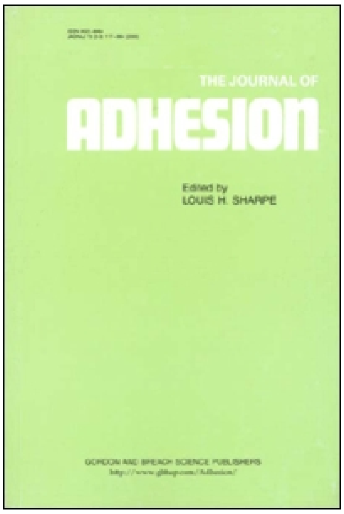

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The Effects of Molecular Weight on the Single Lap Shear Creep and Constant Strain Rate Behavior of Thermoplastic Polyimidesulfone Adhesive

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The bonded shear creep and constant strain rate behavior of zero, one, and three percent end capped Thermoplastic Polyimidesulfone adhesive were examined at room and elevated temperatures. End capping was accomplished by the addition of phthalic anhydrides.

The primary objective was to determine the effects of molecular weight on the mechanical properties of the adhesive in the bonded form. Viscoelastic and nonlinear elastic constitutive equations were utilized to model the adhesive. Crochet's relation was used to describe the experimental creep failure data. The effects of molecular weight changes on the above mentioned mechanical behavior were assessed.

The viscoelastic Chase–Goldsmith and elastic nonlinear relations gave a good fit to the experimental stress strain behavior. Ultimate stress levels and the safe levels for creep stresses were found to decrease as molecular weight was reduced.

KEY WORDS Chase–Goldsmith model; mechanical properties; molecular weight; polyimidesulfone adhesive; single lap shear creep; strain rate behavior.

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INTRODUCTION

The model adhesive for the present investigation, Thermoplastic Polyimidesulfone, is a novel thermoplastic adhesive currently under development at NASA-Langley Research Center. It is solvent resistant and can be used in thermal environments with temperatures up to 450°F (232°C).[1]

In an attempt to maximize mechanical strength and bonding properties, while reducing moisture penetration,[2] different molecular weight versions of the adhesive are being assessed by means of mechanical testing. Molecular weights were varied by the addition of phthalic anhydride end caps. The degree to which changes in molecular weight affects the viscoelastic properties of Thermoplastic Polyimidesulfone are studied at room and high temperatures. Single lap samples (ASTM D1002 standard geometry) are used because they are common to many structural fabrications where adhesives are used.

The designer of adhesively bonded joints must take into account three possible modes of failure:

1) Failure of the adhesive layer when the rupture stress is reached under monotonic loading;

2) Delayed failure caused by creep under a sustained load;

3) Adherend failure.

Adhesive failure in the creep and constant strain rate modes, the effects of molecular weight upon these modes of failure, and the adhesive's stress strain behavior in general, will be discussed in this paper. Adherend failure is not a prominent consideration for the specimens used.

Objectives

The purpose of this investigation is to provide insight into the effects of molecular weight on the creep and constant strain rate behavior of Thermoplastic Polyimidesulfone. The basic objectives are to:

- obtain experimental data on the delayed failure behavior of zero, one, and three percent end capped Thermoplastic Polyimidesulfone/titanium single lap specimens;

- obtain experimental data on the constant shear strain rate behavior of the specimens;
- identify viscoelastic or nonlinear models which will describe the mechanical behavior of the adhesive;
- determine the effects of molecular weight on the above mentioned mechanical properties of Thermoplastic Polyimidesulfone based on experimental data.

Literature review

The magnitude of their molecular weights is known to affect the mechanical strength of adhesives. The effects vary from material to material, and are most evident in the cohesive (pertaining to the adhesive matrix) and adhesive (pertaining to the interface) strengths of the adhesive.

Very little research has been performed on the effects of molecular weight on adhesives in the bonded form, where joint failures may occur at the adhesive/adherend interface. Tsuji, Masuoka and Nakao studied the effects of molecular weight on the peel strength of polyisobutylene used to bond polyethylene films.[3] Their results show increasing peel strength with increasing molecular weight up to a maximum value. Then, with further increases in molecular weight, the peel strength decreases abruptly due to adhesive/adherend interfacial failure. This behavior is attributed to reduced wetting and adhesive behavior of the polymer at increased molecular weight values.

Although molecular rupture has been interpreted as being a process of flow, reorientation, or bond rupture, these phenomena are not clearly separated.[4] Without specific regard to the microscopic behavior of a material, Alfrey[5] offers the opinion that the dependence of mechanical strength of a high polymer upon molecular weight is qualitatively the same for all polymers. Tensile strength is proportional to molecular weight, increasing as molecular weight increases, up to a limiting value. Once a certain level is reached, further increase in molecular weight does not result in appreciable improvements in strength. Alfrey states that low molecular weight polymers break mainly by pulling apart entangled chains, while high molecular weight specimens fail largely by the rupture of primary valence chains.

Schenck[6,7,8] investigated the mechanical behavior of zero percent end capped Thermoplastic Polyimidesulfone at four temperature levels. His results provides a set of data to be used in conjunction with the present investigation.

ANALYTICAL CONSIDERATIONS

A convenient way of relating the mechanical properties of a polymeric material to its molecular weight is through the Young's modulus in the elastic range and viscosity coefficient in the viscoelastic range. The viscosity coefficient in a viscoelastic model represents a dashpot within the schematic of the model, signifying some degree of fluid-like behavior. The Chase–Goldsmith model (which is a modified form of the three parameter solid model) incorporates viscosity into material characterization in this manner (see Figure 1). A sliding element prevents the dashpot from affecting the stress strain behavior until after an initial linear elastic response is seen. A similar behavior was observed during our experiments with Thermoplastic Polyimidesulfone adhesive. This behavior was also similar to that of another polyimide adhesive (LARC-3) which was characterized previously at Clarkson. Sancaktar and Padgilwar successfully used the Chase–Goldsmith model to characterize the constant strain rate behavior of LARC-3 adhesive.[9,10]

In shear, the constitutive equations for the Chase–Goldsmith

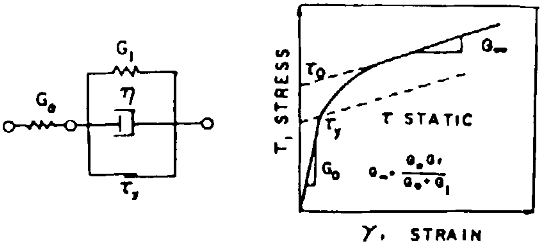


FIGURE 1 The viscoelastic Chase–Goldsmith model used to describe adhesive behavior.

model constant strain rate behavior are:

$$\tau = G_0\gamma; \qquad 0 < \tau \leq \tau_y$$

$$\tau = \frac{G_0}{G_0 + G_1}[(\tau_y + \bar{\eta}R + G_1\gamma)(1 - \exp[-\alpha(\gamma - \gamma_y)])]$$
$$+ \left[\tau_y + \frac{G_1G_0}{G_0 + G_1}(\gamma - \gamma_y)\right]\exp[-\alpha(\gamma - \gamma_y)]; \qquad \tau_y < \tau < \tau_{\text{ult}} \quad (1)$$

where

$$\gamma_y = \tau_y/G_0, \quad (2)$$

$$\alpha = G_0/\bar{\eta}R, \quad (3)$$

and

$$\bar{\eta} = G_0\eta/(G_0 + G_1) \quad (4)$$

The material constants, including the viscosity coefficient, η, appearing in Eqs. (1) can be evaluated from constant strain rate data. The coefficient of viscosity is a function of a number of environmental parameters such as temperature and rate, and process variables such as the polymeric molecular weight. Nielsen[11] suggests the use of the equation

$$\log \eta = \log K + n \log M \quad (5)$$

for relating the viscosity coefficient to the molecular weight M. This relation was derived empirically by Fox and Flory[12] based on viscosity-temperature experiments with polyisobutylene fractions. The constant K of Eq. (5) is dependent on the solvent and temperature, and n was found to be equal to 3.4 for polyisobutylene samples with number average molecular weights above 17,000. Nielsen suggests that Eq. (5) is valid for polymeric materials having molecular weights above the critical molecular weight. The critical molecular weight is defined as the value above which the polymer chains become long enough for chain entanglements to form. Apparently sufficient information on the chemical structure of a material is necessary to evaluate the constant K.

When the effects of rate on stress strain behavior are not strong, the use of a nonlinear elastic-plastic model may be adequate. One such model is the Ramberg–Osgood model which is based on Hencky's stress strain relations. This theory assumes the plastic strains to be functions of the current state of stress and independent of the loading history. In one dimension the Ramberg–Osgood relation has the form:

$$\gamma = \frac{\tau}{G} + D\tau^m \tag{6}$$

where G is the elastic shear modulus, and D and m are material constants. The constant m denotes the plastic flow, and consequently the work-hardening characteristics. For the present investigation, observations revealed that separation of the linear and nonlinear terms of Eq. (6) into the form

$$\gamma = \tau/G \qquad (\tau \leqslant \tau_y) \tag{7}$$

$$\gamma = D\tau^m \qquad (\tau > \tau_y) \tag{8}$$

would provide a more adequate representation of the observed stress strain behavior. It should be noted that this model can be modified to account for rate effects. For example, Brinson and Renieri used a rate dependent version of the Ramberg–Osgood model to characterize Metlbond 1113 and 1113-2 thermosetting (epoxy) adhesives in the bulk form.[13]

The dependence of ultimate shear stress on molecular weight can be described with the use of an empirical relation of the form

$$\tau = u - \frac{v}{M} \tag{9}$$

where u and v are material constants. Equation (9) predicts increasing strength with increasing molecular weight, as the strength approaches an asymptotic level at higher molecular weights. The mathematical form of Eq. (9) is based on the assumption that an increase in the amount of end groups act as imperfections which slow down and eventually halt the increase in strength with increasing molecular weight.[11] In fact, in adhesively bonded joints, further increases in molecular weight can cause reductions in strength due to interfacial failures resulting from poor wetting and low adhesion forces.[14] It should be noted, however, that Eq. (9) does not predict this behavior of decreasing strength, and a second equation is needed to describe it.

Delayed failure of an adhesive can be described with the use of Crochet's creep–rupture equation.[15] This equation can be interpreted[8] to apply to pure shear in the form:

$$\tau = A + B\exp(-CX_s) \tag{10}$$

with

$$X_s = \gamma_{12}^V - \gamma_{12}^E \tag{11}$$

where γ_{12}^V and γ_{12}^E refer to the shear strains, and A, B, and C are material constants. Since delayed failure occurs only in elements loaded up into the viscoelastic region, an expression for X_s can be obtained by subtracting the elastic shear strain from the proposed model's creep equation. Solution of Eq. (10) with Chase–Goldsmith model's creep relation results in:

$$\begin{aligned}\tau = A + B\exp\Bigg[-C\bigg[\frac{\tau_y}{G_1}\bigg(\exp\bigg[-\frac{G_1}{\eta}t\bigg]-1\bigg)\\ +\frac{\tau}{G_\infty}\bigg(1-\exp\bigg[-\frac{G_1}{\eta}t\bigg]\bigg)+\frac{\tau}{G_0}\bigg(\exp\bigg[-\frac{G_1}{\eta}t\bigg]-1\bigg)\bigg]\Bigg].\end{aligned} \tag{12}$$

In Eq. (12) the exponential terms contain viscosity coefficients as viscoelastic models are used. In this case the viscosity affects the failure time of the material. If the material constant C of Eq. (12) were assumed to be a constant for all degrees of end capping, increasing the viscosity would predict a longer time to failure for a given stress level. It should also be noted that the asymptotic value of Eq. (12) represent the maximum safe creep stress levels below which delayed failures are not expected to occur.

When it is desired to describe the constant strain rate stress strain behavior of an adhesive with a nonlinear elastic model (this procedure may be acceptable when the effects of rate are negligible), the simplest model creep relation can be utilized for the evaluation of X_s (Eq. (10)). Following the procedure for the evaluation of X_s in conjunction with the Maxwell model creep relation results in a solution to Crochet's creep–rupture equation of the form:

$$\tau = A + B\exp\left[-C\tau\frac{t}{\eta}\right]. \tag{13}$$

This procedure, however, should be applied only as a first approximation for practical purposes.

EXPERIMENTAL PROCEDURES

Materials and specimen features

The preparation and bonding of Thermoplastic Polyimidesulfone has been described in detail by St. Clair.[1,2] A polyimide acid solution is formed by mixing 0.0569 lb (25.8 gm) of 3,3′,4,4′-benzophenonetetracarboxylic dianhydride (BTDA) in a solution made of 0.0439 lb (19.0 gm) of 3,3′-diaminodiphenylsulfone (3,3′-DDS) and 0.5701 lb (258.6 gm) of bis (2-methoxyethyl) ether. End capping is accomplished through the addition of a phthalic anhydride in concentrations varying from zero to three percent. The acid solution is then brushed onto 112-E-A100 glass carrier cloth. The cloth acts as a control for bondline thickness and as a carrier for the adhesive. The scrim is then heated to 212°F (100°C) for one half hour, 302°F (150°C) for one half hour, and to 392°F (200°C) for one half hour. This heating process removes the solvent and the water from the adhesive.

Titanium (6-AL,4-V) strips are grit blasted with 120 mesh aluminum oxide and treated with Pasa Jell 107. A primer coating of the polyimide acid is brushed onto titanium and heated to 212°F (100°C) for one hour and 392°F (200°C) for one hour. Lap shear specimens are then formed by sandwiching the scrim between the titanium adherends using a 0.5 inch (1.27 cm) overlap. The specimens are heated at 41°F/min (5°C/min) to 392°F (200°C); a 200 psi (1.38 MPa) clamping force is applied at this point. The specimens are then heated further to 635°F (335°C), held for 5 minutes, then allowed to cool to 302°F (150°C) before removing the bonding clamps. The glass transition temperature of Thermoplastic Polyimidesulfone is 552°F (272°C).

All test specimens were prepared at NASA-Langley Research Center, and are in accordance with ASTM D1002 specifications (Figure 2).

Molecular weights were determined with a Brice Pheonix light scattering photometer at Arro Laboratories, Illinois. Information

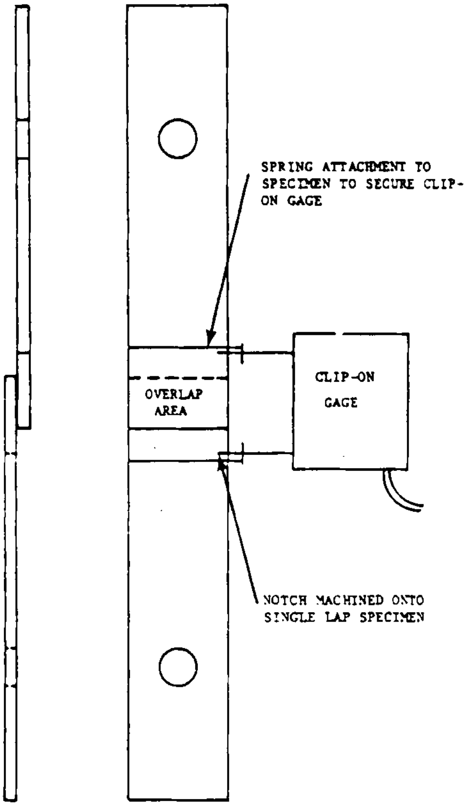


FIGURE 2 Clip-on gage attachment to the single lap specimen for adhesive deformation measurement.

on the number average molecular weight values could not be obtained as the solvent for this process rendered the osmometer inoperable. The molecular weight values reported by NASA are shown in Table I. It should be noted that the molecular weight values listed in Table I actually represent average values of molecular weight bands found for each percent end capping. It is possible (especially for the zero and one percent end cap samples) that specimens with different end cap values may actually have approximately equal molecular weight values as molecular weight bands may overlap for a given set of specimens.

TABLE I
Molecular weight values of zero, one, and three percent end capped Thermoplastic Polyimidesulfone

Percent end cap	Molecular weight (amu)
0.0	19,700
1.0	18,800
3.0	17,400

The bondline thicknesses were measured using a microscope with 40× magnification. Twenty-five measurements were taken along each outside edge of the bonds. The bondline length, adherend width, and adherend thicknesses were measured with a Kanon dial caliper.

The dimensions for each specimen were needed to calculate the adhesive shear stresses and strains. The shear stress was assumed to be uniform and was calculated by dividing the applied load by the overlap area. The shear strain was calculated by the bondline elongation divided by the average bondline thickness.

Testing methods

The mechanical testing of the single lap Polyimidesulfone samples was performed at Clarkson University. NASA-Langley Research Center supplied twenty of each one and three percent end capped specimens. Testing on zero percent end capped specimens was previously performed at Clarkson University by Schenck.[6] All testing was done using a Model 1331 Instron servohydraulic materials testing machine.

Each single lap specimen had notches milled 0.99 inches (2.51 cm) apart on the overlap edge of the bond. The notches were 0.01 inches (0.254 cm) deep by 0.01 inches (0.0254 cm) wide and were used for the placement of a high precision extensometer (Figure 2). The output signal from the extensometer was amplified through the Instron's internal amplification system. The signals from the extensometer and the load cell were then recorded on a strip chart recorder. The extensometer was calibrated using a high magnification calibrator.

Output from the extensometer was used to calculate the strain in the adhesive joint. Deformations in the adherends were subtracted from the total deformation values to obtain adhesive deformations.

Creep and constant strain rate tests were performed at temperatures of 70°F (21°C), 250°F (121°C), and 350°F (177°C). Higher temperature testing was performed with an environmental chamber mounted on the testing frame of the Instron. Room temperature constant strain rate tests were conducted with crosshead rates of 0.001, 0.1, and 10.0 inches/minute (0.00254, 0.254, 25.4 cm/min). High temperature constant strain rate tests were run at a crosshead rate of 5.0 inches/minute (12.7 cm/min). Three to five specimens of each percent end capping were tested in creep at each temperature level. The creep tests had an initial crosshead rate of 0.3 inches/minute (0.762 cm/min), and were performed at stress levels above the elastic limit stress found from the constant strain rate tests.

RESULTS AND DISCUSSION

The constant strain rate behavior of zero, one, and three percent end capped Thermoplastic Polyimidesulfone adhesive at room temperature is shown in Figure 3. The experiments displayed are all at comparable initial elastic strain rates. Experiments at 100 and 10,000 times slower rates did not exhibit strong rate dependence based on ultimate stresses and strains. The observed regions of linear elastic behavior followed by regions of visco-plastic behavior suggest that the Chase–Goldsmith model may be used to describe the constant strain rate behavior of Thermoplastic Polyimidesulfone. Behavior predicted by the Chase–Goldsmith model is also shown in Figure 3. The model provides a good fit to the data at all stress levels observed. The coefficients (G_0, G_1, τ_y, $\dot{\gamma}$, and η) used to fit the model at room temperature are given in Table II.

The elastic shear modulus, G, for each end capped form of Thermoplastic Polyimidesulfone was calculated from the experimental data by averaging the slope of the shear stress strain curve between data points in the initial linear elastic region. G was observed to increase as molecular weight increased. The values of G_0 and G_1 used to fit the Chase–Goldsmith model followed the same trend.

The viscosities found in fitting the Chase–Goldsmith model are

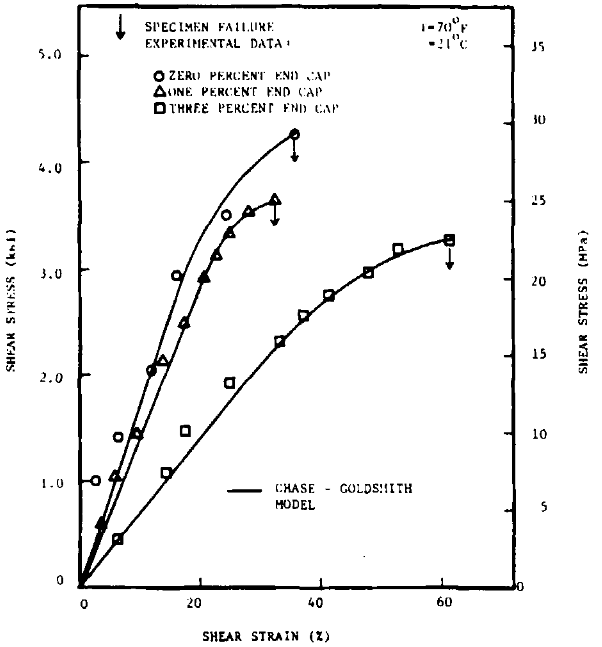


FIGURE 3 Lap shear constant strain rate stress strain behavior of Thermoplastic Polyimidesulfone adhesive film with glass carrier cloth and comparison with viscoelastic theory.

plotted in Figure 4. Equation (5) predicts a much slower increase in viscosity as molecular weight increases. The discrepancy between the two behaviors may partially be attributed to the fact that the adhesive used for experimentation was in the bonded form rather than in the bulk form.

TABLE II
Coefficients used to fit the Chase–Goldsmith model to the constant strain rate behavior of zero, one, and three percent end capped Thermoplastic Polyimidesulfone adhesive at 70°F (21°C) (see Figure 3)

Percent end cap	G_0 ksi (MPa)	G_1 ksi (MPa)	τ_y ksi (MPa)	$\dot{\gamma}$ %/sec	η ksi-sec (MPa-sec)
0	17.1 (117.8)	4.3 (29.6)	3.0 (20.7)	24.6	4.1 (28.2)
1	13.8 (95.1)	1.6 (11.0)	2.9 (20.0)	38.5	2.1 (14.5)
3	7.0 (48.2)	1.2 (8.3)	2.3 (15.8)	125.0	0.8 (5.5)

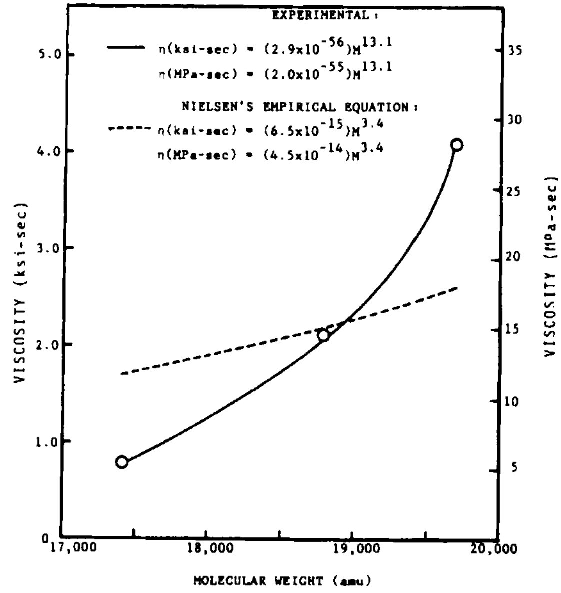


FIGURE 4 Variation of viscosity with molecular weight for Thermoplastic Polyimidesulfone adhesive film with glass carrier cloth tested in the single lap shear mode.

The constant strain rate behaviors of zero, one, and three percent end capped Thermoplastic Polyimidesulfone at 250°F (121°C) and 350°F (177°C) also revealed regions of linear elastic behavior followed by viscoplastic deformation. The zero percent end capped specimens reached higher rupture strains and lower rupture stresses in comparison to the one percent end capped samples. This can be attributed to Schenck's[6] observation of increasing interfacial failure with increasing temperature for the zero percent end capped specimens. This phenomenon causes the coefficients of the Chase–Goldsmith model to change, thus making it impractical to compare viscosities, shear moduli, etc. for the different molecular weights at elevated temperatures.

The ultimate shear stresses under constant strain rate conditions (Figure 5) show the effects of the high temperature interfacial failures on the zero percent end capped specimens. At room temperature, however, the ultimate shear stresses increase with increasing molecular weight. Equation (9) can be used to predict

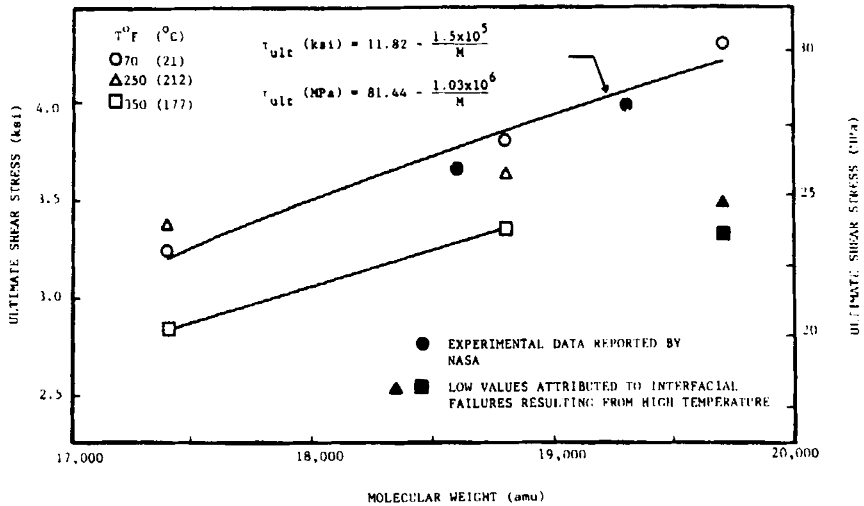


FIGURE 5 Variation of ultimate lap shear stress with molecular weight for Thermoplastic Polyimidesulfone adhesive film with glass carrier cloth.

the variation of ultimate shear stress levels with molecular weight at room temperature. As the molecular weight levels examined fall in a narrow range, Eq. (9), in that range, predicts a near linear response of increasing ultimate stress values with molecular weight (Figure 5). It should be noted that these data represent the maximum ultimate shear stress values even though other (lower) values were also obtained at different test rates. The ultimate shear stress values reported by NASA for one half and two percent end capped Thermoplastic Polyimidesulfone specimens at room temperature also agree with the prediction of Eq. (9). A similar behavior between the ultimate shear stress and molecular weight is observed at 350°F (177°C). These data, however, represent results obtained using only a single crosshead rate.

The constant strain rate behavior of Thermoplastic Polyimidesulfone can also be predicted using a nonlinear elastic approach. In Figure 6 it can be observed that by assuming linear elastic behavior up to the elastic limit stress, and by using a nonlinear power law equation past this limit, the constant strain rate behavior of zero, one, and three percent end capped specimens can be predicted adequately. This application was satisfactory at all temperatures

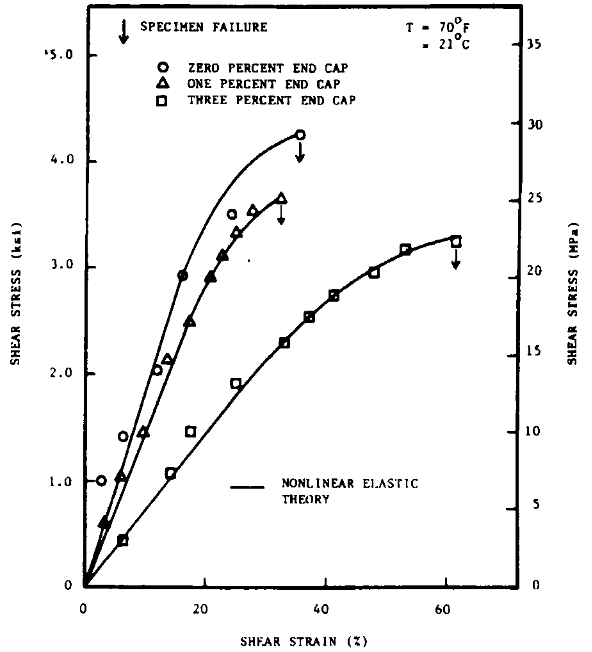


FIGURE 6 Lap shear constant strain rate stress strain behavior of Thermoplastic Polyimidesulfone adhesive film with glass carrier cloth and comparison with nonlinear elastic theory.

tested. The coefficient, m, of the nonlinear relation used (Eq. (8)) is determined from the experimental data (Table III). The material constant m is a relative measure of the amount of work-hardening that the adhesive can withstand before failure occurs. Low values of m indicate soft, ductile materials, and high values of m indicate hard brittle materials. Examination of data (Table III) indicates that room temperature m decreased by 22.5% between zero and one percent end capping, and by 8.0% between one and three percent end capping. As can be seen in Figure 6, toughness decreases considerably when the molecular weight is increased from 17,400 amu to 19,700 amu. This trend, however, was not pronounced at elevated temperatures.

The maximum safe stress levels under creep conditions at room temperature, 250°F (121°C) and 350°F (177°C) are shown in Figure 7. At room temperature there is an increase of approximately 17%

TABLE III

Coefficients used to fit the nonlinear relation (Eqs. (7) and (8)) to the constant strain rate stress strain behavior of Thermoplastic Polyimidesulfone adhesive (see Figure 6)

Percent end cap	T °F (°C)	τ_y ksi (MPa)	G ksi (MPa)	D ksi (MPa)	m
0	70 (21)	3.0 (20.7)	18.1 (124.7)	0.010 (0.069)	2.44
1	70 (21)	2.9 (20.0)	14.3 (98.5)	0.027 (0.186)	1.89
3	70 (21)	2.3 (15.8)	7.3 (50.3)	0.073 (0.503)	1.74
0	250 (121)	2.3 (15.8)	24.2 (166.7)	0.016 (0.110)	2.16
1	250 (121)	3.0 (20.7)	20.7 (142.6)	0.015 (0.103)	2.01
3	250 (121)	2.7 (18.6)	16.5 (113.7)	0.018 (0.124)	2.18
0	350 (177)	2.0 (13.8)	13.3 (91.6)	0.031 (0.214)	2.09
1	350 (177)	2.9 (20.0)	12.2 (84.1)	0.034 (0.234)	1.80
3	350 (177)	1.9 (13.1)	9.6 (66.1)	0.062 (0.427)	1.72

as the molecular weight increases by 8%. The safe creep stress level remains relatively constant when the molecular weight is increased by another 4.8%. The increase is expected due to the increasing chain entanglement and increasing degree of intermolecular forces. The nearly constant values of maximum creep stress, between one and zero percent end capping, on the other hand, may be caused by

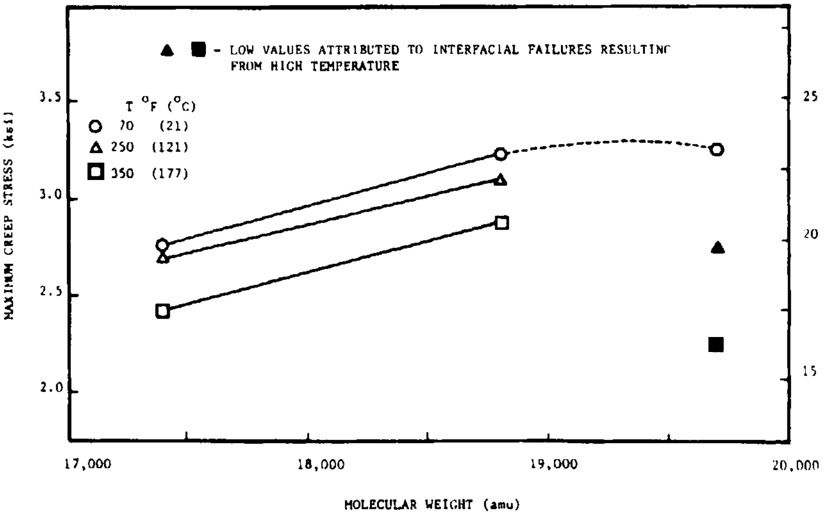
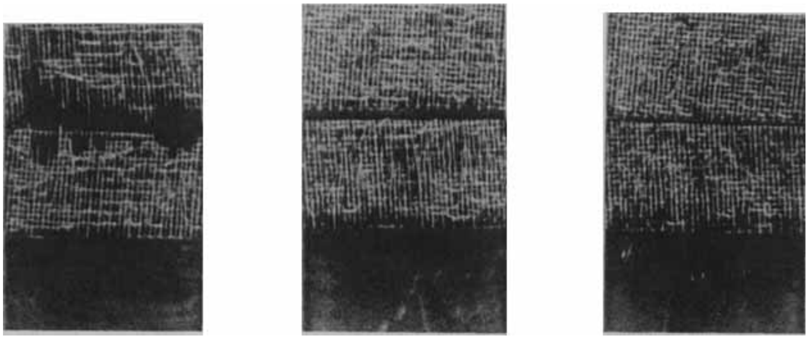


FIGURE 7 Variation of maximum (safe) lap shear creep stress with molecular weight for Thermoplastic Polyimidesulfone adhesive film with glass carrier cloth.

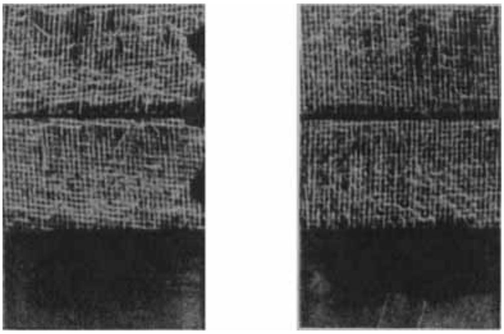
decreasing adhesion as the viscosity increases with molecular weight beyond the point where optimum interfacial bond formation is no longer possible. There is also the possibility that the zero and one percent end capped specimens used are of nearly equal molecular weights as accurate molecular weight control could not be obtained for zero and one percent end capped specimens. The safe creep stress levels are also observed to increase with molecular weight at higher temperatures for the one and three percent end capped specimens. The significant decreases in the safe creep stress levels at zero percent end cap value under elevated temperatures is attributed to the high temperature interfacial failures as discussed previously. It may be assumed that Eq. (9) describes the dependence of safe creep stress levels on molecular weight, but data from the zero percent end capped specimens prohibit an accurate fit to Eq. (9). Data from the zero percent end capped specimens also prohibit determination of a shift factor describing the temperature dependence of the safe creep stress values.

The typical failure surfaces of one percent end capped Thermoplastic Polyimidesulfone at 70°F (21°C), 250°F (121°C) and 350°F (177°C) are shown in Figure 8. Observations reveal some adhesive/adherend interfacial failure at room temperature and at 250°F (121°C). The interfacial failure was common at room temperature, and appeared to decrease in severity with increasing temperature. This indicates the presence of a strong cohesive matrix, with failure initiating at the adhesive/adherend interface due to the relatively high viscosity. The viscosity may have hindered bond formation at the adhesive/adherend interface enough to cause the interface to be the weak link in the system. The decrease of interfacial failure with increases in temperature would be expected as molecular activity increases at higher temperatures causing a limited increase in adhesive forces. Consequently, fracture initiates at points other than the adhesive/adherend interface. In fact, experimental data reveal that adhesive/adherend interfacial failures were uncommon at 350°F (177°C) test temperature for the one percent end capped samples.

Figure 9 shows the typical failure surfaces of three percent end capped Thermoplastic Polyimidesulfone specimens at room and elevated temperatures. Adhesive/adherend interfacial failure was not common to the three percent end capped specimens. Better



T = 70°F (21°C) T = 70°F (21°C) T = 250°F (121°C)

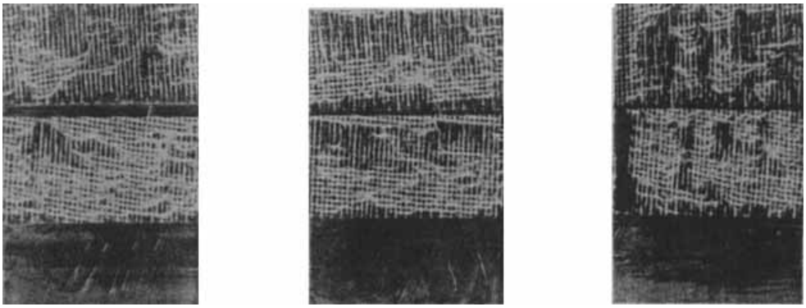


T = 250°F (121°C) T = 350°F (177°C)

FIGURE 8 Typical failure surfaces of one percent end capped Thermoplastic Polyimidesulfone/titanium specimens with glass carrier cloth.

bond formation between the adhesive and adherend may have occurred in comparison to the one percent end capped samples. The lower viscosity of the three percent end capped form of the adhesive would permit better wetting and diffusion during bonding, resulting in a stronger adhesive bond between the Thermoplastic Polyimidesulfone and the titanium adherend. The causes of lower strength in the three percent end capped form may be revealed by comparison

T = 70°F (21°C) T = 70°F (21°C) T = 250°F (121°C)



T = 250°F (121°C) T = 350°F (177°C) T = 350°F (177°C)

FIGURE 9 Typical failure surfaces of three percent end capped Thermoplastic Polyimidesulfone/titanium specimens with glass carrier cloth.

of the failure surfaces shown in Figures 8 (one percent end capping) and 9 (three percent end capping). The one percent end capped specimens appear to form a strong, consistent cohesive matrix, whereas the three percent end capped specimens exhibit some form of cohesive slippage. The carrier cloth exposed by failure of the three percent end capped specimens is generally displaced by slippage along the axis in which the forces were applied. It appears that the molecular chains within the three percent end capped specimens reorient or slip when a load is applied to the adhesive joint. The load must then be carried by the carrier cloth. Since the strands of the carrier cloth are not joined in a design to accommodate stress, the strands affected by the applied load are free to slide. This phenomenon may occur in the three percent end capped specimens because the chain entanglement and viscosity are sufficiently low to allow molecular chains to slip past each other at high stress levels.

CONCLUSIONS

The effects of molecular weight on the shear creep and constant strain rate behavior of Thermoplastic Polyimidesulfone adhesive was studied in the bonded form. For this purpose, constant strain rate, creep, and creep–rupture experiments were performed on zero, one, and three percent end capped specimens of the adhesive.

Mathematical relations were proposed to represent the constant strain rate and delayed failure viscoelastic–plastic behavior of the model adhesive. Based on the mathematical relations, used in conjunction with experimental results, several conclusions can be derived:

- Room temperature constant strain rate results show increases in viscosity with increasing molecular weight. It should be noted, however, that these viscosity coefficient values are calculated parameters obtained on the basis of the Chase–Goldsmith model and they do not represent values measured using standard viscosity measurement techniques;
- Nonlinear relations reveal increasing strain-hardening capacity with increasing molecular weight;
- At all temperature levels of experimentation, ultimate shear stress levels and safe creep stress levels increased with increasing molecular weight. The cases for which the stress levels are the same for zero and one percent end capped specimens are attributed to the possibility of overlapping molecular weight ranges for the given percentages of end capping.
- Reductions in molecular weight revealed increased adhesion (less interfacial failure), but also reduced the cohesive strength of the

model adhesive. In general, the results of this investigation imply that reductions in the molecular weight of Thermoplastic Polyimidesulfone adversely affect the mechanical properties studied. Reductions in molecular weight may increase wetting and diffusion during the bonding process, but these effects are insufficient to override the advantages of increased cohesive strength obtained at higher molecular weights studied.

The authors' recommendations for future studies are to examine different end capped forms of Thermoplastic Polyimidesulfone adhesive in the bulk form. Any geometric and material nonlinearities which may be caused by the use of the bonded form can be avoided in the bulk form. Such a study would also permit the molecular weight effects on the adhesive properties to be evaluated more explicitly, since these effects could then be separated into cohesive and adhesive categories. Also, it may be advantageous to evaluate the mechanical properties of both bonded and bulk forms of the adhesive for a wider molecular weight range thereby permitting a more complete characterization of the adhesive.

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