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Measurements for restorative dentistry: shrinkage and conversion degree of bulk-fill composites / Grassini, S.; ES SEBAR, Leila; Baldi, Andrea; Comba, A.; Angelini, E.; Berutti, E.. - ELETTRONICO. - (2022), pp. 1-6. (17th IEEE International Symposium on Medical Measurements and Applications, MeMeA 2022 UNAHOTELS Naxos Beach, ita 2022) [10.1109/MeMeA54994.2022.9856442].

Availability:

This version is available at: 11583/2972531 since: 2022-10-21T18:36:36Z

Publisher:

Institute of Electrical and Electronics Engineers Inc.

Published

DOI:10.1109/MeMeA54994.2022.9856442

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(Article begins on next page)

Measurements for restorative dentistry: shrinkage and conversion degree of bulk-fill composites

Sabrina Grassini

*Dep. of Applied Science and Technology
Politecnico di Torino
Turin, Italy
sabrina.grassinir@polito.it*

Leila Es Sebar

*Dep. of Applied Science and Technology
Politecnico di Torino
Turin, Italy
leila.essebar@polito.it*

Andrea Baldi

*Dep. of Surgical Science
Università degli Studi di Torino
Turin, Italy
andrea.baldi@unito.it*

Allegra Comba

*Dep. of Surgical Science
Università degli Studi di Torino
Turin, Italy
allegra.comba@unito.it*

Emma Angelini

*Dep. of Applied Science and Technology
Politecnico di Torino
Turin, Italy
emma.angelini@polito.it*

Elio Berutti

*Dep. of Surgical Science
Università degli Studi di Torino
Turin, Italy
elio.berutti@unito.it*

Abstract—The paper deals with a measuring approach based on Raman Spectroscopy and micro-CT imaging for correlating the degree of conversion of bulk-fill composites to the contraction shrinkage and consequently to the internal gap formation in high c-factor dental cavities. The developed study was performed on extracted molars in which a first-class cavity was prepared. A micro-CT scan was performed before and after composite light-curing to tridimensionally measure the interfacial gap between the composite material and the cavity walls. After the complete polymerization of the composite, each sample was sectioned vertically to expose the lateral surface of the restorative material. Raman Spectroscopy measurements were performed along the cross-section of the cavity filled with the restorative material, every 0.5 mm from the occlusal surface. The obtained results showed a minimal gap opening after light-curing and a degree of conversion which was not affected by the bulk-fill composite thickness. Thanks to the 3D rendering, it should be observed that gaps were mostly concentrated at the cavity floor and despite the reduction in the degree of conversion detected in the deeper portions of the restoration, a three-dimensional opening of an interfacial gap was not observed. Therefore, it is possible to assume the presence of a correlation between the degree of conversion and the volumetric interfacial gap could. Further studies are actually in progress to compare these preliminary results with those obtained on other dental composite materials.

Index Terms—Raman spectroscopy, dentistry, composites, restorative materials, interfacial gap, micro-CT imaging

I. INTRODUCTION

Modern resin composites are increasing in popularity as restorative materials, being able to guarantee, if correctly employed, excellent long-term performances both in anterior and posterior teeth. In the posterior region, several studies evaluated longevity and compared properties with other restorative materials [1]. In 2015 Beck et al. [2] published a meta-analysis with a follow-up period of 19 years, observing that the main short-term causes of failure were fractures of the restorations, secondary caries, and marginal gaps, while in the long-term assessment fracture and secondary caries were

similarly distributed. Similar conclusions were reported by Alvanforoush et al. [3] in a recently published paper.

One of the main causes of secondary caries formation is a micro-leakage consequent to the volumetric contraction which occurs in dental composite resins after light-curing. Therefore, the optimization of the curing protocols is, therefore, fundamental in order to achieve good mechanical properties of micro-filled and nano-filled composite materials for restorative dentistry. In addition, it can ensure a sufficient degree of conversion (DC) of the monomers and avoid volumetric contraction of the material, which could result in shrinkage stress at the adhesive interface with the tooth wall [4]. Shrinkage stress can cause deflection of the cusps, enamel, and dentinal cracks, post-operative sensitivity, inflammation of the pulp, and detachment of the adhesive interface. These events, in time, can lead to marginal infiltration and secondary caries, and thus, failure of the dental restoration.

Basically, proper light-curing is performed when a sufficient energy density is delivered to every composite resin layer. The depth of cure is determined by the monomers, polymerization initiators, fillers, shade/opacity, the thickness of the material, and distance of the curing light [5]–[7]. Therefore, one of the primary considerations in designing composite resin systems is the ability to relieve the amount of shrinkage stress upon light curing.

Generally, composite resins are characterized by a curing depth of about 2 mm; recently a new type of light-curing composite resins, so-called bulk-fill composite resins, has been introduced on the market. These new materials, as manufacturers declare, allow increment of the deep of polymerization till 4 mm with a high degree of conversion [8].

If bulk-fill composite resins are to provide a true clinical advantage, they must be characterized by a high depth of cure, leading, therefore, to minimize internal stress and decreasing the incidence of internal gap formation.

In this contest, several in-vitro studies focused on bulk-

fill composites have been recently published in the literature. These studies are mainly focused on the correlation between the micro-mechanical properties and the degree of conversion [9], demonstrating the advantages of these restorative materials such as adequate depth of cure, reduced cuspal deflection, and good marginal integrity. However, although bulk-fill composite resins performed well and are already widely used on patients, the properties of the whole group of bulk-fill materials still need to be further investigated. Moreover, it has to be considered that mechanical performances are particularly important in posterior cavities, so investigating the contraction shrinkage consequences in high c-factor cavities is a further challenge. As a matter of fact, class I cavities represent a complex clinical situation, due to the high c-factor (ratio of internal surface area versus external surface area) of this cavity and the consequent difficulties in managing the composite layering.

In order to evaluate the degree of conversion in restorative dentistry materials several methodologies have been employed in literature, such as differential scanning calorimetry, measurement of the polymerization shrinkage, infrared spectroscopy, and so on [10], [11].

Several studies proved that, among all the analytical approaches, Raman Spectroscopy is an effective and non-destructive technique, that can be applied directly on the surface of interest, both in-vivo and in-vitro and without any preparation of the sample [12], [13]. Raman Spectroscopy allows calculating the degree of conversion of resin-based materials on the basis of the peak height ratio change in intensity of the specific vibration modes. Indeed, the degree of conversion can be calculated as the ratio of C=C double bonds that have been converted into C-C single bonds, after light-curing. Different approaches can be employed to extract this information, namely using the ratio either between the height or the areas of the above-mentioned vibration modes. Several authors [14]–[16] proved that the employing height ratio gives the best results in terms of DC computation. Indeed, it is important to consider that Raman spectra can be complex, thus peak deconvolution could not be straightforward and lead to an increase in uncertainty.

On the other hand, another important issue that has to be evaluated is the 3D interfacial gap presence in deep class I restorations, which could immediately appear after the light-curing process due to the volumetric contraction of the composite. Modern digital imaging techniques such as 3D laser scanners, photogrammetry, and computed tomography (CT) allow the operators to achieve accurate Digital 3D reconstructions of objects [17]. Among the several advantages of these approaches, having 3D replicas of the object allows applying high effective algorithms for the effective evaluation of features and dimensions of the object itself. To this aim, micro-computed tomography (micro-CT) can be employed for interfacial analysis of dental materials and allows for identifying the critical gaps and defining internal and external margin conditions. Indeed, the micro-CT imaging technique enables high-quality 3D reconstructions with a non-destructive approach [18]. However, micro-CT images are usually ana-

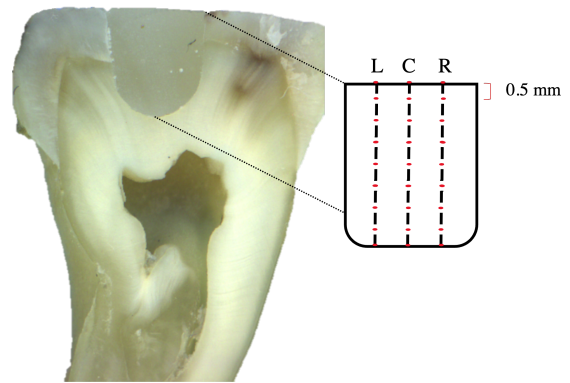


Fig. 1. Cross-section of the tooth with the class I cavity filled with the bulk-fill restorative material (3M™ Filtek™ One Bulk Fill Restorative). The scheme on the right shows the points measured by Raman spectroscopy from the external surface to the bottom, in the center, on the left, and the right of the cavity.

lyzed using linear measurements and two-dimensional reconstructions, which can lead to operator bias. Recent studies have demonstrated a non-destructive standardized 3D method for evaluating gaps. This involves quantitative measurement of the gap volume without operator bias and qualitative evaluation of the gap location through 3D rendering [19], [20]. Above all in bulk-fill materials, which are claimed to show an effective monomer conversion until 4 mm of depth, a narrow interfacial gap formation should be observed.

In this context, a measuring approach based on the combined use of Raman spectroscopy and micro-CT imaging can be an interesting solution to face the challenge of correlating the degree of conversion to the internal gap formation consequent to composite resin polymerization. This approach has been applied to a set of molar teeth and the obtained results are presented and discussed in this paper.

II. MATERIALS AND METHODS

A. Specimens preparation

All teeth selected for this study were collected with informed consent in the Department of Cariology and Operative Dentistry, University of Turin (Italy). The ethical committee of the University of Turin approved the study protocol (DS_00023_2020).

Six intact molar teeth, extracted for periodontal reasons within three months, were stored in 0.5 % chloramine solution at 4 °C, after debridement with an ultrasonic device. The selected teeth satisfy the following criteria: no carious lesions, demineralization, abrasions, or cracks under magnification (20× optical magnification) and transillumination, intact CEJ, dimension of occlusal surface wide enough to allow the preparation of one class I cavity on each tooth.

Then, teeth had their cusps flattened until a regular flat surface was reached, in order to expose middle-deep dentine, and a class I cavity preparation on the occlusal surface was performed, maintaining 360° enamel margins and following standardized parameters: (3 ± 0.1) mm mesiodistal,

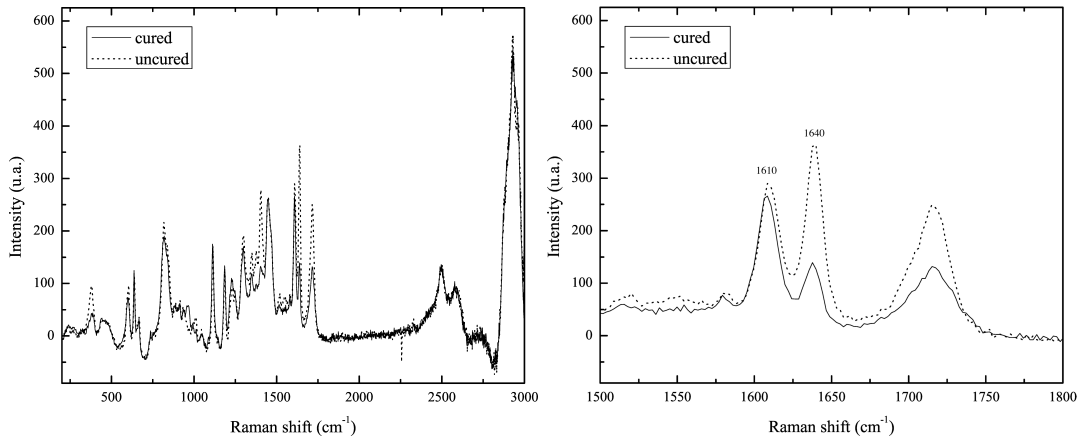


Fig. 2. Representative Raman spectra acquired on a light-cured and uncured bulk-fill composite resin. On the left it possible to observe the spectra in the range between $440\text{ cm}^{-1} - 3000\text{ cm}^{-1}$, while on the right a magnified section, in the range of $1500\text{ cm}^{-1} - 1800\text{ cm}^{-1}$ is shown.

(3 ± 0.1) mm oral-buccal, and (4 ± 0.1) mm depth. After the cavity's preparation, each linear measurement was carefully checked using a periodontal probe.

A universal adhesive was placed along the cavity walls in self-etch mode. After light-curing for 20 s, the cavity was filled with the bulk-fill composite resin (3M™ Filtek™ One Bulk Fill Restorative). The restorative material was applied following a horizontal layering technique and both the bonding and composite layering procedures were performed by an expert operator, following the manufacturer instructions: 20 s brushing primer application, 5 s dry with mild air, bonding application, gentle air-flow to make the layer uniform, light-curing for 20 s with a multi-LED curing unit (Translux 2Wave; Kulzer, Hanau, Germany) at 1400 mW/cm^2 . Specimens were also polished with fine and extra-fine diamond burs, rubber points (Twist DIA, Kuraray Noritake), and nylon brush. A second expert operator confirmed the clinical acceptability of the obtained restorations.

Specimens were then stored in distilled water for 24 h at $37\text{ }^\circ\text{C}$ in order to ensure that the chemical delayed polymerization was completed.

The class I cavities filled with the bulk-fill composite restorative material were then analyzed by Raman spectroscopy and micro-CT. Raman spectroscopy measurements were performed on tooth cross-sections. Teeth were sectioned in two parts along the vertical axis in the tooth mid-line by means of a diamond saw. Finally, all specimens were finished and polished with fine-grit diamond burs and silicon points in order to obtain a smooth surface.

B. Raman spectroscopy

Raman Spectroscopy measurements were performed by means of a BWTEK modular portable Raman spectrometer on the six molar teeth cross-sections. The instrument is equipped with a monochromatic excitation laser (wavelength: 785 nm) and a BTC675N spectrometer (range: $65\text{ cm}^{-1} - 3350\text{ cm}^{-1}$, resolution 6 cm^{-1}) coupled with a CCD sensor. In addition, the instrument was connected to the BAC151 compact Raman

microscope which allows observing the measuring area and carefully focusing the laser spot.

The following parameters were employed: laser power of 100 mW , integration time of 20 s , 24 repetitions for each area, microscopic objective of $80\times$ (analysed area of about $20\text{ }\mu\text{m}$). For each specimen, twenty-seven spectra were acquired. In particular, the points of analysis were chosen along the cross-section of the cavity filled with the restorative material, every 0.5 mm from the external surface to the bottom of the cavity; for each specimen, the Raman profile has been acquired along the center of the cavity, and on the left and right side, respectively. A representative scheme of the points of analysis is shown in Fig. 1. In addition, a specimen of the uncured bulk-fill composite resin was measured as a reference. The acquired spectra were elaborated in Python to subtract the baseline, by means of asymmetric least square smoothing.

The degree of conversion (DC) was evaluated as the ratio of the residual aliphatic and aromatic bonds obtained from Raman spectroscopy. In particular, the height ratio between the aliphatic C=C double bonds (at 1640 cm^{-1}) and the aromatic C=C double bonds (at 1610 cm^{-1}) was calculated in the specimens after curing and also on the uncured resin, to collect a reference spectrum. Then, the degree of conversion of aliphatic carbon-carbon double bond into C-C single bonds was calculated as reported in Equation 1:

$$DC\% = \left(1 - \frac{(I_{1640}/I_{1610})_{cured}}{(I_{1640}/I_{1610})_{uncured}}\right) \times 100 \quad (1)$$

where I is the intensity of the vibration peaks.

C. Micro-CT imaging

Micro-computed tomography (micro-CT) was employed to evaluate the interfacial gap.

Each tooth was scanned twice using a micro-computed tomography machine (Sky Scan 1172 Micro-CT, Bruker Billerica, MA, USA), with the following parameters: 100 kV ; $100\text{ }\mu\text{A}$; aluminium and copper (Al+Cu) filter; pixel size = $8\text{ }\mu\text{m}$; averaging = 5; rotation step = 0.5° . The first scan

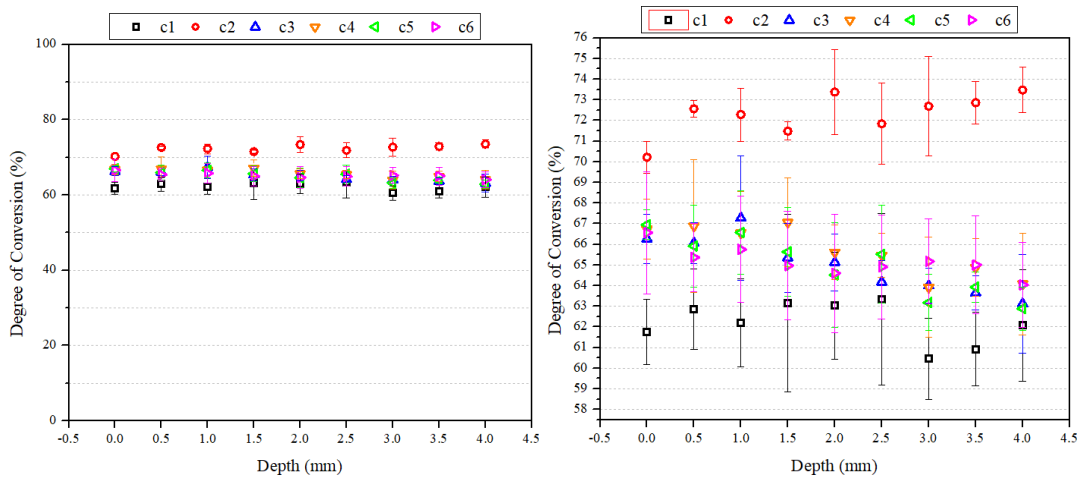


Fig. 3. Scatter plot of the Degree of Conversion (DC %) as a function of the depth (mm) of the analysed point, from the occlusal surface to the bottom of the tooth cavity: in the 0 – 100 % range (left) and a magnified section between 57.5 – 76 % (right). The graph reports the mean value of DC for each sample and the corresponding standard deviation.

was performed after placement of the bulk-fill composite resin in the prepared cavities, before light-curing the restorations. Then, the second scan was performed after the resin light-curing.

Images were reconstructed through NRecon software (Bruker, Billerica, MA, USA) in order to obtain DICOM files with standardized parameters: beam hardening correction = 20%; smoothing = 3; ring artifact reduction = 7. DICOM files were processed with a segmentation software (Mimics Medical 20.0, Materialise) in order to evaluate three-dimensional interfacial gaps and internal voids or bubbles. Finally, superimposition of the two scan images was performed by the software: registered micro-CT data of cured samples were subtracted from uncured samples data, allowing the contraction shrinkage due to the polymerization process to be measured.

III. RESULTS AND DISCUSSION

In Fig. 2 a representative Raman spectrum acquired on the cured restorative material is reported, together with a spectrum collected on the reference uncured sample. On the right side of the figure, a magnified section is shown.

The spectra evidence a clear decrease in intensity of the vibrational mode at 1640 cm^{-1} due to the light-curing. On the other hand, the peak at 1610 cm^{-1} remains stable and thus it has been used as an internal reference for the DC computation as reported in Equation 1 [21].

The degree of conversion, expressed as a percentage, obtained on the bulk-fill composite resin selected for this study is presented in Fig. 3. In particular, the graph shows the mean value of the degree of conversion (DC) for each specimen as a function of the depth of the analyzed point, from the occlusal surface to the bottom of the cavity, computed among the three vertical profiles (Fig. 1), and the corresponding standard deviation. From the results, it is possible to deduce that, in all the tested specimens, the monomer conversion

slightly decreased from the top surface along with the depth of the bulk-fill composite restoration. In particular, for all specimens the DC value is within the range of 60.46-73.48%, thus showing a good level of polymerization.

The results obtained on the composite restoration through micro-CT imaging before and after light-curing are shown in Fig. 4 as a traversal section of the 3D reconstruction. In addition, the interfacial gap before and after light-curing, expressed in mm^3 , is shown in Table I. It is possible to evidence that in all tested specimens the curing process produced an interfacial gap increase, which is strictly related to the volumetric shrinkage of the material. However, despite the reduction in the degree of conversion observed in the deeper portions of the restoration and, therefore, closer to the cavity walls, a three-dimensional opening of an interfacial gap was not detected. It is worth noticing that thanks to the 3D rendering, it was possible to evidence that gaps were mostly concentrated at the cavity floor, as shown in Fig. 5. This may align with the findings of Ausiello et al. [22], who reported a high concentration of stress in this area when applying shrinkage forces on a finite element analysis model. Other studies showed that the shrinkage stress concentrates more in the cavity floor, even with bulk-fill composites [23], [24].

Thus, bulk-fill materials showed that efficient light transmission through the composite mass led to a uniform conversion from monomer to polymer. Consequently, the volumetric contraction and the shrinkage stress are reduced, which corresponds to a reduced gap formation thanks to lower interfacial tensions [19], [25]. It is therefore evident that a polymerization process as uniform as possible along the thickness of a dental composite is a key factor in obtaining a low interfacial gap.

CONCLUSIONS

Based on the study results, it can be concluded that a correlation in evaluating the efficiency of a light-curing process by means of Raman spectroscopy and the three-dimensional

TABLE I
INTERFACIAL GAP BEFORE AND AFTER LIGHT-CURING.

| Sample | Internal gap (mm^3) before ligh-curing | Internal gap (mm^3) after ligh-curing | Gap decrease (mm^3) | Gap decrease % |
|--------|--|---|-------------------------|----------------|
| 1 | 0.1146 | 0.1237 | -0.0091 | -7.9386 |
| 2 | 0.0272 | 0.0289 | -0.0017 | -6.2500 |
| 3 | 0.0674 | 0.0701 | -0.0027 | -4.0232 |
| 4 | 0.0616 | 0.0738 | -0.0122 | -19.7176 |
| 5 | 0.0851 | 0.0984 | -0.0134 | -15.7183 |
| 6 | 0.0238 | 0.0288 | -0.0050 | -21.1013 |

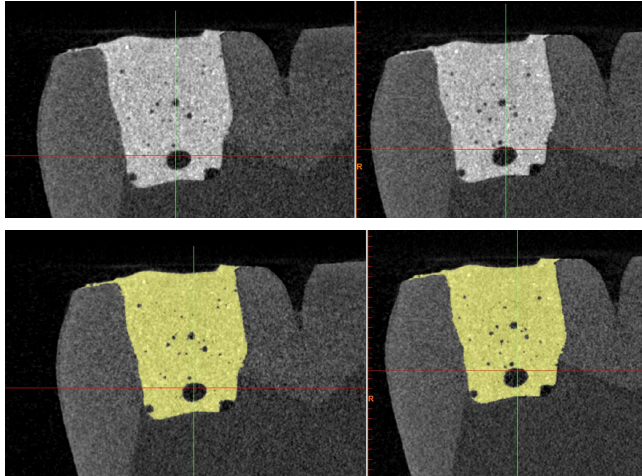


Fig. 4. Traversal section of the 3D reconstruction of the composite restoration before (top) and after (bottom) light-curing.

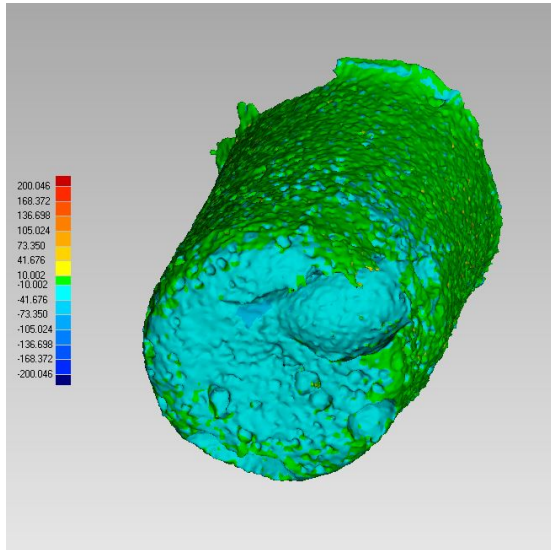


Fig. 5. Color map showing the comparison between the 3D reconstructions of the composite restoration. Color maps evidence positive (red) and negative (blue) deviations (μm) of the composite restoration after light-curing with respect to the uncured one. The shrinkage located at the cavity floor is represented by negative deviations, in light blue.

internal gap measurement due to volumetric contraction was observed. Moreover, the study allowed to prove the efficiency of the coupled use of Raman spectroscopy and micro-CT

imaging. Further measurements are currently in progress on other restorative materials in order to better validate the proposed measuring approach and assess its trustability in dental research.

ACKNOWLEDGMENT

The authors would like to thank Sara Dardanelli, Martina Faletti, and Valentina Zaffalon for their precious contribution to sample preparation.

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