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Depth of cure and surface microhardness of experimental short fiber-reinforced composite

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Abstract

Objectives. The aim of this study was to analyze the depth of cure of a short fiber-reinforced composite (FRC) assessed by microhardness at different curing times and storage conditions. ***Material and methods.*** Experimental composite resin (FC) was prepared by high-speed mixing 22.5 wt% short E-glass fibers (3 mm in length) and 22.5 wt% resin matrix and gradually adding 55 wt% silane-treated silica filler. Half-split cylindrical test specimens were produced from both the FC and from the conventional particulate composite resin (control, Z250, 3M-ESPE). The test specimens ($n = 3$/group) were polymerized at different exposure times (20, 40, 60 s) and then water-stored at 37°C for 24 h and 30 days before testing. A universal testing machine was used for testing Vickers microhardness. All results were statistically analyzed with analysis of variance (ANOVA). ***Results.*** ANOVA revealed that curing time had a significant effect ($p < 0.05$) on the microhardness of both composite resins. Depth of cure of conventional composite resin (control) was significantly greater than that of FC ($p < 0.05$). Microhardness after water storage decreased as curing time increased. ***Conclusions.*** The use of short fiber fillers in interpenetrating polymer network matrix (IPN) achieved the acceptable depth of cure and microhardness values recommended for clinical use, although lower than for commercial composite resin.

Key Words: *Depth of cure, experimental fiber composite resin, microhardness*

Introduction

One of the problems associated with using light-cured composite resin directly in the posterior region is the decrease in curing-light intensity with depth in the material. The intensity of light at a given depth and for a given irradiance period is a critical factor in determining the extent of reaction of monomer into polymer, typically referred to as the degree of conversion [1], and significantly associated with values of mechanical properties [2], biocompatibility [3], color stability [4] and would therefore be expected to be associated with clinical success of the restoration. It is thus important to achieve sufficient irradiance on the bottom surface of each of the incremental layers used in building up the restoration. The concept of the point of sufficiency in this respect is known as depth of cure.

Put simply, depth of cure can be defined as the extent of quality resin polymerization depth from the surface of composite restoratives. The extent of resin cure is affected mainly by filler type and size, monomer and activator type, light source intensity, and duration of exposure [5]. Inadequate polymerization throughout the restoration bulk can lead to undesirable effects, e.g. gap formation, marginal leakage, recurrent caries, adverse pulpal effects and ultimate failure of the restoration [2]. Microhardness testing with increasing depth in a composite has been used in many studies, because surface hardness has been shown to be an indicator of degree of polymerization [6].

For over 30 years now, glass fibers have been investigated with a view to reinforcing dental polymers [7]. Compared to carbon or aramid fibers, glass fibers have documented reinforcing efficiency and good esthetic qualities [8]. The effectiveness of fiber reinforcement is dependent on variables such as the resins used, the quantity of fibers in the resin matrix [9,10], their length [9], their form [11], their orientation [12], their adhesion to the polymer matrix [13], and impregnation of fibers with the resin [14]. Short random fibers provide a thermomechanically isotropic reinforcement effect in multidirections rather than in just one or two directions,

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as described by Krenchel [15]. In terms of optical properties of FRC, there is some information that unidirectional FRC is anisotropic and thus that multidirectional short FRC could be isotropic [16].

Polymethyl methacrylate (PMMA) and dimethacrylate based semi-interpenetrating polymer network (semi-IPN) matrix have been established as a polymer matrix in denture base materials [17]. Although previous investigations [18–21] on the use of experimental semi-IPN matrix in combination with short E-glass fibers in restorative filling composite have shown enhancement in mechanical properties and load-bearing capacity, the effect of short glass-fiber reinforcement on curing depth has not been reported.

It has been hypothesized that using isotropic short-fiber fillers could induce the light transmission of composite resin. The aim of this study was therefore to evaluate the curing depth of experimental short glass-fiber composite resin with different curing times and storage conditions.

Material and methods

Material

Dimethacrylate (BisGMA 67% [bisphenol A-glycidyl dimethacrylate], TEGDMA 33% [triethylenglycol dimethacrylate], CQ and DMAEMA 0.7% [camphorquinone and dimethylaminoethylmethacrylate]) resin consisting of 50 wt% nanofillers (SiO2, 20 nm in size) (Hanse Chemie, Geesthacht, Germany) and E-glass fibers with BisGMA-PMMA [polymethylmethacrylate, M_w 220.000] resin matrix (everStick, StickTech, Turku, Finland) were used. In addition, radio-opacity fillers of $BaAlSiO_2$ (3 ± 2 μm in size) (Specialty Glass Products, Willow Grove, Pa., USA) were incorporated in the resin system. Before the $BaAlSiO_2$ filler particles were incorporated within the resin matrix, they were silane-treated using a previously defined technique [22]. A commercial particulate filler composite (shade A2) (Z250, 3M-ESPE, St. Paul, Minn., USA) was used as control material.

Methods

Experimental fiber composites (FC) were prepared by mixing 22.5 wt% of short E-glass fibers (3 mm in length) and 22.5 wt% of resin matrix and gradually adding 55 wt% of $BaAlSiO_2$ radio-opacity fillers. A high-speed mixing machine was used at 3500 rpm for 5 min (SpeedMixer, DAC; Hauschild Engineering, Hamm, Germany). The dimethacrylate-based resin matrix consisting of PMMA forms semi-IPN polymer matrix for the fiber composite (FC).

Cylindrical test specimens were made by placing the materials in a split metal mold 6 mm in height and 5 mm in diameter. Polymerization of the specimens was done using a hand light-curing unit (Optilux-501; Kerr Corporation, Orange, Calif., USA) for 20 s, 40 s or 60 s from the top of the mold and in close contact. The wavelength of the light was between 380 and 520 nm, with maximal intensity at 470 nm; light irradiance was 800 mW/cm_2.

Once the specimens ($n = 3$) were polymerized, they were removed from the molds and ground longitudinally until half of the specimen was left. The surface was then polished (grit up to 4.000 FEPA) at 300 rpm under water cooling using an automatic grinding machine (Rotopol-1; Struers, Copenhagen, Denmark).

In order to remove polishing debris, all specimens were cleaned ultrasonically (Ultrasonic L&R, Kearny, N.J., USA) for 15 min and then stored in distal water at 37°C for either 24 h or 30 days before testing.

Microhardness was measured using a Struers Duramin hardness microscope (Struers, Copenhagen, Denmark) with a 40 objective lens and a load of 1.96 N applied for 10 s. Each sample was subjected to 10 indentations on each 0.5 mm, starting from the top and moving towards the bottom of the specimen. The diagonal length impressions were measured and Vickers values were converted into microhardness values by the machine.

Microhardness was obtained using the following equation:

$$H = \frac{1854.4 \times P}{d^2}$$

where H is Vickers hardness in N/mm^2, P is the load in N and d is the length of the diagonals in mm [23]. Depth of cure at 1 mm depth should not be less than 70% of the values measured at top surface hardness [24].

Microhardness data were statistically analyzed with analysis of variance (ANOVA) at the $p < 0.05$ significance level with the Statistical Package for Social Sciences, v. 13 (SPSS, Chicago, Ill., USA), followed by Tukey's post hoc analysis to determine the differences among groups.

Results

Figure 1 summarizes and compares the depth of cure of tested materials as evaluated by different curing time. Depth of cure of conventional composite resin (control) was significantly greater than that of experimental FC composite resin ($p < 0.05$). ANOVA revealed that curing time had a significant effect ($p < 0.05$) on depth of cure and microhardness of both composite resins. As curing time increased, depth of cure and microhardness increased. Surface microhardness was significantly ($p < 0.05$) lower in the FC composite group cured for 20 s (mean (SD):

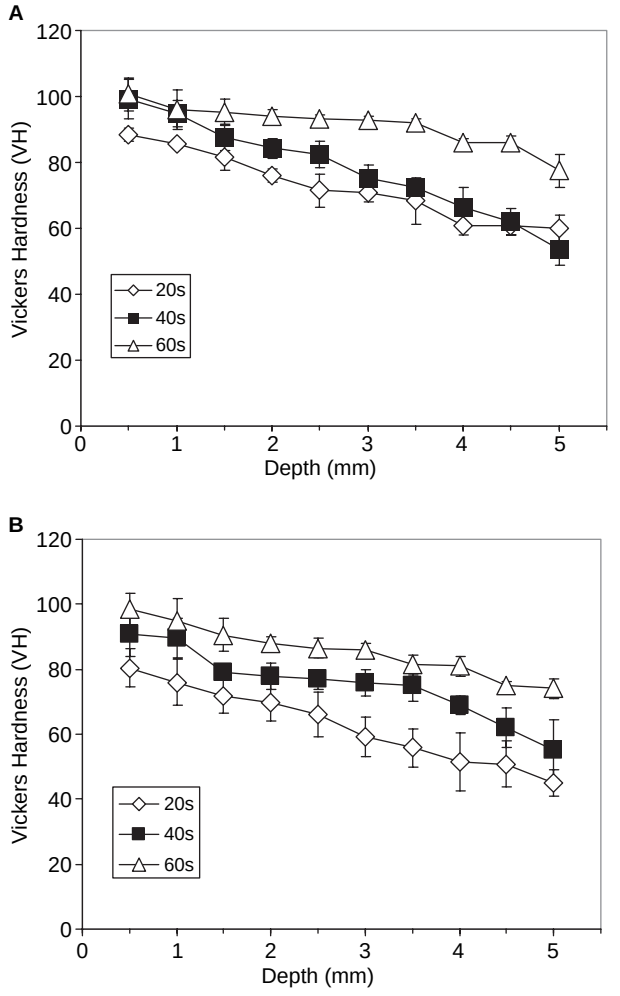

Figure 1. A. Curing depth of conventional restorative composite Z250 stored as dry condition using different curing times. B. Curing depth of experimental FC composite at different curing times stored as dry condition using different curing times.

63 (12) N) than in the control group (73 (10) N). No significant differences were found between the FC and control groups cured for 40 s (mean (SD): 75(11) N and 78(14), respectively) and 60 s (mean (SD): 84(8) N and 91(7) N).

The data from water storage specimens in both materials showed that as curing time increased the microhardness and depth of cure decreased ($p < 0.05$). Microhardness after water storage decrease ranged between 12% and 14% in the group cured for 20 s, between 15% and 17% in the group cured for 40s, and between 21% and 24% in the group cured for 60 s.

Discussion

It has been shown recently that use of short glass fibers with semi-IPN matrix in restorative filling composite resin produces encouraging results [18–21].

Traditional methods for evaluating depth of cure are performed in accordance with ISO 4049. This method is based on the scraping technique, i.e. uncured material is removed with a plastic spatula after curing and the remaining height of the cured cylinder is measured. However, since this technique provides only a rough estimate of depth of cure, as the scraping force is difficult to standardize, a technique using a penetrometer, as suggested by Harrington & Wilson [25,26], was used to enhance the accuracy of the test. The microhardness test was chosen in this study to determine the depth of cure for its clinical relevance, because it provides data about physical properties considered essential for the favorable performance of dental composites [6,27]. ISO 10477 defines the technique, which uses the Vickers microhardness method to evaluate depth of cure, because it is widely used in scientific studies as an indirect method of determining the composite degree of monomer conversion and also because of good correlation with infrared spectroscopy [24,28].

In general, the present study has shown that commercial composite resin achieves higher values of microhardness and depth of cure than experimental FC composite, partly explainable by the difference in filler type and contents between the two materials. In addition, some of the difference could also be explained by the difference between polymer matrices of pure thermoset and semi-IPN. Semi-IPN matrix lowers the cross-linking density of resin matrix, which leads to decreased microhardness of the composite resin. Since both materials achieved the clinical requirement, it may seem the differences found have no clinical relevance because, according to the resin composite manufacturer, the increments should not exceed 2 mm. On the other hand, in some clinical situations the light guide tip cannot be placed in close contact with the restoration surface, as was the case during this study. Therefore, any increase in the depth of cure obtained by curing should be considered important for daily clinical practice. Nevertheless, it has to be kept in mind that the experimental FC composite evaluated in this study showed a lower rate of polymerization shrinkage and microleakage than the commercial composite resin used, although they have the same degree of monomer conversion [18,21].

Microhardness values decreased with increasing depth and improved with rising curing time (Figure 1). Material nearer to the light source underwent more complete polymerization and was thus harder. This finding conflicts with that of Soh et al., who demonstrated that higher hardness values are obtained at 1 mm below the surface compared to the top [6]. Other factors that may influence depth of cure are shade of composite resin, type of curing unit and method of curing, all widely discussed in the literature [5,6,27,28]. Moreover, the light scattering and absorption coefficients of composite resins, which affect the light distribution, should also be taken into consideration. Le Bell et al. [16]

have shown that unidirectional fiber-reinforced composites conduct and scatter the light better than conventional composite resin. However, in this study we used short randomly oriented fibers.

It was no surprise that water storage decreased the surface hardness of the specimens. In the polymer matrix, water acts as a plasticizer in increasing free volume and decreasing the glass transition temperature of the polymer matrix [29]. It has previously been reported that there is a potential deteriorating effect of water on the interfacial adhesion between the polymer matrix to glass fibers through rehydrolysis of the silane coupling agent [29].

Interestingly, the relative softening of microhardness after water storage increased as a function of increased curing time. To our knowledge, this phenomenon is not well documented in the literature. After a short 20 s curing time, more residual monomer remains in the specimen and will leach out and be replaced by water (plasticizing effect) after 30 days' water storage. In this case, water partly replaces residual monomers. Longer curing time (40 or 60 s) increases the degree of monomer conversion and results in a higher degree of conversion and less residual monomer within polymer matrix. Because of this, water diffusion into polymer matrix causes a more plasticizing effect, as it does not replace residual monomer at the same rate but enters as a new component into polymer matrix. This phenomenon leads to a higher relative drop in surface hardness when degree of conversion is increasing with longer curing time of 40s or 60s.

When simulating clinical conditions, aging processes such as thermal stress, mechanical stress and wear must be taken into consideration. It is well known that microhardness of composite resins is of little use as a predictor of the abrasiveness of these products against human enamel. Thus, *in vitro* wearing evaluation of short glass fiber composite resin with semi-IPN resin matrix will be evaluated in further studies.

Based on the results of this study and on our previous published data on short fiber composite resin, it is suggested that experimental FC composite could be used successfully to fulfill the requirements for the ideal posterior restoration. However, it should be emphasized that clinical trials are necessary if the usefulness of FC composite resin in dental restorations is to be evaluated.

In conclusion, E-glass FRC composite resin with semi-IPN-polymer matrix achieved the acceptable depth of cure and microhardness recommended for clinical use. By increasing the polymerization time, the depth of cure can be increased in both composite resins.

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References

[1] Musanje L, Darvell BW. Curing-light attenuation in filled-resin restorative materials. Dent Mater 2006;22: 804–17.

[2] Ferracane JL, Greener EH. The effect of resin formulation on the degree of conversion and mechanical properties of dental restorative resin. J Biomed Mater Res 1986;20: 121–31.

[3] Geurtsen W, Leyhausen G. Chemical-biological interactions of resin monomer triethyleneglycol-dimethacrylate (TEGDMA). J Dent Res 2001;80:2046–50.

[4] Tanaka K, Taira M, Shintani H, Wakasa K, Yamaki M. Residual monomers (TEGDMA and bis-GMA) of a set visible-light-cured dental composite resin when immersed in water. J Oral Rehab 1991;18:353–62.

[5] Rueggeberg FA, Caughman WF, Curtis JW, Davis HC. Factors affecting cure at depths within light-activated resin composites. Am J Dent 1993;6:91–5.

[6] Soh MS, Yap AUJ, Siow KS. Comparative depths of cure among various curing light types and methods. Oper Dent 2004;29:9–15.

[7] Vallittu PK. A review of fiber-reinforced denture base resins. J Prosthodont 1996;5:270–6.

[8] Vallittu PK, Narva K. Impact strength of a modified continuous glass fiber-poly (methyl methacrylate). Int J Prosthodont 1997;10:142–8.

[9] Vallittu PK, Lassila VP, Lappalainen R. Acrylic resin-fiber composite. Part I: The effect of fiber concentration on fracture resistance. J Prosthet Dent 1994;71:607–12.

[10] Stipho HD. Repair of acrylic resin denture base reinforced with glass fiber. J Prosthet Dent 1998;80:546–50.

[11] Ladizesky NH, Cheng YY, Chow TW, Ward IM. Acrylic resin reinforced with chopped high performance polyethylene fiber properties and denture construction. Dent Mater 1993;9:128–35.

[12] Dyer SR, Lassila LVJ, Jokinen M, Vallittu PK. Effect of fiber position and orientation on fracture load of fiber reinforced composite. Dent Mater 2004;20:947–55.

[13] Vallittu PK. The effect of void space and polymerization time on transverse strength of acrylic-glass fiber composite. J Oral Rehabil 1995;22:257–61.

[14] Miettinen VM, Vallittu PK. Water sorption and solubility of glass fiber-reinforced denture polymethyl methacrylate. J Prosthet Dent 1997;77:531–4.

[15] Vishu S. Handbook of plastic testing technology, 2nd edn. New York: John Wiley; 1998.

[16] Le Bell AM, Tanner J, Lassila LV, Kangsniemi I, Vallittu PK. Depth of light-initiated polymerization of glass fiber-reinforced composite in a simulated root canal. Int J Prosthodont 2003;4:403–8.

[17] Lastumäki TM, Lassila LV, Vallittu PK. The semi-interpenetrating polymer network matrix of fiber-reinforced composite and its effect on the surface adhesive properties. J Mater Sci Mater Med 2003;14:803–9.

[18] Garoushi S, Vallittu PK, Lassila LVJ. Short glass fiber reinforced restorative composite resin with semi-interpenetrating polymer network matrix. Dent Mater 2007;23: 1356–62.

[19] Garoushi S, Vallittu PK, Lassila LVJ. Use of isotropic short fiber reinforced composite with semi-interpenetrating

polymer network matrix in fixed partial dentures. J Dent 2007;35:403–8.
[20] Garoushi S, Vallittu PK, Lassila LVJ. Fracture resistance of short, randomly oriented, glass fiber-reinforced composite premolar crowns. Acta Biomater 2007;3:779–84.
[21] Garouoshi S, Vallittu PK, Watts DC, Lassila LVJ. Polymerization shrinkage of experimental short glass fiber-reinforced composite with semi-inter penetrating polymer network matrix. Dent Mater 2008;24:211–5.
[22] Söderholm KJ, Yang MC, Garcea I. Filler particle leachability of experimental dental composites. Eur J Oral Sci 2000;108:555–60.
[23] Beun S, Glorieux T, Devaux J, Vreven J, Leloup G. Characterization of nanofilled compared to universal and microfilled composites. Dent Mater 2007;23:51–9.
[24] International Organization for Standardization. ISO 10477-Dentistry-polymer based crown and bridge materials. 2004(E).
[25] Campregher UB, Samuel SM, Fortes CB, Medina AD, Collares FM, Ogliari FA. Effectiveness of second-generation light-emitting diode (LED) light curing units. J Contemp Dent Pract 2007;2:35–42.
[26] Harrington E, Wilson HJ. Depth of cure of radiation-activated materials. Effect of mould material and cavity size. J Dent 1993;21:305–11.
[27] Tsai PC, Meyers IA, Walsh LJ. Depth of cure and surface microhardness of composite resin cured with blue LED curing lights. Dent Mater 2004;20:364–9.
[28] DeWald JP, Ferracane JL. Comparison of four modes of evaluating depth of cure of light-activated composites. J Dent Res 1987;66:727–30.
[29] Lassila LVJ, Nohrström T, Vallittu PK. The influence of short-term water storage on the flexural properties of unidirectional glass fiber-reinforced composites. Biomaterials 2002;23:2221–9.