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High precision pycnometer for volumetric measurement of polymerization shrinkage in light cured dental composites

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ABSTRACT

Due to the many benefits such as excellent aesthetics, biocompatibility, and wear resistance, the use of light cured composite materials in dentistry has grown in the last few decades. However, the main disadvantage of present composite materials is significant volumetric shrinkage during curing, which leads to void and crack formation that is detrimental to the longevity of the restoration. This work presents a gas pycnometer which was re-designed to achieve high precision measurements of polymerization shrinkage while allowing in-situ curing of small size dental composites. Samples of dentin, enamel and body (DEB) shade of conventionally used restorative nanocomposite material with average mass of ~141.35 mg were in-situ cured at irradiance of ~370 mW/cm^2 using various exposure time cycles, which totaled the manufacturer recommended 60 s. The proposed system enhancements allowed measurement of time-dependent polymerization shrinkage of in-situ light cured dental composites. Volumetric measurements had reproducibility of ~0.005% of nominal full-scale sample cell chamber volume; and the shrinkage measurements did not exceed the average relative standard deviation (RSD) of ~2.8%, which is ~4 times better when compared with conventional techniques. Overall, this newly developed high precision and C-factor independent technique provides better understanding of time-dependent volumetric polymerization shrinkage aiding in development of better clinical procedures and materials to improve health and longevity of composite restorations.

1. Introduction

Composite resin and amalgam are both considered as suitable materials for direct posterior restorations [1]. Growing dissatisfaction of patients with amalgam due to aesthetic demands in molar and premolar areas has led to increased usage of light cured composites. Improved aesthetics, biocompatibility, and wear resistance are some of the benefits attributable to increased use of composite restorations in dentistry [2]. However one of the main disadvantages of present composite materials is significant volumetric shrinkage at curing, which has been linked to numerous issues such as marginal discrepancies, marginal staining, white lines around the restoration, cusp fractures, microleakage, debonding, secondary caries, postoperative sensitivity and pain [3]. Secondary caries that develop at the interface between tooth structure and composite material are responsible for 20–40% of composite restoration failures [4]. Recent advances in dental composite properties made them a more reliable restorative option. It was found that for the combined risk groups and the low-risk group, composite restorations showed better survival at 12 years [1]. However, other studies revealed failure rate twice as high for composite versus amalgam restorations in posterior teeth [5]. Thus, the clinical performance of dental composites is greatly affected by polymerization shrinkage, which has to be further studied using reliable measurement techniques to improve the quality of composite materials.

A good way to observe polymerization behavior and understand curing process of light cured composites is to measure

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time-dependent shrinkage. Methods for polymerization shrinkage measurements can be divided into two groups: one- and two-dimensional strain measurements and volumetric shrinkage measurements.

Previously reported linear strain measurement methods include a variety of techniques using LVDT transducers [6], a linometer [7], a thermomechanical analyzer [8], etc. Two-dimensional strain measurement techniques mainly include the use of strain sensors, e.g., electrical resistance strain gauges [8], and a number of optical techniques such as digital image correlation (DIC) [9], electronic speckle pattern interferometry and digital holography [10], which provide higher precision compared with DIC. Regardless of the technique, all the aforementioned methods have one common issue: strains in light cured dental composite are non-uniform [9] and highly dependent on the C-factor [8], therefore strain conversion to the volumetric shrinkage is quite complex. Boundary conditions or constraints on the specimen are a dominant factor when considering polymerization shrinkage of composites; therefore, various methods can produce significantly different values for the measured magnitude of polymerization shrinkage [8].

The main advantage of volumetric measurements is the ability to measure polymerization shrinkage directly, regardless of boundary conditions or the related C-factor. In this regard, dilatometry and gas pycnometry are conventional techniques, which were implemented for polymerization shrinkage measurements in dental composites.

Because of problems of access of a light source and opacity of mercury, it is more difficult for conventional dilatometers to be applied to light cured dental materials and at the same time to ensure that materials are cured throughout to the 'clinical' degree of conversion [7]. In order to overcome this issue, modified dilatometry systems were developed. However, such modifications have their own disadvantages. One such disadvantage is the danger of mercury vapor poisoning. Further, such systems are not suitable for volumetric measurements of dental composites with boundary constraints. Moreover, dilatometers described in [7,11] suffer from cumbersome sample installation. Conversely, the modified dilatometry system used in [8] does not have the constraint on the body of mercury, which makes it significantly different from conventionally used dilatometers [12]. Elimination of the enclosed chamber for mercury containment introduces additional experimental errors and compromises reliability of the technique.

On the other hand, the gas pycnometer works by measuring the amount of displaced gas. The pressure changes observed upon filling an empty reference chamber and then discharging it into a second chamber, which contains the sample, allow computation of the sample volume. The instrument automatically purges moisture and volatiles from the sample and then repeats the analysis until successive measurements converge upon a consistent result. The gas pycnometer used in [13] for static measurements provided simple, labor efficient, and reproducible results for dental composites shrinkage. Although the reported deviation values for the volume were consistent with the reproducibility of the pycnometer, which is 0.01% of nominal full-scale sample cell chamber volume [14]; shrinkage measurements had average relative standard deviation (RSD) of ~11%. Reported shrinkage precision was not superior compared with conventionally used techniques; therefore gas pycnometry did not receive wide usage for polymerization shrinkage measurements in dentistry.

In this study a conventional gas pycnometer was modified to allow the in-situ light curing dental composites within the sealed pycnometer chamber and to enhance precision using recommendations provided in [15]. This technique provided precise C-factor independent volumetric measurements and allowed straightforward acquisition of time-dependent photopolymerization shrinkage of studied dental composites. The authors believe that such an approach is unique and has a wide range of applications for development of novel dental composites.

2. Materials and methods

To demonstrate the technique, conventionally used restorative nanocomposite material of dentin, enamel and body (DEB) shade was chosen [16]. Evaluated samples of dental composite had elliptic cylinder shape with the average size of $10 \times 3.5 \times 2$ mm (length × width × thickness), which was in the range for class II dental restorations [17]. Dental composite samples were placed in $10 \times 4 \times 5$ mm (length × width × height) aluminum foil cups which fit into the pycnometer sample chamber, as shown in Fig. 1a.

A modified gas pycnometer with sample cups is shown in Fig. 1b. The conventional gas pycnometer was re-designed to allow the light curing of small-size dental composites inside the sample chamber and achieve higher precision, as described below. The system has a custom-made 0.36 cm^3 sample chamber with 5 mm deep insert for sample cups suitable for measurement of relatively small volumes. The proposed system monitors and takes into account the temperature effects on the volume measurements using several temperature sensors installed inside the chamber. The readings were discarded if the temperature changes of surrounding gas exceeded 1 °C during a single volume measurement. Lastly, since the sample chamber of the conventional pycnometer required opening to allow the operator to expose the composite to the curing lamp, the chamber closure may not have returned to the exact position of its previous closed position therefore causing a small volume change of the sample space. This issue is avoided in the modified pycnometer system. It has to be mentioned that the proposed enhancements apply to the pycnometry technique in general rather than for particular pycnometer system. The system described in the work is a prototype and is not for sale presently as a generally available product.

The light source was installed in the pycnometer chamber lid behind the illuminator window (see Fig. 1c), and the exposure time could be controlled directly by pressing the button on the pycnometer control panel (average reaction time was considered to be ~0.25 s). The light source used was high luminous efficacy blue (460 nm) LED emitter with 10 W radiant power and ~7 mm diameter.

Nitrogen was used as the gas displacement medium for all volumetric measurements. Standard equilibration rate of 0.005 psig/min was used for all the measurements. Since the samples studied in this work were not porous, and the instrument was calibrated previously; one purge of 19.5 psig pressure was sufficient for reproducible measurements. At least three samples for each case were tested. Mass of dental composite (~141.35 mg) was recorded and monitored during the experiments using analytical balances with reproducibility of ~0.04 mg. After each sample exposure, 10 consecutive volume measurements were performed at room temperature, and an average volume value was obtained (see Fig. 1d). Since the mass and volume of the dental composites are known, the density was calculated directly and recorded for each measurement. After initial volume measurements, samples were intermittently irradiated. Since various clinicians prefer different curing cycles and exposure times based on their personal experience [2], exposure times selected for the study were varied from 1 s to 60 s. The total exposure time for dental composites curing was 60 s, which is recommended by the manufacturer. The reproducibility of volumetric measurements of post-cured dental composites obtained using the modified system were compared with previously reported data for similar instrument [13], technical specifications [14], and those obtained using the unmodified