

Original Research

Synthesis of new dental nanocomposite with glass nanoparticles

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Abstract

Objective(s): The aim of this study was to synthesis new dental nanocomposites reinforced with fabricated glass nanoparticles and compare two methods for fabrication and investigate the effect of this filler on mechanical properties.

Materials and Methods: The glass nanoparticles were produced by wet milling process. The particle size and shape was achieved using PSA and SEM. Glass nanoparticles surface was modified with MPTMS silane. The composite was prepared by mixing these silane-treated nanoparticles with monomers. The resin composition was UDMA /TEGDMA (70/30 weight ratio). Three composites were developed with 5, 7.5 and 10 wt% glass fillers in each group. Two preparation methods were used, in dispersion in solvent method (group D) glass nanoparticles were sonically dispersed in acetone and the solution was added to resin, then acetone was evaporated. In non-dispersion in solvent method (group N) the glass nanoparticles were directly added to resin. Mechanical properties were investigated included flexural strength, flexural modulus and Vickers hardness.

Results: Higher volume of glass nanoparticles improves mechanical properties of composite. Group D has batter mechanical properties than group N. Flexural strength of composite with 10%w filler of group D was 75Mpa against 59 Mpa of the composite with the same filler content of group N. The flexural modulus and hardness of group D is more than group N.

Conclusion: It can be concluded that dispersion in solvent method is the best way to fabricate nanocomposites and glass nanoparticles is a significant filler to improve mechanical properties of dental nanocomposite.

Keywords: Dental nanocomposites, Dispersion, Glass nanoparticles, Mechanical properties

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Introduction

Dental restorative filling composite resins have been introduced to dental community in 1960s (1). They have good aesthetics and are less expensive compared with cast gold and ceramic materials (2), but still suffer from a key shortcoming: definition of mechanical strength (3). Studies have been undertaken to evaluate and improve restorative composite resin against wear and fracture (4). A new approach aiming to achieve improved performance is related to nanotechnology and uses inorganic nanofillers such as nanoparticles to reinforce the polymer matrix. However, it should be pointed out that the effect of nanoparticles enhancement is highly dependent on the size of particles and the level of their dispersion (2). Nanoparticles can be produced from the gas or liquid phase, in high temperature aerosol or plasma reactors or by the sol-gel route (5). These bottom-up processes for fabrication of nanoparticles have one major disadvantage and limiting factor: it is hardly feasible to synthesize nanoparticles with complex composition, like glasses, which contain 5–10 elements (7). A successful route to achieve nanosized particles of such materials is high-energy comminution of the bulk (top-down process). In comminution processes, stirred media mills are employed for various industrial applications, pharmaceuticals, and agrochemicals (5). Stirred media mills show a higher power density than dry ball mills, and are generally more efficient regarding energy requirement for achievement of nanoscaled powders (7).

It is well known that inorganic nanoparticles tend to agglomerate and finally result in a poor dispersion in polymeric matrix. This poor dispersion makes it difficult to achieve a stable complex system and the desirable properties of final products. Therefore, it is important for nanoparticles reinforced materials to ensure good dispersion and stability of particles in the medium (8). Some efforts, such as the surface modification of nanoparticles (9) and changing the system pH and ionic concentration (10), were made to achieve improved compatibility.

Generally, surface modification is commonly

used to change the surface structure of particles in order to enhance the compatibility between the two phases and the dispersion of particles in organic media (11). The compatibility and adhesion between the organic and inorganic phases can be improved after modification, and the mechanical strength of composites would likely undergo significant enhancement by the addition of nanoparticles in conjunction with coupling agents (12).

In this work, first the wet comminution of an amorphous borosilicate glass to achieve nanoparticles were done and then the two methods of composite production including dispersion and non-dispersion in solvent investigated. The effect of filler content on mechanical properties of composite was studied too.

Materials and Methods

Materials

UDMA, TEGDMA, CQ, 4-EDMAB and MPTMS were purchased from Sigma-Aldrich Co. and used without further purification. Amorphous borosilicate glass from Schott Co. with a composition according to table 1 was used as filler.

Table 1. The composition of glass.

Components	SiO ₂	B ₂ O ₃	Na ₂ O+K ₂ O	Al ₂ O ₃
Weight percent	81	13	4	2

Methods

Milling process

The glass was comminuted in a planetary mill for 30 hours with the speed of 400rpm. Hexane was used as the liquid dispersion medium. The particle size and distribution were determined by particle size analyzer.

Silanization of particles

To overcome the agglomeration of particles and also improve bonding to resin the particles were silanized with MPTMS. The 20g of glass particles dispersed in a mixture of 160cc of ethanol and 5g of MPTMS and then sonicated. Ethanol was removed in a rotary evaporator and glass particles washed in a centrifuge by

distilled water. Finally they were dried in the oven and stored in a desiccator. The particle size distribution and SEM photos of these silanized particles were achieved. The particle size distribution fig. 3 and SEM photos of these silanized particles were achieved fig. 4.

Fabrication of composites by dispersion method (Group D)

For production composites by dispersion in solvent method in solvent (group D) modified glass particles of different fractions (5, 7.5 and 10%w) were dispersed in acetone by sonication. This solution was added to resin (70/30 UDMA,TEGDMA in weight ratio) and sonicated again. The mixture surrounded by an ice enclosure to inhibit heat build-up. After the sonication process, the mixture was placed in the rotary evaporator to remove acetone. Then CQ and 4EDMAB (0.5/0.5 the weight ratio) were added in a dark room. Finally the composite were placed in an ultrasonic bath to exit air bubbles and better mixing.

Fabrication of composites by non-dispersion method (Group N)

For production composites in non-dispersion in solvent method (group N). The resin composition was prepared by UDMA/TEGDMA, 70/30 in weight ratio, then various amount of glass particles (5, 7.5, and 10%w) were added, and mixed with spatula. Then CQ and 4EDMAB (0.5/0.5 in weight ratio) were added in a dark room. Finally the composite were placed in an ultrasonic bath to exit air bubbles and better mixing.

Mechanical properties evaluation

We define 3 groups of samples with 5, 7.5 and 10wt% fillers in each method denoted D₁, D₂ and D₃ (in dispersion in solvent method) and N₁, N₂ and N₃ (in non-dispersion in solvent method) respectively. 3 bar shapes specimens (25×2×2 mm³) were prepared in a stainless steel mold to evaluate flexural strength for all composites. All the samples were light-cured between two glass slides and stored in distilled

water for 24 hours at 37°C. Then flexural strength (FS) and flexural elastic modulus (E_f) of specimens were obtained by three point bending using universal testing machine with a span of 20 mm, at 60 N load cell and a cross-head speed of 0.5 mm/min. calculations were made using formulas as follows(13):

$$(1) FS = 3PL/2WT^2$$

$$(2) E_f = (p/d) (L^3/4WT^3)$$

Where P is the load at fracture, L is the distance between two supports, W is the width of the specimen, T is the thickness of the specimen and d is the deflection at load P. The fractured samples of flexural test were used for microhardness test. The microhardness was evaluated by applying 50 g load for 10 s. VHN was measured at 8 indentation points per cross-section and averaged.

Results and Discussion

The size and distribution of glass particles showed in figure 1. The SEM picture of fabricated particles showed in figure 2.

The particle size distribution figure 3 and SEM photos of these silanized particles were achieved figure 4.

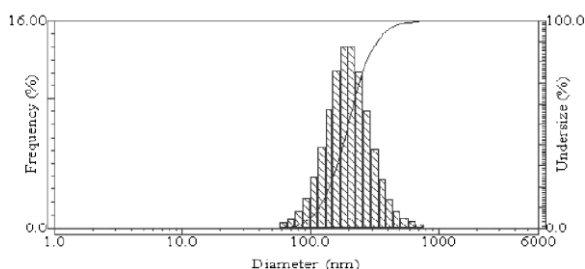


Figure1. The size distribution of milled glass particles.

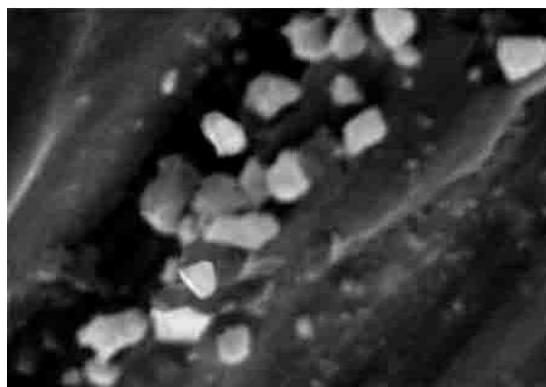


Figure2. SEM photo of milled glass particles.

Dental nanocomposite with glass nanoparticles

Table 2. The mean value of flexural strength, flexural modulus and microhardness among the groups.

composites	D1	D2	D3	N1	N2	N3
Flexural strength(Mpa)	63.98	69.07	75.22	55.83	58.38	59.99
Flexural modulus(Mpa)	1259.53	1295.08	1388.83	1210.78	1280.49	1334.26
Microhardness(VHN)	20.73	21.35	24.56	17.22	19.47	23.23

PSA shows the D50 of glass particles before silanization was 197 nm and the range of particles was 60-600 nm, but D50 decreases to 140 nm after silanization and the range of particle size was 60-500 nm. The SEM photo also shows that agglomeration was removed by silanization. Table 2 shows the flexural strength, flexural modulus and microhardness of all composites in both D and N groups. There is a significant difference between these mechanical properties of composites in two groups. Comparing flexural strength of both groups shows composites in group D have higher mechanical properties than group N. Also results show more filler fraction has positive effect on all mechanical properties, and flexural strength arises with filler content increase.

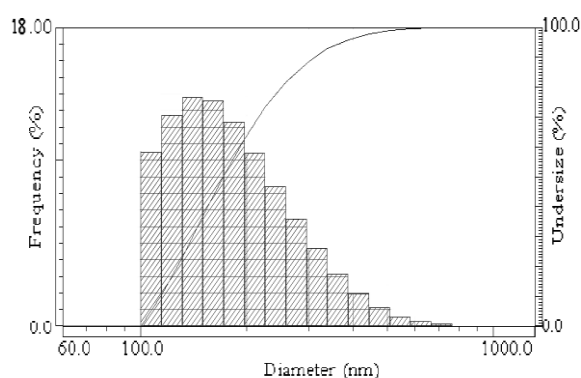


Figure 3. The size distribution of silanized glass particles.

The flexural modulus in groups D and N, arises by increasing filler content, emphasize the great effect of higher filler fraction on flexural modulus. Group D shows higher modulus than group N, again show the good effect of dispersion in solvent method to fabricate composites.

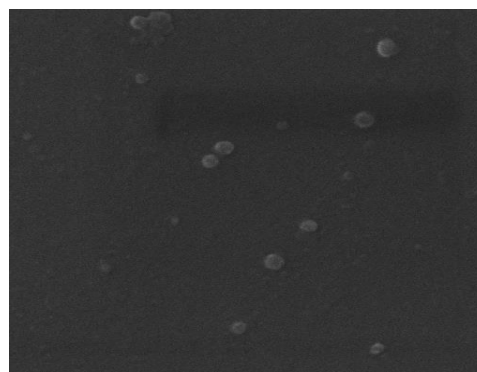


Figure 4. SEM photos of silanize glass particles.

According to our results, it appears that higher hardness values were achieved with more amounts of filler in both groups. The hardness of composites in group D is higher than group N. Flexural strength is a very important property for dental restorations, can reflect the ability that the materials to withstand complex stress (14). That is a criterion of durability and longevity of composites (13). A lot of researches (15–17) indicated that the dispersion of nanoparticles was the key factor for improving the mechanical properties of the matrix. Better mechanical properties of group D show, the dispersion in solvent method is an effective way to make composites with ultrafine particles as filler. The high flexural strength and flexural modulus in this study can be explained as the result of high strength of glass nanoparticles and good interlocking between fillers and resin because of appropriate silanization of fillers. The Hardness is the material resistance to local plastic deformation. There are some factors that influence microhardness, including the size of the filler particles, the weight fraction of the fillers (18).

The good hardness of prepared composites is a result of high hardness of glass nanoparticles. Higher hardness of composite in group D is the results of fabrication method by dispersion, that distribute ultrafine particles more effectively and the particles can act as a good reinforced parameter in smaller size than agglomerate.

Conclusion

Glass nanoparticles were produced by wet grinding in a planetary mill. Dispersion in solvent and non-dispersion in solvent methods were used to fabricate composite. The results showed, by increasing filler content flexural strength, flexural modulus and microhardness arise in both groups. The mechanical properties of composites in group D are significantly higher than group N. It can be concluded that dispersion in solvent method is the best way to fabricate nanocomposites and glass nanoparticles is a significant filler to improve mechanical properties of dental nanocomposite.

Acknowledgments

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References

1. Bowen RL. Properties of a silica-reinforced polymer for dental restorations. *J Am Dent Assoc.* 1963; 66: 57-64.
2. Du M, Zheng Y. Modification of silica nanoparticles and their application in UDMA dental polymeric composites. *Polym Composite.* 2007; 28: 198-207
3. Garoushi S, Vallittu PK, Watts DC, Lassila LVJ. Effect of nanofiller fractions and temperature on polymerization shrinkage on glass fiber reinforced filling material. *Dent Mater.* 2008; 24: 606-610.
4. Garoushi S, Vallittu PK, Lassila LVJ. Short glass fiber reinforced restorative composite resin with semi-inter penetrating polymer network matrix. *Dent Mater.* 2007; 23: 1356-1362.
5. Vitala A, Zürchera S, Dittmann R, Trottmann M, Lienemann P, Bommera B, Graulea T, Apel E and Höland W. Ultrafine comminution of dental glass in a stirred media mill. *Chem Eng Sci.* 2008; 63: 484-494.
6. Tooley FV. *The Handbook of Glass Manufacture.* New York: Ashlee Publishing; 1984.
7. Mende S, Stenger F, Peukert W, Schwedes J. Mechanical production and stabilization of submicron particles in stirred media mills. *Powder Technol.* 2003; 132: 64-73.
8. Verwey EJ, Overbeek JTG. *Theory of Stability of Lyophobic Colloids.* New York: Dover Publications; 2000.
9. Wang H, Zhu M, Li Y, Zhan Q. Mechanical properties of dental resin composites by co-filling diatomite and nanosized silica particles. *Mater Sci Eng.* 2011; C 31: 600-605.
10. Widegren J, Bergstrom L. Electrostatic Stabilization of Ultrafine Titania in Ethanol. *J Am Ceram Soc.* 2002; 85: 523-528.
11. Adler JJ, Singh PK, Patist A, Rabinovich YL, Shah DO, Moudgil BM. Correlation of particulate dispersion stability with the strength of self-assembled surfactant films. *Langmuir.* 2000; 18: 7255-7262.
12. Moszner N, Salz U. New developments of polymeric dental composites. *Prog Polym Sci.* 2001; 26: 535-576.
13. Zandinejad AA, Atai M, Pahlevan A. The effect of ceramic and porous fillers on the mechanical properties of experimental dental composites. *Dent Mater.* 2006; 22: 382.