Optimization of compressive strength of zirconia based dental composites

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Abstract. Dental composites are tooth-coloured restorative material used by dentists for various applications. Restoration of a lost tooth structure requires a material having mechanical as well as aesthetic properties similar to that of tooth. This poses challenges to engineers and the dentist alike. Dental composites consist of a matrix and a dispersed phase called filler, which are mainly responsible for its mechanical properties. Most commonly used matrix is bisphenol glycidyl methacrylate (Bis-GMA) and triethylene glycol dimethacrylate (TEGMA). Silica and glass are conventional fillers used in the past. Recently, zirconia is being used due to its improved mechanical properties. A study was conducted to evaluate the contribution of zirconia to the mechanical properties in general and compressive strength in particular. We have attempted to make an experimental dental composite with a conglomerate of nanofillers, namely, zirconia, glass and silica, and optimize this filler volume percentage and obtain an optimum compressive strength for the experimental dental composite.

Keywords. Optimization; regression; zirconia; nanoclusters; compressive strength; filler.

1. Introduction

Dental composites have been used in dentistry for the past 30 years for tooth restoration, tooth reinforcement and tooth recontouring. Dentists choose a dental composite according to its mechanical and aesthetic needs. The mechanical properties of the composite depend on the filler volume concentration (Manhart *et al* 2000; Ruddell *et al* 2002; Atai *et al* 2004). In the past, many studies have been done on size, shape and types of filler particles to be incorporated into composite resin matrix. Fillers like glass, silica and quartz have been used in sizes varying from 20 to 150 μm^4 to micro fillers of 0.1 μm^4 or less. Recently, nanohybrid fillers have been introduced. This resulted in improved loading, as the same volume of fillers in the matrix accommodated more nanofiller particles and the particles were more in surface contact with the matrix (Mitra *et al* 2003; Soh *et al* 2006). Consequently, there was improvement in the mechanical properties and aesthetic properties without compromising its workability. Contemporary dental composite use silica, glass and quartz either individually or in combination. We have used fillers like silica, glass and zirconia as conglomerates.

This study was conducted to analyse the effect of a specific filler zirconia nanocluster along with glass and silica on the mechanical properties of composite in general

and compressive strength in particular. Mastication is a dynamic process in which compressive forces are the most important forces acting in all the directions. In the past decade, most studies on dental nanocomposites have shown improvement in the mechanical properties, but most of them are focused on fracture toughness. Fewer studies show the influence of particular nanofillers on the compressive strength of dental composites (Eliseu *et al* 2012).

2. Materials and methods

The monomer system can be viewed as the backbone of the composite resin system. Bis-GMA (bisphenol glycidyl methacrylate) and TEGDMA (triethylene glycol dimethacrylate) continue to be the most used monomers for manufacturing present-day composites (Bowen 1964). Recent studies reveal introduction of zirconia as filler for improvement in mechanical properties.

The fillers used were zirconia, barium aluminium fluoride glass and silica in the volumetric percentages as shown in table 1. Nine experimental composites were

Table 1. Percentage of filler volume at each level.

Process parameters	Level 1	Level 2	Level 3
Glass%	23.7	25.7	31
Zirconia%	21	27.5	29
Silica%	13.5	16	20

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Table 2. Taguchi L9 orthogonal array for compressive strength.

Experimental composites	Filler volume levels			Compressive strength (MPa)	Signal-to-noise ratio (S/N ratio)
	Glass	Zirconia	Silica		
1	1	1	1	249.33	47.94
2	1	2	2	579.83	55.27
3	1	3	3	630.08	55.99
4	2	1	2	231.93	47.31
5	2	2	3	657.14	56.35
6	2	3	1	316.97	50.020
7	3	1	3	230	47.23
8	3	2	1	419.41	52.41
9	3	3	2	589.49	55.41

prepared using the Taguchi L9 orthogonal array for deciding the filler volume percentage as shown in table 2. The matrix of experimental photocure composite resins was composed of 2,2-bis[4-(2-hydroxy-3-methacryloxypropoxy) phenyl] propane (*bis*-GMA) and triethylene glycol dimethacrylate (TEGDMA) in 2:1 ratio by weight. One percent of 2-(dimethylamino) ethylmethacrylate and 200 ppm of 4-methoxyphenol are used as a catalyst and an inhibitor, respectively, and added to the mixture of monomers.

The fillers used are hybrids with two sets to improve the loading of the resin. The average size of set 1 is 10–100 nm and that of set 2 is 2–15 micrometers. Hybrid composites contain a broad range of particle sizes. A wide range of particle sizes can lead to high filler loading with resultant high strength. While they may contain a small fraction of filler particles in the nanometer particle size range, they also contain a range of substantially larger filler particles which influence the optical properties of these composites and detract from polish retention. The dispersion of the fillers in the matrix is uniform, and this is improved when the composite is cured with UV radiation at the time of cavity filling. Fully stabilized zirconia is used with dopant levels of 8 mol% yttria, 16 mol% magnesia or 16 mol% calcia. It has a cubic solid solution and has no phase transformation from room temperature up to 2500 °C.

Silica and glass fillers are used in a wide range of dental applications including tooth fillings and tooth build-ups because of its biocompatibility, wear resistance and aesthetics. Important properties of zirconia are exceptional mechanical properties, excellent durability and stability and high biocompatibility, and zirconia is very resistant to corrosion and scratches. Zirconium oxide is ivory in colour and matches perfectly with natural tooth, and also its light reflection is better. It is tough, durable and has superior strength and aesthetics.

2.1 Preparation of experimental composite

A magnetic stirrer was used to agitate the mixture for 3 days, resulting in a homogeneously mixed viscous liquid.

One gram of the monomer mixture and 0.01 g of campherquinone were mixed mechanically for 1–2 min in an automatic mortar. The fillers were gradually added to the monomer mixture and stirred for 15 min. After mixing, the mortar was removed from an automatic apparatus, and then the mortar with the experimental composite resin was evacuated for 1 min in a vacuum desiccator. Silane treatment was given to the fillers.

2.2 Preparation of test specimen for compressive strength

A cylindrical test specimen was made using a glass tube mould of 4 mm inner diameter (6 mm outer diameter) and 8 mm length. Polyester film of 0.1 mm thickness was used to position each mould. The experimental dental composite was slightly overfilled in the mould and all the air bubbles were excluded. The mould was slightly overfilled with the test composite and all air bubbles were excluded. A second piece of polyester film was placed onto the material in the mould and covered with a glass plate, and then pressure was applied gently, thus, exuding excess material from the mould. The specimen was irradiated for 15 min. The light-cured specimen was removed from its mould and stored in distilled water at 37 °C for 24 h prior to testing. The cylindrical specimen was tested in a universal testing machine (Make-Autograph) and used a crosshead speed of 1 mm/min. Three specimens of each experimental group were tested. The mean value of the three specimens was accepted as an observed compressive strength of the tested composite resin.

The ASTM standard (2008) adopted was D695-08. The dimensions of specimens were checked using a digital calliper (digimatic caliper, Mitutoyo Corp., Tokyo, Japan). Test specimen was placed between the surfaces of the compression tool, taking care to align the centre line of its long axis with the centre line of the plunger and to ensure that the ends of the specimen are parallel to the surface of the compression tool. The loads and corresponding compressive strain at appropriate intervals of strain were recorded. After the yield point was reached,

the speed was increased from 5 to 6 mm/min (0.20 to 0.25 in/min) and the machine allowed to run at this speed until the specimen broke.

2.3 Taguchi experiment: Design and analysis

Traditional experimental design procedures are too complicated and not easy to use. When the number of process parameters increases, a large number of experimental works have to be carried out. Dr Genichi Taguchi has designed a method which reduces the number of experiments by the use of an orthogonal array. The information from the behaviour of the process is obtained from a plan of experiments with the objective of acquiring data in a controlled way. This design of high-quality system called Taguchi's design method which reduces efforts of conducting a number of experiments, thus saving time and cost and enabling discovery significant factors quickly.

But the main advantage of Taguchi's method is by conducting fewer experiments, we get a reliable result and also the higher the value of sum of square of an independent variable, the more it has influence on the performance parameter. One can also calculate the ratio of individual sum of square of a particular independent variable to the total sum of squares of all the variables. This ratio gives the percent contribution of the independent variable on the performance parameter. In addition to above, one could also find the optimal solution to the problem.

A statistical analysis of variance (ANOVA) and signal-to-noise ratio (S/N ratio) can be employed to study the influence of different filler volume percentages on compressive strength values. Following are the steps followed in Taguchi's optimization study (Taguchi 1990; Ross 1996). (1) Select noise and control factors. (2) Select Taguchi orthogonal array. (3) Conduct experiments and measure the compressive strength for each of the experimental composites. (4) Analyse results (signal-to-noise ratio). (5) Predict optimum performance. (6) Confirmation experiment.

2.4 Experimental

2.4a Plan of experiments: Here Taguchi method is used, which includes the experiment design theory and the quality loss function concept. The degrees of freedom for three parameters in each of three levels were calculated as follows: degree of freedom (DOF) = number of levels - 1. For each factor, DOF is as follows: for glass, DOF = 3 - 1 = 2; for zirconia, DOF = 3 - 1 = 2; for silica, DOF = 3 - 1 = 2. Taguchi L9 orthogonal array was used, which has three columns at three levels and nine rows corresponding to the number of tests (Ross 1996).

Nine experiments were conducted at different parameters in this study. L9 orthogonal array has eight DOFs, in

which six were assigned to three factors (each one 2 DOFs) and two DOFs were assigned to the error. Table 4 shows the degree of influence of the process parameters, i.e. the filler content (volumetric percentage), three factors, each at three levels, are taken into account (Ross 1996). The compressive strength was tested for each experimental composite with filler volume percentage corresponding to each level. The compressive strength values corresponding to each experimental composite are shown in table 2.

3. Results and discussion

3.1 Regression analysis

A mathematical model was developed with the volumetric percentages of three fillers in the composite, namely, glass (barium aluminium fluoride), zirconia and silica and its effect on the compressive strength. The correlation between factors (glass (barium aluminium fluoride), zirconia, silica) and compressive strength was obtained by multiple linear regressions. To derive the models, the standard commercial statistical software package MINITAB was used

$$CS = 6278 - 883G + 275Z + 269S \\ + 13.5GZ - 8.01ZS + 9.28G^2 - 9.39Z^2.$$

3.1a Analysis of the signal-to-noise ratio: For the minimization of quality characteristic variation due to uncontrollable parameters the response variation is studied using signal-to-noise ratio (S/N ratio) as enumerated in Taguchi's method. In the dental composites, larger compressive strength will be better, so the compressive strength was considered as the quality characteristic and the concept of 'the larger-the-better' (Taguchi 1990; Ross 1996) was used. The S/N ratio used for this type response is taken as 'larger-the-better' and given by the relation

$$S/N = -\log_{10} \left(\frac{1}{n} \sum \frac{1}{y^2} \right), \quad (1)$$

where y is the measured value in a run/row and n the number of measurements in a trial/row ($n = 1$ in this case). Equation (1) was used to calculate the S/N ratio. The S/N ratios and the corresponding values of compressive strength are shown in table 2.

For the volumetric percentages of glass, zirconia and silica concentrations, the compressive strength response table and the ranks are shown in table 3. A greater S/N ratio value will give better performance, irrespective of the performance characteristic. The greater S/N ratio value will give the optimum level filler volume percentage.

The optimal compressive strength was obtained at glass% = 23.7 (level 1), zirconia% = 27.5 (level 2), silica% = 20 (level 3), based on the analysis of the S/N ratio.

Table 3. S/N ratio values for compressive strength by factor level.

Input parameter	Level 1	Level 2	Level 3	Delta (max-min)	Rank
Glass%	53.06	51.22	51.69	1.83	3
Zirconia%	47.49	54.69	53.80	7.19	1
Silica%	50.13	52.66	53.19	3.05	2

Figure 1. The effect of the process parameters on the compressive strength.

The effect of the process parameters on the compressive strength is shown in figure 1, which is evaluated using statistical software package MINITAB.

3.2 Analysis of variance (ANOVA)

For finding out the differences in the average performance of the groups of items tested, a statistical tool named ANOVA, which is an objective decision-making tool is used. At a specific confidence level, the mean square is compared with an estimate of experimental error in ANOVA. The interaction and significance of all the main factors are tested. The following equation is used to find out the total sum of squared deviations SS_T from the total mean S/N ratio, n_m

$$SS_T = \sum_{i=1}^n (n_i - n_m)^2.$$

where n_i is the mean S/N ratio for the i th experiment and n the number of experiments in the orthogonal array. For calculation of P , i.e. percentage contribution

$$P = \frac{SS_d}{SS_T},$$

where SS_d is the sum of the squared deviations.

Table 4 shows the results of ANOVA. To find out the significance of a factor, a ratio of the mean square error to the residual error is taken and called as F -ratio, named after Fisher. Fisher test is used to study the significant effect on quality characteristic by the design parameters. To study the effect of the filler volume percentage on the

compressive strength, we use the percentage (%) shown in table 4. From table 4, it can be seen that nanofiller glass has 13.46%, zirconia has 66.83% and silica has 19.71%, which influences on the compressive strength, respectively.

3.3 Prediction of the optimum results

For the experimental dental composite we know that for the compressive strength, higher is the better (Taguchi 1990; Ross 1996). We have obtained the optimum values of filler volume and, from this, the optimum value of compressive strength is found by the following relation

$$\text{Maximum optimum compressive strength} = A_1 + B_2 + C_3 - 2T.$$

A_1 = Average value of compressive strength at the first level of glass volume percentage. B_2 = Average value of compressive strength at the second level of zirconia volume percentage. C_3 = Average value of compressive strength at the third level of silica volume percentage. T = Average values of compressive strength for all the nine trials. Maximum optimum compressive strength is calculated as 676.68 MPa as shown in table 5. Uncertainty, analysis has been done with a percent uncertainty of 0.00113137.

3.4 Confirmation test

Taguchi has strongly recommended a confirmation test for verifying the results (Taguchi 1990; Ross 1996). The

Table 4. Analysis of variance (ANOVA) results for compressive strength of dental composite.

Source of variation	Degree of freedom (DOF)	Sum of squares (S)	Mean sum of the squares (MSS)	F ratio	Percentage (%)
Glass%	2	12636.93	6318.46	← POOL	
Zirconia%	2	176526.04	88263.02	13.97	66.83
Silica%	2	52070.26	26035.13	4.12	19.71
Residual	2	22912.49	11456.24		13.46
Total	8	264145.72			100

Table 5. Calculation of optimum maximum compressive strength (MPa).

Averages	Maximum average values of compressive strength			Average compressive strength	$A_1 + B_2 + C_3$	2T	Maximum compressive strength (MPa)
A_1	249.32	579.83	630.08	486.41	1544.28	867.59	676.68
B_2	579.83	657.14	419.41	552.12			
C_3	630.08	657.14	230	505.74			

Table 6. Fillers used in the experimental dental composite.

Experimental dental composite no.	Filler volume percentages		
	Glass	Zirconia	Silica
1	23.7	21	13.5
2	23.7	27.5	16
3	23.7	29	20
4	25.7	21	16
5	25.7	27.5	20
6	25.7	29	13.5
7	31	21	20
8	31	27.5	13.5
9	31	29	16

experiments are conducted with the optimum conditions and the compressive strength is found. This compressive strength is compared with that calculated value using the relation enumerated above. We had obtained optimum filler volume as $A_1B_2C_3$ and the compressive strength found experimentally was 680 MPa.

4. Conclusions

(1) In our study, we have prepared a experimental dental composite. Contemporary dental composites use glass and silica individually or in combination as fillers. Zirconia was introduced as nanofiller. We have used three nanofillers, namely, zirconia, glass (Ba–Al–F) and silica in combination (conglomerate).

(2) It is observed that the influence of zirconia as a filler material on experimental dental composite is very significant and highest in our study, effect of zirconia on compressive strength being 66.83%. Statistical results (at a 95% confidence level) show that the glass, zirconia and silica

affect the compressive strength by 13.46%, 66.83% and 19.71% of the experimental dental composites, respectively.

(3) Statistically designed experiments based on Taguchi methods were performed using L9 orthogonal arrays to analyse the compressive strength as a response variable. Conceptual S/N ratio and ANOVA approaches for data analysis drew similar conclusions.

(4) The maximum optimum compressive strength is calculated as 676.68 MPa by Taguchi's optimization method.

(5) In this study, the analysis of the confirmation experiment for compressive strength has shown that Taguchi parameter design can successfully verify the optimum filler volume percentages ($A_1B_2C_3$), which are glass = 23.7% (A_1), zirconia = 27.5% (B_2) and silica = 20% (C_3).

(6) The confirmatory experimental tests results are matching with the optimal value of compressive strength calculated.

(7) The compressive properties of the dental composites mainly depend on the type and size of the filler properties. Many studies have been conducted to investigate the various mechanical properties. The challenge still continues to improve the mechanical properties of dental composite with a predicted clinical life.

It is observed that the influence of zirconia as a filler material on experimental dental composite is very significant and highest in our study. We have used nanoparticles of fillers in conglomerate; this has increased the loading and improved surface finish of the composite. We have optimized the volume fraction of filler content and the consequent compressive strength. This will be useful for the composite supporting compressive and flexural mastication forces more effectively than other restorative materials tested.

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