Application of flake shaped glass (Glass Flake®) filler for dental composite resin

Motohiro UO,† Akiko SASAKI, Junko MASUDA, Juichi INO* and Fumio WATARI

Department of Biomedical Materials and Engineering, Graduate School of Dental Medicine, Hokkaido University, North 13 West 7, Kita-ku, Sapporo 060–8586
***Nippon Sheet Glass, Chitose-cho, Yokkaichi 510–0051**

For dental composite resin (CR), mechanical strength, estheticity and flowability are required. In this study, flake-shaped glass (FSG; Glass Flake®) was employed as the filler of CR. FSG is composed of thin glass platelets with a flat, smooth surface. FSG-filled CR (FSG/CR) showed good transparency compared to an irregularly shaped filler. The Vickers hardness of FSG/CR was increased with increasing FSG content and was comparable to that of a commercial CR at 70 wt % FSG content. The compressive strength of FSG/CR with silanized FSG was also comparable to that of the commercial CR. The flowability of uncured FSG/CR was much higher than that of the CR containing the irregularly shaped filler and that of commercial flowable CR with the same filler content. Thus, FSG will be useful as a filler of dental CR that provides estheticity, mechanical strength and flowability.

Key-words : Flake-shaped glass, Composite resin, Dental material, Mechanical strength, Estheticity, Flowability

[Received February 8, 2010; Accepted April 15, 2010]

1. Introduction

Composite resin (CR) is widely used as a restorative material because of its good estheticity, easy handling and durability. Generally, the mechanical properties, e.g. hardness, compressive strength and elastic modulus, of CR are increased with the filler content.[1)–6)] A smaller filler size or spherical-shaped filler allows close filler packing and improves the mechanical properties. It also contributes a smooth surface and gloss to the polished surface. However, increased filler content also increases the consistency of the uncured composite and the handling properties are negatively affected.[7)] Flowable CR is recently of interest because of its desirable flow for injection into cavities. To obtain the flowablity of CR, filler size or filler content is reduced, and its mechanical properties such as the hardness, abration resistance and compressive strength are decreased.[8)]

On the other hand, flake-shaped glass (FSG; Glass Flake®) particles are used for paint and linings to improve their chemical durability and for plastics to improve their mechanical properties. FSG particles are thin, flat glass platelets. The particle size varies from several to several hundred micrometers and the thickness is about 5 µm. FSG has a quite flat surface, so it mixes well with solvants and the mixture has good flowability. These features of FSG are desirable as a filler of CR to increase the flowability without reducing the filler content. In addition, the flat surface of FSG has high optical transparency, so a contribution to the estheticity of CR can also be expected.

In this study, CRs with various contents of FSG were prepared and their compressive strength, Vickers hardness, optical transparency and flowability were estimated.

2. Experimental procedures

A mixture of bisphenol A-diglycidyl methacrylate (Bis-GMA; Shin Nakamura Kagaku, Wakayama, Japan) and triethylene dimethacrylate (MMA; Kanto Kagaku, Tokyo, Japan) was used as the resin matrix. Bis-GMA and MMA were mixed to a 2:1 weight ratio. Camphoroquinone (Kanto Kagaku, Tokyo, Japan) and benzoil peroxide (Tokyo Kasei, Tokyo, Japan) were added to the mixture at 0.5 wt % as catalysts.

FSG powder (Glass Flake®, approximately 45 µm in diameter and 5 µm in thickness, Nippon Sheet Glass, Tokyo, Japan) was mixed in the above base monomer. The content of FSG filler was varied from 30 to 80 wt %. A part of FSG was silanized with 3-methacryloxypropyl trimethoxysilane (Tokyo-Kasei, Tokyo, Japan) using a reported method.[9)] Crushed silica particles (CSF; Tatsumori, Tokyo, Japan), which are used as a conventional silica filler of commercial CR, were used as a reference for the filler material.

To prepare CR, FSG filler and the base resin matrix were mixed in a mortar to obtain a specific filler content. The mixture was put in a Teflon mold (4 mmϕ * 6 mm$t$) and cured with a light curing unit (αLight II, Morita, Tokyo, Japan) for 15 minutes on both sides. The cured specimen was removed from the mold. The specimens for the mechanical tests were heated at 100°C for 1 hour followed by photo curing to confirm the polymerization in the center of thick specimens. After the polymerization, the specimen was polished and subjected to compression tests and Vickers hardness tests. Four replicates were prepared for each mechanical test. The compression test was carried out with a universal testing machine (Model 4204, Instron, Canton, USA) with a loading speed of 0.5 mm/min. The micro Vickers hardness was measured with a 200g load. (MVK-C, Akashi, Yokohama, Japan). The microstructure of the prepared CR was observed by SEM (S-4000, Hitachi, Tokyo, Japan). The commercial CR (Clearfil AP-X, Kuraray Medical Inc., Tokyo, Japan) was also cured and evaluated with same method as the reference material.

The flowability of the uncured CR was estimated using the method reported by Yoshida et al.[10)] Spread areas with 50 mg of uncured mixtures pressed with a constant load (2g) were used as the index of the flowability. A commercial flowable CR (Metafil Flo, Sun Medical Co. Ltd., Moriyama, Japan) was measured as a reference.

† Corresponding author: M. Uo; E-mail: uo@den.hokudai.ac.jp

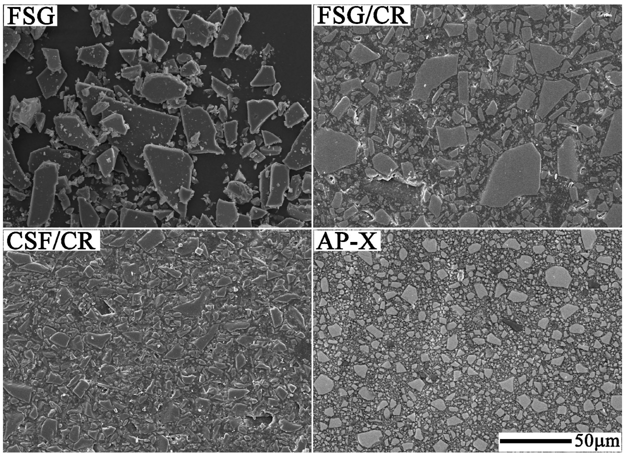

Fig. 1. SEM images of FSG, FSG/CR (60 wt %), CSF/CR (60 wt %) and AP-X.

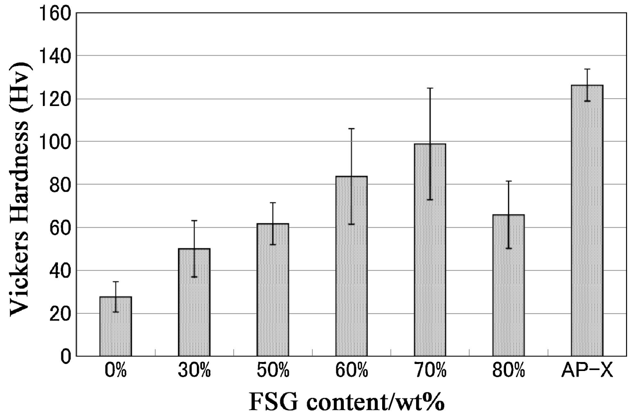

Fig. 3. Differences in Vickers hardness by FSG content.

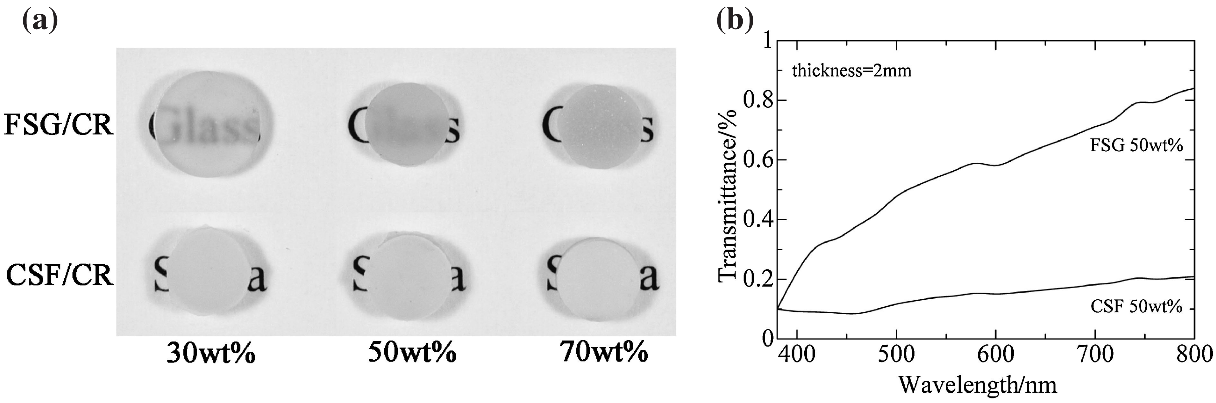

Fig. 2. Optical transparency of polished FSG/CR and CSF/CR disks (2 mm thick). (a) Appearance of disks (b) transmission spectra of disks with 50 wt % of filler content.

In this paper, the following abbreviations are used

CR: composite resin
FSG: flake-shaped glass FSG/CR: FSG-filled CR
CSF: crushed silica filler CSF/CR: CSF-filled CR
AP-X: Clearfil AP-X MF: Metafil Flo

3. Results

SEM images of FSG filler, FSG/CR, CSF/CR and the commercial CR (AP-X) are shown in **Fig. 1**. FSG was platelet-shaped with a flat surface and its size was smaller than 50 μm. The particle size of FSG was larger than that of CSF and the filler contained in AP-X.

Figure 2a shows the transparency of cured FSG/CR and CSF/CR with 30, 50, and 70 wt % filler contents. FSG/CR showed good transparency at 30 wt % filler content and was slightly transparent even at 50 wt % filler content. In contrast, CSF/CR was no longer opaque at 30 wt % filler content. The transmission spectra of FSG/CR and CSF/CR containing 50 wt % filler (2 mm thick) are shown in Fig. 2b. The transmittance of FSG/CR was higher than that of CSF/CR in the entire visible light range.

The Vickers hardness (Hv) of the prepared FSG/CR with various FSG contents and AP-X are shown in **Fig. 3**. Hv was increased with the FSG filler content and the highest Hv was attained at 70 wt % filler content. This Hv was comparable to that of the commercial CR (AP-X). When the FSG content was higher than 70 wt %, Hv decreased as a result of the inhomogeneity of the CR because the filler content was too high.

Figure 4 shows the compressive strength of the prepared FSG/CR with various FSG wt % contents and AP-X. The compressive strength of FSG/CR with less than 60 wt % was slightly lower than that of the base resin without filler. However, with 70 wt % FSG content the compressive strength slightly increased. The silanization of FSG was effective to increase the compressive strength, which was comparable to that of commercial CR (AP-X).

Figure 5 shows the comparison of the flowability of uncured pastes of FSG/CR, CSF/CR and commercial flowable CR (MF). At 60 wt % filler content, FSG/CR showed more than four times higher flowablity than CSF/CR and MF. Even with 70 wt % filler content, FSG/CR showed flowability similar to that of MF.

4. Discussion

The required properties of dental CR are not only mechanical strength (e.g. abrasion toughness, hardness and compressive strength) but also estheticity and flowability. Li et al. reported that a larger filler (15 μm) slightly improved the hardness and the compressive strength of CR compared with a smaller filler (2 μm).[1] Hara et al. reported that an increase of filler content improved the elastic modulus, Rockwell hardness and tensile strength of CR, but the filler size did not affect those properties. The compressive strength was decreased with increasing filler size from 1.3 to 45 μm.[2] Miyasaka also reported decreases of compressive and diametral tensile strengths with the filler size for both irregular and spherical shapes in a binary system of hybrid fillers

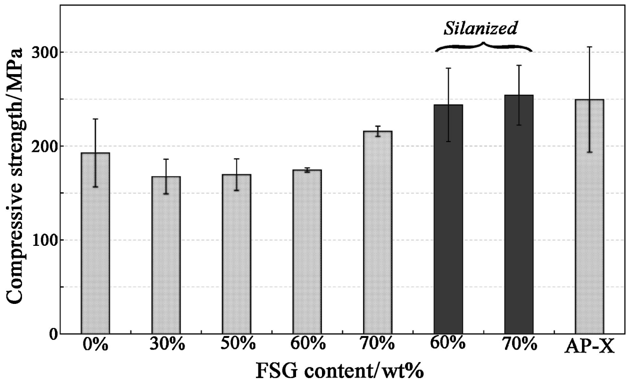

Fig. 4. Differences in compressive strength by FSG content and silanization.

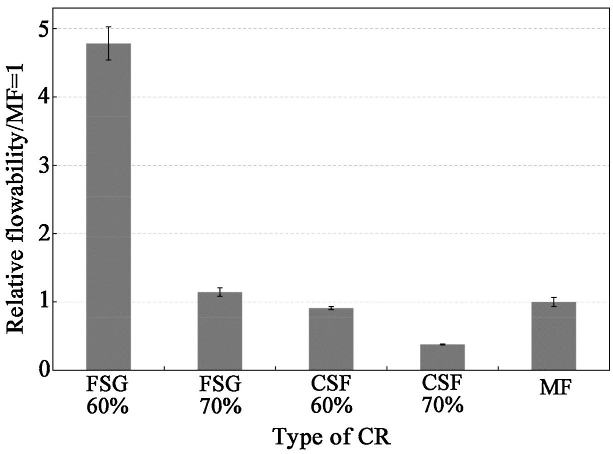

Fig. 5. Relative flowability of uncured FSG/CR, CSF/CR and MF. MF was used as the flowability standard (=1).

and the properties did not depend on the filler shape.[3] Iwasaki et al. reported an increase of the flexural strength and elastic modulus of CR by increasing the size of a spherical fillers and they were slightly higher than with an irregularly shaped filler of similar size.[4] Masouras et al. reported the effect of the filler on the Young's modulus, which was increased with increasing filler content,[6] and an irregular filler had a higher modulus than a spherical filler.[5] Because of their high flowability, recently developed flowable CRs can be operated by injection with a syringe in areas it is difficult or impossible to access with traditional CRs.[8),11] For the flowable CR, low viscosity of the uncured CR paste is required. However, an increase of the filler content increases the viscosity of uncured paste.[7),12] Thus, a filler that does not decrease the flowability with high filler content is required.

In this study, FSG was employed as the filler of CR. The prepared FSG/CR showed good transparency even at 50 wt% filler content compared to CSF/CR, as shown in Fig. 2. The large particle size and quite flat surface of FSG suppressed light scattering, so the optical transparency of FSG/CR was improved compared to the irregularly shaped filler (CSF). High optical transparency would be effective to improve the estheticity of CR. The Vickers hardness and compressive strength of FSG/CR with higher filler content or with silanized FSG were comparable to those of the commercial CR (AP-X), as shown in Figs. 3 and 4. AP-X was reported to have highest Vickers hardness among recent CRs[13] and is used as a posterior filling material. Therefore, FSG/CR could provide sufficient hardness and compressive strength as a dental filling material. On the other hand, the previous studies[2),3] showed that filler size larger than 10 µm decreased the compressive strength. FSG has an anisotropic shape with a large size (45 µm in average) and small thickness (5 µm). Therefore, the compressive strength would not be much decreased in spite of the large filler size of FSG. The flowability of prepared CR was greatly improved by using FSG compared to CSF (Fig. 5). Yoshida et al. reported the filler content and flowability of five commercial flowable CRs.[10] They found that the filler content of MF was 60 wt% and the contents of other flowable CRs varied from 62 to 77 wt%. Their flowablity values varied from 0.82 to 1.2, as normalized using MF as the standard (=1). The 60 wt% FSG-containing CR showed more than four times higher flowability than MF, which contains 60 wt% filler. Even with 70 wt% FSG filler content, the flowability was the same as that of MF. The flat and smooth surface of FSG and large filler size decreased the friction between filler particles in the uncured CR paste. Therefore, FSG filler was effective to improve the flowability of uncured CR paste. Thus, high filler loading without decreasing the flowability could be attained using FSG. This feature would be useful for a CR filler with sufficient mechanical strength and flowability.

5. Conclusions

In this study, FSG, which is comprised of thin glass platelets, was successfully used as the filler of CR. FSG/CR showed good transparency compared to an irregularly shaped filler (CSF). The Vickers hardness and compressive strength were comparable to those of a commercial CR. The flowability of uncured FSG/CR was much higher than those of CSF/CR and the commercial flowable CR with the same wt% filler content. Thus, FSG could be useful as a filler of dental CR that provides estheticity, mechanical strength and flowability.

Acknowledgment This work was supported by Grant-in-Aid for Challenging Exploratory Research No. 21659449, from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

References

1) Y. Li, M. L. Swartz, R. W. Phillips, B. K. Moore and T. A. Roberts, *J. Dent. Res.*, 64, 1396–1401 (1985).
2) R. Hara and K. Nemoto, *J. J. Dent. Mater.*, 5, 1–16 (1986) [in Japanese].
3) T. Miyasaka, *Dent. Mater. J.*, 15, 98–110 (1996).
4) N. Iwasaki, H. Takahashi, Y. Aoyagi, J. Y. Jin, K. Tonami and F. Nishimura, *J. J. Dent. Mater.*, 22, 194–200 (2003) [in Japanese].
5) K. Masouras, R. Akhtar, D. C. Watts and N. Silikas, *J. Mater. Sci. Mater. Med.*, 19, 3561–3566 (2008).
6) K. Masouras, N. Silikas and D. C. Watts, *Dent. Mater.*, 24, 932–939 (2008).
7) J. H. Lee, C. M. Um and I. B. Lee, *Dent. Mater.*, 22, 515–526 (2006).
8) S. C. Bayne, J. Y. Thompson, E. J. Swift, Jr., P. Stamatiades and M. Wilkerson, *J. Am. Dent. Assoc.*, 129, 567–577 (1998).
9) M. Uo, G. Sjögren, A. Sundh, M. Goto, F. Watari and M. Bergman, *Dent. Mater. J.*, 25, 626–631 (2006).
10) E. Yoshida, R. Nomoto, S. Hirabayashi, K. Nitta and S. Hirano, *J. J. Dent. Mater.*, 27, 1–8 (2008) [in Japanese].
11) S. Beun, C. Bailly, J. Devaux and G. Leloup, *Dent. Mater.*, 24, 548–555 (2008).
12) I. B. Lee, H. H. Son and C. M. Um, *Dent. Mater.*, 19, 298–307 (2003).
13) R. J. Scougall-Vilchis, Y. Hotta, M. Hotta, T. Idono and K. Yamamoto, *Dent. Mater. J.*, 28, 102–112 (2009).