

Evaluation of composites light-curing at different times and distances of irradiation

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Received: 25 January 2011 / Accepted: 17 March 2011 / Published online: 1 April 2011
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Abstract The aim of this study is to evaluate light-curing composites polymerization quality (monomer/polymer) carried out at different times and distances of irradiation through the thermal analysis (TG-DTA). Samples have been polymerized at 20, 40 and 60 s (0–2–4–6–8 mm) through a constant polymerization and subsequently analysed by TG-DTA. The TG/DTA analysis shows that different light-curing times and different distance of irradiation affect the quality of polymerization; it is necessary to increase the curing time when the irradiation distance is longer than 2 mm.

Keywords Thermal analysis · TG-DTA · Polymerization · Composite

Introduction

The use of light-cured resin composites is increasing, because of the growing demand for esthetic restorative results. The curing efficiency of light-cured resin composites affects the clinical integrity of resin composite restorations. For this reason, it is important to investigate the factors that control the composite photopolymerization reaction [1].

These materials are composed of basically two phases: a resinous matrix (organic phase), comprising dimethacrylate monomers and/or oligomers, photoinitiator and an inorganic phase, known as filler, generally composed of particulate glass [2]. When the composite resins are irradiated,

the radicals generated attack the double bonds of the monomers, creating cross-linked three-dimensional network polymers [3]. The main monomers/oligomers used in the resinous matrix phase are bisphenylglycidyl dimethacrylate (BisGMA), triethylene glycol dimethacrylate (TEGDMA) and urethane dimethacrylate (UDMA). The photoinitiator system is commonly camphorquinone associated with a tertiary amine. Silicate-based glasses, zirconia, alumina, quartz and barium aluminium silicate are used as fillers which normally are surface modified by a coupling agent aiming to improve mechanical properties [4, 5]. Nevertheless, several problems concerning the lack of mechanical resistance mainly associated with restoration in posterior teeth and heterogeneity in the polymerization are frequently mentioned as drawbacks for the use of polymer restoration dental composites as repairing materials. Both problems are related to the curing process responsible for the formation of the crosslinking network that provides mechanical resistance and hardness to the final composite [6, 7].

The knowledge of polymerization mechanisms offers the possibility to check the properties of the material set in cavity. The degree of conversion (DC) of conventional dental composites depends on several factors: power and intensity of the bright source, time and distance of irradiation, dimensions of the particles of the filler, etc. [8–11]. Therefore, a lower DC value is expected to cause a premature failure of the restoration because of increasing wear, precocious staining, and marginal microleakage [12–16].

Besides, in a wet environment, as the oral cavity, the residual unpolymerized monomer pours out from the polymerized material [17–19] and it is well-known that this can cause several problems, such as toxic effects in the pulpal cells [20–24].

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Studies have demonstrated that the distance from the top of the highest cusp to the cavity floor can reach 8 mm at deep cavities [25, 27], so the light intensity reaching the deepest region can be strongly attenuated. As a consequence, lower degree of conversion is expected from resin cements when the energy is lower than that required for a proper resin cement polymerization, leading to postoperative sensitivity, staining, marginal breaking, poor adhesion between the tooth and the indirect restoration [28], microleakage, secondary caries, and changes in some cement mechanical properties. The degree of conversion depends on the energy supplied during light activation, and can be characterized as the product of light intensity and exposure time [29].

Many experimental techniques have been used to study the degree of conversion (DC) or the number of ethylene double carbon which are converted into single bonds [30–32].

The validity of the thermal analysis has been demonstrated to evaluate the dental materials and light-curing composites conversion degree [33, 34]; differential thermal analysis (DTA) and thermogravimetry analysis (TG) are thermal analysis techniques. DTA is used to gather information on transitions, heats and kinetic of reaction, and others. TG has been used to quantify the inorganic part of dental composites and the rate of mass change, respectively [35, 36].

The aim of this study is to evaluate through the thermal analysis (TG-DTA) the light-curing composites polymerization quality (monomer/polymer), carried out at different times and distances of irradiation (0–8 mm; 20–60 s).

Materials and methods

The materials used were X Duo Ceram (DENTSPLY, UK), Gradia Direct (GC, USA) (Table 1). Samples have been prepared in a 2 mm thickness and a 4 mm diameter stainless steel matrix in order to obtain an equivalent mass of about 50 mg. The light intensity was 1200 mW/cm² (Mini Led Satelec, Acteon Group, Merignac, France).

Table 1 The resin composite composition

Materials	Manufactures	Inorganic filler/wt%
Gradia Direct	GC	Methacrylate monomers 27%, Silica 38%, Prepolymerised filler 35%, Pigments, Catalysts. Inorganic filler: n.d.
X Duo Ceram	Dentsply	Methacrylate modified polysiloxane, dimethacrylate resin, fluorescence pigment, UV stabilizer, stabilizer, Camphorquinone, ethyl-4(dimethylamino)benzoate, barium-aluminium-borosilicate glass, methacrylate functionalised silicon dioxide nano filler, iron oxide pigments and titanium oxide pigments and aluminium sulfo silicate pigments according to shade. Inorganic filler: n.d.

Before proceeding with the TG/DTA analysis, a double weighing with a Gibertini electronic (mod. E42 Milano-Italy) and a TG/DTA scale is made, through which the following thermogravimetric analyses have been carried out. Eight samples of each material (number of materials = 2) for each group (number of groups = 15) have been cured (20 s 0-2-4-6-8 mm; 40 s 0-2-4-6-8 mm; 60 s 0-2-4-6-8 mm).

Moreover, five samples of each material (controls) underwent TG-DTA cycles without light-curing procedures (15 controls total analysis).

Simultaneous thermal analyzer was used to measure the mass change and heat effects (TG-DTA) on dental composites performed by TG/DTA 6300 (Model TG/DTA 6300, Seiko Instruments Inc. Torrance, CA, USA). The samples were heated at a constant rate of 10 °C min⁻¹, from 25 to 600 °C under nitrogen atmosphere (100 mL/min).

In our figures, values are represented up to 500 °C in order to remark peaks, as we did not observe other significant differences between all materials while reaching 600 °C.

In the figures we also used the curves of a single composite because all samples showed a similar behaviour.

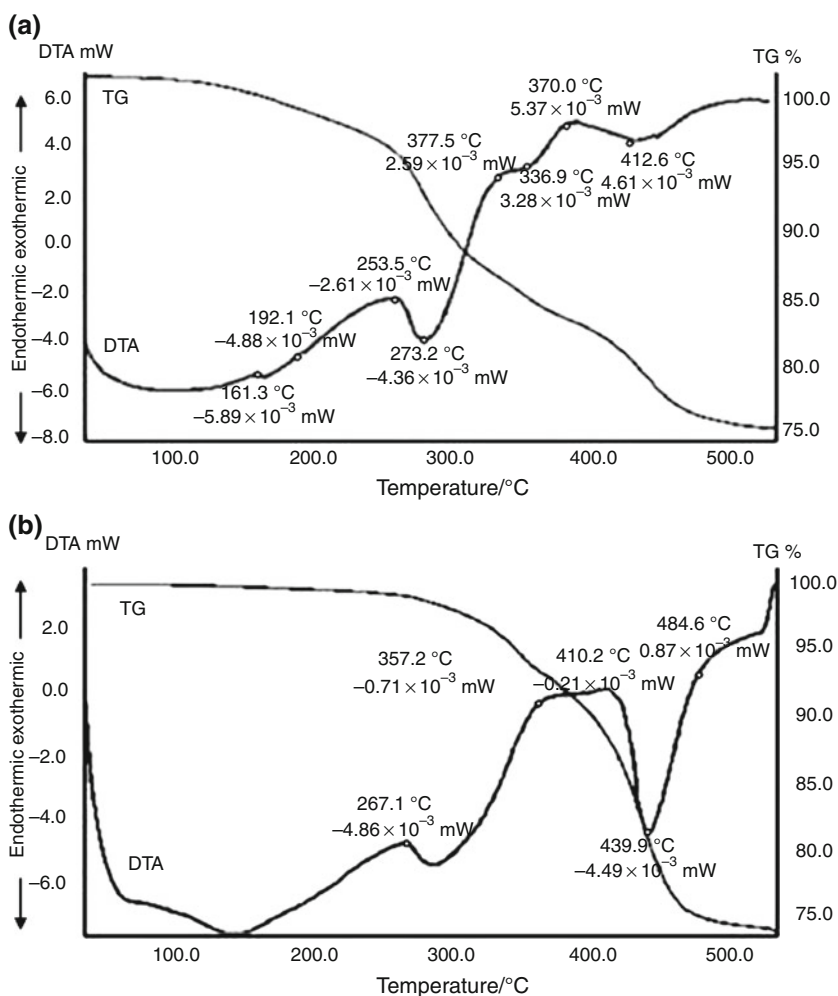
Statistical analysis

Fisher's PLSD, Scheffe and Bonferroni/Dunn were used to evaluate the presence of statistically significant differences. We did not insert the standard deviation because curves often overlapped.

Results and discussion

It is well-known that during the preparation of the light-curing composite resins, the laboratories first work the inorganic part by making it react with the silane (silanization process) and second, in the clinical phase, through the photopolymerization, the reaction of the organic and inorganic part is fulfilled [3, 19]. The final product of these chemical interactions between silane and filler is affected

Fig. 1 TG-DTA curves for uncured (a) and cured (b) composite resins



by the silanization that brings about the creation of reactive molecules on the filler-silane interface; the subsequent photopolymerization of the matrix with the silane-filler reactive molecules gives a more stable final product that improves the composite physical capacity. The TG/DTA analysis allows to highlight the points where the resinous parts (silane and matrix) deabsorb [10, 37, 38].

Figure 1 shows the representatives TG-DTA curves obtained by heating uncured and cured composite resins. Non-cured composite (a) showed a mass loss of 5.7% at 270 °C and 14.9% at 350 °C, whereas cured composite (b) showed a mass loss of 0.4% at 270 °C and 7.4% at 350 °C. At about 270 °C, mass loss is most probably caused by the weak bonds (hydrogen bonds or Van der Waals strengths) break-up between monomer and silane, and by the loss of the same molecules that have not reacted [39, 40]. At about 350 °C, we can also observe another mass loss due to the break-up of the strong (covalent) silane-silane, silane-filler and silane-resin bonds. Thermal stability and dental composite resins degradation were monitored by measuring their TG curves.

Table 2 gathers properties of the composites taken from the TG curves: 5% mass loss temperature (thermal stability) [36], and residues at 600 °C (filler content after burning the polymeric matrix). Mass loss percentage

Table 2 Thermogravimetric (TG) results (%)

Time	Temperature of 5% mass loss/ ± 2 °C					Mass loss at 600 °C/%
	Distance of irradiation					
	0 mm	2 mm	4 mm	6 mm	8 mm	
X DUO Ceram Dentsply						
20 s	389	387	382	380	376	19.45 ± 0.23
40 s	398	392	390	390	384	
60 s	399	392	391	391	389	
Gradia Direct						
20 s	328	317	314	314	312	49.62 ± 0.57
40 s	330	329	327	322	321	
60 s	331	330	329	328	328	

differences were also due to different materials compositions in organic/inorganic components ratio.

These results have to be attributed to the visible light-cure that creates the cross-linked network polymers and due to the interaction between the polymer chains and inorganic particles. Cross-linked network consequently prevents the dental composite resins from thermal decomposition by enhancing their thermal stability.

Figure 2a shows TG curves samples cured in 20 s at 0, 8 mm; it is possible to observe a higher thermal stability of samples cured at shorter distances; however, thermal stability differences between shorter and longer light-cured distances samples decreased if light-curing time was enhanced (Fig. 2b, c).

So the importance of a correct curing time in clinical procedure needs to be evident in order to avoid the release of the monomer; to improve the physical–chemical characteristics of the material and, so to make the restoration more lasting.

It's important to emphasize that in this study a quality comparison between the materials has not been made.

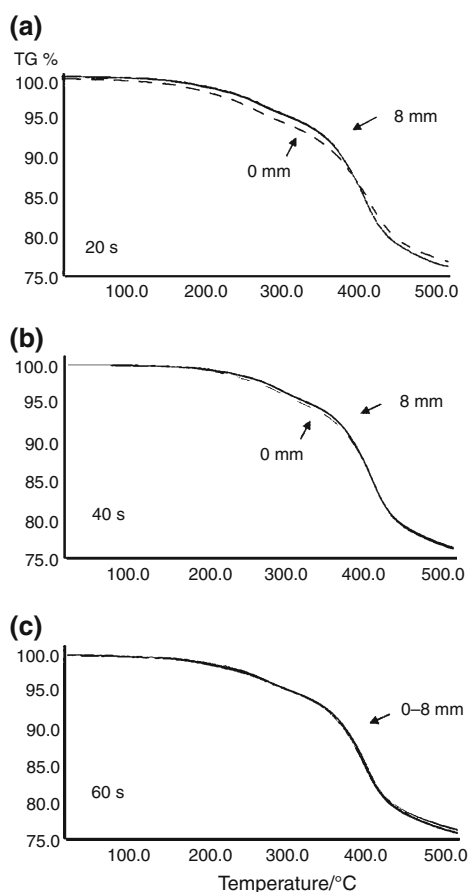


Fig. 2 Comparative TG curves for samples cured in 20 s at 0 and 8 mm (a), 40 s at 0 and 8 mm (b), 60 s at 0 and 8 mm (c)

Conclusions

The thermal behaviour of all samples was investigated using simultaneous TG/DTA methods. The TG/DTA analysis shows different thermal behaviour between samples. Time and distance of irradiation affects the quality of polymerization; it needs to increase the curing time when the irradiation distance is longer than 2 mm.

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