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Polymerization shrinkage kinetics and shrinkage-stress in dental resin-composites

Hanan Al Sunbul^{a,b}, Nick Silikas^{a,*}, David C. Watts^{a,c}

^a Biomaterials Science Research Group, School of Dentistry, University of Manchester, United Kingdom

^b College of Dentistry, King Saud University, Riyadh, Saudi Arabia

^c Photon Science Institute, University of Manchester, United Kingdom

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ABSTRACT

Objectives. To investigate a set of resin-composites and the effect of their composition on polymerization shrinkage strain and strain kinetics, shrinkage stress and the apparent elastic modulus.

Methods. Eighteen commercially available resin-composites were investigated. Three specimens ($n=3$) were made per material and light-cured with an LED unit (1200 mW/cm^2) for 20 s. The bonded-disk method was used to measure the shrinkage strain and Bioman shrinkage stress instrument was used to measure shrinkage stress. The shrinkage strain kinetics at 23°C was monitored for 60 min. Maximum strain and stress was evaluated at 60 min. The shrinkage strain rate was calculated using numerical differentiation.

Results. The shrinkage strain values ranged from 1.83 (0.09) % for Tetric Evoceram (TEC) to 4.68 (0.04) % for Beautifil flow plus (BFP). The shrinkage strain rate ranged from 0.11 (0.01) s^{-1} for Gaenial posterior (GA-P) to 0.59 (0.07) s^{-1} for BFP. Shrinkage stress values ranged from 3.94 (0.40) MPa for TET to 10.45 (0.41) MPa for BFP. The apparent elastic modulus ranged from 153.56 (18.7) MPa for Ever X posterior (EVX) to 277.34 (25.5) MPa for Grandio SO heavy flow (GSO).

Significance. The nature of the monomer system determines the amount of the bulk contraction that occurs during polymerization and the resultant stress. Higher values of shrinkage strain and stress were demonstrated by the investigated flowable materials. The bulk-fill materials showed comparable result when compared to the traditional resin-composites.

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1. Introduction

Polymerization shrinkage of resin-composite occurs due to the conversion of the monomer molecules in to polymer structure which is accomplished by replacing the van der Waals spaces by covalent bonds and consequently reducing the free volume [1,2]. Despite the numerous advances in improving

the bonding mechanism between the tooth structure and the resin-composite material, the failure of the bonding interface remains [2,3]. The defects that develop in the interfacial bonding are due to the polymerization stresses generated during the restorative placement procedure and later due to the functional thermal and mechanical stresses [4,5]. With the introduction of the bulk-fill group of materials the effect of

* Corresponding author at: School of Dentistry, The University of Manchester, Manchester M13 9PL, United Kingdom. Tel.: +44 1612756747.

E-mail address: nick.silikas@manchester.ac.uk (N. Silikas).

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polymerization shrinkage is highlighted as these materials are recommended to be placed in 4 mm increments.

Several studies demonstrated the direct relationship between the stress generated during polymerization and the integrity of the restoration-tooth margins [6,7]. The polymerization process is accompanied by volumetric shrinkage of the material and its magnitude depends on the composition of the material [8,9]. The amount of the polymerization stress generated is the product of many controlling factors including the C-factor, the compliance of the dental substrate and the material properties [4,10–12].

The viscoelastic behavior of the material has an important role in the capacity of the material to flow during early stages of polymerization and on the elastic modulus development. Viscoelastic behavior of the material is another important factor that affects the polymerization stress development [5]. The viscoelasticity of the material and the volumetric shrinkage are both controlled by similar factors that make it difficult to isolate their effect on the polymerization shrinkage stress development [13,14]. In addition, the stress development is affected by the reaction kinetics as a higher polymerization rate is accompanied by high polymerization stress [15,16]. The material plastic deformation is time dependent phenomenon in which the material needs time to flow to accommodate the contraction stresses before the development of the modulus of elasticity [17–20].

Different methods have been proposed to reduce polymerization shrinkage and its resultant stress. These methods include incremental placement of the restoration, soft start curing technique, the use of stress absorbing cavity liners and modification in the material composition [2,21,22]. Polymerization shrinkage for monomers such as Bis-GMA and TEGDMA had been found to be substantially higher than the typically filled composites [13,23]. There is a direct relation between the increase in filler loading and the reduction in polymerization shrinkage [24–26]. The incorporation of pre-polymerized resin fillers (organic fillers) decreases the volume fraction of the polymerizable resin and increases the filler volume fraction resulting in reduction of the polymerization shrinkage. Flowable (low-viscosity) resin-composites have been reported with higher polymerization shrinkage as a result of reduced filler loading [13,27]. Nano-composites are a class of composite that was introduced for its improved mechanical properties, esthetic outcomes and reduced polymerization shrinkage [28–31].

Several modifications have been made to the monomer systems of resin-composite materials to reduce the polymerization shrinkage. Many low shrinkage resin-composites have been commercially introduced to the market. Among these is the introduction of a high molecular based monomer with initially low double bond concentration (Dimer acid based monomer) an example of these materials is N'Durance from Septodont, France. It demonstrated lower polymerization shrinkage and higher degree of conversion when compared with conventional monomers as Bis-GMA and UDMA [32].

SDR[®] bulk-fill material has been introduced with modified UDMA by introducing photo-active groups to the monomer. It has been shown that this material resulted in lower shrinkage stress compared to other flowable and conventional composites [33]. TCD-DI-HEA was also introduced as a low shrinking

monomer and it was incorporated in Venus diamond material from Haeraus Kulzer GmbH, Germany. This low viscosity monomer has a rigid back bone that results in low shrinking behavior [22,34,35]. In addition, ethoxylated bisphenol A dimethacrylate (Bis-EMA) which is a Bis-GMA analog with lower viscosity due to the absence of hydroxyl groups from its structure was introduced for a similar purpose. It has been found to provide lower polymerization shrinkage when combined with other conventional monomers such as UDMA and Bis-GMA [36].

In this study eighteen commercially available materials from different classes were investigated for their shrinkage strain, shrinkage stain kinetics, stress and modulus of elasticity. The null hypothesis was that there are no significant differences in the results between the bulk-fill materials and the other investigated materials.

2. Materials and methods

Eighteen commercially available resin-composites with different viscosities were investigated. The materials represented a wide range of dental applications. Materials' details and manufacturers' specifications are shown in Table 1.

2.1. Shrinkage strain measurement

The 'bonded-disk' method described by Watts and Marouf in 2000 was used to determine shrinkage-strain [37]. Resin-composite paste specimens of circular disk geometry were prepared within 15 mm diameter brass rings of square cross section fixed on a 3.0 mm thick glass slab. The upper surface of the glass slab was lightly grit-blasted with alumina powder to promote bonding of the composite to the glass surface. The specimen was 1.0 mm in height and 8 mm in width. The disks were positioned centrally within the brass rings, leaving a free space around the disk. A flexible 22 mm × 22 mm size and 0.1 mm thick glass cover slip (VWR International Ltd., borosilicate glass, thickness No.0) was placed in close contact above the specimen, supported laterally by the brass ring. A glass plate was used during the preparation of the specimens, in conjunction with the cover slips, to compress the composite pastes into circular disks. Resin-composite specimens of circular disk geometry were of 0.12 g to obtain the required diameter of 8 mm. This value was obtained through pilot testing of different resin-composite weights and direct measurement of the resultant disk.

The bonded-disk arrangement is placed upon a custom jig. The jig was made of an aluminum stand with a horizontal 'stage' for specimen placement with two stainless steel clips to hold the glass slab. Mounted into the stage was a brass ring. The brass ring had a hollow center of 10 mm diameter through which the tip of a light curing unit was fixed in place. A clamp arrangement was attached to the stage to hold the uni-axial LVDT (linear variable displacement transducer) measuring system and allowed for vertical adjustment of its position. The LVDT measuring system, of 8 g active mass, was positioned centrally onto the cover slip. Specimens were light-cured by 20 s direct irradiation from beneath the glass slab at 1200 mW/cm² using an Elipar S10 LED curing light from

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