CAN DIFFERENT POLYMERIZATION TIMES AFFECT THE SURFACE MICROHARDNESS, WATER SORPTION, AND WATER SOLUBILITY OF FLOWABLE COMPOSITE RESINS?

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Abstract

This in vitro study evaluated and compared the effects of different polymerization times on the surface microhardness, water sorption, and water solubility of flowable composite resins. Three flowable composite resins [Es Flow (ESF), IGOS Flow (IGF), Estelite Flow Quick (EFQ)] were tested in this study. Each flowable composite resin ($n = 7$) was polymerized in a disc-shaped mould (1x10 mm) with an LED light-curing unit (D Light Pro) for two different times (20 and 40 sec.). The top surfaces of all specimens were polished (Sof-Lex). The surface microhardnesses of the flowable composite resins were measured with a Vickers HMV microhardness tester. Water sorption and water solubility were calculated according to the ISO 4049 standard. One-way ANOVA and post hoc Tamhane, Dunnett, and Tukey tests were used in the statistical analyses. Pearson's and Spearman's rho correlation tests were used to assess possible correlations between the different variables. The results were evaluated with a significance of $p<0.05$. In terms of microhardness, a significant difference was found between materials at the same polymerization times ($p<0.05$). All materials showed water sorption of less than 40 µg/mm^3 and water solubility of less than 7.5 µg/mm^3 by following ISO 4049. The correlations among surface microhardness, water sorption, and water solubility showed that the differences were determined by the materials and the polymerization times. The physical properties of all flowable composite resin materials were enhanced after polymerization for twice the time recommended by the manufacturers.

Keywords: Flowable composite. Hardness. Solubility. Viscosity.

1. Introduction

Flowable composite resins (FCRs) are restorative materials containing the same small particle sizes as conventional hybrid composites with reduced filler contents and increased resins to lower the viscosity of the mixture (Bayne et al. 1998). When FCRs were first introduced in 1996 (Hervás-García et al. 2006), they initially had inferior physical properties due to the lower filler contents (Baroudi and Rodriguez 2015). To overcome this disadvantage and improve the physical properties, novel FCRs were developed in 2000 (Salerno et al. 2011). Therefore, new-generation FCRs, with increased filler contents, were launched for use as permanent restorative materials for different indications (Zhang et al. 2021). FCRs are ideal for minimally invasive restorations of occlusal Class I and Class III cavities (Bayne et al. 1998), interproximal Class II cavities (Payne 1999), non-carious Class V cavities (Kubo et al. 2010), pits, and fissures (Beauchamp

et al. 2008). These materials can be used as cavity liners (Baroudi and Rodriguez 2015) under composite resin materials to relieve stresses at the gingival margins. In addition, FCRs are preferred for the repair of amalgam (Roberts et al. 2001) crowns and composite restoration margins (Salerno et al. 2011).

The physical properties determine the long-term performance of the dental composites in the oral cavity. One of the restorative material's most important physical properties is surface microhardness, which is related to compressive strength and abrasion resistance (Uslu Cender and Guler 2018). Low water sorption and water solubility are the other features of composite resins serving as restorative materials in the oral environment; in such an environment, the resin-based materials may absorb water or other substances such as saliva, food components or beverages, which can have an important influence on the degradation of the restorative materials (Ferracane 2006).

The physical properties of a restorative material are closely related to the structure of the material. The FCR formulation includes a high proportion of diluent monomers (Cadenaro et al. 2009) with a filler loading of 37–53% by volume. Less inorganic filler content in a composite resin material reduces the surface microhardness of the flowable composite resin. In addition, the high monomer content increases polymerization shrinkage. Each basic monomer, such as Bis-EMA, Bis-GMA, TEGDMA, and UDMA, has a very different degree of conversion, modulus, viscosity, water sorption, and solubility (Sideridou et al. 2003).

The properties of resin-based materials may also be influenced by the polymerization time. A longer polymerization time may result in the formation of more polymeric chains, which produces better features for restorative materials (Alpoz et al. 2008).

As FCRs meet clinicians' expectations and provide more indications during clinical applications, little is known about the physical properties of the flowable composites when the manufacturer's recommended time is doubled. Therefore, the purpose of this in vitro study was to evaluate and compare the effects of different polymerization times on the surface microhardness (VHN), water sorption (WSP), and water solubility (WSL) of flowable composite resins. Hypothesis: 1-The first null hypothesis of this study is that polymerization of flowable composite resins for longer than the recommended time will not improve the surface microhardnesses; 2-The second null hypothesis of this study is that polymerization of flowable composite resins for longer than the recommended time will not improve water sorption; 3-The third null hypothesis of this study is that polymerization of flowable composite resins for longer than the recommended time will not improve the water solubility.

2. Material and Methods

Material development

Three FCRs [EsFlow (ESF), Spident, Korea; IGOS Flow (IGF), Yamakin, Japan; Estelite Flow Quick (EFQ), Tokuyama, Japan] with different viscosities (low, medium, and high) were tested in this study. Seven specimens from each group were prepared (Lassila et al. 2019). The compositions of the tested materials are listed in Table 1.

Preparation of the specimens

In this study, two different polymerization times, 20 and 40 seconds were used during the polymerizations of the disc-shaped samples (*n* = 7). Each FCR material was placed in a disc-shaped acrylic mould (1 x 10 mm), and these moulds were placed between two Mylar strips with two glasses to obtain a flat surface. The specimens were light-cured with an LED light-curing unit (D Light Pro, GC, Belgium) positioned on the top surface with an output of 700 mW/cm2 and a wavelength of 400-480 nanometres. The light intensity was measured with a radiometer (LED Radiometer, SDI, Australia) to provide standardization. Attention was given to touching the tip of the light-curing unit to the surface of the glass. The experimental groups are shown in Table 2.

Table 1. The composition and the inorganic filler content of the experimental materials used in the study.

Material	Manufacturer	Composition	Inorganic Filler Content (vol%)	Lot Number
Estelite Flow Quick (EFQ) (Low viscosity submicron filled flowable resin composite) (A2)	Tokuyama, Tokyo, Japan	Bis-MPEPP, TEGDMA, UDMA, RAP initiator, silica-zirconia filler (supra-nano spherical filler) (mean particle size: 0.3 μm)	53	E517
Es Flow (ESF) (Medium viscosity nano-hybrid flowable resin composite) (A2)	Spident, Korea	UDMA, barium glass fillers (The initiator is not specified by the manufacturer)	50-55	E2F17049 E2F17055
IGOS Flow (IGF) (High viscosity resin-based flowable resin composite) (A2)	Yamakin, Japan	Methacrylate monomer, silica, alumina, zirconia (particle diameter <20μm) (The initiator is not specified by the manufacturer)	42	01051725

Abbreviations: Bis-MPEPP, bisphenol A polyethoxy methacrylate; TEGDMA, triethyleneglycol dimethacrylate; UDMA, urethane dimethacrylate; RAP, Radical Amplified Photopolymerization.

Table 2. Application thickness and different polymerization times for the experimental groups.

Experimental Groups	Application thickness with different polymerization times
EFQ-20	Estelite Flow Quick placed at 1 mm thick and light-cured for 20 sec.
EFQ-40	Estelite Flow Quick placed at 1 mm thick and light-cured for 40 sec.
ESF-20	Es Flow placed at 1 mm thick and light-cured for 20 sec.
ESF-40	Es Flow placed at 1 mm thick and light-cured for 40 sec.
IGF-20	IGOS Flow placed at 1 mm thick and light-cured for 20 sec.
IGF-40	IGOS Flow placed at 1 mm thick and light-cured for 40 sec.

The top surfaces of all specimens were polished with polishing discs (Sof-Lex, 3M ESPE, USA) from coarse to fine for 10 seconds (duration of each disc) with an underwater coolant. No press-on force was applied to the samples. Polishing was simulated as if working on a patient. After every five specimens, the discs were replaced.

Measurements of surface microhardness

The VHNs of the FCRs were measured with a Vickers HMV microhardness tester (Shimadzu, Japan). A 1.961 N weight was applied to the top surface of each sample for 10 seconds and from different points. Measurements were taken from three points on each sample, and the average value was calculated (Chinelatti et al. 2006).

Measurements of water sorption and water solubility

The specimens were kept in dark-coloured glass bottles on which the caps were not completely closed, placed in a desiccator (Vacucell, MMM, Germany) and kept at 37 °C ± 1 °C inside a vacuum oven for 22 hours. After that, the bottles were removed from the oven and left on a bench for two hours at a temperature of 23 °C ± 1 °C to complete a 24-hour cycle. Later, the specimens were weighed daily with an analytical balance (Precisa, ES 225SM-DR, Switzerland) to record the 24-hour weighing cycles. The complete cycle was repeated every day at the same time until a constant (the loss for each specimen was not more than 0.1 mg per 24-hour cycle) mass (M1) was obtained. The diameter and thickness of each specimen were measured three times with a calliper to calculate the specimen's volume (V) in mm^3. Thereafter, the specimens were placed back in the dark-coloured glass bottles. Distilled water (20 ml) was added to the specimens with manual pipettes. The glass bottles were then sealed, returned to the oven, and kept at 37 °C ± 1 °C for 7 days (Stuart Orbital Incubator SI500, Bibby Scientific Ltd., UK). After these procedures, all the glass bottles were removed from the oven and kept at 23 °C ± 1 °C for 2 hours. The specimens were removed from the bottles, dried with absorbent paper for 15 seconds, and left in a sterile

bucket for 1 minute. The identical specimens were weighed again to obtain M2. Afterwards, the specimens were reconditioned in the desiccator until they reached a constant mass (M3) with the cycle described for M1.

The WSP and WSL values of the specimens were calculated as $\mu g/mm^3$ according to the formula specified in ISO 4049 (ISO-Standards 2009).

WSP = (M2 - M3)/V

WSL = (M1 - M3)/V

Statistical analyses

In calculating the sample sizes, the probability of type 1 error (α=0.05) and the power of the test (1-β) were considered to be 0.95. Using the GPower 3.1.9.2 program, it was calculated that the total sample size should be at least 7. Therefore, the sample size used was 7.

The statistical analyses were performed by one-way ANOVA and post hoc Tamhane tests for VHN, Dunnett's test for WSL, and a post hoc Tukey test for WSP. Pearson's and Spearman's rho correlation tests were performed to assess possible correlations between surface microhardness and water solubility, surface microhardness and water sorption, and water sorption and water solubility. The results were evaluated with a significance of $p<0.05$.

3. Results

Surface microhardness

When the VHNs of the FCRs were compared according to the different polymerization times, no statistically significant difference was observed ($p>0.05$), while a significant difference was found between materials subjected to the same polymerization times ($p<0.05$). The VHN numerical values of the experimental materials treated with each polymerization time increased in the order ESF < IGF < EFQ (Table 3).

Table 3. Data obtained from the microhardness process of flowable composites according to different polymerization times and statistical evaluation of the experimental materials. (mean±standard deviation, *n = 7*).

Experimental Material	Microhardness (VHN) Different Polymerization Times	
	20 sec.	40 sec.
EFQ	79.70±9.60 Aa	83.40±7.26 Aa
ESF	27.83±2.90 Ba	29.50±2.52 Ba
IGF	47.60±5.23 Ca	52.10±5.22 Ca

Statistical differences between the experimental materials indicated in uppercase letters and statistical differences between polymerization times indicated in lowercase letters ($p<0.05$), One-Way ANOVA Test.

Water sorption

When the WSP values of the FCRs were evaluated for the different polymerization times, there were no statistically significant differences for EFQ and IGF ($p>0.05$), but a statistically significant difference was calculated for ESF ($p<0.05$). The ESF polymerized for 40 seconds showed a lower WSP value (3.16 ± 0.52) than those polymerized for 20 seconds (4.56 ± 0.62). A significant difference was detected between the materials at both polymerization times ($p<0.05$). The WSP values for the experimental materials increased in the order ESF < EFQ < IGF for each polymerization time (Table 4).

Water solubility

When the WSL of the FCRs were compared with each other for the different polymerization times, there were no statistically significant differences for ESF and IGF (*p*>0.05), but a statistically significant difference was found for the EFQ (*p*<0.05). A statistically significant difference was found between EFQ and ESF (*p*<0.05), while no statistically significant difference was evaluated between the IGF and other experimental materials at both polymerization times (*p*>0.05). The WSL values of the experimental materials increased as ESF < IGF < EFQ for each polymerization time (Table 5).

Correlations between surface microhardness and water solubility, surface microhardness and water sorption, and water sorption and water solubility

For the VHN and WSP properties, a negative correlation was found for IGF-40 (*r* = -0.773, *p* = 0.042). For the WSP and WSL properties, positive correlations were found for IGF-40 (*r* = 0.856, *p* = 0.014) and EFQ-20 (*r* = 0.961, *p* = 0.001).

Table 4. Data obtained from the water sorption process of flowable composites according to different polymerization times and statistical evaluation of the experimental materials. (mean±standard deviation, *n* = 7).

Experimental Material	Water Sorption (μg/mm^3)* Different Polymerization Times	
	20 sec.	40 sec.
EFQ	5.95±0.90 Aa	5.86±0.51 Aa
ESF	4.56±0.62 Ba	3.16±0.52 Bb
IGF	12.95±0.43 Ca	12.21±1.04 Ca

Statistical differences between the experimental materials indicated in uppercase letters and statistical differences between polymerization times indicated in lowercase letters (*p*<0.05), One-Way ANOVA Test.

Table 5. Data obtained from the water solubility process of flowable composites according to different polymerization times and statistical evaluation of the experimental materials. (mean±standard deviation, *n* = 7).

Experimental Material	Water Solubility (μg/mm^3)** Different Polymerization Times	
	20 sec.	40 sec.
EFQ	1.08±0.65 Aa	0.24±0.36 Ab
ESF	-5.62±2.5 Ba	-6.95±2.13 Ba
IGF	-0.75±1.83 ABa	-2.77±2.51 ABa

Statistical differences between the experimental materials indicated in uppercase letters and statistical differences between polymerization times indicated in lowercase letters (*p*<0.05), One-Way ANOVA Test.

4. Discussion

The durability of restorative material in the oral environment is crucial for the longevity of the restoration. The surface microhardness, water sorption, and water solubility of a restorative material are related to durability and significantly affect clinical success.

Although the increased polymerization time caused an improvement in the microhardness, the lack of a statistically significant difference supports the first null hypothesis. Therefore, it was accepted. For the water sorption results, the increased polymerization time did not affect the water sorption values except for that of the ESF material. When the ESF material was polymerized for a longer time (40 sec.), the water sorption value decreased, and a statistically significant difference was found when compared with the recommended polymerization time of the manufacturer (*p*<0.05). Therefore, the second hypothesis was partially accepted. When the water solubility results were evaluated, the increased polymerization time did not statistically affect the solubilities of other materials (*p*>0.05) except the EFQ material (*p*<0.05). Therefore, the third hypothesis was partially accepted.

Mirică et al. (2020) stated that the percentage of inorganic filler was correlated with the physical properties of FCRs. Aung et al. (2021) evaluated the microhardnesses and degrees of conversion for newly developed FCRs. They found that EFQ exhibited a significantly higher hardness when compared with MI FIL

Flow, Estelite Universal Flow, Beautifil Flow Plus, Clearfil Majesty ES Flow, Filtek Supreme Ultra Flowable, and TetricEvo Flow. This result was related to the supra-nano spherical fillers in the EFQ, which resulted in superior matrix filler interaction. In line with the findings of the present study, the EFQ containing one of the higher inorganic filler contents (53% vol) showed the highest microhardness among the materials tested. Chandru et al. (2020) showed that an increase in the polymerization time increased the microhardness of the restorative material. Lima et al. (2012) evaluated the effects of extended curing times (20 and 40 seconds) on the microhardness of a nanofilled composite resin. They found that a longer curing time (40 seconds) increased the microhardness. They speculated that the extended light-curing exposure could have regulated the polymeric formation, leading to a composite with more crosslinked chains and, consequently, higher microhardness and better physical properties. In the present study, all the FCRs showed higher VHN values as the polymerization time was increased, but no statistically significant difference was observed for the different polymerization times ($p>0.05$). Since the samples were prepared at a thickness of 1 mm, it was thought that the material may have polymerized optimally with a 20-second polymerization time. Therefore, the microhardnesses of the materials polymerized for the recommended time (20 sec.) were like the microhardnesses of the material polymerized for 40 seconds. Consequently, the microhardness values of the materials were not affected when the polymerization time was doubled.

Light-curing units in the presence of initiators cure the resin-based materials. The amount of initiator varied from 0.1% to 1% by weight, and this small part of the composition had a considerable influence on the biophysical properties of the material (Kowalska et al. 2021). Unlike the other FCRs, the EFQ contained a radical amplified photopolymerization initiator (RAP technology). Although the ESF had a similar inorganic content (50–55%) to the EFQ, this material exhibited the lowest VHNs at different polymerization times. Unfortunately, the initiator system in the ESF composition was not disclosed. Therefore, one of the significant differences between the VHNs of the EFQ and ESF ($p< 0.05$) can be attributed to the RAP initiator included in the EFQ. Ilie et al. (2014) concluded that the RAP-initiated restorative material demonstrated a higher increase in the degree of conversion with prolonged irradiation time than the CQ/amine-based materials. Generally, a higher surface microhardness is attributed to a higher filler content (Ku et al. 2015). As with Jager et al. (2016), the authors agree that the filler content should not always be seen as the main influencing parameter.

Sorption and solubility values are expected to be correlated since the liquid needs to penetrate the material to leach out the unreacted components. The degree of conversion and crosslinking density are the major factors leading to the correlation between these properties (Floyd and Dickens 2006).

Water sorption is a diffusion-controlled, time-dependent process (Wei et al. 2011), which may decrease the lifespan of the restoration by expanding and plasticizing the resin component and hydrolysing the silane (Ferracane 2006; Barutcigil and Yildiz 2012).

The solubility of a resin composite material is related to the leachability of the unreacted or residual monomers. The increased conversion of monomers to polymers leads to lower solubility with a higher percentage of carbon-carbon bonds. Therefore, the solubility depends mainly on the degree of conversion (Barutcigil and Yildiz 2012). The molecular size of the monomer is another important factor for the decomposition of the residual monomers. Lighter monomers can be decomposed in greater amounts than heavier monomers (Ferracane 1994).

In the present study, all the materials exhibited sorption values of less than 40 µg/mm^3 and solubilities of less than 7.5 µg/mm^3 in the ISO 4049 tests (ISO-Standards 2009). Moreover, it was found that as the polymerization time was increased, the sorption and solubility values of all FCRs decreased.

The ESF showed the lowest sorption value among the materials, and a statistically significant difference was observed between the sorption values of the ESF treated with different polymerization times ($p<0.05$). The water sorption values of a restorative material differ according to the monomer type (Sideridou et al. 2011). UDMA creates more rigid networks than TEGDMA and absorbs less water (Soderholm et al. 1984). One of the reasons for the low water sorption value of the ESF material could be the UDMA content, which was declared by the manufacturer to be the main monomer of the material. Tarumi et al. (1995) stated that the water resistance of composite resin materials containing barium glass fillers was low, but they also emphasized that the amount of the barium glass filler must have a significant

influence on the sorption and physical properties of the composite resin after soaking in water. They stated that the amount of sorbed water obtained by the barium glass-filled resin increased as the particle size decreased. They suggested that the average particle size should be more than 10 µm in diameter. Therefore, although not specified by the manufacturer, the barium glass filler particle size of the ESF should be more than 10 µm in diameter to achieve the lowest water sorption value. It was also reported that (Yap et al. 2000) the water sorption and contact time in the aqueous medium significantly affected the microhardness of restorative material. The ESF showed the lowest surface microhardness at both polymerization times and had a higher inorganic filler content (50-55 vol%). This unexpected situation was attributed to the barium glass filler; when the barium glass is soaked in water, the barium atoms exchange with the hydrogen ions in the water and leak out of the glass. Consequently, the surrounding pH increases (Bowen and Reed 1976), and this accelerates the hydrolysis reactions of silanol bonds between the inorganic filler and the silanizing agent (Soderholm 1981). Then, this hydrolysis leads to the destruction of the bonds between the inorganic filler and the resin matrix. Finally, a decrease in the physical properties of the composite resin occurs.

The EFQ showed higher water sorption values than the ESF, and there were significant differences between the water sorption values of these two materials at both polymerization times ($p<0.05$). According to the manufacturer, the monomer composition of EFQ includes the Bis-MPEPP, TEGDMA, and UDMA monomers. Hiroyuki et al. (2019) evaluated flowable composites and found that Bis-MPEPP-based FCRs showed lower water sorption levels than Bis-GMA-based FCRs. Based on this finding, the EFQ should exhibit higher water sorption values than the ESF. According to Sideridou et al. (2003), TEGDMA has the highest water sorption capability, followed by Bis-GMA and UDMA. Based on these two literature studies, this can be explained by the mixtures of the monomer formulations, since the proportions of the monomers included in the formulations were not declared. In this study, although EFQ presented average sorption values comparable with those of the other FCRs, this material exhibited the highest solubility, and a statistically significant difference was observed for the different polymerization times ($p<0.05$). This may be explained as follows: although the polymeric structure of the EFQ absorbed water moderately, this material contained many unreacted monomers that leached out (Boaro et al. 2013). Additionally, the EFQ contains TEGDMA, which is a low molecular weight monomer that shows high mobility and decomposes more quickly than the larger molecules (Tanaka et al. 1991).

IGF is another material evaluated in this study, and it presented the highest sorption value among the FCRs at both polymerization times. According to the manufacturer, this material only contains methacrylate monomer. No additional information has been provided on whether IGF includes one or more methacrylate monomers. Therefore, from the results obtained, it can be considered that more hydrophilic monomers, such as TEGDMA or Bis-GMA, than hydrophobic monomers were contained in the IGF formulation.

The lowest solubility values were found for the ESF and IGF, and no statistically significant difference was found for the different polymerization times used with each material ($p>0.05$). When the IGF was evaluated in terms of the inorganic fillers, it was determined that this FCR contains silica, alumina, and zirconia particles (42% vol). Abuelenain et al. (2015) and Souza et al. (2012) remarked that zirconia and silica fillers provided greater hardness and lower solubility than barium and strontium fillers. Therefore, one of the lowest values obtained for the IGF could be related to the inorganic filler particles, which made the material less soluble. Another positive effect of these inorganic particles was seen in the microhardness values of the IGF. The microhardness values of the FCRs decreased in the order EFQ > IGF > ESF. Regarding the particles, the EFQ showed the highest surface microhardness values among all the experimental materials. This can also be attributed to the inorganic fillers (silica and zirconia) it has in common with the IGF. Although the ESF contains barium glass fillers, the lowest solubility was obtained with this material, and there was no statistically significant difference between the solubilities resulting from different polymerization times ($p> 0.05$). Yang et al. (2020) stated that the degrees of conversion for UDMA-based composite resin materials were significantly higher than those of methacrylate-based composite resins. ESF is a UDMA-based composite resin material with soluble barium glass fillers. Therefore, it may be that the ESF formulation may be optimally polymerized due to the content of UDMA, which also correlates with the solubility of a material.

The current study found that ESF and IGF presented negative solubilities with no statistically significant differences seen for the different polymerization times (p>0.05). Other studies have reported negative solubilities (Lopes et al. 2009; Barutcigil and Yildiz 2012). The first explanation for the negative solubilities, with which the current authors agree, is that there was incomplete dehydration of the materials. This does not mean that no solubility or eluate was produced from these materials; in contrast, there was a low solubility. Second, these materials were more suitable for water sorption. It was stated that the water sorption capacity was greater than the solubility; therefore, sorption could have masked the actual solubility (Fabre et al. 2007). The current authors disagree with the second statement because, in the present study, the IGF showed the highest sorption value, while ESF exhibited the lowest sorption value.

The current study has some limitations. First, there is the length of the storage time. A one-week (7 days) storage period generally constitutes a short-term evaluation. To provide real-time results, future studies should use longer storage times while evaluating the physical properties of the materials. Under oral conditions, the materials are exposed to temperature changes and physical forces. Therefore, this in vitro study could not simulate the oral environment. The lack of a thermophysical ageing test method and dynamic mechanical analyses of the restorative materials are also limitations of this study. In addition, the degree of conversion measured, for instance, by calorimetric and monomer elution tests, was a shortcoming in the present study.

5. Conclusions

Increased polymerization times did not affect the surface microhardnesses of the flowable composite resins, while it decreased the water sorption and water solubility levels. Regardless of the filler content, the flowable composites containing inorganic silica-zirconia filler particles exhibited the highest surface microhardnesses.

All the materials showed sorption levels of less than 40 $\mu g/mm^3$ and solubility levels of less than 7.5 $\mu g/mm^3$ with the ISO 4049 tests. When all flowable composite resin materials were polymerized twice the time recommended by the manufacturer, only the water sorption and solubility properties were improved.

References

ABUELENAIN, D.A., NEEL, E.A.A. and AL-DHARRAB, A. Surface and mechanical properties of different dental composites. *Austin Journal of Dentistry*. 2015, **2**(2), 1019.

ALPOZ, A.R., et al. Effects of light curing method and exposure time on physical properties of resin based dental materials. *European Journal of Dentistry*. 2008, **2**(1), 37-42.

AUNG, S.Z., et al. The effect of different light curing units on Vickers microhardness and degree of conversion of flowable resin composites. *Dental Materials Journal*. 2021, **40**(1), 44-51. https://doi.org/10.4012/dmj.2019-353

BAROUDI, K. and RODRIGUES, J.C. Flowable Resin Composites: A Systematic Review and Clinical Considerations. *Journal of Clinical and Diagnostic Research*. 2015, **9**(6), ZE18-ZE24. https://doi.org/10.7860/jcdr/2015/12294.6129

BARUTCIGIL, C. and YILDIZ, M. Intrinsic and extrinsic discoloration of dimethacrylate and silorane based composites. *Journal of Dentistry*. 2012, **40**(1), e57-e63. https://doi.org/10.1016/j.jdent.2011.12.017

BAYNE, S.C., et al. A characterization of first-generation flowable composites. *The Journal of American Dental Association*. 1998, **129**(5), 567-577. https://doi.org/10.14219/jada.archive.1998.0274

BEAUCHAMP, J., et al. Evidence-based clinical recommendations for the use of pit-and-fissure sealants: a report of the American Dental Association Council on Scientific Affairs. *The Journal of American Dental Association*. 2008, **139**(3), 257-268. https://doi.org/10.14219/jada.archive.2008.0155

BOARO, L.C., et al. Sorption, solubility, shrinkage and mechanical properties of low-shrinkage commercial resin composites. *Dental Materials*. 2013, **29**(4), 398-404. https://doi.org/10.1016/j.dental.2013.01.006

BOWEN, R.L. and REED, L.E. Semiporous reinforcing fillers for composite resins: I. Preparation of provisional glass formulations. *Journal of Dental Research*. 1976, **55**(5), 738-747. https://doi.org/10.1177/00220345760550050701

CADENARO, M., et al. Flowability of composites is no guarantee for contraction stress reduction. *Dental Materials*. 2009, **25**(5), 649-654. https://doi.org/10.1016/j.dental.2008.11.010

CHANDRU, T.P., et al. Evaluation of effect of polymerization time on curing depth of various bulk fill composites-an in vitro study. *Journal of Research in Dentistry*. 2020, **7**(3), 45-52. http://dx.doi.org/10.19177/jrd.v7e3201945-52

CHINELATTI, M.A., et al. Evaluation of the surface hardness of composite resins before and after polishing at different times. *Journal of Applied Oral Science*. 2006, **14**(3), 188-192. https://doi.org/10.1590/s1678-77572006000300008

FABRE, H.S.C., et al. Water sorption and solubility of dentin bonding agent light-cured with different light   sources. *Journal of Dentistry*. 2007, **35**(3), 253-258. https://doi.org/10.1016/j.jdent.2006.09.002

FERRACANE, J.L. Elution of leachable components from composites. *Journal of Oral Rehabilitation*. 1994, **21**(4), 441-452. https://doi.org/10.1111/j.1365-2842.1994.tb01158.x

FERRACANE, J.L. Hygroscopic and hydrolytic effects in dental polymer networks. *Dental Materials*. 2006, **22**(3), 211-222. https://doi.org/10.1016/j.dental.2005.05.005

FLOYD, C.J.E. and DICKENS, S.H. Network structure of Bis-GMA-and UDMA-based resin systems. *Dental Materials*. 2006, **22**(12), 1143-1149. https://doi.org/10.1016/j.dental.2005.10.009

HERVÁS-GARCÍA, A., et al. Composite resins: A review of the materials and clinical indications. *Medicina Oral Patologia Oral Cirugia Bucal*. 2006, **11**(2), E215-E220.

HIROYUKI, M., et al. Bis-MEPP-based flowable resin has low sorption and high color stability. 2019 IADR/AADR/CADR General Session (Vancouver, BC, Canada). Poster presentation. Final Presentation ID: 3652.

ILIE, N., KREPPEL, I. and DURNER, J. Effect of radical amplified photopolymerization (RAP) in resin-based composites. *Clinical Oral Investigations*. 2014, **18**(4), 1081-1088. https://doi.org/10.1007/s00784-013-1085-1

ISO-STANDARDS (2009) ISO 4049 Dentistry-Polymer-based restorative materials Geneve: International Organization for Standardization 4th edition.

JAGER, S., et al. Filler content, surface microhardness, and rheological properties of various flowable resin composites. *Operative Dentistry*. 2016, **41**(6), 655-665. https://doi.org/10.2341/16-031-l

KOWALSKA, A., SOKOLOWSKI, J. and BOCIONG, K. The photoinitiators used in resin based dental composite-A review and future perspectives. *Polymers (Basel)*. 2021, **13**(3), 470. https://doi.org/10.3390/polym13030470

KU, R.M., et al. Effect of flowability on the flow rate, polymerization shrinkage, and mass change of flowable composites. *Dental Materials Journal*. 2015, **34**(2), 168-174. https://doi.org/10.4012/dmj.2014-178

KUBO, S., et al. Three-year clinical evaluation of a flowable and a hybrid resin composite in non-carious cervical lesions. *Journal of Dentistry*. 2010, **38**(3), 191-200. https://doi.org/10.1016/j.jdent.2009.10.003

LASSILA, L., et al. Characterization of new fiber-reinforced flowable composite. *Odontology*. 2019, **107**(3), 342-352. https://doi.org/10.1007/s10266-018-0405-y

LIMA, A.F., et al. Influence of light source and extended time of curing on microhardness and degree of conversion of different regions of a nanofilled composite resin. *European Journal of Dentistry*. 2012, **6**(2), 153-157.

LOPES, L.G., et al. Influence of pulse-delay curing on sorption and solubility of a composite resin. *Journal of Applied Oral Science*. 2009, **17**(1), 27-31. https://doi.org/10.1590/s1678-77572009000100006

MIRICĂ, I.C., et al. Influence of filler loading on the mechanical properties of flowable resin composites. *Materials (Basel)*. 2020, **13**(6), 1477. https://doi.org/10.3390/ma13061477

PAYNE, J.H. The marginal seal of Class II restorations: flowable composite resin compared to injectable glass ionomer. *Journal of Clinical Pediatric Dentistry*. 1999, **23**(2), 123-130.

ROBERTS, H.W., CHARLTON, D.G. and MURCHISON, D.F. Repair of non-carious amalgam margin defects. *Operative Dentistry*. 2001, **26**(3), 273-276.

SALERNO, M., et al. Surface morphology and physical properties of new-generation flowable resin composites for dental restoration. *Dental Materials*. 2011, **27**(12), 1221-1228. https://doi.org/10.1016/j.dental.2011.08.596

SIDERIDOU, I., TSERKI, V. and PAPANASTASIOU, G. Study of water sorption, solubility and modulus of elasticity of light-cured dimethacrylate-based dental resins. *Biomaterials*. 2003, **24**(4), 655-665. https://doi.org/10.1016/s0142-9612(02)00380-0

SIDERIDOU, I.D., KARABELA, M.M. and VOUVOUDI, E.C. Physical properties of current dental nanohybrid and nanofill light-cured resin composites. *Dental Materials*. 2011, **27**(6), 598-607. https://doi.org/10.1016/j.dental.2011.02.015

SODERHOLM, K.J. Degradation of glass filler in experimental composites. *Journal of Dental Research*. 1981, **60**(11), 1867-1875. https://doi.org/10.1177/00220345810600110701

SODERHOLM, K.J., et al. Hydrolytic degradation of dental composites. *Journal of Dental Research*. 1984, **63**(10), 1248-1254. https://doi.org/10.1177/00220345840630101701

SOUZA, A.B., et al. Color stability of repaired composite submitted to accelerated artificial aging. *General Dentistry*. 2012, **60**(5), e321-e325.

TANAKA, K., et al. Residual monomers (TEGDMA and Bis-GMA) of a set visible-light-cured dental composite resin when immersed in water. *Journal of Oral Rehabilitation*. 1991, **18**(4), 353-362. https://doi.org/10.1111/j.1365-2842.1991.tb00067.x

TARUMI, H., TORII, M. and TSUCHITANI, Y. Relationship between particle size of barium glass filler and water sorption of light-cured composite resin. *Dental Materials Journal*. 1995, **14**(1), 37-44. https://doi.org/10.4012/dmj.14.37

USLU CENDER, E. and GULER, E. An in vitro evaluation of the efects of diferent acidic beverages on the surface hardness of restorative materials. *Yeditepe Journal of Dentistry*. 2018, **14**(1), 35-41. https://doi.org/10.5505/yeditepe.2017.71473

WEI, Y.J., et al. Diffusion and concurrent solubility of self-adhering and new resin-matrix composites during water sorption/desorption cycles. *Dental Materials*. 2011, **27**(2), 197-205. https://doi.org/10.1016/j.dental.2010.10.014

YAP, A.U., LOW, J.S. and ONG, L.F. Effect of food-simulating liquids on surface characteristics of composite and polyacid-modified composite restoratives. *Operative Dentistry*. 2000, **25**(3), 170-176.

YANG, J., et al. Effects of nano-zirconia fillers conditioned with phosphate ester monomers on the conversion and mechanical properties of Bis-GMA- and UDMA-based resin composites. *Journal of Dentistry*. 2020, **94**, 103306. https://doi.org/10.1016/j.jdent.2020.103306

ZHANG, H., et al. Randomized controlled clinical trial of a highly filled flowable composite in non-carious cervical lesions: 3-year results. *Clinical Oral Investigation*. 2021, **25**(10), 5955-5965. https://doi.org/10.1007/s00784-021-03901-z