechanical analysis measurement was made on dynamic mechanical Analyzer DMA 6100 by Seiko Instrument.

2.2 Synthesis of New IPN's

The IPN's considered in this study based on PDMS with epoxy resin were prepared by efficient mixing different quantities shown in Table 1 for a period of 2 min. at room temperature, then tin octaote and TETA were added and mixed gently. The sample were left overnight for curing at room temperature, then at 100C° for 4hr, and at 120C° for 24hrs for post cure.

Table 1: The quantities of the reactant used in the preparation of the new IPN's based on Epoxy resin-PDMS

First component				Second component	
PDMS %	PDMS Mg	TEOS mg	SnOC mg	Epoxy resin mg	TETA mg
0	0	0	0	418	82
5	20.99	3.74	0.25	395	80
10	42.02	7.48	0.5	375	75
15	63.03	11.22	0.75	357	68
20	84.02	14.96	1	335	65

3.0 RESULTS & DISCUSSION

The route for synthesis of tetraethoxy silane, PDMS terminated hydroxyl group are shown in Figure 1, while the network structure of polysiloxane and epoxy resin are shown in Figure 2.

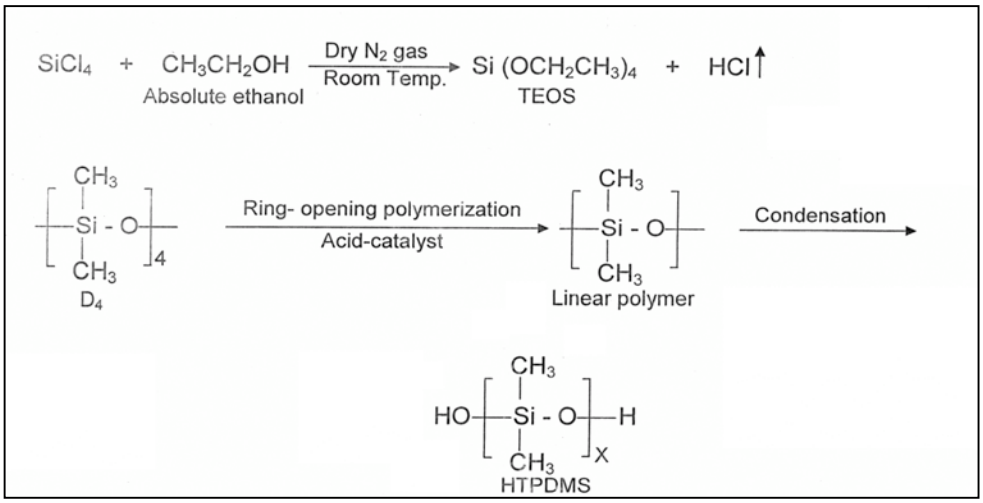

Figure 1: Synthesis of tertraethoxy silane and HTPDMS

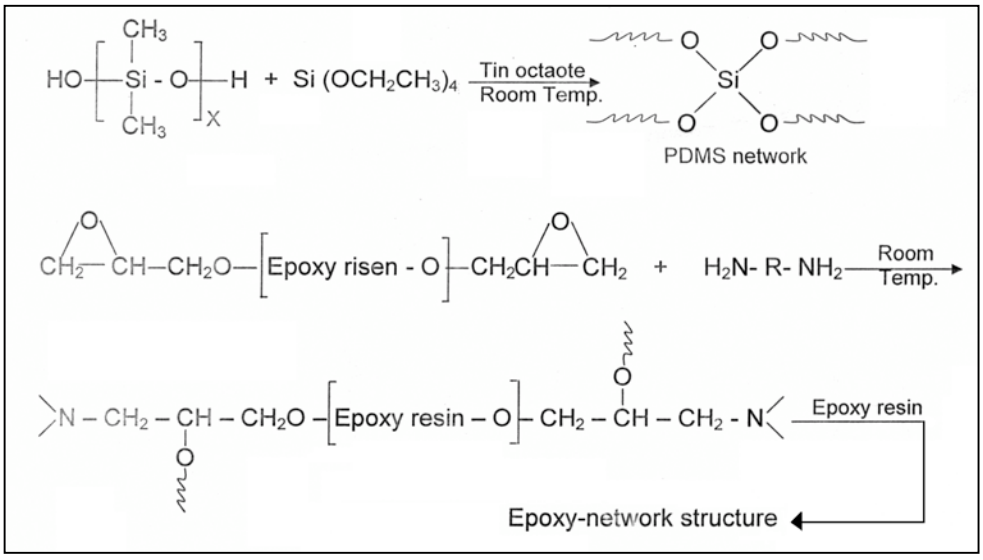

Figure 2: IPN's of PDMS-Epoxy Resin

3.1 Characterization

The PDMS terminated hydroxyl group was characterized by FTIR and H^1-NMR as shown in Figure 3 and Figure 4 respectively. From Figure 3, the presence of hydroxyl group in the silicone polymer are in the region 3400-3600 cm^{-1} as stretching vibration frequency. Further confirmation for terminal Si-OH group is evidence from peak around 1600 cm^{-1} due to the bending motion of Si-OH. There is additional absorption bands in the region 1260 cm^{-1} and 800 cm^{-1} related to the stretching vibration of $Si(CH_3)_2$. The characteristic bands are shown in Table 2.

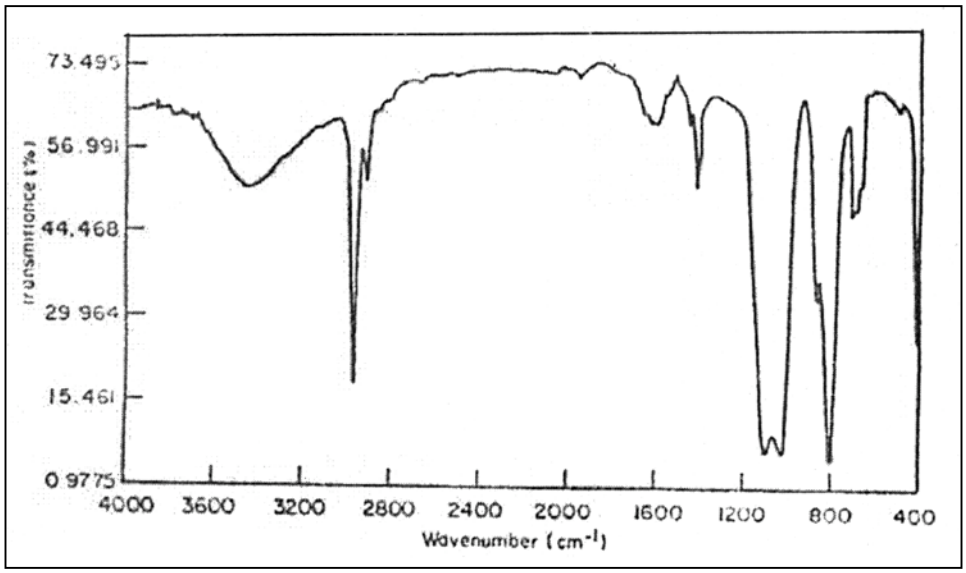

Figure 3: FTIR spectrum of linear polydimethyl siloxane

Table 2: IR bands for PDMS

Si-O	Stretching vibration	1100-1000 cm^{-1}
Si-CH_3	Stretching vibration of Si-CH_3	800, 1250 cm^{-1}
Si-O-Si	Bending motion	475 cm^{-1}
Si-O	Stretching vibration of Si-OH	875 cm^{-1}
C-H	Stretching vibration	2950 cm^{-1}

While the H^1-NMR spectrum in Figure 4 showed the presence of Si-OH is acidic in nature and more deshielded due to differences in screening by the electron from Si-O and C-O group, accounting for the chemical shift difference of 1 ppm (C-OH is at 4.5ppm). The peak corresponding to Si-C1B is seen at 1.8 ppm. In the case of IPN's, the resulting products were transparent at these percentage of silicone polymer (5, 10, 15, and 20%) indicating that no phase separation.

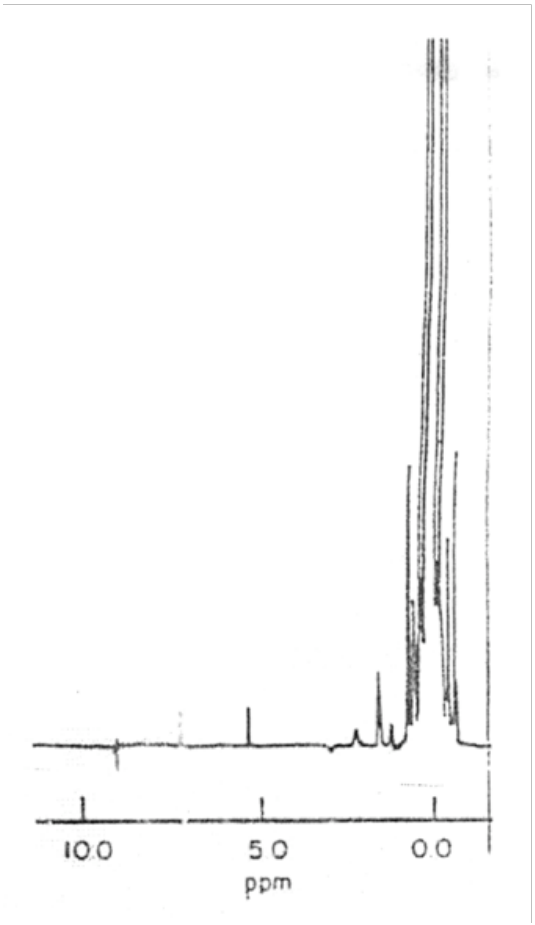

Figure 4: ^{1}HNMR spectrum of polydimethyl siloxane

3.2 Mechanical Properties

The mechanical properties i.e. ultimate compression strength (UCS), ultimate tensile strength (UTS) and percent strain for the material prepared samples with the difference weight percentage of PDMS listed in Table 3 were investigated. The toughness of the epoxy resin increased with the addition of PDMS elastomers.

The UTS of these material (IPN's) decreases with the addition of PDMS but percent strain or percentage of elongation increased as much PDMS incorporated. From the UCS, UTS and percent strain results the optimum toughness was observed and attained the addition of 5% PDM in epoxy resin.

Table 3: Mechanical Properties of the prepared IPN's

No.	Sample ID	PDMS %	Result		
			Ultimate compression strength UCS (Mpa)	Ultimate Tensile Strength	
				UTS (Mpa)	Elongation %
1	0% PDMS	0	124.7	26.86	21.83
2	5%	5	88.3	25.23	25.26
3	10%	10	115.4	19.5	27.84
4	15%	15	125.6	19.58	29.93
5	20%	20	105.4	15.57	38.8

3.3 Dynamic Mechanical Analysis

Epoxy resin ARALDITE 2011A modified by incorporation of PDMS changed the mechanical properties significantly. All the module i.e. storage modulus E′ (Table 4), loss modulus E″ (Table 5) and complex modulus E^* (Table 6) dropped with the increasing ratio of PDMS in the epoxy resin formulation.

A gradual drop of storage modulus E′ with addition of PDMS revaluated that the flexibility of the sample increased with the addition of PDMS epoxy formulation containing 20% PDMS showed maximum flexibility.

Table 4: Storage modulus E′ (GPa) data of epoxy formulation at $1H_Z$ frequency.

Temp. C°	0% PDMS	5% PDMS	10% PDMS	15% PDMS	20% PDMS
28	8.89	7.55	6.76	5.92	2.89
40	4.79	3.84	3.56	3.18	1.18
60	1.01	0.81	0.49	0.50	0.23
80	0.39	0.34	0.23	0.15	0.12
100	0.23	0.21	0.19	0.12	0.07
120	0.16	0.15	0.12	0.08	0.05
140	0.15	0.15	0.12	0.08	0.05
160	0.15	0.14	0.12	0.09	0.05

Temp. Accuracy ±1C°

Table 5: Loss Modulus E″ (GPa) data of epoxy formulation at $1H_Z$ frequency.

Temp. C°	0% PDMS	5% PDMS	10% PDMS	15% PDMS	20% PDMS
28	1.757	1.671	1.614	1.262	1.023
40	1.282	1.191	0.956	0.888	0.499
60	0.341	0.291	0.189	0.205	0.083
80	0.083	0.067	0.037	0.039	0.020
100	0.031	0.027	0.020	0.016	0.012
120	0.006	0.006	0.005	0.004	0.003
140	0.003	0.003	0.003	0.003	0.002
160	0.003	0.003	0.003	0.002	0.002

Table 6: Complex Modulus E^* (GPa) data of epoxy formulation at $1H_Z$ frequency.

Temp. C°	0% PDMS	5% PDMS	10% PDMS	15% PDMS	20% PDMS
28	9.065	7.729	6.95	6.056	3.073
40	4.963	4.025	3.75	3.304	1.280
60	1.067	0.859	0.53	0.543	0.249
80	0.398	0.351	0.24	0.163	0.126
100	0.242	0.225	0.19	0.118	0.086
120	0.156	0.150	0.12	0.080	0.054
140	0.151	0.146	0.12	0.083	0.053
160	0.148	0.145	0.12	0.086	0.050

The stiffness if the resultant material was also temperature sensitive. The storage modulus in Figure 5 decrease with increasing temperature indicating that all epoxy formulations gradually change from stiff, hard solid to soft and flexible material.

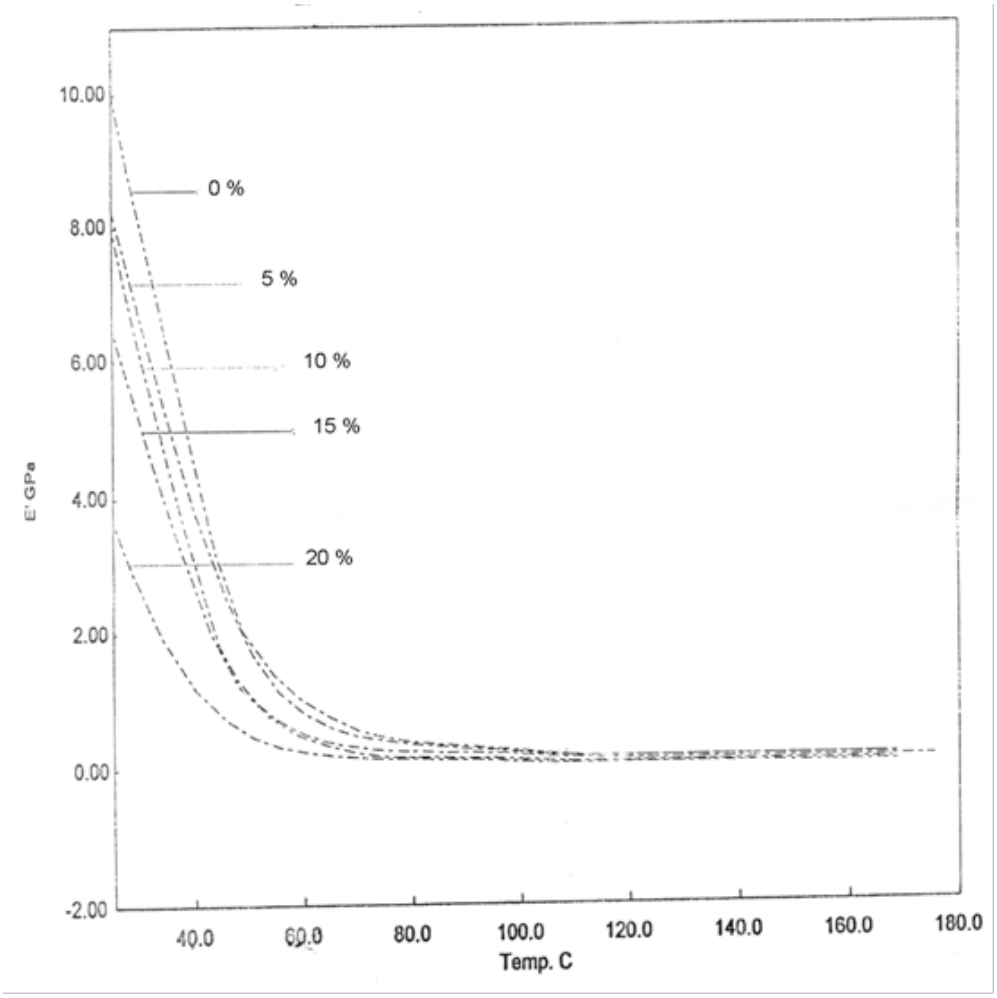

Figure 5: Storage modulus E′ measured at $1H_Z$

The damping characteristics also change with the successive percentage of PDMS in the epoxy matrix as shown in loss modulus E″ Figure 6. All the DMA measurements were sensitive to frequency of oscillation the E′, E″, and E* tan D Figure 7 graphs were also shifted.

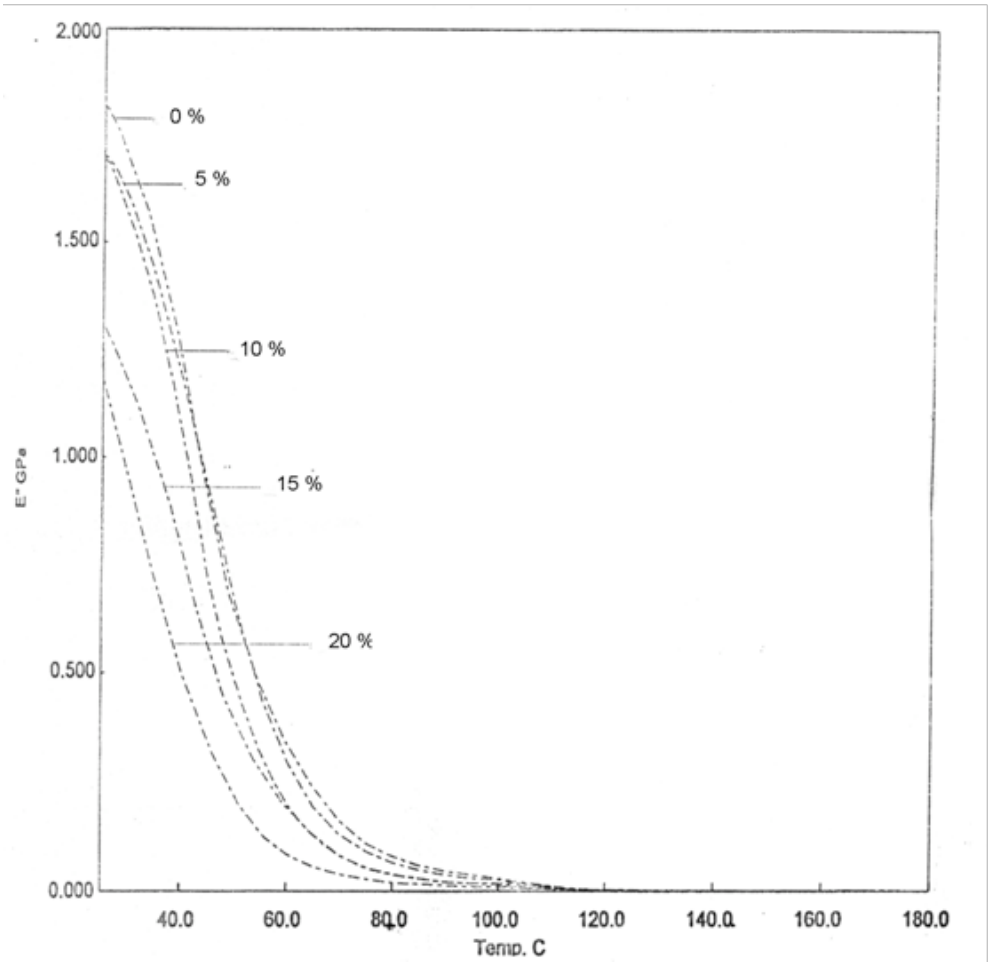

Figure 6: Loss modulus E″ measured at $1H_Z$

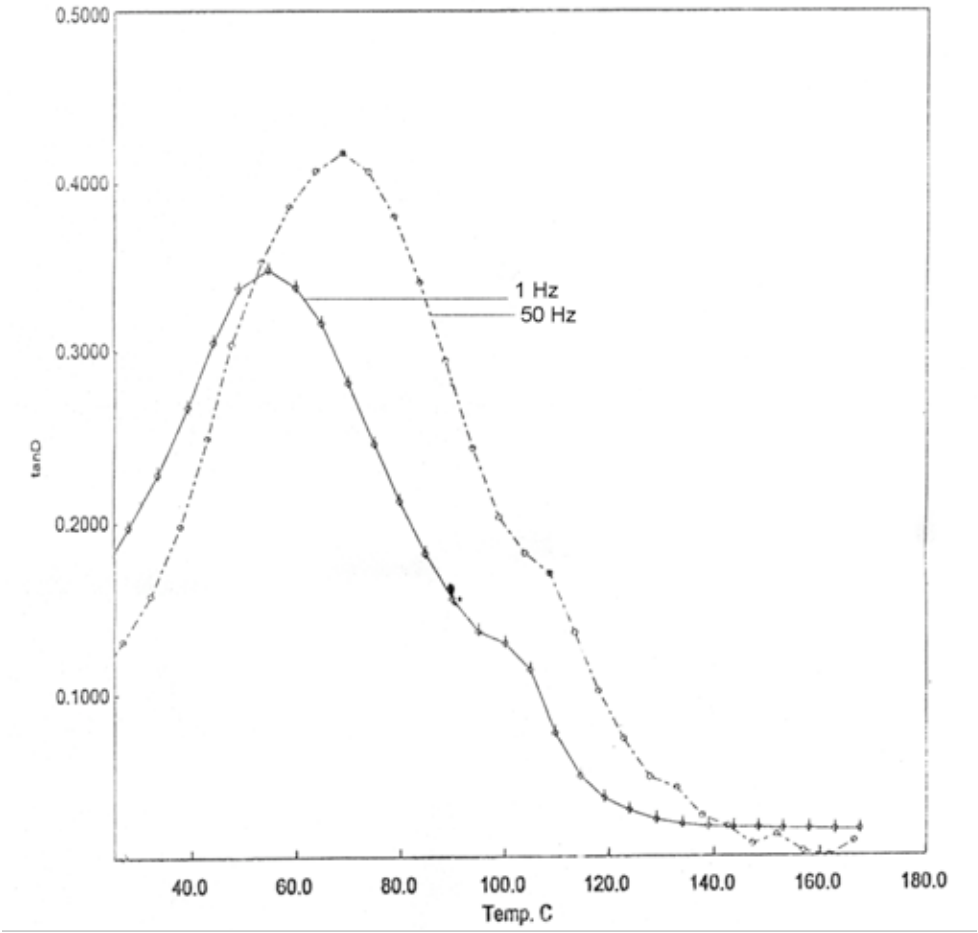

Figure 7: Glass transition tan D of neat epoxy hardener system measured at $1H_Z$ & 50 H_Z

4.0 CONCLUSIONS

PDMS acted here as a potential toughening for epoxy resin. The toughness of the epoxy resin was increased by the addition of PDMS. From UCS, UTS and percent strain results the optimum toughness observed was obtained with the addition of 5% PDMS. Dynamic Mechanical Analysis results showed an increase in flexibility by the addition of PDMS. Stiffness of the resultant material was also temperature sensitive. All the DMA measurements were sensitive to the frequency of oscillation.

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