Flexural strength of direct composites submitted to post-polymerization heat
Resistência à flexão de compósitos diretos submetidos à polimerização térmica adicional

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Abstract

The purpose of this study was to assess the effects of post-polymerization heat on the flexural strength of direct composites. Direct composite specimens (n=10), measuring 25×2×2 mm (Z-250 and P-60), were polymerized by 3 methods: (1) light-polymerized for 40 seconds, (2) (1) + post-polymerization in an oven at 120ºC for 20 minutes, and (3) (1) post-polymerization in an autoclave at 120ºC for 20 minutes. Specimens (n=10) of the indirect composite Artglass (control group) were polymerized in a UniXS oven. Specimens were stored in distilled water for 24 hours at 37ºC and then submitted to a flexural 3-point bending test with a universal testing machine. Flexural strength (MPa) data were analyzed by one-way ANOVA and Tukey´s multiple comparison test (α=.05). Results showed that (1) additional heat polymerization in an oven provided significantly (p<.05) higher flexural strengths (Z250: 158.85 MPa and P60: 147.10 MPa) than that of the Artglass resin (121.52 MPa); (2) with additional heat polymerization in an autoclave, these direct resins presented mean flexural strengths (Z250: 134.54 MPa and P60: 130.18 MPa) similar to that of Artglass (P>.05); (3) groups that were only light-polymerized (Z250: 124.75 MPa and P60: 128.96 MPa) were similar to Artglass (P>.05). When composites were only light-polymerized, they showed a flexural strength behavior similar to that of the indirect composite Artglass. The post-polymerization heat do not enhanced the resistance of the Filtek P60, but increased the strength of the Filtek Z250 when additional heat polymerization in a oven

Uniterms

Dental materials; composite resins; dental esthetics; material testing

Introduction

During the last decade, the development of new esthetic restorative materials and more efficient dentin/enamel bonding systems have provided replacements for metal restorations. Another factor resulting from the increased use of posterior composite restorations is the esthetic advantages of nonmetal restorations[5]. The ideal conditions for posterior composites are small carious lesions in premolars, where there is no evidence of excessive occlusal wear[5]. However, in complex preparations, in areas of high occlusal stress, the use of direct composites may be contraindicated due to their limitations, including insufficient wear resistance, marginal breakdown, difficulty in obtaining a proximal contact, inadequate occlusal adjustment, finishing and color instability[1].

Indirect restorative materials, commercially called "ceromers", were developed in 1995 to overcome some of the undesirable properties of porcelain, such as the difficulty of intraoral repair and occlusal adjustment, and the possibility of wear to opposing teeth[6]. These materials, however, require the use of special equipment for processing, thus increasing the cost of the restoration. Indirect composites present a chemical composition similar to those of the light-polymerized direct composites; yet, the polymerization processes utilized are often more efficient[6].

The indirect composite Artglass is identified by the manufacturer as a non-conventional dental polymer. The filler component is essentially a radiopaque barium glass with a mean particle size of 0.7µm. A moderate amount of colloidal silica is also incorporated for the purpose of enhancing certain handling characteristics.

The formulation of Artglass is considerably more complex. In addition to conventional bi-functional molecules (Bis-GMA or UDMA), it contains 4 to 6 functional groups (manufacturer's literature). This

configuration provides an opportunity for more double-bound conversions, with the potential for creating a higher level of cross-linking (manufacturer's literature). Consequently, this can improve wear resistance and other physical and mechanical properties of the resin matrix[6]. Results from the study by Freiberg & Ferracane[7] (1998), however, do not provide direct evidence between the greater degree of monomer conversion (DC) and greater cross-linking. The composites Artglass and Charisma showed significant differences in DC and physical properties, when compared using similar curing methods. Artglass showed a greater degree of monomer conversion and fracture toughness, but a lower flexural strength, flexural modulus, hardness and wear resistance[7]. The authors pointed out that significant improvements in the mechanical properties of these materials compared to directly placed composites have not been evident.

In light-polymerized composites, 30 to 45% of the monomer remains unreacted[13]. With the aim of improving the mechanical properties of light-polymerized resin composites, allowing their use as inlays/onlays in extensive posterior restorations, the use of additional polymerization procedures has been suggested[8,10,14-16]. Extraoral heat treatment can increase mechanical properties as a result of the increase in DC of the resin matrix[20]. Bagis & Rueggeberg[3] (2000) demonstrated an expressive decrease in the amount of unreacted monomers in a resin composite with post-cure heating and pointed out that this decrease could be due to a number of mechanisms. This reduction can lead to enhanced physical properties and a lower level of leachable material diffusion into the patient's mouth.

This latter consideration would enhance the potential biocompatibility of the restoration[2-3].

Additional polymerization of composite resins have frequently been obtained in special ovens using light and heat, and/or a nitrogen atmosphere[18], with previously determined times and temperatures. Autoclaves and ovens are equipment that are available in dental offices, and are an important source of heat. Studies regarding the effects of temperature associated to the water present in the autoclave seem interesting, since there is a scarcity in literature of data directly related to this information. Hence, the purpose of this study was to evaluate the effects of two post-polymerization methods, oven and autoclave, upon the flexural strength of direct resin composites, in comparison to the indirect composite Artglass.

MATERIAL AND METHODS

Two direct composites, Filtek Z250 and Filtek P60 (3M ESPE, St. Paul, Minn.), and one indirect composite (Artglass; Heraeus Kulzer, Wehrheim, Taunus, Germany) were used in this study (Table 1). Artglass resin contains glass barium (0.7 µm average size) and a special rheological glass (silica; average size approximately 1.0 µm) with microfiller formulations (68 vol %). The resin matrix is a mixture of dimethacrylate and multi-functional metacrylate monomers. Both direct composites (Filtek Z250 and Filtek P60) contain glass silica (0.6 µm average size) and microfiller formulations of 60 vol % and 61 vol %, respectively. The resin matrix contains only dimethacrylate monomers (Bis-GMA, TEGDMA, UDMA, Bis-EMA) (Picture 1).

Picture 1 – Materials tested and composition

	Code	Filler content (% vol)	Resin matrix	Manufacturer
Filtek Z-250	Z250	60	Bis-GMA, TEGDMA, UDMA, Bis-EMA	3M ESPE, St. Paul, Minn.
Filtek P-60	P60	61	Bis-GMA, TEGDMA, UDMA, Bis-EMA	3M ESPE, St. Paul, Minn.
Artglass	Ar	68	Bis-GMA, multifunctional metacrylic ester	Heraeus Kulzer, Wehreim, Alemanha

Note: The abbreviations used above are: Bis-GMA, bisphenol-A glycidyl dimethacrylate, TEGDMA, triethyleneglycol dimethacrylate, UDMA, urethane dimethacrylate, Bis-EMA, bisphenol-A polietileno glycol dieter dimetracrilato.

A stainless steel mold (25×2×2 mm) was utilized to produce flexural test bars, following the procedure described by the International Organization for Standardization[9] (ISO 4049 1988). The unpolymerized material was pressed into a mold and covered with a mylar strip (Polidental Ind. e Com. Ltd, São Paulo, São Paulo, Brazil). One glass slide was placed on top of the mold and light pressure was applied to expel any excess material and trapped air. The specimens (n=10) were pre-polymerized with a continuous light curing unit (XL 2500; 3M ESPE, St. Paul, Minn., USA), with median intensity of 600 mW/cm2, and the tip (7 mm of diameter) was maintained 1 mm from the upper glass surface in 4 locations for the exposure time of 40 seconds. After light-polymeri-zation, the specimen surfaces were ground on #600 grit silicon carbide paper (Buehler, Lake Bluff, Illinois, USA) to remove surface irregularities. The specimens were immediately submitted to additional heating at 120ºC for 20 minutes, either in dry heat (Oven Olidef CZ; Industry and Com. of Appliance Hospital Ltd., Ribeirão Preto, São Paulo, Brazil) or in wet heat (Autoclave; Cientific Equipaments Phoenix, Araraquara, São Paulo, Brazil). The specimens (n=10) of the indirect composite (Artglass; Heraeus Kulzer) were polymerized for a total de 180 seconds (90 seconds per side) in a xenon stroboscopic light curing unit (UniXS; Heraeus Kulzer). The polymerization conditions of the composite materials are shown in Picture 2.

Picture 2 – Polymerization methods

	Polymerization method
Z250 P60	A – light-polymerized for 40 seconds[£] B – light-polymerized for 40 seconds[£] and dry heat at 120ºC for 20 minutes[§] C – light-polymerized for 40 seconds[£] and wet heat at 120ºC for 20 minutes
Artglass	Light-polymerized for 90 seconds

[£] XL 2500 light source, 3M ESPE, St. Paul, Minn.

[§] Oven Olidef CZ, Industry and Com. of Appliance Hospital Ltda., Ribeirão Preto, São Paulo, Brazil.

Autoclave, Cientific Equipaments Phoenix, Araraquara, São Paulo, Brazil.

Oven UniXS, Heraeus Kulzer, Wehrheim, Taunus, Germany.

All specimens were stored in distilled water at 37ºC for 24 hours, prior to testing. The specimens were then submitted to a flexural 3-point bending test with a universal testing machine servo-hydraulic (MTS model 810, MTS System Corporation, Minn., USA). The distance between the supporting wedges was 20 mm and the cross-head speed was .75 mm min-1. The flexural strength σ (MPa) values were calculated using the following equations[9]: $\sigma = 3\,PL/2\,bd^2$, where, P is the applied load at fracture (N), L is the length between the jig wedges (20 mm), b is the width, and d is the thickness.

The flexural strength data were analyzed by factorial analysis of variance (one-way ANOVA) and Tukey multiple comparison tests ($\alpha=0.05$).

RESULTS

Table 1 presents the factorial ANOVA results for flexural strength, which revealed significant differences ($P<.05$).

Table 1 – ANOVA results for flexural strengths

Source of Variation	Sum of squares	Degree of freedom	Mean square	F-value	P-value
Between groups	10832.0000	6	1805.3334	5.96	0.015*
Within groups	19096.2500	63	303.1151		
Total	29928.2500	69			

* Significant (P<.05)

Table 2 shows mean values of flexural strength, standard deviation and grouping for each material/polymerization method combination. Filtek Z250 and Filtek P60 resins, when post-polymerized in an oven, demonstrated similar mean flexural strengths; however, these strengths were significantly (p<.05) higher when compared to the Artglass indirect composite. When an autoclave was used, these resins presented mean flexural strengths similar to that of the only light-polymerized groups and to Artglass. For Filtek Z250, however, post-polymerization in an autoclave resulted in resistance significance lower than that of the group post-polymerized in an oven. When resins were only light-polymerized, there were no significant differences in flexural strength when compared to Artglass.

Table 2 – Mean flexural strength (MPa), standard deviation and grouping

Material/polymerization method	Mean	Grouping
Z-250/light-polymerized + oven	158.85 (15.46)	a
P-60/light-polymerized + oven	147.10 (26.03)	ab
Z-250/light-polymerized + autoclave	124.75 (20.90)	c
P-60/light-polymerized + autoclave	128.96 (11.49)	bc
P-60/only light-polymerized	130.18 (19.48)	bc
Z-250/only light-polymerized	134.54 (14.96)	bc
Artglass	121.52 (10.18)	c

Tukey Value = 21.76.

s.d. are given in parentheses.

Identical letters indicate values are not statistically different (P>.05).

DISCUSSION

In the present study, the flexural strength test was used because both tension and compressive events may occur and flexural stress is developed, which can be used to represent a material's ability of resisting to deformation or fracture[8].

The Filtek Z250 and Filtek P60 resins, which were heat polymerized in an oven, did not present differences between themselves and were significantly stronger than Artglass. This result has important clinical implications, since the heat polymerization in an oven for 20 minutes produced a material with higher flexural strength than Artglass, which requires special oven and techniques.

Venhoven et al.[19] (1996) suggested that the properties of a composite are influenced by type, size and concentration of the inorganic filler, and the degree of conversion of the components of the resin matrix. The type of matrix and degree to which conversion occurs during polymerization also influences composite's properties.

Regarding the treatments applied to the direct composites, the oven increased the strength of the Filtek Z250 resin, when compared to the condition of light-polymerization alone. This increase is in agreement with previous studies, which demonstrated that direct composites submitted to additional heat-polymerization showed an improvement in physical properties[13-4]. Wendt[20] (1987) clinically evaluated heat-treated composite resin inlays for 12-months and compared them with light-polymerized inlays. Heat-treated inlays showed alleviated postoperative sensitivity, better marginal integrity, increased longevity of marginal adaptation, and higher resistance to secondary caries, as well as no change in color and esthetic appearance. Park & Lee[13] (1996) & Park[14] (1996) also demonstrated the effect of heat polymerization on the degree of monomer conversion as an increase in the physical properties of resins employed in the inlay technique, which is also in agreement with a previous report by Asmussen[1].

The possible effect of temperature is the increase in the conversion of residual monomers in the organic matrix. The high temperature of the oven may produce an increase in the mobility of the dimethacrylate monomer[15], and result in a more homogenous polymer matrix with a low content of residual monomers[16]. This may explain the high conversion of the double bonds (C=C to C-C) obtained with additional heat polymerization, which occurred with the Filtek Z250 resin, compared with the resin that was only light-polymerized[15].

Both assessed direct composites (Filtek Z250 and Filtek P60) have a fairly similar composition – the manufacturer, however, does not inform the concentration of each monomer of the matrix (Table 1), but still showed different flexural strength behaviors when submitted to additional oven polymerization. There are evidence in the literature that explain such an event, since the heat treatment itself does not imply better mechanical properties because properties may also be dependent on resin composition, degree of conversion, presence of residual monomers, diluents type and aqueous environment in addition to temperature[8,11].

An autoclave, in order to provide additional heat polymerizations, was used in the present study due to the availability of this equipment in dental offices. Both direct composites (Filtek Z250 and Filtek P60) when treated in an autoclave, presented flexural strength values that were similar to those of the group that underwent only light-polymerization and that of the Artglass resin. Treatment of the Filtek Z250 resin in an autoclave resulted in lower flexural strength values than specimens treated in an oven (Table 4).

Since heating in an autoclave occurs by the boiling of water, according to Ho et al.[8] (2001), the effect of temperature is supplemented by water and the structure goes through softening, which is a coupling of thermal energy and reduced viscous drag forces, because of the plasticizing effect of water. Water absorption can induce a plasticizing effect on the polymer matrix as well as debonding of the filler from the matrix, which can lead to an increased creep formation[12]. This plasticizing effect of the composites occurs by penetrating the matrix and leaching unreacted monomer and filler[11,17], which may explain why the autoclave treatment had no effect on the composites Filtek Z250 and Filtek P60.

TEG-DMA (triethyleneglycol dimethacrylate) used as solvent and as an internal plasticizer because of its flexible nature, associated to UDMA (an aliphatic diacrylate with a urethane group), which, because of the long aliphatic segment in the repeated unit, leads to greater chain flexibility and coiling[4]. This flexible nature of the matrix tends to undergo softening because of water and temperature and thus behaves as a rubbery material[4]. Due to the presence of these components in Filtek Z250 and Filtek P60 composites, the effect of temperature on the increase of the monomer conversion degree may have probably been annulled by the presence of the water. Bagis & Rueggeberg[2] (1997) also pointed out which monomer conversion levels attained by the polymer would not be susceptible to lowering with time after cure or by water storage, as are the physical and mechanical properties of post-cure heating materials. Due to the variety of factors, it is difficult to determine which one is the most responsible for the results obtained in the autoclave.

Direct composites, when only light-polymerized, presented statistically similar flexural strength values when compared to Artglass processed in a laboratory. This was unexpected, since it was imagined that Artglass would show better strength because, besides the bi-functional Bis-GMA, it contains additional

multifunctional monomers. Also, its method of polymerization could lead to a higher degree of conversion and consequently better mechanical properties[6]. The study by Freiberg & Ferracane[7] (1998), which compared Artglass and Charisma polymerized by the same process, showed that, despite the greater degree of monomer conversion, Artglass presented lower flexural strength, flexural modulus, hardness, and wear resistance. According to the authors, these differences between the two composites are probably due to the differences in resin formulation.

A possible explanation for the similar flexural strength values of the indirect composite Artglass and the direct composites Filtek Z250 and Filtek P60, only light-polymerized, is that, just as it occurs with the direct composites that have a higher polymer chain flexibility due to the presence to the monomers TEG-DMA (triethyleneglycol dimethacrylate) and UDMA (an aliphatic diacrylate with a urethane group)[4], Artglass shows a formation of a more flexible, stress-relieving polymer network[7]. These results support the manufacturer's claims that the multifunctional monomer used in Artglass makes this composite more resistant to chipping and breaking as well as being less stiff and more resilient than a typical Bis-GMA-based composite[7].

Although the present study has shown an increase of the flexural strength of the resin composites tested with additional heat polymerization in an oven, there is a need for further investigations of other physical and mechanical properties in order to confirm if the results are, indeed, favorable. Additionally, other indirect composites besides Artglass must be tested, since this material involves different polymerization procedures, including light-polymerization, heat, and/or a nitrogen atmosphere[18].

In addition to the knowledge of physical and mechanical characteristics of the additional heat-cured direct composites, long-term clinical evaluation studies are essential before indicating these materials as reliable replacements of indirect composites.

Conclusions

Within the limitations of this study, the following conclusions were drawn:

1. The Filtek Z250 and Filtek P60 direct composites, when light-polymerized alone or post-polymerized in an autoclave, presented statistically similar flexural strengths to that of the indirect composite Artglass;
2. The additional heat polymerization of the Filtek Z250 and Filtek P60 resins in an oven produced a significantly higher flexural strength than that of Artglass;
3. Neither post-polymerization techniques increased the flexural strength of the Filtek P60 resin, when compared to the light-polymerized alone group;
4. Additional heat polymerization in an oven increased the flexural strength of the Filtek Z250 resin in relation to the light-polymerized alone group.

Resumo

O objetivo deste estudo foi avaliar os efeitos da polimerização térmica adicional na resistência à flexão de compósitos diretos. Os espécimes dos compósitos diretos (Z250 e P60) (n=10), medindo 25×2×2 mm, foram polimerizados por três métodos: (1) fotopolimerização por 40 segundos, (2) (1) + polimerização adicional em estufa a 120°C por 20 minutos, e (3) (1) + polimerização adicional em autoclave a 120°C por 20 minutos. Os espécimes (n=10) do compósito indireto Artglass (grupo controle) foram polimerizados no forno UniXS. Os espécimes foram armazenados em água destilada por 24 horas a 37°C e então submetidos ao teste de resistência à flexão. Os dados de resistência à flexão foram analisados por análise de variância e teste Tukey (α=0,05). Os resultados mostraram que (1) a polimerização térmica adicional em estufa resultou em resistência à flexão significativamente maior (p<0,05) (Z250: 158,85 MPa e P60: 147,10 MPa) que a resina Artglass (121,52 MPa); (2) com a polimerização em autoclave, os compósitos diretos apresentaram médias de resistência à flexão (Z250: 134,54 MPa e P60: 130,18 MPa) similares à resina Artglass (p>0,05); (3) os grupos apenas fotopolimerizados (Z250: 124,75 MPa e P60: 128,96 MPa) foram similares à resina Artglass (p>0,05). Quando os compósitos foram apenas fotopolimerizados, eles mostraram comportamento de resistência à flexão similar ao compósito indireto Artglass.

Unitermos

Materiais dentários; resinas compostas; estética dental; teste de materiais

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