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The effect of ultra-fast photopolymerisation of experimental composites on shrinkage stress, network formation and pulpal temperature rise

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ABSTRACT

Objectives. to complement our previous work by testing the null hypotheses that with short curing times and high DC, TPO-based resin composites would exhibit (1) higher polymerization stresses and consequently display (2) higher temperature rise and (3) higher flexural modulus, flexural strength and hardness, compared to a conventional CQ-based experimental composite.

Methods. Two experimental resin composites using either Lucirin-TPO or camphorquinone/DMAEMA as photoinitiators were prepared. Light curing was carried out using spectral outputs adapted to the absorption properties of each initiator. Different irradiation protocols were selected (0.5, 1, 3, 9 s at 500, 1000 and 2000 mW/cm^2 for Lucirin-TPO based composites and 20 or 40 s at 1000 mW/cm2 for Lucirin-TPO and camphorquinone-based composites). Degree of conversion (DC) was measured in real time by means of FT-NIR spectroscopy. Pulpal temperature rise (ΔT) was studied in a tooth model. Polymerization stress was monitored using the Bioman instrument. For cured specimens, flexural modulus and flexural strength were determined using a three point bending platform and Vickers hardness was determined with a microhardness indentor on samples prior to and after 24 h incubation in 75/25 ethanol/H_2O. Premolars were restored with both materials and microleakage at the teeth/composite interfaces following restoration was assessed.

Results. Lucirin-TPO-based composites irradiated at radiant exposures of 3 J/cm^2 and more exhibited significantly higher DCs, associated with increased flexural moduli and hardness compared to CQ-based composites. For an ultra-short irradiation time of 1 s at 1000 mW/cm^2, TPO-composites displayed similar polymerization stresses compared to CQ-controls with

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yet a 25% increase for flexural modulus and 40% increase for hardness measured after EtOH/H_2O sorption. Higher stress rates were however observed in all curing protocols compared to CQ-composites. Microleakage was similar between TPO and CQ-composites irradiated at 1000 mW/cm^2 for 3 and 20 s respectively, while a significant increase was observed for TPO-composites irradiated for 1 s. ΔT measured through a 0.6 mm thick dentin layer were all below 5.5 °C; TPO-composites exhibited similar or lower values compared to controls.

Significance. The use of Lucirin-TPO in resin composites along with appropriate curing conditions may allow for a major reduction of irradiation time while improving mechanical properties. The amount of stress observed during polymerization in TPO-based composites can be similar to those using CQ and the cohesion at the restoration-tooth interface was not affected by short curing times. Contrary to other studies, we found that the temperatures increases measured during polymerization were all well below the 5.5 °C threshold for the pulp.

1. Introduction

According to clinical studies, the major reasons for failure of resin composite restorations are secondary caries, restoration fracture and need for endodontic treatment [1–4]. From a biomimetic standpoint, the materials used should display the same characteristics as the replaced tissues. For dentin, hardness lies in the range of 50–60 HV [5], elastic modulus is around 20–25 GPa [6]. Current commercial resin composites exhibit flexural moduli and strength in the range 2.4–14.7 GPa and 45–145 MPa, respectively and Vickers hardness falls in the range of 18.6–77.7 HV/20 [7]. The wide variation of results may be explained by the different formulations but also the quality of conversion as higher degrees of conversion (DC) usually lead to more rigid [8] and harder structures [9]. Besides mechanical improvement, the materials should present a reliable interface with the dental tissues (generating minimal shrinkage stress on cure) and be safe for the pulp tissue (low monomer elution, low temperature rise in the pulp chamber, etc.). At the same time, resin composites should meet the patients and practitioners requirements, notably the reduction of chairside procedures times, for example by shortening curing time.

Despite the clear clinical interest of reducing curing time by improving polymerization kinetics, higher rates could result in greater stresses at the dentin-composite interface, which may in turn induce cusp deflection and/or gaps [10]. However the development of polymerization shrinkage stresses is a complex phenomenon, which was shown to be affected by multiple factors including not only polymerization rate, but also degree of conversion, volumetric shrinkage and elastic modulus [11]. For example, polymerization stress was shown to evolve non-linearly with conversion [12] while volumetric changes are the combination of both contraction due to the polymerization and contraction/expansion related to thermal effects [13]. Finally, an additional possible concern of faster polymerization with higher conversion and higher irradiances is the heat generated during polymerization, which could damage the underlying pulp [14,15]. Besides, reaction temperature rise (ΔT) was also positively correlated to polymerization shrinkage [16].

In a recent paper by our group, using a model experimental BisGMA/TegDMA composite formulation, we demonstrated the possibility of improving the degree of conversion (DC) and significantly reducing monomer elution despite a significant reduction of curing time (1 s for a 2 mm composite layer at 1000 mW/cm^2) [17]. This was achieved first by replacing the common ketone (camphorquinone)/amine photoinitiation system (CQ) with a Norrish Type I monoacylphosphine oxide photoinitiator, namely Lucirin-TPO (TPO), and also by choosing appropriate irradiation parameters (wavelength range, irradiance and irradiation time). Several other studies have already demonstrated that the curing light source is critical in realizing the potential of TPO and that switching from CQ to TPO, and other photoinitiators with higher molar absorptivity is advantageous for materials development [18–22]. Further, the clinicians' desire for reduced curing time (3 s, or less) has seen an influx to the dental market of high irradiance (>2000 mW/cm^2) LED light curing units that may not provide sufficient cure of some composite material types [23,24]. If such short curing times are desired, there is a requirement for the development of materials chemistry, rather than simply increasing the power of the curing light source. Overall, both intrinsic and extrinsic parameters should be taken into account in order to optimize the polymerization efficacy and resulting material properties [25].

Despite these clear improvements, it remains unclear whether the dramatic increase in reaction speed seen in TPO-based composites [19,20], associated with improved conversion [17,19,20] and mechanical properties [20], will be detrimental in terms of shrinkage stress at short curing times. Since the generation of polymerization stress throughout cure is not an intrinsic material property, its comprehensive evaluation requires the investigation of other central parameters, i.e. degree of conversion, temperature rise and elastic modulus. Hence, the aim of the present study was to complement our previous work [17] by testing the null hypotheses that with short curing times and high DC, TPO-based resin composites would exhibit (1) higher polymerization stresses and consequently display (2) higher temperature rise and (3) higher flexural modulus, flexural strength and hardness, compared to