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Evaluation of resin composite polymerization by three dimensional micro-CT imaging and nanoindentation

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ABSTRACT

Objectives. Light-cured composites undergo shrinkage during polymerization. The aim of this study was to evaluate regional shrinkage within a light-cured composite during polymerization by microcomputed tomography and mechanical properties by nanoindentation in bonded or non-bonded class-I cavity.

Methods. Zirconium oxide spherical fillers (30 μm diameter) were added as markers to a composite resin, filled into a box-shaped class I cavity with or without a bonding agent. The marker fillers were traced in 3D scans obtained by micro-CT before and after polymerization using a software (TRI/3D-BON). The average hardness of the resin composites determined by nanoindentation at each 250 μm depth was plotted against depth.

Results. In the bonded cavity, the filler particles at the top region moved toward the bottom of cavity, but at deeper depths, the direction of vertical movement changed toward the top of cavity (irradiated surface). A significant linear regression was found between filler displacement and composite depth ($R^2 = 0.9761$). In the unbounded cavity, all the fillers moved toward the light curing source, and a significant power-law regression was found between filler displacement and composite depth ($R^2 = 0.849$). In both groups, the data scattering increased at regions deeper than 3.5 mm, where the hardness, representing degree of conversion of composite, significantly decreased compared to the surface region.

Significance. The magnitude and direction of regional polymerization shrinkage depends on boundary conditions, depth and conversion degree. Polymerization shrinkage effect is most significant at the deepest part of the cavity. The application of micro-CT combined with sophisticated image analysis is a novel approach to investigate shrinkage mechanisms of dental composites.

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1. Introduction

Current dental composites consisting of inorganic fillers and a polydimethacrylate matrix are polymerized by rapid free-radical reactions activated by visible light in situ. These biomaterials are becoming increasingly popular, and preferred over metals for direct restorations [1]. However, currently available dental restorative composites undergo dimensional shrinkage when they are polymerized [2]. The volume loss during the rapid polymerization generates stresses that have been recognized as important factors affecting the marginal integrity of the composite restorations, leading to debonding from the surrounding tooth structure, and formation of gaps [3–5].

Interfacial gap and residual stresses greatly affect the longevity and success of a restoration. Therefore, it is necessary to understand the shrinkage mechanism and strategies to overcome or prevent the detrimental effects [6,7].

The polymerization reaction within a light-cured bulk composite paste varies with depth. The variation is a result of gradual loss of power density deeper inside resin, leading to a slower polymerization reaction for the deeper parts [8]. It was broadly agreed that light-curing resin composites shrink toward the direction of the light-source [9,10]. In this regard, it was suggested that the polymerization of a composite filled into a boxed-shaped (class I) cavity and light-cured from above, would start from superficial area of the composite, and the direction of shrinkage propagation was oriented toward the light source, resulting in shrinkage-generated gaps along the cavity floor [11]. However, hypotheses about contraction patterns of light-curing dental composites are still somewhat controversial.

Shrinkage vectors represent amount and direction of the composite movement during polymerization. The effects of shrinkage have been widely investigated by techniques that require specimen cutting, such as evaluating the gap formation at the cavity floor, composite mechanical properties or bond strength; meanwhile, direct measurement of shrinkage attributes of composite in a cavity has proved to be technically difficult [12,13].

Finite element methods have been utilized as numerical techniques to investigate theories suggested about shrinkage kinetics, effect of light direction and stress development in modeled cavities [14–16]. In addition, high-resolution topographic techniques have enabled nondestructive assessment of the internal structures of biomaterials. Using these techniques, monitoring polymerization shrinkage is possible if the structural features at each spatial location are precisely traceable. Microfocus X-ray computed tomography (micro-CT), is a high-resolution three-dimensional (3D) imaging technique that has been broadly accepted in research on biomedical structures, including dental composites [17–21].

Inai et al. first proposed the idea of tracing fillers in a composite material using micro-CT imaging [22]. More recently, Chiang et al. used more sophisticated analyses of micro-CT images for tracing marker fillers before and after polymerization in a composite, and elaborated on shrinkage vectors in a flowable composite [23,24].

Table 1 – The experimental composite in this study.

Composition	Weight %
Bis-GMA	10.2
TEGDMA	6.8
Silica–zirconia filler (0.4 μm diameter)	56.3
Silica–titania filler (0.1 μm diameter)	24.2
Zirconia marker fillers (30 μm diameter)	2.5
Others (initiator, stabilizer, pigment, etc.)	<1

Bis-GMA: bisphenylglycidyldimethacrylate; TEGDMA: triethylene glycol dimethacrylate.

Development of such computer-aided quantitative techniques is essential to gain a deeper understanding of polymerization shrinkage and minute details of polymerization mechanism for various dental composites. Recent advances in computer software have enabled achieving new insights into polymerization of composites using 3D micro CT imaging.

It has been reported that mechanical properties, curing rate and the boundary conditions significantly affect the polymerization shrinkage, but to date, few studies have attempted to experimentally evaluate all factors in a simulated cavity setup.

Therefore, our aim in the current study was to assess local axial polymerization shrinkage of a universal light-cured resin composite filled into a class-I cavity with or without bonding to cavity walls, in relation to nanoindentation hardness. For this purpose we developed a filler movement tracing method for 3D analysis of the micro-CT image. The null hypothesis was that boundary conditions did not affect local axial polymerization shrinkage of a light-cured composite in a class-I cavity.

2. Materials and methods

2.1. Materials

2.1.1. Experimental resin composite

An experimental universal composite was developed for this study (Tokuyama Dental, Tokyo, Japan). Radio-opaque spherical zirconia fillers 30 μm in diameter were added to the composite as marker fillers to be traced by micro-CT imaging. Composition of the experimental material is detailed in Table 1. Fig. 1 schematically illustrated the experimental procedure.

2.1.2. Cavity preparation

Two composite blocks each with a class I cavity were prepared from a hybrid resin composite (Solare, GC Corp., Tokyo, Japan). For this purpose, a box-shaped cavity (5 mm × 5 mm × 5 mm) was made in an acrylonitrile butadiene styrene (ABS) plastic resin as the model block. Then, an impression of the model block was taken using putty and regular type hydrophilic vinyl polysiloxane impression material (Exahiflex, GC Corp.). Two replicas of the model block were fabricated by incremental filling of the impression mold with Solare composite. Each layer was light-cured for 40 s, and the whole block was finally cured with a laboratory light-curing unit (α-light II, J.MORITA, Tokyo, Japan) for 5 min to ensure complete polymerization of the resin composite.