NPL Report CMMT(A)184
May 1999

Comparison Between Rheological and Bulk Specimen Tests for Creep and Stress Relaxation Properties

B.C. Duncan

Performance of Adhesives Joints Programme
Project PAJ1: Failure Criteria and Their Application
to Visco-Elastic/Visco-Plastic Materials

PAJ1 Report No 15
(milestone 7)

May 1999

SUMMARY

Rheological (shear) and bulk specimen (tensile) tests for determining creep and stress relaxation properties of flexible adhesives have been compared. The rheological creep property measurements were of poor quality. This may have been due to incremental slippage or de-bonding of the adhesive. The rheological creep measurements showed little agreement with the bulk specimen creep measurements. Consequently, the rheological creep measurements appear unreliable. The stress relaxation measurements were more reliable. The rheological and bulk specimen relaxation measurements were comparable.

NPL Report CMMT(A)184
May 1999

ISSN 1361-4061

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

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

Comparison Between Rheological and Bulk Specimen Tests for Creep and Stress Relaxation Properties

B.C. Duncan

PAJ1 Report No 15.

1. INTRODUCTION

Project PAJ1 of the DTI Performance of Adhesive Joints programme focuses on the measurement and analysis techniques required for design with flexible, visco-elastic adhesives. Creep of joints under long-term loading is a potential problem in service. Finite Element Analysis (FEA) can be used to predict joint performance under continuous loading. Time-dependent property measurements - either creep compliance or stress relaxation - are required(1). A previous report(2) described how bulk tension and shear rheometry tests could be used to obtain such data. The purpose of this report is to directly compare the results from these types of measurements.

2. CREEP AND RELAXATION METHODS

The experimental methods have been described in detail in an earlier report(2). Results cannot be directly compared as the levels of stress used in the tension and rheometry tests are significantly different. Data need to be converted into creep compliances or stress relaxation moduli to enable comparison.

The creep test method measures the strain ε(t) in tension (or γ(t) in shear) under constant stress σ (or τ in shear). The tensile creep compliance $J_t(t)$ is calculated:

$$J_t(t) = \frac{e(t)}{s}$$

and the shear creep compliance $J_s(t)$ is determined similarly from γ(t) and τ.

In the relaxation test, the strain is constant and the stress decreases with time. In tension the relaxation modulus E(t) is calculated:

$$E(t) = \frac{s\,(t)}{e}$$

The shear relaxation modulus G(t) is calculated similarly from γ and τ(t).

$J_t(t)$ and E(t) (or $J_s(t)$ and G(t)) are not simply inverse functions of each other. The compliance and relaxation modulus are related through a convolution integral:

$$\int_0^t E(t-u)\,J_t(t)\,du = t$$

In general, $J_t(t) \neq 1/E(t)$ but in the limits of $t \to 0$ and $t \to \infty$ then $J_t(t) \to 1/E(t)$. The same arguments apply in shear.

Shear (G) and tensile (E) moduli are related through Poisson's ratio (ν):

$$G = \frac{E}{2\left(1+n\right)}$$

The adhesives studied in this work had Poisson's ratios of approximately 0.4. Thus tensile moduli should be approximately 2.8 times shear moduli and shear compliances should be approximately 2.8 times tensile compliances.

3. RESULTS

Two flexible adhesives were studied in this work. These were a 2-part polyurethane (3M DP609) and a 1-part epoxy-butadiene (Evode M70). The DP609 adhesive was cured at room temperature (20 - 25 °C). The M70 adhesive was cured at higher temperatures (190 °C for 45 minutes). Bulk tensile test specimens of both were prepared several months before they were tested. Rheometry specimens were prepared prior to testing.

In the case of DP609, the specimens were cured overnight before the tests were performed. It is known that DP609 will continue to cure over much longer periods(3). However, the timescale for these tests made ageing for times comparable with the bulk specimens unfeasible. Specimens were prepared in the rheometer which could not be dedicated for extensive ageing of the test specimens. Therefore, there may be differences between the state of cure of the bulk and rheometer specimens.

For heat cured adhesives, such as M70, the thermal history during cure will influence the final mechanical properties. The bulk and the rheometer test specimens all experienced the same nominal cure cycle (190 °C for 45 minutes). However, the thermal capacity of the plates in contact with the adhesives will differ when preparing bulk and rheometer samples. Thus, the thermal history of the bulk and rheometer specimens may have differed. It was not possible to check the temperature profile during cure of the rheometry specimens.

3.1 CREEP MEASUREMENTS

Figures 1 to 4 show comparisons between the creep compliances determined using the rheometer and bulk tension specimens at 2 temperatures.

Figures 1 and 2 show the compliance measurements made on the DP609 adhesive at 20 °C and 60 °C respectively. All the tension measurements were performed using a lever-arm creep machine(2). At both temperatures, the bulk specimen compliance results are similar. At each temperature, at long times, the creep compliance values tend towards 5 x 10^{-8} Pa^{-1}.

At 20 °C, the initial, elastic compliance is approximately 1 x 10^{-8} Pa^{-1}. This equates to an elastic tensile modulus of 100 MPa. The initial shear compliances are around 3 x

10^{-8} Pa^{-1} (or shear moduli of 33 MPa). The factor of three differences between the shear and the tensile compliances is similar to that expected for a material with a Poisson's ratio of 0.4 to 0.5. The shear compliance curve measured at 0.25 MPa stress shows the same smooth growth in compliance exhibited by the tensile data. The 0.15 MPa shear curve is more discontinuous - there is a large increase then decrease in the creep rate between 1000 and 5000 seconds. However, the initial and final compliances are close to those measured in the 0.25 MPa test.

At 60 °C, the initial, elastic compliance of the tensile specimens is approximately 4 x 10^{-8} Pa^{-1}. This equates to an elastic tensile modulus of 25 MPa. This is in agreement with the decrease in modulus expected from the temperature rise. The four shear curves presented are widely scattered. There is very poor agreement with the tensile data. The initial compliances in the shear tests vary widely. Two of the curves show a rapid rise in the compliance value to ca. 5 x 10^{-6} Pa^{-1} within 10s of the start of the test - failure at the interface is suspected. The other two tests differ by around a factor 10. Both these specimens failed between 1000s and 10000s.

Figures 3 and 4 show the creep compliances determined for M70 adhesive at 20 °C and 60 °C respectively. In contrast to DP609, there is significant scatter between the tensile creep results. Much of the scatter appears to be in the initial, elastic compliance which varies from 1.2 x 10^{-7} Pa^{-1} to 2 x 10^{-7} Pa^{-1} (or an elastic modulus between 5 MPa and 9 MPa). These values are in approximate agreement with the inverse of the initial elastic compliances measured in relaxation tests (and moduli from constant rate tests[(3)]). The elastic compliance values seem to have a low temperature dependence. The shapes of the creep curves at the two temperatures appear similar. However, the times to failure in the tests carried out at 60 °C tended to be much less than those in tests performed at 20 °C. The glass transition temperature of M70 is less than -30 °C. Therefore, limited temperature sensitivity would be expected so far above T_g. The reasons for the scatter in the tensile data are not known. These could be due to differences in the material properties caused by differing cure conditions. Furthermore, it was difficult to prepare specimens free of entrapped voids. High and varying volume fractions of voids could lead to scatter in the mechanical properties of the material. The tensile creep curves are characterised by a sharp 'knee' that is thought to be the transition between the 'elastic' loading and the start of creep. The creep rates are high.

In contrast to the tensile data, the shear creep curves show little creep in the first 100 s of the test. This is followed by a period of rapid creep after which the rate slows. The compliances in the flat part near the start of the test are between 2.5 x 10^{-8} Pa^{-1} to 4 x 10^{-8} Pa^{-1} (equivalent shear modulus of 25 MPa to 40 MPa). These compliances are significantly less than would be expected from the tensile data (by a factor of ca. 30 once the Poisson's ratio has been considered). Again, there are no obvious explanations for this although the cure state of the material may be different to the bulk specimens. The noise seen in the shear data at 60 °C is thought to be caused by temperature fluctuations during the test. The extended temperature module used to achieve the higher test temperature is extremely poor at maintaining a constant temperature. This temperature control system is not suited for maintaining constant temperatures in long duration tests.

The differences between the shear and the tensile data are unlikely to be due to any fundamental differences in the properties of the adhesives in tension and shear. Some preliminary creep studies on lap joint specimens (using the tensile creep equipment) suggest that these creep in a similar fashion to the bulk tensile specimens. The rheometry creep tests contain many potential problems. The actual displacements in the shear creep tests are extremely small. Therefore, any additional displacements, such as slippage of the plates or de-bonding of the adhesive or fillet, will lead to large changes in the determined compliance. These could be responsible for the very different shapes of the creep curves.

3.2. STRESS RELAXATION

Tensile stress relaxation measurements (at 20 °C, 40 °C and 60 °C) for the adhesives M70 and DP609 are shown in Figure 5. The results for DP609 at 20 °C indicate that measurement repeatability is acceptable. The large difference between the relaxation behaviour of DP609 at 20 °C and at 40 °C or 60 °C is due to the proximity of the glass transition temperature, T_g, to 20 °C. Each adhesive seems to relax towards a characteristic relaxation modulus (independent of temperature) at large times. For DP609, this long time relaxation modulus tends to around 2×10^7 Pa. The inverse of this (5×10^{-8} Pa^{-1}) is close to the long term compliance values measured in the tensile creep tests. For M70, the long time relaxation modulus is around 4×10^6 Pa. The inverse of this (2.5×10^{-7} Pa^{-1}) is reasonably close to the compliances measured in the tensile creep tests (despite their scatter). However, the slow rate of relaxation (towards a constant value) does not agree with the high creep rates measured. The high creep rates may be due to the continuing reduction in specimen cross-section as the specimen extends which leads to increasing stress. The initial stress in the creep tests was a significant proportion of the tensile strength of M70. The creep specimens may be experiencing slow rupture and the growth of voids rather than true creep deformation.

Figure 6 shows stress relaxation data for M70 measured at 40 °C. Three shear relaxation curves (at 0.2 %, 1.1 % and 5.1 % strain) and one tensile relaxation curve (13 % strain) are shown. Each of the shear curves shows problems with the data at low times. The 5.1 % strain test saturated the torque transducer until the relaxation had reduced the force into range after 10 s. Both the 0.2 % and 1.1 % strain curves have severe oscillations at low times. The relaxation moduli for the 1.1 % strain test are around a factor of three greater than those in the other two tests. Comparison with the tensile data indicated that the 0.2 % and 5.1 % strain tests give shear relaxation moduli in agreement with expected values from the tensile tests. The 1.1 % strain test gives shear relaxation moduli equal to the tensile moduli - i.e. approximately three times too large. The differences in the shear relaxation moduli measured may be due to uncertainties in the measured angular displacements.

4. CONCLUSIONS

Creep and stress relaxation measurements performed using bulk specimen tensile tests and shear rheometers have been compared. The creep data produced using the rheometer in this work exhibit a wide degree of scatter. In the main, the shear creep data are in poor agreement with the tensile creep data. The quality of the relaxation data produced by rheometry is better than the creep data. There is closer agreement with the bulk specimen tests.

For one of the adhesives (DP609), the long time bulk specimen creep compliances and inverse of the relaxation moduli are in good agreement. For the other adhesive (M70), the continuing high creep rate does not agree with the slowing relaxation processes.

Rheometery can be used to measure creep data. However, the results in this study suggest that the reliability of the data can be suspect. These creep data should not be relied on alone for design but need to be supported by additional measurements (e.g. stress relaxation).

5. ACKNOWLEDGEMENTS

This work was funded by the DTI under the Performance of Adhesives Joints programme. Thanks are due to Tony Maxwell, Keith Robb, Crispin Allen and Bill Nimmo for assistance with the measurements.

6. REFERENCES

1. Hu F. and Olusanya A., Measurement of Creep and Stress Relaxation in Rubber and Rubber Type Materials, PAJ1 Report No 6, NPL Report No CMMT(B)158, December 1997.

2. Duncan B.C. and Maxwell A.S., Measurement Methods for Time-Dependent Properties of Flexible Adhesives, PAJ1 Report No 13, NPL Report No CMMT(A)178, May 1999.

3. Maxwell A.S. and Duncan B.C., Evaluation of a Multi-Functional Adhesives Test Station, PAJ1 Report No 12, NPL Report No CMMT(A)177, April 1999.

Figure 1: Creep Compliance of DP609 at 20 °C

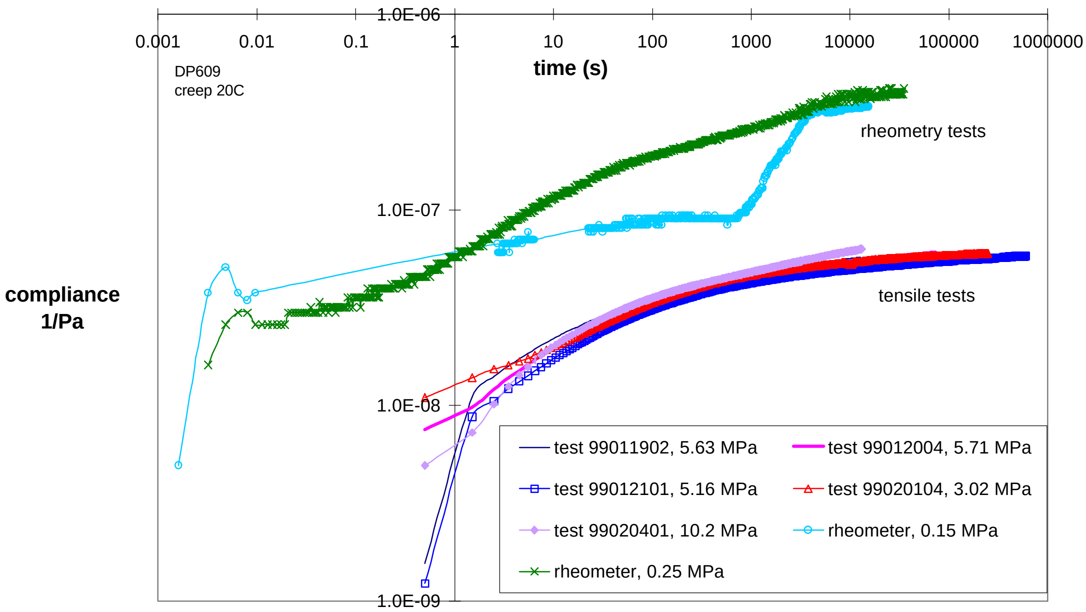

Figure 2: Creep Compliance of DP609 at 60 °C

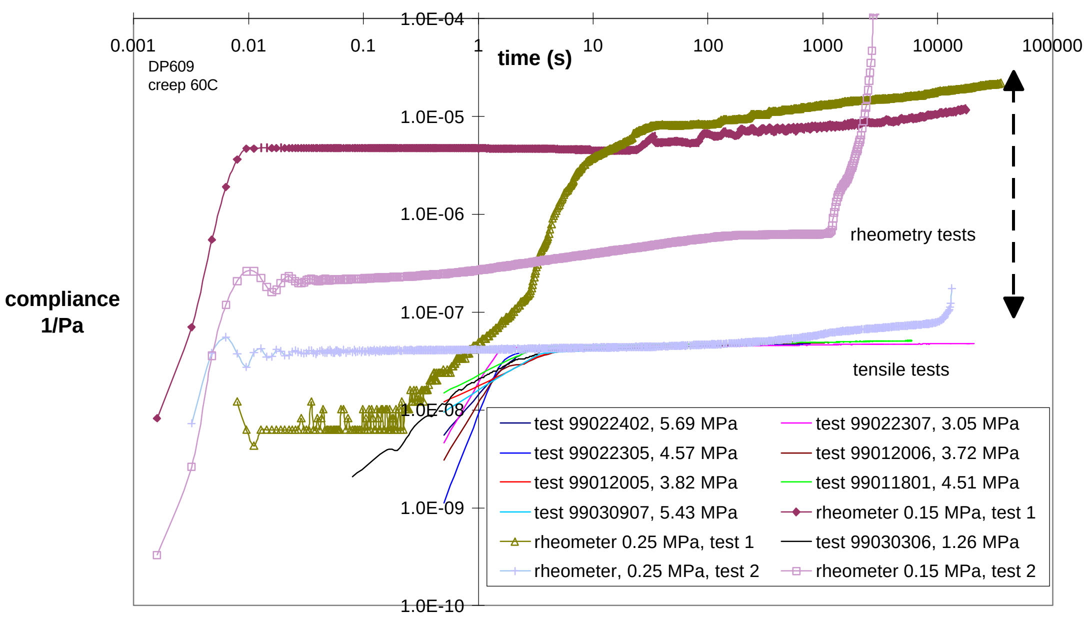

Figure 3: Creep Compliance of M70 at 20 °C

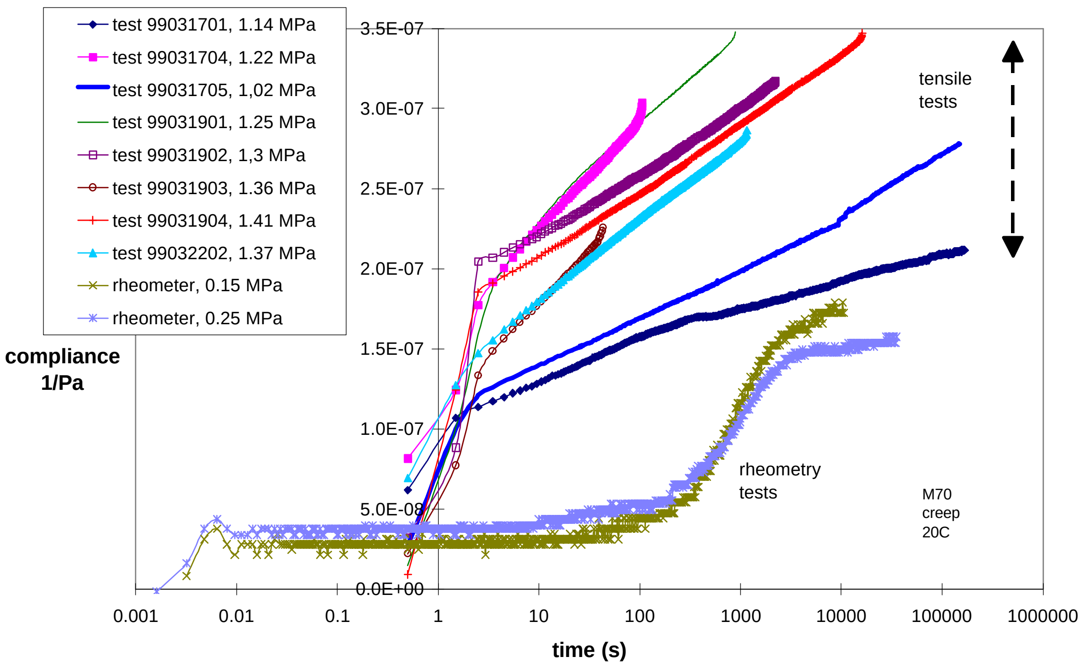

Figure 4: Creep Compliance of M70 at 60 °C

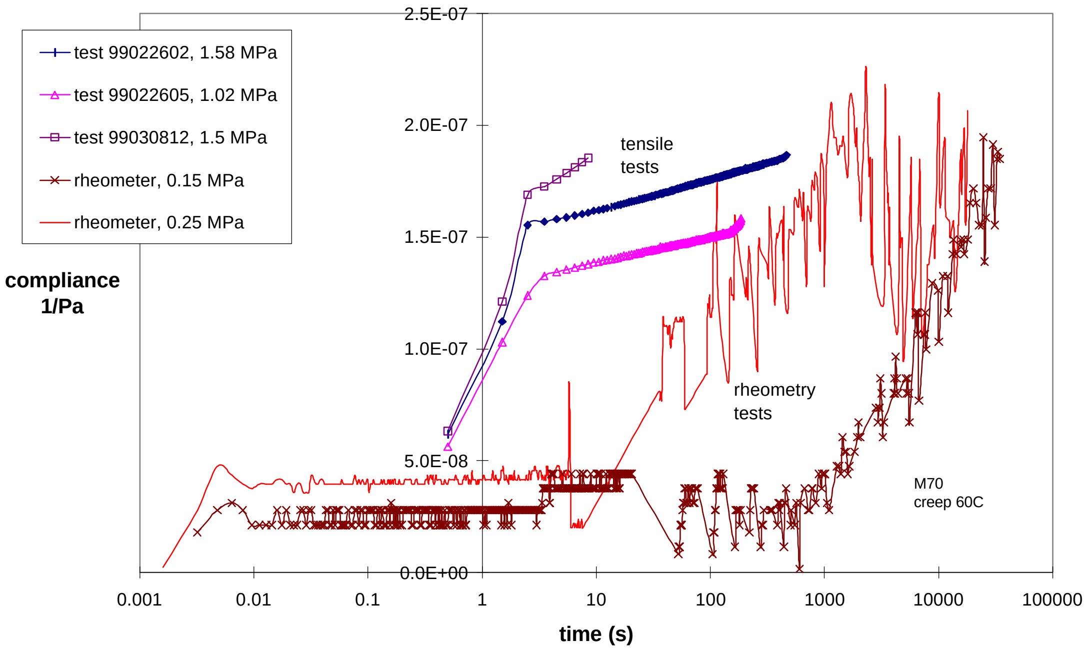

Figure 5: Tensile Stress Relaxation Results for DP609 and M70 Adhesives

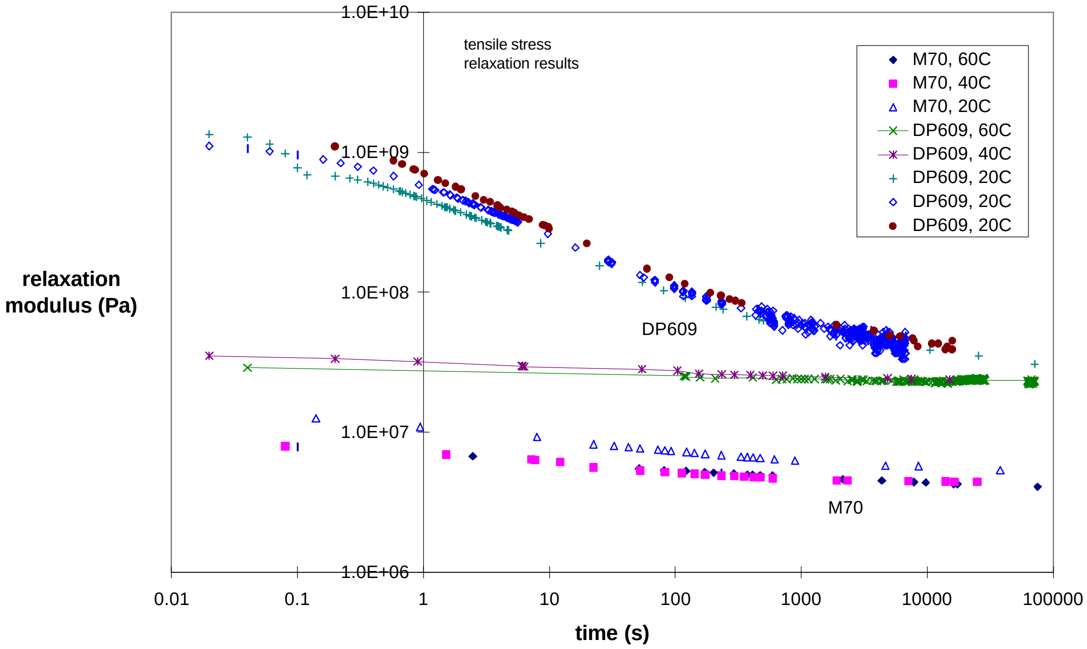

Figure 6: Shear and Tensile Stress Relaxation Results for M70 at 40 °C