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Margaret E. Roylance, David K. Roylance, and Kathryn L. Nasmith

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USE OF ULTRASONIC TAPE LAMINATION FOR IN-PROCESS DEBULKING OF THICK COMPOSITE STRUCTURES

Margaret E. Roylance, Foster-Miller, Waltham, MA
David K. Roylance, Department of Materials Science and Engineering, MIT, Cambridge, MA
Kathryn L. Nesmith, NAWCAD, Patuxent River, MD

ABSTRACT

Foster-Miller has developed an advanced processing technique for organic matrix composites called Ultrasonic Tape Lamination (UTL). This technique employs ultrasonic energy to insonify composite tape (either thermoplastic or B-staged thermoset) to induce controlled and virtually instantaneous viscoelastic and frictional heating in the composite. UTL can be used for in-process debulking and controlled staging of thick filament wound or fiber placed composite structures. During UTL the ultrasonic horn functions as a localized, efficient, and quick response heat source, ideally suited for a feedback control system. The horn heats only the area with which it is in direct contact, and ultrasonic loading can enhance fiber nesting to optimize debulking. With the use of UTL on-the-fly debulking during filament winding or tape placement, a net shape part can be realized without time consuming and expensive repetitive debulking steps. Problems associated with inadequate debulking, including fiber waviness (or marcelling), fiber shifting, resin migration, disbonds and voids, can be eliminated. Higher quality thick composite parts can be achieved with minor process modifications, resulting in a superior and less costly part.

KEYWORDS: Composites, Curing , Modeling

1. INTRODUCTION

The use of ultrasonic welding of metals and unreinforced polymeric materials is an important industrial process, and has been used successfully for many years. The use of ultrasound for the consolidation of polymer matrix composites containing more than 35-40% by volume of reinforcing fiber was viewed as impossible (1) until Foster-Miller developed innovative techniques for ultrasonic tape lamination (UTL) of these materials. Working in cooperation with NAWCAD, Foster-Miller has demonstrated the use of UTL for both consolidation of thermoplastic matrix composite materials and debulking of B-staged thermoset prepreg. The theoretical similarity between the two processes has been confirmed based on experiment and analysis. In both thermoplastic consolidation and thermoset debulking without staging, the ultrasonic loading must generate sufficient heat to inducing flow and provide sufficient support for consolidation. One of the singular advantages of UTL technology is that heating and cooling occur very rapidly and in a more controlled manner than during a hot gas process. Due to this inherent controllability of the process, UTL debulking of B-staged prepreg might also be used to stage the material in a controlled way. In addition, ultrasonic loading can enhance fiber nesting to optimize consolidation.

Figure 1 shows the Foster-Miller UTL head mounted on a filament winder. The horn configuration and stress applied to the composite material is critical to achieving full consolidation without inducing damage in the material. The data in Figure 2 help to clarify the importance of applied stress state to the process. Figure 2 shows the bulk (K) and shear (G) relaxation moduli for a polymeric material (in this case polyisobutylene) on a time scale reflecting the glass transition. The intensity of the relaxation and hence the amount of heat dissipated is much greater in shear or distortional loading than in bulk or dilatational loading. When the relative magnitude of the shear component of loading compared to the hydrostatic pressure is optimized, the material is heated viscoelastically with minimum fiber disruption. This can be accomplished without the use of so-called energy directors, or raised areas which must be formed on the surface of the material.

2. NATURE OF THE UTL PROCESS

Controlled use of UTL for debulking and staging of a reactive material requires a thorough understanding of the UTL process. Careful observation and monitoring of the process was undertaken to clarify the nature of the UTL process and to provide a basis for the simplifying assumptions required for numerical modeling.

One very important question which must be answered in order to develop a quantitative model of heat generation and consolidation in these materials is the relative importance of frictional and viscoelastic effects. These materials are relatively highly damping and significant viscoelastic heating will certainly occur under ultrasonic loading. The extent to which frictional heating occurs as well will change the magnitude and location of heat generation. This is due to the fact that frictional heating should generate high temperatures at the interfaces between horn and tape and between tape and part, whereas viscoelastic heating will occur throughout the volume of the affected material. In an effort to clarify the relative importance of frictional and viscoelastic heating, a study was performed of the effect of static pressure on the heat generation in AS4/PEEK tape.

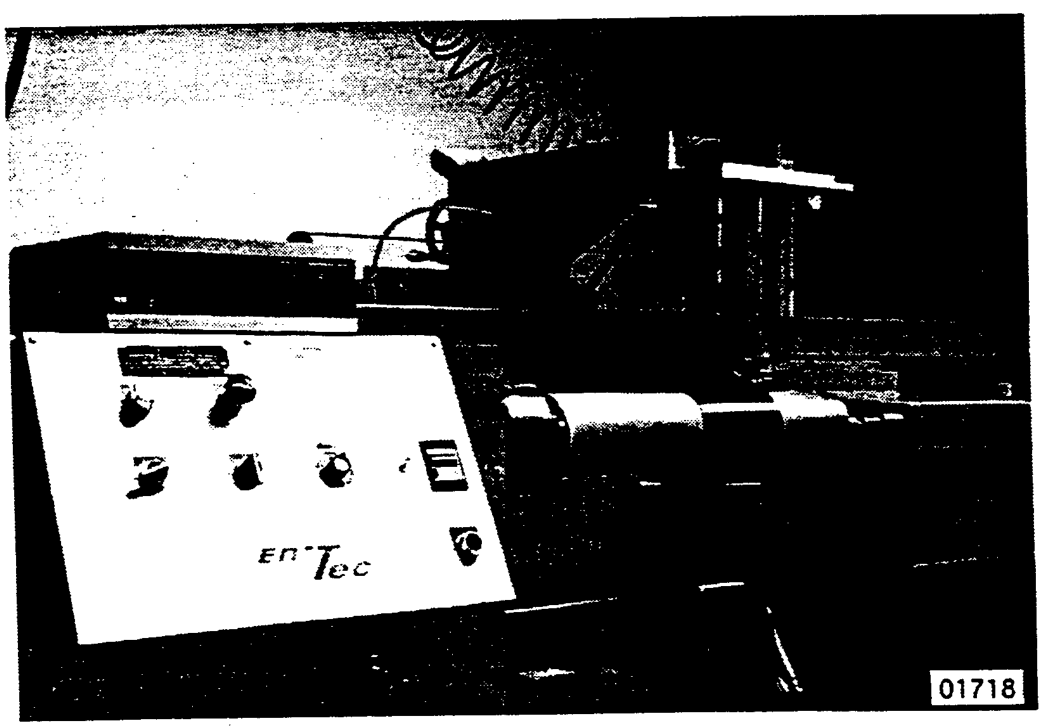


Figure 1. Ultrasonic Tape Lamination during filament winding

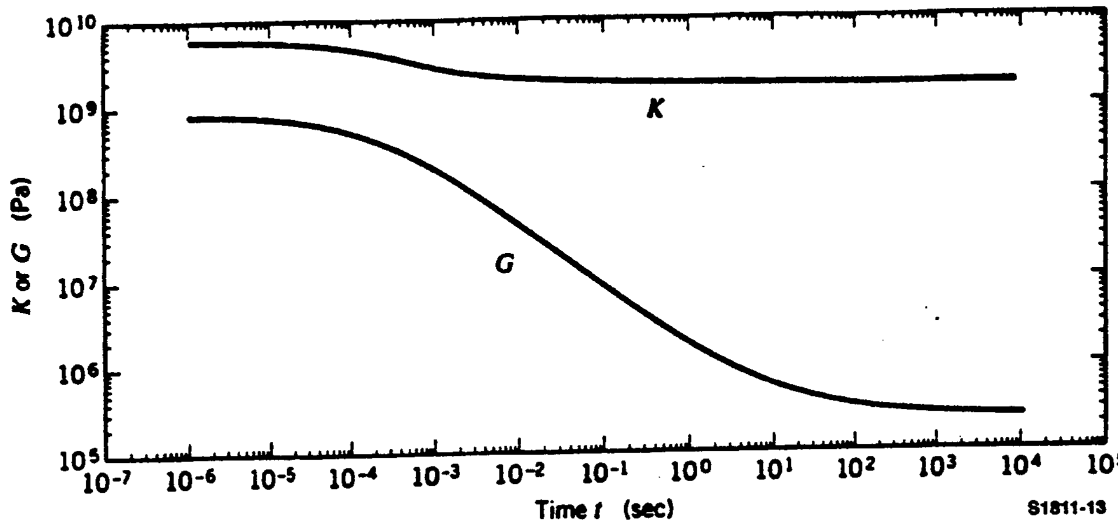


Figure 2. Bulk (K) and shear (G) relaxation moduli of polyisobutylene (from reference 2)

To clarify the relative importance of frictional and viscoelastic heating, the effects of pressure and amplitude on ULT-induced heating were measured. A thermocouple was embedded in the center of a thermoset composite substrate four inches in length. The temperature was measured during UTL of four plies of AS4/PEEK tape. The advance rate of the ultrasonic horn was constant at 2.54cm (1 inch)/minute and an ultrasonic frequency of 40 kHz was used for all the measurements. Figure 3 shows the effect of static pressure and amplitude on the measured heating of the AS4/PEEK tape during insonification.

The data in Figure 3 shows that increasing the static pressure has no observable effect on the heat generation in the material during UTL. If frictional heating were dominating the process, increasing the static pressure should increase the rate and magnitude of heat generation. This result suggests that the viscoelastic heating is dominating the UTL heat generation. It also indicates that the static pressure is sufficient to maintain contact throughout the ultrasonic loading cycle, or the viscoelastic heating would also be a function of static pressure level.

This conclusion is in agreement with the results of Tolunay et al. (3) that, for soft polymers, the interface did not have a significant effect on the amount of heat dissipated during ultrasonic welding of unreinforced materials. They observed that in this case the heating occurs over the whole volume. Further, they observed that intensive heating of the material began only after a certain temperature was reached. This temperature most probably corresponds to the glass transition temperature, since as Tg is approached, the level of viscoelastic energy dissipation as measured by loss modulus increases markedly. As the material continues to heat above Tg, the loss modulus drops again, and Tolunay et al. observed that the heating rate also generally drops until the temperature remains constant. The very rapid heating which occurs at surface asperities should soften the material in these regions quickly and allow for good uniform contact between the horn and the surface of the material.

Based on these results, our model focuses on viscoelastic energy dissipation as a volumetric heat source rather than frictional heating at the interfaces.

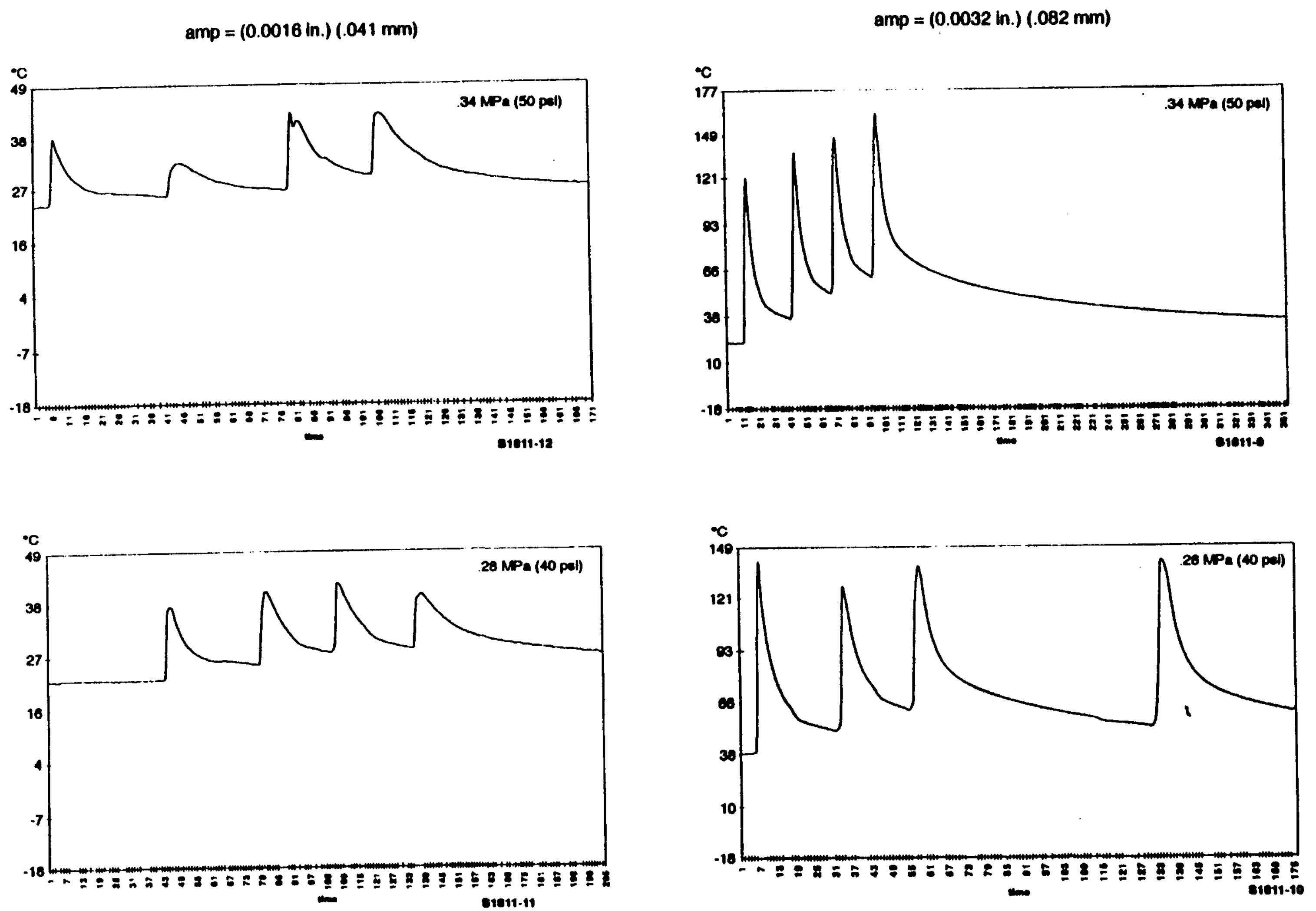


Figure 3. Effect of static pressure and amplitude on heating during insonification

3. FINITE ELEMENT MODELING OF UTL

3.1 Computational Model We have chosen to employ a finite element analysis (FEA) developed by Roylance et al. (4) to model nonisothermal reactive processing operations, and which Benetar et al. (5) have used to predict the behavior of AS4/PEEK during attempted ultrasonic welding using energy directors. One advantage of this model is that it includes the effect of chemical reaction, which will allow us to use it to predict the extent of cure of reactive species in the B-staged prepreg. There are many experimental parameters in ultrasonic heating, to include horn angle, contact pressure, frequency and amplitude, presence of attenuating layers, and others. This makes optimization by purely experimental trials difficult and time-consuming, and computer modeling is useful in this regard.

The FEA code models the equations governing the nonisothermal flow of a reactive fluids, listed in standard texts on transport phenomena and polymer processing (e.g. References 6,7). These are the familiar conservation equations for transport of momentum, energy, and species:

$$\rho\left[\frac{\partial u}{\partial t}+u\nabla u\right]=-\nabla p+\nabla(\eta\nabla u)$$

$$\rho c\left[\frac{\partial T}{\partial t}+u\nabla T\right]=Q+\nabla(k\nabla T)$$

$$\left[\frac{\partial C}{\partial t}+u\nabla C\right]=R+\nabla(D\nabla C)$$

Here u, T, and C are fluid velocity (a vector), temperature, and concentration of reactive species; these are the principal variables in our formulation. Other parameters are density (ρ), pressure (p), viscosity (η), specific heat (c), thermal conductivity (k), and species diffusivity (D). The ∇ operator is defined as:

$$\nabla=\frac{\partial}{\partial x},\frac{\partial}{\partial y}$$

Q and R are generation terms for heat and chemical species respectively, while the pressure gradient ∇p plays an analogous role for momentum generation. The heat generation arises from viscous dissipation and from reaction heating:

$$Q=\tau:\dot{\gamma}+R(\Delta H)$$

where τ and $\dot{\gamma}$ are the deviatoric components of stress and strain rate, R is the rate of chemical reaction, and ΔH is the heat of reaction. R in turn is given by a kinetic chemical equation; in our model we have implemented an m-th order Arrhenius expression:

$$R=k_0\ \exp\left(\frac{-E*}{R_g T}\right)C^m$$

where k_o is a preexponential constant, $E*$ is an activation energy, R_g = 8.31 J/mol-K is the Gas Constant, and m is the reaction order. The material-dependent parameters in this expression (k_o, $E*$, and m) must be determined by experimentation which is able to monitor the reaction as a function of time and temperature. An example of reaction kinetics modeling suitable for this purpose has been presented by Roylance (3,4).

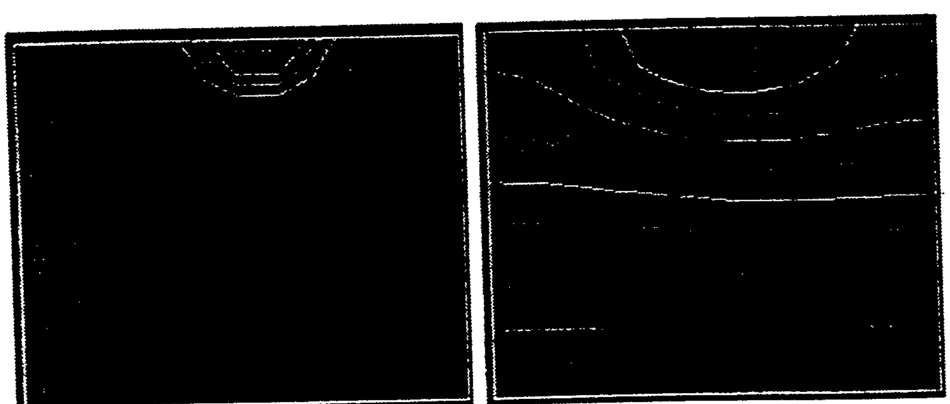
In treating ultrasonic curing, we take the velocities u in the governing equations to be displacements, and suppress the advective flow terms (e.g. $u \nabla u$). In this approach, the dilatational and deviatoric components of stress are considered separately, which leads to convenience in handling incompressible materials such as rubber. It also simplifies the computation of internal dissipative heating, which arises principally from the deviatoric components of stress. Once the deviatoric stresses and strains τ and γ have been computed, the dissipated heat per unit time Q is computed as

$$Q = \omega f \tau \gamma$$

where ω is the cyclic frequency and f is the fraction of strain energy dissipated per cycle. The dissipation factor f is known from dynamic mechanical testing; it is 2π times the tangent of the phase angle between cyclic stress and strain measured in these tests.

The analysis operates in iterative fashion, first solving the force equilibrium equations to determine the distribution of displacement, strain and stress generated in the body by the application of the imposed displacements at the surface. This heat dissipation associated with the viscous loss is used in the heat transfer equation, which is solved for an updated estimate of the temperature in a second iteration. In a third iteration the temperatures obtained in the previous step are used to update the rate of the chemical curing reaction. The equations are strongly coupled, with properties such as the shear modulus and chemical reaction rate depending on temperature, and the temperature depending on both viscous and reaction heating. The computer iterates repeatedly until convergence is reached. The code can also use successive iterations as time steps, so that transient situations can be modeled as well.

3.2 Results of modeling Figure 4 illustrates a typical modeling result. Figure 4a shows a stress distribution (here the vertical stress σ_y) for the case of a horn applying a vertical displacement to a portion of the upper surface of a layered specimen of cured composite and unreinforced polymer. Chemical reaction is not included in this simulation. The stresses are concentrated near the point of application, so the heat generation rate is highest here. However, heat will also be carried to other regions of the specimen by conduction. The temperature field arising from this oscillatory stress field is shown in Figure 4b. These stress and temperature contours were generated from normalized numerical results.



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a) Stress distribution b) Temperature distribution

Figure 4. Typical FEM results

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One way of viewing the effect of ultrasonic heating is to plot the variation of temperature along a vertical line directly beneath the horn as a function of time. In Figure 5 below, the code was set to simulate a multilayer laminate consisting of unreinforced polymer with fiber reinforced composite layers on the top and bottom. A silicone rubber attenuation layer separates the upper face sheet from the ultrasonic horn. The lower composite sheet extends from $y = 0$ to $y = 0.00075$ m (it is convenient to operate the code in SI units), the polymer extends from $y = 0.00075$ to 0.00204 m, the upper face sheet extends from $y = 0.00204$ to 0.00279 m, and the silicone sheet extends from $y = 0.00279$ to 0.00359 m.

3.3 Model development To predict consolidation of composite materials such as AS4/PEEK or B-staged thermosets, the local compressive stress and temperature of the material can be predicted. The stress and temperature required for consolidation can be experimentally determined and the model can be used to identify the ultrasonic parameters which produce these conditions. In order to extend this analysis to staging of prepreg, kinetic parameters appropriate to the epoxy curing reaction must be introduced, and the effects of the curing exotherm on the material temperature calculated. As described above, the model currently has the capability for such analysis, but the kinetic parameters for epoxy reactions have not yet been incorporated.

The extent of consolidation or debulking achieved by UTL in AS4/PEEK has been determined by density and microscopy measurements. Experimental results have shown that good consolidation of AS4/PEEK can be achieved with UTL. Apparent interlaminar shear strength of this material by short beam shear was 82.7 Mpa (12 KSI). This value is within the normal range for autoclave consolidated AS4/PEEK. Results such as these should be correlated with the ultrasonic parameters and the FEA simulations to verify the ability of the FEA model to predict consolidation.

It is clear from the governing equations that accurate modeling of UTL debulking and staging of thermoset composites requires accurate information concerning several materials parameters. These include modulus and log decrement as a function of temperature and frequency, as well as kinetic parameters for one or more epoxy curing reactions.

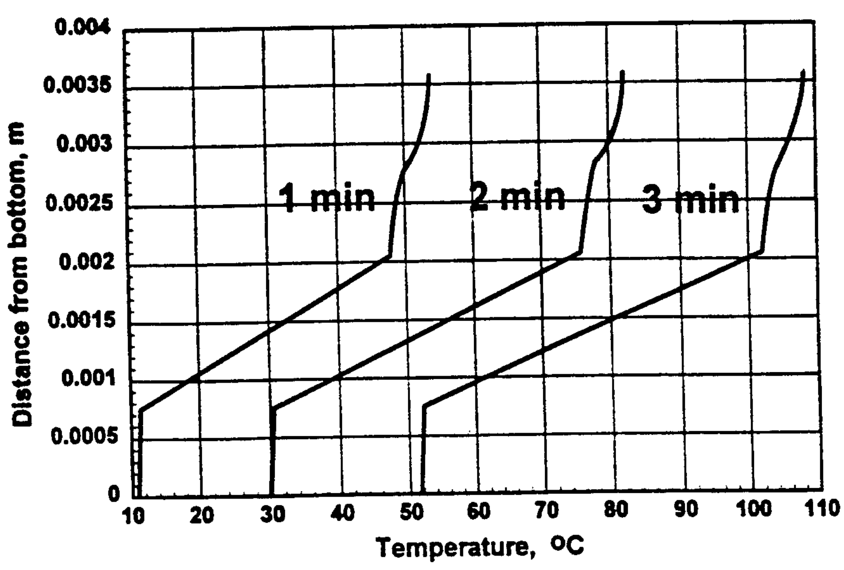
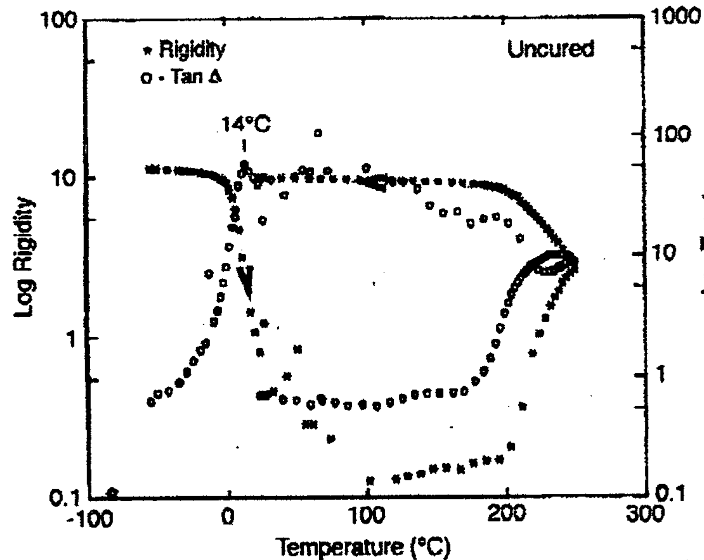
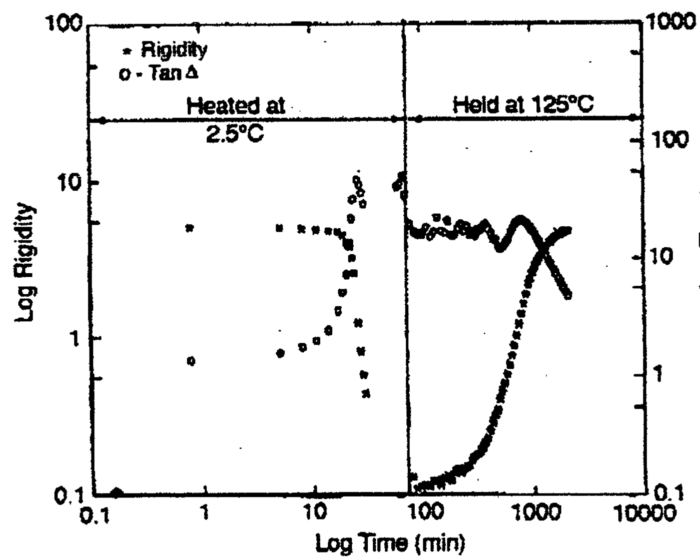


Figure 5. Predicted FEM temperature profiles

Some kinetic data for important epoxy and polyimide curing reactions are available in the literature (8,9). Some other information, such as full characterization of the effect of degree of cure on the dynamic mechanical properties of the curing material must be obtained experimentally. Torsion braid analysis (TBA) can be used to measure rigidity and loss tangent of B-staged prepreg resins during cure. An example of TBA of Cytek Fiberite 977-3 during both a slow heating cycle and staged cure at 125°C is shown in Figure 6. A series of TBA runs of this kind, coordinated with differential scanning calorimetry (DSC) to determine residual exotherm and extent of cure, would be required to characterize the dynamic mechanical properties which would control material behavior during UTL staging.



a) TBA results on B-staged IM7/977-3 prepreg at 2.5°C/min



b) TBA of staging of B-staged 977-3 prepreg at 125°C

Figure 6. TBA of 977-3

4. CONCLUSIONS AND RECOMMENDATIONS

The overall goal of this program was to establish UTL as a feasible approach for in-process debulking and staging of composite structures, and to identify measurable and controllable parameters that can be used for automated control of UTL during automated fiber placement or filament winding. This required development of a more thorough understanding of the nature of the process as it applies to both thermoplastic and B-staged thermoset matrix composites. Quantification of viscoelastic heating and extent of cure of the thermoset matrices were identified as central to the controlled use of UTL.

In pursuit of this goal, a modeling approach has been identified and implemented which is capable of incorporating both viscoelastic heating and chemical reaction, and is thus generally applicable to both thermoplastic and thermoset matrix composites. Future work should seek to extend the FEA to include epoxy curing reactions, and to verify the analysis during model experiments monitored using techniques such as thermometry, dielectrometery and fiberoptic FTIR.

The verified FEA model could then be used to assist in optimization of in-process debulking and staging during automated fiber placement. The model would be used to predict ultrasonic parameters such as amplitude, frequency and horn pressure which will produce consolidation. It could also predict the extent of cure under these conditions, and how far the curing reaction could be advanced during UTL debulking.

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