The influence of three photoactivation techniques on the sorption and solubility of different composite resins

A influência de três técnicas de fotoativação na sorção e solubilidade de diferentes resinas compostas

Marçal Seitaro MATUMOTO[a], Márcio José MENDONÇA[a], Priscilla do Monte Ribeiro BUSATO[a],
Brenda Matsunaga LAURINDO[a], Veridiana CAMILOTTI[a*]

[a]UNIOESTE – Universidade Estadual do Oeste do Paraná, Faculdade de Odontologia, Cascavel, PR, Brasil

Resumo

Introdução: Há uma crescente demanda por restaurações estéticas, sendo as resinas compostas polimerizadas o material mais utilizados nesses procedimentos. Visando uma maior resistência à solubilização, uma polimerização adequada é necessária. **Objetivo:** Avaliar a influência de três técnicas de fotoativação na sorção e na solubilidade de três resinas compostas. **Material e método:** Foram confeccionados 90 corpos-de-prova, com 8 mm de diâmetro e 1 mm de espessura, divididos em 9 grupos (n = 10) de resinas - Filtek Z350 XT, Tetric N-Ceram e IPS Empress Direct, cada uma elas polimerizada com LED Bluephase, utilizando as técnicas: Convencional, Soft-Start (SS) e Pulse Delay (PD). Posteriormente foram colocadas em dessecador com sílica gel a 98,6 F e pesadas diariamente até obtenção de massa constante (m^1). Depois foram imersos em água desionizada durante sete dias e pesados novamente (m^2). O recondicionamento da amostra de teste foi realizado utilizando o procedimento m^1, até que uma massa constante (m^3) fosse obtida. Os valores de sorção e solubilidade foram calculados e submetidos ao teste ANOVA (two-way), seguido por Tukey post hoc, p <0,05. **Resultado:** Não houve diferenças estatisticamente significantes para sorção e solubilidade nas resinas estudadas quanto as técnicas de fotoativação. Para análise de solubilidade, foram observadas diferenças estatisticamente significantes entre os materiais. Os resultados para resina compostaFiltek Z350 XT apresentaram valores de hibridação estatísticos superiores aos das outras resinas compostas avaliadas. **Conclusão:** As diferentes formas de fotoativação não influenciaram a sorção e solubilidade das resinas compostas testadas.

Descritores: Resina composta; polimerização leve; sorção de água; solubilidade.

Abstract

Introduction: There is a growing demand for aesthetic restorations and the polymerized composite resins are the most used material in these procedures. In order to achieve greater resistance to solubilization proper polymerization is required. **Objective:** Evaluate the influence of three photoactivation techniques, on sorption and solubility of three composite resins. **Materials and Method:** 90 test samples measuring 8 mm diameter and 1 mm thick were made and divided into 9 groups (n=10) of resins -Filtek Z350 XT, Tetric N-Ceram and IPS Empress Direct, each of them was polymerized with LED Bluephase, using the techniques: Conventional, Soft-Start (SS) and Pulse Delay (PD). Afterwards they were placed in desiccator with silica gel at 98.6 F and weighed daily until obtaining a constant mass (m^1). Then they were immersed in deionized water for seven days and weighed again (m^2). The test sample reconditioning was performed using the m^1 procedure, until a constant mass (m^3) was obtained. The sorption and solubility values were calculated and subjected to ANOVA test (two-way), followed by Tukey post hoc, p<0.05. **Result:** There were no statistically significant differences in sorption and solubility among the studied resins when photoactivation techniques were compared. For solubility analysis, statistically significant differences were observed among the materials. The results for composite resin Filtek Z350 XT presented higher statistical hybridization values than those of the other evaluated resins. **Conclusion:** The different forms of photoactivation had no influence on the sorption and solubility of the tested composite resins.

Descriptors: Composite resin; light polymerization; water sorption; solubility.

INTRODUCTION

Nowadays there is increasing demand for esthetic restorations, and light polymerized composite resins are the materials most used in these procedures[1-3]. Resin composites are usually composed of an organic matrix and filler particles[3]. In the organic matrix, bisphenol A glycol dimethacrylate (bis-GMA) and triethylene glycol dimethacrylate (TEGDMA) are the combination of monomers most frequently used. The presence of the OH groups in the Bis-GMA molecule is responsible for the characteristic of water absorption and solubility of resin composites. Water absorption by hydroxyl produces plasticization of the polymer, and consequent reduction in the chemical and mechanical properties, in addition to susceptibility to staining[4]. The incorporation of filler particles into the organic matrix of resin composites promotes an increase in physical and mechanical properties. They may consist of quartz, colloidal silicon, barium, and strontium or lithium glass.

The polymerization process occurs in the organic matrix, where monomer-polymer conversion occurs through an activation mechanism. However, an insufficient degree of conversion directly affects the physical properties and chemical stability of material. Thus, when water penetrates into the organic matrix and expands the spaces among the cross links of the polymers, it causes changes in the polymer mass and dimensions. The residual monomers are solubilized by the solvent solution and may be displaced from the polymer mass. Thus, both the processes of sorption and solubility are caused by the diffusion of water into the polymer. Monomers trapped in the region of micropores, and those retained in microgelshave been reported to be more susceptible to leaching. Thus, the displacement of monomers may be affected not only by the quantity of residual monomers, but also by the distribution of monomers inside the polymer formed[5-7].

To make a polymer more resistant to solubilization, appropriate polymerization is required. Polymerization is the process of monomer conversion into polymers causing the change from the viscous to solid state. In photoactivated composite resins, this process starts when sensitization by light occurs, of a light-polymerizing molecule present in the resinous mass. Most of the time, when this molecule -an alpha-diketone (camphorquinone) -is exposed to light at a wavelength between 450 and 500 nm (blue light), it absorbs these photons, entering into a state of excitation, known as the triplet state. This condition, in turn, results in a reaction of camphorquinone with tertiary amine molecules (co-initiator-DMAEMA that produces free radicals. Through these radicals, the cleavage of carbon double bonds of monomers occurs, initiating a polymerase chain reaction. This process creates volumetric shrinkage of composite resin, and it will be at a rate as high as that of the monomer conversion, and therefore, result in the formation of these covalent bonds[8].

In order to reduce the stresses caused during polymerization shrinkage of composite resins, some authors have indicated some photoactivation techniques, such as *soft-star*[9,10] and Pulse Delay[10]. The first type consists of an initial photoactivation atreduced intensity for short period of time, followed by final photoactivationat high intensity. The Pulse Delay technique consists of initial photoactivation at reduced intensity, also for a short period of time, followed by a waiting period, and completing photoactivation at high light intensity[10,11].

Therefore, the aim of this study was to evaluate the influence of three photoactivation techniques (Conventional (CONV), *Soft-Start (SS)* and Pulse Delay (PD)on the sorption and solubility of three different composite resins.

METHODOLOGY

For this research, three light-polymerized composite resins were selected:Filtek Z350 XT (3M ESPE, Saint Paul, Minnesota, USA), (2) Tetric N-Ceram (Ivoclar Vivadent, Schaan, Liechtenstein) and (3) IPS Empress Direct (Ivoclar Vivadent, Schaan, Liechtenstein), all in enamel-shade A1 (Table 1). Each composite resinwas photoactivated with three techniques: Conventional – CONV (1200 mW/cm² for 40 seconds) *Soft-Start* – SS (until 650 mW/cm² for 5 seconds + 35 seconds 1200 mW/cm²) and Pulse Delay – PD (650 mW/cm² for 5 seconds + 5 minutes of waiting + 1200 mW/cm² for 30 seconds). For this purpose, 90 test samples were made by using a metal matrix measuring 1 mm thick and 8 mm in diameter. The composite resins were inserted and accommodated in the matrix in a single increment with the use of a Thompson spatula No.2. A polyester strip and a glass plate were placed on the composite

Table 1. Description of the composite resins used in the research

Material	Producer	Lot	Organic Matrix	Charged particles	Filler /volume
Filtek Z350 XT	3M ESPE, St Paul, MN, USA	11295	BisGMA, TEGDMA, BisEMA	Zirconia/silica with filler particle sizes between 5-20 nm Agglomerated particle varies between 0.6 and 1.4 µm	55.5%
Tetric N-Ceram	Ivoclar Vivadent, Schaan, Liechtenstein	S12951	BisGMA, TEGDMA, BisEMA	Barium glass, Ytterbium Trifluoride, MOX and copolymers With filler particle sizes between 0.04 to 3 µm	56%
IPS Empress Direct	Ivoclar Vivadent, Schaan, Liechtenstein	S17100	Dimethacrylate Cycloaliphatic Bisphenol-A Dimethacrylate propoxylate	Barium glass, Ytterbium Trifluoride, MOX, silicon dioxide and copolymers With filler particle sizes between 40 nm and 3000 nm (3µm) (average 550 nm)	52-59%

Abbreviations: BisGMA: Bisphenol A glycidylmethacrylate: BisEMA: Bisphenol A polyethyleneglycoldietherdimethacrylate; TEGDMA: Triethyleneglycoldimethacrylate. According to producer's technical profile, Giannini et al.[12]

resin, exerting slight digital pressure for 10 seconds and then the glass plate was removed. Photoactivation was performed according to each technique used. Bluephase (Ivoclar Vivadent AG. Schaan, Liechtenstein) light equipment was used, and the power density was verified at the beginning of photoactivation with the help of a radiometer (Ecel RD-7, Ribeirão Preto, SP, Brazil).

On conclusion of photoactivation, the polyester strip was removed and the test sample bases were marked with a scalpel blade No.15. After this, they were stored individually and divided into nine groups of ten samples, according to the photoactivation technique and composite resin.

Sorption and Solubility

The test samples were placed in a desiccator containing dehydrated blue silica gel (Quimidrol Comércio e Indústria Importação Ltda, Joinville, SC, Brazil) and stored in an oven at 98.6 F for 24 hours. After this, they were removed and weighed on an analytical balance accurate to±0.0001g (Marte – AW-220 – Santa Rita do Sapucai, Minas Gerais, Brazil).This cycle was repeated daily until the loss of disk mass was not greater than 0.0001g in a period of 24 hours, thereby obtaining a constant mass m_1. Before the test samples were immersed in the storage solutions, the volume was calculated for each sample. The test samples were measured at: the diameter in two equidistant points; the height in a central point; and in four equidistant points, with a digital caliper accurate to 0.01 mm (Starret, mod. 727 - Itu, São Paulo, Brazil). The volume was calculated according to the following equation:

$$V = (\pi \times r^2) \times t \tag{1}$$

Wherein:
V is the test sample volume (mm³)
$\pi = 3.1416$
r is the radius of each test sample (mm)
t is the thickness of each test sample (mm)

The test samples were immersed in 5 mL of deionized water and placed in an oven at 37°C for 7 days. After this storage period, they were removed, dried with paper towel and then air jets were used for 15 seconds. They were weighed again to obtain m_2.

The test samples were placed in a desiccator with silica gel, in an oven at 37°C for 24 hours to recondition the samples; until a constant mass named $m_3$was obtained, by performing the same procedure as that used in m_1.

The sorption and solubility properties in water were calculated according to the formula proposed by ISO 4049 -2009[13].

For the sorption test, values for W_{sp} were calculated, in micrograms per cubic millimeter, by applying this equation:

$$W_{sp} = \frac{m_2 - m_3}{V} \tag{2}$$

Where:

m_2 is a test sample mass, in micrograms, after being immersed in water for seven days.

m_3 is a test sample reconditioned mass in micrograms, after immersion and desiccation.

V is the test sample volume (mm³)

For the solubility test, W_{sl} values were calculated, in micrograms per cubic millimeter, by applying this equation:

$$W_{sp} = \frac{m_1 - m_3}{V} \tag{3}$$

Where:

m_1 is a test sample mass, in micrograms, before immersion for seven days.

m_3 is a test sample reconditioned mass in micrograms, after immersion and desiccation.

V is the test sample volume (mm³)

Statistical Analysis

Mean values obtained for each test sample of the experimental groups were initially subjected to the Shapiro-Wilk adhesion normality curve test. Positive result. Thus these values were subjected to two-way ANOVA test, followed by the Tukey*post hoc* test, p<0.05.

RESULT

It was possible to observe that there were no statistically significant differences between photoactivation techniques. For the analysis between the materials, composite resin Filtek Z350 XT presented statistically higher sorption (Table 2 and 3) values in comparison with the other composite resins evaluated, for all three polymerization techniques. The composite resins Tetric N-Ceram and IPS Empress Direct presented statistically similar behavior.

No statistical significant differences were observed among groups, when comparing the photoactivation techniques, and between the three composite resins.

DISCUSSION

Suitable polymerization is very important to the longevity of composite resin restorations, because these materials require sufficient light intensity and wavelength to activate the photoinitiator that will react with a reducing agent for the formation of free radicals, and to begin the polymerization process[1]. Within the same energy, the increase in power density makes the degree of photoactivation diminish linearly, causing an increase in strength and flexural modulus[14]. According to the producer, the Bluephase curing equipment used in this study has a maximum power density of 1200mW/cm² and wavelength between 385nm and 515nm; which lies within the absorption spectrum of camphorquinone, the photo activator present in the composite resins used.

The initial high power density increases the polymerization shrinkage stress of composite resins. To prevent this from happening, two techniques are indicated: the *Soft-start* photoactivation technique, and Pulse Delay technique[9,10]. The interval between two pulses

Table 2. Sorption statistical analysis

Resin/Polymerization	CONV		SS		Pulse Delay	
Filtek Z350 XT	21.56	(±2.18)Aa	19.90	(±1.57)Aa	18.77	(±1.68)Aa
Tetric N-Ceram	15.68	(±2.77)Ba	15.22	(±1.85)Ba	12.20	(±2.56)Ba
IPS Empress Direct	13.11	(±1.98)Ba	12.51	(±2.46)Ba	11.74	(±2.18)Ba

Different lower case letters on the same line mean statistically significant differences (p<0.05); Different capital letters on the same column mean statistically significant differences (p<0.05).

Table 3. Solubility statistical analysis

Resin/Polymerization	CONV		SS		Pulse Delay	
Filtek Z350 XT	-3.59	(±1.63)Aa	-0.84	(±2.81)Aa	-5.94	(±1.32)Aa
Tetric N-Ceram	-2.96	(±1.54)Aa	-0.02	(±1.55)Aa	-3.01	(±1.50)Aa
IPS Empress Direct	-1.56	(±1.74)Aa	1.18	(±1.77)Aa	-3.46	(±2.43)Aa

Different lower case letters on the same line mean statistically significant differences (p<0.05); Different capital letters on the same column mean statistically significant differences (p<0.05).

intensifies the composite resinpre-gel phase, and improves its fluidity, relieving stress generated by contraction of the composite[1].

In this study, no significant differences were shown for the photoactivation techniques used. Perhaps the thickness of 1mm is too thin to feel the change in curing techniques for power density devices above 800mW/cm[2,15]. Piccioni et al.[16] also found no significant differences when they evaluated the mechanical properties of different composite resins by comparing the SS technique with a Pulse Delay technique.

Lopes et al.[2], in their study, reported that the Pulse Delay photoactivation technique provided polymers with a linear structure and a lower degree of reticulation, leading to increase in volume. Therefore indicating a higher sorption effect, apart from the significant reduction in the mechanical properties of these composites subjected to solvent action. However these authors used resins without power for the test, and energy density lower than 24J/cm². In this study, the energy density values were higher than values used in other studies[1,2,17] and this may also have contributed to the absence of statistically significant differences between polymerization modes, for sorption and solubility.

N. 4049:2009 ISO[13] specifications states that in a total of five test samples for the sorption test, if four or five of the values obtained are less than or equal to 40 μg/mm³, the material is within the specification. If two or less of values obtained are less than or equal to 40 μg/mm³, the material is likely to show failures; and if three of the values are less than or equal to 40 μg/mm³, the test must be repeated. In this study, the values found for the variable sorption were much lower, demonstrating that these materials were within the standard. For the solubility analysis, if four or five of the values obtained are less than or equal to 7.5 μg/mm³, the material is within the standard. If two or less of the values obtained are less than or equal to 7.5 μg/mm³, the material is likely to show failures; and if three of the values obtained are less than or equal to 7.5 μg/mm³, tests must be repeated. Once again, the composite resins studied were in compliance with the standards.

The properties of sorption and solubility are related to solvent input into the composite resin organic matrix.This process causes swelling and lamination of the polymer; leading to the release of

residual unreacted monomers in the photoactivation process[7]. In this study, composite resin Z350 was the most sensitive to the sorption process. This may have occurred by the presence of TEGDMA in this material composition, since this molecule is hydrophilic and may result in high sorption. Similar results were found by Münchow et al.[18], when they stored composite resin Z350, which has the same organic matrix as Z350, in water for seven days.

There were no statistically significant differences for solubility, and negative values were obtained in most groups. Some studies have also found negative values[2,12]. They were obtained because m³ (after mass storage) values were greater than m¹ (mass after sample preparation). A possible explanation is that the water absorbed during the storage got stuck and included as part of the polymeric structure of the composite[12]. This might indicate that the composite was more prone to water absorption, gaining mass, which could hide the real solubility. This does not mean there was no solubility, it means that there was greater water sorption than solubility. Nevertheless, the photoactivation techniques had no influence on sorption and solubility of composite resins evaluated. However, further researches are required to evaluate the effect of these techniques on other composite resin properties.

CONCLUSION

Considering the methodology for this study, it was possible to observe that:

- The photoactivation techniques used in this study had no statistical influence on sorption and solubility of composite resins evaluated;

- There were no statistically significant differences among the three composite resins for solubility. For sorption, resin Filtek Z350 XT presented higher values compared with resins Tetric N-Ceram and IPS Empress Direct, irrespective of polymerization techniques.

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CONFLICTS OF INTERESTS

The authors declare no conflicts of interest.

*CORRESPONDING AUTHOR

Veridiana Camilotti, Rua Fagunde Varela, 2813, 85807-480 Cascavel - PR, Brasil, e-mail: vericamilotti@hotmail.com