Acta Odontologica Scandinavica, 2005; 63: 371–375

Taylor & Francis
Taylor & Francis Group

ORIGINAL ARTICLE

Effect of 10 wt% spherical silica filler addition on the various properties of conventional and resin-modified glass-ionomer cements

ROSALINA TJANDRAWINATA[1,2], MASAO IRIE[1] & KAZUOMI SUZUKI[1,3]

[1]*Department of Biomaterials, Okayama University Graduate School of Medicine and Dentistry, Okayama, Japan,* [2]*Department of Dental Materials, Faculty of Dentistry Trisakti University, Jakarta, Indonesia, and* [3]*Research Center for Biomedical Engineering, Okayama University, Shikata-cho, Okayama, Japan*

Abstract

In this study, we evaluated the effects of 10 wt% spherical silica filler (SSF) addition on 24-h compressive strength, modulus of elasticity, water uptake, and immediate setting shrinkage of conventional glass-ionomer (Fuji II and Experimental) and resin-modified glass-ionomer (Fuji II LC EM) cements. The glass-ionomer cement powders were modified by being mixed with 10 wt% SSFs with an average particle diameter of 0.3 μm. The materials were mixed to consistencies similar to the flow of Fuji II mixed with a powder-liquid ratio of 2.7 : 1 (w/w). The 24-h compressive strength, modulus of elasticity, water uptake, and immediate setting shrinkage were observed and the results compared with the original materials mixed with similar flow. The addition of SSF increased the compressive strength value to 1.1 times, while the increase of moduli of elasticity was 1.10 to 1.35 times. In general, the addition of SSF decreased the 24-h water uptake to 80–90% and reduced the immediate setting shrinkage to 70–79% of the original materials. The addition of 10 wt% SSF improved the characteristics of conventional and resin-modified glass-ionomer cement.

Key Words: *Glass ionomer, mechanical properties, setting shrinkage, spherical silica filler*

Introduction

Since introduced in the early 1970s, glass-ionomer cement (GIC) has been modified substantially to improve its brittleness, limited mechanical strength, and low wear resistance [1]. To enhance the strength of conventional GICs (CGIC), the polyacrylic acid constituent has been replaced by hydrophilic monomer, and the result is a chemical and light curable material known as hybrid ionomer or resin-modified GIC (RMGIC) which has become popular [2]. However, polymerization results in a greater degree of shrinkage upon setting. The lower water and carboxylic acid content reduces the ability of the cement to wet the tooth substrates, and this can greatly increase microleakage which, in turn, leads to marginal gap formation, marginal discoloration, postoperative sensitivity, and secondary caries [2,3]. Since clinical study of marginal gap formation is more expensive and time-consuming than a laboratory test, an *in vitro* determination of the setting shrinkage of the materials in a Teflon mold is more usual. This has a linear correlation with the marginal gap in the tooth cavity [4,5].

The mechanical properties of RMGICs are superior to those of CGICs [2,6,7]. To improve cement strength and wear resistance, metals or glass short fibers have been added to the powder component of GICs [1,8,9]. In this research, spherical silica fillers (SSFs) were added to GIC powder; up to 20 wt% increases the workability and mechanical properties of RMGIC [10,11]. Although the powder : liquid ratio influenced the strength characteristics of the materials, different GICs with the same powder : liquid ratio reflected different consistency. The consistency of RMGICs correlated with gap formation and influenced the strength characteristics of the specimen [5,11]. However, on increasing the strength by increasing the powder : liquid ratio decreased the flow and made the mixing of GIC more difficult [12].

The aim of this study was therefore to analyze the effect of 10 wt% SSF on the 24-h compressive strength and immediate setting shrinkage of GIC, which are

Correspondence: Masao Irie, Department of Biomaterials, Okayama University Graduate School of Medicine and Dentistry, 2-5-1, Shikata-cho, Okayama 700-8525, Japan. Tel: +81 86 235 6668. Fax: +81 86 235 6669. E-mail: mirie@md.okayama-u.ac.jp

(Received 8 April 2005; accepted 7 June 2005)

ISSN 0001-6357 print/ISSN 1502-3850 online
DOI: 10.1080/00016350500206819

kept at similar flow. The hypothesis is that the addition of 10 wt% of SSF at a similar flow increases the 24-h compressive strength and reduces the immediate setting shrinkage of GIC.

Material and methods

The GIC materials used in this study were from the GC Corporation, Tokyo. They are: (i) CGIC, (a) Fuji II (Code: FII), powder lot no. 9905271, liquid lot no. 0101301, (b) experimental GIC containing fluoroaluminosilicate powder (Code: EXP), powder lot no. 171201, and (ii) RMGIC: Fuji II LC EM (Code: FLC), powder lot no. 0205211, liquid lot no. 0204301.

Experimental GIC was also analyzed, since it contained fluoroaluminosilicate powder with no other elements or coloring agents.

The filler used was unsilanized SSF (KBM 503; Shin-Etsu Chemical, Tokyo, Japan) with an average particle diameter of 0.3 μm. The SSF (20 g) was silanized to modify the RMGIC powder as previously described [11].

The powders were initially mixed with the 10 wt% SSF and 5.0 ± 0.05 g of mixture was shaken in a 50-ml bottle by hand with a frequency of 120 cycle/min and amplitude of 20 cm. The prepared cement powders were noted as FII, FII10, EXP, EXP10, FLC, FLC10 (FLC with 10 wt% untreated silica filler), FLCS10 (FLC with 10 wt% silanized silica filler), which show the type of powder and its filler content. Then the CGIC powders were mixed with Fuji II liquid, while the RMGIC powder was mixed with Fuji II LC EM liquid. The powder and liquid were weighed using an electric balance (AJ 100; Mettler, Greifensee, Switzerland). The consistency of FII mixed with a powder : liquid of 2.7 : 1 (w/w) was chosen as the baseline in this study. The FLC was also prepared with a powder : liquid ratio of 3.0 : 1 (w/w), as recommended by the manufacturer, and was noted as FLCO.

All procedures, except mechanical testing, were performed in a thermo-hygrostatic room kept at 23 ± 0.5°C and 50 ± 2% relative humidity.

Flow test

The powder : liquid ratio of materials was determined based on obtaining a similar flow with FII mixed with a powder : liquid ratio of 2.7 : 1 (w/w) and hand mixed for 30 s. The mixture was poured into a syringe (Centrix C-R Syringe System; Centrix, Shelton, Ct., USA) to ease flow into the mold and to reduce porosity. Then 0.05 cc of mixed GIC was put on a glass plate and covered with another glass plate. Less than 2 min after the start of mixing, pressure (of 127 N) was applied on the top of the plate for 1 min. The maximum and minimum diameters of material were measured using vernier calipers (U39818; Mitsutoyo, Kawasaki, Japan) and the mean result was recorded to the nearest 0.05 mm.

Twenty-four-hour compressive strength and modulus of elasticity

The prepared cylindrical Teflon split mold for compressive strength measurement had a depth of 6.0 mm and diameter of 3.0 mm. Six specimens were made of each material and prepared as outlined in ISO 7489-1986 [13]. The compressive strength measurements were performed after storage in water at 37°C in an incubator for 24 h. Prior to testing, the dimensions of the specimens were measured using a digital micrometer (Mitsutoyo no. 293-421-20, Tokyo, Japan). The accepted specimen size was 3.0 ± 0.03 mm in diameter and 6.0 ± 0.06 mm in height. Strength was measured using a universal testing machine with a cross-head speed of 0.5 mm min^{-1} (Autograph DCS-2000; Shimadzu, Kyoto, Japan).

The modulus of elasticity of the materials was analyzed from the compressive strength measurement chart converted to a stress-strain graph, and the slope of the elastic region was counted as the modulus of elasticity in MPa, which was subsequently converted to GPa [2].

Twenty-four-hour water uptake

Before immersing in distilled water, the specimens for compressive strength measurement were weighed using an electric balance. They were weighed again after immersion in distilled water for 24 h at 37°C in an incubator and dried for 1 min on the Kim Wiper (S-200; Kimberly-Clark Worldwide Inc., Crecia Corp, Tokyo, Japan). The increased weight of the specimen was expressed as a percentage.

Immediate setting shrinkage in the teflon mold

Since it was reported that the marginal gap reduced significantly after 24-h water sorption [14], the setting shrinkage was determined directly after hardening. Hand mixing was for 30 s and preparation time for 30 s. The mixed GIC was put in the Teflon mold (with a depth of 1.5 mm and a diameter of 3.5 mm) placed on a silicone oil-coated glass plate. After setting, the degree of setting shrinkage was inspected under a portable microscope as previously described [4,5]. The sum of the maximum gap width and the opposing gap width (if any) was the marginal gap in the Teflon mold (Figure 1). The percentage mean value of gaps was calculated and expressed as the immediate setting shrinkage of the material.

Statistical analysis

The original and SSF added materials were compared statistically using the *t*-test, while correlations among compressive strength, modulus of elasticity, water sorption, and setting shrinkage were determined using Pearson's product-moment correlation [15].

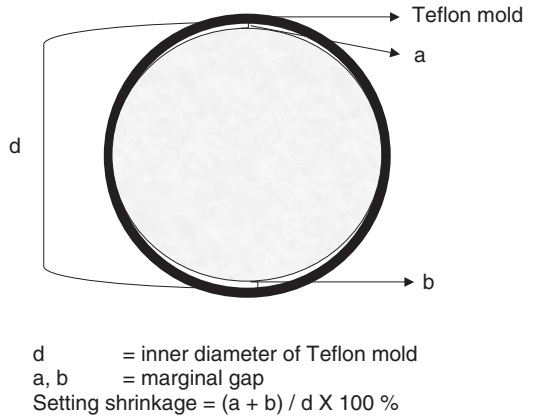

Fig. 1. Immediate setting shrinkage measurement in the Teflon mold.

Results

The results of the flow test are given in Table I, and those of compressive strength test, modulus of elasticity, and water sorption in Table II. Of all materials in this study, EXP10 showed the highest compressive strength and modulus of elasticity values. The increased compressive strength value due to the addition of SSF was about 10%, while for moduli of elasticity it was 10–35%. In CGIC, the specimens with SSF showed compressive strength and modulus of elasticity values higher than the original materials. An addition of 10 wt% untreated filler to FLC resulted in no significant difference in the compressive strength of FLC, but increased the modulus of elasticity significantly. By contrast, the addition of 10 wt% silanized filler increased the compressive strength and modulus of elasticity of FLC significantly.

The highest water uptake value was shown by FLC. The addition of SSF decreased the 24-h water uptake to 80–90% of the original materials except in the FII10 specimens.

The analysis of setting shrinkage gave a better result for the materials with SSF (Table III). The addition of filler reduced the immediate setting shrinkage to 70–79% of the original materials.

The analysis of correlation among the parameters indicated significant correlation ($p<0.05$) between compressive strength and modulus of elasticity, compressive strength and water uptake, and between water uptake and setting shrinkage of the RMGIC group. A lower significant level of correlation ($p<0.10$) was shown between compressive strength and modulus elasticity of the CGIC group, between compressive strength and setting shrinkage of the RMGIC group, and between the increasing rates in compressive strength—decreasing in setting shrinkage.

Discussion

In dental practice, comparing the characteristics of the materials with similar flow is useful. While the manufacturers usually supply the cements as powder and liquid components with a recommended mixing ratio, relative proportions are generally determined by the technical experience of the operator. Variations in hand-mixed cement consistencies utilized by practitioners are expected as a result of proportioning the powder by eye or with the aid of scoops, where the volume of powder dispensed is dependent upon the method of filling the scoop [16–18], on the positioning of the liquid bottle when held to disperse a drop of liquid, and on the drop dispensed, all of which vary due to the inclusion of air bubbles [17,18].

Another aspect to be considered is methacrylate and hydroxyethyl methacrylate (HEMA) content in the liquid component of RMGIC. The allergenicity of methacrylate has been reported and HEMA may change the ability of the monocyte to direct an immune response if challenged by plaque or other agents [2]. From the flow test result, it is obvious that the addition of 10 wt% SSF will increase the flow of GIC and will extend the workability of the material. Thus, increasing the powder : liquid ratio by adding SSF to the powder of RMGICs that do not influence the flow of the material will be more acceptable due to reduction of the amount of liquid used and also the release of free monomers from the filling materials. It was observed that the spherical type of filler facilitated the mixing of powder and liquid of the GIC [11].

The compressive strength test was chosen since this is a standard procedure for measuring the mechanical property of GIC [13]. In the RMGIC group mixed with higher powder : liquid ratio, FLC had a lower flow

Table I. Powder : liquid ratio and flow of glass-ionomer cement

	Material	Code	P/L (w/w)	Flow (mm) Mean (SD)
CGIC	Fuji II	FII	2.7 : 1	34.43 (1.47)
	Fuji II + 10 wt% spherical silica filler	FII10	2.9 : 1	34.39 (1.59)
	Experimental	EXP	2.2 : 1	34.91 (0.70)
	Experimental + 10 wt% spherical silica filler	EXP10	2.7 : 1	34.28 (0.57)
RMGIC	Fuji II LC EM	FLC	3.6 : 1	34.78 (0.95)
	Fuji II LC EM + 10 wt% spherical silica filler	FLC10	4.4 : 1	34.13 (1.07)
	Fuji II LC EM + 10 wt% silanized spherical silica filler	FLCS10	4.2 : 1	34.25 (0.99)
	Fuji II LC EM as a control	FLCO	3.0 : 1	39.52 (1.00)

CGIC = Conventional glass-ionomer cement; RMGIC = resin-modified glass-ionomer cement; P/L = powder : liquid ratio. $n = 6$.

Table II. Twenty-four hour compressive strength, modulus of elasticity, and water uptake characteristics

	Compressive strength (MPa)	Modulus of elasticity (GPa)	Water uptake (%)
Material	Mean (SD)	Mean (SD)	Mean (SD)
FII	186 (19)	3.30 (0.20)	1.07 (0.24)
FII10	214 (14)	3.71 (0.34)	1.19 (0.14)
EXP	202 (5)	3.69 (0.24)	1.09 (0.22)
EXP10	219 (17)	4.15 (0.05)	0.92 (0.18)
FLC	170 (7)*	1.96 (0.04)	3.41 (0.20)
FLC10	174 (10)*	2.37 (0.10)	3.17 (0.16)
FLCS10	193 (5)	2.65 (0.11)	2.58 (0.16)
FLCO	153 (4)	1.59 (0.50)	3.96 (0.33)

$n = 6$, for abbreviations, see Table I.
* No significant difference analyzed using t-test ($P > 0.05$).

Table III. Immediate setting shrinkage in Teflon mold

	Setting shrinkage (%)
Material	Mean (SD)
FII	0.77 (0.11)
FII10	0.56 (0.05)
EXP	0.91 (0.10)
EXP10	0.67 (0.11)
FLC	0.80 (0.08)
FLC10	0.63 (0.07)*
FLCS10	0.56 (0.12)*
FLCO	0.94 (0.10)

$n = 10$; for abbreviations, see Table I.
* No significant difference analyzed using t-test ($P > 0.05$).

but higher compressive strength and modulus of elasticity than the control, FLCO. The better result of FLCS10 was due to the silanization process on SSF, which resulted in chemical bonding between the matrix and silica filler [11]. Filler-matrix coupling enhanced the physical properties of the materials and allowed for adequate wetting and dispersion of the files within the considerably more hydrophobic resin matrices. However, the effect of silane coupling on ion transport from reactive glass has not been thoroughly investigated [19].

As mentioned above, this study showed that correlations among the parameters are significant ($p < 0.05$). Comparison of the control FLCO with FLC showed that increasing powder : liquid ratio reduced the water uptake value. Compared to the CGIC group, the RMGIC group showed more water uptake. The addition of SSF decreased both 24-h water uptake and immediate shrinkage values, since this hydrophobic filler has "no water sorption and no shrinkage" characteristics; besides, this filler also filled the inter-particle space, which could lead to increasing the strength of the material. Actually, water uptake is needed to compensate for the setting shrinkage [14]. Setting shrinkage of the materials in a Teflon mold has a linear correlation with marginal gap in the tooth cavity [4,5]. A gap or microleakage of GIC restoration, especially RMGIC, was created because lower water and carboxylic acid content reduced the ability of the cement to wet the tooth substrates [2]. In addition, a gap was also generated as the adhesion between the tooth cavity and glass ionomer did not resist the stress formed by cement shrinkage [20,21]. After one day of water storage, the curing contraction stresses of the materials are effectively compensated or even converted into expansion stress due to water uptake and swelling [22]. Water absorption of RMGICs and GICs reportedly affects cavity adaptation and reduces microleakage [23–25]. However, clinically, the restoration is usually polished immediately after setting, which results in imperfect closure [26]. A mismatch between the surface of the cavity wall and the opposing surface of the restoration, due to the dimensional changes of the restoration, will prevent this [22]. Although it is suggested that the restoration should be polished after 24 h to prevent gap formation at the material–tooth cavity interface [14,26], the immediate setting shrinkage should be dealt with as soon as possible. An alternative way of compensating for these conditions is therefore to add SSF to the powder of GIC.

Conclusion

Without changing the flow of the mixture, the addition of 10 wt% SSF to both conventional and RMGIC has beneficial effects, especially in increasing 24-h compressive strength and in reducing the immediate setting shrinkage of GIC, which leads to a reduction of the marginal gap in the tooth cavity. In RMGIC, the silanized filler is more advantageous than the addition of untreated filler. Since the clinical success of the GIC is affected by the bonding ability of the material to the tooth structure, these characteristics should be observed in further study.

Acknowledgment

We thank the GC Corporation for providing the materials used in this experiment.

References

[1] McLean JW, Gasser O. Glass-cermet cement. Quintessence Int 1985;16:33–43.
[2] Anusavice KJ. Phillips' science of dental materials, 11th edn. St. Louis: Saunders, Elsevier Science; 2003. p. 179–180. 185–6, 200, 482–4.
[3] Carvalho RM, Pereira JC, Yoshiyama M, Pashley DH. A review of polymerization contraction: the influence of stress development versus stress relief. Oper Dent 1996;21: 17–24.
[4] Irie M, Suzuki K, Watts DC. Marginal gap formation of light activated restorative materials: effects of immediate setting shrinkage and bond strength. Dent Mater 2002;18: 203–10.
[5] Tjandrawinata R, Irie M, Suzuki K. Marginal gap formation and fluoride release of resin-modified glass-ionomer cement: effect of silanized spherical silica filler addition. Dent Mater J 2004;23:305–13.

[6] McCabe JF. Resin-modified glass-ionomers. Biomaterials 1998;19:521–7.
[7] Meyer JM, Cattani-Lorente MA, Dupuis V. Compomer: between glass-ionomer cement and composites. Biomaterials 1998;19:529–39.
[8] Simmons JJ. The miracle mixture: glass ionomer and alloy powder. Texas Dent J 1983;100:6–12.
[9] Kobayashi M, Kon M, Miyai K, Asaoka K. Strengthening of glass-ionomer cement by compounding short fibers with CaO-P_2O_5-SiO_2-Al_2O_3 glass. Biomaterials 2000;21:2051–8.
[10] Miyazaki M, Ando S, Hinoura K, Onose H, Moore BK. Influence of filler addition to bonding agents on shear bond strength to bovine dentin. Dent Mater 1995;11:234–8.
[11] Tjandrawinata R, Irie M, Yoshida Y, Suzuki K. Effect of adding spherical silica filler on physico-mechanical properties of resin-modified glass-ionomer cement. Dent Mater J 2004;23:146–54.
[12] Fleming GJP, Farooq AA, Barralet JE. Influence of powder/liquid mixing ratio on the performance of a restorative glass-ionomer dental cement. Biomaterials 2003;24: 4173–9.
[13] ISO 7489 Dental glass polyalkenoate cements. Technical Committee ISO/TC 106, Dentistry, Switzerland; 1986.
[14] Irie M, Tjandrawinata R, Suzuki K. Effect of delayed polishing periods on interfacial gap formation of Class V restoration. Oper Dent 2003;28:552–9.
[15] Bruning LB, Kintz BL. Computational handbook of statistics, 2nd edn. Glenview: Scot, Foresman; 1977. p. 24–7, 122–4, 240–1, 246–9, 263–5.
[16] Cattani-Lorente MA, Godin C, Meyer JM. Early strength of glass ionomer cements. Dent Mater 1993;9:57–62.
[17] Mount GJ. Glass ionomers: a review of their current status. Oper Dent 1999;24:115–24.
[18] Nomoto R, McCabe JF. Effect of mixing methods on the compressive strength of glass ionomer cements. J Dent 2001;29:205–10.
[19] Tay FR, Pashley EL, Huang C, Hashimoto M, Sano H, Smales RJ, Pashley DH. The glass-ionomer phase in resin-based restorative materials. J Dent Res 2001;80:1808–12.
[20] Feilzer AJ, de Gee AJ, Davidson CL. Curing contraction of composites and glass ionomer cements. J Prosthet Dent 1988;59:297–300.
[21] Sidhu SK. Marginal contraction gap formation of light cured glass ionomers. Am J Dent 1994;7:115–18.
[22] Feilzer AJ, Kakaboura AI, De Gee AJ, Davidson CL. The influence of water sorption on the development of setting shrinkage stress in traditional and resin-modified glass ionomer cements. Dent Mater 1995;11:186–90.
[23] Irie M, Nakai H. The marginal gap and bonding strength of glass ionomers. Dent Mater J 1987;6:46–53.
[24] Fritz UB, Finger WJ, Uno S. Marginal adaptation of resin-bonded light-cured glass ionomers in dentin cavities. Am J Dent 1996;9:253–8.
[25] Irie M, Suzuki K. Marginal seal of resin-modified glass ionomers and compomers: effect of delaying polishing procedure after one-day storage. Oper Dent 2000;25:488–96.
[26] Irie M, Suzuki K. Effect of delayed polishing on gap formation of cervical restorations. Oper Dent 2002;27:59–65.