J. King Saud Univ., Vol. 21, *Dental Sci.* (2), pp. 57-62, Riyadh (2010/1431H.)

The Effect of Light Exposure Time Variations on Shrinkage Strain Kinetics of Resin-based Composite Materials

Abdulaziz A. Al-Kheraif
Assistant Professor, Chairman of Dental Health Department,
College of Applied Medical Sciences, King Saud University,
Riyadh, Saudi Arabia
aalkhuraif@ksu.edu.sa

(Received 15/3/2010; accepted for publication 12/6/2010)

Keywords: Shrinkage strain, Light cure, Resin composite, Exposure time.

Abstract. The aim of this study was to analyze the effect of conventional quartz-tungsten-halogen (QTH) light exposure time variations on shrinkage strain kinetics of resin-based composite materials to determine whether there are differences among them. Two photo-activated resin composites were selected. The bonded-disk technique was used to measure shrinkage-strain kinetics. Measurements were obtained continuously for 30 min by irradiation for 20s, 40s, 60s, 80s, and 100s at 750 mW/cm^2 at 23^{o}C. Three measurements (n = 3) were made per material. The data were analyzed using ANOVA tests. The shrinkage strain of the composite resins at 40s, 60s, 80s, and 100s exposure to light were always significantly higher than the 20s exposure ($p<0.05$) whereas there was no significant difference between 40s, 60s, 80s, and 100s exposure for both the materials ($p>0.05$). The data show that Ecusit® resin had higher shrinkage strain than Gradia ® Direct ($p<0.05$). The extent of polymerization of a light cured resin composite, increased with recommended time, 40s, compared to underexposure time, 20s, and a photo-activated composite restorative resin should be irradiated for 40s to achieve adequate polymerization.

Introduction

Resin composite restorative materials have changed dramatically in composition to obtain better color stability over time (Tyas, 1992), greater wear resistance (Yap, 2002), and clinically acceptable surface smoothness of restorations (Setcos *et al.*, 1999), since their introduction in the 1970s. Dental composites consisting of an organic matrix (e.g., Bis-GMA or UDMA) filled with inorganic filler particles undergo volumetric shrinkage when cured (Peutzfeldt, 1997). Moreover, the kinetics of polymerization and network formation is dependent upon the wavelength, irradiation and duration of the initiating light, and especially upon the ambient and internal temperature levels produced during polymerization (Watts and Marouf, 2000). Shrinkage-strain occurring during polymerization of matrix monomers of restorative materials is a significant phenomenon of the materials and is still one of the main causes of failures in dental composite restorations (Chung *et al.*, 2002). It has been reported that the inorganic filler content of the composite (Iga *et al.*, 1991), the type of monomer, light intensity and curing cycle (Unterbrink and Muessener, 1995; Dennison *et al.*, 2000) affect the polymerization shrinkage. Christensen *et al.* (1999) also showed that the resin formulation plays the major role rather than the curing unit type and curing mode. Shrinkage-strain and initial strain rate were dependent upon monomer functionality, molecular mass and viscosity. Shrinkage-strain rates rose rapidly to a maximum, and then fell rapidly upon vitrification (Atai *et al.*, 2005). Inadequate polymerization can result in loss of biocompatibility, color shifts, loss of retention, breakage, excessive wear and poor physical properties that may lead to early failure (Rueggeberg *et al.*, 1993). A light cured resin composite may also show this problem because conversion in the deep part of the restoration is dependent on light intensity, curing time and other factors related to the polymerization of the materials (Øysaed and Ruyter, 1987). Furthermore, the final degree of conversion depends on the chemical structure of the methacrylate monomer and the polymerization conditions, such as temperature and light intensity (Sideridou *et al.*,

2002). Several publications have studied the influence of polymerization shrinkage on composite resin restorations (Grifftihs *et al.*, 1999; Koran and Kurschner, 1998; Watts and Al-Hindi, 1999). Therefore, the purpose of this study was to analyze the effect of exposure time variations on shrinkage strain kinetics of resin-based composite materials to determine whether there are differences among them. In addition, the polymerization shrinkage rate compared at each exposure time was studied. The null hypothesis is that significant differences will be detected among the tested exposure times.

Materials and Methods

Two photo-activated resin composites were used in this study: Hybrid; Ecusit® resin (DMG Germany) and Microfilled; Gradia ® Direct (GC America, Alsip, IL, USA). The composition of each material is detailed in Table 1. A conventional quartz-tungsten-halogen (QTH) light (Elipar 2500, 3M ESPE, St Paul, MN-Germany) was used in this study.

Table 1. Manufacture's information concerning the compositions of the light cured resin composites used in this study

Restorative Resin	Manufacturer	Composition
Ecusit® resin	DMG Germany	Hybrid universal filling composite , Barium glass in a Bis-GMA based matrix of dental resin, pigments, additives, and catalyst, with filler ratio 77 % by weight (0.02-1.5µm).
Gradia ® Direct	GC America, Alsip, IL, USA	Microfilled Hybrid, Urethane dimethacrylate (UDMA) and dimethacrylate co-monomer matrix, silica and pre-polymerized fillers (0,85µm)

Shrinkage strain measurement

The shrinkage-strain was performed at room temperature using the bonded-disk technique (Watts and Cash, 1991; Watts and Marouf, 2000). This method measures polymerization shrinkage-strain kinetics. A disk-shaped specimen (7 mm × 1.5 mm) was placed at the center of a square cross section circular brass ring acting as a spacer of internal diameter 16 mm and height 1.0 mm. This ring was adhesively bonded onto a rigid glass plate, 74 mm × 25 mm × 3 mm. The top edge of the ring and disc specimen were covered by a flexible 22 mm square cover-slip, thickness 0.1 mm (type 0, Chance-Propper Ltd., Warley, UK). The tip of a QTH curing light was positioned underneath and in close proximity of the glass plate, so that light-cure could be achieved by irradiating from below, through the glass. A linear variable displacement transducer (LVDT) measuring system was positioned centrally onto the cover slip. Following thermal and mechanical equilibration, measurement runs were made by irradiation for 20s, 40s, 60s, 80s, and 100s at 750 mW/cm^2 at 23^{o}C. Measurements were recorded after 30 min from photoinitiation (n = 3 per material).

Shrinkage strain rate measurement

Each material at different curing times was divided into five groups:

1. Group 1: from zero to 20s
2. Group 2: from 20s to 40s
3. Group 3: from 40s to 60s
4. Group 4: from 60s to 80s
5. Group 5: from 80s to 100s

Data were analyzed statistically by one-way ANOVA and Tukey test at 5% significance level.

Results

The means and standard deviations for both resin composites with different exposure times used in this study are shown in Tables 2 and 3. Statistical testing showed that the shrinkage strain of the resin composites at 40s, 60s, 80s, and 100s exposure to light were always significantly higher than the 20s exposure ($p<0.05$) whereas no significant difference between 40s, 60s, 80s, and 100s exposure for both the materials ($p>0.05$) (Figs. 1 and 3). Therefore, extended exposure time had no significant effect on the polymerization shrinkage strain of the tested two photo-activated resin composites. The shrinkage strain rate at 20s, 40s, 60s, 80s, and 100s was not significant difference for both the materials ($p>0.05$) (Figs. 2 and 4).

Discussion

The physical, mechanical and biological properties of a photo-activated composite restorative resin are strongly influenced by the degree of conversion achieved by the organic matrix. Ideally, a resin material would have all of its monomer converted to polymer during polymerization (Eliades and Vougiouklakis, 1987; Ferracane and Greener, 1984; Ruyter and Oysaed, 1988). It is also known that polymerization of a photo-activated composite restorative resin is influenced by the attenuation of the intensity of the photo-activating radiation during its passage through the translucent composite resin.

Table 2. Mean values and SD of maximum shrinkage-strain ε (%) and maximum shrinkage-strain rate (%s^{-1}) and the associated time t (s) with different exposure times

Material	Ecusit® resin			Gradia ® Direct		
Exposure Time	Strain (%)	Rate (%s^{-1})	t (s)	Strain (%)	Rate (%s^{-1})	t (s)
20Sec	1.900± 0.787aA	0.146	0.004	1.211± 0.512bA	0.081	0.002
40Sec	2.178± 0.851aB	0.154	0.004	1.500± 0.287bB	0.082	0.003
60Sec	2.196± 0.461aB	0.156	0.004	1.557± 0.461bB	0.073	0.003
80Sec	2.198± 0.987aB	0.157	0.005	1.589± 0.612bB	0.088	0.003
100Sec	2.212± 0.651aB	0.158	0.005	1.562± 0.432bB	0.079	0.003

Means followed by the same lowercase letters in lines and uppercase letters in columns indicate no statistical difference at 95% confidence level (Tukey's test; p<0.05).

Table 3. Maximum shrinkage strain rate (%s^{-1}) at each group (G) with different exposure times

Material	Ecusit® resin					Gradia® Direct				
Exposure Time	Strain Rate (%s^{-1})					Strain Rate (%s^{-1})				
	G1	G2	G3	G4	G5	G1	G2	G3	G4	G5
20Sec	0.146	0.037	0.007	0.004	0.002	0.081	0.017	0.005	0.003	0.002
40Sec	0.154	0.039	0.012	0.004	0.002	0.082	0.024	0.009	0.003	0.003
60Sec	0.156	0.038	0.013	0.007	0.003	0.073	0.026	0.009	0.007	0.003
80Sec	0.157	0.037	0.012	0.006	0.003	0.088	0.025	0.009	0.009	0.006
100Sec	0.158	0.038	0.012	0.004	0.003	0.079	0.024	0.009	0.008	0.004

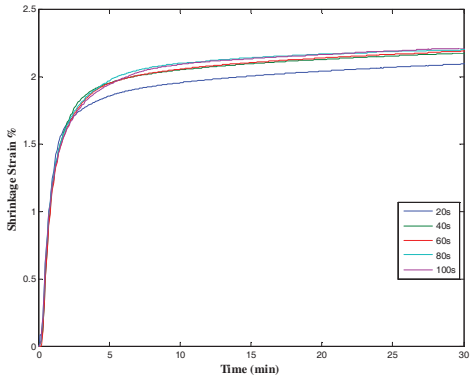

Fig. 1. Polymerization shrinkage strain of Ecusit® resin at 23°C.

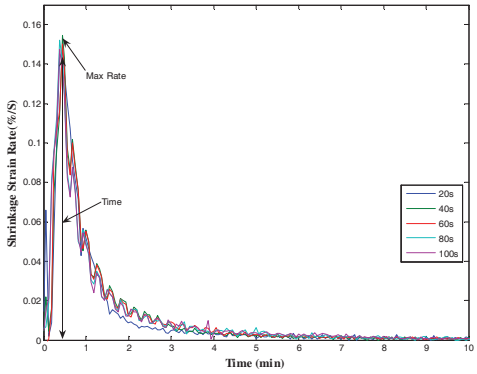

Fig. 2. Polymerization shrinkage strain rate of Ecusit® resin at 23°C.

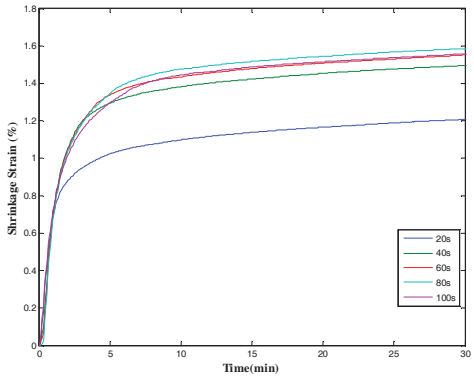

Fig. 3. Polymerization shrinkage strain of Gradia® Direct at 23°C.

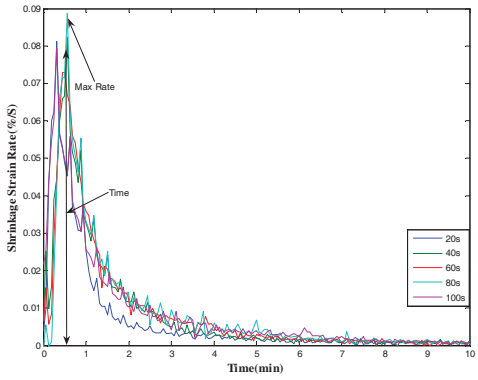

Fig. 4. Polymerization shrinkage strain rate of Gradia® Direct at 23°C.

Hence, the conventional quartz-tungsten-halogen (QTH) light cured composite must be cured for at least 40 seconds to initiate a reaction that ensures the curing will continue to completion (Leung *et al.*, 1983). The amount of exposure time to light is one of the most important operator-dependent factors governing the polymerization of light activated composites. When the light intensity is sufficient to activate the photoinitiator in the restorative material, the duration of exposure to light is the next rate limiting step in the polymerization process (Caughman *et al.*, 1995; Lui *et al.*, 2006). In the present study, the shrinkage strain values at an exposure time of 20s was found to give shrinkage strain values than at an exposure time of 40s. This may be because the system is slightly underexposed for photo-activation, possibly resulting in a low degrees of polymerization. For samples exposed to the photo-activating light for 20s and 40s, an increase in exposure time was accompanied by higher shrinkage strain values, indicating higher degrees of polymerization. Ceballos *et al.* (2009) found, that an increase in irradiation time from 20s to 40s produced a statistically significant increase in hardness values. Silikas *et al.* (2000), and Emami and Soderholm (2003) also showed that the extent of polymerization of a photo-activated resin composite, was influenced by the exposure time to the photo-activating radiation, as the curing time increased from 20s to 40s, the polymerization also increases resulting in higher values of the degree of conversion. However, the differences for samples exposed to the photo-activating light for 40s and 60s were less than the differences between 20s and 40s. This suggested that the effect of extended exposure time above the manufacturer's recommended time was considerably less than the effect of underexposure (Leung *et al.*, 1983). The findings of the present study are in agreement with those of Fan *et al.* (2002), who found that a polymerization light with an intensity of 300 mW/cm^2 would effectively cure most composite shades within the manufacturer's time, but a darker shades might require longer exposure times. The difference in conversion degree could also have been identified in microfilled composites, like Gradia® Direct, because the microfillers particles (silica) cause light scattering, which decreases the effectiveness of the curing light (Leonard *et al.*, 2002; Sobrinho *et al.*, 2000). Christensen *et al.* (1999), and Yazici *et al.* (2008) reported that the resin formulation plays a major role rather than the curing unit type and mode in polymerization. In the current study, Ecusit® resin had higher significant shrinkage strain values than Gradia® Direct. This could be attributed to differences in filler particles of the resin based materials affecting the mechanical properties and the type of polymer employed (Leinfelder, 1991) . Gradia is based mainly on Urethane dimethacrylate (UDMA), this could be attributed to the lower shrinkage strain of Gradia than Ecusit resin. The results of this current study showed that extended exposure time up to 100s was not significantly different from that of 40s exposure. This was in agreement with the results of previous studies (Tanoue *et al.*, 2007; Braga, 2002), and can be explained by the fact that adequate polymerization is not only a function of exposure time to the light, but it is also influenced by the material's composition (Leonard *et al.*, 2001). The result obtained from the shrinkage strain rate of the two light cured resin composites showed that maximum shrinkage strain rate ($\%s^{-1}$) occurred during the first 20s (Group 1), with different exposure times. This might serve as an explanation for extended exposure time did not have any influence on shrinkage strain values of both light cured resin composites. The author believes that possible influences on the shrinkage strain could be the exposure time. However, the exposure times evaluated in this study showed differences in shrinkage strain values (between 20s on the one hand and 40s, 60s, 80s, and 100s on the other hand), therefore the null hypothesis is accepted. Further studies should be conducted to elucidate the effect of light exposure time on shrinkage strain of recent types of low shrinkage resin composite.

Conclusion

Under the limitations of this in vitro study it can be concluded that:

1. The extent of polymerization of a photo-activated composite, increased with recommended time, 40s, compared to underexposure time, 20s.
2. Extended exposure to the photo-activating light did not significantly increase the polymerization shrinkage strain. Light exposures of 60s, 80s, and 100s showed the same shrinkage strain values as that of 40s.
3. Resin composites should be irradiated for 40 s to achieve adequate polymerization.

Acknowledgements. This study was supported by a grant from the College of Applied Medical Sciences Research Center, King Saud University, Riyadh, Saudi Arabia.

References

Atai, M.; Watts, D. C. and Atai, Z. "Shrinkage Strain-rates of Dental Resin-monomer and Composite Systems." *Biomaterials.*, Vol. 26, No. (24), (2005), 5015-5020.

Braga, R. R. and Ferracane, J. L. "Contraction Stress Related to Degree of Conversion and Reaction Kinetics." *J. Dent. Res.*, Vol. 81, No. (2), (2002), 114-118.

Caughman, W. F.; Rueggeberg, F. A. and Curtis, J. W. "Clinical Guidelines for Photocuring Restorative Resins." *J. Am. Dent. Assoc.*, Vol. 126, (1995), 1280-6.

Ceballos, L.; Fuentes, M. V.; Tafalla, H.; Martínez, A.; Flores, J. and Rodríguez, J. "Curing Effectiveness of Resin Composites at Different Exposure Times Using LED and Halogen Units." *Med. Oral. Patol. Oral. Cir. Bucal.*, Vol. 14, No. (1), (2009), 51-6.

Christensen, R. P.; Palmer, T. M.; Ploeger, B. J. and Yost, M. P. "Resin Polymerization Problems: Are They Caused by Resin Curing Lights, Resin Formulation or Both?" *Compend. Contin. Educ. Dent.*, Vol. 25 (Suppl.), (1999), S42-54.

Chung, C. M.; Kim, J. G.; Kim, M. S.; Kim, K. M. and Kim, K. N. "Development of a New Photocurable Composite Resin with Reduced Curing Shrinkage." *Dent. Mater.*, Vol. 18, (2002), 174-8.

Dennison, J. B.; Yaman, P.; Seir, R. and Hamilton, J. C. "Effect of Variable Light Intensity on Composite Shrinkage." *J. Prosthet. Dent.*, Vol. 84, (2000), 499-505.

Eliades, G. C.; Vougiouklakis, G. J. and Caputo, A. A. "Degree of Double Bond Conversion in Light-cured Composites." *Dent. Mater.*, Vol. 3, No. (1), (1987), 19-25.

Emami, N. and Söderholm, K. J. "How Light Irradiance and Curing Time Affect Monomer Conversion in Light-cured Resin Composites." *Eur. J. Oral. Sci.*, Vol. 111, No. (6), (2003), 536-542.

Fan, P.; Schumacher, R.; Azzolin, K.; Geary, R. and Eichmiller, F. "Curing-light Intensity and Depth of Cure of Resin Based Composites Tested According to International Standards." *J. Am. Dent. Assoc.*, Vol. 133, (2002), 429-435.

Ferracane, J. L. and Greener, E. H. "Fourier Transform Infrared Analysis of Degree of Polymerization in Unfilled Resins: Methods Comparison." *J. Dent. Res*., Vol. 63, No. (8), (1984), 1093-5.

Grifftihs, B. M.; Naasan, M.; Sherriff, M. and Watson, T. F. "Variable Polymerization Shrinkage and the Interfacial Micropermeability of a Dentin Bonding System." *J. Adhes. Dent.*, Vol. 1, (1999) ,119-31.

Iga, M.; Takeshige, F.; Ui, T. and Torii, M. "The Relationship between Polymerization Shrinkage Measured by a Modified Dilatometer and the Inorganic Filler Content in Light Cured Composites." *Dent. Mater. J.*, Vol. 10, (1991), 38-40.

Koran, P. and Kurschner, R. "Effect of Sequential versus Continuous Irradiation of a Light-cured Resin Composite on Shrinkage, Viscosity, Adhesion and Degree of Polymerization." *Am. J. Dent.*, Vol. 11, (1998), 17-22.

Leinfelder, K. F. "Using Composite Resin as a Posterior Restorative Material." *J. Am. Dent. Assoc.*, Vol. 22, (1991), 65-70.

Leonard, D. L.; Charlton, D. G.; Roberts, H. W.; Hilton, T. J. and Zionic, A. "Determination of the Minimum Irradiance Required for Adequate Polymerization of Hybrid and Microfill Composite." *Oper. Dent.*, Vol. 26, (2001), 176-80.

Leonard, D. L.; Charlton, D. G.; Roberts, H. W. and Cohen, M. E. "Polymerization Efficiency of LED Curing Lights." *J. Esthet. Restor. Dent.*, Vol. 14, (2002), 286-295.

Leung, R. L.; Fan, P. L. and Johnston, W. M. "Post-irradiation Polymerization of Visible Light-activated Composite Resin." *J. Dent. Res.*, Vol. 62, (1983), 363-5.

Lui, J. L.; Chan, C. L. and Yap, K. T. "Depth of Polymerization of a New Nanocomposite According to Different Exposure Times and Shades." *Annal. Dent. Univ. Malaya.*, Vol. 13, (2006), 6-11.

Øysaed, H. and Ruyter, I. E. "Composites for Use in Posterior Teeth: Composition and Conversion." *J. Biomed. Mater. Res.*, Vol. 21, (1987)**,** 11-23.

Peutzfeldt, A. "Resin Composites in Dentistry: The Monomer Systems." *Eur. J. Oral. Sci.*, Vol. 105, (1997), 97-116.

Rueggeberg, F. A.; Caughman, W. F.; Curtis, J. W., Jr. and Davis, H. C. "Factors Affecting Cure at Depths within Light-activated Resin Composites." *Am. J. Dent.*, Vol. 6, (1993), 91-95.

Ruyter, I. E. and Oysaed, H. "Analysis and Characterization of Dental Polymers." *Critical Rev. Biocompat.*, Vol. 4, No. (3), (1988), 247-79.

Setcos, J. C.; Tarim, B. and Suzuki, S. "Surface Finish Produced on Resin Composites by New Polishing Systems." *Quintessence. Int.*, Vol. 30, (1999), 169-73.

Sideridou, I.; Tserki, V. and Papanastasiou, G. "Effect of Chemical Structure on Degree of Conversion in Light-cured Dimethacrylate-based Dental Resins." *Biomaterials.*, Vol. 23, (2002) ,1819-29.

Silikas, N.; Eliades, G. and Watts, D. C. "Light Intensity Effects on Resin-composite Degree of Conversion and Shrinkage Strain." *Dent. Mater.*, Vol. 16, No. (4), (2000), 292-296.

Sobrinho, L. C.; Lima, A. A.; Consani, S.; Sinhoreti, M. A. and Knowles, J. C. "Influence of Curing Tip Distance on Composite Knoop Hardness Values." *Braz. Dent. J.*, Vol. 11, No. (1), (2000), 11-7.

Tanoue, N.; Murakami, M.; Koizumi, H.; Atsuta, M. and Matsumura, H. "Depth of Cure and Hardness of an Indirect Composite Polymerized with Three Laboratory Curing Units." *J. Oral. Science*, Vol. 49, (2007), 25-29.

Tyas, M. J. "Colour Stability of Composite Resins: A Clinical Comparison." *Aust. Dent. J.*, Vol. 37, (1992), 88-90.

Unterbrink, G. L. and Muessener, R. "Influence of Light Intensity on Two Restorative Systems." *J Dent.*, Vol. 23, (1995), 183-189.

Watts, D. C. and Al-Hindi, A. "Intrinsic 'Soft Start' Polymerization Shrinkage-kinetics in an Acrylate-based Resin-composite." *Dent. Mater.*, Vol. 15, No. (1), (1999), 39-45.

Watts, D. C. and Cash, A. J. "Determination of Polymerization Shrinkage Kinetics in Visible-light-cured Materials: Methods Development." *Dent. Mater.*, Vol. 7, (1991), 281-7.

Watts, D. C. and Marouf, A. S. "Optimal Specimen Geometry in Bonded-disk Shrinkage-strain Measurements on Light-cured Biomaterials." *Dent. Mater.*, Vol. 16, (2000), 447-51.

Yap, A. U. "Occlusal Contact Area (OCA) Wear of Two New Composite Restoratives." *J. Oral Rehabil.*, Vol. 29, (2002), 194-200.

Yazici, A. R.; Celik, C.; Dayangac, B. and Ozgunaltay, G. "Effects of Different Light Curing Units/Modes on the Microleakage of Flowable Composite Resins." *Eur. J. Dent.*, Vol. 2, (2008), 240-246.

تأثير تنوع مدة التعرض للتصلب الضوئي على انكماش مواد الكمبوزيت

عبدالعزيز بن عبدالله الخريف

أستاذ مساعد، قسم صحة الأسنان، كلية العلوم الطبية التطبيقية، جامعة الملك سعود،
ص ب ١٠٢١٩، الرياض ١١٤٣٣، المملكة العربية السعودية

(قدم للنشر في ٢٠١٠/٣/١٥م؛ وقبل للنشر في ٢٠١٠/٦/١٢م)

ملخص البحث. هدفت هذه الدراسة إلى تحليل تأثير تنوع مدة التعرض للتصلب الضوئي على انكماش مواد الكمبوزيت وذلك لتحديد ما إذا كان يوجد اختلاف بينهم، حيث تم استخدام مادتين من الكمبوزيت تعتمدا على التصلب الضوئي. وتم قياس الانكماش بجهاز خاص لمدة (٣٠ دقيقة) بعد تعرض المواد لضوء الهولوجين لمدة (٢٠ ثانية) و (٤٠ ثانية) و (٦٠ ثانية) و (٨٠ ثانية) و (١٠٠ ثانية)، ومن ثم حللت النتائج واتضح وجود اختلاف ملحوظ بين التعرض لفترة (٢٠ ثانية) والتعرض لفترة (٤٠ ثانية) فأكثر وذلك فيما يتعلق بمقدار انكماش المادة. أظهرت المادة (Ecusit® resin) انكماش أعلى من المادة (Gradia® Direct)، كما أن محتوى البلمرة للمواد يزداد مع الوقت الموصى به (٤٠ ثانية) مقارنه بالوقت الأقل (٢٠ ثانية)، وبذلك يجب تعرض المادة للتصلب الضوئي لمدة (٤٠ ثانية) للحصول على بلمرة كافية.