Dental Materials (2006) **22**, 382–387

dental materials

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The effect of ceramic and porous fillers on the mechanical properties of experimental dental composites

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Received 25 November 2004; accepted 7 April 2005

KEYWORDS
Dental composites;
Filler type;
Ceramic filler;
Porous filler;
Mechanical properties

Summary *Objectives*. The purpose of this study was to investigate the effect of ceramic fillers (containing leucite crystals) and their porosity on the mechanical properties of a new experimental dental composite in order to compare with the properties of composites containing conventional glass fillers.
Methods. In this study, experimental composites were prepared by mixing the silane-treated fillers with monomers. Experimental composites were divided into four groups according to their filler type, amount and porosity. The monomers were composed of 70% Bis-GMA and 30% TEGDMA by weight for all groups. Glass and leucite-containing-ceramic were prepared as different filler types. In order to make fillers porous, leucite-containing-ceramic fillers were treated with HF acid. Camphorquinone and DMAEMA were used as photo initiator system. Post-curing was done for all groups before mechanical testing. Degree of Conversion of composites was measured using FTIR spectroscopy. The diametral tensile strength (DTS), flexural strength and flexural modulus were measured and compared among the groups.
Results. The results showed that the stronger and more porous filler has a positive effect on flexural strength. Porosity of filler increased flexural strength significantly. No significant difference was found in DTS tests among the groups. Flexural modulus was affected and increased by using ceramic fillers. The type of the filler affected the DC of the composite and DC increased by post-curing.
Significance: Flexural strength is one of the most important properties of restorative dental materials. Higher flexural strength can be achieved by stronger and more porous fillers. Investigation into the effect of filler on dental material properties would be beneficial in the development of restorative dental material.
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doi:10.1016/j.dental.2005.04.027

Introduction

The ultimate goal of advanced dental composite research is to produce a material that can be used in all circumstances as an amalgam replacement material [1]. Resin composites are used to replace missing tooth structure and modify the color and contour of the teeth in order to enhance esthetics [2].

Basically, a dental composite is a mixture of silicate glass particles within an acrylic monomer that is polymerized during application [3]. In more detail dental composites consist of four major components [2] which are, an organic polymer matrix, inorganic filler particles, coupling agent, and the initiator-accelerator system [2,4]. The resin forms the matrix of the composite material, binding the individual filler particles together through the coupling agent [4]. The most commonly used monomer is Bis- GMA which was invented by Bowen, and has been available for more than 30 years [4,5].

A wide variety of fillers have been employed in composites to improve the properties and developments in filler technology are responsible for many improvements in composites which are used today [4]. Despite many improvements in this field, dental composites do not have enough toughness, strength and durability in order to be used in stress bearing areas [6]. Most of the fillers which are used to reinforce dental composites are silicate glasses. The glass fillers are not strong enough and exhibit cracks that either cut through the glass fillers or propagate around the filler particles [7]. To overcome the problem, much effort has been made into the use of glass fibers, nano-porous fillers, branched fibers or even ceramic whiskers [3,7].

Apart from fillers, a good bond between fillers and the resin matrix is essential in composites. Silane coupling agents provide the bond between two components in dental composites, but this bond can be degraded by water absorbed by the composites [2]. The idea of increasing the micro-mechanical retention between fillers and resin in order to reinforce the coupling agent was first described by Bowen et al. in 1976. Their strategy was to use multi-phase glasses which can be etched and produce porous fillers [8,9].

Glass–ceramics are polycrystalline materials which consist of a glass matrix and one or more crystalline phases [10,11]. IPS-Empress is one type of the glass–ceramics which contains 40–50% of leucite crystals as a reinforcing agent. The flexural strength of this glass–ceramic is 120–140 MPa which is stronger than glasses [12].

As glass fillers are not strong enough to reinforce dental composite for use under all circumstances, the purpose of this study was to investigate the effect of leucite reinforced glass ceramics as stronger and more porous fillers on the mechanical properties of experimental dental composites.

Materials and methods

Sample preparation

IPS glass–ceramic ingots (IPS Empress, D5 0014, Ivoclar Vivadent, Schaan, Liechtenstein) were obtained collected from Ivoclar-Vivadent and ground. The average size of the filler particles was measured using a particle size analyzer (Analysette 22, Frithsch, Germany).

Fillers were acid washed using 10% HCl for 1 h and passed through a 600 mesh sieve in order to be used as fillers. Some of the ground IPS fillers were etched using 10% Hydroflouric acid for 2 min to make the fillers porous. Glass fillers were collected from Specialty Glass Inc., USA. The SEM images of the porous fillers are shown in Fig. 1.

All fillers were surface treated with 1.0% (wt%) of γ-MPS. γ-MPS was prehydrolyzed for 1 h in an aqueous solution of 70 wt% Ethanol, and 30 wt% double distilled water (pH = 3–4). The treated filler was dried for over 20 days at room temperature [13]. The fillers were then hand-mixed with monomers. The monomer system which was used for all samples consisted of 70 wt% Bis-GMA and 30 wt% TEGDMA. Camphorquinone (CQ) of 0.5 wt% and N,N′-dimethyl Aminoethyl Methacrylate (DMAEMA, Fluka, Germany) of 0.5 wt% were added as the photo initiator system. Samples were divided into four groups based on their filler type and amount.

Table 1 shows a complete description of the different groups. To ensure that penetration of resin monomers into the pores of the porous fillers was completed, a solvent evaporation technique under reduced pressure was used. Some of the resin monomers containing the photo initiator system were dissolved in tetrahydrofuran (THF). Porous fillers were mixed with the solution, and the excess solvent was then evaporated under reduced pressure and sub-ambient light.

Diametral tensile strength (DTS)

Cylindrical specimen were prepared in stainless steel molds (3 mm high, 6 mm diameter) [13,14]. The cured specimens (using Optilux 501, Kerr, USA)

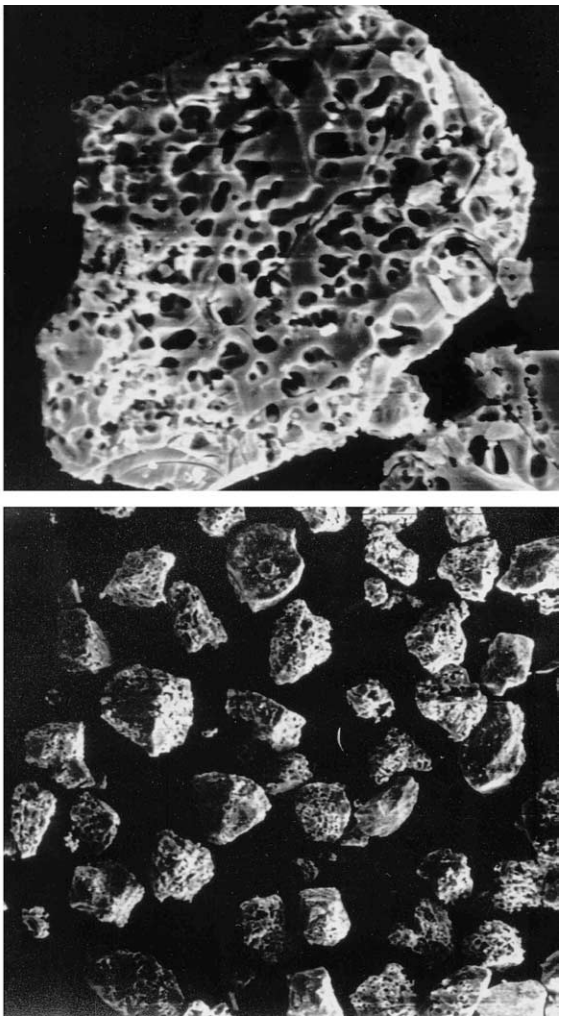

Figure 1 Porous fillers after HF acid etching, used as fillers for experimental dental composites: (a) magnification 2000×, (b) magnification 500×.

were then post-cured at 120 °C for 2 h. All specimens were stored in distilled water for 24 h at 37 °C before the test. DTS was then measured using a universal testing machine (Instron 6025, England) at 10 mm/min cross-head speed and 100 KN load cell. DTS was calculated as $DTS = 2P/\pi DT$, where P, load at fracture; D, diameter; and T, thickness. Six specimen were tested in each group.

Flexural strength

The flexural strength of the bar specimens ($2 \times 2 \times 25\ mm^3$) was measured by a three-point bending test with a span of 20 mm using an Instron 6025 universal testing machine at a cross-head speed of 0.5 mm/min and 1 kN load cell [13,15]. The curing and storage condition of the test specimens were the same as for DTS. The flexural strength was calculated as $3PL/2bd^2$, where P, load at fracture; L, span length; b, width; and d, thickness. Six specimen were tested in each group.

Using this test the flexural modulus was measured for each sample.

Degree of conversion (DC%)

To measure the degree of conversion, the uncured paste of each composite was placed between two polyethylene films, pressed to form a very thin film and the absorbance peaks obtained by the transmission mode of a FTIR spectrometer (EQUINOX 55, Bruker, Germany). The samples were then light-cured for 40 s (Optilux 501, Kerr, USA) and the absorbance peaks were collected for the cured samples. The percentage of unreacted carbon-carbon double bonds was determined from the ratio of absorbance intensities of aliphatic C=C 1638 cm^{-1} against an internal standard before and after the curing of the specimen. The aromatic C···C cm^{-1} absorbance was used as standard. The degree of conversion was calculated as follows: DC% = 100%-percentage of unreacted double bonds.

For all specimens, DC was measured after post-curing using the same method.

Table 1 Composition of the different composite groups used in this study.

Groups composite	Fillers	Resin (matrix)	Other component
G1	Glass (Specialty Glass, USA) 2-4μ. 73 wt% filled	BISGMA/TEGDMA 70/30 wt%	CQ[a] 0.5 wt% DMAEMA[b] 0.5 wt%
G2	Glass-ceramic fillers containing Leucite crystals (IPS.Ingots/Ivoclar-Vivadent) 73 wt% filled	BISGMA/TEGDMA 70/30 wt%	CQ 0.5 wt% DMAEMA 0.5 wt%
G3	Glass-ceramic fillers containing Leucite crystals (IPS.Ingots/Ivoclar-Vivadent) 77 wt% filled	BISGMA/TEGDMA 70/30 wt%	CQ 0.5 wt% DMAEMA 0.5 wt%
G4	Glass-ceramic porous fillers containing Leucite crystals (IPS.Ingots/Ivoclar-Vivadent) 77 wt% filled	BISGMA/TEGDMA 70/30 wt%	CQ 0.5 wt% DMAEMA 0.5 wt%

[a] Camphorquinone.
[b] N,N′-Dimethyl aminoethyl methacrylate.

Statistical method

The reported test values are the average of three for DC and six for mechanical properties.

The results were analyzed and compared using One-way ANOVA, Tukey's test post hoc where there was homogeneity between variances, and Dennett's T3 where there was not. The significant level was considered as 0.05.

Results

Table 2 shows the mean differences in DTS among the groups. As can be seen, there are no significant differences between the DTS of groups ($P>0.05$). Comparison between groups 1 and 2 shows the effect of filler type, and between groups 3 and 4 indicates the effect of filler porosity.

Table 2 shows the flexural strength of all groups. There is a significant difference between the flexural strength of groups 1 and 2 ($P<0.05$), which means the filler type affected this property; in fact, the stronger glass–ceramic filler increased the flexural strength. Comparison between groups 3 and 4 revealed that there is a significant difference between the two groups ($P<0.05$), and porosity of the fillers increased the flexural strength considerably.

Table 2 shows the differences between means considering the flexural modulus. The difference between flexural modulus of groups 1 and 2 is significant ($P<0.05$) which means the filler type affected the modulus of elasticity in the flexural test. There are no significant differences between flexural modulus of groups 3 and 4 which reveals that the modulus has not been affected by the porosity of fillers.

Table 3 shows the DC% of the composites in different groups. Composites containing glass fillers had significantly higher DC% after light curing than group 2, which contained glass–ceramic fillers ($P<0.05$), but there is no significant difference between them after post-curing. Both groups show a significant increase in their DC after post-curing ($P<0.05$).

Table 2 The mean differences of DTS, flexural strength and flexural modulus among the groups.

	Groups	DTS	Flexural strength	Flexural modulus
(1)	Glass, 73 wt%	42.5(3.1)	43.6(3.6)	8.6(0.5)
(2)	Leucite, 73 wt%	39.1(1.7)	56.1(4.4)	10.6(0.8)
(3)	Leucite, 77 wt%	37.5(3.4)	62.6(6.15)	12.6(0.5)
(4)	Leucite-porous, 77 wt%	39.8(1.1)	78.5(8.0)	12.8(0.7)

Table 3 The mean differences of DC (after light and post-curing) among composites containing glass or leucite as fillers.

		DC
Light cured	Glass	71 (2.0)
	Leucite	61 (1.5)
Post-cured	Glass	84 (1.7)
	Leucite	79 (2.5)

Discussion

As dental composites cannot withstand heavy occlusal forces, many ways have been introduced to reinforce them, such as using fibers and whiskers as reinforcing agents. In order to study the effect of filler on the mechanical properties of dental composites other variables should be kept constant, so comparing commercial composites will not give a clear understanding as they differ in other properties, such as amount, size, and shape of the fillers [16,17]. Considering these facts, in this study the effect of filler type and porosity were investigated, while the other variables were kept constant.

Group 2, which contains stronger ceramic fillers, did not show any significant difference in the DTS test. It means that DTS has not been affected by the type of the filler. Material which is going to be tested under compression forces in the DTS test should behave as a brittle material [18]. The load–displacement behavior of tested composites follows a brittle pattern which confirms the reliability of our test results. Although DTS is an acceptable and common test for dental composites [14], it is a separate property and may not be matched with other mechanical properties [18–20]. The DTS values of the experimental composites are in the range of dental composites, 30–55 MPa [2]. The value of DTS for the experimental dental composites was in this range between 30 and 55 MPa which shows acceptable values for the composites.

The flexural strength in group 2 increased significantly. In fact, the ceramic fillers have a positive effect on flexural strength. Flexural

strength is a criterion of durability and longevity of composites [22].

In order to prepare porous filler, they should have two or more phases so that partially elimination of one of the phases makes the fillers porous. Glass–ceramic, which has a crystalline phase, could be a choice as in this study. In working with porous fillers the penetration of resins into the porosities should be considered, otherwise the porosity will reduce the mechanical properties such as flexural strength and fracture toughness, which is a result of incomplete micro-mechanical retention of fillers and resin [23]. One of the ways to mix porous fillers with the resin matrix is by using a vacuum [24]. Decreasing the viscosity of resin with fast evaporating agents is another [25], but it seems that using a vacuum is the best way [1]. In this study, a combination of both methods was used, which is described in Section 2. In the study conducted by Ruddel et al. [1], porous fillers did not increase mechanical properties and it was explained as the result of an incomplete penetration of resins into the pores of the fillers and was assumed that empty pores acted as voids in the system, which resulted in weak points in the composite structure. In their study, results showed that the porous fillers did not significantly affect the DTS. Therefore, considering the condition in the current study it can be suggested that the porosity of fillers has no significant impact on DTS. It has been shown that the DTS alone gives no direct indications as to the particular use of a composite or its potential clinical performance [4].

Flexural strength was compared between groups 3 and 4, in which the porosity of fillers was the main variance. Results showed that the flexural strength increased significantly in group 4. In fact, porous glass–ceramic fillers can be considered as an increasing factor for the flexural strength of dental composites. This property was far above all other groups. The bond between fillers and resins in dental composites is very important, and it is believed to be one of the main factors in reinforcing composites. The high flexural strength in this study can be explained as the result of good interlocking between fillers and resin because of porosity as well as a good bond due to the use of silane coupling agents. In fact, by using porous fillers and with good penetration of resin into the porosity of fillers, a reliable and strong bond between two components can be achieved. The elastic modulus in flexural test or flexural modulus was not affected by porosity.

The DC% of different dental composites varied from 40 to 70% [26]. The DC% of both composites (groups 1 and 2) after light curing remains a high level. The results showed that the DC% of the composite consisting of glass fillers (group 2) is significantly higher than that of composite with ceramic as filler (group 1). As the light transmission in glass fillers is higher than in ceramic fillers, the DC% of the composites containing glass is higher after light curing, but there is no significant difference between the DC% of the groups after post-curing. Post-curing significantly increases DC% in both groups, which is in agreement with the results of other studies [27]. Post-curing is important as it can increase DC and affect the mechanical properties positively [28], even though some papers do not agree with it in general for all mechanical properties [29].

Conclusion

An experimental study was conducted in order to measure the effect of filler type and porosity on the mechanical properties of dental composites. The comparison between composites containing conventional glass fillers and those containing glass–ceramic revealed that the latter increased flexural strength and modulus significantly although it did not affect DTS. In the other part of this study porous fillers was also prepared out of glass–ceramic fillers. The porosity increased flexural strength significantly but did not affect flexural modulus and DTS. Therefore, porous fillers can be considered as an important and applicable way to reinforce dental composites.

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