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INFLUENCE OF IRRADIATION EXPOSURE TIME ON THE DEPTH CURE OF RESTORATIVE RESIN COMPOSITE

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ABSTRACT. A study was conducted to evaluate the degree of conversion by the hardness measurements of a commercial resin composite. The specimens were prepared according to ISO 4049 and photo-activated for 20s – 40s – 60s with a light-emitting diodes (LEDs). To establish the optimal increment technique mono-layers 1 mm and 2 mm thick were tested. The ratio bottom-to-top was assessed for the mono-layers groups. Vickers hardness profiles were measured for mono-layer, bi-layer and tri-layer along the cross-section. The microhardness map showed difference in the mechanical characteristic of overlying resin confirmed by SEM images analysis of the fracture mechanics. Curing effectiveness of resin composite is not only dependent on the curing light unit but also from thickness of the resin composite and the duration of the exposure. The data suggest that an exposure time of 40 s or higher is required to provide composites with a homogeneous and high hardness, moreover, a 1 mm buildup multi-layering technique results in adequate curing of the bottom layer and better mechanical properties.

1. Introduction

Aesthetic composite resin restorative materials, in addition to their use in anterior teeth, are mainly being used to restore shape and function in posterior teeth. Posterior composite resin restorations, however, are submitted to very large masticatory forces (Moore *et al.* 2008). Resin-based composites are constituted by a polymeric matrix based on dimethacrylate monomers, such as 2,2-bis[p-(2'-hydroxy—3'methacryloxypropoxy)phenyl]propane (bis-GMA) that is most widely used monomer in polymeric matrices of resin-based composites. Due to intermolecular hydrogen bonding, its high molecular weight and the presence of two aromatic rings in its molecule, this monomer shows a high viscosity and a low mobility. These characteristics could interfere with the degree of conversion of the polymeric matrix; for this reason low-molecular-weight monomers such as TEGDMA (triethylene glycol dimethacrylate) are added to reduce the polymeric matrix viscosity and to obtain an increase filler incorporation and degree of conversion of the composite (Filho *et al.* 2008). In addition to the resin matrix, diketone camphorquinone serves as photoinitiator in almost all commercially available composite materials. While the composition of light cure composites, including the quantity and size of the fillers, the amount and type of photoinitiators,

and resin matrix are determined by the manufacturer, the degree of final cure depends on the quality of the curing light and the duration of cure (Rueggeberg, Caughman, *et al.* 1993).The optimal degree of curing throughout the bulk of a visible light activated dental resin composite is acknowledged to be important to the clinical success of a resin composite restoration. The LED curing units produced light with a narrow spectral distribution falling within a specific wavelength with a 400-500nm range, with a peak wavelength at 460nm as this conveniently fell within the absorption range of camphorquinone (Wiggins *et al.* 2004). This meant that no filters were needed hence the risk of degradation of the filter that can cause decreased efficiency (as in QTH halogen) lights was removed. As the lights had a higher irradiation in the region of the peak absorption of CQ, it was expected that these lights would be more effective than QTH lights to provide a greater depth of cure (Mills, Jandt, and Ashworth 1999). Hardness is commonly correlated to physical properties of composite resins like mechanical strength, rigidity and resistance to intra-oral softening (Uhl, Mills, and Jandt 2003). The depth of cure and microhardness testing have been used to assess, as indirect methods, the degree of polymerization and, thus, the efficiency of light sources (Ceballos *et al.* 2009). The scraping technique, according to ISO 4049, allows to measure the depth of cure and thereby the degree of conversion of double bonds in the polymer for dental resins. To define depth of cure, based on top and bottom hardness measurements, it is a common procedure to calculate the ratio of bottom/top hardness and to give an arbitrary minimum value for this ratio in order to consider the bottom surface as adequately cured. Values of 0.80 and 0.85 have often been used (Moore *et al.* 2008). This study compared the depth of cure (DC) of a commercial resin with 1 mm and 2 mm layer thickness at different activation times; hence having established the relationship between exposure time and layer thickness, to obtain curing efficiency, a multi-layering procedure has been carried out and was characterized by micro-hardness test and SEM analysis.

2. Material and methods

A commercial light-cured resin (Quadrant Universal LC) was chosen for the tests. It is based on a Bis-GMA matrix loaded with 72% by weigth of filler such as Ba-Al-F silicate glass (0.02-2 mm) and highly dispersed silicon dioxide (0.02-0.07 mm). The depht of cure is dependent on the optical translucency of the composite. Therefore in this study it was utilize the shade A3 of resin composite with a polymerization time of 20 seconds as reported in manufacturer informations. The rectangular specimens of the composite were prepared with stainless steel molds of 25 mm of lenght, 1.0 -2.0 - 3.0 mm of thickness and 4 mm of width and irradiated with light-emitting diodes (LEDs). Micro-hardness tests were investigated by using a Future-Tech Micro-hardness Tester FM-300. We used a Vickers indenter and a compressive load of 100g. Before the tests, the specimens were polished using a sequence of 800-1200-4000 grit silicon carbide paper. Ten indentations were made through a pyramidal diamond point on each specimen from the top and the bottom of the composite. Afterward Micro-hardness tests were performed along the cross section of each sample. The Vickers hardness, HV, was calculated as average of 100 replicas for mono-layers, bi-layers and three-layers samples. In order to evaluate the morphology of the composite and to study the fracture mechanics Scanning electron microscope (JEOL 5600LV operated at 20 kV) images of the fracture surface in a bi-layer sample was analysed.

3. Results and discussion

The means and standard deviation of VHN measurements for both ticknesses layers are reported in Table 1. Regarding 1mm mono-layer the light-emitting diodes provided, with an exposure time of 40 sec, a significant increase in microhardness if compared to the standard mode of 20 sec. In fact the bottom hardness was an acceptable 89.8% of the top hardness.

Specimens are considered to be adequately cured when the B/T ratio exceeds 80%. Furthermore, a B/T ratio of 80% corresponds to 90% of the maximum conversion possible at the top surface of a composite (Bouschlicher, Rueggeberg, and Wilson 2004). However with regard to the rate bottom/top surfaces, a further increase from 40 sec to 60 sec did not result in significant microhardness improvements (David *et al.* 2007) in which it was achieved an exceed of 6.2%.

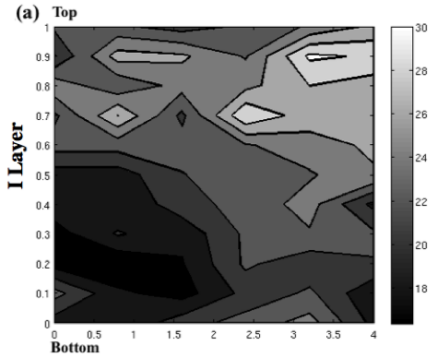
Concerning 2 mm specimen the gap between lower and upper surfaces was found to be significantly enhanced in comparison with a thinner layer (i.e. 1mm). Consequently the bottom/top ratio showed to be lower, reaching at a cure time of 40 seconds the ratio of 84.7% in comparison with 89.8% of 1mm specimen. The results of this investigation are in disagreement with some studies that have shown that 2 mm increments are well polymerized (Lindberg, Peutzfeldt, and Dijken 2005; Rueggeberg, Ergle, and Mettenburg 2000) but agree with other studies that show inefficiency of polymerization at a 2 mm depth (Fan *et al.* 2002). Confirming that those curing parameters are strictly related with the resin composite employed for dental restoration. Consequently when a minimal radiant exposure is used with increments higher than 1 mm this combination tends to yeld a lower conversion and to an undercured bulk in comparison with increments of 1 mm with a cure time longer than 20 sec. For this reason 1 mm buildup layering technique was proposed in order to optimise the mechanical performances. The efficiency was evaluated by micro hardness maps (Figure 1).

When the clinical technique of incremental packing occur the intensity of the light through the bulk of the resin composite decreased due to light absorption and scattering by restorative material attenuating its potential to cure (Ruyter and Oysaed 1982)

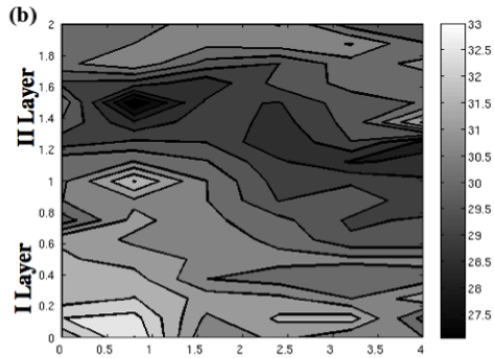
Furthermore Quadrant Universal LC is made by microparticles thus increasing light dispersion, leading to a lower degree of polymerization in the deepest layers. As a result fewer CQ molecules are being activated in these parts, resulting in potentially fewer free radicals. Therefore curing time must be increased in order for the lower number of activated CQ molecules to diffuse and successfully collide with the reducing agent to form free radicals (Rueggeberg, Caughman, *et al.* 1993).

Figure 1a shows an increment along the cross section in 1 mm sample after 20 sec of irradiation exposure. The superficial surface of Quadrant Universal LC hardens more (29.2 HV) than the bottom (17.7 HV).

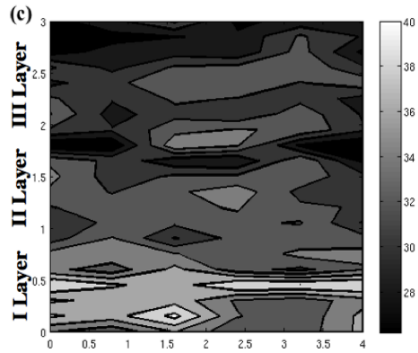
A relatively high degree of cure can be obtained on the surface of restorations, even when a low intensity curing light is used for a short exposure time (Rueggeberg, Caughman, *et al.* 1993). Hence, the influence of exposure duration cannot be assessed by top surface hardness. Consequently the method of top and bottom measurements overestimates depth of cure compared to the determination by Vickers hardness bulk profile along the cross-section.



(a) Mono-layer



(b) Bi-layer



(c) Tri-layer

FIGURE 1. Vickers hardness maps for each resin composite.

To overcome the low depth of cure with 20 s curing time of a 1mm mono-layer a multi-step layering phase was implemented. In fact 1 mm increments allow for further cross-linking of the underlying layers.

The figures 1b and 1c show the comparison between bi-layer and tri-layer maps. In the bi-layer map (Fig. 1b) the two laminae composite can be clearly distinguished. The layer I received two curing steps (20 seconds each) despite layer II that received only one curing step (Bonaccorsi *et al.* 2012). The sample exhibits an asymmetrical structure, with a very hard region with high micro-hardness values and a soft superficial region. The marked gap in the microhardness values lead to an evident difference in the mechanical characteristic of the bi-layer. On the contrary in the tri-layer map the layer I was cured 60s, while the II and III layers, 40 s and 20 s respectively. Accordingly, the gap in depth of cure between the layers cured 40-60 sec is not significant as the gap between the layers with a time of cure of 20 sec and 40 sec. This latter region, characterized by only 20s of curing time, have a micro-hardness value lower than that of the hardest layer, characterized by a 60 sec of curing time. This intrinsic anisotropy influences the stress distribution and could increase the interlaminar stresses, favoring debonding or delamination (Di Bella, Borsellino, and Calabrese 2012) of the restored tooth. Therefore a post-curing of the superficial layer is required to minimize the mechanical performances discrepancies among the laminae and to provide a homogeneous hardness. Indeed differences in microhardness measurements between layers of composite restorations can increase the flexure of the material under masticatory forces, leading to open margins or fracture of the bulk of restoration (David *et al.* 2007).

For this reason, the fracture surface of the bi-layer commercial composite was analyzed through SEM images (Fig. 2) in order to evaluate the morphology of the composite and to study the fracture mechanic.

The fracture starts in the area above and to the right of the image: from this point, it propagates along the cross section of the sample. In the area of fracture initiation, a rough surface is observed, characterized by the presence of many hanckle markings. At the top of the crack, along the direction of its propagation in the sample, there is a zone characterized by the presence of striations (scallops and rivers) typical of brittle fractures (B). Such striations take place in thermoset resins when shear stresses dominate the fracture process, they are attributed to the coalescence of many tension-induced microcracks disposed at an angle to the overall fracture plane. These lines extend approximately along the crack-propagation direction and are related with the point of arrest of the crack front (Wang and Chen 1995). Extended whitening zones (C) are also found due to the scattering of the visible light from the plan of the centres of scattering. This phenomenon is associated to the local plastic deformation of the front of the crack (Fellahi, Chikhi, and Bakar 2001). The fracture lines are regular and this indicates that there is a good adhesion between the filler and the polymer matrix. In the above reported microhardness map (Fig. 1b) an evident difference in the mechanical characteristic of two different layers was observed. By analyzing the fracture surface a slight visible transition in the morphology was identified: on the right, rivers and scallops, related to an elasto-plastic behavior, while on the left there is a smooth and shiny fracture surface typical of cleavage-like brittle fracture (Brown, White, and Sottos 2004). Furthermore, there is an outer edge (figure 2, point A), which has a an anomalous morphology, different from the adjacent areas (in such region the fracture

surface is not rough). This behavior can be related with the presence of a soft thin external layer characterized by a low cross-linking, confirming the above reported results obtained by the study of VHN maps.

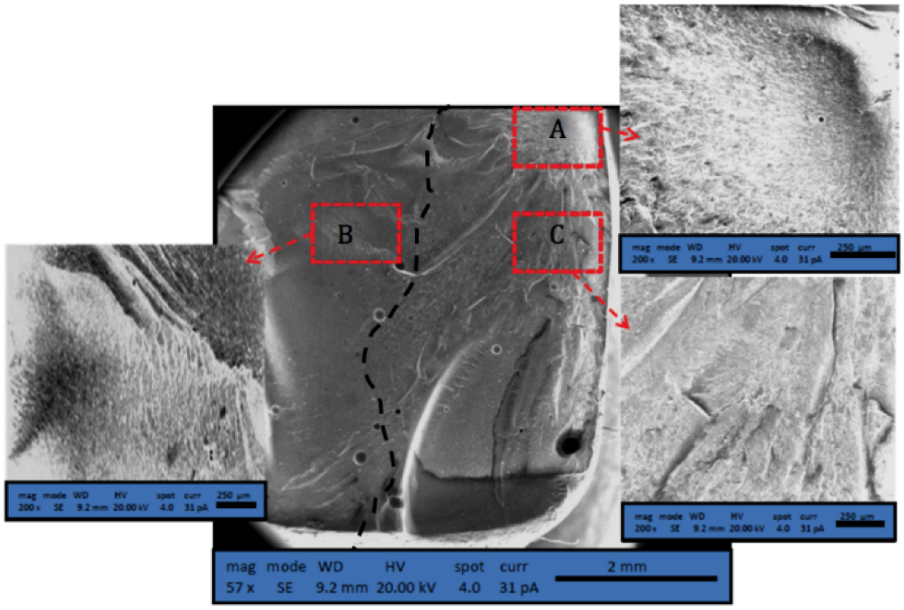


FIGURE 2. SEM micrographs of the fracture surface of the commercial composite (bi-layer).

4. Conclusions

On the basis of results achieved in this investigation it is possible to affirm that the method of top and bottom measurements overestimates depth of cure compared to the determination by Vickers hardness bulk profile along the cross-section. Furthermore, the resin-based composite tested did not achieves a 2 mm depth of cure with 20 seconds irradiation exposure. Therefore a 1 mm buildup layering technique with a post-curing phase of the superficial layer, maintaining the 20 s standard mode, was proposed. The efficiency in curing has been determined by the evaluation of VHN maps and the study of fracture surface of the specimen. The results showed much uniform hardness of multi-layer specimen that is necessary condition to increase the mechanical performances during clinical application.

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