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ORIGINAL ARTICLE

The diffusion kinetics of a nanofilled and a midifilled resin composite immersed in distilled water, artificial saliva, and lactic acid

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Abstract This study investigated the diffusion kinetics of a nanofilled (Filtek Z350) and a midifill (Filtek P60) resin composite immersed in distilled water, artificial saliva and lactic acid. Resin composite specimens were desiccated, immersed in the media, weighed at suitable time intervals until they reached sorption equilibrium and were then desiccated again. Sorption and solubility (µg/mm^3) were calculated based on ISO 4049:2000(E). The diffusion coefficient (m^2.s^{-1}) was determined according to Flick's second law. The degree of conversion (DC%) was evaluated by FT-IR and the action of the media on the surfaces of the resin composite was evaluated by SEM. Z350 immersed in lactic acid presented the highest sorption (25.9±1.3). The highest solubility was presented by Z350 immersed in lactic acid (5.6±0.9), followed by P60 immersed in lactic acid (4.4±0.5). The other groups presented no significant difference among them. The diffusion coefficients of both resin composites immersed in lactic acid and that of Z350 immersed in artificial saliva were significantly higher. The lowest diffusion coefficient was presented by P60 immersed in distilled water. The DC % was not significant, ($p>0.05$). The SEM analysis showed that the effect of lactic acid on the resin composites was more deleterious than those of water and artificial saliva.

Keywords Resin composites · Solubility · Sorption · Diffusion coefficient · Degree of conversion

Introduction

In an endeavor to gain a better understanding of the resin composite degradation process, several researchers have exhaustively studied the sorption, solubility, and mechanical properties of many resin composite after immersion in water, artificial saliva, or ethanol [1–11]. Resin composites are generally constituted of a polymeric matrix, filler particles, and a silane-coupling agent that links the matrix to the fillers. The current resin composites are classified according to the size of their filler particles, as microfilled, hybrid, microhybrid, and nanofilled [12]. Filtek Z350 (the universal restorative that contains the same patented and proprietary nanotechnology used to create Filtek™ Supreme Universal Restorative, 3M-ESPE) is a typical nanofilled composite, which has a filler particle system that combines silica nanofillers with a primary particle size of 20 nm and zirconia-silica nanoclusters of 0.6–1.4 µm [13]. Although some published studies have shown that this type of resin composite presents mechanical properties similar to those of the hybrid or midifill type [14, 15], the large surface area to volume ratio derived from the silica particles may increase its water uptake, leading to degradation of the filler-polymeric matrix interface [5, 9], and to a probable reduction in some of its mechanical properties [1].

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The sorption phenomenon in resin composite is a diffusion-controlled process that causes chemical degradation due to residual monomer release and filler-polymeric matrix debonding [4, 10]. Resin composite solubility is reflected by the amount of leachable unreacted monomers. During polymerization, these unreacted monomers are trapped in microgels between the polymer chains and adsorbed to the surrounding network, or are trapped in nanopores. The latter are more susceptible to leaching out of the resin composite than the monomers inside the microgels [7]. Although sorption and solubility are important to understanding the behavior of resin composites as regards degradation in the oral environment, both properties represent the final stage of fluid flow inside the material. During the polymerization reaction, the monomers present in the polymeric matrix of resin composites are converted to a rigid-elastic network [16]. However, even when light-activated, the monomer conversion is never immediately complete and the matrix may undergo a post-curing effect [17]. In other words, the polymeric network undergoes re-arrangement until it reaches a maximum degree of conversion (DC%) [16].

In addition to sorption and solubility, the diffusion coefficient that represents the rate at which the fluids penetrate the polymer network could be crucial to increasing knowledge regarding resin composite degradation [18–21]. Most of the previous researches that have analyzed the sorption, solubility and diffusion coefficient of resin composites have used immersion media such as water [1, 4, 6–11] artificial saliva [3–5, 10, 11] and ethanol [2, 4]. However, to date, few studies have analyzed the effect of acids produced by human dental plaque on these properties [22]. Indeed, several earlier studies have shown that lactic and other acids produced by human dental plaque had detrimental effects on softening [23], wear [24], and surface degradation [25] of polymeric resins and resin composites. Moreover, some published studies have shown that lactic acid is one of the main acids produced by human dental plaque [26, 27]. Thus it is important to analyze the sorption kinetics of resin composites when they are immersed, not only in water and artificial saliva, but in lactic and other acids produced by human dental plaque.

Therefore, the aim of this research was to analyze the sorption kinetics (sorption, solubility and diffusion coefficient) of a nanofilled and a midifill resin composite when immersed in saliva artificial and lactic acid 0.01 M. Distilled water was used as control. The research hypotheses were: (1) the nanofilled resin composite would present higher sorption, solubility and diffusion coefficient than the midifill one, (2) the lactic acid would increase the sorption, solubility, and diffusion coefficient of the resin composites tested.

Materials and methods

The resin composites Filtek Z350 (nanofilled) and Filtek P60 (midifill), (3M ESPE St. Paul, MN, USA) were investigated in the present study. According to the 3 M ESPE, the two resin composites have qualitatively the same polymeric matrix (Bis-GMA, Bis-EMA, UDMA, and TEGDMA). Their compositions are disclosed in Table 1.

All the resin composite specimens used in the experimental protocol were light-activated with a quartz-tungsten-halogen unit (Optilux 501, Kerr, Danbury, CT, USA) using an irradiance of 650 mW/cm^2 for 40 s. The radiant exposure (26 J/cm^2) was calculated as the product of the irradiance of the curing unit, by using a radiometer (model 100, Demetron Inc. Danburry, USA), and the time of irradiation.

Degree of conversion—DC%

Spectra of the unpolymerized and polymerized specimens of each resin composite were recorded by a FT-IR spectrometer (Varian 3100 FT-IR, Varian Inc, Palo Alto, CA, USA), equipped with an attenuated total reflectance crystal—ATR (MIRacle ATR, Pike Technologies, WI, USA) operating with 120 scans at a resolution of 4 cm^{-1}. Standard Increments of each resin composite were compressed between two polyethylene strips and two glass slides to produce a thin film. Five films of each resin composite were then light-activated with the light tip in contact with the glass slide. FT-IR spectra of the polymerized specimens were recorded 24 h after dry storage at 37 C. The DC% was calculated from the ratio between the height of absorbance peaks of the aliphatic C = C bond (1,638 cm^{-1}) to the aromatic C = C bond (1,608 cm^{-1}),

Table 1 Composition of the resin composites investigated in this study

Material	Composition
Filtek P 60 (A3 shade)	Filler: 61 vol.% silica/zirconia filler with mean particle size of 0.6 µm Polymeric matrix: Bis-GMA, Bis-EMA, UDMA, and TEGDMA
Filtek Z350 (A3 shade)	Filler: 59.5 vol.% combination of aggregated zirconia/silica cluster ranging from 0.6 to 1.4 µm with primary particles size of 5–20 nm and nonagglomerated 20 nm silica filler Polymeric matrix: Bis-GMA, Bis-EMA, UDMA, and TEGDMA

The resin composite compositions are given according to the manufacturer's information (3M ESPE, St. Paul, MN, USA) used as an internal standard, obtained from the polymerized and unpolymerized specimens by the following equation:

$$\mathrm{DC}(\%) = 100\ \mathbf{x}\left[1 - \left(R_{\text{polymerized}}/R_{\text{unpolymerized}}\right)\right]$$

where R = peak at 1,638 cm^{-1}/ peak at 1,608 cm^{-1}

Diffusion kinetics

Disc-shaped specimens were built up by filling an aluminum mold (1 mm thick and 6 mm in diameter). After filling the mold to excess, the material surface was covered with a polyester strip and a glass slide, compressed with a device (500 g) for 20 s to avoid porosities, and then light-activated from the top. The discs were assigned in groups of five specimens (n=5), placed in a desiccator containing freshly dried silica gel, and transferred to an oven at 37 C. After 24 h, the discs were repeatedly weighed in an analytical balance (AX 220, Shimadzu, Tokyo, Japan) until a constant mass (m_1) was attained, i.e., disc mass variation was less than ±0.1 mg in any 24 h period. After that, the discs were individually placed in plastic vials and immersed in 10 ml of distilled water, artificial saliva (KCl, NaCl, MgCl, CaCl, Nipagin, CNC, Sorbitol, and deionized water - neutral pH) and lactic acid 0.01 M (pH=4) at 37±1 C. Subsequently, the discs were removed from the vials at fixed time intervals, washed in distilled water, blotted dry with absorbent paper, air dried for 15 s, weighed and returned to the vials containing 10 ml of fresh fluids. On the first day the discs were weighed at time intervals of 5, 10, 15, 30, 60, 90, 120, 150, and 180 min. After the first day, the discs were weighed daily until the sorption equilibrium was reached, (mass variation less than ±0.1 mg (m_2)). This process lasted 16 days. After this, the discs were placed in a desiccator and weighed daily until the mass variation was less than ±0.1 mg (m_3). The thickness and the diameter of the discs were measured at four points, with a digital caliper (MPI/E-101, Mitutoyo, Tokyo, Japan), and the volume (V) was calculated in mm^3.

Sorption and solubility

The sorption (Sp) and the solubility (Sl) during the course of the 16 days of media immersion were obtained using the following formulae (µg/mm^3):

$$Sp = \frac{m_2 - m_3}{V}$$

$$Sl = \frac{m_1 - m_3}{V}$$

Where m_1 is the specimen mass (mg) after drying, m_2 is the specimen mass (mg) at equilibrium uptake (maximum sorption), m_3 is the mass (mg) of redried specimen and V is the specimen volume (mm^3).

Diffusion coefficient

For a solid bounded by two parallel planes, the Fick's second law relates the diffusion coefficient in one dimension (x) as a function of the time, and its solution is expressed as follows:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2}$$

Here, D is the diffusion coefficient and C is the concentration (%) of the diffusing specimen at time t.

For longer times of diffusion, the solution to this differential equation is expressed as follows:

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2}\sum_{n=0}^{\infty}\left[\frac{1}{(2n+1)^2}\right]e\left[-\frac{(2n+1)^2\pi^2}{L^2}Dt\right]$$

Where M_t is the mass uptake (g) at time t, M_∞ is the mass uptake (g) at equilibrium, and L is the specimen thickness (m).

However, for the initial stages of uptake (when $M_t/M_\infty \leq 0.6$), the above equation is reduced to:

$$\frac{M_t}{M_\infty} = \frac{4}{L}\left(\frac{Dt}{\pi}\right)^{\frac{1}{2}}$$

The diffusion coefficient D (m^2.s^{-1}) was obtained from the initial curve slope of the plot of M_t/M_∞ against $t^{1/2}$.

SEM analysis

In order to characterize the effect of media on the surface of the resin composites, scanning electron microscopy (SEM) images of the composite surfaces were taken. For this purpose, two specimens of each material were analyzed before and after immersion in the media. The specimens were air dried for 48 h in a desiccator containing dried silica gel, then mounted on aluminum stubs and sputter-coated with Au-Pd (EMITECH model K550, Emitech, Ashford, Kent, UK). The specimen surfaces were observed using a SEM (JSM 5310, Jeol Ltd, Akishima Tokyo, Japan) operating in the backscattered electron mode.

Statistical analysis

The obtained data were analyzed using Statgraphics 5.1 Software (Manugistics, Rockville, MD, USA). Initially, the normal distribution of the errors and the homogeneity of variances were checked, respectively, by Shapiro–Wilk's test and Levene's test. Based on these preliminary analyses, the DC% data were analyzed by Student's t test and the sorption, solubility and diffusion coefficient data were analyzed by two-way analysis of variance (ANOVA) and

Student-Newman–Keuls test for multiple comparisons. The analyses were performed at a significance level of $\alpha=0.05$. In addition, the SEM images were analyzed qualitatively.

Results

Table 2 shows the means and standard deviations of the DC %, sorption and solubility. As regards DC%, Student's *t* test detected no statistically significant difference between the resin composites ($p>0.05$).

With respect to sorption, two-way ANOVA identified statistical significance for resin-based composite independent factor ($p=0.004$) as well as for the double interaction (resin composite vs. immersion medium, $p=0.0368$). On the other hand, the immersion medium was shown to be not significant ($p=0.2436$). Student-Newman–Keuls test showed that the sorption of Z350 immersed in lactic acid was significantly higher than that in the all other groups ($p=0.0074$).

As regards solubility, two-way ANOVA detected a significant influence for the two main factors: resin-based composite ($p=0.003$) and immersion medium ($p<0.05$). The double interaction was not significant ($p=0.7154$). According to Student-Newman–Keuls test, Z350 immersed in lactic acid presented the highest solubility, followed by P60 immersed in lactic acid ($p<0.05$). It was found that the other groups did not differ from each other, ($p<0.05$).

The diffusion coefficients calculated from the plots of M_t/M_∞ against the $t^{1/2}$ (Figs. 1 and 2) are shown in Table 3. Two-way ANOVA showed a significant influence for the two main factors: resin-based composite ($p=0.0032$) and immersion medium ($p<0.05$). According to the Student-Newman–Keuls test both resin composites immersed in lactic acid and Z350 in artificial saliva presented the highest diffusion coefficient, followed by Z350 immersed in distilled water, P60 immersed in artificial saliva and P60 immersed in distilled water ($p<0.05$).

Figures 3 and 4 show representative SEM images of resin composites before and after 16 days of immersion in the tested media.

Discussion

Resin composite degradation is a somewhat complex phenomenon that involves mechanisms such as the polymeric matrix plasticization, hydrolysis of silane bonds between the fillers and the matrix, as well as filler particle corrosion [7, 10, 28]. These phenomena are strongly influenced by the chemical structure of monomers present in the polymeric matrix and features of the filler particle system [4, 5, 25], and may diminish some mechanical properties such as hardness, flexural strength and elastic modulus [1, 4]. The rationale to analyze Z350 (nanofilled) and P60 (midifill) resin composites in the present study was due because, according to their manufacturer (3M ESPE, St Paul, MN, USA) both material have qualitatively the same polymeric matrix (Bis-GMA, Bis-EMA, UDMA, and TEGDMA). In other words, the findings about the properties assessed here were focused on the influence of the filler particle systems present in these resin composites.

Earlier studies have reported that organic acids produced by the human dental plaque caused adverse effects on resins and resin-based restorative materials [22, 25]. Asmussen [23] analyzing the effect of propionic, acetic and lactic acids on the softening of dimethacrylate resins showed that the former two acids produced a higher softening effect than lactic acid and claimed that this reflects the similarity between the solubility parameters of these acids and those of dimethacrylate monomers. At a first glance, this result leads to accepting that propionic and acetic acids have more deleterious effects on resin composites than lactic acid has. However, in an in vivo study, Distler and Kröncke [26] showed that in addition to acetic acid, lactic acid accounts for about 70% of the total acids present in the human dental plaque of the subjects examined. Furthermore, in the same study, there was a steeper increase in lactic acid than acetic and propionic acids after sucrose rinsing. According to these authors, this result "is due to rapid sucrose degradation by lactate–producing bacteria such as *Streptococcus mutans*, and is associated with a decrease in the percentages of the other acids". Based on this, it is reasonable to speculate that during sugar insult, the bacteria present in

Table 2 Means and standard deviations of DC%, sorption and solubility (µg/mm^3) of the resin composites

Media	Z350		P60	
	Degree of conversion			
	55.7±3.8		59.0±4.6	
	Solubility	Sorption	Solubility	Sorption
Artificial saliva	3.4±0.7 C	22.1±1.9 b	2.8±0.3 C	21.0±2.7 b
Lactic acid	5.5±0.9 A	25.9±1.3 a	4.5±0.5 B	18.9±3.3 b
Distilled water	3.5±0.9 C	21.0±3.5 b	2.7±0.4 C	19.7±3.1 b

The capital letters represent the statistical significance for solubility values and the lower case letters represent the statistical significance for sorption values. Means followed by the same letters are not statistically different (ANOVA/Student-Newman–Keuls test, $\alpha=0.05$)

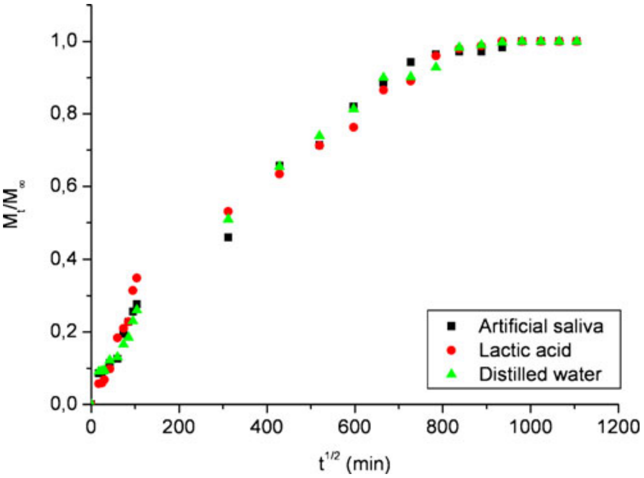

Fig. 1 Plot of M_t/M_∞ against the $t^{1/2}$ for Z350 resin composite

human dental plaque, e.g., *S. mutans*, would increase the production of lactic acid, therefore increasing the amount of it in the dental plaque. Principally in the areas where the buffer action of saliva is less effective and where the dental plaque is adherent to the composite surfaces, i.e., proximal areas, this increase in the amount of lactic acid could increase the sorption and the degradation of composite. Thus, studies about the action of this acid on resin composites may increase the basis of knowledge about resin composite degradation in the oral environment. The pH of the lactic acid used in the present study was adjusted to four because, according to an earlier study, this is the lowest pH found in human dental plaque [29].

Among other aspects, the DC% may influence the sorption and the solubility the resin composites undergo [5, 7]. In the present study, the difference in DC% between resin composites was found to be not significant (Table 2). Since materials have qualitatively the same polymeric matrix, this result was expected. Based on this, it is reasonable to claim that the DC% did not influence the sorption and the solubility of resin composites analyzed here.

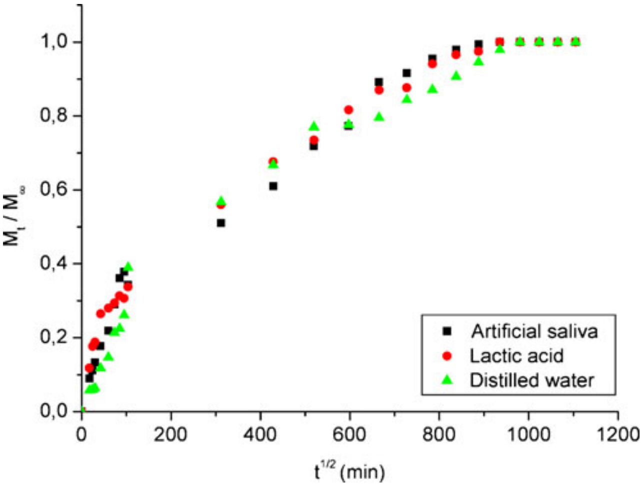

Fig. 2 Plot of M_t/M_∞ against the $t^{1/2}$ for P60 resin composite

In the present study, Z350 presented a higher sorption than P60. This result supports the first research hypothesis. In lactic acid, its sorption (25.9 µg/cm^3) was significantly higher than that of P60 (18.9 µg/cm^3), (Table 2). These values represent a difference of 37% in lactic acid sorption between the two resin composites. Moreover, although the sorption in artificial saliva and distilled water was found to be not statistically significant between resin composites (Table 2), the values of Z350 (22.1 µg/cm^3/21.0 µg/cm^3) were numerically higher than that of P60 (21.0 µg/cm^3/ 19.7 µg/cm^3). This could be interpreted as a trend of nanofilled resin composite to be more prone to absorb fluids than the midifill type. Even considering some differences in terms of methodology, e.g., size of specimens, light-polymerization mode used and time of immersion, these results are supported by previous studies. Silva et al. [5] showed that when immersed in artificial saliva, Supreme resin composite, i.e., a nanofilled composite similar to Z350, presented a sorption 9% higher than that of P60. Furthermore, in the study of Curtis et al. [1] Supreme resin composite had a higher water uptake than Z250, i.e., a midifill resin composite similar to P60, after 6 and 12 months of storage. Two mechanisms can be used to explain this higher sorption suffered by nanofilled resin composite. Firstly, the greater surface area to volume ratio, derived from the nonagglomerated 20 nm silica filler, could have allowed more fluids to accumulate at the filler particle-polymeric matrix interfaces [1, 5, 9]. Secondly, analyzing dental composites with hydroxyapatite, Santos et al [18] showed that the presence of porosity and formation of filler particle aggregates increased the water uptake into composites. These authors claimed that this increase in water sorption was due to the poor impregnation of aggregates by the polymeric matrix. According to them, this weak link could also provide paths of facile diffusion towards the inside of aggregates, where the presence of microvoids was quite probable due to a lack of impregnation of filler particles with the polymer matrix. Since Z350 also has filler aggregates in its composition, we hypothesized that the fluids accumulated at the interface between the aggregated zirconia/silica cluster filler-polymeric matrix could have diffused through paths created due to poor impregnation of 5–20 nm-sized primary particles by polymeric matrix.

The higher sorption of Z350 in lactic acid was most probably influenced by the chemical structure of this acid. Lactic is a carboxylic acid which has –OH and –COOH functional groups in its molecule. It is most probable that these functional groups had established a high level of hydrogen bonds with polar sites of dimethacrylate mono-

Table 3 Means± standard deviations of the diffusion coefficient ($10^{-13} \times m^2.s^{-1}$)

RBC	Immersion media		
	Artificial saliva	Lactic acid	Distilled water
Filtek Z350	7.6±0.4 (0.98) a	7.7±0.4 (0.99) a	5.6±0.3 (0.97) b
Filtek P60	4.9±0.1 (0.98) c	7.7±0.2 (0.92) a	3.6±0.1 (0.97) d

The *r* parameter values for each condition are presented in parentheses. Mean values with identical letters indicate no statistically significant difference (Student-Newman–Keuls, $p > 0.05$)

mers present in Z350 polymeric matrix, i.e., -OH- in Bis-GMA, -O- in TEGDMA and Bis-EMA and –NH- in UDMA, thus increasing the acid uptake by the matrix. The micromorphology showed in the Fig. 3c reinforces this possibility. In other words, the large surface area and the microvoids formed in the Z350 polymeric matrix after immersion in lactic acid could have increased the trapping of acid molecules that established hydrogen bonds with methacrylate monomers. Taking into account the results presented in the study of Bagheri et al. [25], it is reasonable to claim that this higher lactic acid sorption will increases the degradation of Z350. Analyzing the surface degradation of some resin-based materials, these authors showed that Supreme resin composite immersed in lactic acid presented a higher silver penetration depth than it presented when it was immersed in distilled water. According to these authors the possible factor that influenced this finding was the degree of sorption suffered by Supreme resin composite.

The solubility phenomenon in resin composites reflects the release of residual monomers and oligomers as well as filler particles and ions from its surfaces [30]. In the present study, the highest solubility was presented by Z350 (5.5 µg/mm^3)

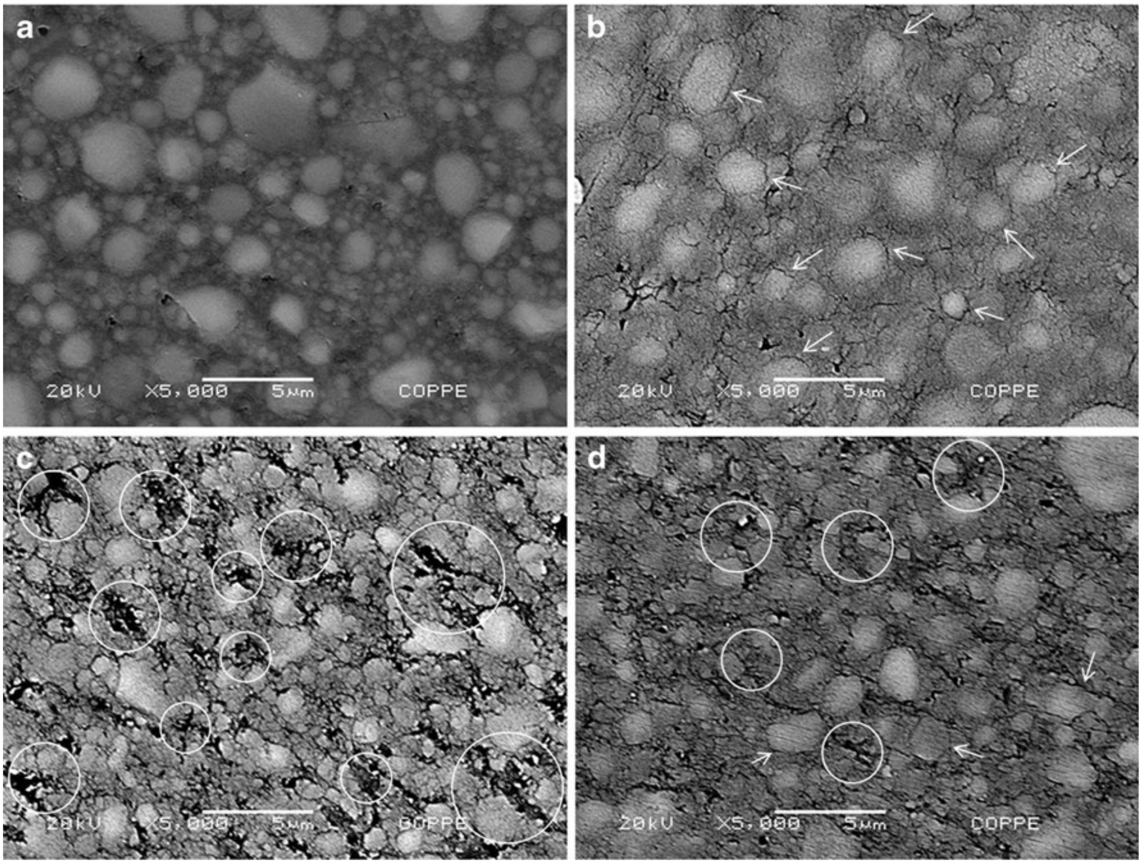

Fig. 3 Representative SEM micrographs of the Z350 specimens. **a** Before immersion; and after immersion **b** in distilled water; **c** in lactic acid; and **d** in artificial saliva. Severe superficial damage, suggesting aggregated zirconia/silica cluster filler loss can be seen in the specimen after lactic acid and artificial saliva immersion (*white circles*). Microcracks between filler-polymeric matrix can be noted in the specimens after immersion in distilled water and artificial saliva (*white arrows*)

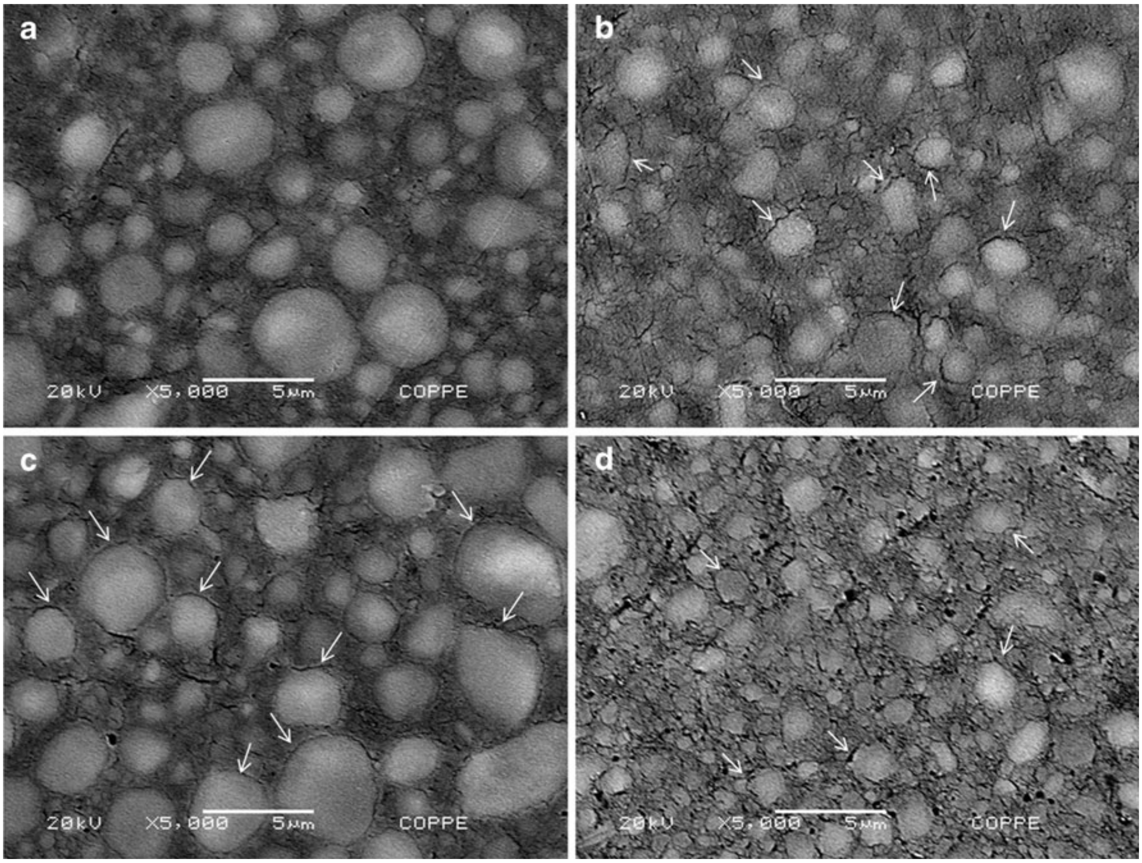

Fig. 4 Representative SEM micrographs of the P60 specimens. **a** Before immersion; and after immersion **b** in distilled water; **c** in lactic acid; and **d** in artificial saliva. Microcracks between filler particle-polymeric matrix can be seen in all the specimens after immersion in the media (*white arrows*)

immersed in lactic acid. This supports the first research hypothesis. In addition, the solubility of both resin composites in lactic acid was higher than those in distilled water and artificial saliva (Table 2). Thus, the second research hypothesis was partially proved. It seems obvious that this action of lactic acid on the solubility of resin composites was influenced by its low pH (= 4). Initially, the low pH may have acted in the polymeric matrixes through catalysis of ester groups from dimethacrylate monomers. The hydrolysis of ester groups leads to the formation of alcohols and carboxylic molecules that may accelerate degradation due to lowering the pH inside the matrix [31]. On the other hand, the low pH may also have caused erosion on the filler surfaces, accelerating their debonding or, at least, increasing the release of ions from their surfaces [30]. The high level of porosities shown in Fig. 3c could reinforce this possibility.

Specifically in the case of the highest solubility presented by Z350 in lactic acid, discussed for sorption, here the role the greater surface area to volume ratio derived from the nonagglomerated 20 nm silica filler plays in this property is clear. The larger amount of silane-coupling agent probably pre-mixed with the filler particle system of this resin composite, e.g., γ-methacryloxypropyltrimethoxysilane, certainly suffered a high level of hydrolysis via ester linkages, contributing to an increase in its solubility.

The diffusion coefficient values obtained in the present study (ranging from $3.6\times10^{-13}\mathrm{m}^2.\mathrm{s}^{-1}$ to $7.7\times10^{-13}\mathrm{m}^2.\mathrm{s}^{-1}$), (Table 3) are in good agreement with other values published in the literature [18–20, 32]. Based on Figs. 1 and 2 the authors assumed that for all the media the plots of M_t/M_∞ against $t^{1/2}$ were linear in the initial stages of sorption. In Table 2 the goodness-of-fit parameter for the linear approximation (*r* parameter), are shown for each linear fit. It can be observed that all *r* values are higher than 0.9, showing that the sorption processes showed a Fickian behavior, confirming the authors' assumption. Coincidently, the two resin composites presented equal diffusion coefficient when immersed in lactic acid ($7.7\times10^{-13}\mathrm{m}^2.\mathrm{s}^{-1}$). Most probably, this was due to the fact that the two resin composites have similar polymeric matrixes (Bis-GMA, Bis-EMA, UDMA, and TEGDMA). Santos et al. [18] found similar values of diffusion coefficients for experimental composites based on BisGMA/TEGDMA matrixes (66.5/33.5 wt.%) irrespective of presence of hydroxyapatite filler particles ($3.7\times10^{-13}\mathrm{m}^2.\mathrm{s}^{-1}$ for unfilled resin and 3.1 to $4.5\times10^{-13}\mathrm{m}^2.\mathrm{s}^{-1}$ for filled materials). According to these authors this suggests that water diffuses principally through the resin phase of composite filling materials.

The two resin composites presented the highest diffusion coefficient when immersed in lactic acid (Table 2). This finding contributes to acceptance of the second research hypothesis and may be interpreted as a faster diffusion of lactic acid inside the resin composites than distilled water and artificial saliva. We hypothesized that this behavior was influenced by the plasticizing effect of lactic acid on the polymeric matrixes of materials [33]. Again, the devastating effect seen in Fig. 3c could be interpreted as a result of plasticization in the polymeric matrix of Z350. Furthermore, Z350 presented a higher diffusion coefficient than P60 when immersed in distilled water and artificial saliva (Table 2). Although the magnification of SEM micrographs seen in Fig. 3 (×5.000) is not sufficiently high to identify details at the filler-matrix interface, it is possible that these higher diffusion coefficients could have been influenced by some areas of ineffective impregnation of nanofillers and nanoclusters by the polymeric matrix of Z350 nanocomposite [18].

The Figs. 3 and 4 highlight the effects of media on resin composite surfaces. It is worth noting that the image seen in Fig. 4a (P60 before immersion) shows cracks at some filler particle-polymeric matrix interfaces. At a first glance, this could suggest that the specimen processing produced some alterations in the surfaces of this resin composite. However, when making a comparison between Figs. 3a and 4a, it can be noted that the surface of Z350 resin composite did not suffer any damage during the specimen processing. Based on this, the authors assumed that most of the aspects seen in Figs. 3b, c, d and 4b, c, d were produced or, at least, increased after immersion in the media. For both materials, the worst damage was produced by lactic acid and artificial saliva (Figs. 3c, d, 4c and d). Although the ISO 4049:2000 (E) for polymer-based filling, restorative and luting materials determines that the sorption and solubility phenomena in resin composites must be conducted using water, these findings suggest that the behavior of resin composites when immersed in oral-like fluids, i.e. artificial saliva and acids produced by human dental plaque, are also crucial for understanding their degradation in the oral environment.

Conclusion

Within the limits of the experimental protocol used in this study it was concluded that due to the greater surface area to volume ratio, the nanofilled resin composite may suffer a higher degradation than the midifill type in the oral environment. In addition to sorption and solubility, the diffusion coefficient must be taken into account in the analysis of resin composite degradation. The effect of acid lactic on resin composites may be more deleterious than that of distilled water and artificial saliva in the degradation process of resin composites. Therefore, further investigations using other acids produced by human plaque are needed to increase the knowledge base about resin composite degradation.

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Conflict interest declaration The authors declare that they have no conflict of interest.

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