INFLUENCE OF TEMPERATURE ON COHESIVE PARAMETERS FOR ADHESIVES

Anders Biel¹ and Thomas Carlberger²

¹University of Skövde, PO Box 408, SE-541 28 Skövde, SWEDEN
anders.biel@his.se, telephone: +46 500-44 85 27
²SAAB Automobile AB, SE-461 80 Trollhättan, SWEDEN
thomas.carlberger@se.saab.com, telephone: +46 520-84 119

ABSTRACT

Experiments are performed to evaluate the temperature dependence of the stress-elongation relation for an engineering epoxy adhesive. Seven temperatures from -40°C to 80°C are considered. At each temperature, about seven experiments are performed with a double cantilever beam specimen. The experiments are evaluated using an inverse solution. The results show that the peak stress decreases monotonically with temperature, from about 55 MPa at -40°C to about 11 MPa at 80°C. Thus, the shape of the stress-elongation relation varies with the temperature. At higher temperatures, the fracture energy decreases slightly.

1. INTRODUCTION

Polymers are known to be strongly temperature dependent. Adhesives, which consist mainly of polymers, are therefore believed to be temperature dependent. In an automotive car body, the material temperature may span a significant temperature range. Excluding the engine compartment, measurements show temperatures in the range -40°C to 125°C. Thermo-set adhesives typically show a glass transition temperature, \( T_G \), between 85°C and 120°C. Thus, the temperature effect is expected to be significant in the present temperature range.

An automotive structure subjected to impact is supposed to fulfil the crash requirements throughout its specified temperature range. In the open literature, only limited information is available concerning temperature dependencies of the stress-elongation relation. Some temperature studies have been performed cf. e.g. Ashcroft et al. (2001) which show a transition in fracture mode from brittle at -50°C, stick-slip at 22°C to ductile at 90°C. To this end, it is noticeable that recent developments of crashworthy adhesives are focusing on increasing fracture energy rather than ultimate strength. This new behaviour is created through alteration of...
the chemical mixture e.g. by adding minerals or thermoplastics. Such chemical modifications are likely to influence the temperature dependency of the adhesive, inducing the need for renewed temperature studies.

It is well known that adhesives in the form of a thin layer between stiffer adherends behave differently from the adhesive as a bulk material. This is due to the constrained state of deformation. An effect of the influence of the constraint is the variation of the fracture energy with the thickness of the adhesive layer; cf. e.g. Kinloch (1987).

A convenient method to determine the adhesive properties is the use of simple test geometries and inverse formulas as described in e.g. Andersson and Biel (2006) and Leffler et al. (2007). Olsson and Stigh (1989) develop an inverse method for pure peel using the double cantilever beam (DCB) specimen, cf. Fig. 1.

![Fig. 1. Double cantilever beam specimen. Unbonded part of the specimen can be considered as a crack, i.e. $a$ is the crack length.](image)

During an experiment, the applied force, $F$, and the rotation of the loading point, $\theta$, are measured as functions of the elongation $w$ of the adhesive at the start of the adhesive layer. The inverse formula reads

$$\sigma = \frac{dJ}{dw} = -\frac{d}{dw}\left(\frac{2F\theta}{b}\right),$$

(1)

where $b$ is the width of the specimen. The result is the stress-elongation relation for the adhesive layer. This relation is closely related to the fracture energy of the adhesive. An application of the path independent $J$-integral shows that the area under the stress-elongation relation equals the fracture energy, cf. e.g. Andersson and Stigh (2004). The present adhesive, DOW-Betamate XW 1044-3, has been extensively studied at room temperature, cf. Andersson and Stigh (2004); Andersson and Biel (2006).

In the present study we study the influence of temperature on the stress-elongation relation. The temperature range is $-40^\circ$C to $80^\circ$C in intervals of $20^\circ$C. Special consideration is given the variation of fracture energy and peak stress with temperature.
2. EXPERIMENTS

2.1 Experimental setup and procedure
The test machine is specially designed for testing of DCB-specimens, cf. Fig. 2. The specimen is oriented vertically. Both crossheads are moving symmetrically around the centre of the machine. Two horizontally working ball screws powered by an electric motor control the motion. A load cell measures the force $F$. The angle, $\theta$, at the loading point is measured with an incremental shaft encoder. The position of the crossheads is measured with a linear potentiometer. Two Linear Voltage Differential Transformers (LVDT) are applied at the outside of the adherends to measure the elongation of the adhesive layer at the start of the layer (not shown in Fig. 2).

![Test machine without LVDT:s and specimen](image)

The specimens and the entire tensile test machine, except the control box and the computer for data acquisition, are placed in a climate chamber. When the desired temperature is stabilised the test equipment is calibrated. Subsequently a specimen is mounted. After an additional temperature check the experiment is performed. The temperature is measured individually for each experiment using a thermocouple attached to a dummy specimen in the climate chamber. A temperature variation of $\pm 2^\circ$C is allowed.

2.2 Specimens
The adhesive is an engineering epoxy, DOW-Betamate XW 1044-3. The curing condition is $180^\circ$C for 30 minutes. At room temperature the adhesive has a viscosity of 4 kPas (similar to toffee) and it is preheated to about $50^\circ$C before application. After curing, $T_G \approx 90^\circ$C.

The dimensions of the specimens are $L = 165$ mm, $a = 80$ mm, $b = 5$ mm, $H = 6.6$ mm, and $h = 0.2$ mm, cf. Fig. 1. The adherends are made of tool steel, Rigor Uddeholm, with a yield-strength larger than 500 MPa securing elastic behaviour of the adherends during the experiments. Two plates are joined with the adhesive. The initial crack length is created by a 0.2 mm thick PTFE-film that also works as a distance giving the correct layer thickness. Before applying the adhesive, the plates are cleaned with heptane and acetone. The adhesive is cured for 30 minutes at $180^\circ$C. Slow cooling is allowed in order to minimise the influence of residual stresses. After curing, the plates are cut into specimens by use of band saw. Subsequently, the specimens are machined to final dimensions.

2.3 Experiments
The experiments are performed with a nominally constant crosshead velocity, $d\Delta/dt = 10$ $\mu$m/s.
Due to friction in the machinery, the crosshead velocity varies with the temperature. This is indicated in Fig. 3 where the strain rate at the peak stress is shown at the different temperatures considered. It may be noted that a constant crosshead velocity results in an increasing strain rate during an experiment. A total of 53 experiments are performed with about seven experiments at each temperature. The mean strain rate at peak stress is shown in Fig. 3 for each temperature.

![Fig. 3. Strain rate, $\varepsilon$, at the moment of peak stress in the adhesive vs. temperature, $T$.](image)

### 2.4 Experimental results

Figure 4 shows $J$ and $\sigma$ as functions of $w$ for all temperatures. At each temperature, an average curve is calculated. Evaluation of $J$ is performed by fitting a Prony-series to the $J$-$w$ curve of each specimen cf. Andersson and Biel (2006). Differentiation of the Prony-series gives the constitutive relation, $\sigma$-$w$, of the adhesive according to Eq. (1).

![Fig. 4. Left: $J$ as a function of $w$; Right: $\sigma$ as a function of $w$.](image)

As shown in Fig. 4, the peak stress and the critical elongation are strongly dependent on the temperature. The elongation at the peak stress occurs at about 2-6 µm for all experiments.

Four of the experiments, three at -40ºC and one at 0ºC, show an abrupt cusp in the $J$-$w$-curve. These four experiments are considered as failures. Initial studies show no signs of initial flaws in the adhesive that might explain this behaviour. Further investigations are required. The result is interpreted as due to a zone of weakness at the start of the adhesive layer. These four experiments are neither included in Fig. 4 nor in the further evaluation of the experiments. The
number of evaluated experiments is given in Table 1.

<table>
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<th>$T$</th>
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Table 1. Number of evaluated test specimens at each temperature.

Figure 5 shows the fracture energy for all evaluated experiments; both the average value at each temperature and the standard deviation, $s$, are shown. At lower temperatures, the fracture energy seems independent of the temperature. For the experiments performed at 60°C and 80°C the fracture energy is slightly lower indicating the vicinity to the glass transition temperature. The maximum temperature for automotive applications is around 120°C. The present test equipment is not specified for this temperature, but future research should include at least this temperature.

Figure 5 shows the peak stress, $\sigma$, of the adhesive. The peak stress decreases monotonically with increasing temperature. Thus, there is a substantially difference in the temperature dependence of the fracture energy and the peak stress. An explanation might be that the fracture energy is associated with the covalent bonds in the molecular structure while the peak stress is dependent on both the van der Waals and covalent bonds. The number and stiffness of the van der Waals bonds are expected to decrease with temperature and disappear at $T_G$. This can be the reason for the monotonic decrease in peak stress. Experiments at temperatures above $T_G$ would then result in a temperature independent peak stress.

Figure 6 shows two fracture surfaces put together and photographed. The fracture surface at 80°C shows more stress-whitening than the fracture surface at -40°C. With the interpretation of the effects of the van der Waals bonds given above, the increase in stress whitening would be associated with increased molecular mobility due to a gradual decrease of van der Waals bonds.
3. CONCLUSIONS

Experiments have been performed to evaluate the temperature dependence of the stress-elongation relation for an engineering epoxy adhesive. The area under the stress-deformation relation, i.e. the fracture energy is virtually independent of the temperature at the lower temperatures considered. At higher temperatures, $J_c$ decreases slightly with $T$. At the highest temperature, 80ºC, $J_c$ has decreased 20 %. The peak stress decreases monotonically for all temperatures considered.

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REFERENCES