DIAMETRAL TENSILE AND COMPRESSIVE STRENGTH OF RESIN BASED COMPOSITE CONTAINING BIOACTIVE GLASSES AND SILVER NANOPARTICLES

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ABSTRACT

Study was carried out to determine the diametral tensile strength (DTS) and compressive strength (CS) of resin based composites (RBC) impregnated with bioactive glass (BAG, 45S5) and silver nanoparticles (AgNPs).

RBC specimens in the commercially available RBC were prepared to comprise the control group (G1). The experimental composite containing 70 wt % filler was prepared to comprise the experimental control group (G2). The experimental RBC (G2) was modified by incorporating various quantities of BAG (5, 10 & 15 wt% respectively) and a fixed amount of AgNPs (0,009%) to make the specimens of the experimental groups comprising of G3, G4 and G5 respectively (n=6). DTS and CS of the specimens were determined by using universal testing machine. SEM analysis and dynamic light scattering of the specimens were done to see the morphology of the synthesized silica particles. Data were analyzed using one-way ANOVA and post hoc Tukey test. $p<0.05$ was taken as significant.

Mean values of DTS for RBC specimen of G1, G2, G3, G4 and G5 were 45.71±11.52, 26.83±4.58, 24.40±3.30, 21.22±3.60 and 22.36±3.47 respectively. DTS of commercial RBC was significantly higher than rest of the groups. DTS showed decrease with the addition of BAG and AgNPs when compared to experimental RBC (G2) ($p>0.05$) but the results were statistically insignificant. Mean values of CS for RBC specimen of G1, G2, G3, G4 and G5 were 361.49±20.06, 215.19±48.22, 190.59±40.63, 216.94±17.81 and 180.10±27.86 respectively. CS decreased with the addition of BAG and AgNPs in G3 and G5 as compared to G4 ($p>0.05$). The shape of the silica particles was found to be round with the size ranging up to 0.9 -1 μm. Uv-vis spectroscopy showed round shape of the AgNPs with size ranging up to 20nm.

Within the limitations of this study, the DTS and CS tested for the various RBC groups are significantly less than the commercial RBC while showed decrease in experimental RBC after substitution by AgNPs and BAG therefore, experimental RBC might be beneficial only in low stress bearing areas such as class III, class V cavities.

Keywords: *Bioactive glass, Re-mineralizing resin based composite, Silver nanoparticles, Diametral tensile strength, Compressive strength.*

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INTRODUCTION

Resin based composites (RBCs) are popular restorative materials due to pleasing appearance and direct restorative capabilities, however, resin based composites attract more plaque and microorganisms with acid formation resulting in the initiation of secondary caries at the margins. A study showed that percentage of streptococcus mutans of total colony forming unit in plaque was higher on composite resin than glass ionomer and amalgam. Due to polymerization shrinkage in RBCs, marginal gap develops at tooth restoration interface that allows entry of microorganisms and fluids resulting in the formation of secondary caries which is cited as the major reason for failure of composite restorations.[1]

Both releasing (such as chlorhexidine) and non-releasing agents (such as quaternary ammonium

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methacrylate) have been added to resin to impart antimicrobial property.[1] These agents show excellent antibacterial properties, however, other properties such as mechanical behavior and optical properties could be affected.[2] Due to broad spectrum antimicrobial activity of silver ions, several studies have been conducted by incorporating fillers such as silver doped silica and silica glass into resin based composite.[3] Silver nanoparticles shows strong antibacterial capability when introduced into resin composite without significantly affecting mechanical properties due to larger surface area to mass ratio.[4] Cheng et al reported that the silver nanoparticles added to resin composites in amount as low as 0.03% showed adequate antimicrobial activity.[5] The problem associated with introduction of nano scale fillers into resin is the agglomeration of particles. Some studies have reported excellent dispersion of AgNPs in resin formed in situ while using silver 2 ethyl hexanoate as a precursor.[4]

In early 1970s, Hench and his coworkers introduced bioactive glass that possess the ability to form chemical bond with both hard and soft tissues. Bioactive glass contains sodium, calcium, phosphorous and silicon in a proportion that encourages bioactivity. Due to its biocompatible nature and remineralising effect, the interest has developed among researchers to utilize it for dental applications.[6] Khovestenko et al reported that after the addition of custom made bioactive glass into resin based composites, the mechanical properties tested were suitable as those of commercially available resin based composites.[7] The addition of bioactive fillers such as CaP particles to the resin results in low strength of the resin composite. Significant reduction in strength is attributed to the lack of durable chemical bond between the resin phase and bioactive particles.[8]

Suppression of biofilm acids and promotion of remineralisation are the two main approaches that could be used to inhibit caries process therefore it would be advantageous to combine the benefits of both the strategies.[1] Langhorst et al[2] incorporated amorphous calcium phosphate into composite resin, however, such materials were found to be weak and could not be employed as restorative materials in stress bearing areas.[3] Xu et al synthesized composite resin containing amorphous calcium phosphate nanoparticles. They reported that the mechanical properties of the synthesized resin composite were superior than the commercially available composite resin.[3] A major short coming of calcium phosphate containing resin composite is that Ca and P ions release is for a limited period and declines with the passage of time.[4]

Due to interference in ion release ability, bioactive glasses are not silanized prior to incorporation in resin. These bioactive glasses are not well bonded to the resin matrix which might lead to inferior mechanical properties of the resulting resin composite. The aim of this study is to determine the effect of replacement of silica by bioactive glasses and silver nanoparticles on the compressive and diametral tensile strength of resin based composite.

METHODOLOGY

Tetraethyl orthosilicate (98%), cyclohexane (99.5%), 3-methacryloxypropyl trimethoxysilane (99%), n-propylamine (99%), and bisphenol A glycerolate dimethacrylate (BisGMA), tri (ethylene glycol) dimethacrylate (TEGDMA, 95%), and camphorquinone(97%) were purchased from Sigma–Aldrich USA. Ethyl 4-dimethylamino benzoate (99%) was purchased from Alfa Aesar USA. Ammonium hydroxide (28–30 wt %) (EMD Chemical Inc. Darmstadt, Germany). Silver 2 ethyl hexonoate (Strem Chemicals USA). Bioactive glasses 45S5 were provided by Denfotex Research Ltd UK.

Resin for the experimental composite was prepared by mixing 1:1 mass ratio of Bis-GMA and TEGDMA monomers with 0.4 wt% of camphoroquinone, 0.8 wt% of 4- dimethylaminobenzoic acid ethyl ether. In order to prepare 0.03% mass fraction of silver salt in resin, 10% solution of silver 2 ethyl hexanoate was prepared in 2-tert butyl amino ethyl methacrylate (TBAEMA). Then 1 % of this solution was added to Bis GMA, TEGDMA resin. Mass fraction of 0.03 % of silver in resin was used following a previous study5. Final concentration of AgNPs in resin composite was 0.009%. Silica particles used in the study were synthesized using Stober method9.

In order to improve the interfacial bond between resin matrix and inorganic particles, the surface of the particles was treated with ɤ-MPS using a previous procedure10. The silane (0.50±0.01g), the silica (5.0±0.05g), n-propylamine (0.1±0.01g) and solvent (100ml cyclohexane) were mixed for 30 min at room temperature and then for additional 30 min at 60±5◦C. Rotary evaporator at 60±◦C was used for removal of the byproducts and solvent. The powder obtained was then heated for 1 h at 90±5◦C in rotary evaporator and then dried at 80◦C in a dry heat oven for about 20 hours.

Control experimental dental composite (G2) was prepared by mixing the resin mixture with 70 wt% filler comprising of silica (60% synthesized silica and 9% commercial nano silica, size 50nm (Aerosol OX50) zirconia particles 1% (size 1-5 μm). First both the resin mixture and filler particles were mixed manually in a plastic container. Then the material was transferred to three roll mill (Exakt, TRM, Norderstedt, Germany) to obtain a homogenous material. Control experimental composite resin was modified by the substitution of silica by 0.009% AgNPs and 5, 10 and 15 wt% of 45S5

(BAG) (Table 1).

Compressive and diametral tensile strength

Specimens for determination of compressive11 and diametral tensile strength were made using mould made from silicone rubber. The mould was cylindrical in shape having dimensions of 4mm diameter and 6mm height for conducting both the tests. In order to prevent interference in polymerization by inhibiting effect of oxygen, the specimens were covered on top and bottom with a cellulose acetate strip. A clear rigid microscope glass slide having thickness about 1 mm was placed on top and bottom of the matrix strip. Both top and bottom of the specimen surface was cured by overlapping radiations for 20 seconds by using portable LED light cure unit (Dmetec co., Ltd.Korea). The intensity of curing light was 750 mW/cm2. The intensity of light was periodically noted with the help of digital radiometer (Optilux Radiometer US). The diameter of the light guide tip was 10 mm. The light guide of the light curing unit was placed at right angle to the specimen's surface. Specimens were placed in distilled water at 37C for 24 h before testing. Surplus material was removed by using 600 grit Silicon Carbide paper. Compressive strength was determined using a universal testing machine (Instron 5565.USA). The compressive strength (CS) was calculated by dividing the applied force (F) by the cross-sectional area.

$CS = F/\varpi R2,$

Where R is the radius of the cross-section of the specimen

F is the applied load.

Diametral tensile strength (DTS) was determined under compressive stress in UTM (Instron5565.USA). Force was applied on the side of the cylinder at a cross head speed of 1mm/minute. After each test the force in Newtons (N) was recorded and the (DTS) was calculated using the equation.

$\sigma = 2F/\varpi dh$

d: diameter (4 mm);

h: height (6 mm) of specimens;

ϖ: 3.1416.

The size of silica particles and BAG was determined by dynamic light scattering (DLS) (Malvern Zetasizer UK). Silver nanoparticles were characterized with the help of uv-vis spectroscopy (Cary uv-vis Agilent technologies, USA) in the wavelength range 300-850nm.

Statistical analyses of the data were conducted using analysis of variance (ANOVA) and Tukey's post hoc method to calculate the differences between groups of materials ($P < 0.05$) by statistical package for social sciences SPSS software version 19 for windows.

RESULTS

Table 2 and Fig:1(A) show the results of diametral tensile strength (DTS) of various resin based composite groups. One-way ANOVA indicated statistically significant difference between various resin composite groups ($p< 0.001$). Tukey HSD post hoc test showed that the diametral tensile strength of resin composite groups (G2, G3, G4, and G5) was significantly less than commercial resin composite G1 ($p < 0.001$). The addition of BAG and AgNPs reduced the DTS ($p>0.05$) of G3, G4 and G5 when compared with G2.

Results for compressive strength are presented in Table 2 and Fig 1 B. Post hoc Tukey test showed highly significant difference between G1 and rest of the groups ($p<0.001$). The replacement of silica by bioactive glass and AgNPs reduced the compressive strength for G3 and G5 while G4 showed increase in compressive strength when compared to G2 but the results are statistically not significant ($p>0.05$).

Scanning electron microscopy (SEM) analysis reveal round shape of the particles. The sizes of the particles ranged from 0.9 to 1µm. Furthermore, the sizes of the particles evaluated by dynamic light scattering (DLS) also ranged from 0.9 to 1µm. Similarly, the average size of BAG particles determined by DLS was found to be about 512nm.

Uv-vis spectroscopy showed the surface plasmon resonance absorption peak centered at about 428nm wavelength which indicated round shape of the particles. Small spherical silver nanoparticles (less than 20nm) exhibit single surface plasmon resonance peak. In addition, silver salt solution dissolved in resin mixture displayed colour change from transparent to light brown when exposed to visible light from dental light cure unit which indicated the formation of silver nanoparticles.

Table 2. Mean and SD values for compressive and diametral tensile strength. Values in parenthesis ( ) indicate SD. Different case letters indicate statistically significant difference

DISCUSSION

Several studies have been conducted to develop new antibacterial and bioactive restorative material for the prevention of recurrent decay, but most of them show compromised physical properties.[12] Diametral tensile strength provides information about the performance of brittle material such as resin based composites subjected to tensile stresses. It is therefore clinically a relevant aspect since resin based composites could fail under stresses due to tensile forces during chewing.[13,14]

In the present study diametral tensile strength (DTS) values of RBC specimens in the various groups

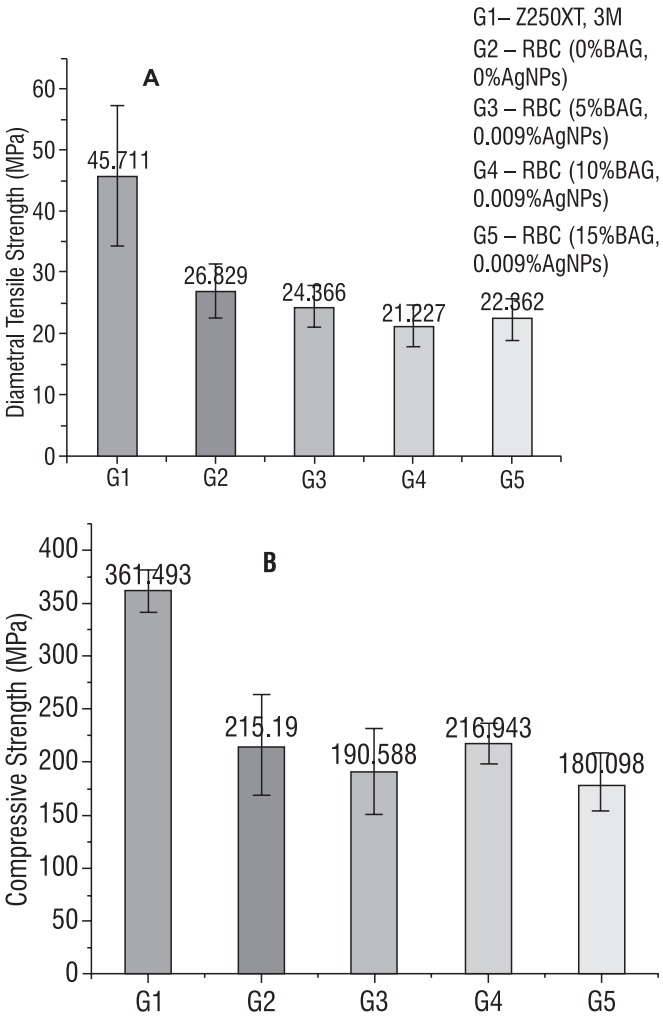

Fig: 1: Bar chart showing mean values for diametral tensile strength (A) and compressive strength (B) for the various resin composite groups

TABLE 1: COMPOSITE RESIN GROUPS AND THEIR CONTENTS

Group 1 (G1)	Commercial resin composite (3M Filtek Z250XT)
Group 2 (G2)	Experimental resin composite (0%BAG, 0%AgNPs)
Group 3 (G3)	Experimental resin composite (5%BAG,0.009%AgNPs)
Group 4 (G4)	Experimental resin composite (10%BAG,0.009%AgNPs)
Group 5 (G5)	Experimental resin composite (15%BAG, 0.009%AgNPs)

(G2—G5) ranges between 21-26 MPa which is significantly lower than the DTS value (40-50 MPa) reported in the literature for a typical hybrid RBC.[15] The DTS of experimental RBCs (G2-G5) was significantly lower than G1. The dominant factor responsible for lower diametral tensile strength in experimental RBC groups

TABLE 2. MEAN AND SD VALUES FOR COMPRESSIVE AND DIAMETRAL TENSILE STRENGTH. VALUES IN PARENTHESIS ( ) INDICATE SD. DIFFERENT CASE LETTERS INDICATE STATISTICALLY SIGNIFICANT DIFFERENCE

Resin Based Composite Groups	Compressive Strength	Diametral Tensile Strength
G1	361.493(20.061) a	45.71(11.524) a
G2	215.19(48.224) b	26.829(4.585) b
G3	190.588(40.635) b	24.366(3.309) b
G4	216.943(17.809) b	21.227(3.607) b
G5	180.098(27.860) b	22.362(3.466) b

seems to be the lower filler content when compared to commercially available RBCs.[7] Figure 1A shows that there was slight decrease in tensile strength values with replacement of silica by BAG and AgNPs but the results are statistically not significant (Table 2). Possible explanation might be that BAG particles incorporated in RBC have been in quantity and size that cannot considerably interfere with the RBC system and can maintain the mechanical properties.[12] This study has not investigated the influence of incorporation of BAG more than 15% on the mechanical properties of RBC therefore it cannot be predicted that what is the limit of addition of BAG in RBC after which the mechanical properties of RBC are deteriorated. Korkut et al[6] reported non-significant results when BAG 5 % and BAG 10% added to RBC, however decrease in strength was noticed when the percentage of BAG was raised up to 30% filler mass fraction. The results of this study are also in accordance with another study which also mentioned non-significant results when the BAG was added up to 15%.[7] The underlying reason for this behavior might be that at low concentration the bioactive glasses and silver nanoparticles are well distributed but at higher concentration these particles tend to aggregate and lead to defects and flaws which ultimately deteriorates its mechanical properties[6] Although BAG particles were not silanized prior to addition in RBC, the results of this study revealed that strength of the RBC was not affected significantly by the replacement of silica by unsilanized BAG up to a certain limit.

Compressive strength is a useful property for assessment of brittle materials such as amalgam, cements or composite resins.[16] Compressive stress has a predominantly essential role in the chewing process[17]

and is an important indicator of fracture resistance of brittle material.[18] The key limitation of the compression test is that most of the researchers have not used the same protocol for determination of compressive strength.[19] Therefore it was difficult to compare the values obtained in this study (Table 2)

In this study, the results obtained for compressive strength for groups (G2, G3, G4, and G5) are significantly lower (Table 2, Fig 1A) than the commercially available RBC (G1) but approach the values (150-250 MPa) reported for some conventional RBCs containing hybrid fillers and bulk filled RBCs.[20,21] Similar to other mechanical properties such as flexural and tensile strength, high compressive strength of RBC specimens in the G1 can be attributed to the high filler amount and reactions between resin components after the light curing process.[22] The first obvious difference between the microstructures is that the experimental RBCs (G2-5) had significantly lower filler particles concentration than G1 (70 wt% versus 82wt%, respectively). The higher strength values for the G1 are thus in agreement with opinions in the literature for resin based restorative composites that higher strength is due to higher filler content and that filler amount is the leading element regardless of filler having different size and shape.[7] Another factor, which might have contributed to low mechanical properties of experimental RBCs in comparison to commercially available RBC (G1), is the presence of unsilanised nanosilica (9%, size 50nm), which was added to facilitate manipulation and adjust viscosity.

The results for compressive strength of experimental RBC groups (G2-G5) ranges from 180-216MPa are better than another similar study (118-175MPa) conducted by Korkut et al6 which incorporated 5%,10% and 30% BAG in RBC with a total resin/filler ratio of 30:70%. G2 and G4 has almost the same compressive strength while G3 and G5 had slightly lower strength than G2 and G4 but the results are statistically not significant (Table 2). This implies that the replacement of silica by BAG (5-15%) and 0.009%AgNPs has minimal negative impact on the compressive strength of RBC. The other reason for this behavior might be that at low concentration the bioactive glasses and silver nanoparticles are well distributed but at higher concentration these particles tend to aggregate and lead to defects which ultimately deteriorates its mechanical characteristics. In another study, the flexural strength and elastic modulus was not decreased after the addition of 0.042% AgNPs to RBC 4. This also supports that at such a low concentration (0.009%AgNPs) in RBC does not negatively influence the mechanical properties of RBC.

CONCLUSION

The Diametral tensile and compressive strengths tested for the various experimental resin based composite groups are significantly less than the commercial resin composite therefore experimental resin composite might be beneficial only in clinical circumstances such as class III, class V cavities that are low stress bearing areas and also restoration of deciduous teeth, however, for application in anterior teeth, shade and colour of the experimental resin composite need to be optimised.

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1 Amjad Hanif:	Conceived the research idea, designed, conducted the research, analysed the data and wrote the manuscript.