

Time-dependent fracture behavior of acrylic resins

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An investigation was conducted into the effects of time, temperature, and different toughening mechanisms on plain and toughened polymer samples.

Acrylic resins are used in several modern applications (such as bone cement in orthopedics and dentures) because of their desirable properties, i.e., they are stiff, transparent, and have good weathering properties. Furthermore, these polymers have recently found use as matrices in continuous fiber composite materials (produced via reactive processing methods). However, despite their advantageous characteristics, acrylic resins generally exhibit brittle behavior.

To improve the fracture toughness of acrylic resins, rubber inclusions are commonly added to the polymer matrix.^{1,2} It is important, however, to consider the viscoelastic nature (i.e., the time dependence of the elastic properties) of the materials when designing such composites for structural applications. To that end, several theories have previously been proposed to explain the fracture mechanics of viscoelastic materials. For example, linear elastic fracture mechanics have been applied to viscoelastic materials to predict the relationship between fracture toughness and the time of crack onset, or between fracture toughness and crack propagation speed.^{3,4}

To experimentally test the existing fracture behavior theories, we have studied the fracture toughness behavior of two thermoplastic acrylic resins.⁵ We obtained both the resins—Elium (plain) and Elium Impact (toughened with about 10wt% of an acrylic block copolymer)—from Arkema. We then prepared the specimens for our mechanical tests by casting the monomers into a closed mold. We left the Elium samples to polymerize for 24 hours at room temperature and then applied a thermal treatment at 80 and 120°C (both for 1 hour) to complete the polymerization. For the Elium Impact samples, we used a thermal cycle of 5 hours at 80°C and 1.5 hours at 125°C to perform the polymerization.

For the first mechanical measurements, we conducted double torsion tests^{6,7} at different temperatures. This test allows the speed of crack propagation to be controlled and is thus suitable for studying the rate dependence of fracture toughness. The results of these tests—fracture

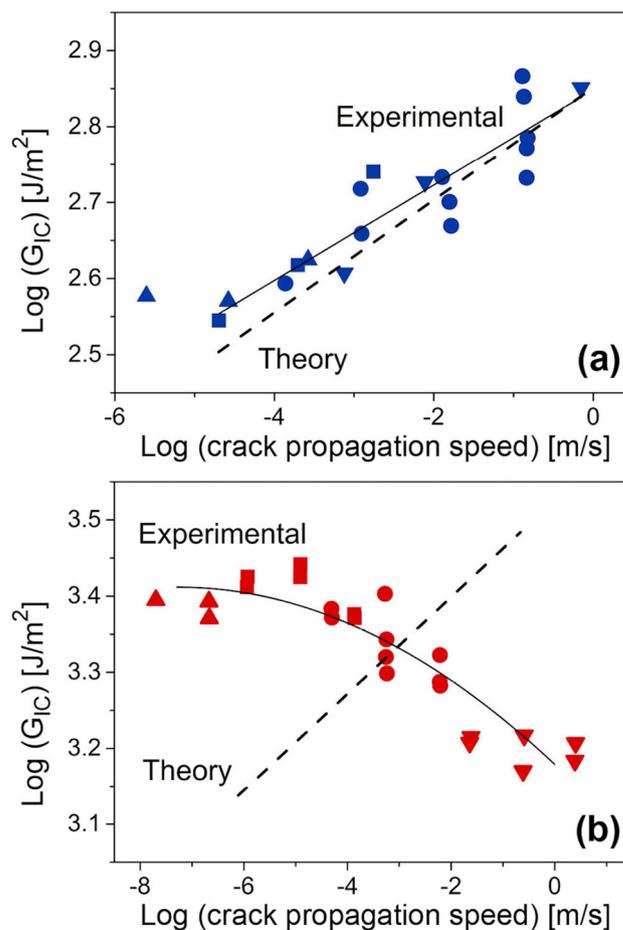


Figure 1. Fracture toughness (G_{IC}) as a function of crack propagation speed (both on a log scale and for a reference temperature of 23°C) for (a) the plain resin and (b) the toughened resin. Experimental results are given by the colored symbols, and the dotted lines mark the predicted theoretical trends.

toughness as a function of crack propagation speed—are shown in Figure 1 for both of our acrylic resin samples. In addition, by applying the time-temperature equivalence, we obtained master curves

Continued on next page

for our experimental data. We find, as expected, that the toughened resin exhibited higher levels of fracture toughness. Furthermore, for the plain resin—see Figure 1(a)—we observe that the fracture toughness increased with increasing crack speed. This result is in good agreement with the theoretical viscoelastic fracture predictions,³ i.e., that are based on the hypothesis that the crack tip opening displacement (CTOD) is constant, irrespective of time and temperature. For the toughened resin, however, the trend of our experimental results—see Figure 1(b)—clearly does not agree with the theoretical predictions.

To investigate the difference between the predicted and measured behavior of the toughened resin, we further studied the fracture processes that occurred at the crack tip. Specifically, we performed digital image correlation analyses during single-edge notched bending tests to measure the CTOD. Our results (see Figure 2) show that the CTOD remained constant with increasing temperature for the plain resin, whereas it increased with temperature for the toughened sample. We hypothesize that this CTOD variation arises because of a change in the damage mechanism that occurs at the crack tip. Indeed, it has previously been reported that two main failure mechanisms—multiple crazing and cavitation of the rubbery phase—can occur in toughened polymers.^{8,9} The latter mechanism facilitates shear yielding of the matrix and is associated with higher levels of energy dissipation.

To identify which of the two possible damage mechanisms occurred in our toughened resin, we also measured the volume change of the samples during tensile tests (under different loading conditions). Based on the previous studies,^{8,9} it is known that high volume changes occur during crazing. In contrast, cavitation causes only a limited volume

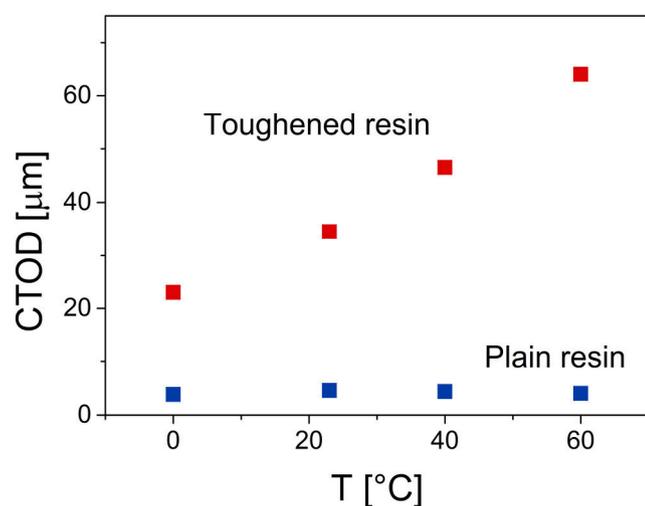


Figure 2. Crack tip opening displacement (CTOD) measured at different temperatures (T) for the plain resin (blue) and the toughened resin (red).

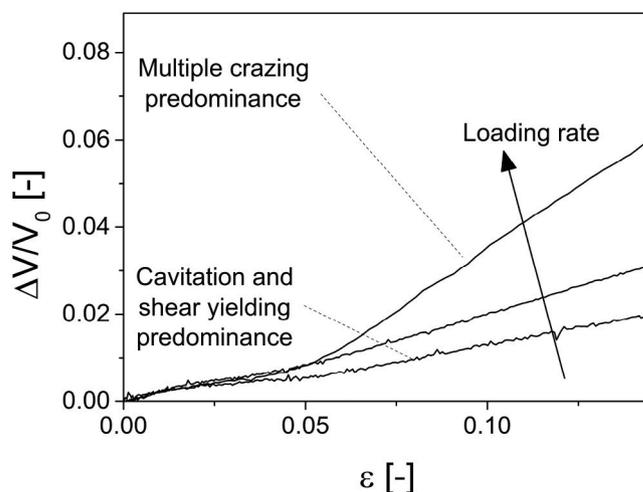


Figure 3. Experimentally measured volume change (ΔV) of the toughened resin as a function of strain (ϵ) for three different loading rates. V_0 : Initial volume.

change. Our results (see Figure 3) indicate that at slow strain speeds (i.e., low crack speeds), a major contribution to toughness is provided by the cavitation of the rubbery domains and shear yielding. In addition, we find that multiple crazing tends to occur at higher crack speeds. Our observations can thus explain the results of Figure 1(b), i.e., where the fracture toughness of the toughened resin was higher at lower crack propagation speeds.

In summary, we have investigated the time-dependent fracture toughness behavior of a plain and a toughened acrylic resin. On the basis of our experimental results, we find that the fracture characteristics of the plain resin are governed by the viscoelastic properties of the bulk material. In contrast, for the toughened resin, our results indicate that the time dependence of the fracture toughness is determined by changes in the damage mechanism that occurs at the crack tip (i.e., under different loading conditions). In our ongoing work, we are using transmission electron microscopy to further investigate the changes in volume that occur in the toughened resin at different rates and temperatures.

The authors thank the Arkema Company and the Groupement de recherches de Lacq (France) for providing materials and support, as well as Gerald Pinter, Steffen Stelzer, and staff at Montanuniversität Leoben (Austria) for their help with this work.

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