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Material behavior of resin composites with and without fibers after extended water storage

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The objective of this study was to determine the long-term water sorption, solubility and hygroscopic expansion of resin composites with and without incorporated short fibers. Three resin composites incorporating fibers were examined: everX Posterior (EVX), NovoPro Universal (NPU) and NovoPro Flow (NPF). Four Particulate filled composites were used as controls: Filtek bulk Fill (FBF), Filtek one bulkfill (FBO), Filtek Supreme XTE (XTE), and Filtek Supreme Flow (XTEF). For sorption and solubility measurements, specimens were immersed in water for 140 days, weighed at intervals, then dried for a further 42 days at 37±1°C. Laser micrometer measured diametral expansion. XTEF exhibited the highest sorption. The solubility range was between -1.4 to 4.1 µg/mm; XTEF had the highest solubility, with EVX demonstrating negative solubility. Hygroscopic expansion ranged between 1.4% for hydroxyapatite fiber reinforced composite (NPU) and 2.2% for E-glass fiber reinforced composite (EVX). A nano-fiber containing composite (NPU) had the most favorable outcomes compared to a range of composites.

Keywords: Resin composites, EverX Posterior, Sorption, Solubility, Bisphenol A-glycidyl methacrylate

INTRODUCTION

The majority of resin composites are known to be chemically stable; however, chemicals present in the oral environment can be absorbed by composite polymer networks, and some of the components from the resin matrix and filler could be released into the surrounding area¹. These occurrences are known as sorption and solubility, and these processes into and out of the network structures can result in undesirable physical and biological effects². Resin composites are significantly affected by sorption and solubility in terms of longevity, as these processes affect the material's physical and mechanical properties such, color and dimensional stability, strength, and hardness²⁻⁵.

A wide range of chemicals are found in the oral environment, including alcohol, acids and bases. The effects that these chemicals have on resin composites depend on a number of factors such as the nature of the chemicals and the length of time the material is exposed to them². While the oral environment does play a significant role, the nature of the resin composite is also a factor; the filler system's nature, the porosity, crosslink density and hydrophilicity of the network, and the quality of the filler interface all have a considerable effect⁶.

Long-term clinical success of a resin composite depends significantly on its dimensional changes, both during and after curing⁷. Unreacted monomer can be gradually released from these materials, water can also be absorbed and take up all of the free volume of the

network structure which may lead to swelling through the separation of chains in the polymer network⁸. The elastic modulus of the polymer is known to be affected by the uptake of water, which coupled with hygroscopic expansion could potentially relax the internal stresses created by constrained shrinkage⁷. It is not as straightforward as this, however, because the expansion caused by water uptake is not controlled and can result in alternative deleterious stresses. Moreover, each of these phenomena follow very different timescales. For instance, shrinkage takes place within seconds—days at a maximum^{9,10}; on the other hand, water absorption occurs many days, and saturation typically takes weeks^{1,5}. Prior research found that material expansion is not controlled and can lead to potential stresses on the cavity walls, which may subsequently cause micro-cracks in the restored tooth⁵. These findings demonstrate the importance and complexity of dimensional changes in resin composite, highlight their unpredictability and that these changes depend on both the material and solvent involved^{2,5,7}.

A promising type of resin composite includes fibers as reinforcement¹¹. These fibers enhance composite properties by acting mainly as crack stoppers^{12,13}. They can offer enhanced mechanical properties that can be very close to those of the natural tissues¹⁴. This approach was first reported for reinforcement of polymethyl methacrylate (PMMA), which was later utilized in a different aspect of clinical dentistry¹³. Furthermore, reinforcing the resin matrix with fibers improves the capacity of distributing the stress more efficiently when the loads are concentrated on the restoration¹². Having said this, several factors play an important role in

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ensuring the efficiency of fiber reinforcement, such as fiber type and aspect ratio¹⁵.

Several fiber materials have been used as reinforcement; carbon/graphite, for example, has been utilized in post and core systems. Unfortunately, the dark color of the fibers restricts their clinical use as a tooth-colored restorative material¹⁶. However, inorganic glasses may have favorable esthetic, mechanical and chemical properties. Thus, glass fibers have been used as reinforcement for direct restorations^{13,17}. Electrical/E-glass is the most commonly used glass fiber due to its low cost¹³. Due to the biocompatibility of hydroxyapatite¹⁸, it has been used in resin composites to improve their mechanical properties. This material could be deployed in various forms, such as particulate fillers¹⁹ or fibers²⁰.

According to Callister and Rethwisch, short fibers with a subcritical length are not effective and significantly lower the reinforcement effect of any resin containing such²¹. This length may be defined as the minimum fiber length required for optimal stress transfer within the resin matrix²². It is the minimum length at which a fiber will fail, midway along its length in an fiber reinforced composite (FRC), rather than as interfacial fracture between the matrix and the fiber²³. For example the diameter of glass fibers currently used in dental FRCs is 15–18 μm and the critical fiber length should be, between 0.75–0.9 μm ²³.

The critical fiber length should be 50 times greater than the diameter of the fiber to allow homogenous stress transfer within the resin matrix²³.

The present objective was to determine time-dependent water sorption and related properties of seven resin composites with and without incorporated short fibers. The null hypotheses were as follows:

1. No difference in either water sorption or solubility between the evaluated resin composites after 140 days water exposure.
2. No difference in hygroscopic expansion between the evaluated resin composites after 140 days water storage.

MATERIALS AND METHODS

The resin composite investigated are presented in Table 1. Three fiber containing composites and four composites reinforced exclusively with particulate fillers. They were chosen to represent composites used for different clinical applications with varying percentages of resin matrix and filler.

Measurement of filler content

To measure each resin composite's mass percentage of inorganic filler, the ISO 1172:1996 standard ash method was followed²⁴. For each composite (Table 1) two specimens were made ($n=2$). Polytetrafluoroethylene (PTFE) molds were used to prepare the specimens (2 mm thickness, 4 mm diameter) and they were placed between two sections of clear Mylar strip with glass slides on each side (1 mm in thickness) and then squeezed together. An LED light curing unit with an

output irradiance of 1.2 W/cm² was used to irradiate the specimens for 20 s on one side (Elipar S10, 3M Espe, Seefeld, Germany). The irradiance was measured every time the light cure unit was utilized, using a calibrated radiometer (MARC™ Resin Calibrator, BlueLight Analytics, Halifax, Canada). The specimens were then stored for 24 h at 37°C. An electric furnace (Programat EP 5010, Ivoclar Vivadent, Schaan, Liechtenstein) was used to keep a silica crucible at 630°C for 30 min. Once the crucible had been cooled to ambient temperature in a desiccator containing silica gel at 37±1°C, a precision digital balance (BM-252, A&D, Tokyo, Japan) was used to determine its weight. Each of the composite specimens was placed in the crucible and the balance was used again to weigh the specimen, including the crucible. To burn out the organic matrix, the specimen-containing crucible was placed in the electric furnace for 30 min at 630°C. Once cooled to ambient temperature in a desiccator, the crucible and residue were reweighed. The following equation was used to determine the inorganic filler content:

$$\text{Filler content (\%)} = \frac{a_3 - a_1}{a_2 - a_1} \times 100$$

Equation 1: Filler content formula

Where a_1 is the mass of the crucible, a_2 is the mass of the crucible plus the specimen; a_3 is the final mass of the crucible plus the residue after heat treatment.

Sorption and solubility

1. Specimen preparation

Using brass molds, five disc-shaped specimens were produced for each material. The molds, with dimensions of 15×2 mm, were placed between two sections of clear Mylar strip with glass slides on each side (1 mm in thickness) and then squeezed together. The thickness of specimen was modified from 1 mm to 2 mm. This increased thickness, corresponding more closely to clinical setup, allowing water sorption studies over a longer period. An LED curing unit with measured average tip irradiance of 1.2 W/cm² (as mentioned above) was used to irradiate five sections of each side for 20 s. The irradiance was measured every time the light cure unit was utilized, using a calibrated radiometer (as mentioned above). The specimens were taken out of their molds with care, and 1000 grit silicon carbide paper was used to smooth out any rough edges. Following this, the specimens were placed in a desiccator containing silica gel at 37±1°C. After a period of 24 h a precision-calibrated balance was used to weigh each specimen, accurate to ±0.01 mg (BM-252, A&D). The cycle was duplicated repeatedly until a constant mass was acquired (m_1)—in other words, until the mass loss of the specimens was no more than 0.2 mg over 24 h.

For the thickness measurement, a digital caliper was used (Absolute Digimatic, Mitutoyo, Kanagawa, Japan) to obtain two measurements of the height. After taking the dimensions of the specimen, the volume (V) was calculated in mm³ through the following formula:

Table 1 Composition of materials investigated according to manufacturers' information

Material		Manufacturer	Lot number	Type and shade	Filler load		Filler type	Resin matrix
Code	Name				vol%	wt%		
Fiber and particulate reinforced composite								
NPU	NovaPro Universal	Nanova, MO, USA	30001	Nano-fiber reinforced, nano-hybrid Conventional A2 shade	—	77	Barium Borosilicate Glass, Hydrophobic Amorphous Silica, Hydroxyapatite fibers.	Bis-EMA, TEGDMA, UDMA
NPF	NovaPro Flow	Nanova,	2001	Nano-fiber reinforced, nano-hybrid Flowable A2 shade	—	60	Barium Borosilicate Glass, Amorphous Silica, Hydroxyapatite fibers.	Bis-EMA, TEGDMA, UDMA
EVX	ever X Posterior	GC, Tokyo, Japan	1701101	Fiber reinforced BulkFill Universal shade fiber length 0.3–2.0 mm, diameter 16–17 μm	53.6	74.2	E-Glass short fibers, Barium Borosilicate Glass,	Bis-GMA, TEGDMA, PMMA
Particulate reinforced composite								
XTE	Filtek Supreme XTE	3M Oral Care, St.Paul, MN, USA	N836906	Nano-hybrid Conventional A2 shade	63.3	78.5	Zirconia filler Silica fillers/ Zirconia and silica clusters.	Bis-GMA, Bis-EMA, UDMA, PEGDMA
XTEF	Filtek Supreme Flowable	3M Oral Care	N522058	Nano-hybrid Flowable A2 shade	46	65	Zirconia filler Silica fillers/ Zirconia and silica clusters	Bis-GMA, Bis-EMA, TEGDMA
FBF	Filtek Bulk fill	3M Oral Care	N838840	Nano-hybrid Bulk fill A2 shade	58.4	76.5	ytterbium tytterbium trioxide and zircon silica	DDDMA, UDMA, AUDMA
FBO	Filtek One Bulk fill	3M Oral Care	N859232	Nano-hybrid Bulk fill A2 shade	58.4	76.5	ytterbium tytterbium trioxide and zircon silica	DDDMA, UDMA, AUDMA

Bis-GMA: bisphenol-A-diglycidyl methacrylate; Bis-EMA: bisphenol-A-polyethylene-glycol-diether dimethacrylate; TEGDMA: triethyleneglycol dimethacrylate; PMMA: polymethyl methacrylate; UDMA: urethane dimethacrylate, DDDMA (1,12-Dodecanediol dimethacrylate), AUDM: Aromatic urethane dimethacrylate.

$$V = \pi r^2 t$$

Equation 2: Volume calculation formula

Where $\pi=3.14$, r is the radius of cross section; t is the thickness of specimen.

2. Sorption measurement

All five specimens were submerged in 10 mL of distilled water within separate glass bottles, which were sealed with polyethylene caps. The bottles were kept at 37°C for 1 h, 3 h, and 1, 2, 3, 4, 5, 6, 7, 14, 21, 28, 56, 84,

112, and 140 days. After each time period, a tweezer was used to take each specimen from the bottles. They were dried using filter paper before being weighed 1 min after removal from the water. The recorded mass is denoted as $m_2(t)$. All five specimens were then returned to aqueous storage. This was replenished every week, with the total volume of water maintained at 10 mL.

3. Solubility measuring

After the sorption cycle was complete, specimens were dried using a desiccator and weighed at time points of 1, 2, 3, 4, 5, 6, 7, 14, 21, 28, 35 and 42 days. Once the mass loss of the specimens was no more than 0.2 mg within any 24 h period, the constant final mass was then obtained (m_3).

Weight increase $W_i(\%)$ and water sorption WSo were calculated through the following formulae:

$$W_i(\%) = 100 \left[\frac{m_3 - m_1}{m_1} \right]$$

Equation 3: Weight increase calculation formula

m_1 is the conditioned mass prior to immersion in water; m_2 is the mass after to immersion in water for 140 days.

$$WSo = \left[\frac{m_2 - m_3}{V} \right]$$

Equation 4: Water sorption calculation formula

m_2 is the mass after to immersion in water for 140 days, m_3 is specimens' mass after desorption, and V is the volume of the specimen.

The percentage amount of water absorbed by a composite at the end of the storage period was calculated by the following formula:

$$WSoC(\%) = \left[\frac{m_2 - m_3}{m_1} \right] \times 100$$

Equation 5: Water sorption % calculation formula

m_1 is the conditioned mass prior to immersion in water; m_2 is the mass after to immersion in water for 140 days, m_3 is specimens' mass after desorption.

Providing sorption has occurred principally by the polymer matrix component, the following equation was used to measure the percentage amount of water the polymer matrix absorbed²⁵.

$$WSoM(\%) = \left[\frac{mSoC\%}{a} \right] \times 100$$

Equation 6: Water sorption % in the resin matrix calculation formula

In this formula a represents the proportional weight of the polymer matrix in the composite.

The following equation was used to calculate the solubility (Sol) values:

$$Sol = \left[\frac{m_1 - m_3}{V} \right]$$

Equation 7: Solubility calculation formula

m_1 is the conditioned mass prior to immersion in water; m_3 is specimens' mass after desorption, and V is the volume of the specimen.

Hygroscopic expansion

Hygroscopic dimensional changes were measured in parallel with the water sorption measurements. A custom built noncontact laser micrometer was utilized to measure the dimensional changes of the specimens⁵. The initial mean diameter d_1 of each specimen was measured. After each time period had elapsed, specimens were dried using filter paper then measured 1 min after removal from the water. Mean diameter ($d_{2(t)}$) was recorded at each time interval, and then returned to aqueous storage. An average of 600 diametral values was recorded for each specimen at each time point.

The percentage diametral change was calculated:

$$d(\%) = \frac{d_{2(t)} - d_1}{d_1} \times 100$$

Equation 8: Diametral change calculation formula

In this formula d_1 represent the mean diameter before water storage, while $d_{2(t)}$ represents the mean diameter which was recorded at each time interval.

The following equation was used to calculate volumetric change, assuming isotropic expansion behavior²⁶:

$$V(\%) = \left[\left(1 + \frac{d(\%)}{100} \right)^3 - 1 \right] \times 100$$

Equation 9: Volumetric change calculation formula

Statistical analysis

SPSS v.23 (IBM, Armonk, NY, USA) was used to analyze the data. The mean and standard deviations were calculated for the water solubility, water sorption, hygroscopic expansion and mass change. One-way ANOVA was carried out at 140 days followed by Tukey *post-hoc* tests (at $\alpha=0.05$) for the hygroscopic expansion, water sorption, and mass change. For the solubility, the same statistical test was applied to evaluate differences in weight after 42 days of desorption cycle. Pearson correlation coefficients were calculated to express the correlation between hygroscopic expansion and mass change for each material during 140 days water immersion.

RESULTS

Filler content

Table 2 shows the mean and standard deviations of the filler wt%, using the ashing technique, and the manufacturers reported values.

Sorption and solubility

As can be seen from Fig. 1, each of the resin composites

Table 2 Filler by weight percentage

Materials	Filler (wt %) after ashing in air	Manufacturer reported filler (wt%)
NPU	69.6 (2.3)	77
NPF	59.6 (1.3)	66
EVX	72.8 (1.2)	74.2
XTE	74.1 (1.3)	78.5
XTEF	63.1 (1.1)	65
FBF	74.6 (1.4)	76.5
FBO	73.2 (1.0)	76.5

Table 3 Water sorption (*WSo*) and solubility (*Sol*), water sorption in composite (*WSoC%*), water sorption in polymer matrix (*WSoM%*), of resin composites after 140 days storage in distilled water at 37°C

Materials	<i>WSo</i> ($\mu\text{g}/\text{mm}^3$)	<i>WSoC%</i>	<i>WSoM%</i>	<i>Sol</i> ($\mu\text{g}/\text{mm}^3$)
NPU	19.96 (3.32) ^a	1.04 (0.18) ^a	3.43 (0.59) ^{a,b}	2.63 (0.13) ^{a,b}
NPF	28.88 (0.11) ^{b,c}	1.62 (0.13) ^{b,d}	3.94 (0.32) ^a	3.59 (0.44) ^e
EVX	30.00 (0.28) ^{b,c}	1.29 (0.05) ^{a,d}	4.75 (0.20) ^{a,c}	-1.49 (0.41) ^d
XTE	21.11 (1.62) ^a	1.17 (0.11) ^a	3.95 (0.37) ^a	3.19 (0.19) ^a
XTF	30.11 (0.28) ^{b,c}	1.60 (0.11) ^{b,c,d}	4.44 (0.33) ^a	4.18 (0.47) ^e
FBF	22.24 (3.63) ^{a,b}	1.20 (0.16) ^{a,c}	4.55 (0.63) ^a	3.43 (1.09) ^{a,e}
FBO	24.70 (3.25) ^{a,b,c}	1.17 (0.13) ^a	4.37 (0.51) ^a	3.32 (0.57) ^{a,e}

The same superscript lower case letters indicate a homogeneous subset (columns) ($p > 0.05$)

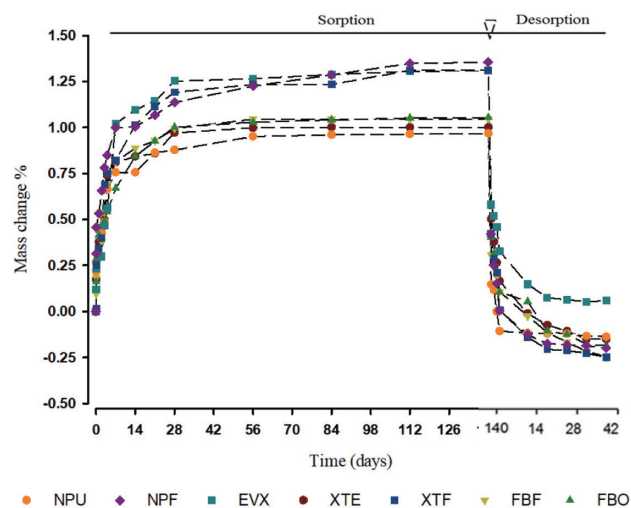


Fig. 1 Mass change percentage with water sorption and desorption cycles.

exhibited a percentage mass change throughout the water sorption/desorption cycle. All of the composites demonstrated an increase in mass of various degrees by their water uptake, up to the point of equilibrium which occurred after 140 days. All of the examined

composites showed a higher initial mass (m_1) than their reconditioned mass (m_3), with the exception of EVX whose initial mass was lower than its reconditioned mass.

At 140 days, water sorption ranged between 19.96 and 30.11 $\mu\text{g}/\text{mm}^3$ (Table 3). The highest sorption was observed in XTEF followed by EVX and NPF which exhibited similar results. Conversely, XTE, NPU, FBO and FBF exhibited lower water sorption levels, with no significant differences between each other ($p \geq 0.05$).

The solubility for the resin composites was found to fall between -1.49 to 4.18 $\mu\text{g}/\text{mm}^3$, as shown in Table 3. The most soluble materials were NPF and XTF; they had higher levels of solubility when compared with their packable counterpart. A negative solubility value was observed for EVX (-1.49 $\mu\text{g}/\text{mm}^3$).

Hygroscopic expansion

One-way ANOVA conducted after 140 days of immersion in water showed that EVX had a significantly higher hygroscopic expansion when compared to the rest of the materials. Table 4 provides the mean and standard deviation for all materials for their volumetric hygroscopic expansion, taken after 140 days at 37°C. The percentage hygroscopic expansion for each material is shown in Fig. 2.

Table 4 The percentage increase in mass and volume of the investigated materials after 140 days, filler wt% after air ashing. Person correlation coefficient between hygroscopic expansion and mass change during 140 days sorption period in water at 37°C

Materials	% Mass Change	% Volumetric Change	Pearson correlation coefficient
NPU	0.93 (0.18) ^{a,b}	1.40 (0.17) ^a	0.83
NPF	1.34 (0.11) ^{a,c}	1.70 (0.19) ^a	0.90
EVX	1.35 (0.08) ^{a,c}	2.21 (0.26) ^b	0.97
XTE	1.00 (0.10) ^a	1.54 (0.05) ^a	0.87
XTF	1.32 (0.14) ^{a,c}	1.72 (0.11) ^a	0.91
FBF	1.01 (0.12) ^a	1.49 (0.19) ^a	0.93
FBO	1.00 (0.12) ^a	1.51 (0.15) ^a	0.94

The same superscript lower case l letters indicate a homogeneous subset (columns) ($p < 0.05$)

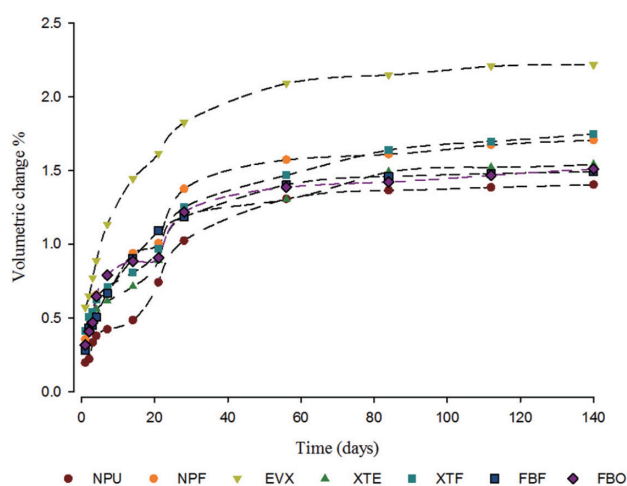


Fig. 2 Hygroscopic expansion from 1 h to 20 weeks.

The final hygroscopic expansions ranged between 1.40 and 2.21% at 140 days. According to the method of application of the resin composite, the composites can be categorized into one of three bands: conventional composites (NPU and XTE), with expansions of 1.40% and 1.54%; bulk fill composites (EVX, FBO and FBF), with the greatest expansion being 2.21% for EVX; and lastly, flowable composites (NPF and XTEF), with expansions of 1.70% and 1.72%, respectively.

Figure 3 shows the relationship between the mass and the changes in volume over the period of 140 days revealing that the relationship was almost linear.

DISCUSSION

This study evaluated water sorption, solubility and hygroscopic expansion of a number of resin composites immersed in water over 140 days including fiber reinforced materials. Considerable differences were identified between the materials, leading to a rejection of the first and second null hypotheses. ISO Standard

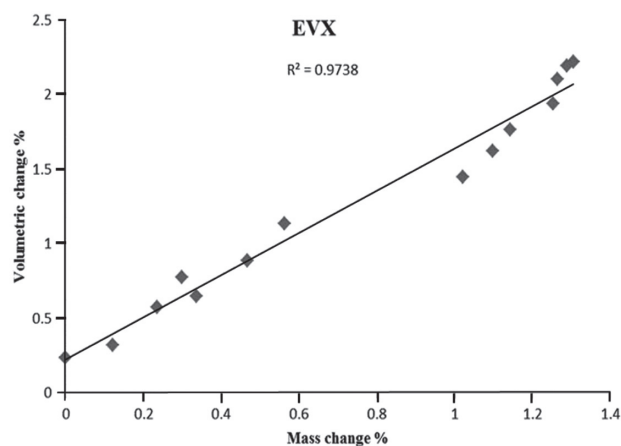


Fig. 3 The relationship between the mass and volumetric changes during the 140 days sorption period in EVX.

4049 permits a sorption limit of 40 $\mu\text{g}/\text{mm}$ and solubility of less than 7 $\mu\text{g}/\text{mm}$ after a period of 7 days storage. Each of the composite materials satisfied this standard, despite being exposed to an extended period of water sorption. Thus the aqueous challenge was more stringent than the 7-day ISO process.

Hydrophilicity and crosslinking of the network structure are the two main factors affecting the solubility and water sorption of resin composites. Moreover, the amount of solvent taken up by the composite during the exposure period depends on both the porosity of the material itself and the nature of the filler matrix^{27,28}.

Two different types of fiber were incorporated in the tested materials: short E glass fibers (EVX) and Nano-hydroxyapatite fibers (NPU and NPF), which vary in their composition, configuration and amount; thus, could show different behavior. Therefore, the focus of this study was on the main factors in overall degradation resistance: the polymeric matrix and filler amount.

The results in the present study regarding sorption values correlated negatively with the amount of filler

loading (with the exception of EVX). This corresponds to the results observed in other studies²⁹. The wt% of the polymeric matrix decreases as the weight percentage of filler increases, and so the water sorption also decreases as this phenomenon is known to occur within the polymeric phase^{30,31}. Although glass fillers (particulates or fibers) are known to not contribute to the sorption process, it is still possible that water is adsorbed onto their surface —this depends on the integrity of the interface between the resin matrix and the glass fillers³².

In the case of NPF and XTEF, the high sorption can be attributed to their filler content; however, when the influence of fillers was removed through the calculation of the percentage of water absorbed by the polymer matrix alone (*WSoM*) rather than by the composite (*WSoC*), no significant differences were found between these materials when compared with their packable counterpart (NPU, XTE).

The properties of FRCs deteriorate in water, similarly to particulate filled composites. Water diffuses *via* the resin matrix and leaches from the fiber surface³³. Areas with poorly impregnated fibers will be more prone to water uptake³⁴. Water sorption is influenced by the hydrophilicity of the resin matrix, and the amount of the inorganic phase (fibers and the particulates) and the quality of silanization. Additionally, water uptake may be accentuated by capillary action of the fibers, resulting in mass increases³⁵. When water comes into contact with glass fiber by exposing the glass fibers during finishing and polishing procedures, water sorption along the interface is much greater than the diffusion through the polymer matrix. This is due to the capillary effect of the glass fiber, which may be seen in E-glass fiber reinforced composites. Moreover, polymerization shrinkage could cause capillaries between the resin matrix and the glass fibers, especially with resin systems of high polymerization shrinkage, such as monomers of TEGDMA and MMA.

EVX was shown to have the highest polymer sorption value (*WSoM*) (4.75%) and NPU exhibited the lowest value (3.43%) out of all the composites studied. These results confirm that water storage has a significant effect on polymeric matrix properties as noted by much previous research on dimethacrylate-based resins^{1,4,35}. The higher sorption rate of EVX, therefore, may be explained by the highly hydrophilic PMMA and TEGDMA monomers that constitute its resin matrix. Similarly, the significantly low sorption of NPU could be associated with the fact that its main monomers are Bis-EMA and UDMA, which are highly hydrophobic. Prior studies found that Bis-EMA homopolymer has a much lower water sorption (1.8 wt%) than UDMA, Bis-GMA, and TEGDMA homopolymers (2.6, 3.05, and 6.3 wt%, respectively)³⁶.

logP (octanol-water partition coefficient) is a quantity that is widely employed in medicinal chemistry and pharmaceuticals to measure hydrophilicity³⁷. Research on resin composites suggests that logP is an effective predictor of water sorption³⁸. Alshali *et al.* listed the logP values of the following monomers in ascending

order: TEGDMA < DEGDMA < UDMA < Bis-GMA < Bis-EMA³⁹. This offers further explanation for the outcome of this study.

Incomplete dehydration of the EVX material may explain its negative solubility. This does not imply that EVX is insoluble, although it suggests that its solubility is low. Another possible explanation is that hydrolytic chemical reactions between metal oxides, glass fillers and water result in metal hydroxides forming within the composite material⁴⁰.

There was a significant variation between the final hygroscopic expansions leading to rejection of the second null hypothesis. Hygroscopic expansion happens when water diffuses into the polymer network separating the chains —particularly when the water molecules bond to hydrophilic groups within the polymer^{5,35,41,42}. Absorbed water can, however, often reside in free volume and micro-voids between polymer network chains, to an extent that does not increase the macroscopic volume^{5,43}. It is well established that water diffuses into the resin phase, and that the matrix expands to accommodate absorbed water³⁰. Hirasawa *et al.* were the first to establish an association between water sorption and volumetric expansion in resin composites⁴⁴.

Water sorption thus affects the mass and dimensions of composites and previous studies highlight relationships between their volume and mass changes^{2,5}. The present study also found high correlations between percentage mass and volumetric changes (Table 4).

EVX is a resin composite incorporating short glass fibers within a matrix of PMMA, TEGDMA, and Bis-GMA.

Dental manufacturing companies introduced glass fiber reinforced materials to try to improve the strength of resin composites, especially fracture toughness and flexural strength^{45,46}. The current study observed that EVX did not behave in the same manner as the other resin composites, as it exhibited the greatest volumetric changes. Research on PMMA acrylic denture-base resins indicated that, in the case of fiber-reinforced materials, the resin matrix determined material behavior during water storage more than the glass fibers⁴⁷. EVX polymer matrix consists of polymer PMMA and copolymer Bis-GMA/TEGDMA. The structure of Bis-GMA incorporates hydroxyl groups that raise its susceptibility to water diffusion and bonding. The sorption behavior of Bis-GMA may also be affected by the co-monomer PMMA present in the organic matrix^{35,48}. According to its “Instructions for Use” EVX should only be used as dentin replacement and thus should be covered by a conventional particulate filled composite. However, in certain clinical situations such a procedure may not be feasible^{49,50}.

NPU was found to have lower expansion, along with XTE, FBO, and FBF. If the material is hydrophobic, reduced hygroscopic expansion is known to take place^{5,35}. NPU does contain Bis-EMA, and this material is more hydrophobic than Bis-GMA⁵¹. It may also be that differences in polymerization can be a factor, although we have no evidence that is actually the case.

From the results of the current study, we can

conclude as follows:

1. There were some variations in the water sorption/desorption cycles of all the resin–matrix composites investigated. Nonetheless, they all complied with the requirements set out by ISO 4049 for water solubility and sorption, despite the sorption period.
2. The greatest changes in volume and water sorption were seen in the millimetre scale glass-fiber reinforced composite (EVX), whereas the greatest stability in an aqueous environment was seen in the nano-fiber hydroxyapatite reinforced composite (NPU).

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