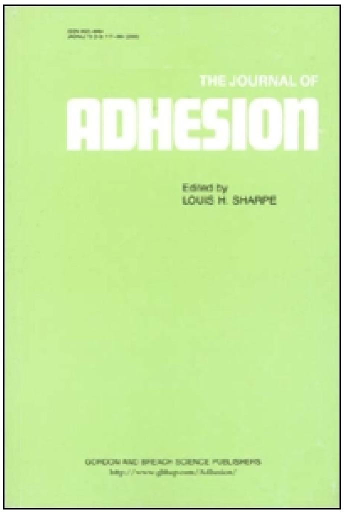

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Observations on Finger-like Crack Growth at a Urethane Acrylate/Glass Interface

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Interfacial crack growth behavior along a urethane acrylate/glass interface is characterized by the development of finger-like perturbations along the advancing crack front. The finger-like perturbations grow from a slightly irregular crack front until they reach a steady-state where the velocity of the finger tips equals the velocity of the finger valleys. Once the fingers reached steady-state, the crack velocity was dependent on the applied strain energy release rate *via* a power law relationship where the exponent was independent of test humidity; however, the multiplicative constant A decreased by an order of magnitude from 80 to 15% RH. The spacing of the fingers was found to be independent of the crack's velocity and the relative humidity of the environment.

KEY WORDS interfacial crack growth; urethane-acrylate; glass; finger-like cracks; viscous adhesive interface; crack growth instability; relative humidity; applied strain energy release rate.

INTRODUCTION

The purpose of the present study was to study finger-like crack growth at a viscous adhesive/glass interface. A four-point flexure apparatus coupled with an inverted optical microscope was used to characterize and measure finger-like crack growth at a urethane acrylate/glass interface as a function of the applied strain energy release rate and relative humidity. With this experimental apparatus the crack will grow stably along the adhesive/glass interface and the rate of growth is directly dependent on the applied strain energy release rate and relative humidity.

Of particular interest in this study is the finger-like profile of the interface crack that grows in a stable fashion after forming. Finger-like surfaces of separation have been observed in a number of other physical systems including interpenetration of fluids,[1,2] peeling of flexible adhesive tape,[3–6] ductile crack growth in glassy polymers[7–9] and intergranular creep fracture in ceramics.[5,10]

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EXPERIMENTAL PROCEDURE

The specimen consists of two cleaned annealed glass plates 40 mm long by 8 mm wide by 2 mm thick glued together with a polymer adhesive, see Figure 1. The glass is cleaned by immersing for 10 minutes in an ultrasonic detergent bath and then hot air dried before adhering. The polymer adhesive used is a urethane acrylate (950-133 Desoto, Inc.). This material is claimed by the manufacturer to have good moisture resistance with the water absorption being 1.1 wt%/24 h and an elastic modulus of 0.26 GPa. This acrylate was cured under ultraviolet light for 24 hours. The exact curing procedures followed the manufacturer's recommendations. The edges of the specimens were polished (Buehler LTD, standard grinder/polisher) to eliminate fillets of polymer on the edges that could affect the crack growth behavior.

A four-point flexure test was used to study crack growth at the interface between the adhesive and glass.[11,12] Figure 2 shows a schematic of the bend flexure test apparatus that was constructed to fit on the stage of an inverted optical microscope (Zeiss IM35). The inner and outer spans are 22.225 and 31.750 mm, respectively. A button load cell (Entran ELF-1000 Flatline) was used in conjunction with a multimeter (Keithley 175) to record the load, P, being applied to the specimen. The load was applied by means of turning a micrometer. This apparatus was used both to precrack the specimen and to measure and characterize crack growth as a function of time at a given applied strain energy release rate. Crack growth measurements were made as a function of humidity at various applied loads by constructing a plastic envelope around the fixture. The high relative humidity (80%) was achieved by flowing of nitrogen gas through a water bath and piping it into the plastic envelope. The low relative humidity (15%) was achieved by piping the nitrogen gas directly into the plastic envelope. Tests were also done in ambient air (60% RH). The humidities were measured with a hygrometer (Hanna Instruments HI 88064).

The specimens were precracked by first placing an array of Vickers indents along the width of the top layer of the glass. An indent load of 15 N was chosen by initial experimentation, since an excessive load would produce an interfacial crack too large to allow for subsequent crack measurements and too small a load would not given an observable interfacial crack. The indented specimens were placed in the

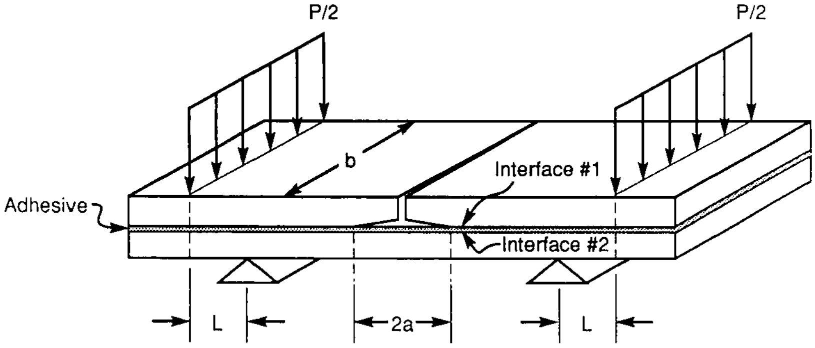

FIGURE 1 Schematic of the four-point flexure specimen.

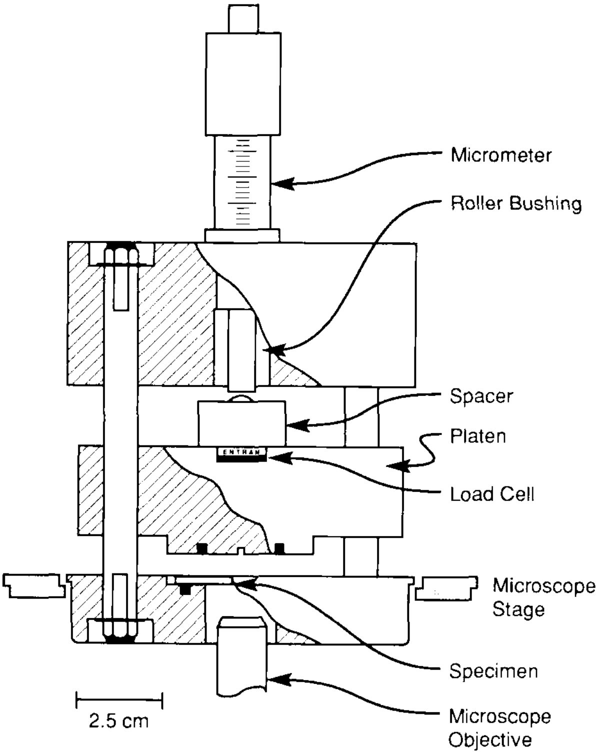

FIGURE 2 Schematic of the four-point flexure apparatus.

apparatus with the indented surface on the tensile side. Upon slowly loading the specimen in three-point bending, a crack propagated downward from the array of indents and then branched symmetrically into interface #1 before arresting.

Before crack growth testing, the samples were preconditioned at the test humidity by storing them in a bell jar with a saturated aqueous salt solution to give the test relative humidity of 15 or 80%. The lower humidity was obtained by using an aqueous Potassium Acetate solution and for the 80% RH an aqueous Potassium Chloride solution was used.[13] The pre-conditioned specimens were then placed in four-point bending with the precracked side of the specimen in tension and the interfacial crack tips within the inner loading span. The interfacial crack produced an interference pattern due to the gap separating the crack surfaces that allowed it to be easily observed. The crack length was measured from the line of indents to the leading edge of one of the two crack tips. The applied strain energy release rate, G, is related to the applied load, P, by:[11,12]

$$G = \frac{(PL)^2}{8Eb}\left[\frac{1}{I_c} - \frac{1}{I_2}\right] \qquad (1)$$

where L is the distance between inner and outer loading points, E is the elastic modulus of glass, I_2 and I_c are the moment of inertias of the lower glass plate and the composite sample, respectively, and b is the width of the specimen. With this specimen, the phase angle of loading (a measure of the ratio of the shear stress to tensile stress at the crack tip) is approximately 45–50°, indicating that the shear to tensile stress ratio is about one.[11,12] Since G is constant within the inner loading span and is independent of crack length, crack growth could be measured as a function of constant applied G, time and relative humidity.

RESULTS AND DISCUSSION

After precracking, the interfacial crack front at the urethane acrylate/glass interface often contained several small irregular perturbations. On subsequent crack growth at a constant applied G, this slightly irregular front would develop well-defined fingers with a given periodicity, *i.e.* finger spacing. It is important to realize that this finger-like crack front would develop independently of the initial, irregular perturbations. For example, precracking by scoring the glass rather than indenting it caused a relatively smooth initial crack front to form; however, under the same test conditions the finger-like pattern that developed was similar to that resulting from the more irregular interface caused by indenting. Figure 3 shows the development of the finger-like crack front at an applied G of 2.4 J/m^2 in ambient air (60% RH) and Figure 4 shows the development at 1.5 J/m^2 in 15% RH. During this developmental stage, two fingers would sometimes grow into one (see third and fourth finger from bottom of Fig. 3c and 3d) while at other times a finger would bifurcate (see Fig. 4). Also, a void occasionally would appear ahead of a growing finger (Fig. 3d). After the fingers became fully developed, the crack front would grow linearly with time and the finger length and periodicity would remain constant. Figure 5 summarizes the crack length data, as measured at the finger tips and valleys, for the experiments shown in Figures 3 and 4. Note that the steady state velocity for the finger growth data is given.

Although the above observations on the finger-like crack growth at the urethane acrylate/glass interface are similar to peeling of adhesive tape,[3–6] there are some important differences. In the peeling of adhesive tape, the finger-like crack growth occurs within the adhesive layer and not at the interface. Also, separation of the adhesive from the substrate generally occurs under a constant peeling rate; consequently, the driving force for this crack/void growth is not constant during the experiment. In addition, the exact nature of the finger-like void growth is dependent on the peeling rate.[4,6] For example, Reference 4 found that finger-like growth was observed for peeling if the product of the peeling rate and viscosity of the adhesive was high; conversely, if this product was low, a straight crack front was produced. In the experiments described here, finger-like crack growth is occurring stably under a constant driving force, G, at the adhesive/glass interface. Also, in the present study it can be estimated from the birefringence pattern seen in Figures 3 and 4 that the crack opening displacement at the interface is approximately 0.3 μm. Separation displacements in the peel test are much greater than this.

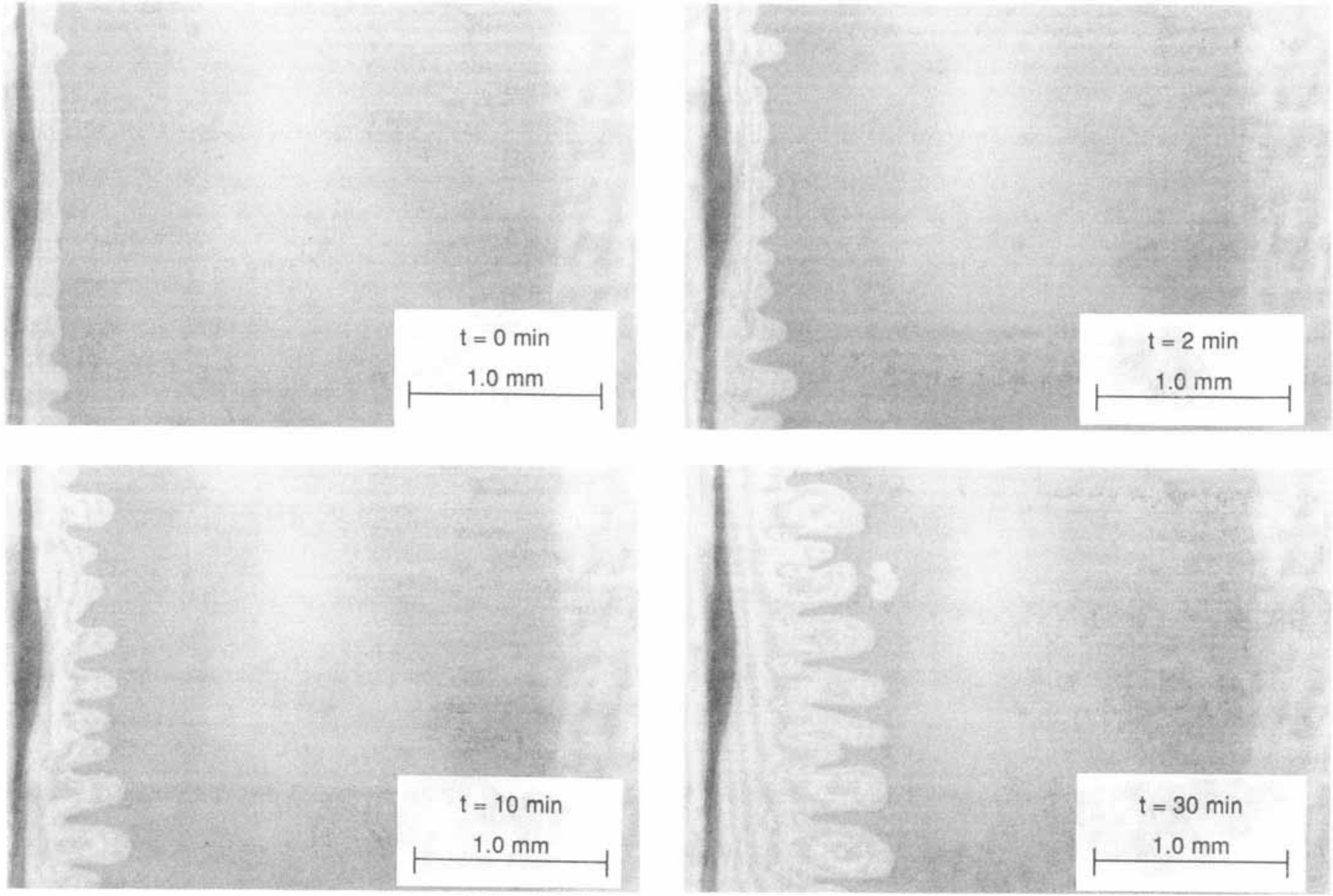

FIGURE 3 Sequence of micrographs showing the development of fingers in a urethane acrylate/glass interface under an applied G of 2.4 J/m^2 and at 60% RH.

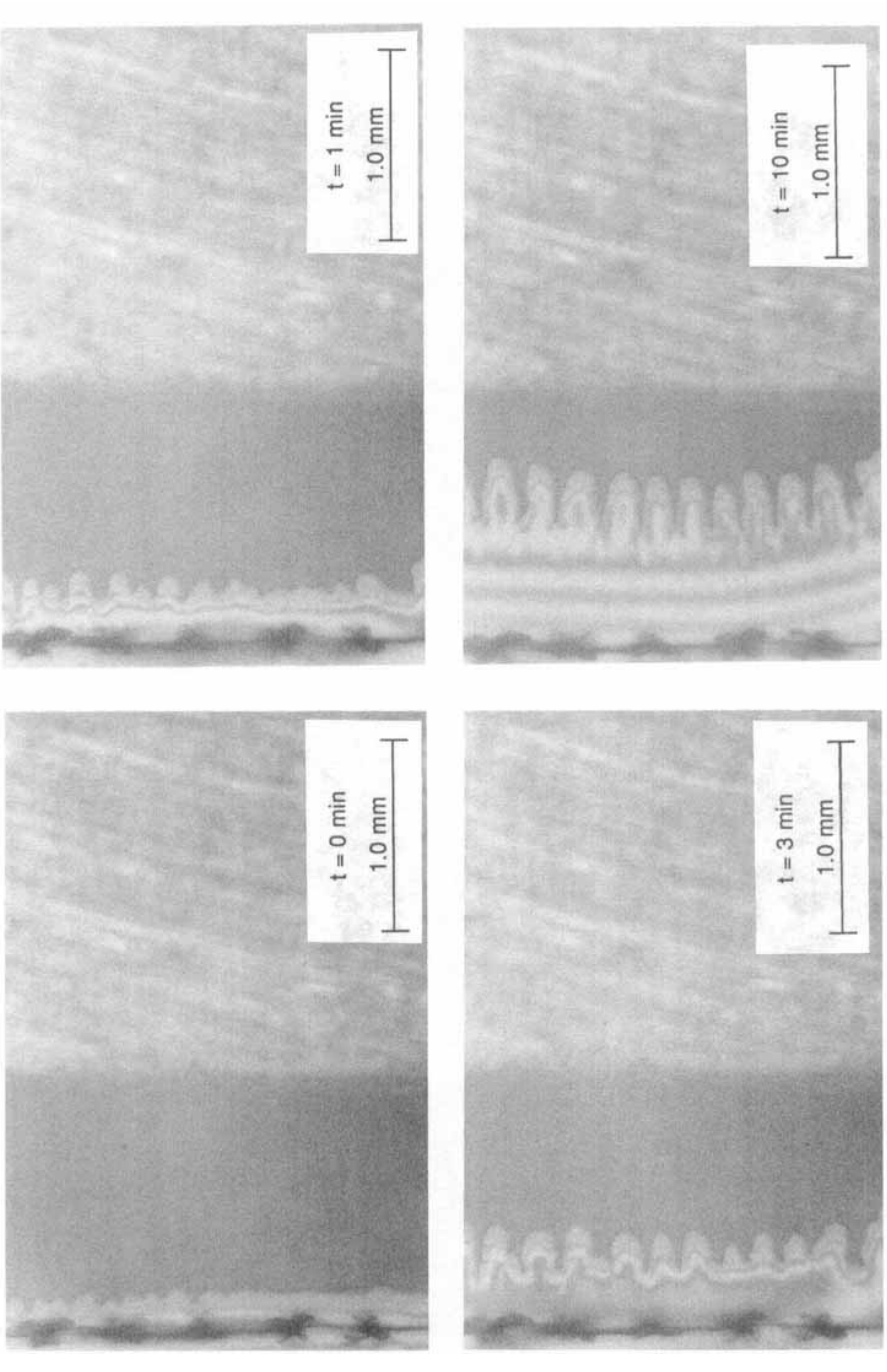

FIGURE 4 Sequence of micrographs showing the development of fingers in a urethane acrylate/glass interface under an applied G of 0.72 J/m^2 and at 80% RH.

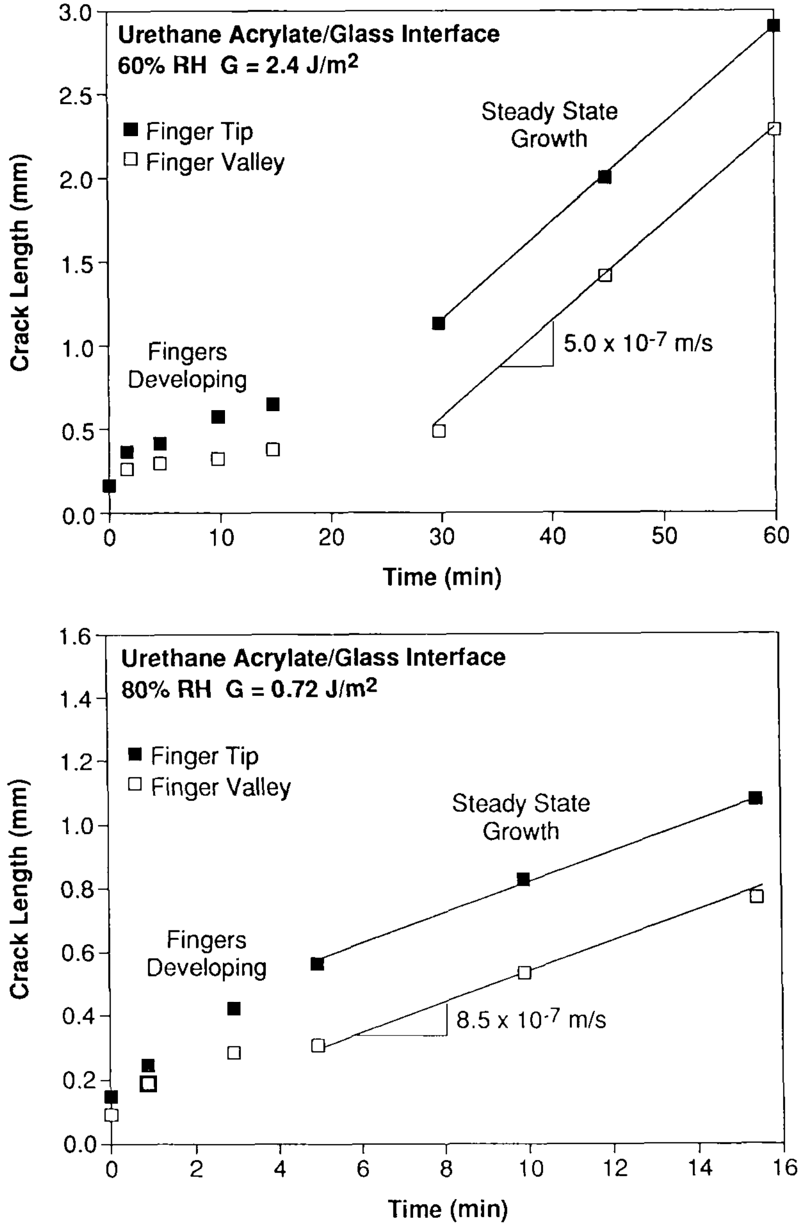

FIGURE 5 Crack length *versus* time graph of the sample with developing fingers shown in (a) Fig. 3 and (b) Fig. 4.

The steady state velocity for this finger-like crack growth in the urethane acrylate/glass interface increased with increasing applied G at constant RH and was greater in the high humidity test environment. Figure 6 compares crack growth at 80 and 15% RH for a G = 1.4 J/m^2. It is evident that the growth rate is greater in

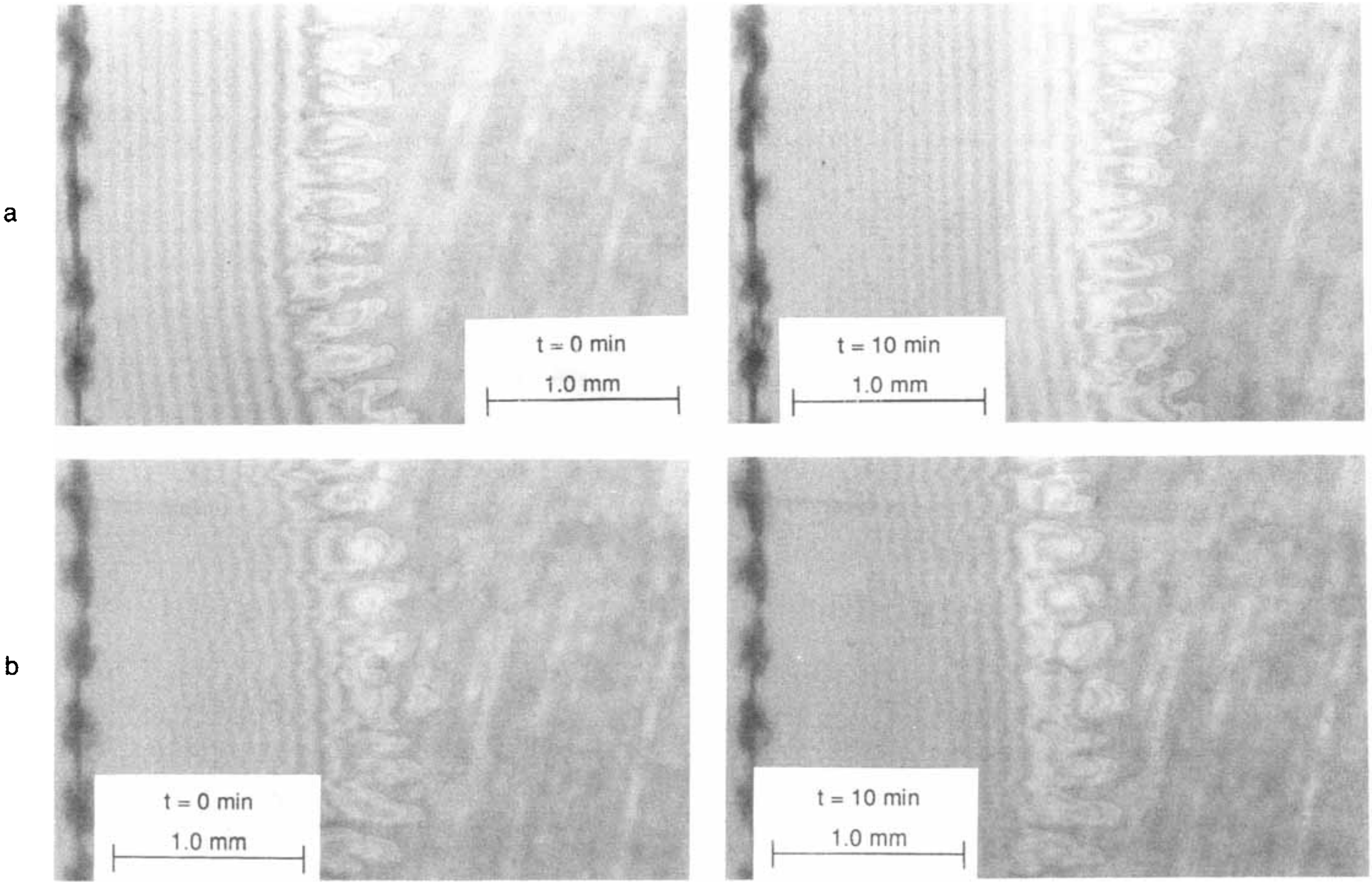

FIGURE 6 Micrographs of crack growth in the urethane acrylate/glass interface at (a) 80% RH and (b) 15% RH under a G of 1.4 J/m^2. Note that time is relative to when steady-state growth is obtained.

the higher humidity. Figure 7 shows a log-log plot of the growth rate (v) data as a function of the applied G. Linear aggression of the data gave

$$80\%\ \mathrm{RH} \qquad v = (1.76 \times 10^{-6})\, G^{2.5}$$

$$15\%\ \mathrm{RH} \qquad v = (1.23 \times 10^{-7})\, G^{2.5}$$

Note that since the slope of the data in Figure 7 were not significantly affected by humidity, the data were regressed to a common slope (2.5). From these results it can be seen that the finger-like crack growth rate was about an order of magnitude greater in the 80% RH environment as compared with 15% RH.

The finger spacing for steady crack growth was not a function of either the test humidity or the applied G, *i.e.* the spacing was independent of the crack growth rate. Figure 8 shows that the measured finger spacing was 0.29 (±0.06) mm.

This moisture-assisted, finger-like crack growth at the urethane acrylate/glass interface contrasts sharply with crack growth at an epoxy acrylate/glass interface studied previously in our laboratory.[14] Cracks in the epoxy acrylate/glass interface grow with a relatively straight, smooth crack front. Figure 9 illustrates this for a crack growing in an epoxy acrylate/glass interface at an applied G of 2.2 J/m^2 at 80% RH. This figure shows that the crack front grows quite uniformly with no localized section of the crack front growing faster or slower then the rest.

The mechanical properties of the urethane acrylate adhesive differ in several important ways from that of the epoxy acrylate. The urethane acrylate has a lower elastic modulus (260 *vs.* 655 MPa) and lower hardness (25 *vs.* 140 MPa).[15] Moreover, the hardness indent in the urethane acrylate tends to disappear within 30 minutes

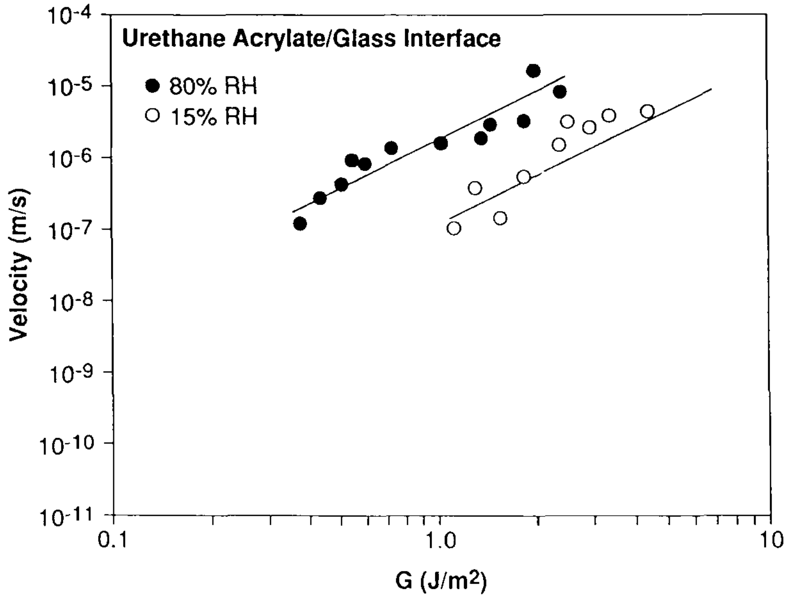

FIGURE 7 Log-log plot of velocity *versus* G for the urethane acrylate/glass interface.

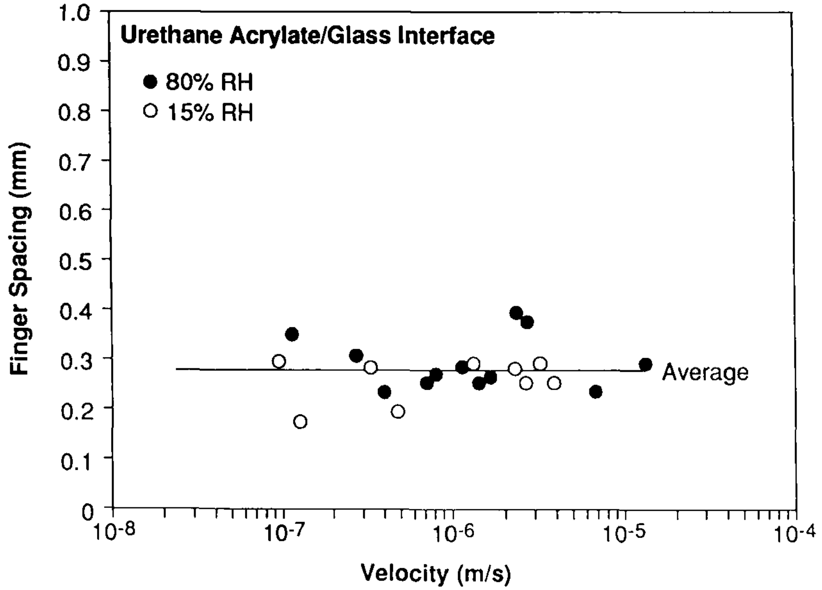

FIGURE 8 Graph of finger spacing *versus* velocity for the urethane acrylate/glass system.

due to viscoelastic recovery and under a constant indent load the urethane acrylate material will creep.[15] Preliminary results showed that indentation creep at 80% RH was greater than at 15% RH. Although these results demonstrate that the adhesive must be viscous to cause finger-like crack growth, an exact relationship between the viscous properties and the parameters influencing finger-like crack growth is not known at present. Both References 3 and 5 have proposed a model for finger-like crack growth in the peeling of flexible adhesive tape; however, their models are only applicable to cracks growing entirely within the viscous adhesive layer. In the research described here, finger-like crack growth occurs in the urethane acrylate/glass interface so that both the properties of the viscous urethane acrylate and the interface should be important in any model. In this regard it should be noted that the critical stress energy release rate, *i.e.* interfacial fracture energy, G_c, was measured in four-point flexure for fast, catastrophic crack propagation by loading a urethane acrylate/glass sample quickly up to the point at which the interfacial crack propagated spontaneously. At 80% RH, G_c was 8.3 J/m^2 and at 15% RH, G_c was 13.8 J/m^2. It is important to note that even at this fast catastrophic crack propagation, the crack front contained finger-like perturbations, similar to that observed at the low, stable crack velocities.

SUMMARY

Finger-like crack growth behavior at a urethane acrylate/glass interface was studied as a function of humidity and applied strain energy release rate. A four-point flexure apparatus coupled with an inverted microscope allowed for observation in-situ of

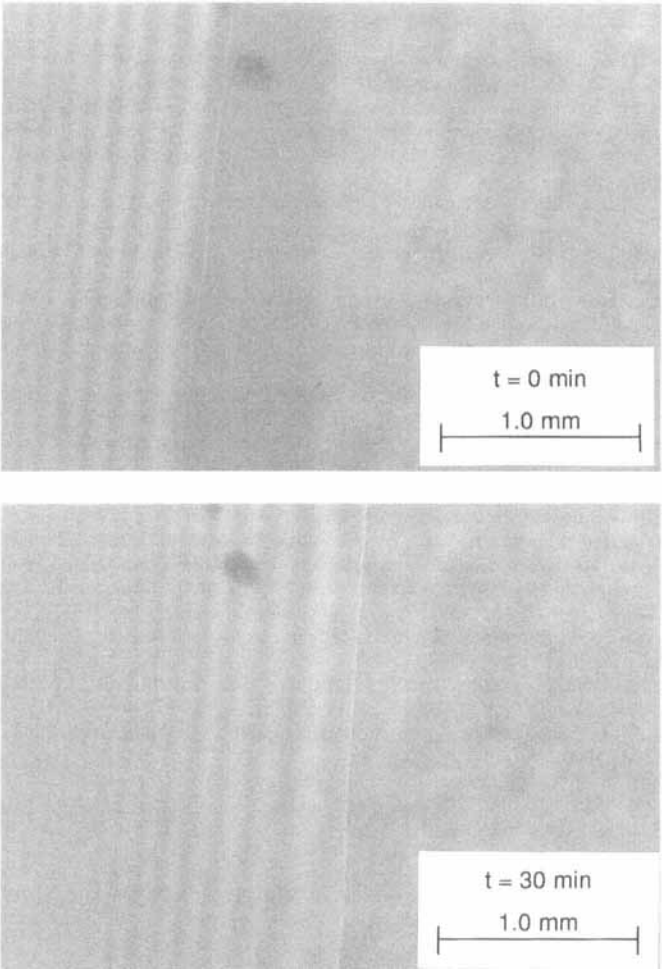

FIGURE 9 Micrographs of a smooth interface crack front in the epoxy acrylate/glass interface under an applied G of 2.2 J/m^2 and at 80% RH.

the crack growth at this polymer adhesive/glass interface. The four-point flexure specimen consisted of two glass plates bonded together with a crack introduced at the adhesive/glass interface. Interfacial crack growth was accompanied by the development of well-defined fingers with a given periodicity. Once the fingers reached steady-state, crack velocity was dependent on the strain energy release rate *via* a power law function where the proportionality constant was sensitive to test humidity but the exponent was not. Finger spacing was independent of crack velocity.

Acknowledgement

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