Original paper

Dental Materials Journal 19(1) : 22–33, 2000

Microfracture Mechanisms of Composite Resins Containing Prepolymerized Particle Fillers

Kyo-Han KIM, Young-Bin KIM and Osamu OKUNO[1]

Department of Dental Biomaterials, College of Dentistry and Institute of Biomaterials Research and Development, Kyungpook National University, 2-101 Dong-In Dong, Jung-Gu, Taegu 700-422, Korea

[1]Department of Dental Materials Science, School of Dentistry, Tohoku University, 4-1 Seiryo-Machi, Aoba-Ku, Sendai 980-8575, Japan

Received August 2, 1999/Accepted December 9, 1999

The effect of prepolymerized particle fillers on fracture toughness of four commercial composite resins was studied. The ground surface morphology was examined with a field emission scanning electron microscope. Fracture toughness was determined by using single edge notched specimens. Acoustic Emission (AE), which is the elastic wave due to the release of energy from the localized sources of material, was detected by sensors of a high-sensitivity and low-noise resonance type during the fracture toughness test. Acoustic Emission signals detected were analyzed for parameters such as amplitude and energy. Fracture surfaces were examined with a scanning electron microscope. The fracture toughness values, AE releasing patterns during fracture toughness test, and the fracture surface findings were analyzed to understand the fracture behavior of composite resins containing prepolymerized particle fillers. A microfracture model and fracture mechanisms to increase the fracture toughness of this type of composite resins are proposed.

Key words : Prepolymerized particle filler, AE (Acoustic emission), Microfracture mechanism

INTRODUCTION

Dental composite resins have established of important role as an esthetic restorative material in anterior and posterior areas over the last 10 years. Nevertheless, there are many problems with composite resins used as anterior and posterior restorative materials, such as high polymerization shrinkage[1,2], low wear resistance[3,4], and low fracture resistance[5,6]. Recently, there have been many studies on the development of fillers[7–9], resin monomers[10,11], polymerization initiator[12–14], and silane coupling agents[15–17] to solve the above-mentioned problems. Among them, the research on submicron sized fillers has been focused on increasing the wearability and the polishability.

To achieve a higher filler load with high mechanical properties, there is a limit to the ratio of submicron sized filler load to resin matrix. Prepolymerized particle fillers[18], which are solidified and ground polymers heavily loaded with submicron sized filler particles have been used to overcome this problem. The strength of the prepolymerized particle filler, the bonding strength of the prepolymerized particle filler to the resin matrix and the morphology of the prepolymerized particle filler, are all important factors affecting the properties of composite resins.

In brittle materials such as dental ceramics and composite resins, the evaluation of the resistance of materials to the crack propagation is very important since catastrophic failure can easily occur due to inherent internal flaws or surface cracks formed during grinding and polishing procedures[19-22]. Clinically, the bulky composite resins often form the surface cracks first during wear, finally leading to fracture[23-25].

To evaluate the resistance of materials to crack extension, the stress intensity factor K_I, is used. The stress intensity factor reflects the stress distribution around the main crack tip. The critical stress intensity factor (fracture toughness K_{IC}) is a maximum value of resistance of material to crack extension. An increase in fracture resistance is required, due to the low fracture toughness of dental composite resin ($0.8\text{-}1.5 MPa\sqrt{m}$)[19-22].

It has been reported that phase transformation[26], generation of microcracks[27], and deflection of crack tips[28] are very effective in increasing the fracture resistance of brittle materials. Apparently, the stress relaxation around the main crack tip by microcrack nucleation and deflection of the crack tip can increase the fracture resistance of dental composite resins. These phenomena such as microcrack nucleation before the main crack extension and crack deflection after the main crack extension can be influenced by the internal structure, for example, the size and the morphology of prepolymerized particle fillers in the case of composite resins containing prepolymerized particle fillers.

In this study, the microfracture process of dental composite resin containing prepolymerized particle fillers was clarified from fracture surface findings, a fracture toughness test, and AE pattern generation during the fracture toughness test. Mechanisms to increase the fracture resistance of this type of composite resins are proposed.

MATERIALS AND METHODS

Materials and specimen preparation

Four commercial composite resins containing prepolymerized particle fillers used as experimental materials are listed in Table 1 together with code, manufacturer and Lot number (Table 1).

Table 1 Composite resins containing prepolymerized particle fillers used in this study

Materials	Code	Manufacturer	Lot No.
Matafil CX	MC	Sun-Medical[1]	70301
Silux Plus	SP	3M[2]	5702XL
Heliomolar	HM	Vivadent[3]	818828
Palfique Estelite	PE	Tokuyama[4]	282

[1] Osaka, Japan
[2] St. Paul, MN, USA
[3] Schaan, Liechtenstein
[4] Tokuyama, Japan

Specimens for SEM observation of ground surface texture made by filling composite resins into a metal mold (5 mm width, 2.5 mm thickness) and light curing for 1 min were embedded in a chemical cured epoxy resin and stored in water at 37℃ for 24 hrs. The surface of the composite resins was polished with water/proof silicon-carbide paper in sequence of #800, #1000 and #1200 and then polished with diamond pastes (6 μm→3 μm→1 μm→0.25 μm→0.1 μm). After vapor coating with carbon, the surfaces were examined with a field emission scanning electron microscope (S-4200, Hitachi Co., Japan).

Determination of weight filler contents

The weight filler contents of the composite resins were determined by the standard ash method of ISO NO.4049. The weight of a rectangular specimen (2.5 × 5 × 10mm) (W_0) was measured with an electric balance, and after heating the specimen in an electric furnace at 600℃ for about 30 min to burn out the organic matrix, the weight of the specimen (W_1) which consisted only of inorganic filler was calculated. The weight percentage of filler content of composite resins was determined from the weight difference between W_0 and W_1. The filler volume fraction of inorganic filler was calculated by Eq.1, where the densities of resin matrix and inorganic filler were 1.2 g/cm^3 and 2.5 g/cm^3 respectively. The density, d_r, is the density of the resin and d_f is that of the filler. Three specimens were used to determine the filler contents for each composite resin.

$$\text{Vol.}\% = \frac{d_r \times \text{Wt.}\%}{d_r \times \text{Wt.}\% + d_f(100 - \text{Wt.}\%)} \times 100 \qquad \text{(Eq.1)}$$

Fracture toughness test

Specimens for fracture toughness were made in a split mold (30 mm (L) × 5 mm (W) × 2.5 mm (B)) containing a razor blade notch fixed into a slit in the mold (single edge notch specimen). The resulting notch/width ratio of the specimen was controlled in the range of 0.45-0.55. After packing the resin into the mold, the specimens were illuminated in five 30 s illumination steps along each side with a light curing machine (Powerlite 100, USA). The light-cured specimens were removed from the mold, trimmed and stored in distilled water at 37℃ for 24 hrs before testing.

The specimens were tested by three-point bending in an Instron testing machine (4202, Instron Corp., Canton, MA, USA) at a cross-head speed of 0.1 mm/min. The stress intensity factor, K_Q (MPa · m$^{0.5}$) was obtained from the peak load (P_Q) and specimen configuration by Eq.2.

$$K_Q = [(P_Q \cdot S)/(B \cdot W^{1.5})] \times f(a/W) \qquad \text{(Eq.2)}$$

$$f(a/W) = 3(a/W)^{0.5}[1.99 - (a/W)(1-a/W)(2.15 - 3.93a/W + 2.7a^2/W^2)]/2(1+2a/W)(1-a/W)^{1.5}$$

Where P_Q is the peak load (N), S is the span (m), B is the specimen thickness (m), a is the crack length, and W is the specimen width (m). If K_Q satisfies the condition, B, $a \geq 2.5(K_Q/\sigma_{ys})^2$, then K_Q is prescribed as K_{IC} (the fracture toughness) (ASTM E-399). Five specimens were used for each fracture toughness test.

Acoustic Emission Detecting

Two sensors attached to the ends of the specimen detected acoustic emission signals released during the fracture toughness test. The sensors used were of a high sensitivity, low-noise resonance type, equipped with an amplifier of 54 dB gain (M204, Fuji Ceramic Co., Japan). The signals from the sensors were sent to an AE analyzer (NF9861, NF Instrument Co., Japan) and further amplified by 30 dB. Signals recorded by the AE analyzer were analyzed for various parameters, such as the amplitude and the events (numbers of released signals). The fractured surfaces of fracture toughness specimens were examined under a scanning electron microscope (S-2300, Hitachi Corp., Tokyo, Japan).

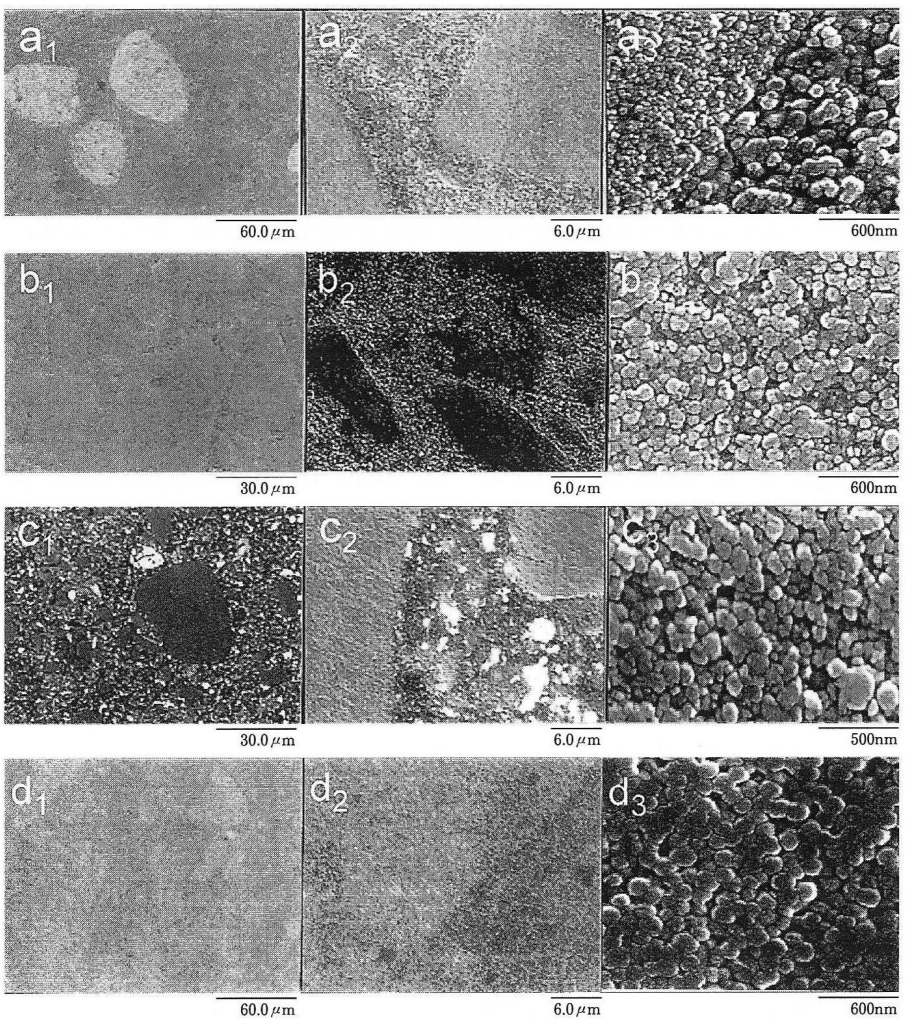

Fig. 1 Scanning electron micrograph of the ground surface of composite resins (a_1, a_2, a_3: MC, b_1, b_2, b_3: SP, c_1, c_2, c_3: HM, d_1, d_2, d_3: PE).

RESULTS

Microstructure of composite resins containing prepolymerized particle fillers

SEM photographs of composite resin containing prepolymerized filler particles are shown in Fig. 1. Round shaped prepolymerized filler particles with sizes of 30-60 μm (a_1), and irregular shaped prepolymerized filler particles with sizes of 5-10 μm, are shown in MC. Round fillers with sizes of 0.1-0.2 μm are included in the resin matrix and round fillers smaller than 0.05 μm are seen in irregular shaped prepolymerized particle filler (a_3). In SP, prepolymerized filler particles of various shapes and sizes, ranging from 5 to 30 μm, are seen (b_1). Round filler of 0.1-0.2 μm are included in the prepolymerized particle fillers and the resin matrix (b_3).

HM includes round and irregular shaped prepolymerized filler particles, sized 30 μm and smaller than 10 μm respectively (c_1). These prepolymerized filler particles and matrix consisted of round fillers, sized 0.05-0.1 μm (c_3). Irregular shaped prepolymerized particle fillers of various sizes, less than 30 μm (d_1) and round fillers of 0.05-0.1 μm, are included in prepolymerized filler particles and the resin matrix (d_3), respectively, in PE. In these types of composite resins, the shapes of prepolymerized filler particles include round and irregular. The size of round shaped prepolymerized filler particles was about 30-60 μm and the size of irregular shaped prepolymerized filler particles, being crushed, was smaller than 10 μm. The fillers included in the prepolymerized filler particles and resin matrix were round shaped, with sizes smaller than 0.2 μm.

Filler fractions and fracture toughness

Fracture toughness is listed in Table 2. The filler fraction ranged from 25.7% to 51.1% in volume and from 41.8 to 68.5% in weight. PE shows the highest filler content and MC shows the lowest filler content. The fracture toughness showed values in the range of 0.63 to 0.85 MPa$\sqrt{m}$. MC has the lowest value of fracture toughness, second is SP, next is HM, and PE shows the highest fracture toughness of 0.85 MPa$\sqrt{m}$.

Fractography

SEM fractographs of the fracture surfaces of composite resins containing

Table 2 Filler fractions and fracture toughness of composite resins containing prepolymerized particle fillers

Materials	Filler Vol.% (Wt.%)	K_{IC}*(MPa$\sqrt{m}$)
MC	25.7 (41.88)	0.63±0.01
SP	37.1 (55.14)	0.81±0.07
HM	41.2 (59.31)	0.84±0.02
PE	51.1 (68.55)	0.85±0.01

* Mean and standard deviation

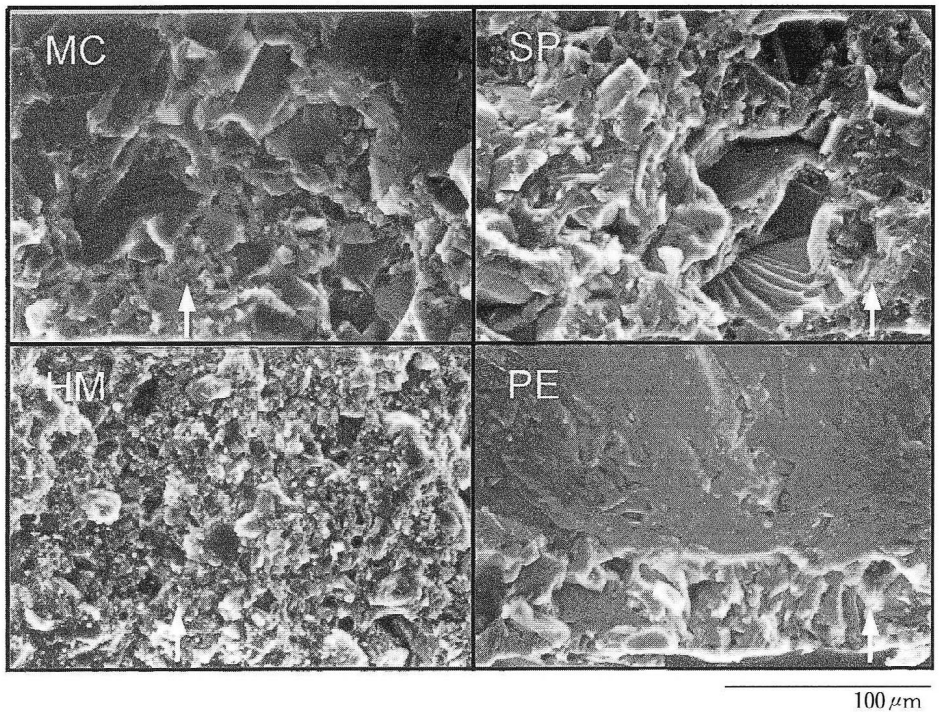

Fig. 2 Scanning electron micrograph of the fracture surface of composite resins (Arrows show the direction of crack extension).

prepolymerized filler particles are shown in Fig. 2. MC and SP reveal very rough fracture surfaces while HM and PE show rather flat fracture surfaces. In MC, cleavage planes sized 30-60 μm were observed, and these cleavage planes were linked by the interface of irregular shape filler and matrix. A brittle fracture surface was also observed in SP. The cleavage planes, sized 30 μm, and rough fracture surface in the cleavage planes were observed. In HM, fracture units of 30 μm were locally seen, and the greatest portion of the fracture surface was composed of the interface of fractured fillers and matrix. In PE, a different fracture surface, being very flat, was observed.

Acoustic Emission

The results of amplitude of acoustic emission released during the fracture toughness test are shown in Fig. 3. Load-Time curve and accumulated AE events and AE energy released during fracture toughness test are also depicted in Fig. 4. A large number of AE events was released during the fracture toughness testing of MC. AE release commenced at 70-80% of maximum load. AE events released during toughness testing of SP were almost the same as those of MC. Low amplitude and low energy AE were released starting at about 80-90% of maximum load. In HM, low amplitude AE were released, and the number of events was smaller compared to MC and SP. Also, the

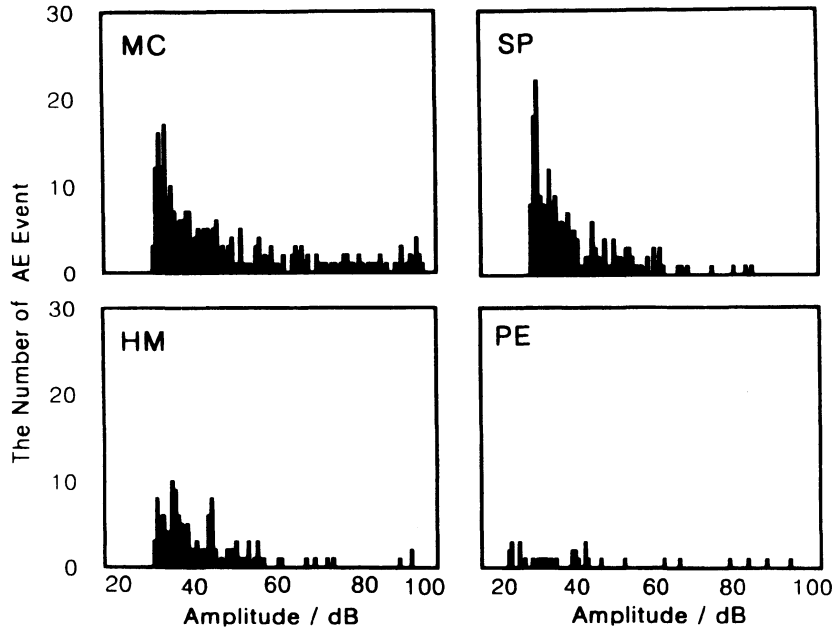

Fig. 3 Released AE events and their amplitude during fracture toughness test.

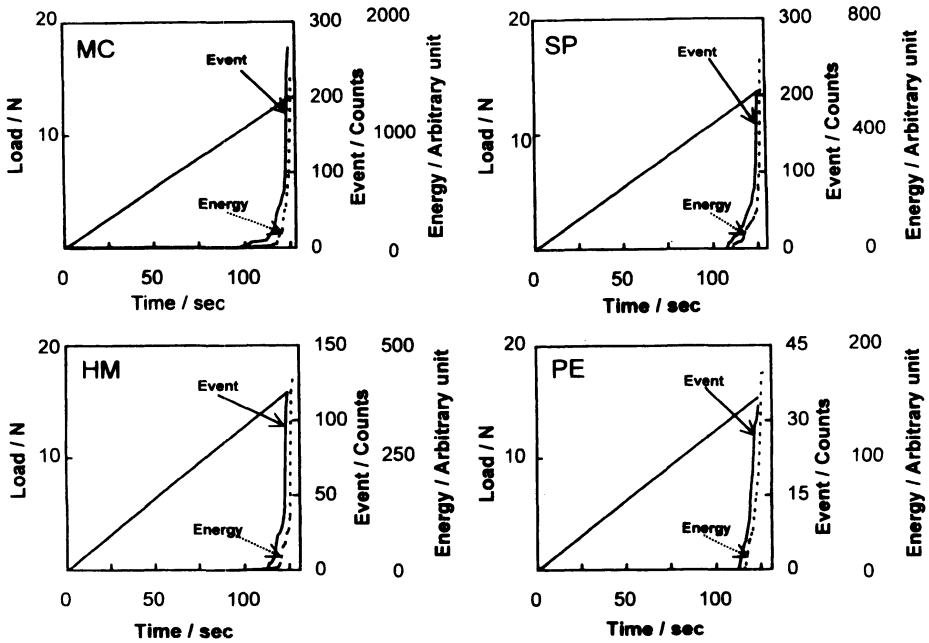

Fig. 4 Releasing behavior of AE during fracture toughness test.

released AE were detected almost at the maximum load. The smallest number of AE, abruptly released near the maximum load, was detected in PE.

DISCUSSION

Microfracture Mechanism

Generally, it is known that fracture toughness is an increasing function of filler content in dental composite resins. However, Kim *et al.*[19,20] reported that in dental composite resins, fracture toughness increases until it reaches a certain limiting filler content, after which the fracture toughness decreases because the crack front overlaps with an other crack front. In that experiment, the fracture toughness decreased with filler content exceeding 60% of the volume filler fraction, rather than increasing the bending strength. The composite resins examined here showed a volume fraction in the range of 25 to 51%, so the fracture toughness of the composite resins tested in this experiment was increased with increasing filler volume fraction.

In MC, cleavage fracture surfaces of size 20-60μm, which can be assumed to have been caused by the fracture of round shaped prepolymerized filler particles of size 30-60μm, were observed, resulting in a rough fracture surface. Comparing this with the AE detection results (Fig. 3, 4), it is assumed that the round shape prepolymerized filler particles of size 30-60μm started to fracture at 70-80% of maximum load. Before the main crack extension, round shape prepolymerized filler particles, located above and below the main crack, were preferentially fractured by stress concentration around the main crack, and the main crack extended to coalesce three-dimensionally with microcracks that were already formed by prepolymerized filler particles. The resultant fracture surface was very rough. The high-amplitude AE events of 60-80 dB corresponded to the fracture of round shape prepolymerized filler particles, and low amplitude AE events corresponded to the separation of irregular shaped prepolymerized filler particles of size 5-10μm, from the matrix.

The fracture surfaces of 20-30μm observed coincided well with the size of prepolymerized filler particles seen in the surface structure of SP. The number of AE events released during this experiment is smaller than that for MC, but the AE release commenced at 80% of maximum load in the same way as MC. In the AE amplitude distribution of HM, the release of a small number of high amplitude AE coincided well with the number of cleavage planes formed by the round-shaped fracture of prepolymerized filler particles on the fracture surface. This suggests that the generation of low amplitude AE corresponded to the separation of small sized irregular prepolymerized filler particles from matrix. The most characteristic fracture surface and AE release pattern were observed in PE. The number of AE events was very small, and the AE were released abruptly at the maximum load. The main crack extended almost linearly regardless of prepolymerized filler particles.

Taking the ground surface texture, fracture surfaces, and AE release patterns into account, it is considered that high amplitude AE are related to the fracture of prepolymerized filler particles themselves, and low amplitude AE to the interface

separation of prepolymerized filler particles from the matrix. Lange[29], Range and Radford[30], and Young and Beanmont[31] have suggested the following possible mechanisms for the increase of fracture energy, *i.e.*, fracture toughness, when a second inorganic filler was incorporated into the brittle resin matrix.

1) Fracture energy is increased due to the increase in the fracture surface area by adding a second phase in the resin matrix. This increase is independent of particle size and only dependent on the volume fraction of the second phase.

2) Fracture energy can be increased by plastic deformation of the second phase during fracture.

3) Fracture energy can be increased by friction between parting fracture surfaces during fracture.

4) Fracture energy can be increased by the interaction between a crack front and the second phase.

During the fracture, a moving crack front is momentarily pinned at positions of heterogeneity within the brittle matrix. This interaction leads to bowing, out of the crack front between the pinning positions, thus increasing its total length. On breaking away from these pinning positions, the crack front creates the characteristic steps on the fracture surface. These steps are formed by overlapping of the crack front as it bows between the dispersed particles.

Besides the mechanisms listed above, Kim *et al.*[19,20] confirmed other toughening mechanisms in dental composite resin, such as the microcrack-induced toughening effect and the crack-deflection-induced toughening effect. Irregular shaped fillers of size 5-10μm cause the nucleation of microcracks around the crack tip before crack extension, which releases the stress concentration, and this apparently increases the fracture resistance of the material (microcrack-induced toughening effect). When these microcracks are distributed broadly around the main crack, the crack tip extends by deflection from one direction to another, and coalesces with microcracks, resulting in a very rough fracture surface crack-deflection that induces the toughening effect.

In this experiment, the lowest filler volume fraction was MC with 25.7%, followed by SP with 37.1 Vol.%, HM with 41.2 Vol.%, and PE with the highest filler volume fraction of 51.1%. The fracture toughness value of MC reached almost 80% of that of PE regardless of its filler fraction being almost half that of PE. Likewise, SP and HM showed the similar fracture toughness to PE in spite of their low filler fractions. The reason for the high fracture toughness value of MC in spite of its low filler fraction was understood as follows. First, round shape prepolymerized filler particles were fractured before main crack propagation and formed microcracks that relieved the stress concentrations. Second, the main crack extended with deflection when it coalesced with pre-formed microcracks, resulting in an apparent increase of fracture toughness, which was confirmed by the AE release patterns.

From the fracture surface observation and AE release pattern results, the stress relaxation effect in SP by fracture of irregularly shape filler particles was not so remarkable as that of MC since fracture of irregular filler particles and high amplitude

AE release were very low compared to MC. HM showed a similar fracture toughness value to PE. Regardless of its low filler fraction, the toughening effect was induced by microcrack formation of round shaped prepolymerized filler particles, and crack deflection induced the toughening effect when the main crack extension was deflected.

The microfracture mechanism of composite resins containing prepolymerized filler particles can be understood as follows. The main crack extends three-dimensionally to coalesce with microcracks formed by round shaped prepolymerized filler particles before main crack extension, causing branching of the main crack in MC, SP, and HM. In the case of PE, the main crack extended almost linearly through the prepolymerized filler particles and matrix without fracture of prepolymerized filler particles before the main crack extension.

The factors which contribute to the increase of fracture toughness are the filler addition effect, the stress relaxation effect by fracture of prepolymerized filler particles around the main crack, and the branching effect of the main crack by coalescing three-dimensionally with prepolymerized particle fillers (Fig. 5).

The fracture of round-shaped prepolymerized filler particles sized 30-60μm before main crack extension was essential to increasing the fracture toughness in MC and SP. The other toughening mechanism was that the main crack extended three-dimensionally through the interface between the small irregularly shaped prepolymerized filler particles and the matrix resulting in a rough fracture surface. Fracture of prepolymerized filler particles did not occur until the maximum load since the strength of the prepolymerized filler particles and the interface strength between prepolymerized filler particles and matrix were both high in PE. When the main crack extended it, advanced linearly through the prepolymerized filler particles and matrix, forming a flat fracture surface. Consequently, this type of composite resin is not additive to the toughening mechanism, except for the filler adding effect, in spite of it having the highest volume fraction among the composite resins examined.

From the above results, it was concluded that in order to obtain an apparent

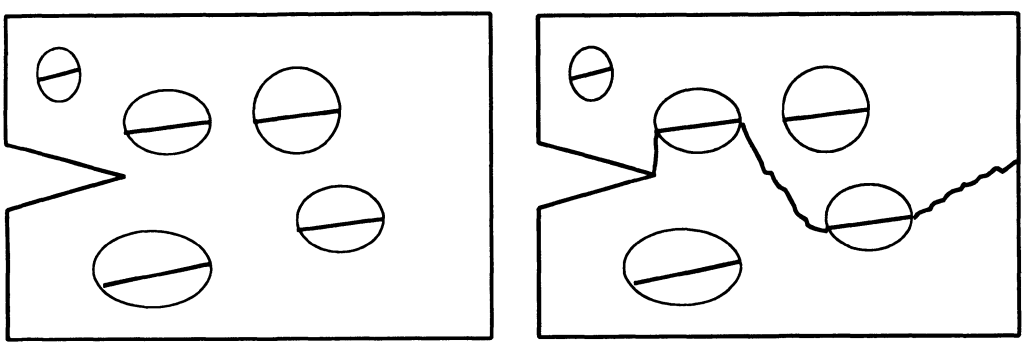

Fig. 5 Microcrack formation and crack extension mechanism of composite resins containing prepolymerized particle fillers.

toughening effect, the round-shape prepolymerized filler particles sized 30-60μm, must have a low strength to fracture easily by stress concentration before main crack extension. The small irregularly shaped prepolymerized filler particles must be strong and the interface strength weak for main cracks to propagate easily between the matrix and interface, which increases the resistance to crack extension, the fracture toughness.

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