

# Prediction of Thermoset Viscosity Using a Thermomechanical Analyzer

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## Synopsis

A thermomechanical analyzer (TMA) in the parallel-plate configuration was employed to study the viscosity behavior of an unfilled, FR-4 type epoxy resin by monitoring its thickness change with time under a constant compressive force at various temperatures. With a dual-Arrhenius rheology model, we were able to predict viscosities under nonisothermal conditions of high heating rate from more easily obtained isothermal viscosity data. Due to its simplicity and speed, this technique is well suited for investigating resin viscosity behavior during composite lamination in a manufacturing environment.

## INTRODUCTION

During printed circuit composite construction, epoxy/glass sheets with surface copper patterns are bonded together using B-stage epoxy prepreg (i.e., glass fabric impregnated with partially cured epoxy) under the application of heat and pressure. The flow characteristics (particularly, the viscosity-temperature-time relationship) of a B-stage resin affect many ultimate composite properties, including dielectric strength and thermal expansion coefficient.<sup>1</sup> Due to the time and expense involved in full-scale lamination, it would be desirable to develop a laboratory-scale approach for modelling the resin viscosity behavior under any chosen lamination conditions.

Two such approaches exist for the determination of resin viscosity: one involves the use of a cone-and-plate viscometer<sup>2,3</sup>; the other utilizes a parallel-plate rheometer.<sup>4</sup> While the cone-and-plate approach estimates the viscosity from the cone speed and torque of the spring, the parallel-plate rheometer [Fig. 1(a)] derives viscosity from the thickness change of a specimen placed between a pair of flat parallel plates under a constant force. Similar in operation to the cone-and-plate device is a parallel-disk system.<sup>5</sup> The cone-and-plate viscometer is capable of obtaining both the viscous (real) and elastic (complex) components of the viscosity (i.e., dynamic or complex viscosity) under a wide range of strain rates. In contrast, the parallel-plate rheometer measures absolute viscosities under very low shear rates, in the neighborhood of  $0.1 \text{ s}^{-1}$ .<sup>6</sup> In spite of this, the ease of operation and similarity to a lamination process make parallel-plate rheometry a useful alternative to the more commonly used cone-and-plate approach for monitoring resin viscosity during lamination. Using parallel-plate rheometry, melt viscosities ranging from  $10^2$  to  $10^9$  P have been reported<sup>7,8</sup> for materials as diverse as glass and polymers.

Most previous parallel-plate work has involved use of large samples

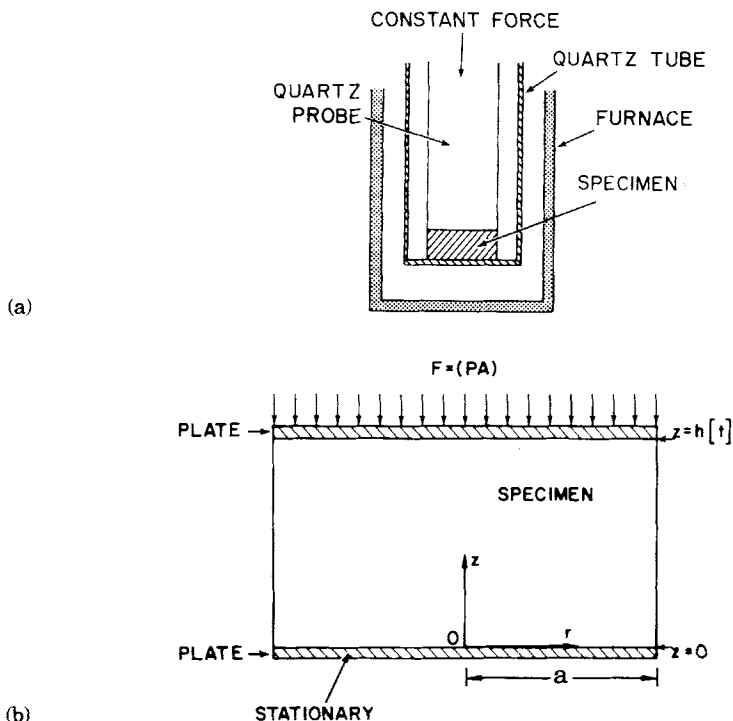


Fig. 1. Schematics of (a) a thermomechanical analyzer and (b) a parallel-plate rheometer.

(> 2 g). When a thermomechanical analyzer (TMA) is employed as a parallel-plate rheometer [Fig. 1(a)], much smaller samples (< 0.1 g) are possible. Small samples allow a quick measurement and better temperature uniformity in the sample (and therefore higher accuracy). Cessna and Jabloner<sup>8</sup> pioneered the application of the TMA technique to characterize the viscosity behavior of powdered polyaromatic thermosets. We have now extended their work to estimate the viscosity history of a B-stage, FR-4 epoxy resin under both isothermal and constant-rate-change thermal histories. Using the dual-Arrhenius rheology model derived for treating viscosity data from a cone-and-plate viscometer,<sup>2</sup> the viscosity changes under conditions of high heating rate (> 10°C/min) can be predicted from isothermal TMA viscosity data. This, in effect, generalizes the Cessna-Jabloner (C-S) approach to cover a wider range of heating schedules. In a subsequent paper, we will derive a model which allows estimation of the resin flow field in prepreg layers during lamination using the TMA thickness-time relationships for both neat resin and prepreg samples.

## THEORY

The foundation of parallel-plate rheometry was laid by Dienes and Klemm.<sup>4</sup> In this technique, the specimen to be tested is placed between two parallel circular plates and a constant force is applied perpendicular to the top plate to deform the specimen as the top plate approaches the stationary bottom plate [Fig. 1(b)]. Under the assumptions that the flow is Newtonian and that the time-dependent inertia terms in the Navier-Stokes (momentum) equation can be neglected (steady-state assumption), expressions re-

lating the sample thickness to viscosity have been given for two modes of experiment: (a) when the sample completely fills the space between the plates with the sample radius equal to the plate radius [Fig. 1(b)] and (b) when the sample never completely fills the space between the plates and the sample volume is constant. Some parallel-plate rheometers which operate predominantly in one of these two modes are available.<sup>9</sup> For mode (a) experiments, the viscosity  $\eta$  of a sample under a force  $F$  satisfies

$$\eta = \frac{-2\pi h^3(P/A)}{3(dh/dt)} \quad (1)$$

where  $h$  is the sample thickness,  $A (= \pi a^2)$  is the overall area of the sample,  $a$  is the plate radius,  $P (= F/A)$  is the applied pressure, and  $t$  is the time. The wall shear rate [i.e., the gradient of  $r$ -direction velocity with respect to the coordinate  $z$  in Fig. 1(b)] at the upper plate equals

$$\frac{-3hR}{2} \frac{d(1/h^2)}{dt}$$

For mode (b) experiments,

$$\eta = \frac{-2\pi h^5 F}{3V^2(dh/dt)} \quad (2)$$

where  $V$  is the sample volume. Rectangular samples in mode (a) experiments can be approximated using an elliptical model for which the radius  $a$  in eq. (1) is replaced by

$$bc\sqrt{2/(b^2 + c^2)}$$

where  $b$  and  $c$  are the radii of the ellipse.<sup>10</sup> When all assumptions are met, eq. (1) implies that sample deformation is determined by the  $P/A$  (pressure to area) ratio and the viscosity  $\eta$  (which, in turn, depends on the temperature schedule). In other words, the thickness history obtained from a laboratory parallel-plate experiment simulates the results of larger-scale resin lamination processes under constant temperature schedule and ( $P/A$ ) ratio. This is similar to the scaled flow concept for the lamination of prepreg materials.<sup>1,11,12</sup> Although eqs. (1) and (2) have been developed for systems with constant viscosity and force ( $F$ ), they are equally applicable to situations where both parameters vary with time. As in the C-S approach, only mode (a) experiments have been performed in the present case.

The sample deformation process is governed by elastic, time-dependent elastic and viscous components of viscosity.<sup>4,9</sup> The measurement of viscosity using the parallel-plate method requires that the viscous component dominate the deformation and that the flow be Newtonian or approximate Newtonian over the relatively small range of shear rates available. Integration of eq. (1) gives

$$\frac{1}{h^2} = \left(\frac{4\pi}{3A}\right) \int_0^t \frac{P}{\eta} dt + \text{const.} \quad (3)$$

where the pressure  $P$  can depend on time. When the viscosity and applied force are constant, the criterion for the validity of the Newtonian assumption is therefore the constancy of the respective slope ( $1/h^2$ ) with time  $t$  according to eq. (3). In other situations, no proper criterion has yet been

established. At very low shear rates (as in the present case), however, many polymer systems exhibit Newtonian behavior.<sup>13</sup> This is assumed in the following.

The modification or omission of the inertia terms in the momentum equation when the Reynolds number is sufficiently small has greatly facilitated the solution of a vast number of fluid dynamics problems.<sup>14</sup> In order to check the steady-state assumption for Newtonian fluids, a numerical method has been developed<sup>15</sup> for analyzing the complete Navier-Stokes equation (including the inertia terms). The result indicates that negligence of fluid inertia is better justified as the Reynolds number  $Re (= F\rho/\eta^2$ , where  $\rho$  is the sample density) is lowered and/or the aspect ratio  $\theta (= R/h)$  increases. The Reynolds number represents the relative contribution between inertia and viscous driving forces and decreases as the viscous part becomes more important. Based on the argument that erratic viscosity data were not obtained when the sample radius  $R$  was at least 10 times the sample thickness  $h$ , Dienes and Klemm<sup>4</sup> were able to establish  $\theta > 10$  for the steady-state assumption to hold with  $Re < 0.45$  and for viscosities ranging from  $10^4$  to  $10^9$  P. Since  $Re$  in the present case is smaller than  $3 \times 10^{-5}$  (corresponding to  $F = 3$  g, the minimum viscosity of  $10^4$  P and  $\rho = 1$  g/cm<sup>3</sup>), it is likely that the steady-state assumption remains valid even when the aspect ratio  $\theta < 10$  (see below).

To cover all thicknesses, Gent<sup>16</sup> has proposed

$$\eta = \frac{-2\pi h^5 F}{3V^2(dh/dt)(1 + 2\pi h^3/V)} \quad (4)$$

for mode (b) experiments where a thickness-dependent term,  $(2\pi h^3/V)$ , has been incorporated in eq. (2). When a specimen was actually compressed, the deformation in eq. (4) was treated as the result of the vertical compressive force, and the horizontal force which kept the specimen in a cylindrical shape during flow. Equation (3) was then obtained as a result of substituting the viscosity for the rigidity modulus and the deformation rate  $(dh/dt)$  for the elastic displacement. With a limited amount of data, Gent found that the viscosity ( $\sim 10^8$  P) of coal-tar pitch obtained from parallel-plate experiments is in accord with that measured by Couette method.<sup>17</sup> Gent's method was later adopted by Fontant<sup>7</sup> for measuring glass viscosities between  $10^5$  and  $10^9$  P up to  $1000^\circ\text{C}$ . Although the counterpart of eq. (4) for mode (a) experiments does not exist, the thickness effects in these two modes of experiments are probably comparable because of the similarity in flow mechanism. Equations (2) and (4) predict viscosities differing by 2% maximum within the thickness range of interest here (6–12 mils). In this study, eq. (1) has been used to estimate resin viscosities.

## EXPERIMENTAL

### Sample Preparation

The FR-4 epoxy system is a tertiary amine-catalyzed, dicyandiamide-cured epoxy system. It is similar to NEMA grade FR-4 commonly used in the industry for printed circuit composites except that the present system

utilizes a brominated epoxy based on tetrabromobisphenol A, further modified with a higher functionality epoxy to improve heat and solvent resistance. The FR-4 epoxy resin samples for TMA experiments were prepared by coating the FR-4 solution on a circular glass coverslip, followed by heating the solution on a Mettler FP5-FP52 programmable hot stage under a given schedule to remove the volatiles and partially cure the epoxy. Samples of thicknesses varying from 6 ( $\theta = 12$ ) to 12 mils ( $\theta = 6$ ) were prepared. During heating, a Fluke 2165A Digital Thermometer was used to monitor the temperature of the coverslip. The glass coverslip was chosen because of its relatively low thermal expansion coefficient in the temperature range between 22 and 190°C (the linear thermal expansion coefficients of various glasses are, on the average,  $9 \times 10^{-6}\text{C}^{-1}$  between 0 and 100°C<sup>18</sup>). Each coverslip had an overall area close to that of quartz probe (0.145 in. in diameter) with its thickness individually measured by the TMA or a micrometer before solution coating.

Subsequently, the partially cured resin samples were kept in vacuum before TMA experiments to remove residual volatiles (< 6% by weight using thermogravimetric analysis). Without this step, the presence of solvents and humidity would adversely affect the TMA viscosity determination (see below). After removal from vacuum, each sample was used immediately.

### Thermomechanical Analysis

A Perkin-Elmer (P-E) TMS-2 thermal mechanical analyzer was employed in the present study using the furnace temperature controller in a P-E DSC-2C Differential Scanning Calorimeter. Figure 1(a) shows a schematic drawing of the microparallel plate assembly of a TMA, where the specimen on the platform of a nonmoving quartz tube is under a force exerted by a flat-ended, cylindrically shaped quartz probe. The probe assembly is supported by means of a plastic float which is totally immersed in a high-density fluorocarbon fluid. The net force on a sample was obtained by subtracting the weight needed to balance the upward buoyancy force of the probe assembly from the total weight exerted by the weight tray and/or a P-E DMA-2 Dynamic Mechanical Analyzer. As the sample deformed, its thickness displacement as a function of temperature (or time) was monitored by a highly sensitive linear variable differential transformer (LVDT) and was recorded with a P-E 3600 Thermal Analysis Data Station. Both the furnace temperature and LVDT displacement were calibrated for accuracy according to standard P-E procedures.

Before each TMA experiment, the probe position was zeroed against the bottom of quartz platform under some net force (3 g). The epoxy sample on the coverslip was then centered directly under the probe. To reduce problems caused by the nonflatness of sample surface, a light net force (0.5 g) was applied to the sample at a temperature (50–60°C) just enough to soften the resin. The selection of proper force and temperature depends on the initial cure extent of the resin. The duration of this treatment should be kept short ( $\sim 5$  s) to avoid significantly altering the resin cure state. Following this step, the force was released and the sample as well as the furnace was quickly cooled to room temperature.

Throughout this study, a net constant force of 3 g was applied by the

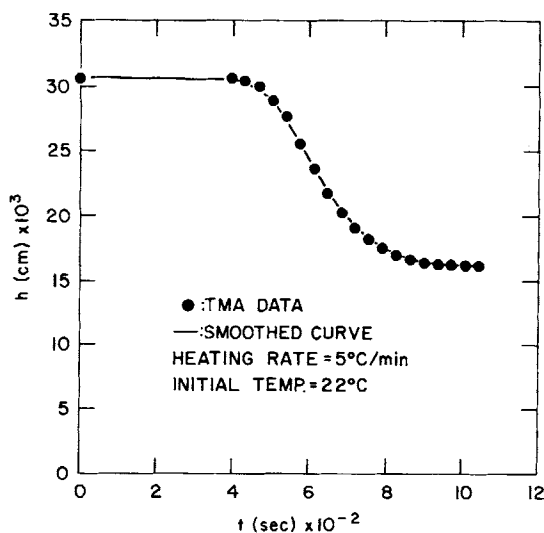
probe to the resin sample. The total sample thickness (including the coverslip) was measured by the displacement of the probe from its zeroed position at room temperature. Using the coverslip thickness, the initial sample thickness was estimated. After adjustment to the desired initial temperature (at a rate of 320°C/min for isothermal situations), the TMA monitoring commenced. In the ensuing section, the nonisothermal viscosity results under heating rates of 5, 10, and 20°C/min will be discussed and compared using a rheological model with predictions from isothermal TMA experiments performed at 60, 90, 120, and 150°C.

## RESULTS AND DISCUSSION

Figure 2(a) shows the thickness-time relationship from a TMA experiment (●) and the results (—) obtained by local least-square smoothing (first-degree polynomials relative to three points) for a 12.1-mil thick epoxy sample under a force of 3 g and a heating rate of 5°C/min from room temperature (22°C). From the smoothed curve, slopes at various times were determined by quadratic interpolation for estimates of viscosity using eq. (1) [Fig. 2(b)]. The thermal expansion coefficient of quartz glass is very low ( $6 \times 10^{-7} \text{C}^{-1}$  within 16–1000°C<sup>18</sup>), thus its contribution to thickness variations was neglected. Defining

$$\lambda = \int_0^t \frac{1}{\eta} dt$$

the  $(1/h^2)-\lambda$  relationship corresponding to Figure 2(b) was plotted in Figure 2(c). The linearity is expected from eq. (3) for constant  $F$  and  $R$ . Using Figure 2(c), a very small wall strain rate, of the order of  $10^{-2} \text{ sec}^{-1}$ , was obtained. Small strain rates are characteristic of parallel-plate experiments.



(a)

Fig. 2. (a) Thickness-time, (b) viscosity-time, and (c)  $1/h^2-\lambda$  relationships of a vacuum dried epoxy sample under a heating rate of 5°C/min and a force of 3 g during the TMA experiment: (●) TMA data; (—) smoothed curve.

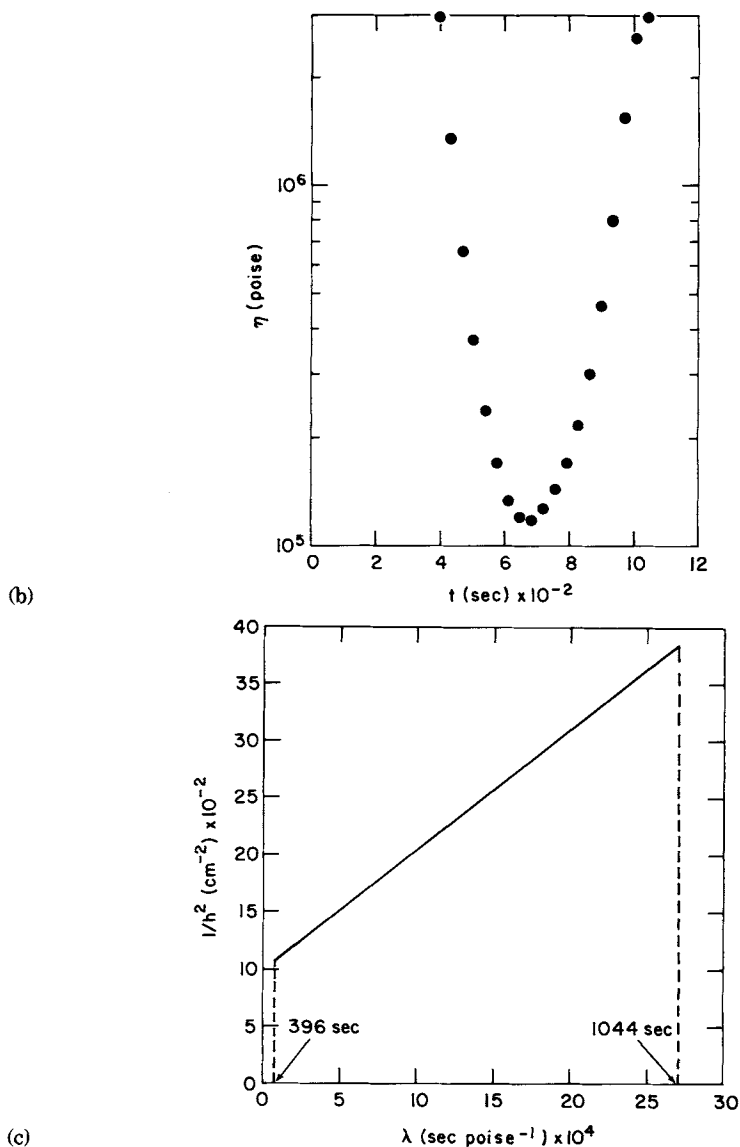


Fig. 2. (Continued from previous page.)

When the initial sample thickness was varied between 6 and 12 mils, the results shown in Figure 3 were obtained. The consistency here indicates that within this range the effect of thickness in determining viscosity is not important.

Generally speaking, the viscosity history of a thermosetting resin (e.g., epoxy) is controlled by competition between thermal softening and the thermally-induced crosslinking (curing) reaction. During the early stages of the experiment [Fig. 2(b)], the thermal softening effect is dominant because the resin viscosity decreases with temperature (or time). After the viscosity minimum is reached at about 700 s, an increase in viscosity with time indicates that the curing reaction dominates the deformation process as the

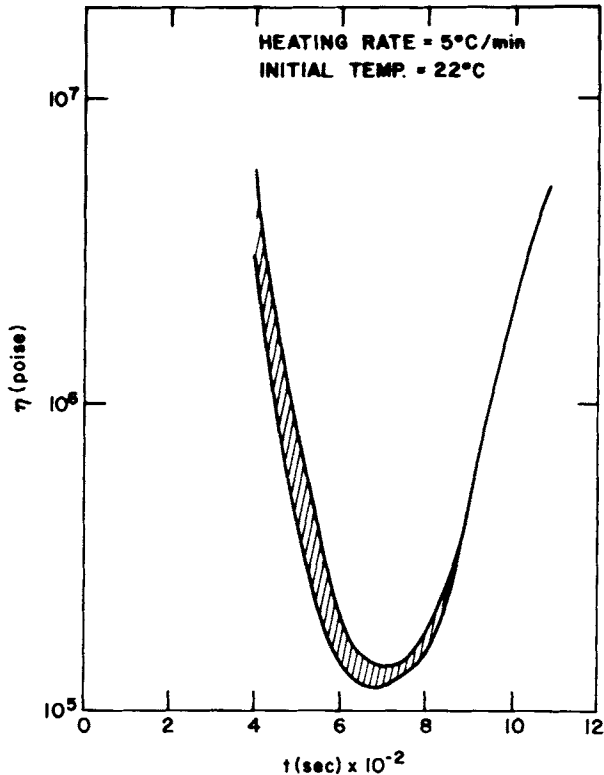


Fig. 3. Viscosity-time relationship of epoxy samples with thicknesses ranging from 6 to 12 mils under identical conditions as in Figure 2.

resin gel point is approached. Similar viscosity-time relationships have also been observed for polyaromatic resins<sup>8</sup> and various other epoxy systems.<sup>2,5</sup>

Figures 2 and 3 show data for resin samples which have been vacuum dried (for removal of volatiles) before the TMA experiments. If this step is omitted and the sample is left in laboratory ambient, the residual volatiles and moisture uptake in the sample may change the viscosity-time curve in Figure 2(b) to that shown in Figure 4(b) (with poorer reproducibility). A similar viscosity-time relationship was obtained from cone-and-plate studies of a different epoxy resin system cured in a high humidity environment.<sup>19</sup> In Figure 4(b), the viscosities at shorter times are lower than those in Figure 2(b) for vacuum-dried samples, indicating possibly a water-induced plasticization effect. The peak appearing between 800 and 1000 s probably relates to water vaporization. Corresponding to this peak is the presence of a bump in the thickness curve [Fig. 4(a)]. For dicyandiamide cured epoxy systems (as in the present case), a mechanism accounting for the water-epoxy interactions during curing has been proposed<sup>20</sup> in terms of reaction between the dicyandiamide and water. Given the large discrepancies between Figures 2(b) and 4(b), it is obvious that care must be taken to remove the volatiles before each TMA experiments to ensure reproducibility.

During a composite lamination cycle, the heating rate frequently varies. A varying rate heating schedule cannot be readily implemented using the



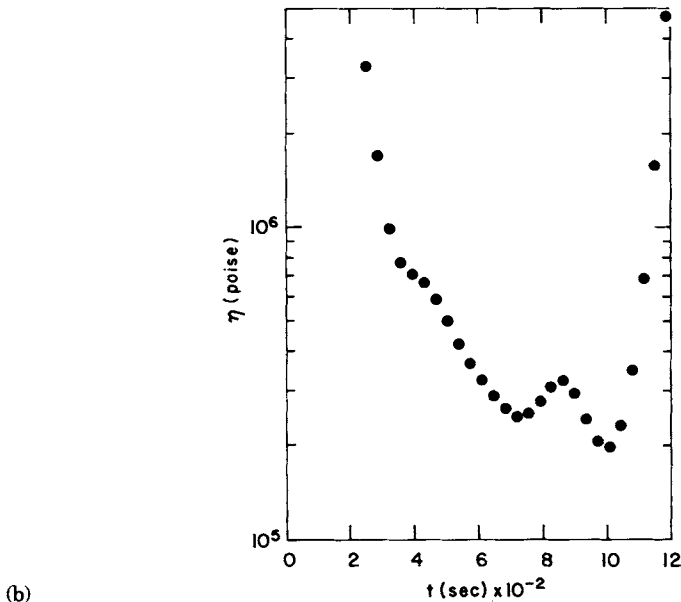
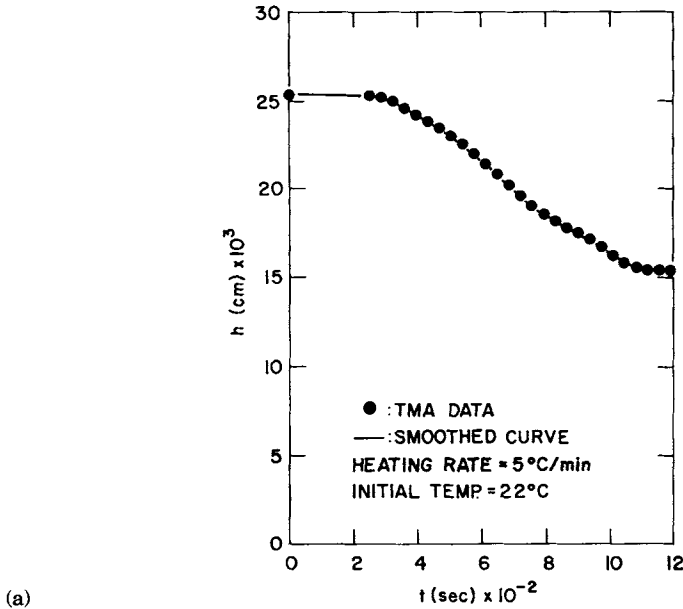


Fig. 4. (a) and (b) were obtained under identical conditions as in Figure 2 without vacuum drying the sample: (●) TMA data; (—) smoothed curve.

DSC temperature controller. In order to follow the viscosity history and actual lamination conditions, it is desirable to have a rheological model which is capable of estimating viscosity changes under any type of dynamic temperature schedule using viscosity data from a limited number of isothermal TMA experiments. To meet this end, we have used the rheology model developed by Roller<sup>2</sup> for predicting thermoset viscosity from a cone-and-plate viscometer. The applicability of this model can be confirmed by com-

paring viscosity data from dynamic TMA experiments at constant heating rates (covering the rate range in question) with those predicted by the model using data from isothermal experiments.

In the Roller model, the isothermal viscosity data are first correlated with an empirical dual-Arrhenius expression

$$\ln \eta[t] = \ln \eta_{\infty} + \frac{\Delta E_{\eta}}{RT} + tk_{\infty} \exp\left(\frac{-\Delta E_k}{RT}\right) \quad (5)$$

which is equivalent to

$$\eta = \eta_0 \exp(kt) \quad (6)$$

where  $\eta_0$  and  $k$  satisfy the first-order activation energy expressions

$$\eta_0 = \eta_{\infty} \exp(\Delta E_{\eta}/RT)$$

and

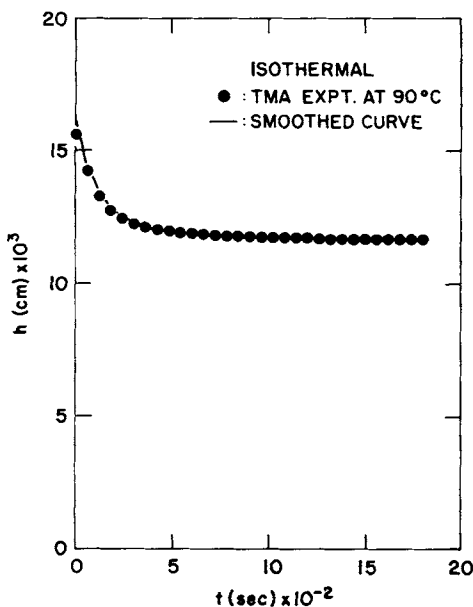
$$k = k_{\infty} \exp(-\Delta E_k/RT)^1$$

$\eta_{\infty}$  is interpretable as the resin viscosity at zero time and infinite temperature,  $R$  is the universal gas constant,  $k_{\infty}$  is the apparent reaction constant at infinite temperature, and  $\Delta E_{\eta}$  and  $\Delta E_k$  are the activation energies for viscosity and reaction rate constants. Thus, for each isothermal TMA experiment, the logarithm of the viscosity  $\eta$  plotted versus time should yield a straight line with slope and intercept, respectively, equal to  $k$  and  $\ln \eta_0$ . Using the values of  $\eta_0$  and  $k$  obtained from a number of isothermal experiments covering the temperature range of interest, the parameters  $\eta_{\infty}$ ,  $k_{\infty}$ ,  $\Delta E_{\eta}$ , and  $\Delta E_k$  can be determined by least-square fitting. In order to model the viscosity  $\eta[t, T]$  under transient temperature conditions, Roller<sup>2</sup> proposed

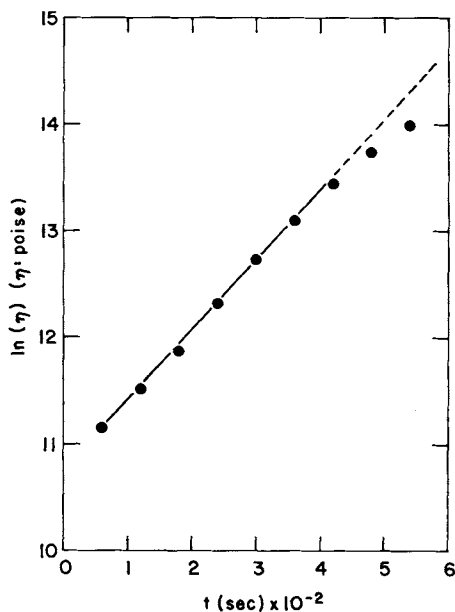
$$\ln \eta[t, T] = \ln \eta_{\infty} + \frac{\Delta E_{\eta}}{RT} + k_{\infty} \int_0^t \exp\left[\frac{-\Delta E_k}{RT}\right] dt \quad (7)$$

Although modifications to eq. (7) have been suggested, for example, by the introduction of a chain entanglement factor<sup>21</sup> and a reaction-order term,<sup>5</sup> Roller's initial model was employed in this study. Instead of obtaining the parameters from a single dynamic run,<sup>22</sup> the values were estimated from isothermal viscosity data following the procedure described previously in order to check the applicability of Roller's model.

Isothermal TMA experiments were performed at 60, 90, 120, and 150°C. Figures 5(a) and 5(b) show the results at 90°C. The viscosity in Figure 5(b) increases monotonically with time and a linear relationship between  $\ln \eta$  and  $t$  holds up to  $t = 400$  s. The break point is typical of the approach to the gel point. It defines the upper time limit, beyond which eq. (6) no longer applies. The values of  $\eta_0$  and  $k$  in this case were determined to be  $6.8 \times$



(a)



(b)

Fig. 5. (a) Thickness-time and (b)  $\ln \eta$ - $t$  relationships for an isothermal TMA experiment at 90°C under a force of 3 g: (●) TMA data; (—) smoothed curve.

$10^4$  P and  $5.4 \times 10^{-3} \text{ s}^{-1}$  by eq. (6). From the values of  $\eta_0$  and  $k$  at all four temperatures, we obtained:  $\Delta E_\eta = 1.733 \times 10^4 \text{ cal/mol}$ ,  $\eta_\infty = 1.879 \times 10^{-6} \text{ P}$ ,  $\Delta E_k = 1.104 \times 10^4 \text{ cal/mol}$ , and  $k_\infty = 2.845 \times 10^4 \text{ s}^{-1}$ . The activation energies are of the same order of magnitude as those given by Roller<sup>2</sup> for his epoxy system.

The experimental viscosity-time relationships (●) at heating rates of 20, 10, and 5°C/min are shown in Figures 6, 7, and 8 along with the theoretical

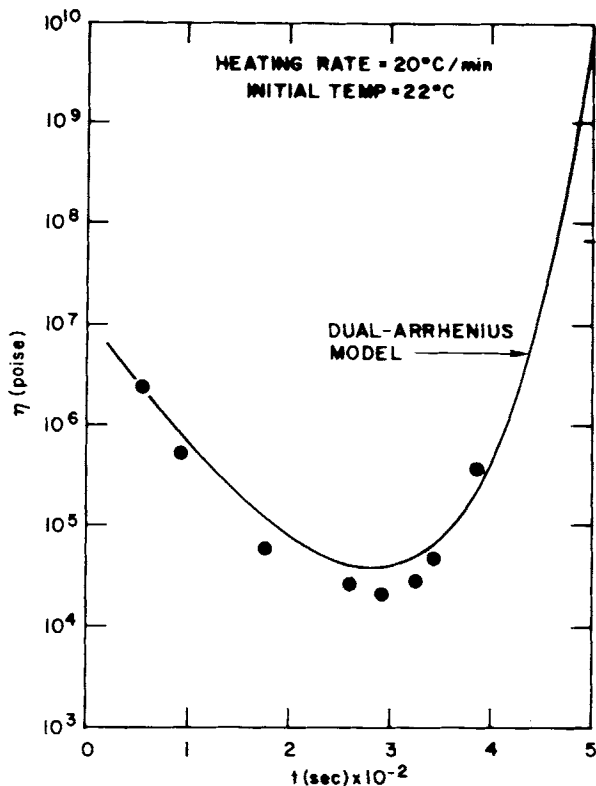


Fig. 6. Viscosity-time relationships from a dynamic TMA experiment (●) and the dual-Arrhenius model (—) at a heating rate of 20°C/min.

predictions (—) from the dual-Arrhenius model. While good agreement between experiment and theory is obtained with 20 and 10°C/min, the agreement in the case of 5°C/min is poor. The discrepancies between model and viscosity data under conditions of low heating rate can be explained following Roller.<sup>2</sup> According to Nielsen,<sup>23</sup> the Arrhenius expression

$$\eta_0 = \eta_\infty \exp(\Delta E_\eta/RT)$$

in eq. (6) holds for temperatures considerably above the material's glass transition temperature  $T_g$  (which increases with the degree of resin cure). At relatively low heating rates, the sample temperature remains closer to the material's  $T_g$ , and therefore the measured viscosities are higher than the predicted values (Fig. 8).

The actual temperature profiles during the lamination of printed circuit composites are typically nonlinear with an initial heat-up rate as high as 20–30°C/min, followed by an asymptotic decrease as the press temperature is approached.<sup>2</sup> The important region for viscosity flow is the early time, low viscosity region when the heat-up rates are still high.<sup>11</sup> Although the higher viscosity region (corresponding to a relatively low heating rates) in the proximity of the gel point may be important to final composite properties, it is of little importance to flow. Since Roller's model predicts well the viscosity—time relationship at high heating rates, it can be used to

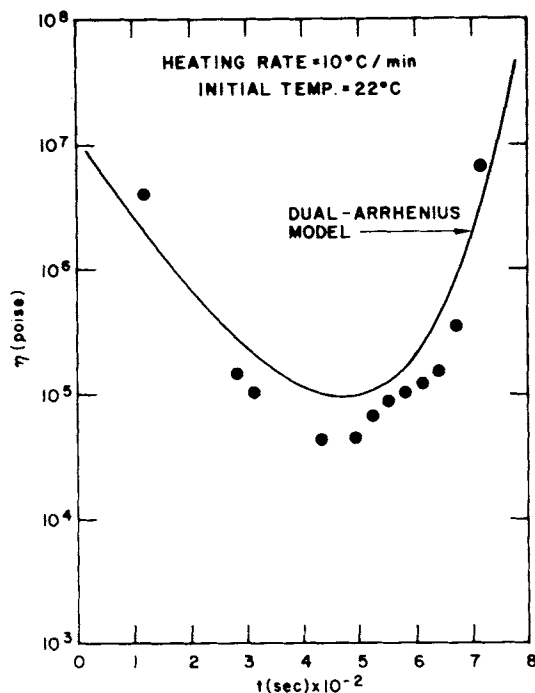


Fig. 7. Viscosity-time relationships from a dynamic TMA experiment (●) and the dual-Arrhenius model (—) at a heating rate of 10°C/min.

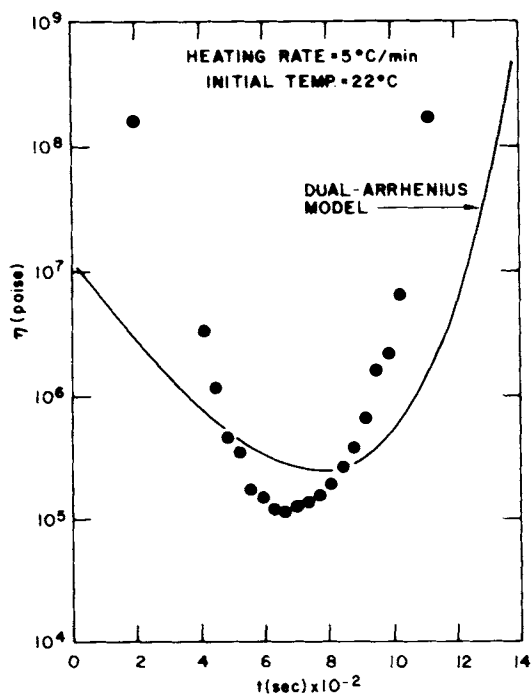


Fig. 8. Viscosity-time relationships from a dynamic TMA experiment (●) and the dual-Arrhenius model (—) at a heating rate of 5°C/min

study the viscosity behavior of the present FR-4 resin system for composite manufacture.

### CONCLUSION

A simple and quick approach has been presented for the prediction of thermoset viscosity under composite lamination conditions using a dual-Arrhenius rheology model and the isothermal TMA viscosity data. The small samples required can be prepared either from solution or from the resin powder flaked off the resin/glass prepreg. Although only an FR-4 resin system was considered here, it is obvious that other resin systems can also be studied. With a controlled humidity environment, it is possible to use the present approach for investigating the humidity effects on resin viscosity history during lamination.

The authors are very grateful to T. C. Clarke for many stimulating discussions and suggestions. The useful comments from C. Feger are also acknowledged.

### References

1. D. P. Bloechle, *J. Elast. Plast.*, **10**, 377 (1978).
2. M. B. Roller, *Polym. Eng. Sci.*, **15**, 406 (1975).
3. R. R. Eley, in *Chemorheology of Thermosetting Polymers*, C. A. May, Ed., ACS Symposium Series 227, Am. Chem. Soc., Washington, DC, 1983.
4. G. J. Dienes and H. F. Klemm, *J. Appl. Phys.*, **17**, 458 (1946).
5. M. R. Dusi, C. A. May, and J. C. Seferis, in *Chemorheology of Thermosetting Polymers*, C. A. May, Ed., ACS Symposium Series 227, Am. Chem. Soc., Washington, DC, 1983.
6. J. H. Dillon and N. Johnson, *Physics*, **4**, 225 (1933).
7. E. H. Fontana, *Ceramic Bull.*, **49**(6), 594 (1970).
8. L. C. Cessna, Jr., and H. Jabloner, *J. Elast. Plast.*, **6**, 103 (1974).
9. R. B. Prime, in *Thermal Characterization of Polymeric Materials*, A. Turi, Ed., Academic, New York, 1981.
10. W. Engelmaier and M. B. Roller, Proc. NEPCON, Anaheim, CA, 1975, pp. 86-96.
11. C. J. Barlett, *J. Elast. Plast.*, **10**, 367 (1978).
12. J. Del, P. Marx, and J. Sallo, IPC-TP-420, 1982.
13. R. Darby, *Viscoelastic Fluids*, Marcel Dekker, New York, 1976, pp. 60-65.
14. J. Happel and H. Brenner, *Low Reynolds Number Hydrodynamics*, Prentice-Hall, Englewood Cliffs, NJ, 1965.
15. R. J. Grimm, *Appl. Sci. Res.*, **32**, 149 (1976).
16. A. N. Gent, *Br. J. Appl. Phys.*, **11**, 85 (1960).
17. J. R. Van Wazer, J. W. Lyons, K. Y. Kim, and R. E. Colwell, *Viscosity and Flow Measurement—A Laboratory Handbook of Rheology*, Wiley-Interscience, New York, 1963.
18. R. H. Perry and C. H. Chilton, *Chemical Engineers' Handbook*, McGraw-Hill, New York, 1973.
19. R. Hinrichs and J. Thuen, *SAMPE J.*, (Nov.-Dec., 1979) pp.12-21.
20. R. A. Pike, F. P. Lamm, and J. P. Pinto, *J. Adhesion*, **13**, 229 (1982).
21. J. D. Keenan, SAMPE Educational Workshop, Sunnyvale, CA, 1980.
22. R. R. Eley, *Org. Coatings Appl. Polym. Sci., Proc.*, **47**, 503 (1982).
23. L. E. Nielsen, *Mechanical Properties of Polymers*, Reinhold, New York, 1962.

Received March 6, 1985

Accepted October 5, 1985