Effect of particle size on impact properties of epoxy resin filled with angular shaped silica particles

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The effect of particle size on the impact properties of cured epoxy resin has been studied. This resin is filled with angular shaped silica particles that were prepared by crushing fused natural raw quartz. These particles were sorted into six groups having different mean sizes ranging from 2 to 47 μm. Specimens having a U-shaped blunt notch were prepared. The impact properties were measured by an instrumented Charpy type impact tester which can record a load–displacement curve at impact fracture. The impact absorbed energy increased with a decrease in silica particle size. The fractured surfaces were studied using a scanning electron microscope to clarify the initiation point of fracture.

(Keywords: epoxy resin; silica particle; instrumented Charpy type impact test; impact absorbed energy; fractography; blunt notch)

INTRODUCTION

In our previous papers[1,2], the effects of particle size on static physical properties, such as crack propagation[1], and on the mechanical properties[2] of cured epoxy resin filled with angular shaped silica particles have been studied. The angular shaped silica particles were prepared by crushing fused natural raw quartz and were sorted into six groups having different mean sizes ranging from 2 to 47 μm and dispersed in epoxy resin prior to curing. The mechanical strength increased with decrease in particle size[2], whereas the critical stress intensity factor decreased[1]. Using scanning electron microscopy (SEM), the reason why particle size affected these quantities was evaluated.

The impact test measures the ability of a material to withstand a sudden blow[3]. In many applications a satisfactory resistance to impact loading is an important performance requirement and, indeed, impact toughness is often an important criterion in materials selection. Impact strength is generally measured using an ordinary Charpy or an Izod type impact tester. However, since the values obtained include, in addition to the true impact absorbed energy, energy stored in the tester, toss energy of broken halves, etc.[3–5], the testers are not suitable for the quantitative analysis of impact fracture. Recently, an instrumented tester which can record a load–displacement curve at the impact fracture by a load sensor on the hammer has been developed. This enables the true impact absorbed energy to be measured from the area under the load–displacement curve[3–7]. However, the impact properties of particulate filled cured epoxy resin have not yet been extensively studied using this tester[5].

In this study, therefore, the effect of particle size on the impact properties of cured epoxy resin filled with angular shaped silica was studied with an instrumented Charpy type impact tester using the same silica particles used in our previous papers[1,2].

EXPERIMENTAL

Materials

The angular shaped silica particles were prepared by crushing amorphous silica that was made by fusing natural raw quartz at 1900°C (RD-8, Tatsumori Ltd). The crushed particles were sorted into six groups by air separation. As shown previously[1], the particle size at which the accumulated distribution values reached 50% was defined as the mean particle size. The mean sizes of the particle classifications are 2, 5, 13, 18, 33 and 47 μm.

The epoxy resin used was bisphenol A type (Epikote 828, Shell Chemical Co.; equivalent weight per epoxy group 190 ± 5, average molecular weight 380). 1,2-Cyclohexanedicarboxylic anhydride and tri-n-butylamine were used as a hardener and an accelerator for curing the epoxy resin, respectively.

Sample preparation

The silica particles (204 or 296 parts per hundred resin by weight, phr) were dispersed in the mixture of epoxy resin (100 phr) and hardener (66 phr) by stirring at room temperature for 1 h under vacuum. The accelerator (0.5 phr) was added to the mixture and stirred for 10 min. The final mixture was cured in a mould (15 mm × 15 mm, height 100 mm) at 120°C for 2 h, followed by 140°C for 21 h.

Instrumented Charpy type impact test

The shape and dimensions of the impact test specimen

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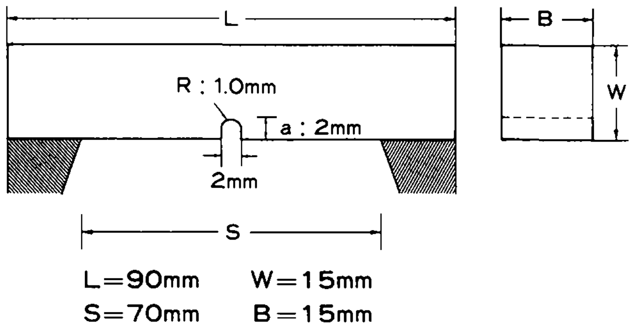

Figure 1 Shape and dimensions of Charpy type impact test specimen

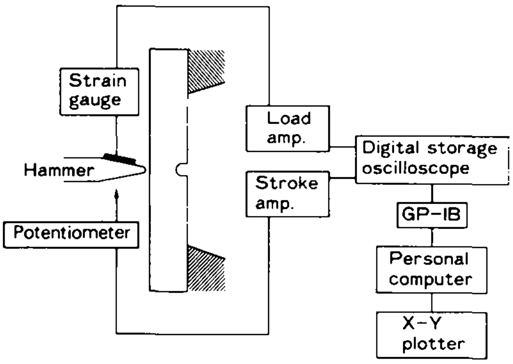

Figure 2 Block diagram of the instrumented Charpy type impact tester

used are shown in *Figure 1*. A U-shaped blunt notch (depth 2 mm, radius 1 mm) was machined at the centre of the specimen. The support span was 70 mm. The impact properties were measured by the instrumented Charpy type impact tester[3-7] (CAI-AC5-CZ(A) type, Japan Sensor Corp.; capacity 14.7 J) at room temperature. The weight of the hammer was 6 kg, the length of the hammer arm was 0.6 m and the hammer speed at strike was 1.10 m s^{-1}. The block diagram of the tester is shown in *Figure 2*. The impact load was resolved by a semiconductor strain gauge on the hammer and the displacement was measured by a potentiometer on the pivot of the hammer. The signals detected were amplified by load and stroke amplifiers. The load–time curve and the displacement–time curve at impact fracture were monitored by a digital storage oscilloscope (VC-8240 type, Hitachi Ltd). Both curves were recorded and analysed by computer and the load–displacement curve was obtained.

RESULTS AND DISCUSSION

Figure 3 shows a typical load–displacement curve measured by the instrumented Charpy type impact tester. The fracture load and the displacement at specimen break were obtained from the load and the displacement values at the last peak, respectively[3]. Further, the impact absorbed energy was measured from the area under the load–displacement curve[4]. The shape of the curve was similar to that obtained for epoxy resins filled with silica particles[5] or modified with rubber particles[7] measured using a similar instrumented Charpy test impact tester.

Figure 4 shows the effect of particle size on the load at break. For particle contents of 55 and 64 wt%, the load values increased with decrease in particle size. The values for the former case were always higher than those for the latter. The values for the cured epoxy resins filled with small particles (2, 5 and 18 μm) were higher than that for unfilled cured epoxy resin.

Figure 5 shows the effect of particle size on the displacement at break. The value increased with decrease

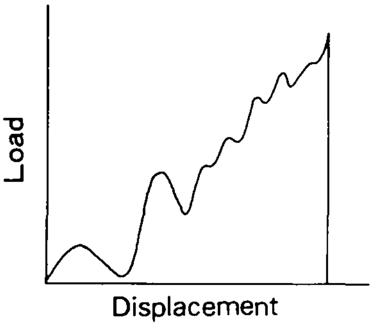

Figure 3 Typical load-displacement curve measured by the instrumented Charpy type impact tester

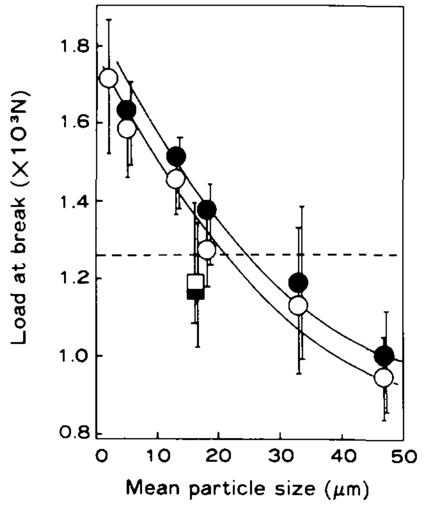

Figure 4 Effect of particle size on load at break measured by instrumented Charpy type impact test of cured epoxy resins filled with unsorted original (□, ■) and sorted (○, ●) angular shaped silica particles with particle contents of 55 wt% (□, ○) and 64 wt% (■, ●). Broken line indicates the load at break for unfilled cured epoxy resin

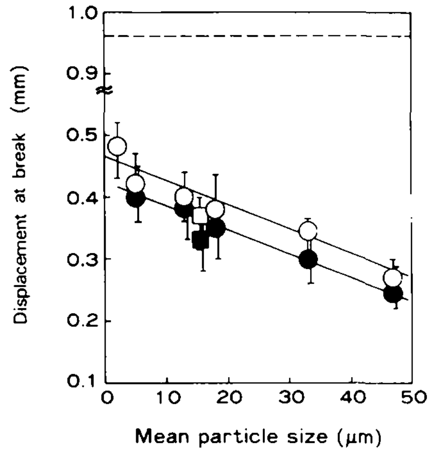

Figure 5 Effect of particle size on displacement at break measured by instrumented Charpy type impact test of cured epoxy resins filled with unsorted original (□, ■) and sorted (○, ●) angular shaped silica particles with particle contents of 55 wt% (□, ○) and 64 wt% (■, ●). Broken line indicates the displacement at break for unfilled cured epoxy resin

in particle size although the values for filled cured epoxy resins were lower than that for unfilled cured epoxy resin. The values for particle content of 55 wt% were slightly higher than those for particle content of 64 wt%.

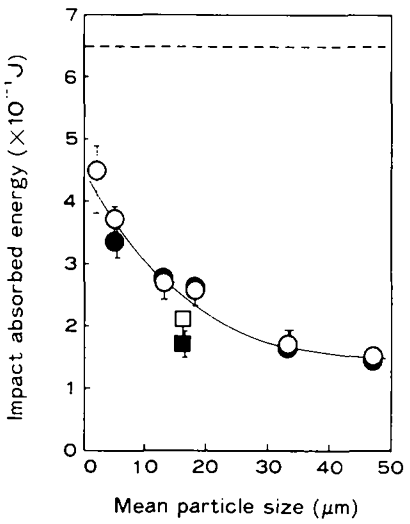

Figure 6 Effect of particle size on impact absorbed energy measured by instrumented Charpy type impact test of cured epoxy resins filled with unsorted original (□, ■) and sorted (○, ●) angular shaped silica particles with particle contents of 55 wt% (□, ○) and 64 wt% (■, ●). Broken line indicates the impact absorbed energy for unfilled cured epoxy resin

Figure 6 shows the effect of particle size on the impact absorbed energy. The impact absorbed energy increased with decrease in particle size. There was no difference between the values for particle contents of 55 and 64 wt%.

From the above results, it was clear that impact properties improved with decrease in particle size and the tendency was the same as that for static mechanical strength shown previously[2].

The fractured surfaces were subsequently observed by SEM. *Figure 7* shows the U-shaped notch tip regions of fractured surfaces of impact specimens. In all the fractured specimens, the fracture initiated at the bottom of the U-shaped notch. The characteristic fractured marks which started from the initiation point of fracture[8,9] were observed in both unfilled (*Figure 7a*) and filled resins (*Figure 7b–d*). This was the same as for the flexural test specimens shown elsewhere[2]. In order to clarify the effect of particle size on the initiation mechanism of fracture, the region around the initiation point of fracture was magnified.

Figure 8 shows the magnified regions. For the cases of unfilled (*Figure 8a*) and filled with small size particles (mean size 5 μm, *Figure 8b*), the initiation point of fracture was the defect which had existed at the bottom of the U-shaped notch. A similar observation was obtained in the cured epoxy resin filled with 2 μm particles. By increasing the particle size to >5 μm, the

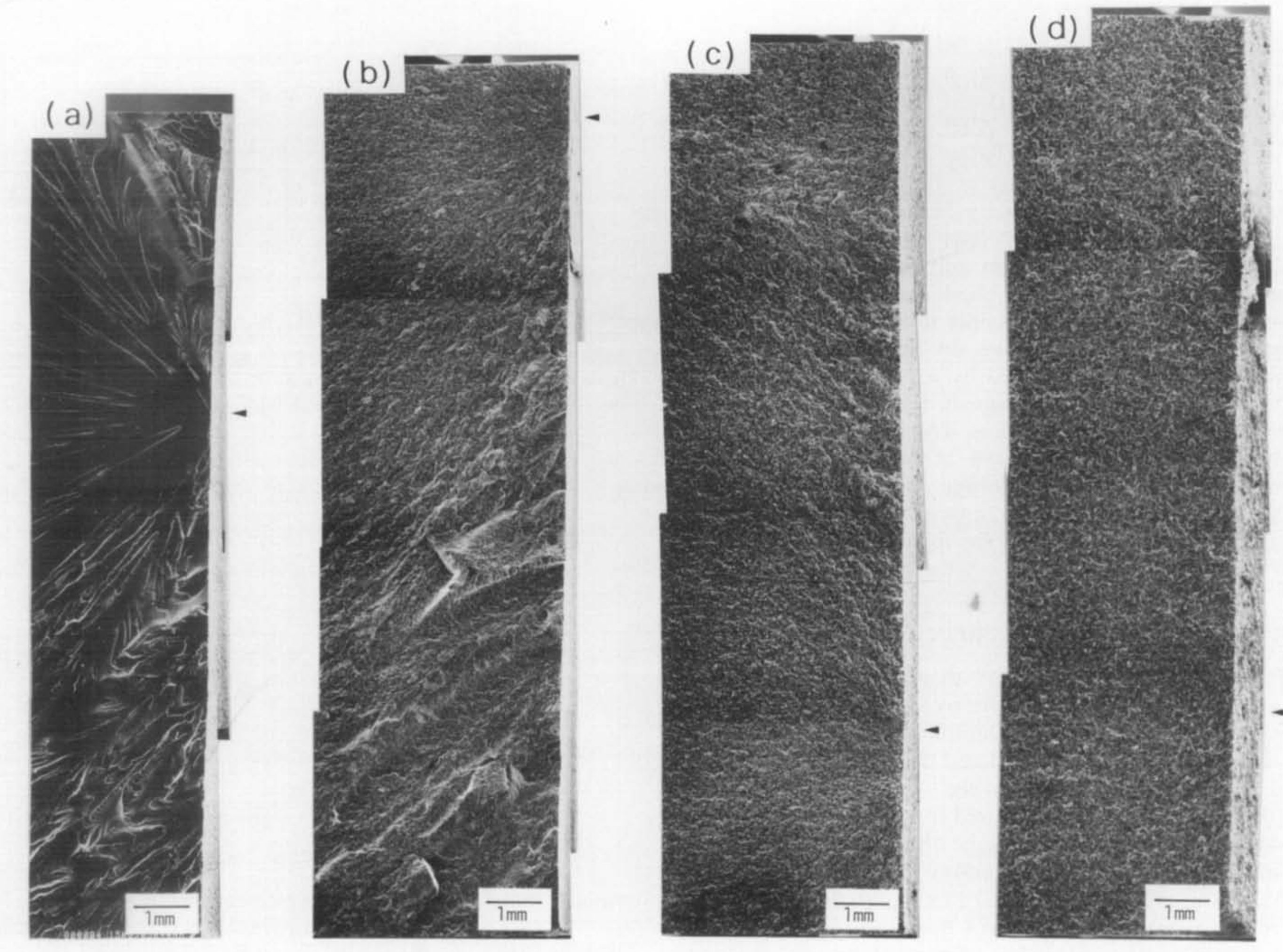

Figure 7 Fractured surfaces for impact test specimens of unfilled cured epoxy resin (a) and those filled with angular shaped silica particles ((b)–(d)) with a particle content of 55 wt% observed by SEM. Mean particle size of filled silica: (b) 5 μm; (c) 18 μm; (d) 47 μm. Arrow head indicates the initiation point of fracture

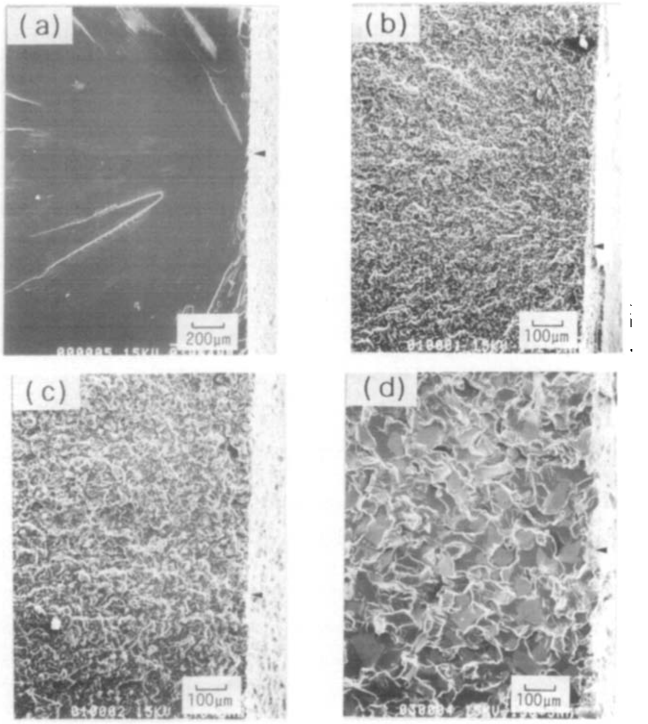

Figure 8 Regions around the initiation point of fracture for fractured impact test specimens of unfilled cured epoxy resin (a) and those filled with angular shaped silica particles ((b)–(d)) with a particle content of 55 wt% observed by SEM. Mean particle size of filled silica: (b) 5 μm; (c) 18 μm; (d) 47 μm. Arrow head indicates the initiation point of fracture

fracture of the specimen tends to be initiated more favourably from the fracture involved in the particles (*Figures 8c* and *d*, mean size 18 and 47 μm, respectively). In all cases for the resins filled with 33 and 47 μm particles, the fracture was initiated from the particle fracture. As mentioned in our previous papers[1,2], since the silica particles were prepared by crushing fused raw quartz, the shapes were more irregular in the larger sized particles, and the number of particles containing cracks increased with particle size.

From the above results, it is concluded that the reason that the impact properties improved with decrease in particle size is the same as the reason for the mechanical strength increasing with decrease in particle size as given in a previous paper[2]. That is, the particle fracture due to the inherent flaws (cracks) initiated fracturing in the impact test and decreased strength.

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