

Fracture Toughness, Compressive Strength and Load-bearing Capacity of Short Glass Fibre-reinforced Composite Resin

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Objective: To investigate the reinforcing effect of short E-glass fibre fillers on fracture related mechanical properties of dental composite resin with a semi-interpenetrating polymer network (IPN) polymer matrix.

Methods: Experimental short fibre composite (FC) resin was prepared by mixing 22.5 wt% of short E-glass fibres, 22.5 wt% of IPN-resin and 55 wt% of silane treated silica fillers using a high speed mixing machine. Test specimens were made bar shaped ($3 \times 6 \times 25 \text{ mm}^3$), cylindrical (6 mm length \times 3 mm diameter) and cubic ($9.5 \times 5.5 \times 3 \text{ mm}^3$) from the experimental FC resin and conventional particulate composite resin (Grandio) as control. The test specimens ($n = 8$) were either dry stored or water stored (37°C for 30 days) before the mechanical tests. A three-point loading test and compression test were carried out according to ISO 10477 and a static loading test was carried out using a steel ball ($\varnothing 3.0 \text{ mm}$) with a speed of 1.0 mm/min until fracture.

Results: Experimental fibre composite had a significantly higher mechanical performance for fracture toughness ($14 \text{ MNm}^{-1.5}$), compression strength (129 MPa) and static load-bearing capacity (1584 N) than the control composite ($2 \text{ MNm}^{-1.5}$, 112 MPa and 1031 N).

Conclusion: The resin with short E-glass fibre fillers and IPN-polymer matrix yielded improved mechanical performance compared to the conventional particulate composite resin.

Key words: compression strength, fibre-reinforced composite, fracture toughness, restorative composite

The increasing attractiveness of tooth-coloured restorations has promoted research in this particular area of dental materials during last few years. After many significant material improvements, restorative composite resin still suffers from a lack of mechanical properties and problems related to polymerisation shrinkage. Clinical studies have shown that direct composite fillings fail predominantly because of occlusal wear or secondary

caries^{1,2}. However, fracture of restorative composite is also reported as a common reason for replacement². Due to the failures of these kinds, it is still controversial whether restorative composites should be used in large high-stress bearing applications such as in direct posterior restorations. The relatively high brittleness and low fracture toughness of current composites still hinder their use in the large stress-bearing restorations³.

Many studies on the fracture strength of the composite resins have been carried out. Attempts have been made to change the type of fillers or filler size and their surface silanisation. By changing the polymerisation kinetics of resins, matrices and the degree of monomer conversion have been attempted to be influenced⁴. Reinforcing the resin with micro-glass fibres⁵, with fibre-reinforced composite (FRC) substructure^{6,7}, whiskers³, particulate ceramic fillers (dense and porous)⁸ and optimisation of filler content are among the methods that have been studied⁴. However, further significant improvements are

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still needed. In addition to flexural strength, the material properties that represent resistance to fracture are fracture toughness (K_{IC}) and compression strength. They are important material characterisation parameters required for the prediction of the mechanical performance of structural materials⁹.

Glass fibres have been investigated to reinforce dental polymers for over 30 years¹⁰. They have documented reinforcing efficiency and good aesthetic qualities, compared to carbon or aramid fibres¹¹. The effectiveness of fibre reinforcement is dependent on many variables, including the resins used, the quantity of fibres in the resin matrix^{12,13}, length of fibres¹², form of fibres¹⁴, orientation of fibres¹⁵, adhesion of fibres to the polymer matrix¹⁶ and impregnation of fibres with the resin¹⁷. Short random fibres provide an isotropic reinforcement effect in multi-directions instead of one or two directions, as described by Krenchel¹⁸.

Polymethyl methacrylate (PMMA) and dimethacrylate-based semi-interpenetrating polymer network (semi-IPN) matrix has been established as a polymer matrix in denture base materials¹⁹. Also some products of fibre-reinforced composite (FRC) use semi-IPN polymer in the matrix²⁰.

Although previous investigations^{21,22} on the use of experimental semi-IPN matrix in combination with short E-glass fibres in restorative filling composite show enhancement of flexural strength, the effect of short glass-fibre reinforcement on fracture toughness and compressive strength has not been reported.

Thus, the aim of this study was to investigate the reinforcing effect of short E-glass fibre fillers on fracture-related mechanical properties, such as fracture toughness and compression strength, of dental composite with a semi-IPN polymer matrix.

Materials and methods

Dimethacrylate (BisGMA 67% [bisphenol A-glycidyl dimethacrylate] and TEGDMA 33% [triethylenglycol dimethacrylate]) resin consisting of 50 wt% nanofillers (SiO_2 , 20 nm in size) (Hanse Chemie, Germany) and E-glass fibres with BisGMA-PMMA (polymethyl methacrylate, Mw 220.000) resin matrix (everStick, StickTech Ltd, Turku, Finland). In addition, radiopacity fillers of BaAlSiO_2 ($3 \pm 2 \mu\text{m}$ in size) (Specialty Glass, USA) were incorporated to the resin system. Before the BaAlSiO_2 filler particles were incorporated into the resin matrix, they were silane treated using a previously reported technique²³. Commercial particulate filler composite (Grandio Caps, VOCO, Germany) was used as a control material.

Experimental fibre composites (FC) were prepared by mixing 22.5 wt% of short E-glass fibres (3 mm in length) with 22.5 wt% of resin matrix, and then 55 wt% of BaAlSiO_2 radiopacity fillers were added gradually to the mixture. The mixing was carried out by using a high-speed mixing machine for 5 minutes (SpeedMixer, DAC, Germany, 3500 rpm). The dimethacrylate-based resin matrix consisting of PMMA forms a semi-IPN polymer matrix for the FC.

Rectangular bar (single-edge notched) specimens to measure the K_{IC} ($3 \times 6 \times 25 \text{ mm}^3$) (Fig 1a) were fabricated according to British standard 54479. Specimens were prepared using a metal brass mould, so that no force was required to remove the cured bars. A sharp central notch of specific length (a) was produced by inserting a straight-edged blade into an accurately fabricated slot at mid-height in the mould; the slot extended down half the height to give $a/W = 0.5$. The crack plane was perpendicular to the specimen length.

K_{IC} was calculated from the following formula²⁴:

$$K_{IC} = [3PL / BW^{3/2}] Y$$

Where P = peak load at fracture; L = length; B = width; W = height; and Y = calibration functions for a given geometry, $(1.93[a/W]^{1/2} - 3.07[a/W]^{3/2} + 14.53[a/W]^{5/2} - 25.11[a/W]^{7/2} + 25.80[a/W]^{9/2})$.

Cylindrical test specimens for the compression strength (CS) test were made by incrementally placing the materials in a silicon mould of 6 mm in height and 4 mm in diameter (Fig 1b).

The compressive strength was calculated using the following formula²⁵:

$$CS = 4F/\pi d^2$$

Where F is the maximum applied load (N) and d is the diameter of the specimen (mm).

Cubic test specimens to measure the static load-bearing capacity ($9.5 \times 5.5 \times 3 \text{ mm}^3$) were fabricated in an open silicon mould covered with a Mylar sheet and a glass slide of 1 mm thickness (Fig 1c).

Polymerisation of the specimens was made using a hand light-curing unit (Optilux-501, Kerr, CT, USA) for 40 s from both sides of the metal mould and incrementally from the top of silicon mould. The wavelength of the light was between 380 and 520 nm with maximal intensity at 470 nm, and light irradiance was 800 mW/cm^2 . The specimens from each group ($n = 8$) were either stored dry or water stored (37°C for 30 days). The dry-

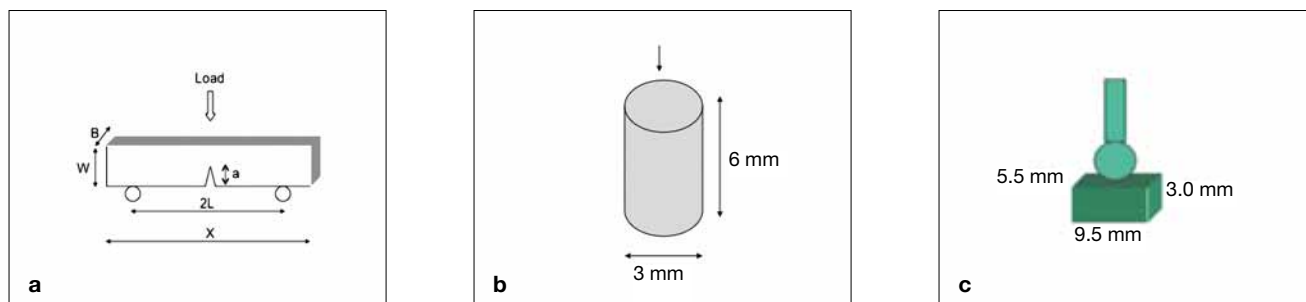


Fig 1 a) Schematic illustration of a specimen used for fracture toughness testing ($B = 3$ mm; $W = 6$ mm; $X = 25$ mm; $L = 10$ mm; $a = 3$ mm). b) Schematic diagram of the cylindrical test specimen used in the compression test. c) Schematic drawing of the test specimen and the static load-bearing capacity test.

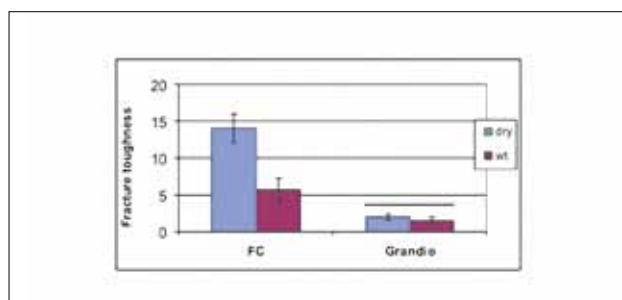


Fig 2 Fracture toughness ($\text{MNm}^{-1.5}$) of the experimental FC composite and conventional restorative composite (Grandio). Vertical lines represent standard deviations. Horizontal line above the bars indicates groups that do not differ statistically significantly from each other.

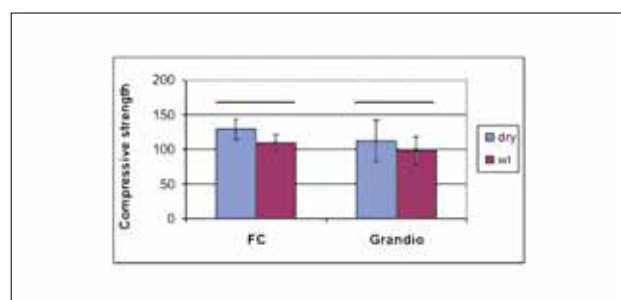


Fig 3 Compressive strength (MPa) of experimental FC composite and conventional restorative composite (Grandio). Vertical lines represent standard deviations. Horizontal line above the bars indicates groups that do not differ statistically significantly from each other.

stored (room temperature) specimens were tested 24 hours after their preparation.

A three-point loading test and compression tests were conducted according to ISO 10477 (cross-head speed of 1 mm/min). All specimens were loaded in a material testing machine using a spherical loading tip ($\varnothing 2.0$ mm) (model LRX, Lloyd Instruments, Fareham, England) and the load-deflection curves were recorded with PC computer software (Nexygen 4.0, Lloyd Instruments). A static load fracture test was carried out to determine the load-bearing capacity of each group using a steel ball ($\varnothing 3.0$ mm) until fracture.

Mean values of fracture toughness, compressive strength and load-bearing capacity were examined with analysis of variance (ANOVA) at the $P < 0.05$ significance level with SPSS version 13 (Statistical Package for Social Science, SPSS, Chicago, IL, USA) to determine the differences between the groups.

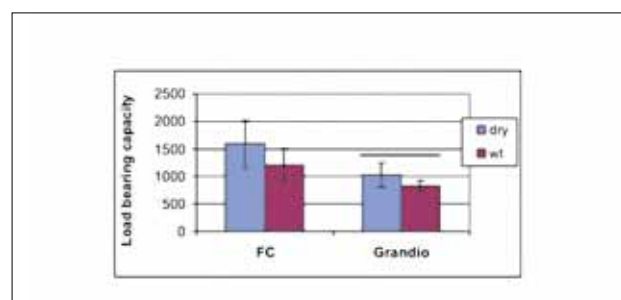


Fig 4 Static load bearing capacity (N) of experimental FC composite and conventional restorative composite (Grandio). Vertical lines represent standard deviations. Horizontal line above the bars indicates groups that do not statistically significantly differ from each other.

Results

The mean values of fracture toughness, compressive strength and load-bearing capacity of tested groups with standard deviations (SD) are summarised in Figures 2 to 4.

One-way ANOVA revealed that experimental FC composite had statistically significantly higher fracture toughness ($14 \text{ MNm}^{-1.5}$), compressive strength (129 MPa) and static load-bearing capacity (1584 N) compared to the control ($2 \text{ MNm}^{-1.5}$, 112 MPa and 1031 N) ($P < 0.05$) in dry conditions. Water storage decreased the mechanical properties in both materials as compared to dry storage.

Discussion

Currently, the performance of biomaterials is most often evaluated using laboratory tests. Teeth and restorations are always subjected to both flexural and compressive forces during chewing. Moreover, these stresses may be present to a higher degree during parafunction (e.g., bruxism and clenching). Clinical studies, after years of follow-up of indirectly or directly made conventional composite restorations in high stress-bearing areas, have shown that fracture of the restoration was a common type of failure.

Recently, the present authors showed that the use of semi-IPN matrix in combination with short glass fibres in restorative filling composite resin gives encouraging results^{21,22}.

Fracture toughness and compressive strength tests are important techniques used for screening of resin-based restorative materials⁹. However, it is likely that fracture toughness is more meaningful than strength for brittle materials as the critical effect of surface defects is accounted for by the presence of a notch.

The results of this study showed improvements in fracture toughness, compressive strength and load-bearing capacity of dental composite resin reinforced with short E-glass fibre filler in comparison with conventional restorative composite. However, the effect of fibre reinforcement on the fracture toughness test was clearer than the compression test, where fibres were oriented mostly in the same load direction. The fibres of the FC composite resin were oriented from an original 3D random orientation (isotropic) to a 2D random orientation (anisotropic) with the long axis of the specimens. It has been shown early on that fibre orientation is an important factor influencing the mechanical properties of FRC^{15,18}.

The present findings are in agreement with a study by Kim and Watts, which showed that fracture tough-

ness of polymer-based materials was improved when they were reinforced with glass fibre-reinforced composite⁹. In contrast, Drummond et al have shown low values of fracture toughness of a fibre containing dental composite²⁶. However, the length of fibres used were in micro-scale. A clinical study reported by Van Dijken showed that a restorative composite resin with micro-scale fibres suffers extensive wear and fracture²⁷, which can be partly explained by the fact that the fibre length used was well below the critical fibre length. It was measured by using a fibre fragmentation test in which the critical fibre length of E-glass with Bis-GMA polymer matrix varied between 0.5 and 1.6 mm²⁸. In order for a fibre to act as an effective reinforcement for polymers, stress transfer from the polymer matrix to the fibres is essential⁵. This is achieved by having a fibre length equal to or greater than the critical fibre length⁵. Therefore, the length of the fibres used as fillers in this study was chosen to be 3 mm, thus exceeding the critical fibre length. The reinforcing effect of the fibre fillers is based on stress transfer from polymer matrix to fibres, but also the behaviour of individual fibre as a crack stopper. Random fibre orientation and lowered cross-linking density of the polymer matrix by the semi-IPN structure is likely to have had a significant role in the mechanical properties.

Stress applied to the teeth and dental restorations is generally low and repetitive rather than high and isolated in nature. However, because of a linear relationship between fatigue and static loading, the compressive static test also gives valuable information concerning load-bearing capacity⁷. Water storage decreased the mechanical properties in all of the specimens. In the polymer matrix, water acts as a plasticiser, increasing free volume and decreasing the glass transition temperature of the polymer matrix²⁹. Also, previously it has been reported that there is a potential deteriorative effect of water on the interfacial adhesion between polymer matrix to glass fibres through rehydrolysis of the silane coupling agent²⁹. However, a recent study by Vallittu showed only minor reduction in flexural strength of E-glass fibre-reinforced composite after 10 years of water storage³⁰. Composite FC might absorb more water than conventional composite, due to the lower filler content of FC compared with conventional composites²¹. The amount of the absorbed water is also affected by the hydrophilicity of the polymer matrix and the chemical stability of the filler particle in water²¹.

Based on the results of this study and previously published data of short-fibre composite resin, it is suggested that experimental FC composite could be used successfully to fulfil the requirements for the ideal

posterior restoration. However, it should be emphasised that clinical trials are necessary in order to evaluate the usefulness of FC composite resin in dental restorations.

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