In situ straining of epoxy adhesives

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Received 2 October 1990; accepted in revised form 22 January 1992

The microstructure of an adhesive can have a significant effect on the fracture of bonded joints. In the present work, the effect of large second-phase filler particles (approximately 3 to 100 μm) has been studied using a scanning electron microscope fitted with an in situ *straining stage. Observations of the fracture of bonded joints and bend bars containing blunt notches have been combined with stress analysis to determine the fracture stress of the filler particles.*

Key words: adhesives; filler particles; fracture stress; *in situ* straining; scanning electron microscopy; epoxies

The use of an *in situ* straining stage in a scanning electron microscope (SEM) has proved to be a useful technique in determining fracture mechanisms in particulate-filled epoxy resins[1], high resolution strain measurements in carbon fibre-reinforced epoxy specimens[2] and crack tip opening in brittle epoxy systems in order to evaluate strain fields at crack tips and hence the fracture toughness (K_{IC}) of adhesives[3].

Many epoxy adhesive systems contain rigid filler particles such as silica, alumina, glass and dolomite. These fillers have been cited to increase the fracture energies[4-6] and the fracture toughness[4] of epoxy systems, but reduce both the flexural strength and the tensile strength of the same systems[4]. However, the fracture toughness of these epoxy systems depends on both the volume fraction of particles and the particle size of the filler[7]. The increase in fracture energy has been proposed to be due to the addition of second-phase particles which act to pin cracks[8]. The mechanical properties of the particle/matrix interface also control the properties of the epoxy system[7]. For a crack to propagate in a cross-linked polymer network, all the bonds of a chain lying between its junction and points in the network have to be stressed to their breaking point before the chain eventually breaks[9]. This bond stressing also occurs in an epoxy system and results showed that the small epoxy resin bonds were capable of storing energy when the main chain is stressed[10].

The aim of the present study is to investigate the fracture mechanisms of an epoxy resin matrix containing calcium silicate filler particles. Qualitative observations of bond and adhesive failure mechanisms using an *in situ* SEM straining stage have been combined with a quantitative analysis of local failure stresses. The results are assessed in terms of the influence of second-phase particles on the failure of bonded joints.

Experimental procedure

The yield stress (σ_{ys}) of the adhesive was measured using a screw-driven 100 kN Zwick testing machine. Strain rates were derived from a cross-head displacement rate of 1 mm min^{-1} and specimen elongation was measured using a dual averaging MTS extensometer with a 25 mm gauge length. The extensometer was attached to the gauge of the tensile test-piece machined from rods of adhesive. The ends of the adhesive bars were glued into metal bolts (Fig. 1(a)), which in turn were fitted into the grips of the testing machine. Load and elongation measurements were made during a test and from these a value of the yield stress associated with a 0.1% plastic strain offset was defined.

Two geometries were tested in the straining stage of an SEM: single lap joints loaded in tension (Fig. 1(b)) and notched bend bars loaded in four-point bend (Fig. 1(c)). Single lap joints were made from 6082 aluminium treated with a conversion coating and bonded using a hot-cure epoxy paste adhesive. Notched bend bars were produced from discs of adhesive pressed in a steel mould and tensile test-pieces were machined from rods of adhesive pressed in the same mould. In both cases the adhesive was cured under identical conditions to the bonded lap joints.

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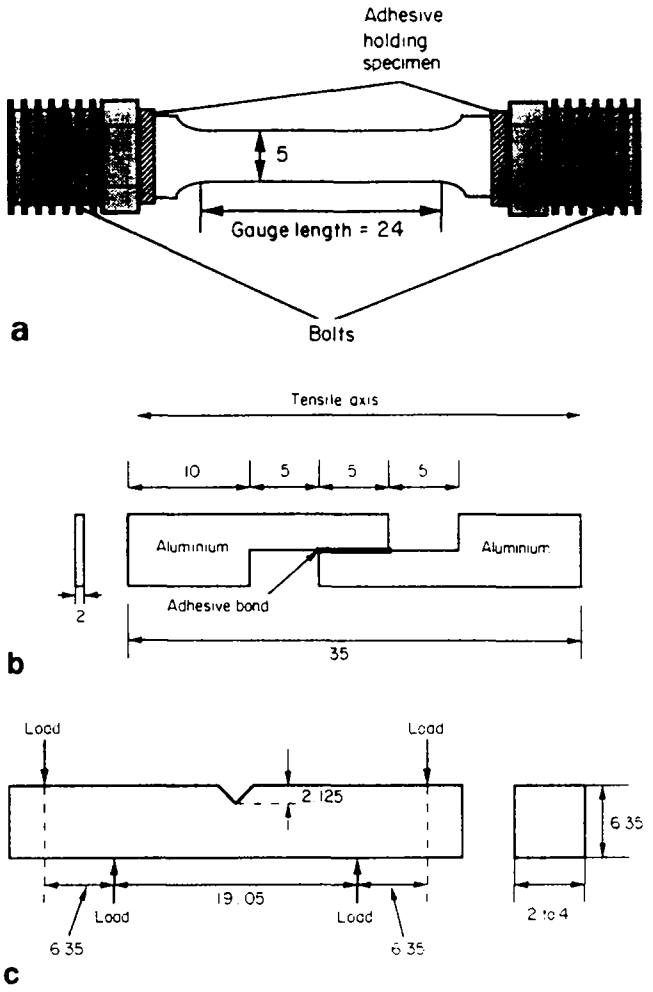

Fig. 1 (a) Tensile test specimens; (b) SEM tensile bonded specimens; and (c) notched bend bars used for evaluation of fracture stress. All dimensions in mm

The failure process was studied using an ISI DS130 SEM operating at an accelerating voltage of 20 kV and 0° tilt. The straining stage used in the SEM was manufactured by Hexland with a maximum load capacity of 785 N. Strain rates used on the adhesive joints and bars were derived from a cross-head displacement rate of 100 μm min^{-1}.

Prior to mechanical testing in the SEM straining stage, all specimens were polished to a 1 μm finish. A gold coat was applied to one side of both the joints and the bend bars so that charging of the adhesive surface was prevented when viewed using secondary electron imaging (SEI). This gold coat proved to be a useful bonus; when cracking within the adhesive occurred, the coat was ruptured and 'flaring' was seen on the surface of the bar, indicating the presence of a crack. In order to evaluate the length and position of this crack, back-scattered electron imaging (BEI) was used.

Adhesive and particulate fracture stresses were evaluated from bend bars containing blunt notches. These act as stress raisers when loaded; the magnitude of the stress is dependent on the load applied and the position below the notch root, the maximum stress being achieved below the root of the notch. Loads were applied to the bend bars in increments of 18.6 N; after each increment, loading was stopped and the region below the notch was analysed for failure of filler particles. When a cracked filler particle was observed, the load at which this occurred and the position below the notch were noted. These parameters were used together with specimen dimensions, the yield stress of the adhesive and the stress distribution obtained from finite element analyses[11] to calculate the fracture stress (σ_f) of the filler particles and the fracture stress of the bulk adhesive.

Results and discussion

The observed deformation up to the onset of failure of a bonded joint is shown in Fig. 2 and may be separated into two processes. Fig. 2(a) shows the bond under zero load in which the angular particles are the calcium silicate filler particles. On loading it is observed that the particles catastrophically fail through their bulk at a critical load (Fig. 2(b)). The particles were generally observed to fail at an angle to the bondline but at right angles to the direction of maximum principal stress. With continued loading, the cracks within the filler particles extend into the epoxy matrix (Fig. 2(c)) before final catastrophic failure of the bondline occurs by interlinkage of the defects when the fracture stress of the adhesive is attained (Fig. 2(d)).

Measurements of the fracture stresses (σ_f) of the adhesive and the filler particles were made using notched bend bars. The nominal bending stress (σ_{nom}) taken at the notch root is calculated from Equation (1):

$$\sigma_{nom} = \frac{3PL}{B(W-a)^2} \qquad (1)$$

where P is the load, L is the loading arm, B is the specimen depth, a is the depth of the notch and W is the width of the specimen (Fig. 1(c)). Although the original analysis was derived for stresses in the bulk of the material, the model was verified using etch-pitting techniques on the *surface* of bend bars to indicate plastic zone sizes at a variety of different loads revealing that surface effects are similar to bulk material effects[11]. To evaluate the appropriate fracture stress it is necessary to know the yield stress of the adhesive, which in this case was measured to be 39 MPa. It is assumed that failure takes place at the position of maximum stress below the notch and, by determination of the position and comparison with finite element analyses, the local fracture stress can be defined.

An example of the application of this technique can be seen in Figs 3 and 4. In Fig. 3, prior to catastrophic failures, particle failure was observed at an applied load of 52 N and at a position approximately 400 μm below the notch tip. Using Fig. 5, together with the calculated nominal bending stress-to-yield stress ratio, a stress intensification (σ_{1max}/σ_y) of 1.53 is obtained. This represents a fracture stress for the calcium silicate particle of 60 MPa. With continued loading, failure of the bend bar occurs (Fig. 4) and the fracture stress of the bulk adhesive can therefore be defined as 74 MPa.

The quantitative measurements of the fracture stress of the particulate and the epoxy matrix support the qualitative examinations of the bond failure. The particulate has a lower fracture stress than the bulk

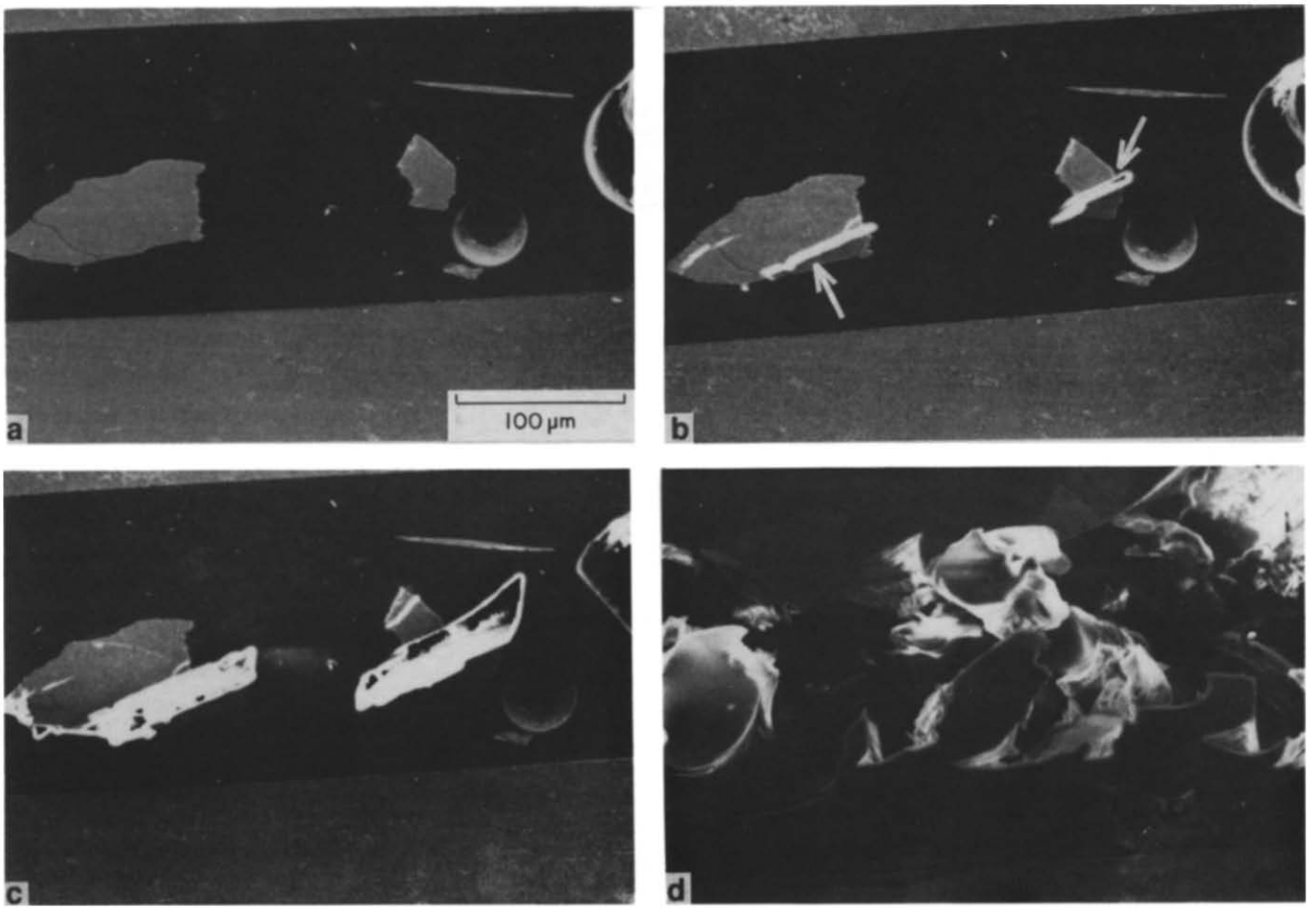

Fig. 2 Scanning electron micrographs of bonded tensile specimens at: (a) 0 N; (b) 392 N; (c) 540 N; and (d) 579 N. Cracked particles are arrowed. All images are viewed using secondary electron imaging

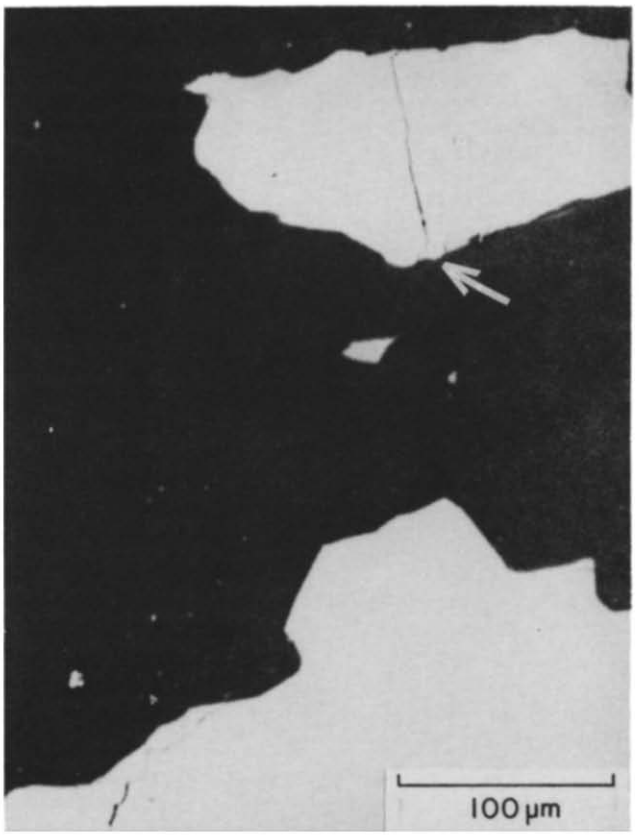

Fig. 3 Cracked filler particle in a notched bend bar at an applied load of 52 N (arrowed)

Fig. 4 Fractured bulk adhesive, cracked particle arrowed approximately 400 μm below the notch of the bend bar

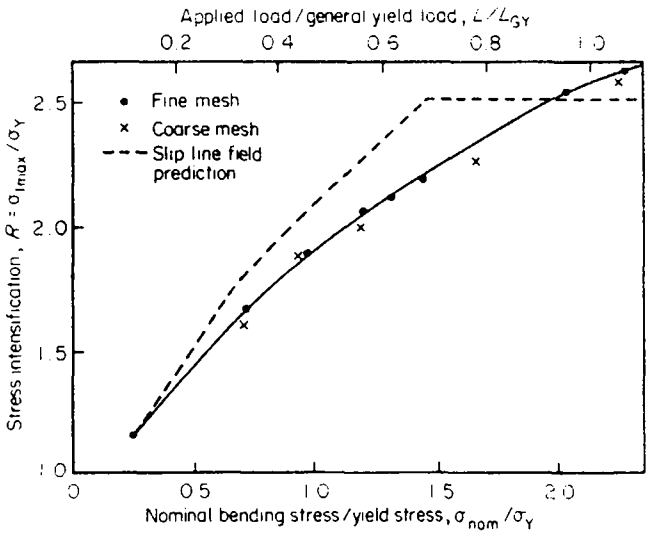

Fig. 5 Variation of stress intensification with applied load for a notch with flank angles of 45° and a root radius of 0.25 mm (after Reference 11)

adhesive and therefore fails at a load prior to bond failure. This observation supports work carried out on the same adhesive in the region of stable crack growth. Catastrophic adhesive failure occurs when the ligament of adhesive between the particles fails by ductile tearing[12]. Unlike the observations made in the SEM, where surface effects are observed, the failure processes in this case occur in the bulk of the adhesive. The value of 74 MPa for failure of the epoxy matrix is in line with previous findings[13,14] and indicates that the final failure of the joint under tensile loading is related to epoxy matrix properties. The significance of this to the nature of crack propagation in epoxy resins has been addressed separately[12].

Conclusions

1) The fracture stress of calcium silicate filler particles was found to be 60 MPa and that of the bulk adhesive was found to be 74 MPa.
2) Ultimate failure of the joint is determined by the epoxy matrix properties since some crack extension from the particle to the matrix occurs before final tensile failure.
3) With continued loading the cracks extend into the matrix, interlink and catastrophic bond failure takes place.

References

1 **Roulin-Moloney, A.C., Cudre-Mauroux, N. and Kausch, H.H.** 'In-situ fracture tests in the scanning electron microscope' *Polymer Composites* **8** No 5 (1987) pp 324-330

2 **Kortschot, M.T.** 'High resolution strain measurements by direct observation in the scanning electron microscope' *J Mater Sci* **23** (1988) pp 3970-3972

3 **Mao, T.H., Beaumont, P.W.R. and Nixon, W.C.** 'Direct observations of crack propagation in brittle materials' *J Mater Sci Lett* **2** (1983) p 616

4 **Roulin-Moloney, A.C., Cantwell, W.J. and Kausch, H.H.** 'Parameters determining the strength and toughness of particulate-filled epoxy resins' *Polym Composites* **8** No 5 (1987) pp 314-323

5 **Lange, F.F. and Radford, K.C.** 'Fracture energy of an epoxy composite system' *J Mater Sci* **6** (1971) pp 1197-1203

6 **Young, R.J. and Beaumont, P.W.R.** 'Failure of brittle polymers by slow crack growth. Part 3. Effect of composition upon the fracture of silica filled epoxy resin composites' *J Mater Sci* **12** (1977) pp 684-692

7 **Spanoudankis, J.** 'Fracture in particle filled epoxy resins' *PhD Thesis* (University of London, UK, 1981)

8 **Lange, F.F.** 'Interaction of a crack with a second phase particle' *Phil Mag* **22** (1970) pp 983-992

9 **Lake, G.J. and Thomas, A.G.** 'Strength of highly elastic materials' *Proc Roy Soc London A* **300** (1967) pp 108-119

10 **King, N.E. and Andrews, E.H.** 'Fracture of energy of epoxy resins above T_g' *J Mater Sci* **13** (1978) pp 1291-1302

11 **Griffiths, J.R. and Owen, D.J.R.** 'Elastic-plastic stress analysis for a notched bar in plane-strain bending' *J Mech Phys Sol* **19** (1971) pp 419-431

12 **Dickson, R.F., Ellis, M.B.D., Hepples, W. and Crompton, J.S.** 'The effect of cure on the failure properties of a rubber toughened epoxy resin' *'Adhesion 90', 10-12 September 1990, Cambridge, UK*

13 **Kinloch, A.J. and Young, R.J.** *Fracture Behaviour of Polymers* (Elsevier Applied Science Publishers, London, UK, 1985)

14 **Riew, C.K., Rowe, E.H. and Siebert, A.R.** in *Toughness and Brittleness of Plastics* edited by R.D. Deanin and A.M. Crugnola (Academic Press, New York, USA, 1978)

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