Effects of heat treatment on the microhardness of direct composites at different depths of restoration

Efeito do tratamento térmico na microdureza de resinas compostas diretas em diferentes profundidades de restauração

Abstract

Purpose: The mechanical properties of direct composites may be improved by heat treatment. This study aimed to verify whether heat treatment influences the Knoop microhardness (KHN) of direct composites and whether there are differences among photoactivated areas before and after the application of heat.

Methods: A total of 84 rectangular cross-sectional specimens ($10 \times 2 \times 2$ mm) was prepared, 14 per composite. After photoactivation (600 mW/cm^2 - 40 s), seven specimens per composite were heat-treated ($170\,°C/10$ min), whereas the others remained unheated (control). KHN values were obtained for three sites on each specimen at specified distances from the irradiated surface (0.2, 1.0, and 2.0 mm). The data were analyzed by two-way ANOVA and Tukey's test at with a significance level set to 5%.

Results: The improvement in the KHN values depended on the composites studied. The KHN values of TE-Economic, Natural Look, and Prisma were significantly improved by heat treatment ($P < 0.05$), whereas FillMagic and Glacier remained unchanged ($P > 0.05$) after heating. The distance from the irradiated area significantly influenced the KHN of all of the composites, regardless of heating application ($P < 0.05$). Sites that were closer to the irradiated surface exhibited a higher KHN ($P < 0.05$).

Conclusion: The choice of the composite is important when intending to improve the KHN by heating. Sites that are closer to the irradiation source exhibit greater hardness, even after thermal treatment.

Key words: Dental composite; thermal treatment; hardness

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Resumo

Objetivo: Este estudo procurou verificar se o tratamento térmico influencia na microdureza Knoop (KHN) de resinas compostas, e observar se existem diferenças entre áreas fotoativadas antes e depois da aplicação do calor.

Metodologia: Oitenta e quatro amostras retangulares transversais (10×2 mm $\times 2$ mm) foram preparadas, 14 por resina composta. Após fotoativação (600 mW/cm^2 - 40), sete espécimes por compósito foram tratados termicamente ($170\,°C/10$ min), enquanto os demais permaneceram sem tratamento térmico (controle). Os dados de KHN foram obtidos em três áreas de cada amostra, de acordo com a distância a partir da superfície irradiada (mais próxima, intermediária e mais distante). Os dados foram analisados por ANOVA a dois critérios e teste de Tukey a 5%.

Resultados: O aumento da KHN foi dependente dos compósitos estudados. Os valores da KHN da TE-Econômico, Natural Look e Prisma foram significativamente melhorados por tratamento térmico ($P < 0,05$), enquanto que na FillMagic e Glacier permaneceram inalterados ($P > 0,05$) após o aquecimento. A área mais distante irradiada influenciou significativamente a KHN para todos os compósitos, independentemente da aplicação de tratamento térmico ($P < 0,05$). Quanto mais próximo da superfície irradiada, maior foi a KHN observada ($P < 0,05$).

Conclusão: A escolha do compósito é importante quando é proposta a melhora da KHN por aquecimento. Quanto mais próximo da fonte de irradiação, maior a dureza mesmo após tratamento térmico.

Palavras-chave: Resina composta; tratamento térmico; dureza

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Introduction

Since the introduction of direct composites as restorative materials, many efforts have been made to increase the longevity of dental restorations. Although a number of advances have been made, mechanical properties, such as wear and fracture resistance, are still challenged by shrinkage caused by polymerization contraction (1). In addition, adequate photoactivation of polymers is important to ensure optimum durability, as incomplete curing of the deeper portions increases the risk of marginal fracture, secondary caries, and decreased hardness. These risks are among the drawbacks that may compromise the clinical performance of dental composite, especially when large restorations are necessary (2).

To expand the range of clinical applications, the concept of indirect application of composites was developed. Indirect composites minimize the detrimental effects related to polymerization shrinkage that are observed in direct composites due to the additional laboratory treatment received (3). In the indirect technique, an initial photoactivation is applied, followed by an additional heat treatment using special ovens (4,5). The heat treatment results in an increase in the degree of conversion (1,6-9), improving the mechanical properties (5,6,9) and contributing to the relief of the stresses that originated during the polymerization and finishing procedures (10). However, the disadvantages of this technique are its high cost, as special equipment is required for the laboratory procedures (3-5), and the increase in the number of appointments.

The composites used for direct and indirect applications display similar compositions (11,12). Therefore, many authors have proposed that the association of direct composites with simple thermal treatments may produce results similar to those of the indirect system, such as improvement in material curing, thus enhancing clinical properties (7,8,11-17). An advantage would be the low cost, as the special ovens can be replaced by other heat sources, such as casting furnaces or autoclaves, that are commonly available in prosthetic laboratories and dental offices (14-16).

There is no consensus in the literature about the ideal time and temperature for the heat treatment of direct composites after photoactivation. To determine the appropriate time and temperature, several aspects must be observed, including the glass transition temperature (T_g), measured by differential scanning calorimetry (DSC), and the initial degradation temperature, calculated by thermogravimetric analysis (TGA) (15). Temperatures above T_g produce a greater filler × matrix interaction and consequently increase several properties (6,11,12,14-16), such as the diametral tensile strength (6,12), flexural strength (6,15,16), and fracture toughness (14). Nonetheless, it has rarely been investigated whether this process can result in improvements in the Knoop microhardness (KHN). In addition, it remains unknown whether heat treatment may help to improve KHN values at different distances from the photoactivator source.

The purpose of this study was to verify whether heat treatment is capable of improving the KHN values of direct composites and whether there are differences among photoactivated areas before and after heat application.

Materials and methods

The direct composites used in this study are described in Table 1.

Table 1. Descriptions of the direct resin composites studied.

Composite	Manufacturer	Characteristic	Composition		Batch #
			Organic Matrix	Filler	
FillMagic	Vigodent S.A. Ind. Com. Bonsucesso, RJ, Brazil	Micro-hybrid composite	bis-GMA, bis-EMA, TEGDMA and UDMA	Barium and aluminum silicate glass and pyrogenic silicon (0.5 μm). 80% wt.	129/08
Glacier	SDI Limited Bayswater, Victoria, Australia	Condensable micro-hybrid composite	Multifunctional methacrylic ester	Strontium glass and amorphous silica (0.4-1.0 μm). 81% wt.	07/0507
Natural Look	DFL, Rio de Janeiro, RJ, Brazil	Micro-hybrid composite	bis-GMA, bis-EMA and TEGDMA	Silanized barium crystals, hydrophobic amorphous fumed silica (1.0 μm). 70% wt.	08091231
Prisma AP.H	Dentsply International Inc., The L.D. Caulk Division, Milford, USA	Hybrid composite	bis-GMA and Urethane modified	Barium, boron and alumino silicate glass, and pyrogenic silicon (1.0 μm). 50-60%wt.	979320
TE-Economic	Ivoclar Vivadent, Schaan, Liechtenstein	Hybrid composite	bis-GMA, TEGDMA and UDMA	Barium glass, aluminum and barium fluorosilicate glass, high/dispersed silicon dioxide and mutual spheroid oxides (1.0 μm). 72% wt.	16366

Specimen preparation

A stainless steel split mold was used to manufacture 14 specimens per composite (n=84) with a rectangular cross-sectional area ($10 \times 2 \times 2$ mm). The mold was positioned over a polyester film (Mylar, Labordental Ltda., São Paulo, Brazil) that was lying on a glass slab. A single mass of each composite was inserted into the mold and then covered with another polyester film. To standardize the composite thickness, a glass slide was pressed over the film, leading to the extrusion of excess material from the mold. After excess removal, the upper surface was irradiated for 40 s using an Ultraled light-curing unit in contact with the polyester film. After being removed from the mold, the specimens were subjected to finishing with a number 15 scalpel (Lamedid Comercial Ltda., Barueri, Brazil). Immediately after the specimens were prepared, they were stored in dark boxes at 37°C for 48 h.

Heat treatment

Based on the recent literature (17), 170°C was standardized as an effective temperature for the heat treatment of all of the resins studied, as it is above the observed T_g and below the initial degradation temperature. The specimens were randomly distributed, and seven specimens per composite (the experimental groups) were dry-heated (170°C) in a casting furnace (OVMAT 7, Manfredi S.p.A., Torino, Italy) for 10 min (15,16), whereas the other seven specimens per composite remained unheated (the control groups). After the heat treatment, the specimens were stored again in dark boxes at 37°C for 72 h.

Microhardness measurements

To obtain microhardness values, the specimens were embedded in acrylic resin, using PVC cylinders as a muffle in such a way to expose their lateral surface. This surface was chosen to allow the investigation of different depths of restoration, depending on the distance from the irradiated surface: 0.2, 1.0 and 2.0 mm. The KHN was evaluated using a microhardness tester (model HMV-2/2T, Shimadzu Corporation, Tokyo, Japan) at a 100 g load and a dwell time of 15 s. Three indentations, spaced 200 μm from each other, were created at each distance region from the irradiated surface, resulting in nine indentations per specimen. For each specimen surface, the mean value of the KHN was obtained from the three readings.

Statistical analysis

The KHN data were subjected to two-way ANOVA to assess the possible effects of the heat treatment on the direct composite and to verify the differences among the irradiated areas in the heated and control groups. Multiple comparisons were performed using Tukey's test. The level of significance was set at 5%.

Results

The statistical analysis indicated that the effect of heat treatment on the KHN values depends on the composite studied ($P<0.05$). The resins TE-Economic, Natural Look and Prisma AP.H displayed significant improvements in the KHN values after heat treatment ($P<0.05$), whereas FillMagic and Glacier remained unchanged ($P>0.05$, Table 2).

Table 2. The effects of heat treatment on the different composites studied.

	Heated	Control
TE-Economic	$23.2 \pm 0.15^{A,a}$	$18.8 \pm 0.28^{B,b}$
FillMagic	$23.2 \pm 0.27^{A,a}$	$22.3 \pm 0.35^{A,a}$
Glacier	$21.9 \pm 0.23^{A,a}$	$21.2 \pm 0.17^{A,a}$
Natural Look	$22.0 \pm 0.18^{B,a}$	$19.9 \pm 0.21^{B,b}$
Prisma	$18.1 \pm 0.26^{C,a}$	$16.0 \pm 0.31^{C,b}$

Different superscripted uppercase letters indicate significantly different means within each column; different superscripted lowercase letters indicate significantly different means within each row ($P < 0.05$).

Significant differences in the KHN values were observed for the irradiated surfaces in both the heated and control groups, regardless of the composite studied ($P<0.05$). The KHN values were significantly higher for closer areas than for midway and farther areas in both the heated and control groups ($P<0.05$), i.e., the closer to irradiated surface a site was located, the higher the KHN values measured in both the heated and control groups, indicating that the application of heat does not change the differences in the KHN values among the irradiated areas (Table 3).

Table 3. The differences in the KHN values among the irradiated surfaces.

	0.2 mm	1.0 mm	2.0 mm
TE-Economic	$23.2 \pm 2.1^{A,a}$	$21.1 \pm 2.1^{A,a}$	$18.6 \pm 3.1^{B,a}$
FillMagic	$24.1 \pm 1.6^{A,a}$	$22.6 \pm 1.5^{A,a}$	$21.3 \pm 1.9^{A,a}$
Glacier	$23.7 \pm 2.0^{A,a}$	$22.0 \pm 2.1^{A,a}$	$18.4 \pm 1.6^{B,a}$
Natural Look	$23.1 \pm 1.2^{A,a}$	$21.6 \pm 2.1^{A,a}$	$18.1 \pm 2.2^{B,a}$
Prisma	$20.1 \pm 2.7^{A,a}$	$16.8 \pm 2.6^{B,b}$	$14.1 \pm 3.4^{B,a}$

Different superscripted uppercase letters indicate significantly different means within each column; different superscripted lowercase letters indicate significantly different means within each row ($P < 0.05$).

Discussion

DSC is a technique that, by energy variation, assesses the temperature difference between the sample and a reference with a temperature-controlled program. Using this method, it is possible to identify critical temperatures, such as T_g, for the analyzed material[18]. T_g can successfully be used as a reference to design the ideal heat treatment for direct composites (15,18). Above T_g, secondary molecular interactions are weakened, which allows for molecular relaxation through the bending of lateral groups. As a consequence, the mechanical properties are optimized (3,6) once trapped radicals are given the opportunity to react (3), which allows for partial relief of the stress induced during the polymerization process (19).

Prior to the heat treatment, the maximum temperature for heating without damaging the composites needs to be determined to avoid weight loss. The TGA technique provides the change in weight of a specimen as a function of temperature. This technique is a precise quantitative method for determining combustion, volatilization and decomposition by analyzing weight changes (18). In this study, this test assisted in determining the maximum secure temperature to which it is possible to subject the composites without the occurrence of marked degradation.

Most conventional post-curing heat treatments use temperatures below 130°C (3,6-9,11-14). However, the determination of specific critical temperatures for each composite studied, such as T_g and the degradation temperature, was not a concern. In the present study, an adequate heat treatment methodology was already identified, as a thermal characterization had been previously conducted (17). This prior work assures us that the heat treatment was conducted using an effective temperature.

According to the current study, the effect of heat treatment on KHN values depends on the composite studied (Table 2). Compared with the control group, TE-Economic, Natural Look and Prisma AP.H displayed significant enhancement in microhardness, whereas FillMagic and Glacier exhibited no significant improvement. Differences in the polymer structure, organic matrix composition (6), and type and content of inorganic filler (14) may explain these results.

Post-curing heating has been suggested to improve the homogenization of the composite structure (18). It is possible that initially cured FillMagic and Glacier display a more homogeneous structure than initially cured TE-Economic, Natural Look and Prisma AP.H; thus, the latter group would be more prone to display increases in the degree of conversion, and consequently on microhardness, as a result of the heat treatment (6).

In addition, differences in the amount of filler loading may play a role in the results, as this parameter has been directly related to surface hardness (14,18-19). The greater the percentage of filler, the higher the homogeneity and therefore the higher the hardness. It was observed that composites with higher inorganic filler loading, such as FillMagic (80% wt.) and Glacier (81% wt.), displayed no changes in the microhardness after the heat treatment; whereas composites with less inorganic filler loading, such as TE-Economic (72% wt.), Natural Look (70% wt.) and Prisma AP.H (60% wt.), displayed significant improvements in microhardness after heat application.

Another explanation may be due to the relief of polymerization shrinkage stresses. Initially concentrated around filler particles, the polymerization shrinkage became more homogeneously distributed after heating application, reducing filler/matrix interfacial stresses (1,2). As the TE-Economic, Natural Look and Prisma AP.H composites display larger filler size (Table 1), which accounts for a larger filler/matrix interface area, the post-curing mechanism may have largely benefited the KHN values of this group rather than those of the other composites.

Differences in organic matrix, such as the type of monomers and photoinitiators (6,19) also contributes to the results. Variations among monomers from different composites result in distinct hydrophilic properties, degree of conversion and capacity to form crossed chains during polymerization (18). Little variation in the organic matrix composition can be observed, except for Glacier. This composite is presented with ester-methacrylic acid, a low-molecular-weight monomer, which likely decreased the KHN values, with or without heating. As a general rule, it is accepted that a lower mean molecular weight of the monomer is associated with a greater percentage of shrinkage. In addition, this composite also utilizes a co-initiator with a lower absorption range (below 410 nm) (19) that is not polymerized when a 600 mW/cm^2 photoactivator source is used, resulting in a reduced degree of conversion.

Significant differences in microhardness among the irradiated surfaces were observed in both the heated and control groups. Greater distances to the irradiation sources were associated with lower KHN values (Table 3). These results indicated that the application of heat does not change the differences in the KHN values among irradiated areas.

The potential for the additional conversion of different levels in heat-treated composites depends on the elapsed time between the initial light-curing and the subsequent exposure to heat application (6-8). Regarding the treatment itself, post-cure heating of a composite decreases the levels of unreacted monomer after the initial photoactivation stage. Two basic mechanisms may be involved in this phenomenon. Firstly, the residual monomers could be covalently bonded to the polymer network as a result of the heating, leading to an increase in conversion itself. Secondly, unreacted monomers could be volatilized during the heating process. It is possible that both mechanisms act simultaneously, but their relative contributions have still not been completely elucidated (8)

The number of free radicals created during the initial light-curing decreases with time. Authors affirm that heat application aiming to increase the degree of conversion must be ideally performed immediately after photo-irradiation (7,8). Elevated monomer conversion occurs up to 6 h after photoactivation. After that period, the heat treatment did not lead to an increase in the mechanical properties (7).

In the present study, however, the heat was applied 48 h after photoactivation, but the temperature for the treatment, in contrast to those of the other studies (6-9,11-14), was higher than the T_g, which may be the most relevant mechanism responsible for maintaining the improvement in the degree of conversion, and in the hardness, at different levels of composites. Because the heat treatment was conducted above T_g, it should allow for increased radical mobility, enhancing conversion (15).

dependent. The heat treatment maintained the differences in the microhardness of different photo-activated regions in different composites, and the sites closer to the irradiated surface exhibited greater microhardness, even after thermal treatment. The association of direct composites with simple heat treatments would be an alternative to the current indirect composite systems. However, further studies with standardized methodologies are needed to verify other properties of composites for this application.

Conclusions

Based on the results, it may be concluded that the effect of heat treatment in enhancing microhardness was material-

Acknowledgments

References

1. Mante F, Saleh N, Mante M. Softening patterns of post-cure heat-treated dental composites. Dent Mater 1993;9:325-31.
2. American Dental Association. Statement on posterior resin-based composites. ADA Council on Scientific Affairs; ADA Council on Dental Benefit Programs. J Am Dent Assoc 1998;129:1627-63.
3. Eldiwany M, Powers JM, George LA. Mechanical properties of direct and post-cured composites. Am J Dent 1993;6:222-4.
4. Terry DA, Touati B. Clinical considerations for aesthetic laboratory-fabricated inlay/onlay restorations: a review. Pract Proced Aesthet Dent 2001;13:51-8.
5. Cesar PF, Miranda Jr WG, Braga RR. Influence of shade and storage time on the flexural strength, flexural modulus, and hardness of composites used for indirect restorations. J Prosth Dent 2001;86:289-96.
6. Peutzfeldt A, Asmussen E. The effect of postcuring on quantity of remaining double bonds, mechanical properties, and in vitro wear of two resin composites. J Dent 2000;28:447-52.
7. Loza-Herrero MA, Rueggeberg FA, Caughman WF, Schuster GS, Lefebvre CA, Gardner FM. Effect of heating delay on conversion and strength of a post-cured resin composite. J Dent Research 1998;77:426-31.
8. Bagis YH, Rueggeberg FA. The effect of post-cure heating on residual, unreacted monomer in a commercial resin composite. Dent Mater 2000;16:244-7.
9. Lovell LG, Lu H, Elliott JE, Stansbury JW, Bowman CN. The effect of cure rate on the mechanical properties of dental resins. Dent Mater 2001;17:504-11.
10. Sideridou I, Achilias DS, Kyrikou E. Thermal expansion characteristics of light-cured dental resins and resin composites. Biomaterials 2004;25:3087-97.
11. Wendt Jr SL. The effect of heat used as a secondary cure upon the physical properties of three composite resins. II. Wear, hardness, and color stability. Quintessence Int 1987;18:351-6.
12. Wendt Jr SL. The effect of heat used as secondary cure upon the physical properties of three composite resins. I. Diametral tensile strength, compressive strength, and marginal dimensional stability. Quintessence Int 1987;18:265-71.
13. Wendt Jr SL, Leinfelder KF. The clinical evaluation of heat-treated composite resin inlays. J Am Dent Assoc 1990;120:177-81.
14. Ferracane JL, Condon JR. Post-cure heat treatments for composites: properties and fractography. Dent Mater 1992;8:290-5.
15. Miyazaki CL, Medeiros IS, Santana IL, Matos JR, Rodrigues Filho LE. Heat treatment of a direct composite resin: influence on flexural strenght. Braz Oral Res 2009;23:241-7.
16. Santana IL, Lodovici E, Matos JR, Medeiros IS, Miyazaki CL, Rodrigues Filho LE. Effect of experimental heat treatment on mechanical properties of resin composites. Braz Dent J 2009;20:205-10.
17. Santana IL, Gonçalves LM, Ribeiro JJS, Mochel Filho JR, Cabral Júnior AA. Thermal behavior of direct resin composites: glass transition temperature and initial degradation analyses. Rev Odonto Cienc 2011;26:50-5.
18. Ferracane JL. Correlation between hardness and degree of conversion during the setting reaction of unfilled dental restorative resins. Dent Mater 1985;1:11-4.
19. Viljanen EK, Skrifvars M, Vallittu PK. Dendritic copolymers and particulate filler composites for dental applications: degree of conversion and thermal properties. Dent Mater 2007;23:1420-7.