

International Conference on Manufacturing Engineering and Materials, ICMEM 2016, 6-10 June 2016, Nový Smokovec, Slovakia

Polymerization mechanics of dental composites - Advantages and Disadvantages

Milos Milosevic

University of Belgrade, Innovation Centre of Faculty of Mechanical Engineering, 11120 Belgrade, Serbia

Abstract

Dental composite materials which are used as tooth fillings have numerous advantages and disadvantages in comparison to previously used alternative materials. Effects of polymerization shrinkage (light curing) of composite polymer materials which occur after illuminating with a LED lamp can cause problems in dental practice. Considered in this paper were the mechanical consequences which can occur due to light curing, along with strain fields for composites Z250, TetricEvoCeram and Silorane, as well as strain on a separate circular section that have occurred before and after light curing, by using the DIC method.

© 2016 Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

Peer-review under responsibility of the organizing committee of ICMEM 2016

Keywords: Dental composites, polymerization mechanics, shrinkage

1. Introduction

Resin based composite materials (hereinafter referred to as composites), due to their favourable physical and mechanical properties, including high resistance to compression and wear, relatively low costs and simple application, recently emerged as a substitute for amalgams. As for amalgams, despite their good properties, they have numerous disadvantages, such as: bioelectric potentials in the oral medium, potential risk of mercury poisoning, corrosion and impossibility of bonding with hard tooth tissues. Due to the aforementioned, radical cavity preparation is necessary, which involves sacrificing of a considerable amount of tooth tissue in order to ensure satisfying filling integrity, wherein a completely non-aesthetic form is obtained, which significantly reduces its overall value [1, 2].

Composites are essentially made of three basic components: resin based organic matrix, nonorganic filler particles or nonorganic dispersed phases and organic-nonorganic bonding agent, silane [3-5]. Organic matrix is made of monomers, that, due to polymerization, bond into polymers and form a three-dimensional network, which is

filled with fillers, and in this way the physical and mechanical properties of the network are improved (Figure 1). In addition to the mentioned components, composites contain smaller amounts of additional materials which contribute to the overall material quality, such as: polymerization initiators, various additives, stabilizers, inhibitors, pigments etc. Filler materials typically include glass or quartz particles, or fused glass particles. Organic-nonorganic adhesive is typically added to filler particles themselves, and the nonorganic end of the molecule bonds with it, wherein the organic end of the molecule tends to bond with the resin matrix, thus unifying the organic and nonorganic phase of the composite.

The composite is non-metallic, contains no mercury, is thermally and electrically inert, possesses the ability to directly bond with hard tooth tissues, and ensures a satisfying aesthetic appearance of a natural tooth [3, 4].

The bond between tooth and composite has always been a sensitive issue in science, and the introduction of adhesive dentistry represents a huge step towards solving the issue of restoration conservation. One of the biggest disadvantages of restorative materials is their limited lifespan upon restoration, which may result as a consequence of occurrence of light curing and related stresses [6].

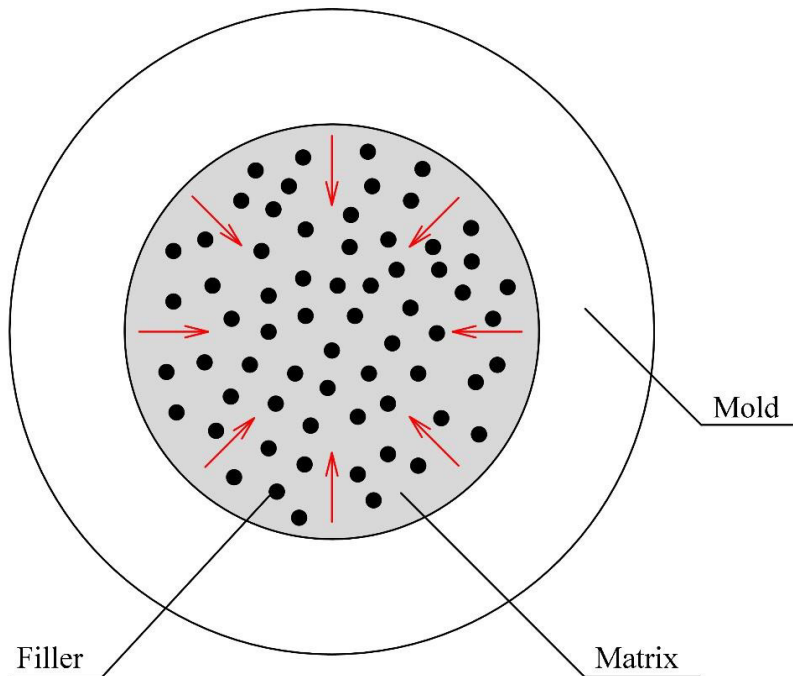


Figure 1. Light curing of a composite

2. Hardening mechanics and light curing

Displacement and spatial organisation of monomer molecules are responsible for volumetric changes during polymerization. At the beginning of the polymerization process, the resin enters the pre-gel stage, during which the organic matrix is in viscous plastic form, which allows it to “spill”, i.e. “flow”. In this stage the monomers can still move or “slip” into new positions within the organic matrix. The polymerization process continues, wherein larger molecules are formed, and the composite hardens and homogenizes into a solid body. The point at which any and all movement is no longer possible is referred to as the gel point, and denotes the transition from pre-gel to post-gel stage. Material is in a stiff elastic state, but is still contracting. This shrinkage causes stresses to occur. Gelation can be seen as the moment in which molecules within the material can no longer compensate the shrinkage. Total material shrinkage is determined by the pre-gel stage, during which the material can still be controlled and is

capable of compensating light curing [7, 8]. Post-gel stage, also known as vitrification stage, is considered responsible for the occurrence of residual stresses.

Effects of light curing are typically decreased in two ways:

1. by reducing the reaction surface per unit volume,
2. by using different types of resin.

In order to overcome issues related to light curing, experts choose between a variety of techniques, including agents use for bonding with dentine [9], low-shrinkage composite materials, glass-ionomer cement coatings [10] and various ways of inlaying [11].

It has been shown that none of the techniques mentioned above are capable of completely preventing the occurrence of shrinkage and stresses.

There are 3 basic forms of light curing:

- Free shrinkage. In case the composite material does not lie on a solid surface, it will contract towards the centre. Since the shrinkage is not prevented in any way, there will be no residual stresses.
- Effective shrinkage. If the composite material is connected to a single solid surface, newly formed boundary conditions will affect the shrinkage, and there will be barely any residual stresses, since the loss of volume will be compensated by the shrinkage opposite the bonding surface.
- Shrinkage between opposite cavity walls. Stress caused by light curing will occur if the shrinkage is impeded by the opposite walls, as a force pulling the composite from the cavity walls [12, 13]. If the stresses exceed the adhesive bond strength, the bond will fail and marginal micro-cracks will occur. Light curing of composite materials may cause shrinkage forces which could damage the bond with tooth tissue, cause marginal cracks and micro-permeability [14].

3. Consequences of occurrence of polymerization stresses and volumetric shrinkage of composites

Most of resin based composite materials, which are used in restorative dentistry have a common base of polymerizing free methacrylate radicals or creating networks during cationic polymerization with ring opening [15]. These processes change the volume of the material, which causes stresses at the restoration-tooth bond, which are known as “curing (shrinkage) stresses” [13] (Figure 2.3). Such stresses occur in the composite mass and are transferred to the adhesive bond [8, 16] and the tooth surface, which often leads to cusp damage [17] and micro-cracks in the enamel and dentine [18], and results in postoperative sensitivity in patients [19]. Polymerization stress can compromise the marginal integrity of the tooth and restoration, enable bacteria to penetrate the area between the filling and tooth [20] and finally, cause marginal discoloration, secondary caries, partial tooth damage [21], tooth pulp inflammation [22].

Loads on the adhesive composite-tooth bond and the tooth depend on cavity shape, size, C-factor, elasticity module of the tooth and composite, polymerization rate and level of conversion [23, 24]. All of the abovementioned factors are mutually related and act in a complex way to produce polymerization stresses, Figure 2.

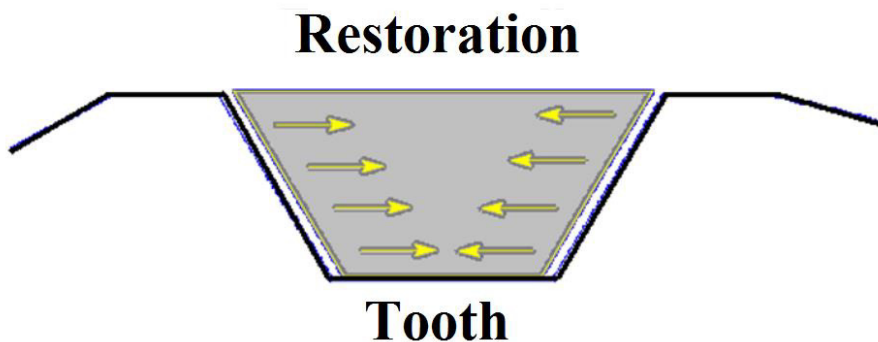


Figure 2. Stresses due to light curing

Occurrence of stress is mainly influenced by:

- cavity configuration and volume
- resin material properties
- material implanting technique
- resin material polymerization.

Modern composites cause shrinkages of 1 to 3% during polymerization, wherein even adhesive systems which create strong bonds, above 20 MPa, which are higher than shrinkage stresses (13-17) MPa, cannot always bear the load, and in this case separations and cracks begin to occur [14].

Deforming of cavity walls can partially compensate shrinkage stresses in case of composites in lateral tooth cavities, although light curing may cause cracks and complete cusp fractures [25-27].

Finite element method (FEM) represents a modern numerical method, which is applied to computer aided design and calculations of structures and elements and solving of continuum mechanics problems, and in dentistry it is not uncommon to use it for stress calculations [28-33].

In order to obtain dimensional characteristics of the composite, the method for obtaining mean values of polymerization shrinkage as a comparative characteristic is shown in this study. This method can, for a set of various materials, provide a more realistic insight into composite behaviour, thus aiding the dentist in selecting the adequate composite. In addition, examples of shrinkage stress calculations, obtained by using finite element method, are shown.

4. Materials and method

The method for three-dimensional (3D) optical strain and displacement analysis is based on digital image correlation (DIC). This technique involves digitalization of a prepared measuring surface of an image of the specimen before and after the load has been applied, i.e. displacement or strain. All changes of initial points or small surfaces in the images are compared to the previously recorded ones, using correlation functions, until an accurate similarity can be achieved. Each pixel in the image has its numerical value and by comparing these images, series of these values are obtained.

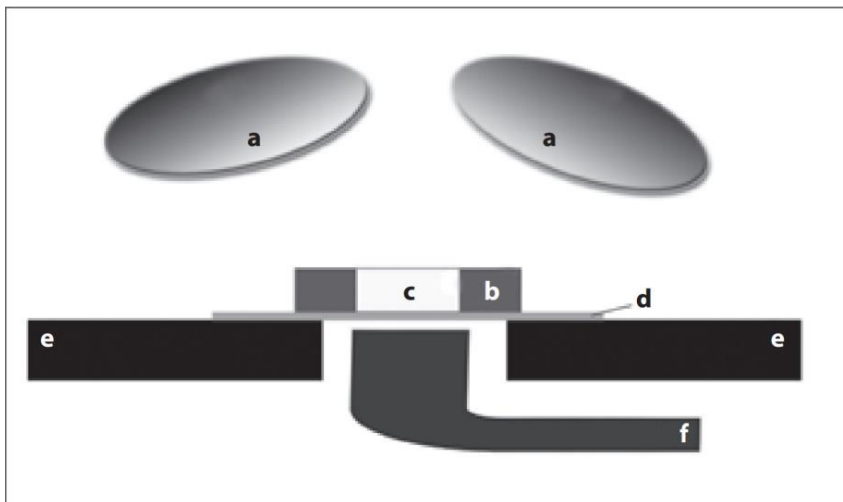


Figure 3. Scheme of the experimental setup. (a) Cameras, (b) Teflon mould, (c) sample, (d) Mylar strip, (e) sample holder, and (f) light-curing unit [116]

For the purpose of determining the mean values of polymerization shrinkage in this paper, DIC method with an experimental setup shown in Figure 3 and described in [34] was used in this paper. Materials used for determining the values of shrinkage include TetricEvoCeram (Ivoclar Vivadent), Filtek Silorane (3M, ESPE) and Filtek Z250 (3M, ESPE).

5. Results

Strain fields of the representative samples are shown for composites Z250 (4.1), TetricEvoCeram (Figure 4.2) and Silorane (Figure 4.3), before and after light curing in Teflon moulds. Blue colour in images (Figure 4.A) denotes strain fields before light curing, wherein the colour spectre in images (Figure 4.B) denote strain field after light curing.

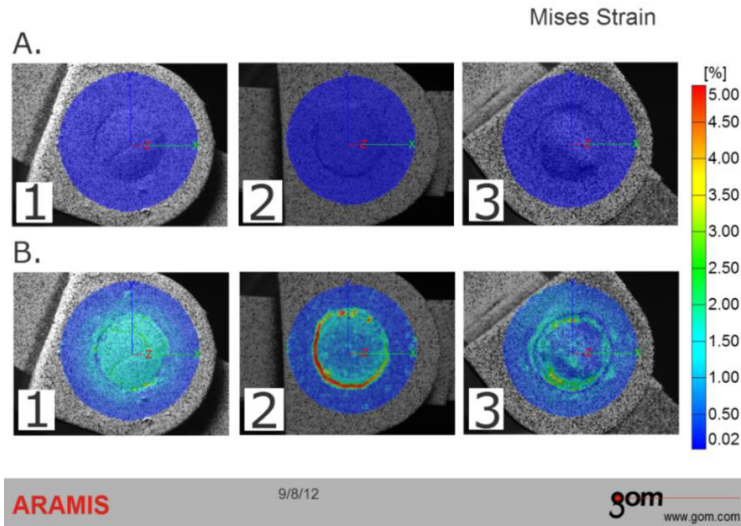


Figure 4 Strain field for composites in Teflon moulds. Sample: 1 – Z250; 2- Tetric EvoCeram; 3 - Silorane; A. Before light curing; B. After light curing

All three composites have exhibited non-homogeneous strain field.

Each composite sample in Teflon moulds was tested by using a circular section whose diameter was 1 mm (Figure 5), which consisted of 30 measuring points per sample. For each point, the coordinates before and after light curing were measured, along with resulting displacement, and strain was calculated based on it. Experimentally obtained data are graphically represented for each central section of a representative sample in order to obtain curves that show strain as a function of circular central section length.

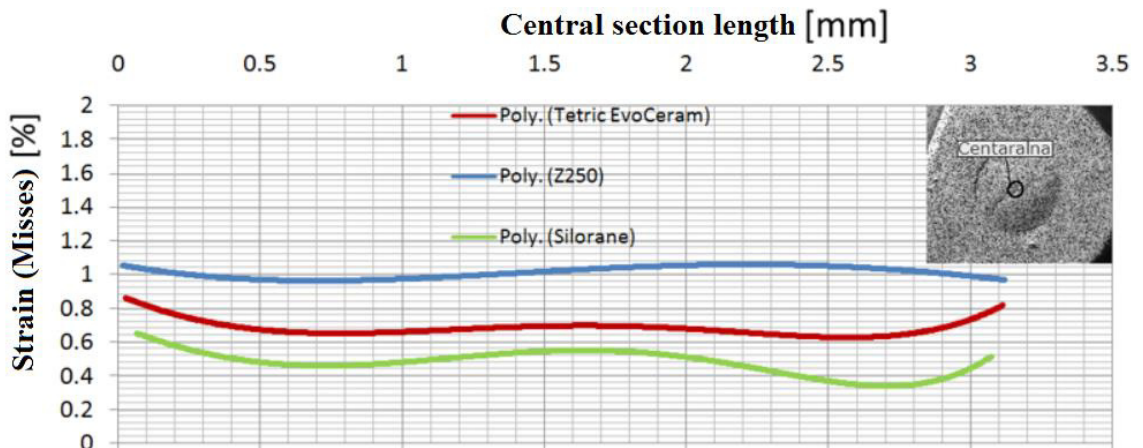


Figure 5 Functional dependence of strain from circular central section length of the defined composite

6. Discussion

Digital image correlation is a non-contact method which was used in this paper to measure volumetric shrinkage. It is based on monitoring of the position of surface markers before and after light curing, based on which strain is determined using a specialised software. The use of digital image correlation during this study involved the use of two cameras, in order to obtain shrinkage data in three-dimensions.

During the last few decades, several methods for measuring polymerization shrinkage and analysing its effects were developed. Developing of monomers attracted significant attention from chemists, which were investigating free polymerization shrinkage that can be measured by using, for example, Archimedes method, mercury dilatometer or optical monitoring of volumetric changes [35-37]. For direct application in dentistry, scientists were more interested in investigating polymerization shrinkage in tooth tissue cause by shrinkage stresses [16, 38, 39]. Polymerization shrinkage can be measured using a micro tensile test machine [40, 38], strain gauges [41], digital image correlation [36], linear variable differential transformers (LVDT), i.e. by applying Watts and Cash method [42]. Some of these displacements do not completely correspond to clinical situations, since the experimental setup typically idealises these conditions. Showing of the consequences of polymerization shrinkage is possible to achieve by using the FEM method, with limitations in calculation accuracy and necessary verification via physical experiment. In addition, contact methods used until now can cause the occurrence of additional gravitational or adhesive forces, which could deform the composite material before light curing begins [43, 44-46]. Currently available literature does not contain many data on three-dimensional, optical, non-contact analysis of composite strain which occurs due to light curing. Understanding of polymerization shrinkage, local strain and displacement field in accordance with the structural properties of the composite is of great importance for further application of existing and improving of new composite materials.

Chuang et al. [47] investigated the 2D influence of cavity size on polymerization shrinkage and cavity wall displacement, by using digital image correlation method with one camera. Shrinkage magnitude on the restoration surface depended on cavity geometry, as well as free and bonded tooth surfaces. Initial displacement was measured after light curing and ranged between 19 I 37 μm , and half an hour after illuminating with the lamp, it increased by additional 2-5 μm . Obtained results have shown that polymerization and resulting shrinkage do not end after the light is turned off, which represents the basis for further development of the methodology for 3D shrinkage measuring once the illuminating stops, also known as the post-polymerization effect.

By comparing the results of strain fields for all three samples, it can be noticed that strain values shown in the central part of the field (Figure 4) correspond to mean values shown in the diagram given in Figure 5. Weinmann et al. (3M, ESPE, Seefeld, Germany) have shown that values according to Watts & Cash method for Silorane are around 1%, and for Filtek Z250 around 1,8 % [48]. Mean strain values obtained in this dissertation for Silorane and Filtek Z250 correspond to values obtained in research conducted by Weinmann et al., except for composite TetricEvoCeram, which represents a new generation of composites, and was not tested at that time.

7. Conclusion

Presented in this paper was the testing of dental resin based composite materials, i.e. the analysis of light curing of these materials due to polymerization by means of LED diode in Teflon moulds. Strain fields and values are shown in the central part of the measuring area for 3 composites, Z250, TetricEvoCeram and Silorane. All three composites exhibited a non-homogeneous strain field, wherein highest polymerization shrinkage was present in Z250, and the lowest in Silorane.

Advantages and disadvantages of composite materials were also shown in this paper. It was observed that the most common consequences of light curing include shrinkage stress and cracks, which may cause marginal gaps between fillings and tooth, and failure even in case of stresses significantly lower than the yield strength of the material.

Acknowledgements

This study was supported by Research grant TR35040 from the Ministry of Education, Science and Technological Development, Republic of Serbia. The authors are grateful to 3M ESPE for a generous donation of the materials used in the study.

REFERENCES

- Papadogiannis DY, Lakes RS, Papadogiannis Y, Palaghias G, Helvatjoglu-Antoniades M. The effect of temperature on the viscoelastic properties of nano-hybrid composites. *Dent Mater*. 2008 Feb;24(2):257-66.
- Arola D, Galles LA, Sarubin MF. A comparison of the mechanical behavior of posterior teeth with amalgam and composite MOD restorations. *J Dent*. 2001 Jan;29(1):63-73.
- Maserejian NN, Hauser R, Tavares M, Trachtenberg FL, Shrader P, McKinlay S. Dental Composites and Amalgam and Physical Development in Children. *J Dent Res*. 2012 Sep 12.
- Alb C MM, Alb S, Moldovan M, Dudea D. The structure, composition and visual opacity of some dental composite resins. *European Cells and Materials* 2005;10:10.
- Tamas C, Moldovan M, Prejmerean C, Colceriu A, Furtos G, Vezensyi L, et al. Structure and properties of inorganic fillers for dental composites. *Journal of Optoelectronics and Advanced Materials*. 2005 Dec;7(6):2849-52.
- Nakabayashi N. Dental biomaterials and the healing of dental tissue. *Biomaterials*. 2003 Jun;24(13):2437-9.
- Masaki K, Nomura Y, Bhawal UK, Sawajiri M, Hirata I, Nahara Y, et al. Apoptotic and necrotic influence of dental resin polymerization initiators in human gingival fibroblast cultures. *Dent Mater J*. 2007 Nov;26(6):861-9.
- Joe C, Ontiveros RP. Light-emitting diode polymerization: a review of performance, part I. *Acta Stomatologica Naissi*. 2006;22(53):601-10.
- Bowen RL, Nemoto K, Rapson JE. Adhesive bonding of various materials to hard tooth tissues: forces developing in composite materials during hardening. *J Am Dent Assoc*. 1983 Apr;106(4):475-7.
- Yap AU, Soh MS. Post-gel polymerization shrinkage of "low shrinkage" composite restoratives. *Oper Dent*. 2004 Mar-Apr;29(2):182-7.
- Alomari QD, Reinhardt JW, Boyer DB. Effect of liners on cusp deflection and gap formation in composite restorations. *Oper Dent*. 2001 Jul-Aug;26(4):406-11.
- Janda R, Roulet JF, Kaminsky M, Steffin G, Latta M. Color stability of resin matrix restorative materials as a function of the method of light activation. *Eur J Oral Sci*. 2004 Jun;112(3):280-5.
- Tjan AH, Bergh BH, Lidner C. Effect of various incremental techniques on the marginal adaptation of class II composite resin restorations. *J Prosthet Dent*. 1992 Jan;67(1):62-6.
- Davidson CL, Feilzer AJ. Polymerization shrinkage and polymerization shrinkage stress in polymer-based restoratives. *J Dent*. 1997 Nov;25(6):435-40.
- Gallo JR, Burgess JO, Ripps AH, Walker RS, Maltezos MB, Mercante DE, et al. Three-year clinical evaluation of two flowable composites. *Quintessence Int*. 2010 Jun;41(6):497-503.
- Fabianelli A. A study into the significance of tracing microleakage by color die infiltration Siena: School of dental medicine, University of Siena. 2004.
- Davidson CL, de Gee AJ, Feilzer A. The competition between the composite-dentin bond strength and the polymerization shrinkage stress. *J Dent Res*. 1984 Dec;63(12):1396-9.
- Jorgensen KD, Asmussen E, Shimokobe H. Enamel damages caused by contracting restorative resins. *Scand J Dent Res*. 1975 Mar;83(2):120-2.
- Eick JD, Welch FH. Polymerization shrinkage of posterior composite resins and its possible influence on postoperative sensitivity. *Quintessence Int*. 1986 Feb;17(2):103-11.
- Kemp-Scholte CM, Davidson CL. Marginal integrity related to bond strength and strain capacity of composite resin restorative systems. *J Prosthet Dent*. 1990 Dec;64(6):658-64.
- Tantbirojn D, Versluis A, Pintado MR, DeLong R, Douglas WH. Tooth deformation patterns in molars after composite restoration. *Dent Mater*. 2004 Jul;20(6):535-42.
- Letzel H. Survival rates and reasons for failure of posterior composite restorations in multicentre clinical trial. *J Dent*. 1989;17 Suppl 1:S10-7; discussion S26-8.
- Ferracane JL. Developing a more complete understanding of stresses produced in dental composites during polymerization. *Dent Mater*. 2005 Jan;21(1):36-42.
- Yamamoto T, Nishide A, Swain MV, Ferracane JL, Sakaguchi RL, Momoi Y. Shrinkage stresses in dental composites adjacent to and at the bonded interface as measured by crack analysis. *Acta Biomater*. 2011 Jan;7(1):417-23.

25. Soares CJ, Martins LR, Pfeifer JM, Giannini M. Fracture resistance of teeth restored with indirect-composite and ceramic inlay systems. *Quintessence Int.* 2004 Apr;35(4):281-6.
26. Ausiello P, De Gee AJ, Rengo S, Davidson CL. Fracture resistance of endodontically-treated premolars adhesively restored. *Am J Dent.* 1997 Oct;10(5):237-41.
27. Brackett WW, Covey DA, St Germain HA, Jr. One-year clinical performance of a self-etching adhesive in class V resin composites cured by two methods. *Oper Dent.* 2002 May-Jun;27(3):218-22.
28. Stansbury JW, Trujillo-Lemon M, Lu H, Ding X, Lin Y, Ge J. Conversion-dependent shrinkage stress and strain in dental resins and composites. *Dent Mater.* 2005 Jan;21(1):56-67
29. Feilzer AJ, De Gee AJ, Davidson CL. Setting stress in composite resin in relation to configuration of the restoration. *J Dent Res.* 1987 Nov;66(11):1636-9.
30. Watts DC, Marouf AS, Al-Hindi AM. Photo-polymerization shrinkage-stress kinetics in resin-composites: methods development. *Dent Mater.* 2003 Jan;19(1):1-11.
31. Kinomoto Y, Torii M. Photoelastic analysis of polymerization shrinkage stresses in resin composite restorations. *J Dent.* 1998 Mar;26(2):165-71.
32. Kinomoto Y, Torii M, Takeshige F, Ebisu S. Comparison of polymerization shrinkage stresses between self- and light-curing composites. *J Dent.* 1999 Jul;27(5):383-9.
33. Kinomoto Y, Torii M, Takeshige F, Ebisu S. Polymerization shrinkage stresses of resin-based composite restorations within beveled cavity preparations of Class I restorations. *Am J Dent.* 2003 Apr;16(2):139-43.
34. Miletic V, Manojlovic D, Milosevic M, Mitrovic N, Stankovic TS, Maneski T. Analysis of local shrinkage patterns of self-adhering and flowable composites using 3D digital image correlation. *Quintessence Int.* 2011 Oct;42(9):797-804.
35. de Gee AJ, Davidson CL, Smith A. A modified dilatometer for continuous recording of volumetric polymerization shrinkage of composite restorative materials. *J Dent.* 1981 Mar;9(1):36-42.
36. de Gee AF, Feilzer AJ, Davidson CL. True linear polymerization shrinkage of unfilled resins and composites determined with a linometer. *Dent Mater.* 1993 Jan;9(1):11-4.
37. Sharp LJ, Choi IB, Lee TE, Sy A, Suh BI. Volumetric shrinkage of composites using video-imaging. *J Dent.* 2003 Feb;31(2):97-103.
38. Davidson CL, de Gee AJ. Relaxation of polymerization shrinkage stresses by flow in dental composites. *J Dent Res.* 1984 Feb;63(2):146-8.
39. Fard MY, Liu YT, Chattopadhyay A. Characterization of Epoxy Resin Including Strain Rate Effects Using Digital Image Correlation System. *J Aerospace Eng.* 2012 Apr;25(2):308-19.
40. Lohbauer U, Frankenberger R, Kramer N, Petschelt A. Strength and fatigue performance versus filler fraction of different types of direct dental restoratives. *J Biomed Mater Res B Appl Biomater.* 2006 Jan;76(1):114-20.
41. Clifford SS, Roman-Alicea K, Tantbirojn D, Versluis A. Shrinkage and hardness of dental composites acquired with different curing light sources. *Quintessence Int.* 2009 Mar;40(3):203-14.
42. Watts DC, Cash AJ. Determination of polymerization shrinkage kinetics in visible-light-cured materials: methods development. *Dent Mater.* 1991 Oct;7(4):281-7.
43. Boaro LC, Goncalves F, Guimaraes TC, Ferracane JL, Versluis A, Braga RR. Polymerization stress, shrinkage and elastic modulus of current low-shrinkage restorative composites. *Dent Mater.* 2010 Dec;26(12):1144-50.
44. Fleming GJ, Hall DP, Shortall AC, Burke FJ. Cuspal movement and microleakage in premolar teeth restored with posterior filling materials of varying reported volumetric shrinkage values. *J Dent.* 2005 Feb;33(2):139-46.
45. Hertele S, de Waele W, Denys R, Verstraete M. Investigation of strain measurements in (curved) wide plate specimens using digital image correlation and finite element analysis. *J Strain Anal Eng.* 2012, Jul;47(5):276-88.
46. Sakaguchi RL, Versluis A, Douglas WH. Analysis of strain gage method for measurement of post-gel shrinkage in resin composites. *Dental Materials.* 1997 Jul;13(4):233-9.
47. Chuang, S. F., Chang, C. H. & Chen, T. Y. 2011. Spatially resolved assessments of composite shrinkage in MOD restorations using a digital-image-correlation technique. *Dent Mater*, 27, 134-43.
48. Weinmann W, Thalacker C, Guggenberger R. Siloranes in dental composites. *Dent Mater.* 2005 Jan;21(1):68-74.