The Monomer Elution of Bulk-Fill Sculptable Composite and Flowable Composite after Photopolymerization

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Abstract

Introduction: Composite resins have excellent mechanical properties, including fast polymerization, aesthetic quality, easy handling, and the ability to adhere to the enamel surfaces. However, there are also some disadvantages from this material, one of them is residual monomers. Bulk-fill composite resins are available in sculptable (solid) and flowable (liquid) form. Most composite resin matrix is aromatic or aliphatic acrylates. Bis-GMA, UDMA, and TEGDMA are the matrix monomers that are commonly used but show a high cytotoxicity level. They also have poor mechanical properties such as wear resistance, hardness, tendency to change color, and may cause pulp reactions. Aim: The aim of this study was to investigate the number of residual monomers in bulk-fill sculptable and flowable composites in the submersion of 75% ethanol solution (10 minutes, an hour, and 24 hours). Methods: Sculptable and flowable composites were used in this study. Samples (4 mm thick, 5 mm diameter) were prepared and polymerized for 10 seconds with an intensity of 1025 mW / cm2 of a light emitted diode (LED). After the sample was made, each sample was immediately immersed in a 75% ethanol solution as an extraction liquid and stored in amber-colored bottles at room temperature. Samples were taken as many as 7 cc at intervals of 10 minutes, an hour, and 24 hours. Samples were analyzed by High-Performance Liquid Chromatography (HPLC). The data obtained were analyzed using the Kruskal-Wallis Test, Independent T-Test and Mann-Whitney Test at a significance level of p <0.05. Result: The amount of elusion remaining monomers of the bulk-fill flowable composite was higher than bulk-fill sculptable composite. UDMA composite of bulk fill flowable submersed in 75% ethanol solution for 24 hours was the highest among other monomers. Conclusion: Residual monomers eluted inside bulk-fill composite resins in all time periods and the amount of eluted monomers increases with time.

Keywords: Residual monomer, monomer elution, bulk-fill composite, High-Performance Liquid Chromatography (HPLC)

Introduction

Composite resins have excellent mechanical properties, including fast polymerization, aesthetic

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Nanik Zubaidah, Departement of Conservative Dentistry, Faculty of Dental Medicine, Universitas Airlangga, Mayjend Prof. Dr. Moestopo Street 47, Surabaya, Indonesia, 60132. Phone Number: +628165450924. E-mail: nanik-z@fkg.unair.ac.id quality, easy handling, and the ability to adhere to the enamel surfaces. However, there are still concerns about the clinical condition of composite resin. Unreacted and residue of monomers in a set composite resin were known as residual monomers. The residual monomer can lead to oxidation and hydrolytic degradation, which can cause discoloration and worn out of the composite restoration. Moreover, residual monomers can cause soft tissue irritation, stimulate bacterial growth, and increase allergic reactions¹.

The monomer elution process is the release of monomers from polymer composite resins. The amount of monomer elution is influenced by (a) the number of unreacted monomers, which is determined by the chemical structure of the monomer and the polymerization conditions; (b) solvents used for monomer elution; and (c) the size and characteristics of the chemical structure of the monomer. Some studies found that small molecules, such as TEGDMA (having a molecular weight of 286 g/ mol) has a higher mobility monomer and faster elution process than larger molecules such as bis-GMA (having molecular weights of 512 g/mol). There are different views about the time taken to complete the elution of unreacted monomers. Some studies state that elution takes 1 to 7 days to complete, while other studies state that the complete elution process takes approximately 30 days^{2,3}.

Bulk-fill composite resins are available in sculptable (solid) and flowable (liquid) form. Most composite resin matrix is aromatic or aliphatic acrylates^{4,5}. The characteristic feature of bulk-fill composites is its increased depth of cure up to 4 mm and reduced polymerization shrinkage stress⁶. Tetric N-Ceram Bulk-fill has a patented filler which is partially functionalized by silanes, acts as a unique shrinkage stress reliever which essentially "holds on" to the cavity walls along with the matrix and the adhesive. Tetric N-Ceram Bulk Fill also utilizes the initiators like camphorquinone plus an acyl phosphine oxide, together with a recently patented initiator Ivocerin dibenzoyl germanium derivative which allows the application and curing in larger increments of up to 4 mm, without compromising the optical properties of the composite such as translucency or colour⁵.

Bis-GMA, UDMA, and TEGDMA are matrix that are commonly used in dentistry. Bis-GMA and UDMA are thick monomers to facilitate their clinical application. It is necessary to add monomers with a lower molecular weight called viscosity controllers. One of the diluting monomers used is TEGDMA. The majority of bulk-fill sculptable resin composites contain Bis-GMA monomers which are high viscosity due to its high molecular weight. While the majority of flowable bulk-fill composite resins contain TEGDMA monomers which are the opposite of Bis-GMA monomers. TEGDMA is an aqueus monomer because of its low viscosity and low molecular weight. The factory recommended polymerization time for 4 mm layer thickness is 20 seconds with light intensity 500 mW/cm² and 10 seconds with light intensity \geq 1000 mW/cm^{4,5,7}.

Recommended energy density for adequate polymerization of conventional composite resin at 2-mm depth is 21–24 J/cm^{2 8}. The present in vitro study has proven that energy density plays a pivotal role in adequate photopolymerization of bulk-fill composite resins. The high energy density (20 J/cm 2) derived from both increased curing time and low power of the curing device results in a faster release of free radicals and, consequently, a higher DC of the material and increased depth of cure⁹. This energy (E) is calculated by multiplying the irradiance level (I) coming from the light control unit (LCU) (mW/cm²) by its duration (T). Curing time is set depending on the irradiance level. The higher the irradiance level, the shorter the curing time needed¹⁰. El-Damanhoury and Platt (2014) Comparing bulk-fill resin-based composite (RBCs) with conventional RBCs and a micro-hybrid RBC (Filtek Z250) as a control. They reported adequate DC of all bulk-fill RBCs at 4 mm RBC thickness However, Filtek Z250 RBC showed inadequate DC of the bottom surface at 4 mm RBC thickness⁶. In conclusion, although 2 mm incremental thickness is still the regular standard for RBC increment placement, using bulk-fill allows placement of RBCs in more than 2 mm increments (up to 4 mm) while maintaining an adequate DC. This is due to higher light transmission through the more translucent bulk-fill RBC thickness when compared to conventional RBCs⁶. According to Sideridou et al., monomers bis-GMA, TEGDMA, UDMA, and bis-EMA dissolves in 75% ethanol, because 75% ethanol solution has the ability to penetrate the composite resin matrix, then expand and enlarge the polymeric bonds consisting of monomers which cause the remaining monomers to come out, 75% ethanol solution can also be used removing the residual bulk-fill resin monomer, Therefore, 75% ethanol solution was used in this study¹.

In addition, from the results of Cebe et al., revealed that residual monomers (BIS- GMA, UDMA, and TEGDMA) were found in all bulk-fill composite resins studied and the longer storage time in solvents caused the remaining monomer numbers to increase¹¹. The aim of the study was to know the difference in the number of residual monomers in the sculptable and flowable bulk-fill composite resins under immersion in 75% ethanol solution for 10 minutes, 1 hour, and 24 hours.

Materials and Methods

This was a laboratory experimental research with post-test only group design.

1. Sample Preparation

A glass-lab (Duran, Indonesia) with a size of 15x30 cm and 5 mm thickness was put at the bottom of the mold. Bulk-fill sculptable (Tetric N-Ceram Bulk-Fill, Ivoclar, Vivadent, Liechtenstein) and flowable composite (Tetric N-Ceram Bulk-Fill flowable (Ivoclar, Vivadent, Liechtenstein) were inserted into the mold using plastic filling instrument until full, then condensed to avoid air-trapping and then flatten the top and given a transparent plastic that functions as an oxygen inhibiting layer and weight of 0.25 kg on the top. The intensity of Light Emitted Diode (LED) (DBA Woodpecker, China) light cure was confirmed with a light meter tool (DBA Woodpecker, China), then samples were cured for 10 seconds with radiation intensity of 1025 mW *I* cm2.

The composite discs were removed from the mold, and then the thickness was checked using a caliper (Nankai, China). Samples then were submerged in 75% ethanol solution (Ensure CAS 64-17-5, Germany) in an amber-colored bottle (Duran, Indonesia) and then 7 cc of the solution were taken with 10 cc syringe at 10 minutes (Group 1 = TI), 1 hour (Group 2 = T2) and 1 day (Grop 3 = T3).

2. Sample Analysis

Standard solutions of TEGDMA, UDMA, and BIS-GMA were examined using High-Performance Liquid Chromatography (HPLC) (Agilent 1100 series, Korea). Each standard solution TEGDMA, UDMA, and BIS-GMA (Aldrich chemistry, United States of America) was weighed using milligram scale (Mattler Toledo, Singapore) then dissolved in solvents, respectively, standard TEGDMA and UDMA were dissolved in methanol, and standard BIS-GMA was dissolved in chloroform (CHCb) (Emsure CAS 65-66-3, Germany). Each standard solution was placed on an ultrasonic machine (ELMA, China) for 5 minutes, then was chilled to room temperature and was calculated for the concentration using the formula:

$standard \ solution \ concentration = \frac{standard \ solution \ weight}{standard \ solution \ volume} \times 1000$

The next step was the dilution of a standard solution of 100 μ l into 1000 μ l solvents, respectively, in the HPLC vial. Dilution aimed to decline the peak area of the standard solution, preventing it from being too far from the sample area. Calculation the concentration of a standard solution was using the formula:

$$standard \ solution \ concentration = \frac{100 \mu l \ standard \ solution}{1000 \mu l \ solvent \ solution} \ x \ concentration \ 1$$

note: Concentration 1 = Concentration of standard solution before dilution

Each standard solution was taken as much as 1 ml to be transferred to the HPLC vial. TEGDMA standard solution examination was done by injecting a standard solution for 20μ l, resulting in a retention time peak of 2,153 minutes. The same injection volume was also done for UDMA, resulting in a retention time peak

of 2.768 minutes. Finally, for the standard solution of the Bis-GMA examination, the retention time peak was obtained at 10,219 minutes.

The next step was an examination of samples in HPLC. Each sample solution was taken for 1ml, then transferred to the HPLC vial. The sample solution was injected for 20 μ l and peak obtained was recorded. The peak of residual monomers was confirmed if the

peak was at retention time \pm 0.30 minutes from the standard peak. Percentage of residual monomers was calculated using the formula:

$$Residual \ monomers = \frac{sample \ area}{standard \ solution \ area} \times standard \ solution \ concentration \ \times 100\%$$

3. Statistical Analysis

The normality test is carried out with the Kolmogorov-Smirnov test. The homogeneity of the variance of data between groups, the Levene test was used. The Kruskal-Wallis test was done to analyze the differences between the groups. The differences between the experimental groups, an analysis was carried out with the Independent T-test and the Mann-Whitney test ($p \le 0.05$).

Results

Group means and standard deviation can be seen in Table 1 to Table 3.

Table 1. Mean and standard deviation of residual monomers in 3 different submersing times of sculptable bulk-fill composite, Mann-Whitney test.

Groups	N	$X \pm SD$			n voluo
		TEGDMA	UDMA	BIS-GMA	p-value
T1	5	$1,390 \pm 0,004a$	5,616 ± 1,225a	$0,130 \pm 0,048a$	
T2	5	$1,408 \pm 0,086a$	$8,302 \pm 2.270b$	0,145± 0,104a	< 0,05
Т3	5	$3,119 \pm 1,301b$	$15,201 \pm 1,625c$	$0,810 \pm 0,096b$	

Note: Words printed in superscript show significant differences within the same column with Mann-Whitney test.

Tl: Flowable bulk-fill composite submersed for 10 minutes

T2: Flowable bulk-fill composite submersed for 1 hour

T3: Flowable bulk-fill composite submersed for 24 hours

X: Mean

SD: Standard deviation

Mean and standard deviation of residual monomers expressing TEGDMA, UDMA, and BIS-GMA in sculptable bulk-fill composite represented by a graph in Figure 1.



Figure I. Diagram of mean and SD residual monomer that expressed TEGDMA, UDMA and bis-GMA in a bulk fill sculptable composite resin.

From the table above (Table 1), it can be seen that a 10-minutes sculptable TEGDMA has no significant difference with a 1-hour sculptable TEGDMA, but there was a significant difference with a 24-hours sculptable TEGDMA. In 1-hour sculptable TEGDMA, there was a significant difference with 24-hour sculptable TEGDMA. In a 10-minutes sculptable UDMA, there was a significant difference with a 1-hour and 24-hours sculptable UDMA, while in a 1-hour sculptable UDMA there was a significant difference with a 24-hours sculptable UDMA. In a 10-minutes sculptable Bis-GMA, there was no significant difference with a 1-hour sculptable Bis-GMA, but there was a significant difference with a 24-hours sculptable Bis-GMA. In a 1-hour sculptable Bis-GMA, there was a significant difference with a 24-hours sculptable Bis-GMA.

 Table 2. Mean, and standard deviation number of residual monomers of flowable bulk-fill composite in three different submersion time, Mann- Whitney test

Groups	Ν		n voluo		
		TEGDMA	UDMA	BIS-GMA	p-value
T1	5	$1,430 \pm 0,093a$	7,932 ± 1,729a	0,198 ± 0,145a	
T2	5	$1,447 \pm 0,282a$	$10,604 \pm 1,429b$	0,238 ± 0,095a	< 0,05
Т3	5	$3,773 \pm 1,293b$	$18,243 \pm 4,495c$	$1,417 \pm 1,183b$	

Note: Words printed in superscript show significant differences within the same column with Mann-Whitney test.

Tl: Flowable bulk-fill composite submersed for 10 minutes

X: Mean

SD. Standard deviation

T2: Flowable bulk-fill composite submersed for l hour

T3: Flowable bulk-fill composite submersed for 24 hours

Mean and standard deviation of residual monomers expressing TEGDMA, UDMA, and BIS-GMA in flowable bulk-fill composite represented by a graph in Figure 2.



Figure 2. Means of residual monomers expressed by TEGDMA, UDMA, and BIS GMA in a flowable bulkfill composite.

From the table above (Table 2, it can be seen that the 10-minutes flowable TEGDMA has no significant difference with a 1-hour flowable TEGDMA, but there was a significant difference with a 24-hours flowable TEGDMA. In a 1-hour flowable TEGDMA, there was a significant difference with a 24-hours flowable TEGDMA. In 10-minutes flowable UDMA, there was no significant difference with a 1-hour flowable UDMA, but there was a significant difference with a 24-hours flowable UDMA. In 1-hour flowable UDMA, there was a significant difference with 24-hours flowable UDMA. In the 10-minutes flowable Bis-GMA, there was no significant difference both with 1-hour flowable Bis-GMA and 24-hours flowable Bis-GMA. In a 1-hour flowable Bis-GMA, there was no significant difference with a 24-hours flowable Bis-GMA.

Correct	N	T1	Τ2	Т3	p-value
Group		$X \pm SD$	$X \pm SD$	$X \pm SD$	
TEGDMA Sc TEGDMA Fl	5 5	$1,390 \pm 0,004^{a}$ $1,430 \pm 0,093^{a}$	$\begin{array}{c} 1,408 \pm 0,086^{a} \\ 1,447 \pm 0,282^{a} \end{array}$	$3,119 \pm 1,301^{a}$ $3,773 \pm 1,293^{a}$	
UDMA Sc UDMA Fl	5 5	5,616 ± 1,225 ^a 7,932 ± 1,729 ^b	$8,302 \pm 2.270^{a}$ $10,604 \pm 1,429^{a}$	$15,201 \pm 1,625^{a}$ $18,243 \pm 4,495^{a}$	P < 0,05
Bis-GMA Sc Bis-GMA Fl	5 5	$\begin{array}{c} 0,130 \pm 0,048^{a} \\ 0,198 \pm 0,145^{a} \end{array}$	$\begin{array}{c} 0,145\pm 0,104^{a}\\ 0,238\pm 0,095^{a} \end{array}$	$\begin{array}{c} 0,810 \pm 0,096^{a} \\ 1,417 \pm 1,183^{a} \end{array}$	

 Table 3. Means and standard deviation number of residual monomers of bulk-fill composite by same submersion time, Independent T-test.

Note: Words printed in superscript show significant differences within the same cell with Independent T-test

Tl: Submersed for 10 minutes

T2: Submersed for I hour

T3: Submersed for 24 hours

Sc: Sculptable bulk-fill composite

Fl: Flowable bulk-fill composite

X: Mean

SD: Standard deviation

The mean and standard deviation of the residual monomer graphics that express differences between groups of similar residual monomers in the immersion time of 75% ethanol solution for 10 minutes can be seen in the diagram below (Figure 3).



Figure 3. A diagram of the average amount of residual monomers bulk-fill sculptable composites resin and bulk fill composites resin flowable (a) TEGDMA, (b) UDMA, and (c) bis-gMA in soaking 75% ethanol solution for 10 minutes.

The mean and standard deviation graphic of residual monomers expressed the differences between groups of similar residual monomers in the immersion time of 75% ethanol solution for 1 hour can be seen in the diagram below (Figure 4).



Figure 4. A diagram of the average amount of residual monomers bulk fill sculptable composites resin and bulk fill composites resin flowable (a) TEGDMA, (b) UDMA, and (c) bis-gMA in soaking 75% ethanol solution for 1 hour.

The mean and standard deviation graphic of residual monomers expressed the differences between groups of similar residual monomers in the immersion time of 75% ethanol solution for 24 hours can be seen in the diagram below (Figure 5).



Figure 5. A diagram of the Means of residual monomers of flowable and sculptable composite (a) TEGDMA, (b) UDMA, (c) bis-GMA submerged in ethanol 75% solution for 24 hours.

From the table above (Table 3), it can be seen that 10-minutes sculptable TEGDMA has no significant difference with 10-minutes flowable TEGDMA. There was no significant difference between 1-hour sculptable TEGDMA and 1-hour flowable TEGDMA, and also, there was no significant difference between 24-hours sculptable TEGDMA and 24-hours flowable TEGDMA. In a 10-minutes sculptable UDMA, there was a significant difference with a 10-minutes flowable UDMA. While there was no significant difference between 1-hour sculptable UDMA and 1 hour flowable UDMA, and there was no significant difference between 24-hours sculptable UDMA and 24-hours flowable UDMA. In the 10-minutes sculptable Bis-GMA there was no significant difference with 10-minutes flowable Bis-GMA. There was no significant difference between 1-hour sculptable Bis-GMA and 1-hour flowable Bis-GMA, and also there was no significant difference between 24-hours sculptable Bis-GMA and 24 hours flowable Bis-GMA.

Discussion

Optimal polymerization is an important factor to obtain both physical and mechanical properties and optimal clinical performance. Adequate radiation intensity and exposure time affect the degree of polymerization. The success of the polymerization depends on the filler, light intensity, and the irradiation time^{13,14}. Polymerization of the composite resin occurs through monomers transformation into polymers, followed by polymerization shrinkage¹⁵.

There were various solvents used to elute residual monomers, including artificial saliva, distilled water, methanol, and acetonitrile which already used in the study to investigate monomer elution. Previous studies showed that the type of solvent affected the number of residual monomers eluted from resin composite^{16,17}. Ethanol 75% solution is recommended by FDA as food-oral simulating liquid due to its relevancy, and therefore already used in several studies ¹¹. According to Sideridou and Achilias, Bis-GMA, TEGDMA, UDMA, and bis-EMA monomers were able to dissolve

in ethanol 75% solution due to its ability to penetrate resin composite matrix, followed by expansion and enlargement of polymer bonds which were consisted of monomers, causing residual monomers to be released. Therefore, ethanol 75% solution was chosen as a solvent in this study¹.

High molecular weight monomers such as Bis-GMA and UDMA will decompose in gas chromatography examination, and only decomposition products can be detected, However monomers with a low molecular weight such as TEGDMA will be difficult to be detected by gas chromatography examination¹⁸. For this reason, the quality and quantity of the residual monomers eluted from composite resin materials were usually determined by examination using HPLC because it was a very potent and commonly used separation method. HPLC was preferred over gas chromatography because it provided a greater degree of control in the separation process, due to the dissolved monomers in the mobile phase¹⁹. Therefore, the HPLC examination was used in this study to evaluate the release of monomers from bulk-fill resin material.

From this study, it was found that in sculptable and flowable bulk-fill composites, residual monomers were detected at 10 minutes, 1 hour, and 24 hours. More monomers of the remaining flowable bulk-fill composite were detected compared to the residual monomers of a sculptable bulk-fill composite. This result was in accordance with previous studies conducted by Cebe et al. (2015), which stated that after polymerization of bulk-fill resin composite, elution of residual monomers was found in solution. Polymer networks that were formed during the initial polymerization due to free radical reactions were densely crosslinked (microgels) and some other parts are loosely crosslinked (micropore), which contained non-reacting monomers¹¹. The polymer network morphology takes the form of agglomerating microgels into clusters and these clusters were interconnected ²⁰. These polymer networks almost always contain a number of unreacted monomers, usually trapped inside microgels and micropores which cluster between polymer chains. Polymer crosslinking is generally insoluble but has a tendency to expand in a good solvent. The solvent penetrates

into the composite matrix and expands the space between the polymer chains, which will cause changes in mass and dimensions. If the monomers were not dissolved in the solvent, they could be removed from the polymer mass. The process of enlarging polymer crosslinking and the process of removing unreacted monomers occur due to the diffusion process. Solvents can penetrate into the composite matrix and expand the opening of the polymer chain, causing unreacted monomers to dissolve and diffuse out. It was reported that monomers trapped in micropore were more susceptible to diffusion than monomers in microgels and it was known that the volume of micropore is very large in heterogeneous polymer nentworks¹. The majority of sculptable bulk-fill composites contain monomer Bis-GMA, which comes from a mixture of bisphenol A with glycidyl methacrylate. Bis-GMA has a very high molecular weight, therefore the shape of the monomer is very thick because of its high viscosity. The majority of flowable bulk-fill composites contain TEGDMA monomers. TEGDMA has a very low molecular weight, therefore the shape of the monomer is very flowable because of its low viscosity. These are the reasons why the residual monomer of flowable bulk-fill composites was more than the sculptable bulk-fill composites.

The results showed that in TEGDMA sculptable, Bis-GMA sculptable, TEGDMA flowable, and UDMA flowable groups at 10 minutes submersion compared to 1 hour submersion had no significant differences in the number of residual monomers. However if 10 minutes submersion was compared to 24 hours submersion, it showed a significant difference in the number of residual monomers, as well as if 1 hour submersion compared to 24 hours submersion showed a significant difference. This indicates that there were more residual monomers released in the span of 1 hour to 24 hours than in 10 minutes to 1 hour and the longer the submersion time in 75% ethanol solution, the more residual monomers released. This happened because of the accumulation time, the time between 10 minutes and 1 hour was shorter than the time between 1 hour and 24 hours. Another reason was that the ability to penetrate the ethanol solution took more than 1 hour. The swelling ability to crosslink polymer and the process of releasing unreacted monomers by the diffusion process took more than 1 hour.

In the analysis of the data of this study, it was also shown that the UDMA sculptable group at 10 minutes submersion compared to 1 hour submersion had a significant difference in the number of residual monomers, this also occurred in 10 minutes submersion compared to 24 hours submersion and I hour submersion compared to 24 hours submersion. This means that from the beginning of submersion there were already many residual monomers diffused out, this happened because the residual UDMA monomers had morphology with high crosslinking density. The higher the density of the crosslinking, the higher the heterogeneity and the greater the volume of the micropores. It was reported that monomers trapped in micropores were more susceptible to diffusion than monomers in microgels, causing UDMA to diffuse more. UDMA also have weaker bonds between monomers, this resulted in the solvent penetrating the composite matrix easy to expand and release the bonds between the monomers to make them dissolve and diffuse more. UDMA also has an affinity between monomers and 75% ethanol solution, therefore when ethanol solution penetrated, the intermonomer bond of UDMA broke and directly bind with 75% ethanol solution because of its high affinity for 75% ethanol solution, therefore, the number of residual UDMA monomers released were more than TEGDMA and Bis-GMA.

Data analysis showed that the flowable Bis-GMA group at 10 minutes submersion compared to 1 hour submersion showed no significant difference in the number of residual monomers, this also occurs in 10 minutes submersion compared to 24 hours submersion and 1 hour submersion compared to 24 hours submersion. This means that less residual monomer released, therefore, there is no difference between submersion with 75% ethanol solution in 10 minutes, 1 hour and 24 hours. This happened because of the site and chemical structure characteristics of the BIS-GMA monomer, the molecular size of BIS-GMA was very large (molecular weight 512 g/mol)^{2,3}. The Bis GMA molecule is a very compact resin therefore the monomer shape is very thick due to its high viscosity, structurally, those molecules consist of several monomer molecules chemically bonded to each other after the polymerization process. Bis-GMA also has a strong hydrogen bonding ability, hence the bond is not easily released even though it is penetrated by 75% ethanol solution. The remaining monomer molecules can be physically described as trapped/sandwiched molecules between the arrangement of the molecules of the Bis-GMA and making it difficult to release.

In this study, it was also found that in general, the number of residual monomers increased with increasing submersion time in 75% ethanol solution. Composite resins consist of polymers containing unreacted monomers and trapped inside. The solvent penetrated into polymer networks and extended the space between the polymer chains, causing unreacted monomers to elute¹. The longer the composite submersed in 75% ethanol solution, the more space between the polymer chain and the more residual monomers diffused out. Previous research had shown that the number of eluted monomers increased with increasing submersion time^{21,22}. In accordance with these findings, the highest number of monomers was detected after 24 hours of submersion of 75% ethanol solution.

Conclusion

From the results of this research, it can be concluded that (1) the amount of residual monomer of flowable bulk-fill composite resin was greater than the sculptable bulk-fill composite resin on submersion in 75% ethanol solution for 10 minutes, 1 hour and 24 hours². The number of residual monomers in sculptable bulk-fill composite resin at submersion in 75% ethanol solution for 24 hours was greater compared to 1 hour and 10 minutes submersion time³. The number of residual monomers in flowable bulk-fill composite resin at submersion time³. The number of residual monomers in flowable bulk-fill composite resin at submersion in 75% ethanol solution for 24 hours was greater compared to 1 hour and 10 minutes in flowable bulk-fill composite resin at submersion in 75% ethanol solution for 24 hours was greater compared to 1 hour and 10 minutes in flowable bulk-fill composite resin at submersion in 75% ethanol solution for 24 hours was greater compared to 1 hour and 10 minutes in flowable bulk-fill composite resin at submersion in 75% ethanol solution for 24 hours was greater compared to 1 hour and 10 minutes in flowable bulk-fill composite resin at submersion in 75% ethanol solution for 24 hours was greater compared to 1 hour and 10 minutes submersion for 24 hours was greater compared to 1 hour and 10 minutes submersion for 24 hours was greater compared to 1 hour and 10 minutes submersion for 24 hours was greater compared to 1 hour and 10 minutes submersion time.

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