JOURNAL OF MATERIALS SCIENCE: MATERIALS IN MEDICINE **10** (1999) 375–378

Effect of filler content and size to properties of composite resins on microwave curing

H. URABE[‡], Y. NOMURA*, K. SHIRAI, M. YOSHIOKA, H. SHINTANI
*Department of Operative Dentistry and *Department of Dental Materials, School of Dentistry, Hiroshima University, Hiroshima, Japan*

The effect of filler content and size to property of composite resin using high purity spherical silica particles on microwave curing was examined by mechanical means and electron microscope observation to develop microwave-curing composite resin inlay. Increasing filler content in three kinds of filler particles (0.45, 0.96 and 1.46 μm) resulted in increasing compressive strength, diametral tensile strength and knoop hardness and indicated the highest values at the operative mixing limit. The effect of particle size was that the mechanical property of the 0.96 μm filled resin was highest, followed by 1.46 μm and 0.45 μm on the same filler content. The 0.96 μm filled resin had about the same knoop hardness on both surfaces of the cured sample, which means that uniform polymerization occurred in the cured sample.

With electron microscope observation, the bubbles of about 1.0 μm and the cracks between the fillers and the matrix were observed in each cured resin. This is caused by the excessive absorption of microwave energy in fillers and surface treatment materials of fillers. Therefore, improvement of the composition of filler and surface treatment material of filler are needed, which makes it harder to absorb the microwave energy.

1. Introduction

Visible-light (VL) cured dental composite resins have been widely used as restorative materials for their ease of handling and aesthetic merits [1–6]. However, several clinical faults such as limited depth of cure and insufficient wear resistance, particularly in the posterior teeth resins, have been indicated [7–9].

To offset these drawbacks, the so called inlay/onlay technique, by which cured composite resins are made adding heat after VL curing out of the mouth, has been introduced [10, 11]. This method certainly improved the properties of the composite resin, but it needed more curing time and was cumbersome compared with only the VL curing method [12]. Thus, we tried to use microwave energy, which was very useful on curing denture base resins as reported previously [13–15], for curing the composite resin inlay/onlay instead of VL.

On the other hand, much effort has been devoted to investigations of the relationship between filler particles in composite resins and their properties. The information provided by past research suggests that the filler plays an important role in properties of the composites [16–20].

These above-mentioned studies were for VL curing, so we studied the influence of filler content and size to properties of composite resins on microwave curing in this research.

2. Materials and methods

2.1. Base monomer and filler

Monomers used were 2,2-bis[4-(3-methacryloxy-2-hydroxypropoxy) phenyl] propane (Bis-GMA) (Shin Nakamura Chemistry Co., Tokyo, Japan) and triethyleneglycol dimethacrylate (TEGDMA) (Tokyo Chemical Co., Tokyo, Japan) (Fig. 1). Bis-GMA and TEGDMA were mixed in the ratio of 60 : 40 wt %, to which was added benzoyl peroxide (BPO) (Tokyo Chemical Co., Tokyo, Japan) in the ratio of 0.5 wt % as the polymerization initiator for the base monomer. This ratio was the best, as we reported previously [21, 22].

Fillers were prepared with three kinds of high pure spherical silica particle having average sizes of 0.45, 0.94 and 1.46 μm (Adma Fine, Tatumori Co., Tokyo, Japan), which were surface-treated with γ-methacryloxypropyl trimethoxysilane (γ-MPTS) (Shin-Etus Chemical Co., Tokyo, Japan). The filler of 0.45 μm was mixed 70 wt %, 75 wt %, 80 wt % and 85 wt % in the base monomer (S05). Similarly the filler of 0.94 μm was added to 80 wt % (S10) and the filler of 1.46 μm to 82 wt % (S15), as shown in Table I, which were the ranges of the operative limit of mixing.

[‡] To whom correspondence should be addressed: Hidenori Urabe, Department of Operative Dentistry, School of Dentistry, Hiroshima University, 1-2-3 Kasumi-cho, Minami-ku, Hiroshima-shi, 734 Japan.

0957–4530

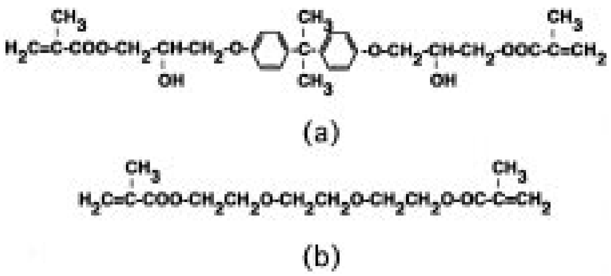

Figure 1 Molecule structure of the base monomers: (a) Bis-GMA; (b) TEGDMA.

TABLE I Fillers of three particle sizes were mixed for base monomer (Bis-GMA: TEGDMA = 60 : 40 wt % added 0.5 wt % BPO) as following

Filler size (μm)	Filler content (wt %)	Sample code
0.45	70 75 80 85	S05
0.96	70 75 80	S10
1.46	70 75 80 82	S15

2.2. Polymerization method

S05, S10 and S15 were filled in a Teflon mold, cover glasses put on the upper and lower surfaces, and irradiated for 5 min by the microwave of oscillation frequency 2450 MHz in a conventional microwave oven (EM-107S type, Santo Co., Tokyo, Japan). The cured samples (6 mm diameter × 3 mm, 3 mm diameter × 6 mm) were detached from the Teflon mold after keeping at room temperature for 30 min. They were supplied to each examination after 24 h five times.

2.3. Compressive strength test

The cured samples (3 mm diameter × 6 mm) were measured at a crosshead speed of 2.0 mm min^{-1} with a versatile examination machine (Autograph DCS-500, Shimadzu Co., Kyoto, Japan).

2.4. Diametral tensile strength test

Diametral tensile strengths of the cured samples (6 mm diameter × 3 mm) were measured at a crosshead speed of 0.5 mm min^{-1} based on the standard of ADA specification No. 27 [23] with a versatile examination machine (Autograph DCS-500, Shimadzu Co., Kyoto, Japan).

2.5. Knoop hardness

Knoop hardnesses of the cured samples (6 mm diameter × 3 mm) were measured on the upper and the lower surfaces by the condition of load (10 g) and load maintenance time (15 s) with a microhardness tester (MVK-H1, Akashi Co., Tokyo, Japan) using a Knoop diamond indentor.

2.6. Electron microscope observation

The cured samples (6 mm diameter × 3 mm) were sanded flat with #1200 silicon carbide paper and microcloth. These were sputter-coated with a 50 nm layer of Au (TWIN COATER JEC-550, JEOL, Tokyo, Japan) for analysis by scanning electron microscopy (SEM) (JSM-6300, Jeol, Tokyo, Japan). SEM photomicrographs with a magnification of 2500 × were taken.

3. Results

3.1. Compressive strength

The compressive strengths of the cured samples as related to filler content level and particle size are presented in Fig. 2. On three resin series (S05, S10 and S15), the compressive strengths of the cured samples increased with the filler content level and the maximum values were 297.2 ± 10.6 MPA (S05), 347.0 ± 8.3 MPa (S10) and 304.2 ± 11.1 MPA (S15), which were on the maximum filler addition. At the same filler content the mechanical properties of S10 were highest, followed by S15 and S05.

3.2. Diametral tensile strength

The result of diametral tensile strength test is shown in Fig. 3. Filler addition (80 wt %) of S10 indicated a maximum value 17.5 ± 3.0 MPa and 70 wt % addition of S15 indicated a minimum value 8.0 ± 1.2 MPa. S10 showed the highest diametral tensile strength, following S15, and S05 on the same filler content.

3.3. Knoop hardness

Fig. 4 shows the result of the Knoop hardness tests. S15 indicated the higher value on the upper rather than lower surface, while S05 and S10 indicated about the same values. With an increase in the amount of the filler addition, the Knoop hardnesses of the cured samples increased and S10 indicated the maximum value (83.0 ± 6.2 on the upper surface, 84.1 ± 3.7 on the lower surface), with added 80 wt % fillers. The minimum values of Knoop hardness were S05 of 70 wt % filler content on both surfaces (57.8 ± 4.2 on the upper surface, 53.2 ± 2.7 on the lower surface).

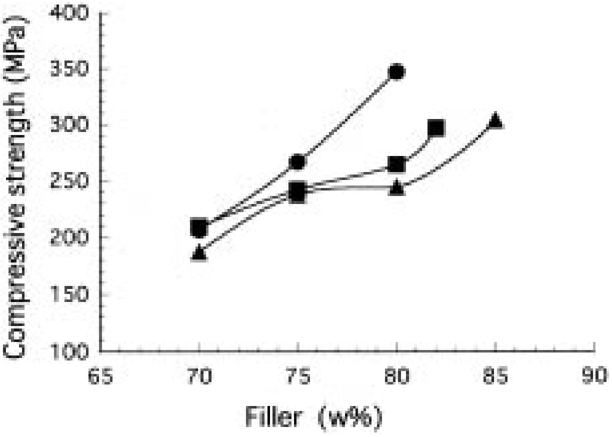

Figure 2 Graphical comparison of compressive strength values of the cured samples investigated. (▲) S05, (●) S10 and (■) S15.

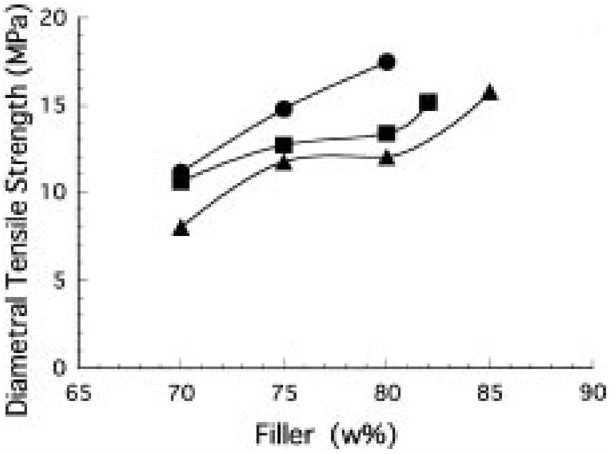

Figure 3 Graphical comparison of diametral tensile strength values of the cured samples investigated. (▲) S05, (•) S10 and (■) S15.

3.4. Scanning electron microscope (SEM) observation

Fig. 5 shows the SEM image of S05, S10 and S15 which contain 80 wt % fillers, respectively.

Bubbles of about 1.0 μm in diameter and the cracks between the fillers and the matrix were admitted in each cured sample, no matter what the filler content was.

4. Discussion

In the development of microwave-curing composite resin inlay, the form, composition, particle size and content of fillers are very important factors, as well as resin matrix [24–26].

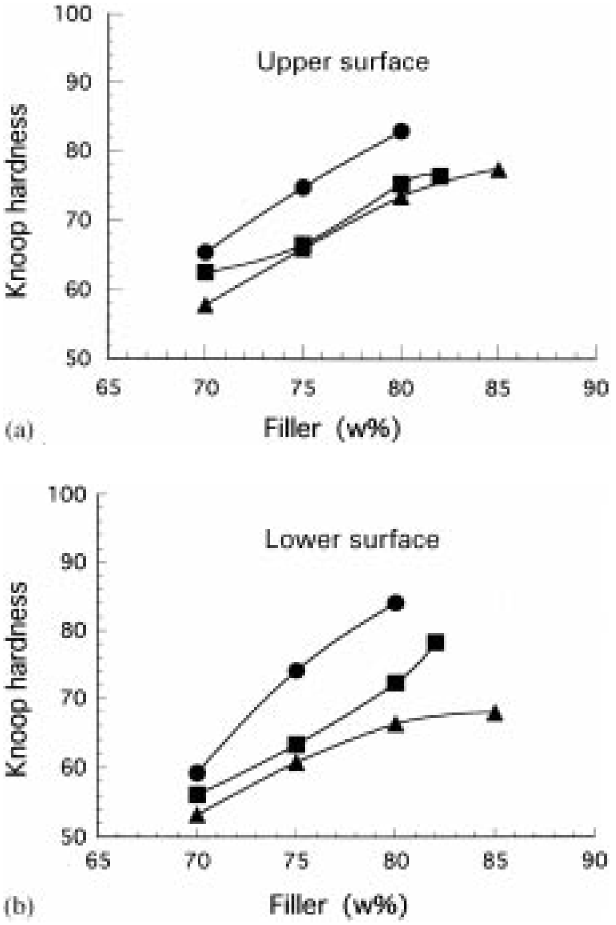

Figure 4 Graphical comparison of Knoop hardness values of the cured samples: (a) on the upper surface, (b) on the lower surface investigated. (▲) S05, (•) S10 and (■) S15.

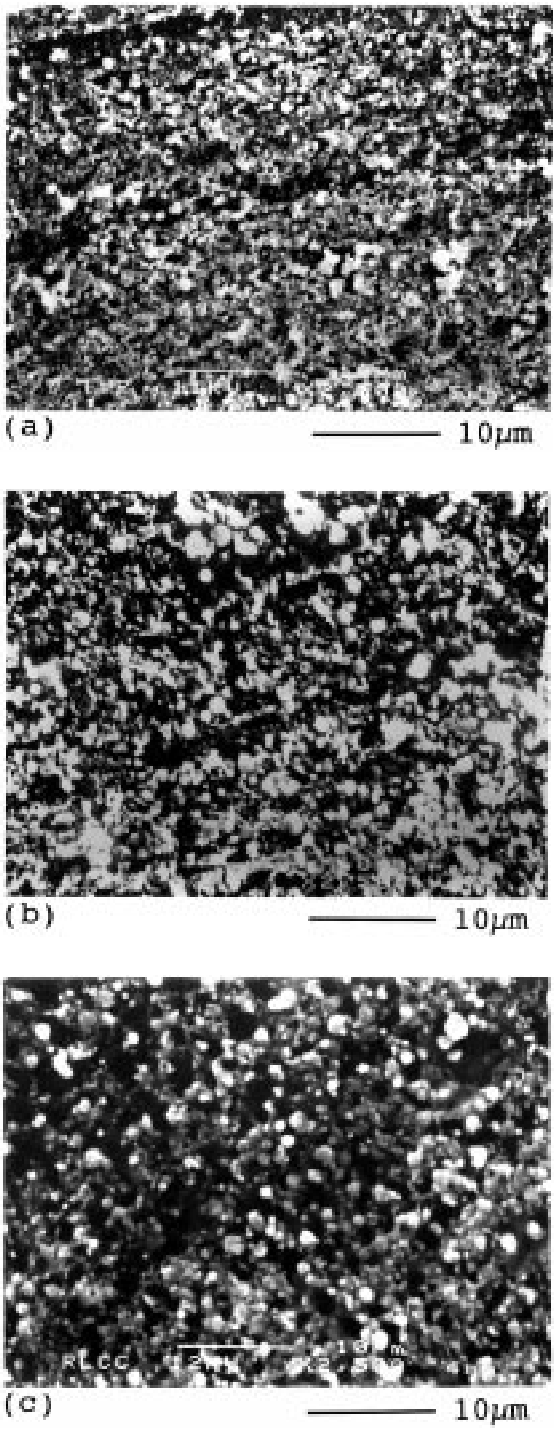

Figure 5 SEM photomicrographs of the cured samples containing 80 wt % quantities of: (a) 0.45 μm; (b) 0.96 μm and (c) 1.46 μm filler particles.

The fillers used in this research were spherical silica fillers, which had the advantage of high density filling, without making fillers to the hybrid unlike the irregular type fillers, examined the effect of filler size and content to the properties of composite resins. As a result, the filler content level had a significant effect on the mechanical properties of composite resins as well as filler particle size. The increased filler content in three kinds of filler particles (0.45, 0.96 and 1.46 μm) resulted in increasing compressive strength, diametral tensile strength and Knoop hardness and indicated the highest values at the operative mixing limit, respectively. This suggests that the polymerization by the microwave was intensely affected by viscosity of base monomer connected with the operative mixing limit. The effect of particle size was that the mechanical properties of S10 were highest, followed by S15 and S5 on the same filler content. S10 had about the same Knoop hardness on both surfaces of the cured sample, which means uniform polymerization occurred in the cured sample.

The microstructures of these cured samples was observed by SEM. Bubbles of about 1.0 µm in diameter were admitted as well as cracks between the fillers and the resin matrix in each sample, while there were no bubbles in the cured samples without fillers. This is caused by the excessive absorption of microwave energy in fillers and surface treatment materials of fillers. Therefore, the composition of the filler and surface treatment material of filler need to be improved, to make it harder to absorb the microwave energy.

References

1. K. F. LEINFELDER, T. B. SLUDER, J. F. F. SANTOS and J. T. WALL, *Oper. Dent.* **5** (1980) 57.
2. F. LUTZ and R. W. PHILLIPS, *J. Prosthet. Dent.* **50** (1983) 480.
3. M. BRAEM, P. LAMBRECHTS, V. VAN DOREN and G. VANHERLE, *Dent. Mater.* **2** (1986) 106.
4. I. E. RUYTER and H. OYSAED, *J. Biomed. Mater. Res.* **21** (1987) 11.
5. P. M. LAMBRECHTS and G. VANHERLE, *Oper. Dent.* **12** (1987) 53.
6. K. F. LEINFELDER, *Adv. Dent. Res.* **2** (1988) 115.
7. F. LUTZ, R. W. PHILLIPS, J. F. ROULET and J. C. SETCOS, *J. Dent. Res.* **63** (1984) 914.
8. J. L. FERRACANE, H. MATSUMOTO and T. OKABE, *ibid.* **64** (1985) 1332.
9. I. E. RUYTER and H. OYSAED, *J. Biomed. Mater. Res.* **21** (1987) 11.
10. J. F. MCCABE and S. KAGI, *Br. Dent. J.* **171** (1991) 246.
11. A. PEUTZFELDT and E. ASMUSSEN, *J. Prosthet. Dent.* **66** (1991) 322.
12. M. BAFILE, G. N. GRASER, M. L. MYERS and E. K. H. LI, *ibid.* **66** (1991) 269.
13. P. W. WALLACE, G. N. GRASER, M. L. MYERS and H. M. PROSKIN, *ibid.* **66** (1991) 403.
14. R. A. DYER and J. A. HOWLETT, *J. Dent.* **22** (1994) 236.
15. R. E. MCKINSTRY and I. ZINI, *J. Prosthet. Dent.* **66** (1994) 269.
16. M. L. SWARTZ, B. K. MOORE, R. W. PHILLIPS and B. F. RHODES, *ibid.* **47** (1982) 163.
17. M. CROSS, W. H. DOUGLAS and R. P. FIELDS, *J. Dent. Res.* **62** (1983) 850.
18. A. J. DE GEE, E. C. TEN HARKEL and C. L. DAVIDSON, *ibid.* **63** (1984) 532.
19. Y. LI, M. L. SWARTZ, R. W. PHILLIPS, B. K. MOORE and T. A. ROBERTS, *ibid.* **64** (1985) 1396.
20. H. ST. GERMAIN, M. L. SWARTZ, R. W. PHILLIPS, B. K. MOORE and T. A. ROBERTS, *ibid.* **64** (1985) 155.
21. M. TAIRA, H. URABE, T. HIROSE, K. WAKASA and M. YAMAKI, *ibid.* **67** (1988) 24.
22. H. URABE, K. WAKASA and M. YAMAKI, *J. Mater. Sci.: Mater. Med.* **1** (1990) 55.
23. Council on Dental Materials and Devices, *J. Amer. Dent. Ass.* **94** (1977) 1191.
24. F. LUTZ, J. C. SETCOS, R. W. PHILLIPS and J. F. ROULET, *Dent. Clin. North Amer.* **27** (1983) 697.
25. F. LUTZ and R. W. PHILLIPS, *J. Prosethet. Dent.* **50** (1983) 480.
26. R. G. GRAIG, in ''Posterior composite resin dental restorative materials'' (3M Co., St. Paul, 1986) p. 199.

Received 5 January
and accepted 3 August 1998