

**RATE AND TEMPERATURE DEPENDENT MECHANICAL
PROPERTIES OF A FLEXIBLE ADHESIVE**

by

Bruce Duncan, George Hinopoulos, Keith Ogilvy-Robb and Elena Arranz

Project PAJex2 - Flexible Adhesives

PAJex2 Report No 1

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**Performance of Adhesive Joints Programme
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(milestone T1/M3)**

Summary

Design stress analyses of adhesively bonded structures may be performed using Finite Element methods provided that suitable material models and mechanical properties data are available. Flexible adhesives appear to be best modelled by hyperelastic models. However, their properties are dependent on strain rate and temperature. The aim of this work is to characterise the behaviour of a typical flexible adhesive.

Statistical analysis of bulk specimen tensile and single-lap joint shear tests shows that stress at failure increases with decreasing temperature and increasing strain rate. Strain-to-failure shows less significant dependence. Within the hyperelastic temperature region and at strain rates up to 10^{-1}s^{-1} , the statistical analyses show that temperature has a higher influence than strain rate on the material properties. There is a reasonable correlation between failure stress values in tensile and single-lap joint tests. The mechanical properties have been investigated using visco-elastic models. Rate dependence of the adhesive stress-strain response can be described using a relaxation model. Time-temperature superposition shows some promise for modelling the failure of the adhesive specimens.

This work has shown that there is no single property that will describe the mechanical or failure properties of the flexible adhesive studied since these properties vary with temperature and strain rate. Appreciation of this is vital when load bearing joints are designed so that all possible service conditions can be considered. However, these findings indicate that there are methods that can model the rate and temperature behaviour of the adhesive and of the bonded joint. These would enable interpolation of properties to conditions not measured and, potentially, allow extrapolation to conditions beyond the range measured.

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National Physical Laboratory
Teddington, Middlesex, UK, TW11 0LW

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Approved on behalf of Managing Director, NPL, by Dr C Lea,
Head of Centre for Materials Measurement and Technology

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1. INTRODUCTION

Accurate Finite Element (FE) modelling of the performance of adhesively bonded structures is hindered by the lack of accurate material properties data and uncertainties over the most appropriate material models. The Performance of Adhesive Joints programme aims to assist in the design process through the identification of valid material models and development of test methods for obtaining the input data required. Previous work^(1, 2) has identified that flexible, elastomeric adhesives, with low moduli and large strains to failure, are better represented by hyperelastic models developed for rubbers than by the elastic-plastic models used for structural adhesives (e.g. epoxies).

The objective of the current work is to continue the development of the hyperelastic approach for characterising flexible adhesives. It is recognised that the properties of the flexible adhesives will be influenced by strain rate and temperature. This report discusses the rate and temperature dependence of a typical elastomeric adhesive. Dynamic measurement techniques such as Dynamic Mechanical Thermal Analysis (DMTA)⁽³⁾ or oscillatory rheometry⁽⁴⁾ can be used to characterise the frequency and temperature dependence of the low strain properties of polymeric materials. However, they do not obtain the large strain properties required for design. Conventional mechanical tests are required for obtaining these properties. In this work, uniaxial tension and lap joint shear tests have been carried out at different strain rates and temperatures.

Since flexible adhesives are visco-elastic material, one aim of this work was to investigate the application of visco-elasticity concepts such as time-temperature superposition to the measured data. Statistical analysis methods have been used to investigate the dependence of material properties and potential failure criteria on temperature and strain rate.

2.1 EXPERIMENTAL

2.1 SAMPLE PREPARATION

The adhesive used in this study was a 1-part polybutadiene elastomer M70 supplied by Evode Ltd. Bulk tensile test specimens were prepared for obtaining material properties and lap joint specimens for assessing rate and temperature dependence of the adhesive performance.

2.1.1 Bulk Tensile Specimens

Bulk adhesive specimens were used for generating material properties data for hyperelastic models⁽¹⁾. Sheets of bulk adhesive were cast^(5, 6) onto flat metal plates covered with a PTFE release film. The adhesive was applied with a spatula and scraped to an even layer thickness taking care to remove any visible large voids. A sheet of PTFE film was placed over the top of the cast adhesive and pressed on using a roller to squeeze out any entrapped air. A second metal plate was then pressed down on top of this sheet. Strips of plastic (either 0.5 mm or 1.0 mm thick) were inserted between the plates to control specimen thickness. The sheet was then cured in an oven (pre-heated to 200 °C) for 45 minutes. Thermocouples embedded in the adhesive monitored the temperature of the adhesive during cure.

Half sized ISO 3167⁽⁷⁾ multi-purpose test specimens (90 mm long, 10 mm wide tabs, 5 mm wide by 30 mm long gauge section, as shown in Figure 1) were punched from bulk sheets using a shaped cutter. This is the most efficient method for preparing specimens from such thin flexible

sheets. However, it is not suitable for stiffer structural adhesives that are more brittle. Cured sheets and test specimens were stored in a dessicator prior to testing to prevent adsorption of moisture.

2.1.2 Lap Joint Specimens

The adherends for the lap joint specimens were 1.5 mm thick mild steel strips 100 mm x 25 mm. The areas to be bonded were prepared in four stages:

1. degreased by acetone wipe;
2. grit blasted using 80/120 alumina;
3. cleaned in acetone in an ultrasonic bath for 5 minutes; and,
4. final degreasing using 1,1,1-trichloroethane wipe.

(Trichloroethane is a hazardous chemical and must be used with the appropriate COSHH controls to prevent operator exposure. Therefore, general use of trichloroethane in surface preparation is not recommended. Trichloroethane was used in this study to maintain continuity with previous studies.)

The adherends were bonded with a 12.5 mm long overlap. The bondline thickness was controlled by mixing 1% by weight 250 μm ballotini spheres into the adhesive. As shown in Figure 1, tabs (25 mm long cut from the same steel sheet) were bonded onto the ends of the adherends (using a 1 part epoxy AV119 supplied by Ciba Speciality Chemicals). These were to ensure that the direction of applied force was through the centre line of the grip assembly. Excess adhesive was wiped off the joints before cure and any remaining fillets were removed after cure.

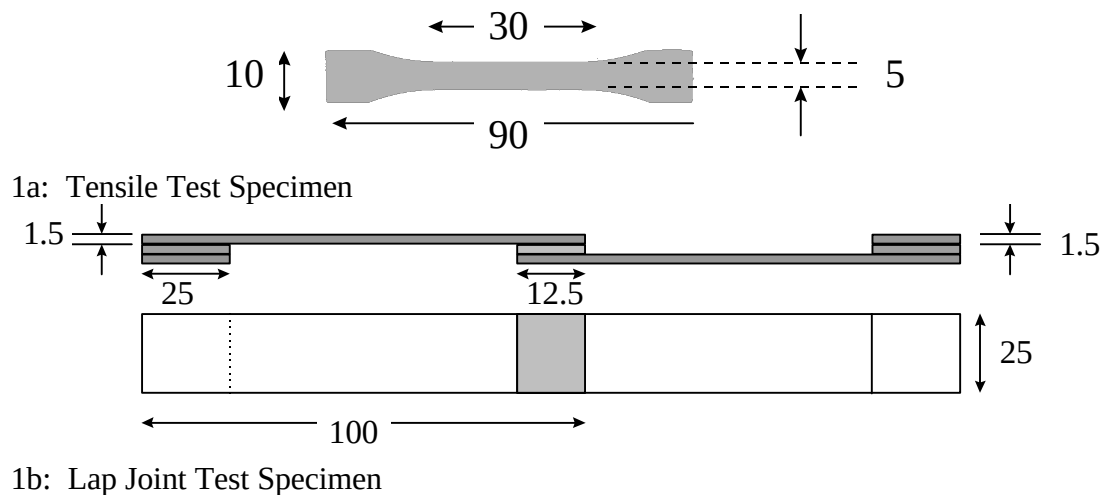


Figure 1: Test Specimens

Specimens were bonded and cured in a special jig to maintain alignment and bondline thickness. Since the mass of this jig differed from that of the plates used in the preparation of bulk specimens the adhesive heated at a slower rate than in the bulk specimens⁽⁸⁾. Various cure temperatures were tested. The rates of temperature increase in the adhesive layer were monitored using a thermocouple attached to the adherend close to the bond line. This study suggested that an oven temperature of 215 °C produced a thermal history similar to the bulk

adhesive. Glass transition temperatures (T_g) of adhesive cured in the lap joint configuration measured by DMTA were around -30 °C. These were similar to the bulk adhesive values.

2.2 TENSILE TESTS

Tensile tests were carried out at four speeds (1, 10, 100 and 250 mm/min) and five temperatures (-40, 0, 20, 40 and 80 °C) using Instron 4505 series test machines. The four speeds selected gave approximate strain rates of $3 \times 10^{-4} \text{ s}^{-1}$, $3 \times 10^{-3} \text{ s}^{-1}$, $3 \times 10^{-2} \text{ s}^{-1}$ and $8 \times 10^{-2} \text{ s}^{-1}$ in the gauge section of the specimen. Plots of measured strain against crosshead movement were linear until the specimens failed. Slopes calculated by regression were approximately 0.018 strain per mm. This correction factor was independent of temperature and strain rate.

Tests at 0, 20 and 40 °C were performed on a test machine fitted with a Climatic Systems temperature chamber and remote temperature conditioning unit that provide an extremely stable test temperature over the range -10 °C to 70 °C. In these tests, tensile and transverse strains were measured using a Messphysik video extensometer.

The low (-40 °C) and high (80 °C) temperature tests were performed on a different test machine fitted with an Instron temperature chamber. This chamber is able to sustain a wide range of temperatures. However, temperature stability is not as good near room temperature as the other chamber owing to the use of liquid nitrogen as the cooling agent. Tensile strains in these tests were measured using an Instron video extensometer. The accuracy of strain measurement appears to be relatively poor owing to the low strains to failure. Generally, video extensometry is less accurate in the low strain region^(6, 9). Bending of the specimen in the test was an additional problem. Specimens were conditioned at low temperature for at least 30 minutes before testing. When the specimen was being positioned in the grips it heated and became flexible. The specimen became more rigid as it cooled, thereby 'freezing-in' any distortion. There were also problems imaging the specimen as:

- frost forming on the window of the temperature chamber, blurring the image;
- frost forming on the sample surface, reducing the contrast of the gauge marks; and
- some mist occurring in the chamber when liquid nitrogen is injected.

Higher rate tests⁽¹⁰⁾ were performed at room temperature (22 °C) using an Instron 1343 servo-hydraulic test machine. The speed settings used were 4.17, 41.7, 417 and 4170 mm/s. The slowest speed (4.17 mm/s) is equal to the highest speed used on the 4505 machines (250 mm/min). The test machine was unable to reach the highest speed setting although speeds in excess of 3.5 m/s were achieved. Strains were estimated from the movement of the test machine crosshead using a correction factor of 0.018 strain/mm. The low stiffness of the test specimens meant that the compliance of the test machine would have negligible influence on the results.

2.3 LAP JOINT TESTS

The lap joints were tested on an Instron 4507 test machine fitted with an Instron temperature chamber. The shear extension was measured using a pair of 25 mm gauge length Instron type 2602 extensometers attached straddling the bondline. Specimens were tested at the same temperatures as the tensile specimens (-40, 0, 20, 40 and 80 °C). Test speeds were selected,

depending on the thickness of the adhesive layer, in order to obtain shear strain rates in the bondline that were approximately the same as the strain rates in tensile tests.

There was a fair degree of variability in the bondline thickness measured for the bonded joints. This may reflect uncertainties in the measurement of a thin bondline through subtracting the adherend thickness from the total thickness of the bonded joint. However, it may also reflect an uneven distribution of the ballotini beads in the adhesive. The adhesive was a thick paste which made it difficult to mix in the ballotini. Specimens that seemed to have an uneven bondline thickness were excluded from the main test programme.

3. RESULTS

Owing to the scatter in the stress-strain curves measured in the tests all the stress-strain response results for the tensile and lap joint tests are presented as averages of the curves measured. The stress values measured from all the tests performed at a temperature and strain rate were averaged at selected strains in order to produce the average stress-strain curve. A minimum of 5 tests was performed at each test condition. These averaged sets of data will be used as the input data for FE stress analyses in later parts of the project.

3.1 TENSILE TEST DATA

3.1.1 Low Temperature (-40 °C) Data

The lowest test temperature was below the glass transition temperature of the M70 adhesive. Therefore, the material is likely to be glassy. Therefore, it is unlikely to be modelled by hyperelasticity. Stress-strain curves measured in tension are shown in Figures 2a - 2d. Given the low strains to failure very few of the strain values chosen to provide average curves for the rest of the temperatures would be relevant. Hence, the curves are presented unaveraged. In many tests the strains measured using video extensometry were unreliable and therefore strain data were calculated from the crosshead movement using correction factor of 0.018 strain/mm. Regression of strain against displacement in tests where the measured strains appeared more reliable confirmed the validity of this correction factor.

The strains to failure of the specimens seem to decrease with increasing test speed. At the low speeds the adhesive appears to yield and flow plastically. At higher speeds it tends to fail in the elastic region. These curves suggest that the low temperature response of the M70 adhesive will be best modelled by elastic-plastic models. Further work would be needed to determine which yield model would be most suitable.

The oscillations in the stress values measured in the 1 mm/min tests (and to a lesser extent in the 10 mm/min tests) are real effects, which are due to fluctuations in the specimen temperature during the tests. It was noted earlier, that the stability of the temperature chamber is not perfect. The test temperature will over- and under-shoot as the controller tries to maintain the set temperature. At -40 °C the adhesive is close to its glass transition temperature (ca. -30 °C) and, since the material modulus as measured by DMTA⁽⁵⁾ decreases by a factor of 10 between -40 °C and -30 °C, the material properties will be extremely sensitive to temperature in this region. The small fluctuations of the temperature of the air in the chamber will be mirrored in the specimen temperature since it has a low thermal mass. Therefore, the adhesive will stiffen and soften

cyclically as the temperature changes. These oscillations are not seen at the highest speeds since the test is over in a period very much shorter than the period of the temperature fluctuation.

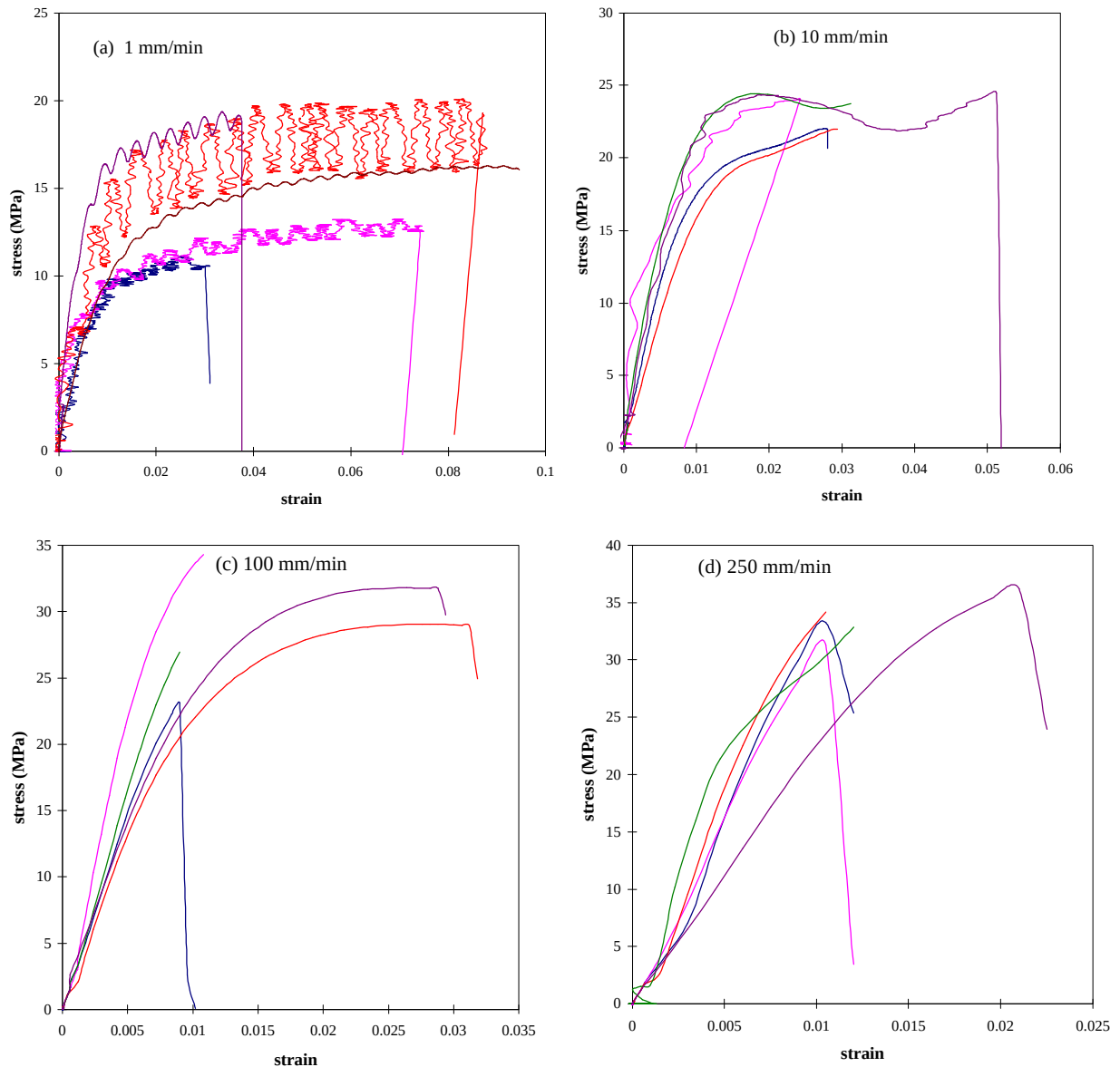


Figure 2: Uniaxial tensile test results of the flexible adhesive at -40 °C

3.1.2 Tensile Data in the Hyperelastic Region

The average of the tensile test curves measured at 0 °C, 20 °C, 40 °C and 80 °C are shown in Figures 3a - 3d. At these temperatures the adhesive is comfortably above T_g . The adhesive will behave hyperelastically. Therefore this temperature range is referred to as the hyperelastic region. Strains to failure are significantly higher than at -40 °C although failure stresses are very much lower. The data determined from the high rate machine are plotted with the 20 °C data from the lower rate tests. The test data show that the material is strain rate sensitive. The strain rate sensitivity diminishes with increasing temperature. Strains to failure also seem to decrease with increasing temperature. There is also some evidence of increasing strains to failure at higher strain rates. Analysis of the specimen failure data is discussed in Sections 3.1.5 and 4.

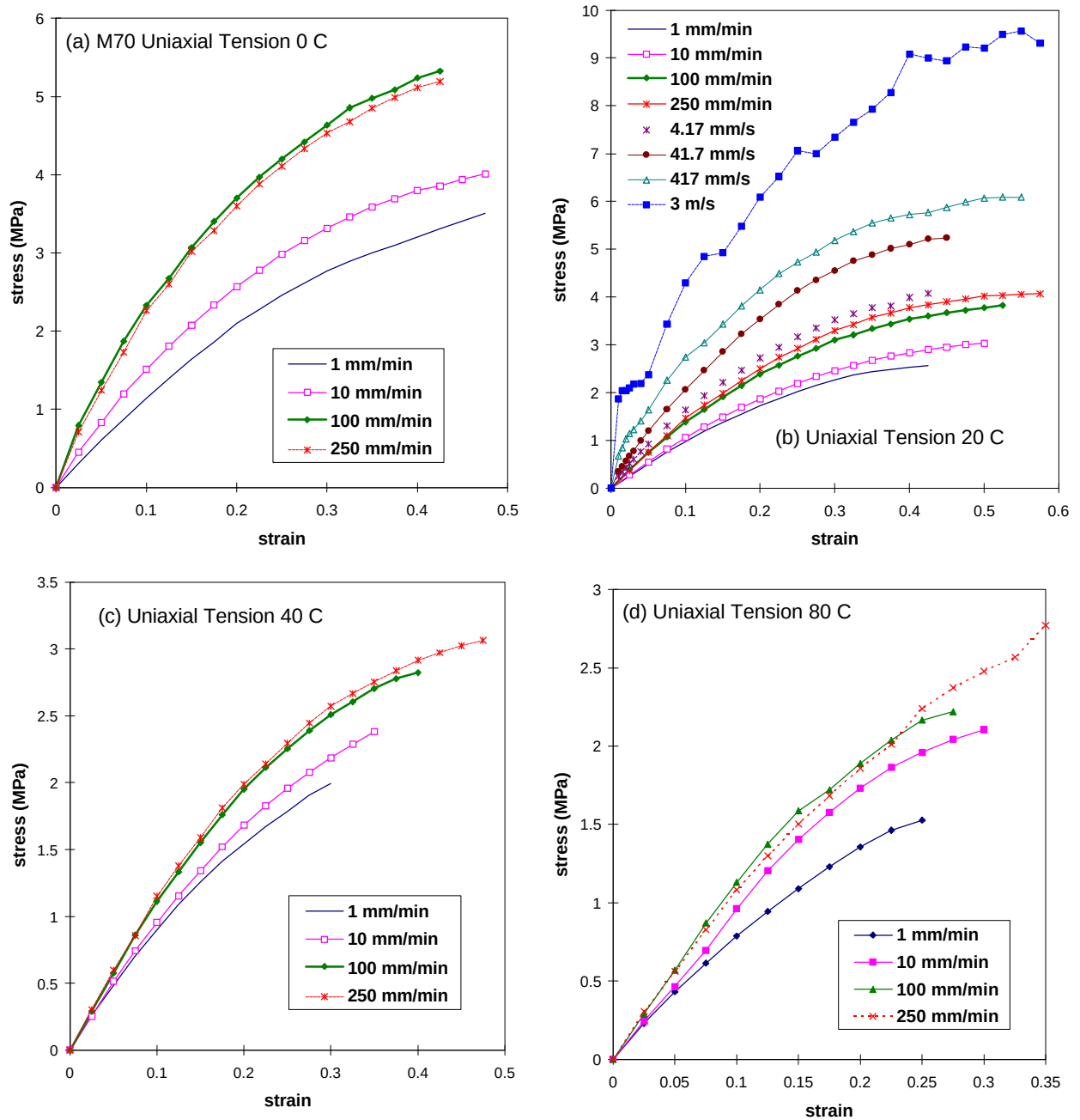


Figure 3: Tensile test data measured in the hyperelastic temperature region (0 °C - 80 °C)

The uniaxial tension data were also analysed to obtain elastic-plastic properties suitable for FE analysis. The data were formatted as true stress-true strain curves. The measured Poisson's ratio of 0.35 (Section 3.1.4) was used to obtain the reduction in specimen cross-section area required to calculate true stress. A typical set of true stress-strain curves obtained at a single temperature is shown in Figure 4. These true stress-true strain curves appear linear. Elastic modulus values were calculated from the regression of the true stress-true strain curves between 0 % and 10 % true strain. These are summarised in Table 1. The modulus depends on temperature and strain rate. The linearity of the true stress-true strain curves suggests that little plastic yielding occurs. Thus, elastic-plastic material models offer little advantage over elastic models in the way of improved fits to the data.

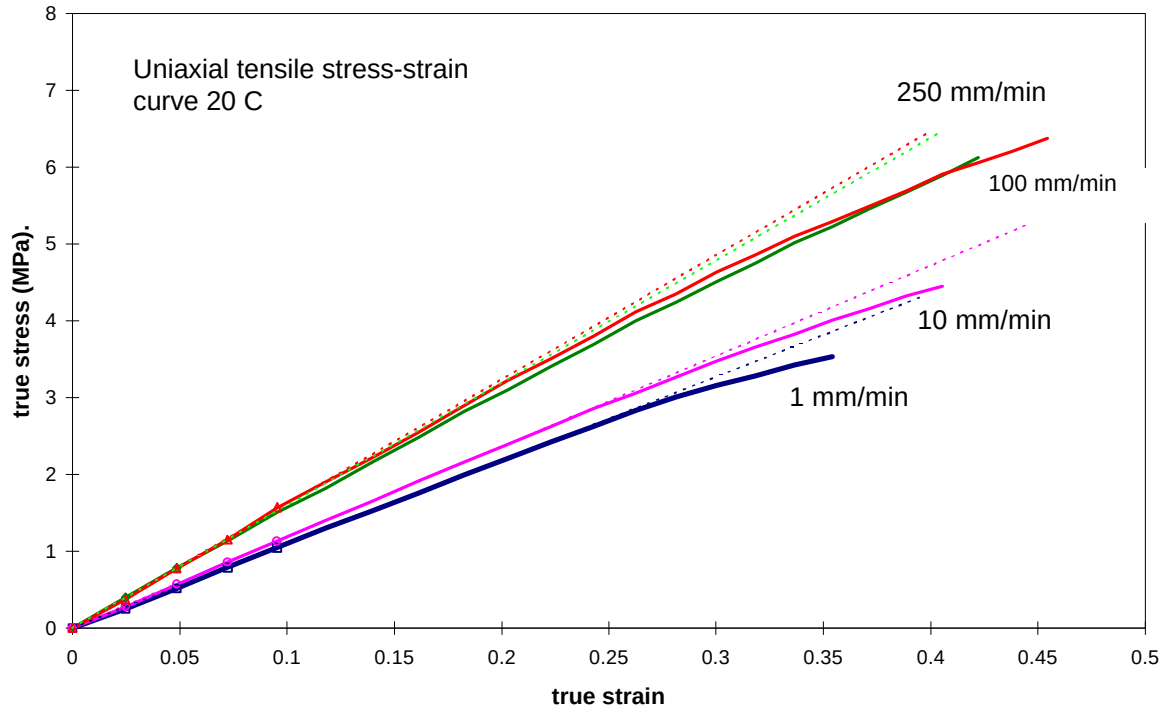


Figure 4: True stress-true strain data obtained in tensile tests

Table 1: Elastic modulus values (MPa) measured in uniaxial tension tests

Temperature (°C)	speed (mm/min)			
	1	10	100	250
0	12.8	17.3	27.1	25.7
20	10.9	11.8	15.7	16.2
40	10.2	10.8	12.5	12.8
80	9.0	10.5	12.6	12.1

3.1.3 Time Dependent Data

Stress relaxation measurements were made in tension at a series of different strains and temperatures. Strains, ϵ , were applied at a speed of 100 mm/min. The extensions were held constant and stress, $\sigma(t)$, monitored over a period of 2 hours. At the highest level of strain used (25 %), some specimens failed during the stress relaxation. The time dependent relaxation moduli, $E(t) = \sigma(t) / \epsilon$, measured at 5 % strain (0 °C, 20 °C and 40 °C) are shown in Figure 5a. The data were fitted adequately by Eqn 1, the visco-elastic function⁽³⁾, as shown in Table 2:

$$E(t) = E_0 + Dt^{-n} \quad (1)$$

E_0 , D and n are relaxation constants obtained by fitting the function to the data. Time dependent data can also be obtained from constant rate tests. Effective time values are obtained by dividing strains by the strain rates. Stress values at a constant strain (10 %) determined from tests at different speeds and temperatures (0 °C, 20 °C, 40 °C and 80 °C) are shown in Figure 5b. Data from the 20 °C tests were fitted using Eqn 1.

Figure 6 shows stress strain curves calculated from Eqn 1 using the sets of relaxation constants determined at 20 °C that are shown in Table 2. Two test speeds (1 mm/min and 0.4 m/s) are shown. The plots indicate that the measured stress-strain curves are not well modelled from the relaxation data. There was a better correlation with the curves determined from time-dependent data from the constant rate tests (at 10% strain). However, measured stress-strain curves still show significantly greater curvatures than the modelled curves. The agreement between the modelled curves and the measured data is significantly better when true stress data, calculated using a Poisson's ratio value $\nu = 0.35$, are used. True stress, where the change in specimen cross-section is incorporated, should give the better agreement since the visco-elastic function models the time-dependence of the modulus. Interestingly the predicted curve from constant rate data is very close to the measured curve if the material is assumed to be incompressible (i.e. $\nu = 0.5$). The differences between the measured and predicted curves at large strains may be due to stress/strain dependence of the relaxation function constants that is not obtained using data at only a single strain.

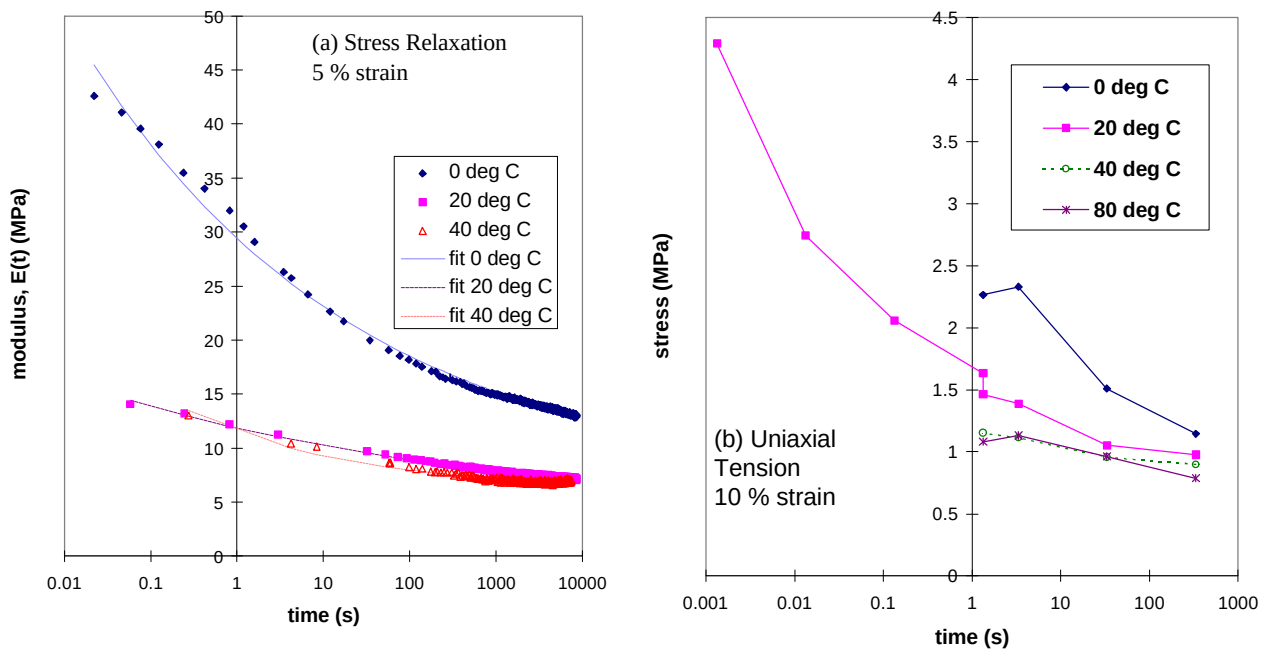


Figure 5: Time dependent material properties obtained from tensile tests

Table 2: Fitted visco-elastic parameters

Test	Fit Parameter			
	E_0	D	n	Goodness of fit (r^2)
0 °C relaxation	6.4070	22.9891	0.139019	0.993761
20 °C relaxation	3.7767	8.0717	0.096894	0.991789
40 °C relaxation	6.0495	5.4873	0.230096	0.898549
20 °C const rate	7.6415	7.9274	0.224163	0.995566

There is relatively poor agreement between the measured stress-strain curves and those predicted from the stress relaxation in comparison to those predicted from constant strain rate tests. This is probably due to uncertainties in the data measured at the start of the relaxation test. The strain takes a finite time to apply. During this period the material is relaxing and, thus, the point where the test strain is reached is not the beginning of the relaxation. Therefore, the

initial point on the relaxation curve is not the instantaneous modulus. It is actually a relaxed modulus at a higher effective time. In contrast, the times at fixed strains in the constant rate tests can be established reliably. The constant rate data measured at 20 °C are compared with the relaxation data in Figure 7. The curve lies between, and is approximately parallel to, the relaxation curves at 0 °C and 20 °C.

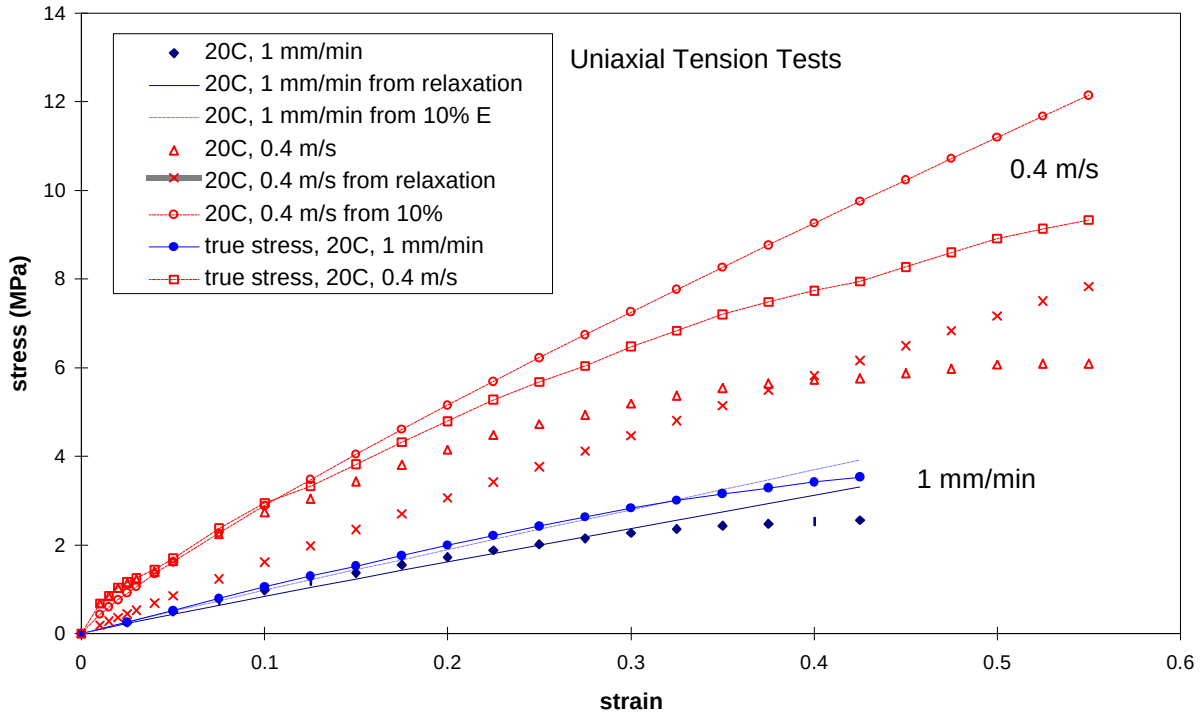


Figure 6: Predictions of constant strain rate tests from time-dependent data

Visco-elasticity theories imply that there are relationships between the time- and temperature-dependent behaviour of polymers. The visco-elastic function (Eqn 1) that earlier fitted the data can be re-written:

$$E(t) = E_0 + A \left(\frac{t}{t} \right)^{-n} \quad (2)$$

The parameter τ defines the position of the curve on the time axis and represents a relaxation time for the mechanism giving rise to the time-dependence of the elastic modulus. When $t \ll \tau$ the material is glassy, when $t \gg \tau$ the material should be rubbery and the glass transition occurs when $t \approx \tau$. The parameter τ increases with decreasing temperature. Thus temperature and rate dependence are linked. The variation of τ with temperature (T) is described⁽³⁾ by the Vogel-Fulcher equation:

$$t = t_0 \exp \frac{\Delta H}{R(T - T_\infty)} \quad (3)$$

where ΔH is the activation energy/mole for the glass-rubber relaxation mechanism, R is the universal gas constant/mole and T_∞ is the temperature (below T_g) where τ approaches an infinite value. If the parameters E_0 , A , n and ΔH are independent of temperature then the horizontal shift factor (a_T) on a log(time) axis between curves at temperatures T_1 and T_2 is:

$$a_T = \ln(t_{e1}) - \ln(t_{e2}) = \frac{\Delta H}{R} \left(\frac{1}{T_1 - T_\infty} - \frac{1}{T_2 - T_\infty} \right) \quad (4)$$

t_{e1} and t_{e2} are arbitrary effective times at which the modulus is the same at temperatures T_1 and T_2 respectively. Thus, if time-dependent modulus curves at different temperatures are parallel on a log(time) axis then a_T is constant. If a_T values from a reference temperature (T_R) are evaluated over a range of temperatures (T) then Eqn 4 can be expressed as the WLF shift function⁽³⁾:

$$a_T = \frac{C_1(T - T_R)}{C_2 + T - T_R}, \quad (5)$$

$$C_1 = \frac{\Delta H}{R(T_R - T_\infty)} \text{ and } C_2 = T_R - T_\infty$$

and the slope of a_T against $(T - T_R)/(T - T_\infty)$ should be linear. Thus, the shift factor can be used to calculate modulus data at any temperature and time/frequency from a reference curve measured over a range of temperatures at a single frequency. This time-temperature superposition approach is only valid around and above the glass transition temperature.

The relaxation and constant rate data are curved when plotted as E or stress against log(time), Figure 6. It is difficult to make out whether there is a constant shift between the curves. These curves become straighter if the data are plotted as $\log(E)$ or $\log(\sigma)$ against $\log(\text{time})$ as shown in Figure 7. This shows that the relaxation curves measured at 0 °C and 20 °C are approximately parallel. However, the 40 °C curve is not parallel to the others. It seems to intersect the 20 °C around 1000 s. Since there are few points and some degree of scatter it is difficult to determine if the $\log(\sigma)$ -log(time) data from constant rate tests are parallel. However, the trends in the 0 °C, 20 °C and 40 °C curves suggest that the curves will intersect after 1000 s.

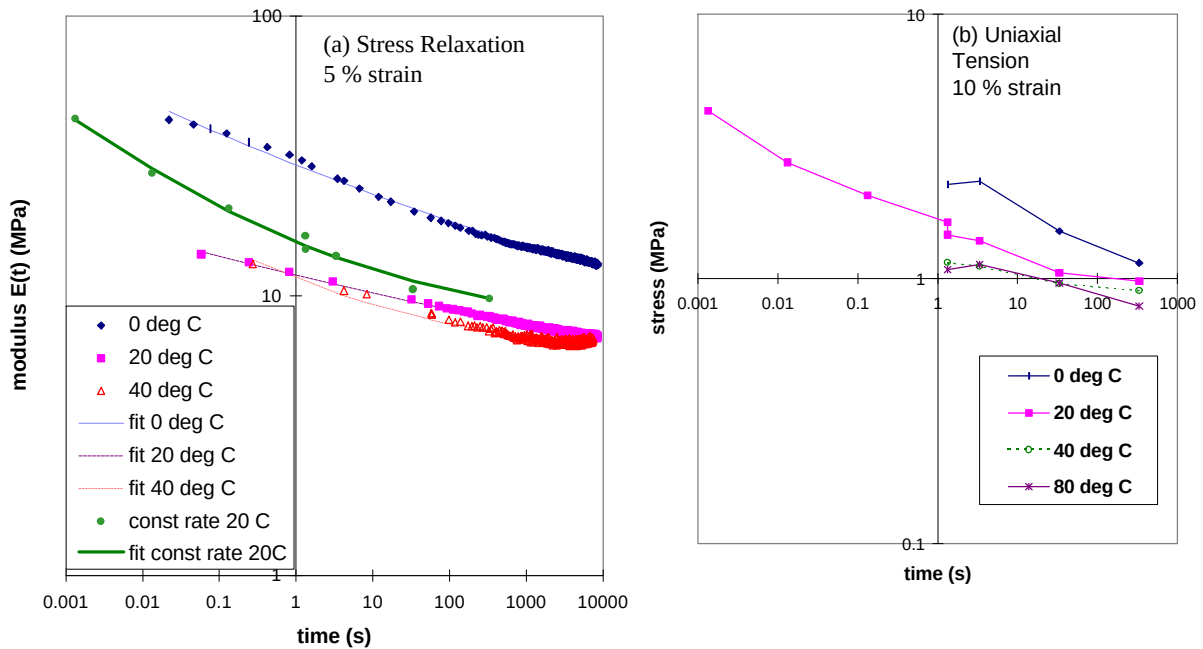


Figure 7: Logarithmic plots of visco-elastic properties for time-temperature superposition

3.1.4 Poisson's Ratio Measurements

Poisson's ratio measurements were made in tests performed at 0 °C, 20 °C and 40 °C at speeds between 1 mm/min and 250 mm/min. The data were averaged and presented in Figure 8. All curves have similar features. The initial Poisson's ratio, ν , is close to 0.4 and then decreases approximately linearly with increasing strain (reaching around 0.3 at strain = 0.4). It appears that Poisson's ratio is lower for the higher test speeds. Although not shown, plots of Poisson's ratio against strain at equal test speeds indicate that Poisson's ratio is lowest at 0 °C. However, the temperature and strain rate dependence of Poisson's ratio do not appear to be very significant. In analyses where Poisson's ratio was required (e.g. calculation of true stress values) an approximate average value of 0.35 was used. The variation of Poisson's ratio with strain is likely to have implications for accurate modelling the adhesive using FEA. A common assumption to simplify data requirements is that $\nu = 0.5$ and, thus volume is conserved. The value and variation of Poisson's ratio indicate that this assumption is invalid for this material. Volumetric components will need to be included in the hyperelastic models. Use of volumetric properties in FE analyses will be described in a future report.

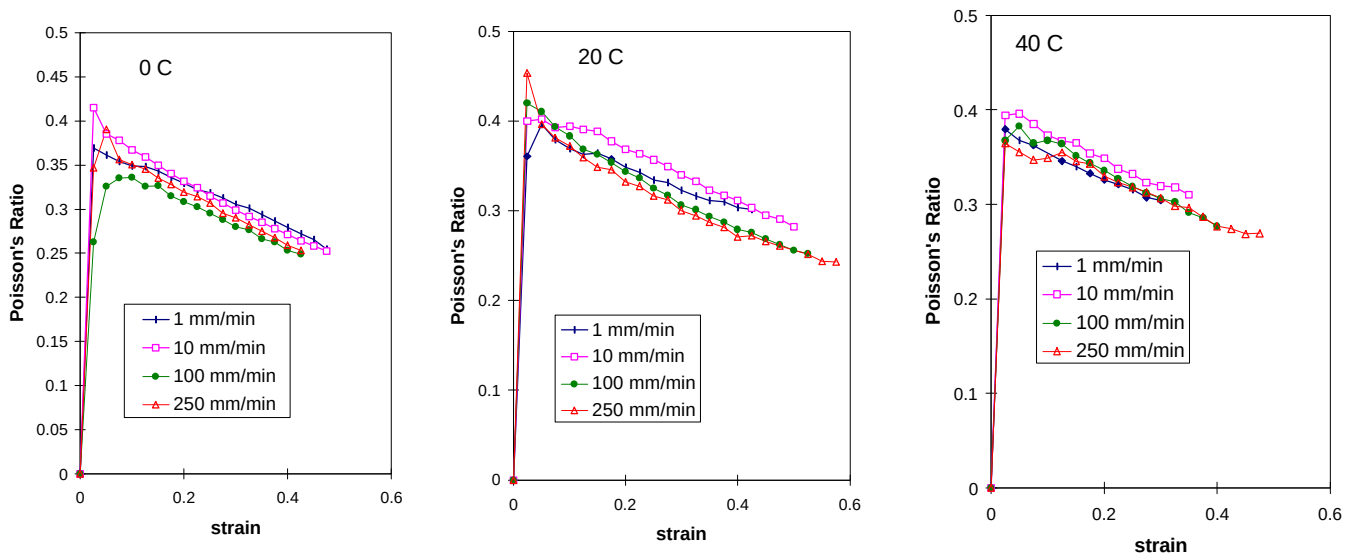


Figure 8: Strain, strain rate and temperature dependence of Poisson's ratio of M70

3.1.5 Failure in Uniaxial Tension Tests

The stress and strains measured at specimen failure are shown in Table 3. These are shown graphically in Figure 9. The data presented are the average of a minimum of 5 tests. The 'stiffness' value is calculated from the ratio of stress to strain at failure (i.e. it represents the slope of a secant drawn from the origin of the stress-strain curve to the point of failure). The product of stress and strain at failure represents an estimate of the 'strain energy' density (strictly speaking the stress-strain curve should be integrated in order to obtain the strain energy but obtaining such data from each test would be a laborious task).

The data in Figure 9a show that the stress at failure decreases with increasing temperature at each test speed. Figure 9b shows that the failure stress values, σ_f , also increase with increasing

test speed, U (proportional to strain rate). The failure stress-test speed curves can be fitted with a function of the form:

$$s_f = z \ln(U) + s_0 \quad (6)$$

The slope of the logarithmic function, z , decreases with increasing temperature. The fitted stress at $\ln(U)=0$ (i.e. when $U = 1$ mm/min) is represented by σ_0 .

The strains at failure do not follow such simple trends. The low strains at failure of the adhesive at -40 °C occur because the adhesive is in the glassy region as the test temperature is below T_g . However, the strain to failure is greater at 0 °C and 20 °C than it is at higher temperatures (Figure 9c). The effect of strain rate on failure strain is more difficult to interpret (Figure 9d). The coefficients of variation in the failure strain are large compared to the stress data. Failure strains at the higher temperatures appear to increase with test speed (presumably due to the higher rate lowering the effective test temperature through time-temperature superposition). However, the data for 20 °C appear to indicate that the strains to failure show no significant dependence on rate over more than 5 decades of test speed.

Table 3: Failure data measured in uniaxial tension tests

Temp	Speed	Stress	+/-	Strain	+/-	stiffness	strain energy
(°C)	(mm/min)	(MPa)	(MPa)	(%)	(%)	(MPa)	(J/mm ³)
-40	1	15.33	3.5	7.3	3.2	211.0	1.11
-40	10	23.40	1.0	3.3	1.1	717.8	0.76
-40	100	29.00	4.3	1.8	1.1	1621.9	0.52
-40	250	33.58	1.8	1.3	0.5	2665.1	0.42
0	1	3.12	0.2	42.2	6.7	7.4	1.31
0	10	3.87	0.2	46.8	8.2	8.3	1.81
0	100	5.51	1.6	49.1	12.1	11.2	2.71
0	250	5.04	0.9	34.0	14.0	14.8	1.71
20	1	2.19	0.6	39.0	9.7	5.6	0.85
20	10	2.87	0.3	48.7	5.5	5.9	1.40
20	100	3.50	0.3	48.4	8.8	7.2	1.69
20	250	3.79	0.4	42.0	7.4	9.0	1.59
22	251	3.92	0.2	38.5	7.7	10.2	1.51
22	2500	4.90	0.8	37.7	9.3	13.0	1.85
22	25000	6.02	0.3	46.6	9.0	12.9	2.80
22	217000	7.23	2.1	39.6	12.8	18.2	2.86
40	1	1.92	0.3	28.9	5.5	6.7	0.56
40	10	2.26	0.2	34.4	6.2	6.6	0.78
40	100	2.77	0.3	40.3	9.4	6.9	1.12
40	250	2.87	0.2	35.3	4.2	8.1	1.01
80	1	1.46	0.3	22.1	3.6	6.6	0.32
80	10	2.05	0.1	26.8	2.9	7.6	0.55
80	100	2.11	0.3	23.7	5.7	8.9	0.50
80	250	2.39	0.5	31.0	5.6	7.7	0.74

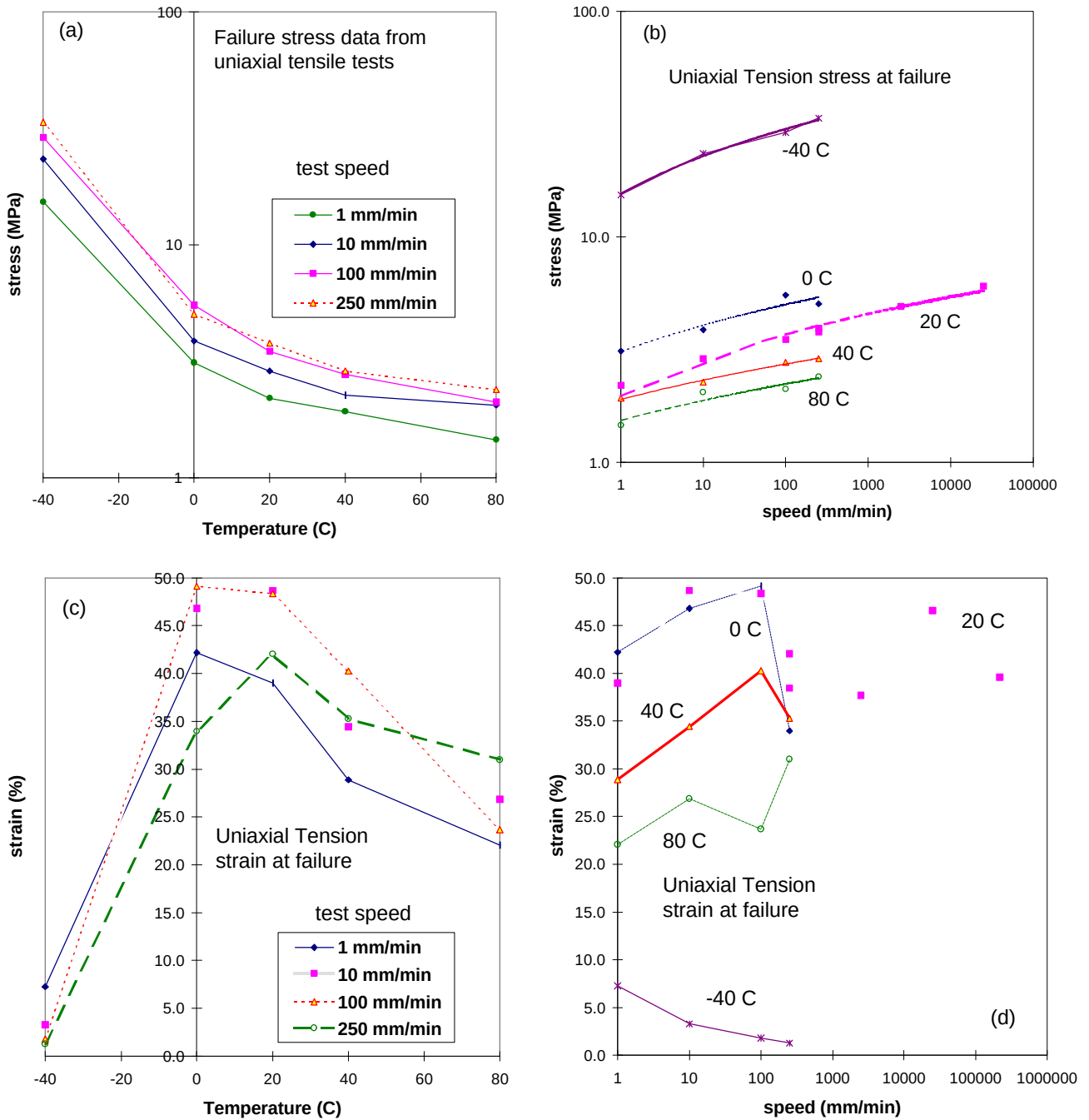


Figure 9: Tensile specimen failure data

3.2 LAP JOINT DATA

3.2.1 Lap Joint Force-Strain Response

The test data for the lap joint specimens are presented as plots of load/bond length against strain (shear strain, γ , = extension/bond thickness). This representation of the data accounts for the variability in the bond dimensions of the specimens (bond length and bondline thickness). The average shear stress in the lap joint is the load/bond length (normalised load) divided by the adherend width (25 mm). Thus, the normalised load is directly proportional to the shear stress and conclusions made about rate/temperature dependence will be applicable to either quantity.

The data determined at each of the test temperatures are shown in Figures 10a-10e. Tests were performed at shear strain rates equivalent to those in the tensile tests (3×10^{-4} , 3×10^{-3} , 3×10^{-2} and $8 \times 10^{-2} \text{ s}^{-1}$) at all temperatures. Additional tests at higher rates (0.8 and 8 s^{-1}) were carried out at 20°C .

The tests at -40°C are characterised by a significantly stiffer response than the tests performed above T_g . The properties at -40°C show the most significant strain rate dependence. This is expected given that -40°C is close to the glass transition temperature. There is no indication in the single-lap joint results of the thermal oscillations seen in the bulk tensile test data. It is thought that the adherends act as a buffer, effectively insulating the adhesive from any fluctuations in the temperature of the air in the chamber.

The measured curves shown in Figure 10 indicate that the sensitivity of the joint response to strain rate falls with increasing temperature. It is noticeable that the low strain results ($\gamma < 0.2$) at the higher temperatures seem to be independent of strain rate.

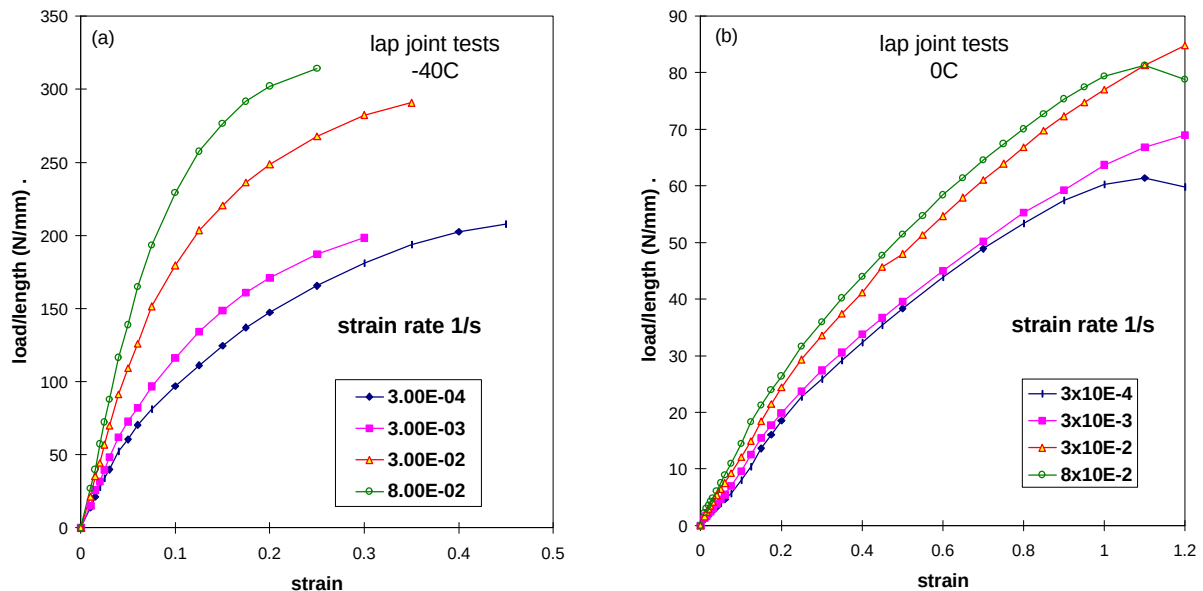


Figure 10: Lap joint test results

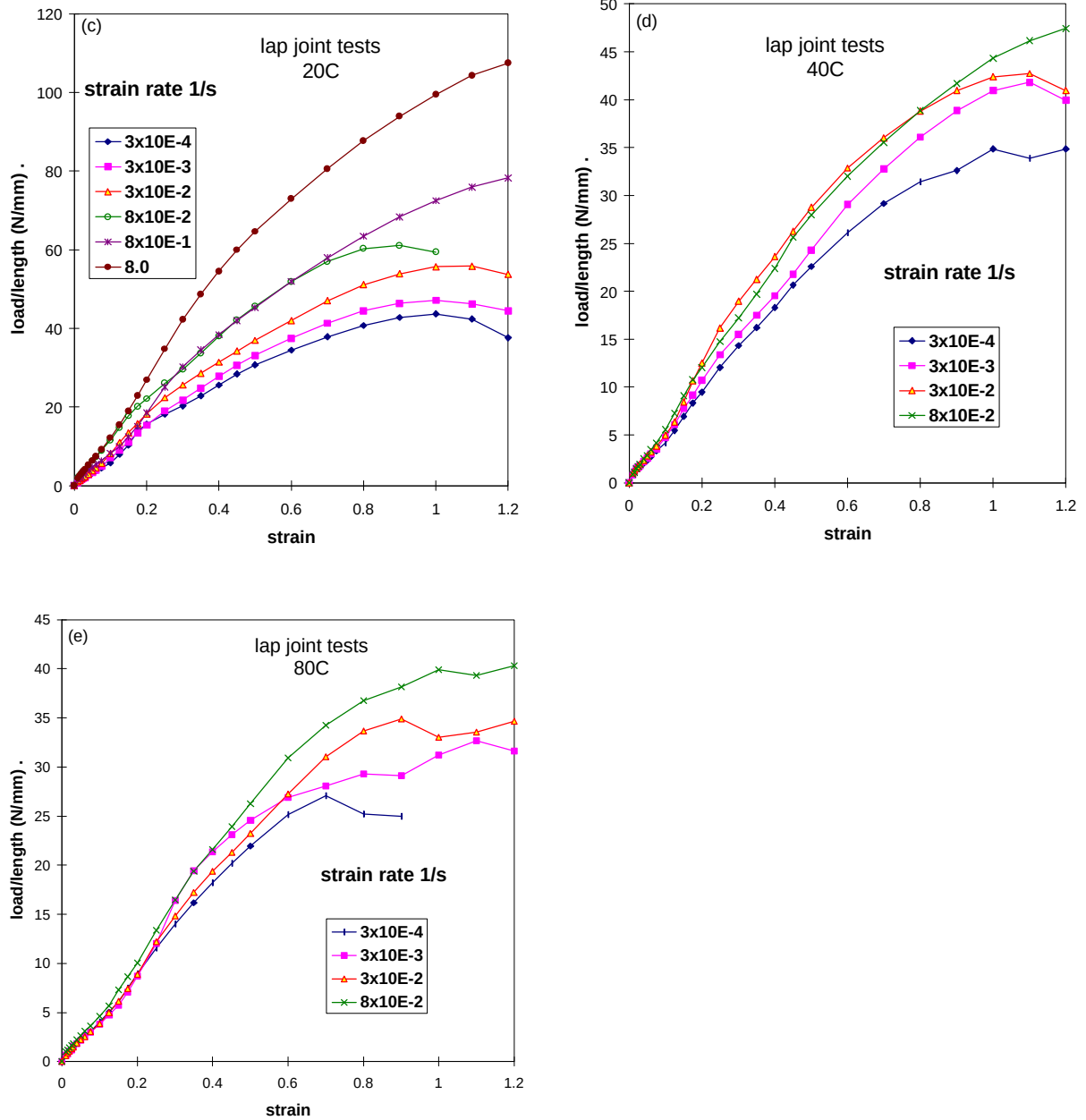


Figure 10: Lap joint test results:

3.2.2 Lap Joint Failure Data

All the lap joint specimens failed as a result of cohesive rupture of the adhesive layer. There was no indication of failure at the adhesive-metal interface or in the adherends. This is the desired situation from a design viewpoint and indicates that the surface preparation gives a good bond in the short term. Longer term durability has not been considered in this work.

The quantities calculated for the failure parameters in the lap joint specimen tests are the same as those calculated from the uniaxial tensile tests. The data are tabulated in Table 4 and shown graphically in Figures 11 and 12. One noticeable difference between the uniaxial tension data and the lap joint data is that strains to failure are significantly higher for the lap joint specimens (by a factor of 2 or more). This suggests that use of material properties derived from the tensile data to model the stress state in the adhesive bond near failure may lead to significant uncertainties due to the amount of extrapolation required.

Table 4: Lap joint failure results

Temp (°C)	strain rate (1/s)	load (N)	load/length (N/mm)	extension (mm)	strain (ext/thick) (mm/mm)	stiffness (N/mm)	strain energy (J/mm ³)
-40	3x10 ⁻⁴	2791	224.6	0.13	0.55	427.7	4.9
-40	3x10 ⁻³	2945	235.1	0.11	0.57	485.7	5.4
-40	3x10 ⁻²	3421	301.7	0.09	0.48	673.8	5.8
-40	8x10 ⁻²	4004	326.1	0.07	0.31	1126.8	4.1
0	3x10 ⁻⁴	791	64.5	0.27	1.18	52.8	3.0
0	3x10 ⁻³	864	73.6	0.27	1.44	51.9	4.2
0	3x10 ⁻²	1092	90.4	0.29	1.50	60.7	5.4
0	8x10 ⁻²	1109	92.5	0.31	1.56	65.3	5.8
20	3x10 ⁻⁴	535	44.8	0.18	1.01	44.4	1.8
20	3x10 ⁻³	612	51.0	0.23	1.08	60.4	2.2
20	3x10 ⁻²	729	59.8	0.23	1.11	49.3	2.7
20	8x10 ⁻²	831	68.7	0.22	1.04	43.6	2.9
20	8x10 ⁻¹	981	81.9	0.28	1.49	56.0	4.9
20	8.0	1335	118.7	0.27	1.51	80.9	7.2
40	3x10 ⁻⁴	413	32.9	0.19	0.90	38.0	1.2
40	3x10 ⁻³	496	39.9	0.23	1.12	35.1	1.8
40	3x10 ⁻²	543	45.0	0.21	1.22	39.3	2.2
40	8x10 ⁻²	596	49.7	0.23	1.39	37.5	2.8
80	3x10 ⁻⁴	354	29.1	0.19	0.85	36.7	1.0
80	3x10 ⁻³	407	32.6	0.18	0.89	39.3	1.2
80	3x10 ⁻²	463	39.3	0.22	1.12	38.0	1.8
80	8x10 ⁻²	510	41.6	0.22	1.04	41.5	1.7

Definitions for Table 4 and Figures 11 - 16

load/length = maximum load divided by bond length, proportional to stress since adherend width was constant;
strain = shear extension at maximum load divided by bond line thickness;
stiffness = load/length divided by strain; proportional to a secant modulus to the point of maximum stress; and

strain energy = product of load/length and strain divided by adherend thickness, this value is an energy density (i.e. energy per mm^3) and is more accurately determined by integrating the stress-strain curve.

The strain rate and temperature dependence of load/bond length and strain at failure shown in Figures 11 and 12 appear to follow similar trends to the tensile specimen data. However, the strains to failure of the lap joint specimens at -40°C are a much higher proportion of the strains to failure in the higher temperature tests than is the case for the tensile test specimens.

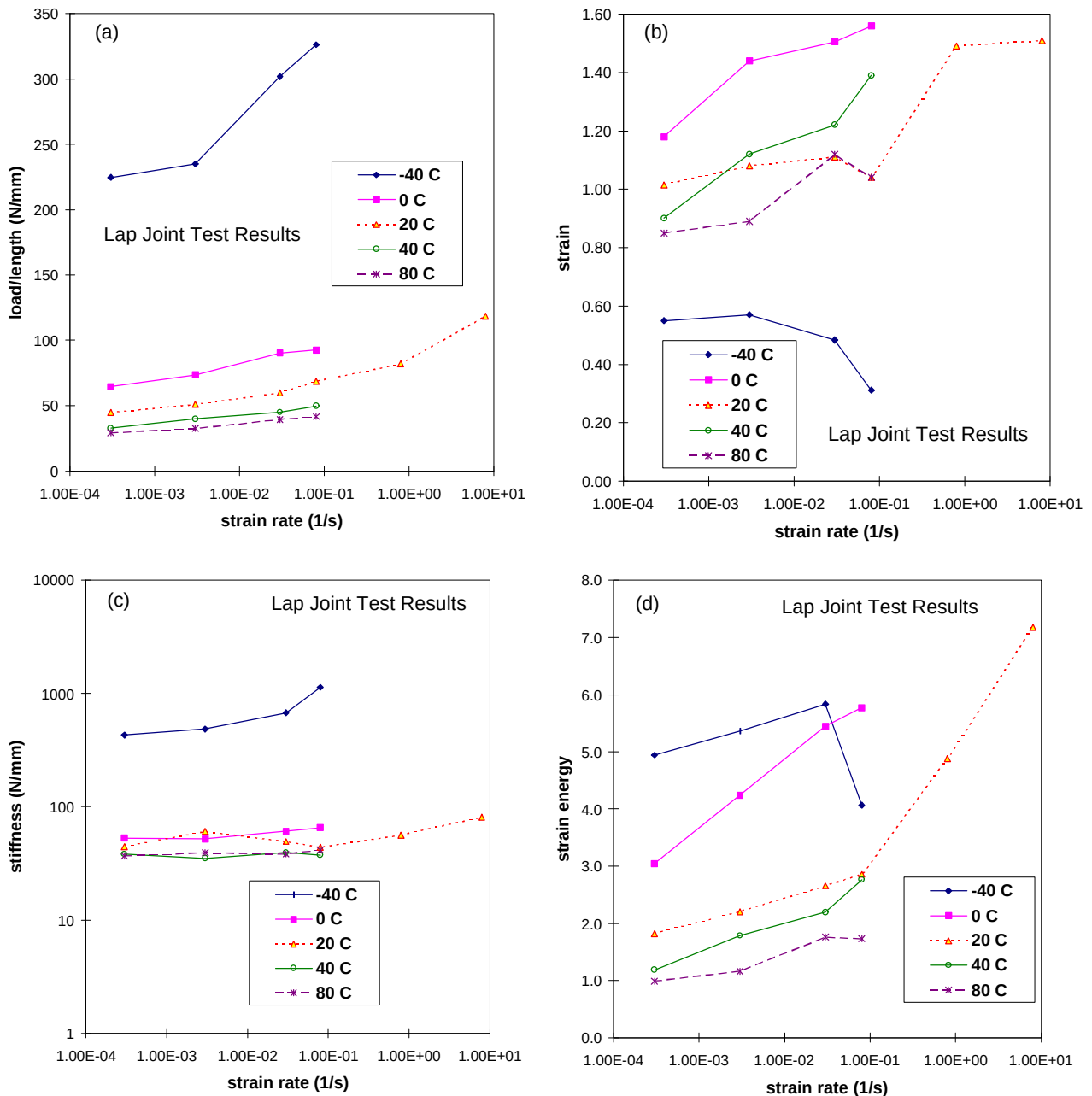


Figure 11: Strain rate dependence of lap joint failure properties

Load (and stress) at failure increase with increasing strain rate and decreasing temperature. In the hyperelastic temperature region, strains to failure also seem to increase with increasing strain rate and decreasing temperature. In this temperature range, there appears to be little dependence of 'stiffness' at failure on strain rate. The strain energy follows the same trends as stress and strain.

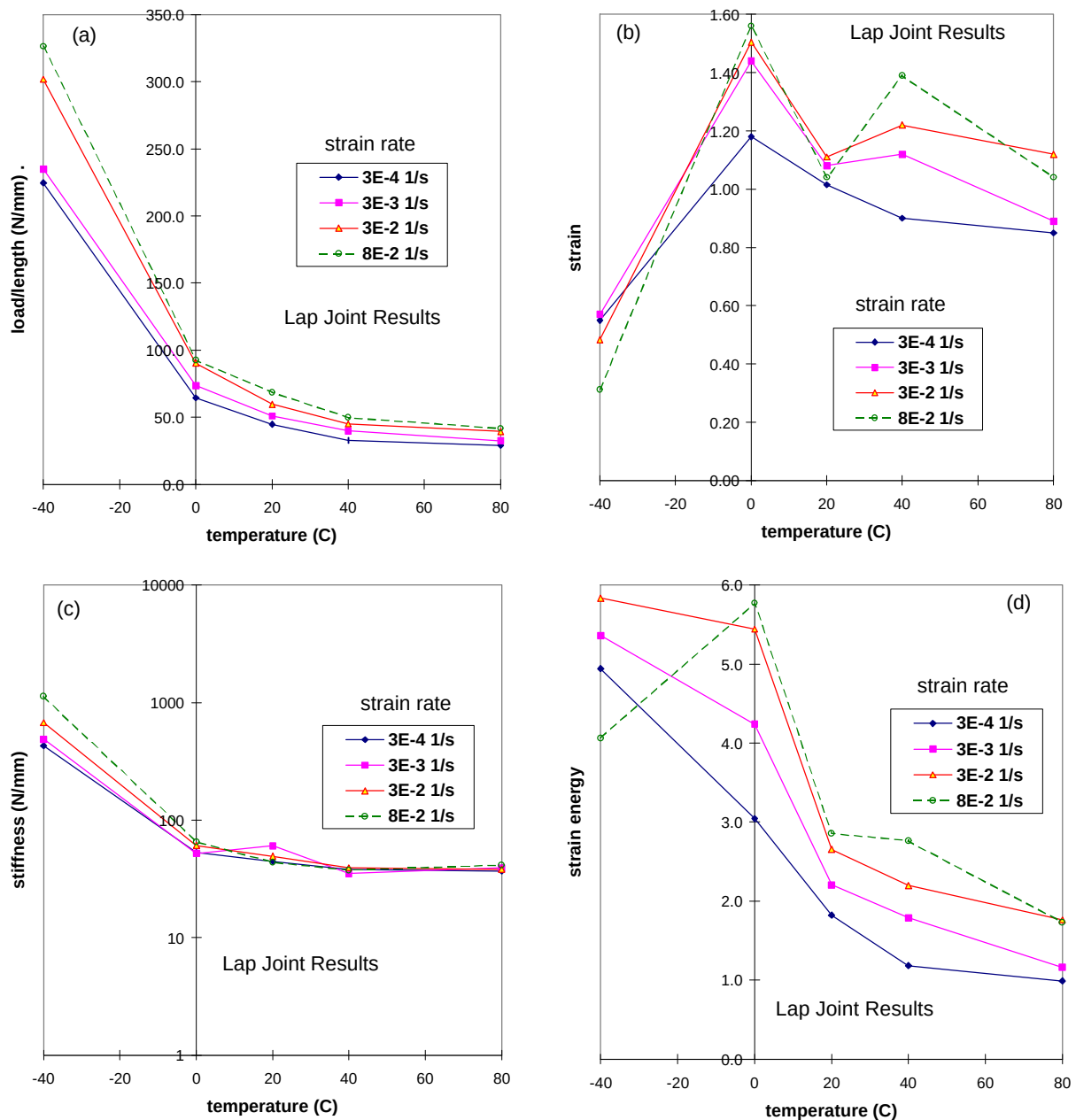


Figure 12: Temperature dependence of lap joint failure properties

4. DISCUSSION

4.1 STATISTICAL ANALYSIS OF FAILURE DATA

Statistical analyses of the data were performed using Qualitek-4 (version 4.75.0001) software supplied by Nutek Inc^(11, 12). The data were analysed using an experimental matrix of 2 factors at 4 levels (Table 5). Data analysis was restricted to the region of results above T_g (i.e. the data at -40°C have been excluded).

Table 5: Factors and levels in statistical analyses

Factor	Level 1	Level 2	Level 3	Level 4
Temperature	0 °C	20 °C	40 °C	80 °C
Test Speed (strain rate)	1 mm/min ($3 \times 10^{-4} \text{ s}^{-1}$)	10 mm/min ($3 \times 10^{-3} \text{ s}^{-1}$)	100 mm/min ($3 \times 10^{-2} \text{ s}^{-1}$)	250 mm/min ($8 \times 10^{-2} \text{ s}^{-1}$)

The various 'failure' parameters were analysed for their dependence on strain rate and temperature. The analysis of variations (ANOVA) results for the tensile tests are shown in Table 6 and those for the lap joint tests in Table 7. The averaged responses of failure loads and strains to temperature are shown in Figure 13. The averaged responses to strain rate are shown in Figure 14.

Table 6: Statistical analysis results - tensile specimen

Parameter	Temperature Significance (%)	Test Speed Significance (%)	Other Factors (%)	Temperature-Speed Interaction Severity (%)
Failure Stress	65.5	24.0	10.5	0.98
Failure Strain	68.8	6.3	24.9	9.44
Stiffness at Failure	33.4	28.0	38.6	3.42
Failure Strain Energy	67.6	16.1	16.3	1.04
Elastic Modulus	55.9	22.8	24.3	9.94

Table 7: Statistical analysis results - lap joint specimens

Parameter	Temperature Significance (%)	Test Speed Significance (%)	Other Factors (%)	Temperature-Speed Interaction Severity (%)
Failure Stress	79.7	16.6	3.7	1.81
Failure Strain	59.3	22.2	18.5	14.08
Stiffness at Failure	66.3	11.4	22.2	9.26
Failure Strain Energy	79.7	16.6	3.7	1.77

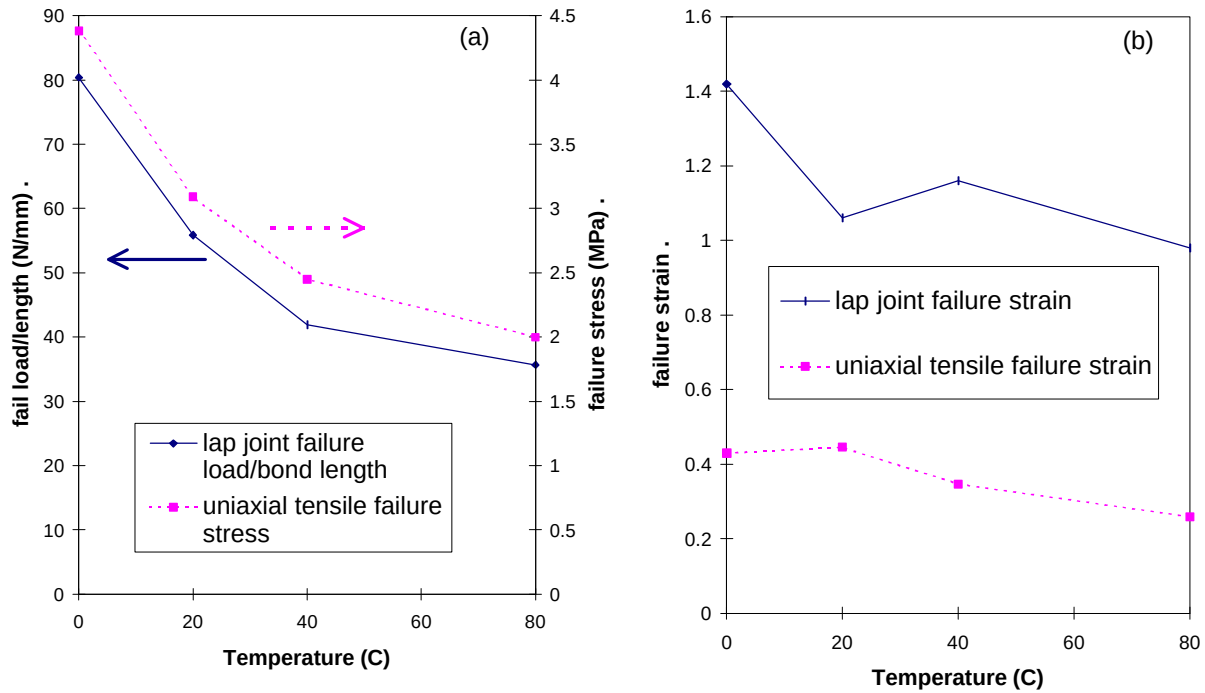


Figure 13: Temperature dependence

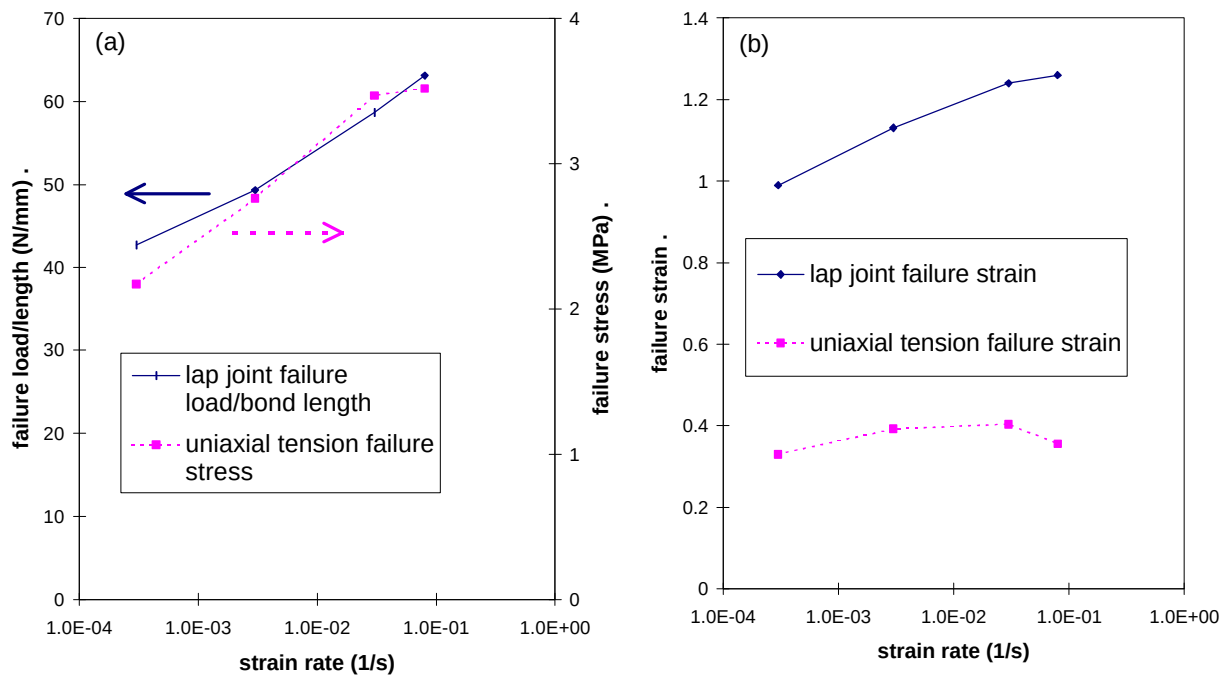


Figure 14: Strain rate dependence

These results show that, over the range of temperatures and strain rates analysed, most of the variance in the data from the experimental mean depends on the test temperature. The strain rate has less influence. The analysis shows that the interaction severity between temperature and test speed is typically less than 10 % indicative of only a weak interaction. Figures 13 and 14 demonstrate the trends of increasing failure loads with decreasing temperature and increasing strain rate. The averaged data show decreasing failure strains with increasing temperatures. Failure strain seems to increase with strain rate in lap joint tests. However, there is little sign of

significant strain rate dependence in the uniaxial tension results (this agrees with the very small contribution to the variance calculated from the ANOVA analysis).

4.2 RELATIONSHIP BETWEEN FAILURE IN LAP JOINT SPECIMEN AND FAILURE IN TENSILE SPECIMEN

Various failure parameters for the tensile and lap joint test specimens are correlated in Figure 15. These plots indicate that the data points for the low temperature tests (-40 °C) do not follow the same trends as the tests in the hyperelastic regions. There is a good correlation between failure stress in tension and failure load/bond length in the lap joint at temperatures in the hyperelastic region. The correlation between failure strains, failure strain energies and stiffness at failure is not as clear.

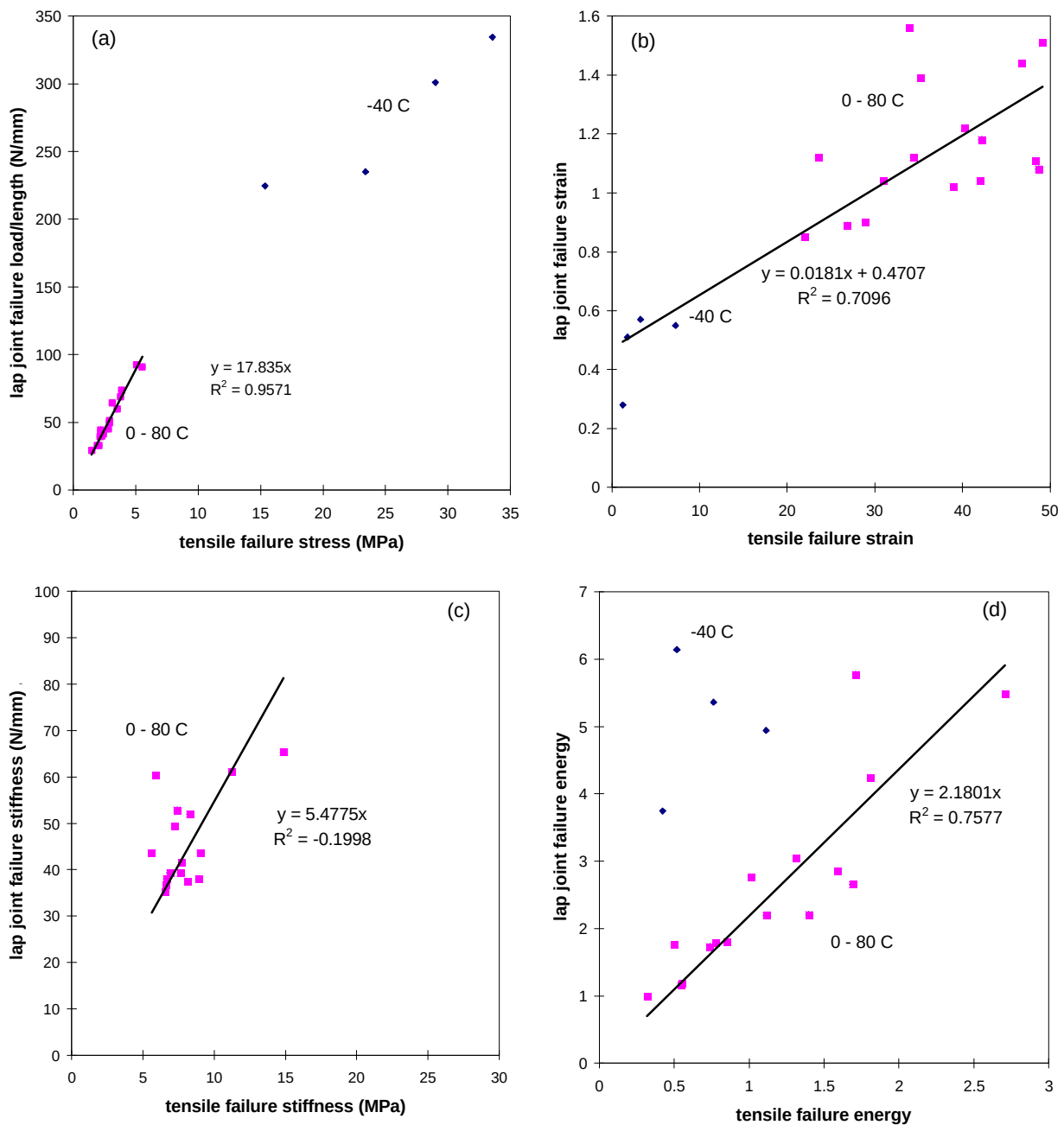


Figure 15: Relationships between failure properties of lap joint and tensile specimens

4.3 TIME-TEMPERATURE ANALYSIS OF FAILURE LOADS

The time-temperature superposition methods outlined in Section 3.1.3 in the analysis of modulus data may also apply to the failure criteria. The failure load data can be plotted as a function of the test time to failure (as calculated from the failure strain and strain rate). The resulting plots for the uniaxial tension and lap joint tests are shown in Figures 16a and 16b respectively.

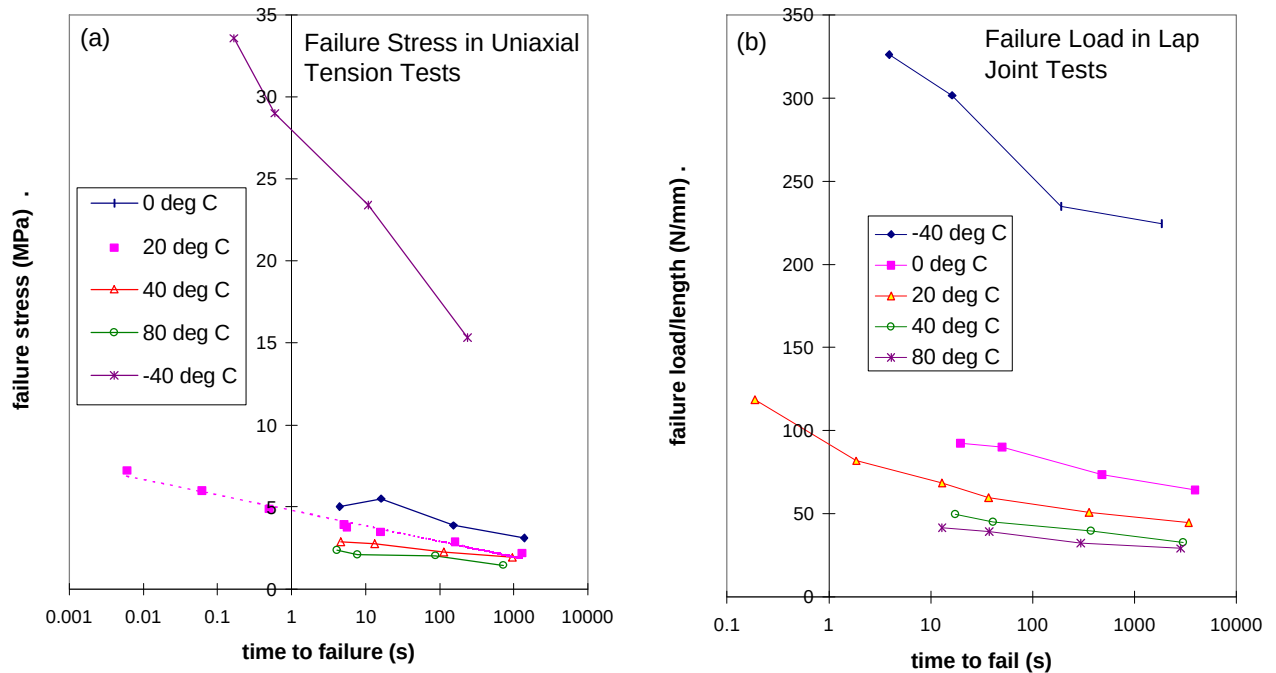


Figure 16: Time dependence of failure loads

The lowest failure loads at -40 °C are significantly greater than the failure loads at any of the other temperatures. Thus, it is not possible to make definite conclusions about how this data relates to the rest. However, the slope of the 20 °C curve for lap joint results is significantly steeper at low times than is at larger times. This suggests that the slopes of the two curves may be similar at the times where the failure loads are comparable.

The curves for the data determined in the hyperelastic temperature range have similar slopes at times to failure greater than 10 s. This suggests that it may be possible to use time-temperature superposition to predict failure loads and times over extended ranges of time and temperature. However, closer examination of the slopes of failure load against $\log(\text{time})$ indicates that the logarithmic slope of the curves seems to decrease with temperature. This implies that the size of the logarithmic time shift between points at equal failure load at different temperatures will decrease as the failure load decreases (i.e. increasing time to failure). The data also tend to imply that the size of the time shift increases as the temperature decreases. Hence, simple linear superposition would not be 100 % reliable. Additional data covering more temperatures and a wider range of times would be needed to fully investigate a quantitative relationship between failure time, sample temperature and failure load.

5. CONCLUSIONS

The effects of strain rate and temperature on the mechanical properties of a typical flexible adhesive have been investigated:

- The results show that the mechanical properties and failure points of flexible adhesives are influenced by both strain rate and sample temperature.
- Both bulk tensile specimens and lap shear joints have similar rate and temperature dependence.
- Stress at failure increases with decreasing temperature and increasing strain rate.
- Above T_g , strain at failure falls with increasing temperature.
- Statistical analysis of the failure values suggests that the hyperelastic temperature range investigated (0 °C to 80 °C) has significantly greater influence on failure criteria than the range of strain rates used ($3 \times 10^{-4} \text{ s}^{-1}$ to $8 \times 10^{-2} \text{ s}^{-1}$). However, it should be recognised that the temperature range covers a significant proportion of the likely service temperatures whilst the adhesive could experience rates of strain running from several decades lower (creep) to several decades higher (impact).
- Poisson's ratio is weakly sensitive to temperature and strain rate but depends significantly on the tensile strain. This will need to be included in any modelling of material properties for FE stress analyses.

Visco-elastic theories have been investigated as means of predicting the stress-strain response and failure properties of flexible adhesives:

- Reasonable predictions of constant rate responses can be made using relaxation constants derived from time-dependent data obtained at a fixed selected strain from tests performed at different rates.
- Slopes of modulus and failure stress against $\log(\text{time})$ at different temperatures above T_g are similar suggesting that time-temperature superposition may be used to predict both of these properties over wider ranges of time or rate. However, slight differences in logarithmic slope suggest that a non-linear superposition approach may be needed for best accuracy. Such an approach would require more data than have been generated in this study.

Analysis of relationships between failure properties of the tensile and lap joint specimens shows that:

- Relationships between the failure properties differ at temperatures above and below T_g .
- Analysis of the various failure properties determined for bulk tensile and lap joint tests suggests that stress and normalised load at failure are well correlated in the temperature range 0 °C to 80 °C.
- Other properties, such as strain, show a lesser correlation between 0 °C to 80 °C.

In general, this work has shown that there is no single property that will describe the mechanical or failure properties of the flexible adhesive studied. These properties vary with both temperature and strain rate. Appreciation of this is vital when load bearing joints are designed so that all possible service conditions can be considered. The findings of this investigation indicate that there are methods that can model the rate and temperature behaviour of the adhesive and of the bonded joint. This will enable interpolation of properties to conditions not measured and, potentially, allow extrapolation to conditions beyond the range measured.

6. ACKNOWLEDGEMENTS

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**RATE AND TEMPERATURE DEPENDENT MECHANICAL
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by

Bruce Duncan, George Hinopoulos, Keith Ogilvy-Robb and Elena Arranz

Project PAJex2 - Flexible Adhesives

PAJex2 Report No 1

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