Dielectric Properties of Epoxy Resins at Different Extents of Cure at 2.45 GHz

Liming Zong[1], Rensheng Sun[2], Leo C. Kempel[2], and Martin C. Hawley[1],*
[1]Department of Chemical Engineering and Materials Science, Michigan State University, East Lansing, Michigan 48824
[2]Department of Electrical and Computer Engineering, Michigan State University, East Lansing, Michigan 48824

Microwave processing of polymers has been studied as an attractive alternative to conventional thermal processing [1]. Information regarding dielectric properties of materials at microwave frequencies is important for control of electromagnetic absorption during microwave processing. The Debye model [2] works well for systems containing small molecules in gas media or media with low viscosity. Several empirical models, e.g. Havriliak and Negami (H-N) model [3] and Schonhals and Schlosser (S-S) model [4], were proposed to describe the dielectric properties of a complicated system. The study of dielectric properties of epoxy resins is an interesting topic [1, 5]. We reported the study on the dielectric properties of diglycidyl ether of bisphenol-A (DGEBA)/DDS epoxy resins at 2.45 GHz [6]. This paper focuses on the dielectric properties of DGEBA/ *m*-phenylenediamine (mPDA) epoxy resins.

EXPERIMENTAL

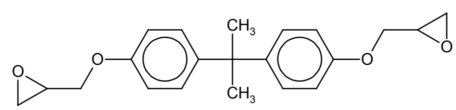

Figure 1. Chemical Structure of DGEBA

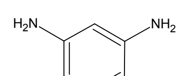

Figure 2. Chemical Structure of mPDA

The epoxy resin is DGEBA (DER 332 by Dow Chemical) with an epoxy equivalent weight of about 173. The molecular structure of DGEBA is shown in Figure 1. The curing agent is mPDA from Aldrich Chemical with amine hydrogen equivalent weight (AHEW) of 27. The molecular structure of mPDA is shown in Figure 2. All of the materials were used as received without further purification. In preparing the neat epoxy resin, stoichiometric DGEBA/mPDA were mixed. The mixture was stirred for 3 minutes with a magnetic bar at 70°C and degassed at 0.02 bar and room temperature for 5 minutes. Fresh samples were kept in a -20°C freezer and used within three days. Details of experimental procedure can be found in [6, 7]. When uncured DGEBA/mPDA samples were measured, fresh samples were first heated to 80^{o}C; when cured DGEBA/mPDA samples were measured, fresh samples were first heated to 110^{o}C for a designated time. Then the single frequency microwave curing system was switched to a low-power swept frequency diagnostic system. Measurements of temperature and dielectric properties using the swept frequency method were made during free convective cooling. The uncured and cured samples were analyzed with Differential Scanning Calorimetry (DSC) to determine the residual heat of reaction per gram of resin and then the curing extent of a sample can be calculated.

RESULTS AND DISCUSSION

Dielectric properties of DGEBA/mPDA epoxy resins at different extents of cure vs. temperature at 2.45 GHz are presented in Figure 3. The dielectric properties, including dielectric constant (ε') and dielectric loss factor (ε''), increase as temperature increases and decrease as extent of cure increases.

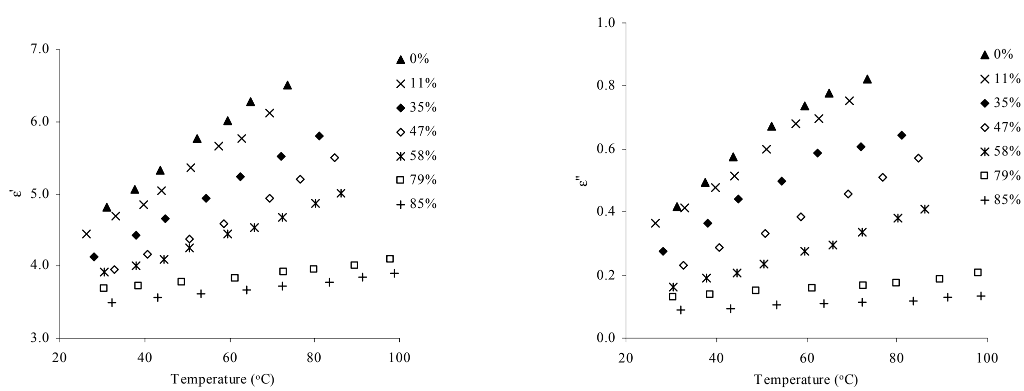

Figure 3. Dielectric properties vs. temperature for DGEBA/mPDA at different extents of cure.

A simple model was proposed to describe the dielectric properties of curing epoxy resins at a high frequency microwave [6]. The model is shown in eq. (1)-(3), where ε^* is the complex dielectric constant; ε' is the dielectric constant, ε″ is the dielectric loss factor; ω is the radial frequency of the electric field in s^{-1}; τ is the relaxation time in s; ε_0 is the low frequency dielectric constant; ε_∞ is the high frequency dielectric constant; and n is the shape parameter.

$$\varepsilon^* - \varepsilon_\infty = \frac{(\varepsilon_0 - \varepsilon_\infty)}{(j\omega\tau)^n} \quad (1)$$

$$\varepsilon' = \varepsilon_\infty + \frac{(\varepsilon_0 - \varepsilon_\infty)\cos(n\frac{\pi}{2})}{(\omega\tau)^n} \quad (2)$$

$$\varepsilon'' = \frac{(\varepsilon_0 - \varepsilon_\infty)\sin(n\frac{\pi}{2})}{(\omega\tau)^n} \quad (3)$$

The relaxations occurred during this experiment are mainly γ relaxation. The γ relaxation time fits the Arrhenius rate law [6]. Plots of ln (ε'-ε_∞) versus 1/T and ln (ε'') versus 1/T should yield straight lines if the proposed simple model is correct and the Arrhenius rate law is applicable to γ relaxation. Figures 4 and 5 show that this is indeed the case.

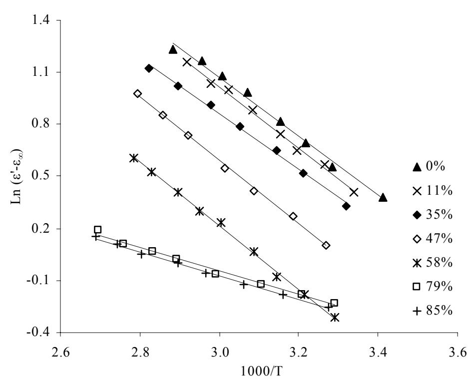

Figure 4. ln (ε'-ε∞) vs. 1000/T.

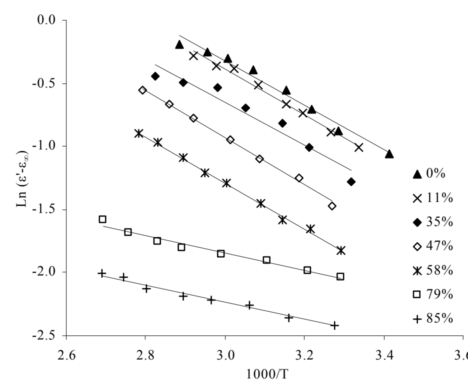

Figure 5. ln (ε″) vs. 1000/T.

The calculated values of parameters of the simple model, except for ε_0 from reference [5], are listed in Table 1.

Table 1. The values of parameters

Extent of Cure	n	(ε_0–ε_∞)	τ (s) 300K	τ (s) 353K
0%	0.153	5.68	1.9E-07	6.5E-10
11%	0.152	5.49	2.5E-07	6.9E-10
35%	0.138	4.95	9.3E-07	2.4E-09
47%	0.137	4.48	4.3E-06	4.9E-09
58%	0.139	3.84	1.5E-05	2.1E-08
79%	0.108	3.45	9.9E-05	3.9E-06
85%	0.073	3.47	9.8E-02	8.6E-04

CONCLUSIONS

The dielectric properties of DGEBA/mPDA epoxy resins at different extents of cure have been studied at the temperature range of 25 to 80^{o}C at 2.45 GHz microwave. The data fit our model very well. γ relaxation process was used to fit the data. The Arrhenius rate law is applicable to describe the γ relaxation.

ACKNOWLEDGEMENTS

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