Effect of Displacement Rate on the Mode I Fracture Behavior of Rubber Toughened PMMA*

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Mode I fracture behavior of rubber toughened PMMA (RT-PMMA) was studied over five decades of displacement rates (10^{-4} –13 m/s) using a high speed tensile testing machine. Two kinds of RT-PMMA, extruded RT-PMMA containing 20 wt.% rubber particles (EM20) and pressed RT-PMMA containing 40 wt.% rubber particles (PM40), exhibited different rate dependencies of the maximum mode I stress intensity factor, $K_{I\max}$, and the maximum mode I energy release rate, $G_{I\max}$. $K_{I\max}$ and $G_{I\max}$ of EM20 decreased rapidly at high rates (1–11 m/s). This suggests that the toughening effect due to rubber blending was dramatically reduced at the high rates. PM40, on the other hand, retained high $K_{I\max}$ and $G_{I\max}$ values at all the rates tested, thus, maintaining the toughening effect even at a rate of up to 13 m/s. Neat PMMA was also examined, for comparison, and its $K_{I\max}$ was found to increase with increase in the displacement rates while $G_{I\max}$ remained constant. The rate-dependent $K_{I\max}$ and $G_{I\max}$ values are discussed, taking $(K_{I\max})^2/G_{I\max}$ into account, and the fracture surface morphology of the specimens is also considered.

Key Words: High Polymer Materials, Stress Intensity Factor, Crack Propagation, Fractography, Rubber Toughened PMMA, Toughening Mechanism, Rate Dependence, High-Speed Tensile Testing

1. Introduction

A transparent thermoplastic poly (methyl methacrylate) (PMMA) is in wide use in the automobile, electric, optical and architectural industries. The greater is the spread of PMMA, the higher are the mechanical properties demanded; impact resistance is one of the most desired properties for such engineering plastics. To increase the impact resistance of this material without sacrificing its transparency, different types of rubber toughened PMMA (RT-PMMA) have been developed by blending rubbery phases with PMMA matrices, and attempts made to characterize

their mechanical properties.

The effect of loading rate on the fracture behavior of RT-PMMA and neat PMMA have been investigated^{(1)–(5)}. For example, by comparing a quasi-static fracture test result with an impact test result obtained using an instrumented drop weight testing apparatus, Hooley et al. found that the dynamic fracture toughness of RT-PMMA was higher than the static toughness⁽¹⁾. Goolsby and Lin found a similar result for a neat PMMA⁽²⁾. Sahraoui and Lataillade compared a quasi-static result with dynamic test results obtained using a Sharp impact tester with a gas gun and a Hopkinson bar type tensile testing machine, and showed that the fracture toughness of PMMA increased with increase in strain rate. Our research group recently found that if indirectly measured load data was used to evaluate the dynamic fracture toughness, K_{Id} , of polycarbonate (PC), K_{Id} tended to increase with increase of loading rate⁽⁶⁾; however, if directly measured loading-point displacement was used for the analysis, K_{Id} of PC actually decreased as loading rates increased. Thus, dynamic

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effects appearing on the measured load data can possibly lead to erroneous interpretation. It is therefore difficult to determine the actual loading-rate dependence of the fracture properties using a standard impact testing system in which load is measured indirectly at the loading dart. To address this, Béguelin and Kausch developed a dynamic testing system consisting of a modified servohydraulic testing machine and a self-made dynamic displacement measuring apparatus^{(4),(5),(7),(8)}. They applied the system to the study of loading-rate dependencies of the fracture toughness and mechanical properties of polymers and composite materials from quasi-static rates to the maximum rate of 14 m/s.

In this study, the effects of loading rate on the mode I fracture behavior of two kinds of RT-PMMA's and two different neat PMMA's were examined using the dynamic testing system developed by Béguelin and Kausch at testing velocities from 1×10^{-4} to 13 m/s. Maximum mode I stress intensity factor and maximum mode I energy release rate were the fracture mechanics properties evaluated, and their loading rate dependence was determined.

2. Experimental

2.1 Materials

Two types of RT-PMMA were studied: one was manufactured by extrusion and rubber particles were blended with a PMMA matrix in a weight fraction of 20 wt.%, and the other was fabricated by press and the weight fraction of rubber particles was 40 wt.%. These materials are hereafter denoted as EM20 (extruded modified, 20 wt.%) and PM40 (press modified, 40 wt.%), respectively. Each rubber particle has a three-layer structure in which a PMMA core is surrounded by an inner shell of crosslinked butyl acrylate-co-styrene with an outer shell of grafted PMMA. The diameter of the rubber particles is approximately 300 nm. Field emission scanning transmission electron microscope (FE-STEM) was used to identify the microstructures of the two materials.

Two types of neat PMMA, extruded and cast materials, were also examined for comparison; their molecular weights were $M_w = 1.5 \times 10^5$ and 1.6×10^6 , respectively. These PMMA's are hereafter denoted as EU (extruded unmodified) and CU (cast unmodified), respectively.

2.2 Specimens

Compact tension (CT) specimens were prepared for mode I fracture testing. The CT specimen geometry is shown in Fig. 1. Specimens were cut out from molding plates 5 mm thick. A sharp notch was machined for each specimen so that for the extruded materials (EM20 and EU) the notch direction coincided

Fig. 1 CT specimen geometry (width W : 12 mm, thickness B : 5 mm, crack length a :)

ed with the direction of extrusion. For each of the neat PMMA specimens, a new razor blade was tapped into the machined notch to introduce a natural starter crack. For each RT-PMMA specimen, a prenotch was carefully introduced by drawing a new razor blade along the front of the machined notch.

2.3 Testing apparatus

Mode I fracture testing was carried out at a wide range of displacement rates from 10^{-4} to 13 m/s using a modified servohydraulic high speed testing machine originally manufactured by Schenck. A piezoelectric load cell, designed to reduce the inertial effect of the fixture assembly, was used to measure the load with high sensitivity. Crack opening displacement was measured by an optical device assembled using a laser light beam with an optical fiber⁽⁷⁾.

Béguelin and Kausch successfully employed a mechanical assembly, called a pick-up unit, as the link between the specimen fixture and the accelerating piston, to avoid strain induced in the attached specimen during the acceleration of the servohydraulic actuator at high displacement rates (> 0.5 m/s)^{(4),(8)}. In this pick-up unit, force is applied to the specimen by striking the specimen fixture with the piston accelerated to the desired velocity; this usually results in generation of a mechanical noise due to the impact at the high testing rates. This inertial effect can be reduced by decreasing the contact stiffness using a viscoelastic damper in the pick-up unit. In this study, viscoelastic dampers were used for all the tests performed at displacement rates greater than 1 m/s. Assuming that a quasi-static stress state was achieved even at those high rates, static fracture mechanics was applied to the analysis of the mode I fracture properties.

2.4 Quantitative analysis of mode I fracture properties

The mode I stress intensity factor, K_I , and the mode I energy release rate, G_I , were evaluated to study the effects of displacement rates on the mode I fracture properties quantitatively. K_I for a CT specimen can be expressed as⁽⁹⁾

$$K_I = f(x) \frac{P}{B\sqrt{W}}, \quad x = a/W \quad (1)$$

$$f(x) = [(2+x)(0.886 + 4.64x - 13.32x^2 + 14.72x^3 - 5.6x^4)] / (1-x)^{3/2}$$

where f is a calibration factor depending upon the specimen geometry and crack length, a , and P the applied load. B and W are the thickness and the depth of the CT specimen, respectively, as shown in Fig. 1. G_I is given by⁽⁹⁾

$$G_I = \frac{U}{BW\phi} \quad (2)$$

$$\phi = \frac{(1.9118 + 19.118x - 2.5122x^2 - 23.226x^3 + 20.54x^4)(1-x)}{(19.118 - 5.0244x - 69.678x^2 + 82.16x^3)(1-x) + 2(1.9118 + 19.118x - 2.5122x^2 - 23.226x^3 + 20.54x^4)}$$

where U is strain energy and can be estimated as the area under measured load-displacement curve; ϕ is the energy calibration factor. The maximum mode I stress intensity factor, $K_{I\max}$, and the maximum mode I strain energy release rate, $G_{I\max}$, corresponding to the maximum load, $P_{\max}$, were evaluated. $P_{\max}$ was obtained as the maximum value of the load from each of the load-displacement curves, and $U_{\max}$ was calculated by numerically integrating the load-displacement curve up to $P_{\max}$. It is noted that for the neat PMMA specimens exhibiting brittle fracture, the maximum point of load coincided with the initiation point of crack growth; therefore, $K_{I\max}$ and $G_{I\max}$ could be considered as the mode I fracture toughness K_{IC} and G_{IC} , respectively.

3. Results and Discussion

3.1 Microstructure of RT-PMMA

FE-STEM photographs of EM20 and PM40 are shown in Figs. 2(a) and (b), respectively. For these observations samples about 40 nm thick were prepared using a ultra-microtome. The parallel lines leaning to the left in the photos were scratches made by the diamond knife, and the rubber particles were deformed in an elliptical shape due to the compressive force in the cutting direction. It can be seen that the rubber particles were distributed uniformly in the PMMA matrices for both materials, and the sizes of the particles were almost equal. It is also observed that EM20 and PM40 contain rubber particles of the same structure, and the white regions giving the outlines of the particles are the rubbery phases, that is, crosslinked butyl acrylate-co-styrene.

3.2 Displacement-rate dependence of load-displacement curves

Load-displacement curves obtained at three different displacement rates are shown in Fig. 3. For CU, EU and PM40, the maximum load, $P_{\max}$, increased with increase in rate, while EM20 exhibited a decrease of $P_{\max}$ after initially increasing with the rate, as a

result, $P_{\max}$ at the highest rate (10 m/s) was equivalent to that at the lowest rate (1.1×10^{-4} m/s). For all the materials, the compliances as evaluated from the initial linear parts of the curves tended to decrease with increase in rate. For the two RT-PMMA's the decreasing behaviors of the load after $P_{\max}$ differed: for EM20 the load rapidly decreased at high rates, while for PM40 it decreased gradually at all rates. This difference is closely related to the various behaviors of crack growth described in detail in paragraph 3.5.

Oscillation as an effect of impact was observed in the beginning of the load-displacement curve at 10 m/s for all the materials. This dynamic effect was

(a) EM20

(b) PM40

Fig. 2 Microstructures of RT-PMMA

Fig. 3 Load-displacement curves at different displacement rates

observed in all the results at rates greater than 3 m/s, although a viscoelastic damper was used in each of the tests. This effect was assumed to be negligible in the following analysis.

3.3 Displacement-rate dependence of mode I fracture properties

Displacement-rate dependence of the maximum Mode I stress intensity factor, $K_{I\max}$, is shown in Fig. 4. For the two neat PMMA's $K_{I\max}$ slightly increased with increase of displacement rates. Béguelin et al. reported that K_{Ic} of a PMMA of low molecular weight (1.3×10^5) increased with rise in loading rate, while K_{Ic} of a PMMA of high molecular weight (4.0×10^6) depended little on loading rate^{(4),(5)}. The molecular weights of the present PMMA's were lower: 1.5×10^5 for EU and 1.6×10^6 for CU. Thus, dependence of K_{Ic} of a neat PMMA on displacement rate appears to be closely related to the molecular weight, although detail of the mechanisms remains to be clarified.

The two RT-PMMA's, EM20 and PM40, exhibited different displacement-rate dependencies of $K_{I\max}$ as shown in Fig. 4. $K_{I\max}$ of PM40 increased to 2 m/s, and then slightly decreased to 10 m/s. $K_{I\max}$ of EM20

Fig. 4 Effect of displacement rate on maximum mode I stress intensity factor $K_{I\max}$

increased with increase in rate up to 1 m/s, and at the higher rates (>1 m/s) rapidly decreased. This suggests that the toughening effect of EM20 achieved through the rubber modification is suddenly reduced at a rate greater than 1 m/s. It is also noted that $K_{I\max}$ of EM20 at the highest rate was almost equivalent to that of the neat PMMA's.

Displacement-rate dependence of the maximum

Fig. 5 Effect of displacement rate on maximum mode I energy release rate $G_{I \max}$

mode I energy release rate, $G_{I \max}$, is shown in Fig. 5. In both EM20 and EM40, $G_{I \max}$ was almost constant at rates up to 1 m/s, and increased above that (>1 m/s). This increase is believed due to the effect of kinematic energy described in the following paragraph. The two RT-PMMA's showed slight increases of $G_{I \max}$ up to 0.1 m/s and different rate dependencies at rates greater than 0.1 m/s: $G_{I \max}$ of EM20 rapidly decreased at high rates (>0.1 m/s), while $G_{I \max}$ of PM40 increased up to 8 m/s and then decreased at the higher rates (>8 m/s). It is concluded that EM20 having a lower content of rubber particles tends to lose the toughening effect at a high rate greater than 1 m/s, while PM40 maintains the effect at a rate up to 13 m/s. Toughness of PM40 is also thought to decrease at rates greater than 13 m/s. Fracture testing of PM40 at these higher rates should be done to clarify this.

Difference of the rate dependent behaviors between $K_{I \max}$ and $G_{I \max}$ is discussed in the following paragraph.

3.4 Displacement-rate dependence of $(K_{I \max})^2/G_{I \max}$

If linear fracture mechanics is applicable, the following relation between K_I and G_I holds:

$$K_I^2/G_I = E^* \quad (3)$$

where $E^* = E/(1-\nu^2)$ for plane strain and $E^* = E$ for plane stress and E is the Young's modulus and ν Poisson's ratio. In general, for neat PMMA exhibiting brittle fracture Eq. (3) approximately holds, although it may not be directly applied to the present RT-PMMA's exhibiting relatively ductile fracture. Therefore, the values of $(K_{I \max})^2/G_{I \max}$ were used in conjunction with Figs. 4 and 5 only to see the tendency of the rate dependence of the modulus.

Displacement-rate dependencies of $(K_{I \max})^2/G_{I \max}$ are shown in Fig. 6. For the neat PMMA's $(K_{I \max})^2/G_{I \max}$ increased with rise in rate up to 1 m/s. This seems reasonable, since it is well known that the moduli of glassy polymers including PMMA generally increase

Fig. 6 Dependence of $(K_{I \max})^2/G_{I \max}$ on displacement rate

with increase of strain rates because of their viscoelastic nature. However, $(K_{I \max})^2/G_{I \max}$ suddenly decreased at rates greater than 1 m/s. The primary reason for this behavior appears to be the increase of $G_{I \max}$ at rates higher than 1 m/s shown in Fig. 5. This increase might be caused by an overestimation of $G_{I \max}$ which implies that the measured $U_{\max}$ contained kinematic energy in addition to strain energy. It is understood that the contribution of the kinematic energy cannot be negligible under a high speed tensile testing because of the high speed deformation of the specimen.

$(K_{I \max})^2/G_{I \max}$ of EM20 increased with increase in rate up to about 5 m/s and then suddenly decreased above that, while PM40 showed a monotonic increase of $(K_{I \max})^2/G_{I \max}$ as the rate increased. It is thus thought that the modulus of RT-PMMA generally tends to increase with increase of displacement rate. The sudden decrease observed in EM20 appears to be associated with the sudden decrease of $K_{I \max}$ shown in Fig. 4. The main toughening mechanisms of RT-PMMA are generally believed to be the cavitation of rubber particles and microcrazes generated around the particles in the vicinity of the crack tip at a fracture initiation⁽¹⁰⁾. The rapidly decreasing $K_{I \max}$ of EM20 at high rates was likely due to the reduction of such toughening effects. On the basis of these observations, it is understood that the initiation and growth of microcrazing and cavitation in a RT-PMMA is significantly affected by the rate of deformation in addition to the rubber content, and the toughening mechanisms work effectively at a strain rate lower than a critical rate that definitely depends upon the rubber content.

The increase of $G_{I \max}$ of PM40 at rates higher than 1 m/s might also be due to the effect of kinematic energy, and $G_{I \max}$ of EM20 possibly received the same contribution, although it was not detectable under the process of the significantly decreased $G_{I \max}$ because of the reduction of the toughening mechanisms. Thus,

Fig. 7 Fracture surfaces of EU at different displacement rates

the measured $G_{I\max}$ of the present RT-PMMA's appears to contain the effects of both the reduction of the toughening mechanisms and kinematic energy, and thus the rate dependent behavior was complicated. Further study is needed to clarify this.

3.5 Displacement-rate dependence of crack growth behavior

Fracture surfaces of EU and CU at three different displacement rates are shown in Figs. 7 and 8, respectively. For both EU and CU the patterns of the surfaces depended upon the rates. At the low rate of 1×10^{-4} m/s both EU and CU exhibited surfaces created by stable fracture; this corresponded to the gradually decreasing load after the crack initiation observed in the load-displacement curves (Fig. 3). As the rates increased, transition of the fracture process

Fig. 8 Fracture surfaces of CU at different displacement rates

from stable to unstable occurred. The lines perpendicular to the direction of crack growth seen in Fig. 7(c) are typical patterns observed in extruded PMMA, and are thought to appear on a fracture surface when the rate of crack growth reaches about 350 m/s⁽¹¹⁾. Parabolic patterns typical of PMMA are seen in Fig. 8(c). The rate of the unstable crack growth can be estimated as 150 to 200 m/s on the basis of the parabola density.

SEM photographs of the fracture surfaces of EM20 and those of PM40 at three different displacement rates are shown in Figs. 9 and 10, respectively. As seen, EM20 and PM40 exhibited a flat fracture surface at all rates, and no shear lip indicating macroscopic plastic deformation was noted. It therefore appears that quasi-static linear fracture mechanics

(a) 1×10^{-4} m/s

(b) 0.9 m/s

(c) 10 m/s

500 μ m

Fig. 9 Fracture surfaces of EM20 at different displacement rates

was applicable to the fracture initiation in the RT-PMMA's. As seen in Fig. 9(a), at 1×10^{-4} m/s the entire fracture surface of EM20 was flat and smooth and seen as a whitening region under light. This whitening phenomenon in RT-PMMA is generally due to the cavitation of rubber particles and microcraze formation caused by local stress concentration in the vicinity of the particles. It is therefore understood that in the fracture process shown in Fig. 9(a) the crack propagated in a stable manner accompanied by the cavitation and microcrazing in the vicinity of the crack tip. Similar fracture surfaces were observed at

(a) 1×10^{-4} m/s

(b) 0.96 m/s

(c) 13 m/s

2 mm

Fig. 10 Fracture surfaces of PM40 at different displacement rates

rates lower than 0.1 m/s, and the intensity of whiteness decreased with increase in rate. As seen in Figs. 9(b) and (c), at higher rates a transition from smooth to rough surface was observed on each fracture surface, and the smooth region narrowed as the rates increased. This type of rough surface usually appears following brittle fracture of RT-PMMA⁽¹²⁾, and corresponds to the sudden decrease of the load in the load-displacement curve as shown in Fig. 3(c). PM40, however, exhibited whitening smooth surfaces at all rates tested, with the intensity of whiteness decreasing as the rate increased.

It is thus concluded that in an RT-PMMA a crack propagates accompanied with cavitation and

microcrazing in the vicinity of the crack tip, and that these toughening mechanisms are affected by displacement rate, i.e. strain rate in the crack-tip region. It is also understood that the increase of strain rate tends to reduce the generation of the toughening mechanisms. The effects of strain rate on the mechanisms of the initiation and growth of cavitation and microcrazing must be understood in order to know the limits of the toughening effects.

4. Conclusions

Dependence of the mode I fracture properties of two different RT-PMMA's (EM20 and PM40) and two neat PMMA's (EU and CU) on displacement rates was investigated over five decades of displacement rates (1×10^{-4} –13 m/s) using a high speed testing machine, and the following conclusions were reached:

(1) The maximum mode I stress intensity factor, $K_{I\max}$, and the maximum mode I energy release rate, $G_{I\max}$, of EM20 increased with increase in rate up to 1 m/s and then rapidly decreased above that (>1 m/s), suggesting the sudden reduction in toughening effects. PM40 showed increases of $K_{I\max}$ and $G_{I\max}$ as the rates increased, and then they decreased slightly at high rates (>1 m/s). Thus PM40 maintained the toughening effect relatively well within the rates examined in this study. It is understood that the toughening effects due to the cavitation of rubber particles and microcraze formation around the particles in the vicinity of the crack tip are restrained and reduced by the increase of displacement rate, i.e. strain rate in the crack-tip region.

(2) No traces of extensive ductile fracture were observed on the fracture surfaces of the RT-PMMA's or neat PMMA's, suggesting the applicability of linear fracture mechanics in this study. However, the effect of kinematic energy on the mode I fracture properties must be considered for the neat PMMA's at a high rate greater than 1 m/s.

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