GLASS TRANSITION AND DEGREE OF CONVERSION OF A LIGHT-CURED ORTHODONTIC COMPOSITE

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ABSTRACT

Objective: This study evaluated the glass transition temperature (Tg) and degree of conversion (DC) of a light-cured (Fill Magic) versus a chemically cured (Concise) orthodontic composite. Material and Methods: Anelastic relaxation spectroscopy was used for the first time to determine the Tg of a dental composite, while the DC was evaluated by infrared spectroscopy. The light-cured composite specimens were irradiated with a commercial LED light-curing unit using different exposure times (40, 90 and 120 s). Results: Fill Magic presented lower Tg than Concise (35-84oC versus 135oC), but reached a higher DC. Conclusions: The results of this study suggest that Fill Magic has lower Tg than Concise due to its higher organic phase content, and that when this light-cured composite is used to bond orthodontic brackets, a minimum energy density of 7.8 J/cm2 is necessary to reach adequate conversion level and obtain satisfactory adhesion.

Key words: Orthodontic composite. Glass transition temperature. Degree of conversion. Mechanical spectroscopy. Infra-red spectroscopy.

INTRODUCTION

Orthodontic cements, composites, and hybrid resin cements are used to bond orthodontic components to teeth4. Two types of orthodontic composites are available in the market: light cured and chemically cured materials. Light-cured composites are preferred by orthodontists due to their longer working time for optimal bracket placement, before the curing process is initiated by exposure to visible light. The disadvantage of these composites is that, in general, orthodontic brackets are made of materials with low visible transmission coefficient, and the composite layer is irradiated through the contour of the bracket edges1,6.

The importance of the curing efficiency with regards to the performance of composites is well established. The physical and mechanical properties of these materials are influenced by the level of monomer to polymer conversion during the polymerization process6. Light intensity and irradiation time are important factors to achieve an appropriate degree of conversion (DC)15 of photoactivated composites. Mechanical properties, such as tensile and compressive strengths, depend on the degree of the resin matrix cure1. On the other hand, the bond strength of orthodontic composites must be sufficient to withstand the masticatory forces, the stresses exerted by orthodontic mechanics, and variations in the oral environment.

An important physical property of the cured matrix is indicated by the glass transition temperature (Tg). Inadequate polymerization determines a low final Tg of a material. The Tg of an orthodontic composite is only of relevance if it lies within the range of intraoral temperatures7,11. Intraoral temperatures that exceed the Tg may result in material softening and consequently in bracket debonding14.

The purpose of this study was to evaluate the Tg and DC of a light-cured versus a chemically cured orthodontic composite using different exposure times.

MATERIAL AND METHODS

The specifications, main ingredients and manufacturers of the orthodontic composites used in this study (Fill Magic and Concise) are displayed on Table 1.

Beam-shaped specimens of each material were prepared in a brass mold with internal dimensions of 40 x 4 x 4 mm for Tg measurements. The samples were kept at room temperature and subjected to the measurements only after completely cured. Equal volumes of Concise’s Paste A
(containing the accelerator) and Paste B (containing the catalyst) were mixed according to the manufacturer’s instructions. Samples of Fill Magic composite were irradiated on the opposite faces of the lateral surface for 40, 90, and 120 s. A LED light curing unit (Gnatus - LDIII model) with a power density of 65mW/cm² was used as visible light source. The power output was measured with a power meter (Model 13PEM001; Melles Griot, Irvine, CA, USA).

Measurements of anelastic relaxation (internal friction) were performed in a torsion pendulum, operating at 1.4 and 9.4 Hz frequency range. Internal friction as a function of temperature was carried out between 100 and 380 K, with a heating rate of 1 K/min and pressure of approximately 10⁻⁶ mbar. The sample is placed to vibrate at a fixed frequency, and the sample temperature is changed so that mechanical relaxation can be observed. The technique enables measuring the elastic modulus (related to the oscillating frequency) and the internal friction (the elastic energy loss, $Q^{-1}$) as a function of the temperature. The internal friction is determined by the free decay method:

$$Q^{-1} = \tan \delta = \frac{1}{\pi N} \ln \frac{A_0}{A_N}$$  \hspace{1cm} (1)

where $N$ is the number of oscillating cycles, during which the amplitude decreases from $A_0$ to $A_N$. The peak observed on the curve $\tan \delta$ versus temperature reflects the glass transition temperature.

For the Fourier transform infra-red (FTIR) analysis, a small amount of each material was placed between two microscope slides that were compressed to produce 0.4-mm-thick films. For Concise, films of uncured pastes A and B as well as films of the cured mixture were prepared. For Fill Magic, five films of 0.4 mm of uncured were prepared for each exposure time. These five films were superposed one on the other to form a sample of 2 mm of thickness. In this way, it is believed that the reached conversion level is similar that obtained in the sample used for TG measurements. After each sample was irradiated, the first and the last films were used for DC measurements. Five samples for each exposure time were prepared.

The DC was evaluated using two absorption bands. One at 4740 cm⁻¹ associated to a combination of $=\text{CH}_2$ aliphatic bands and an aromatic band at 4623 cm⁻¹. This latter band is used as an internal standard of normalization. The DC was determined base on the decrease of 4740 cm⁻¹ absorption on the FTIR spectra as follows:

$$\text{DC} = \frac{(A_0/A_N)_{\text{uncured}} - (A_0/A_N)_{\text{cured}}}{(A_0/A_N)_{\text{uncured}}} \times 100$$  \hspace{1cm} (2)

where $A_0$ and $A_N$ are the height of aliphatic and aromatic absorption, respectively. Spectra of uncured and cured films were recorded by transmission method in a FTIR spectrometer (Nexus 670; Nicolet Ramsey, MN, USA), using 64 scans at a resolution of 4 cm⁻¹.
The statistical analysis of the DC results was performed by one-way ANOVA and Tukey multiple comparison test at \( p = 0.05 \) significance level.

RESULTS AND DISCUSSION

Tg and DC values obtained for the two composites are presented in Table 2. The DC of each sample is the average between the DC value at the surface (first film) and at the depth of 2mm (last film). The DC of Concise was similar to that obtained by Eliades, et al.\(^2\) (2000) (52.0 ± 6.0). On the other hand, the Fill Magic presents a DC statistically higher than Concise only for the exposure time of 120s. For the Fill Magic, the Tg value changed with the exposure time, providing an increase of almost 50 °C when the exposure time was increased from 40 to 120 s, while the DC increased 13.9%. Thus, the two studied properties were influenced by irradiation time suggesting the necessity of a least energy quantity for the material to reach an appropriate level of polymerization and consequently a bigger value of Tg. From the results it can be observed that the best DC and Tg values were obtained with an energy density (\( = \) power density \(\times\) exposure time) of 7.8 J/cm\(^2\).

The differences observed in the Tg and DC of the studied composites can be attributed to the structural characteristics of each material. Factors such as monomer type and inorganic filler type and content, determine the physical and mechanical properties of cured composite. Concise contains the BisGMA and TEGDMA monomers in its organic matrix and 77.4 wt% of quartz as inorganic filler. Fill magic contains BisGMA and methacrylic acid ester as monomers and 38.1 wt% of fluorsilicate glass as inorganic filler.

\( T_g \) determines the physical state of a polymer and influences several properties. Tg variation has been attributed to various molecular parameters, such as molecular weight, stiffness of the crosslinked chains and free volume entrapped in the network.\(^{19}\) Other investigations have shown that \( T_g \) can be affected by crosslinked chains and filler content.\(^{3,8,13,16}\) Figure 1 shows the loss tangent curve of Fill Magic and Concise composites. The analysis of curve behavior shows that Concise has a lower and broader loss tangent peak and a higher Tg than Fill Magic. An increase of filler content causes a broadening\(^{18}\) and a decreasing\(^{13}\) in the loss tangent peak. Therefore, the higher filler content of Concise explains the observed difference in the loss tangent curve when compared to Fill Magic. On the other hand, the different organic/inorganic phase concentration ratios between the composites explain the difference in the Tg observed for the materials. The larger inorganic phase concentration of Concise (77.4 wt%) promotes a decreased of polymeric chain mobility and consequently increases its Tg.

Figure 2 shows the loss tangent \( versus \) exposure time curve obtained with Fill Magic. A larger exposure time increased the DC (Table 2) and the crosslinked chain concentration, thus promoting an increase of \( T_g \). It is important to observe that the Tg values obtained with 40 and 90 s are within the range of oral environment temperature. The fact that Fill Magic presented a higher DC and smaller Tg than Concise suggests that, despite the greater level of conversion, its final structure might have a larger mobility and, consequently, a smaller cross-link chains density.

CONCLUSIONS

Within the limitations of this study, the following conclusions may be drawn: 1. Fill Magic presents a lower Tg than Concise due to its higher organic phase content; 2. When Fill Magic is used to bond orthodontic brackets, a minimum energy density of 7.8 J/cm\(^2\) is necessary to reach adequate conversion level and obtain satisfactory adhesion. Only with the results obtained in this work, it is impossible to predict which of the two composites will promote better bracket to tooth adhesion. Further studies, as for instance bonding strength, are necessary to be performed.

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