

# Study of Physicochemical Properties of Two Current Commercial Dental Self-Curing Resin Composites

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**ABSTRACT:** The aim of the present work is the study of some physicochemical properties of two current commercial dental self-curing two component composites, i.e., of Concise™ (3M™, Dental Products, St. Paul, USA), and Alfacomp (VOCO GmbH, Germany). Based on their filler type Concise is characterized as “conventional” or “macro-filled” composite and Alfacomp as “hybrid.” The resin matrix of Concise is a copolymer of 2-bis[4-2-hydroxy-3-(methacryloxy)-propyl]phenyl propane (Bis-GMA)/triethyleneglycol dimethacrylate, while that of Alfacomp a homopolymer of Bis-GMA. The composites were prepared in accordance with the manufacturers’ instructions by mixing equal amounts of the two components at room temperature. The degree of conversion of double bonds of resin matrix during curing was determined in thin film of composites using FTIR transmission spectroscopy. The degree of conversion of Concise and Alfacomp was found to be correspondingly  $(73.63 \pm 4.33)\%$  and  $(47.75 \pm 1.80)\%$  after a day-polymerization. Sorption, solubility, and volumetric

change were determined after storage of composites in water or ethanol/water solution 75 vol % at 37°C (a good food/oral simulating fluid) for 30 days. These properties for both composites were higher in ethanol/water solution than in water. Also these properties for Concise were lower than those for Alfacomp in both liquids. Thermal stability of composites was studied by thermogravimetric analysis which performed in air atmosphere from 50 to 800°C. This analysis of composites showed the degradation of their resin matrix in three steps. The organic resin content of Concise was found to be 18.0% w/w and that of Alfacomp 20.1% w/w. Concise showed generally better properties than Alfacomp. This behavior must due to the differences in the chemical structure of the organic resin matrix and the type of inorganic filler. © 2012 Wiley Periodicals, Inc. *J Appl Polym Sci* 000: 000–000, 2012

**Key words:** self-curing composites; degree of conversion; sorption; solubility; volumetric change; thermal stability

## INTRODUCTION

Currently, resin composites are commonly used as direct restorative materials. Although light-cured resin composites are generally used because of their numerous advantages,<sup>1</sup> self-cured and dual-cured resin composites still have important applications in contemporary restorative dentistry, including core buildup, luting of indirect restorations, and post bonding.<sup>2–4</sup> Self-curing composites are relatively easy to use and require no additional curing equipment. So, they often have been used in developing countries due to a lack of facilities, especially electricity, necessary to cure light-cured composites. Self-curing composites came as two separate putty-like pastes (a base and a catalyst) containing correspondingly the benzoyl peroxide (BPO) as initiator and an aromatic tertiary amine as activator (usually *N,N*-dimethyl-*p*-toluidine, DMT). The dentist would take equal amounts of each of these pastes and mix

them together just prior to their use. The combination of the two pastes would activate the composite’s curing reaction. This reaction involves the polymerization of the organic matrix of composite, which consists mainly of dimethacrylate monomers. The most used monomers are the 2-bis[4-2-hydroxy-3-(methacryloxy)-propyl]phenyl propane (Bis-GMA) and the triethyleneglycol dimethacrylate (TEGDMA) (Fig. 1). These monomers have a double bond in common, which is opened up to allow the monomer to bond to a neighboring monomer (free-radical addition polymerization).<sup>5</sup>

The inorganic filler is the second phase those composites bear. Silicon dioxide is the most common filler due to its properties which can suit for biomaterials. It is used in quartz or colloidal silica form and the grains’ size specify the composite type: macro-, micro-, nano-filled, or hybrids. Other ingredients like organosilanes, pigments, and stabilizers are present in small quantities.

In our previous work, we have studied the kinetics of the BPO/amine-initiated free-radical polymerization of dental dimethacrylate monomers<sup>6</sup> and the effect of amine and monomer chemical

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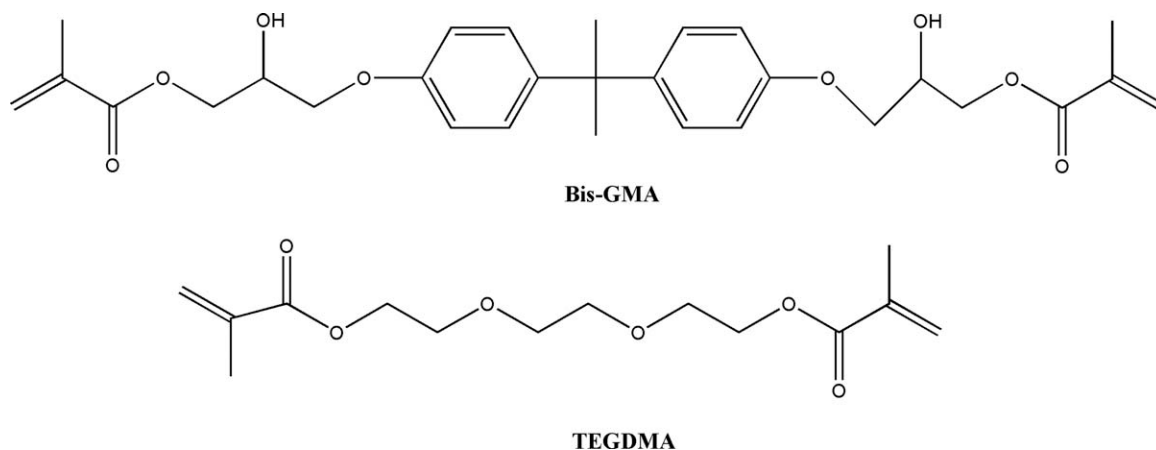


Figure 1 Structure of Bis-GMA and TEGDMA monomer.

structure on this kinetics.<sup>7</sup> Also in another work, we have studied the copolymerization kinetics of dental dimethacrylate monomers initiated by a BPO/amine redox system.<sup>8</sup>

In the present work, we have studied some properties of two current commercial dental self-curing composites, i.e., of Concise (3M, Dental Products, St. Paul, USA), and Alfacomp (VOCO GmbH, Germany). The composites were prepared in accordance with the manufacturer's instructions.

The degree of conversion of double bonds during curing was determined using FTIR spectroscopy. The room-temperature polymerization of dimethacrylates usually leads to glassy resins in which only a part of the available double bonds are reacted. Before the completion of conversion, the vitrification process decelerates the reaction to a hardly perceptible rate. Only very flexible monomers in which the reactive methacrylate groups are relatively far apart can be completely reacted at ambient temperature. The degree of conversion of resins is a major factor influencing their bulk physical properties. In general, the higher the conversion of double bonds is, the greater the mechanical strength. The unreacted double bonds may either be present in free monomer or as pendant groups on the network. The unreacted monomer may leach from the polymerized material and irritate the soft tissue. For example, TEGDMA is suspected to be propitious to bacterial growth around the restoration. Furthermore, monomer trapped in the restoration may reduce the clinical serviceability of composite through oxidation and hydrolytic degradation, which may be manifested in forms such as discoloration of the fillings and accelerated wear. The final degree of conversion of a resin depends on the chemical structure of the dimethacrylate monomer and the polymerization conditions, i.e., atmosphere, temperature, and initiator concentration.<sup>9</sup>

Sorption, solubility, and volumetric change were determined after storage of composites in water or

ethanol/water solution 75 vol % at 37°C for 30 days. Dimensional changes of dental resin composites during and after setting are a source of concern for clinicians. It has been shown clinically that long-term dimensional instability might lead to post-operative pain, tooth staining, marginal breakdown of the restorations and fractured teeth. The dimensional stability of resin composites is affected by polymerization shrinkage, thermal contraction, and expansion and interaction with an aqueous oral environment. Water or solvent uptake into the resin phase of resin composites causes two opposing processes. The solvent will extract unreacted components, mainly monomer, resulting in shrinkage, loss of weight, and reduction in mechanical properties. Conversely, solvent uptake leads to a swelling of the composite and an increase in weight. The solvent diffuses into the resin network and separates the chains, creating expansion. However, since the resin network contains microvoids created during polymerization and free volume between chains, a part of the solvent is accommodated without creating a change in volume. Thus, the dimensional change of a resin composite in a solvent is complex and difficult to predict and depends on the chemical structure of the resin. The hydrophilicity of the resin needs to be of sufficient magnitude to distend the resin. In addition, the mean elastic modulus of the resin needs to be sufficiently low to accommodate the distension. Hence, the ratio between the elastic modulus of the resin and the strength of its hydrophilic attraction may determine the capacity to alter the dimensions of the resin. Expansion resulting from water sorption can be a clinically desirable phenomenon if it fully counteracts the effects of shrinkage. A coefficient of expansion that exceeds the shrinkage value is not desirable, as further stresses may be introduced into the teeth. Several researchers have studied the dimensional changes of various dental resin composites after immersion in

water for a certain time. However, intraoral conditions are clearly more complex than those achieved with distilled water in the laboratory. According to Food and Drug Administration Guidelines of the United States, ethanol in water is a recommended food simulator and may be considered clinically relevant. A 75 vol % ethanol/water solution has been the solvent of choice to simulate accelerated aging of restorations, because it has a solubility parameter which matches that of Bis-GMA.<sup>10</sup>

Thermogravimetric analysis of composites was performed in oxygen atmosphere from 50 to 800°C. It is well known that dental composites when used as indirect composites can be improved by heat treatment, as a possible way to increase mechanical properties due to additional cure (post-cure). So it is useful to know the thermal stability of dental composites.

## MATERIALS AND METHODS

Concise™ (3M™, Dental Products, St. Paul) (Lot No. N154570) is a “conventional” or a “macrofilled” composite. The major component of organic matrix is Bis-GMA and TEGDMA which is used as diluent. In the literature is reported that Bis-GMA is 75% and TEGDMA 25% of the organic matrix.<sup>11</sup> Organic matrix content is 22% w/w according to the manufacturer. The filler powder is in form of ground quartz with average particle diameter of about 9 μm (the particle sizes ranges from 1 to 40 μm). Filler content is 78% w/w (which corresponds with approximately 67% v/v). A small percentage of microfill particles are added to the filler fraction. It is treated with a silane so that it is chemically bound to the resin. Other ingredients are present in very minor amounts. The “catalyst” (more correctly the initiator of polymerization) is BPO and the accelerator is DMT. Inhibitors are present in parts-per million. Titanium dioxide and stable iron oxides are present for pigmentation and provide a consistent shade. Concise is widely used for Class III, IV, and V restorations or crown built up, etc, in conjunction with the acid etch technique.

Alfacomp (VOCO GmbH, Germany) (Lot No. 0910388) is a Bis-GMA-based, self-curing two-component “hybrid” composite with 81% w/w filler content (ca., 0.05 and 3–5 μm) according to the manufacturer. Alfacomp is recommended for: Class III–V fillings, Class I and II fillings which are not exposed to occlusal forces, extended fissure sealing, core build-up, reconstruction of traumatically affected teeth, improving aesthetics (e.g., diastema, hypoplasia, conical teeth, and erosions). It shows high abrasion resistance and stability, easy mixing and homogeneous consistency.

The composites were prepared by mixing equal quantities of base and catalyst paste on the mixing pad with a disposable spatula for 15–30 s until homogeneous.

## FTIR analysis—degree of conversion

Fourier’s transform infrared (FT-IR) transmission spectroscopy is well used for measuring the degree of conversion of the monomers in resin composite materials.

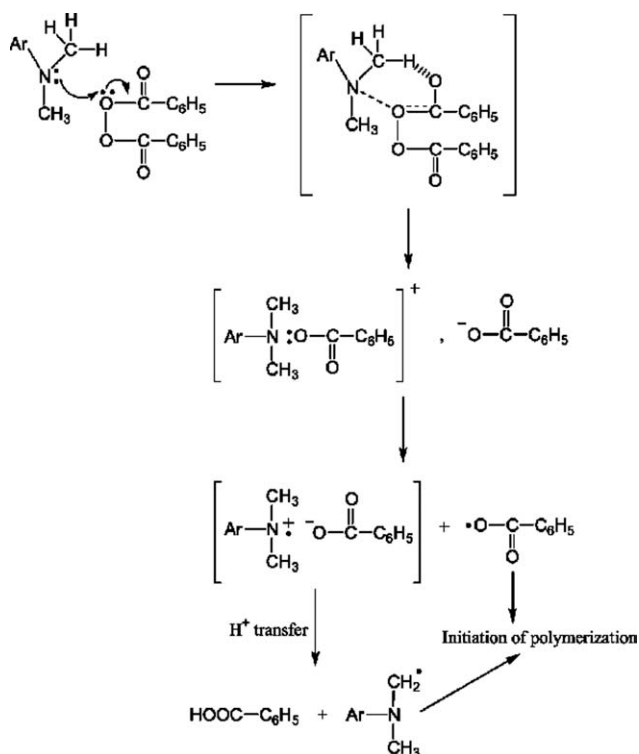
The FTIR analysis was conducted in a FTIR spectrometer, Spectrum One Model of Perkin–Elmer Company (Massachusetts, USA). Spectra were obtained over 4000–600 cm<sup>-1</sup> region and were acquired with a resolution of 4 cm<sup>-1</sup> and a total of 32 scans per spectrum. Equal proportions of the two pastes were well mixed with a spatula and a small amount of each mixed composite was placed between two NaCl crystals, covered with a transparent film of polyethylene, and pressed to produce a thin film. The FTIR spectrum was recorded 24 h after initial mixing of the pastes. Four samples were studied in each case.

For each spectrum it was determined the height of aliphatic C=C peak absorption at 1637 cm<sup>-1</sup>, and the aromatic C..C peak absorption at either 1609 cm<sup>-1</sup>, utilizing a baseline technique which proved the best fit to the Beer–Lambert law.<sup>12</sup> The aromatic C..C vibration is used as internal standard. The percent monomer conversion of the cured specimen, which expresses the percent amount of double carbon bond reacted, is determined according to the equation:

$$\text{Degree of conversion (\%)} = 100 \left[ 1 - \frac{(A_{1637}/A_{1608})_{\text{polymer}}}{(A_{1637}/A_{1608})_{\text{monomer}}} \right]$$

## Sorption of water and ethanol/water solution—solubility–volumetric change

No matter the results which resin-composite materials show in laboratory, it is basic to know how they will behave *in vivo*. The mouth conditions are rather complicated, as they should since biological terms control many parameters. Different organisms, various eating, and hygiene habits result in unlike environments. The food simulating liquids (FSLs) are a model of chemical simulation in testing. Solvents like water, ethanol, ethanol/water, oils, artificial saliva, etc., are chosen to transfer the mouth scene *in vitro*. After solvent sorption during storage in the solvent 37°C for one month and isochronal desorption circle at 37°C, physical properties can be determined for characterizing the materials towards their chemical tolerance.



**Figure 2** Mechanism of curing initiation by the BPO/amine system.

Sorption and solubility tests were determined according to the method described in ANSI/ADA Specification No. 27-1993 for resin-based filling materials which is identical to ISO 4049-1988. Specimen discs were prepared by filling a Teflon mold (15 mm in diameter and 1 mm in thickness) with the mixed material. After removing excess polymer by polishing them and extra moisture by leaving them at oven for a week, they were inserted in capped vessels and kept for a month. Weight and dimensions were determined before and after sorption/desorption took place according to our previous work. Four specimen discs were prepared for each composite material.

The percentage amount of water or ethanol/water solution (75 vol %) sorbed (WS (%) or EWS (%)) and desorbed (WD (%) or EWD (%)), the solubility (SL (%)) in these liquids and the volumetric change (VI (%)) were determined according to the method described in details in our previous works.<sup>10,13,14</sup>

### Thermogravimetric analysis

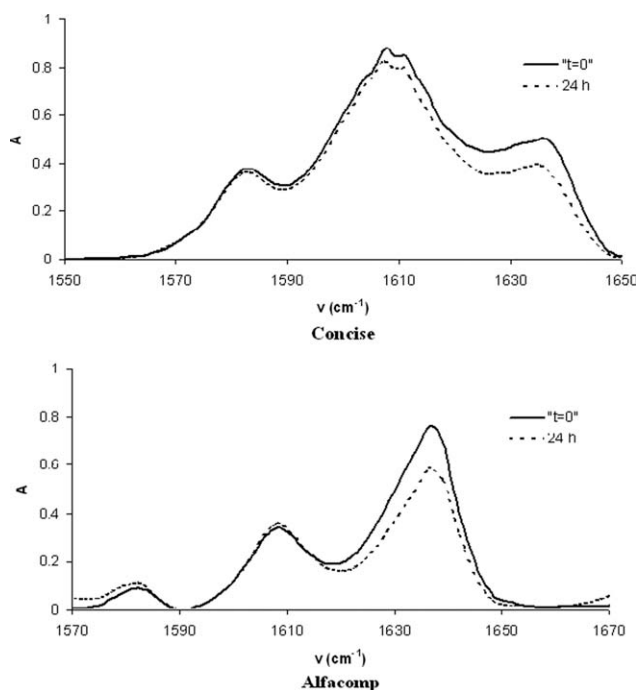
A simple but fundamental analysis is the Thermogravimetric one (TGA). The samples go through heating under specific conditions in order to decompose, when their weight is constantly recorded. It allows the analyst to characterize them as for the heating-resistance, the number of the steps of decomposition the temperature ranges where the phenomena occur

and the corresponding mass losses, whether plateaus are formed on the TG curves and more. Important parameters are the purge gas flowing in the Thermobalance (nitrogen leads to pyrolysis, while oxygen to combustion), the heating rate (since the slower the rate is the more definite facts take place), the amount, and the shape of the sample (affecting the thermal conductance). When decomposition appears multiple steps, differential analysis is useful for clearing data up.

TGA was performed on a Pyris 1 TGA (Perkin-Elmer, USA) thermal analyzer. Samples are prepared following manufacturer's instructions and are placed in desiccator for removing moisture. A day later a cubic piece weighting 10 mg approximately is transferred at the platinum pan for the thermoanalysis. The purge gas was oxygen with flow rate 20 mL/min while the temperature range was 50–800°C with heating rate 10 °C/min. After data collecting, cooling and pan cleaning, the next sample was inserted. Two specimens were used for each composite.

### Statistical analysis

The values reported in all following Tables and Figures represent mean values  $\pm$  standard deviation of replicates. One-way analysis of variance (ANOVA) test, followed by a Tukey's test, for multiple comparisons between means to determinate significant differences was used at a significance level set at  $P \leq 0.05$ , for analysis of the experimental results.



**Figure 3** The FTIR spectra of composites obtained immediately after the mixing of two pastes and 24 h later.



**TABLE I**  
Sorption/Desorption of Water by the Studied Composites After Immersion in Water for 30 days [means (S.D.)]\*

Composite	Sorption on composite (%)	Desorption on composite (%)	Sorption on polymer matrix (%)	Sorption on composite ( $\mu\text{g}/\text{mm}^3$ )
Concise	0.76 (0.00) <sup>A</sup>	0.79 (0.01) <sup>B</sup>	4.2 <sup>a</sup>	13.12 (3.78) <sup>C</sup>
Alfacomp	1.47 (0.68) <sup>A</sup>	1.46 (0.41) <sup>B</sup>	7.3 <sup>b</sup>	21.28 (2.35) <sup>C</sup>

Number of specimens  $n = 4$ .

Common corresponding uppercase letters in a given column indicate no significant difference ( $P \leq 0.05$ ).

<sup>a</sup> It was calculated based on the polymer matrix content 18% w/w obtained by TGA analysis (Table V).

<sup>b</sup> It was calculated based on the polymer matrix content 20.1% w/w obtained by TGA analysis (Table V).

**TABLE II**  
Solubility and Volume Increase of the Studied Composites After Immersion in Water for 30 Days [Means (S.D.)]\*

Composite	Solubility (%)	Solubility ( $\mu\text{g}/\text{mm}^3$ )	% Volume increase
Concise	0.15 (0.03) <sup>A</sup>	4.15 (0.94)	0.70 (0.19) <sup>B</sup>
Alfacomp	1.24 (0.69) <sup>A</sup>	12.89 (1.34)	1.09 (0.91) <sup>B</sup>

Number of specimens  $n = 4$ .

Common corresponding uppercase letters in a given column indicate no significant difference ( $P \leq 0.05$ ).

## RESULTS

The mechanism of self-curing of composites initiated by the BPO/DMT system is presented in Figure 2.

The degree of carbon-carbon double bond conversion of Concise  $73.63\% \pm 4.33\%$  and Afacomp  $47.75\% \pm 1.80\%$  was determined by transmission FTIR spectroscopy in thin films, 24 h after the mixing of the two pastes. The FTIR spectra of composites were obtained immediately after the mixing of the two pastes and 24 h after mixing are shown in Figure 3.

In Table I the sorption of water ( $37 \pm 1$ )°C by the studied composites after immersion in water for 30 days is shown. The solubility in water and the volume increase is shown in Table II.

Analogously, the results obtained for the sorption of EtOH/H<sub>2</sub>O solution 75% v/v ( $37 \pm 1$ )°C are shown in Tables III and IV.

In Figure 4(a) the sorption (%) of studied composites after immersion in water or EtOH/H<sub>2</sub>O solution 75% v/v ( $37 \pm 1$ )°C is compared, in Figure 4(b) the solubility (%) is compared and finally in Figure 5 the volume increase (%) is compared.

In Figure 6 the thermograms of the TGA and the thermograms of the first derivative (dTGA) of composites are shown.

The temperatures with the maximum degradation rate of each step of thermal degradation and the corresponding weight loss of the studied composites are shown in Table V.

## DISCUSSION

In the self-curing "two part" composites one of the parts incorporates the amine and the other the BPO and a stabilizer to control the start of polymerization, usually butyl hydroxytoluene (BHT). When the two parts are mixed in air at room temperature, the monomers of the mixture start to polymerize after a short time and the material starts to harden. The desired mixing time and hardening time, which is defined by the clinical application, can be adjusted by varying the content of the BPO, stabilizer, and the amine. The BPO/DMT redox system has been used and studied for a long time. The results obtained showed that a cyclic transition molecular

**TABLE III**  
Sorption/Desorption of Ethanol/Water Solution 75% v/v by the Studied Composites After Immersion for 30 Days [Means (S.D.)]\*

Composite	Sorption on composite (%)	Desorption on composite (%)	Sorption on polymer matrix (%)	Sorption on composite ( $\mu\text{g}/\text{mm}^3$ )
Concise	1.25 (0.28) <sup>A</sup>	1.31 (0.39) <sup>C</sup>	6.9 <sup>a</sup>	22.65 (0.04) <sup>E</sup>
Alfacomp	2.19 (0.35) <sup>B</sup>	2.34 (0.42) <sup>D</sup>	10.9 <sup>b</sup>	41.60 (5.82) <sup>F</sup>

Number of specimens  $n = 4$ .

Common corresponding uppercase letters in a given column indicate no significant difference ( $P \leq 0.05$ ).

<sup>a</sup> It was calculated based on the polymer matrix content 18% w/w obtained by TGA analysis (Table V).

<sup>b</sup> It was calculated based on the polymer matrix content 20.1% w/w obtained by TGA analysis (Table V).

**TABLE IV**  
Solubility and Volume Increase of the Studied Composites After Immersion in Ethanol/Water Solution 75% v/v for 30 Days [means (S.D.)]\*

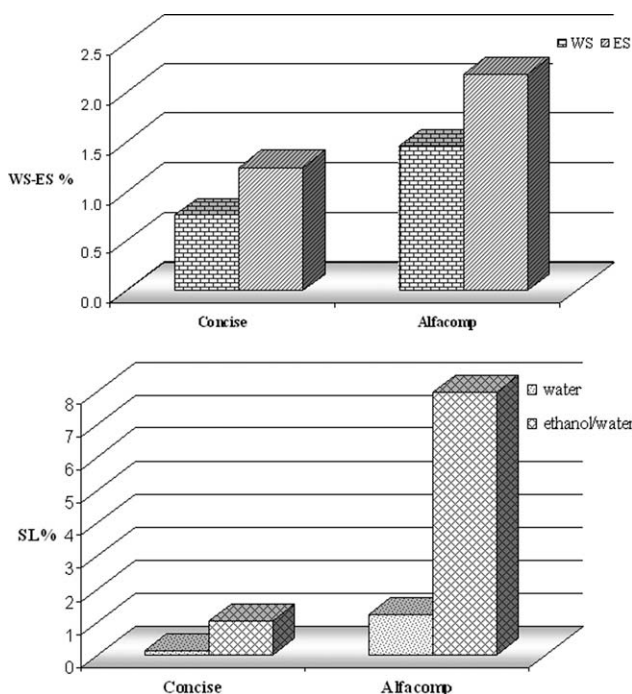
Composite	Solubility (%)	Solubility ( $\mu\text{g}/\text{mm}^3$ )	% Volume increase
Concise	1.04 (0.67) <sup>A</sup>	15.89 (13.91) <sup>C</sup>	1.52 (0.31) <sup>E</sup>
Alfacomp	7.97 (1.95) <sup>B</sup>	150.76 (33.62) <sup>D</sup>	2.81 (0.29) <sup>F</sup>

Number of specimens  $n = 4$ .

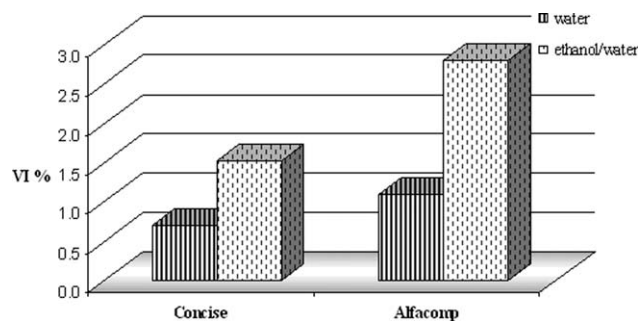
Common corresponding uppercase letters in a given column indicate no significant difference ( $P \leq 0.05$ ).

complex and the subsequent ion pair are formed as shown in Figure 2. It is accepted that both the aminoalkyl radicals and benzyloxy radicals (RO $\cdot$ ) are efficient initiators for free-radical polymerization.

The free-radical polymerization of the dimethacrylate monomers leads to a three-dimensional network. In general, even at a high monomer conversion, not all double bonds are consumed. In present commercial composites, it has been verified by infrared spectrophotometry that 25–55% of the methacrylate groups remain unreacted after polymerization.<sup>9</sup> In the literature, it is reported that Concise showed a degree of conversion  $59\% \pm 4\%$  obtained by multiple internal reflection FTIR spectroscopy.<sup>15</sup> Since this technique is essentially a surface measuring technique, the spectrum reveals the absorption in the

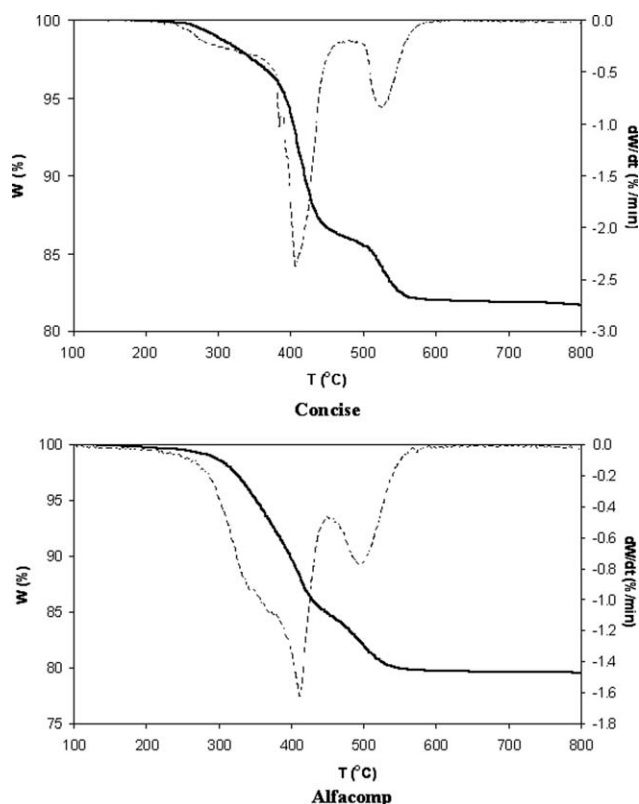


**Figure 4** (a) Sorption of water (WS) or ethanol/water solution 75% v/v (ES) 37°C by the studied composites after immersion in the liquid for 30 days. (b) Effect of the liquid medium aging on solubility (SL) of studied composites after immersion in water or ethanol/water solution 75% v/v for 30 days.



**Figure 5** Volume increase (%) (VI) of studied composites after immersion in water or ethanol/water solution 75% v/v at 37°C for 30 days.

actual region representative of the top five microns of the specimen. The composite was kept at 37°C for 24 h after mixing of the two pastes and then the spectrum was recorded. The degree of carbon-carbon double bond conversion of Concise was also determined by Eliades et al. using also the multiple internal reflection FTIR spectroscopy and found a lower value of  $52 \pm 5.6$ .<sup>16</sup> Ferracane et al. determined the degree of conversion in the unfilled resin of Concise after removal of the filler. Thin film of the resin was cured at 37°C for 24 h. The degree of conversion determined by transmission FTIR was found to be  $63\% \pm 1.4\%$ .<sup>17</sup> Finally, the degree of



**Figure 6** TGA and dTGA curves of studied composites Concise and Alfacomp in oxygen atmosphere. W%: weight loss and  $dW/dt$  (% weight loss/min).

**TABLE V**  
**Temperatures (°C) with the Maximum Degradation Rate of Each Step of Thermal Degradation of Studied Composites**

Composite	1st weight loss		2nd weight loss		3rd weight loss		Three steps WL <sub>1-3</sub> (%)	At 800°C WL <sub>800</sub> (%)
	T <sub>1</sub> (°C)	WL (%)	T <sub>2</sub> (°C)	WL (%)	T <sub>3</sub> (°C)	WL (%)		
Concise	298	1	406	13	528	3.9	17.9	18.0 <sup>a</sup>
Alfacomp	341	4	413	11.3	502	4.8	20.1	20.1 <sup>b</sup>

<sup>a</sup> The content of the organic matrix according to the manufacturer is 22% w/w.

<sup>b</sup> The content of the organic matrix according to the manufacturer is 19% w/w.

conversion of Concise was also determined in thick films (0.4 mm) produced by placing a small amount of composite between two microscope slides that were then compressed. Degree of conversion was found to be 57.7%.<sup>18</sup> No data were found in literature for degree of conversion of Alfacomp.

The degree of conversion of Concise and Aflacomp obtained in the present work in thin films is correspondingly 73.63% ± 4.33% and 47.75% ± 1.80%. The higher degree of conversion of Concise than that of Alfacomp must due to the different structure of their polymer matrix. In Concise, the polymer matrix consists of Bis-GMA and TEGDMA, while in Alfacomp it consists only of Bis-GMA. It is known that Bis-GMA with the rigid aromatic nuclei shows much lower degree of conversion than TEGDMA with the flexible aliphatic group, due to the lower mobility of the polymerizing system, which depends on the glass transition temperature ( $T_g$ ) of the formed polymer network and mainly on the  $T_g$  of the unreacted monomer.<sup>9</sup> The much higher degree of conversion of Concise than that of Alfacomp shows that it is a better material and is expected to show also better mechanical and physical properties and probably better clinical performance.

The water absorption of any polymeric material, whether filled or not, is of importance for dental applications. The water ingress into polymers can have both detrimental and beneficial effects on the properties of the material. Water exposure also may decrease the life of resin composites by silane hydrolysis and microcrack formation. Excessive water uptake can promote breakdown causing a filler-matrix debonding. It is thus important to investigate the water absorption behavior of resin composite filling materials.

The water sorption of the studied composites was determined after immersion of composites in water at 37°C for 30 days, because it has been found that this time is enough in order to achieve about sorption-equilibrium.<sup>19</sup> Assuming that the polymer matrix absorbs the bulk of the water, the values for the water uptake on the polymer matrix alone were also calculated (Table I). Concise showed a lower water sorption on polymer matrix (4.2% w/w) than

Alfacomp (7.3% w/w). We must remember that in Concise the polymer matrix consists of Bis-GMA and TEGDMA, while in Alfacomp it consists only of Bis-GMA. In a recent work, we have found that poly-TEGDMA absorbs higher amount of water (6.23% ± 0.15%) than poly-Bis-GMA (3.73% ± 0.11%).<sup>20</sup> This behavior was attributed to the fact that poly-TEGDMA network is more heterogeneous than poly-Bis-GMA and this higher heterogeneity seems to favor the higher water sorption of the former. In a more heterogeneous network, the space created between the polymer clusters (microporous) is larger and can accommodate a larger quantity of water. The higher water sorption of poly-TEGDMA than that of poly-Bis-GMA may also be due, to some extent, to the higher flexibility of the network of the former than the latter which permits the higher swelling of polymer chains by water.<sup>20</sup> According to these data, we would expect Alfacomp to show lower water sorption on polymer matrix than Concise and close to that of poly-Bis-GMA. Such higher water absorption values for the filled specimens than which would be expected on the basis of the resin content was reported for Bis-GMA-based composites incorporating untreated and surface-treated hydroxyapatite.<sup>21</sup> This was attributed to the fact that additional amount of water can be accommodated at the interface between filler particles and the matrix, where the presence of microvoids is quite probable due to a lack of impregnation of filler particles with the polymer matrix. On the other hand, we have to allow for the possibility that the filler phase may contribute to the water absorption due to its surface coating of silanol (Si—O—H) groups, which will absorb water onto its surface. Also in two paste systems, as our composites, which require mixing manually, have air incorporated into the matrix. The presence and size of these air bubbles can be minimized with an appropriate mixing technique but not eliminated. It can be hypothesized that these air bubbles may be filled with water.<sup>22</sup>

For both studied composites, the amount of water which desorbed was similar with that sorbed (Table I).

The solubility of composites is concerned to the leachable by the water amount of unreacted

monomer. Alfacom showed higher solubility than Concise (Table II) and this must be due to the lower degree of conversion observed for Alfacom. It has been found that the higher the degree of conversion and thus higher the amount of unreacted monomer, the lower the solubility value.<sup>23</sup> It is obvious that from clinical point of view it is better for a composite to have a lower solubility.

The absorbed water diffuses into the polymer network of composites and separates the chains creating expansion. The percentage volume increase of Alfacom was higher than that of Concise most probably due to the higher amount of absorbed water by Alfacom. It has been found that the percentage volume increase of dental resins follow linear dependence with the percentage water absorbed.<sup>10</sup>

The study of sorption by the composites of ethanol/water solution 75% (v/v) 37°C, which is a good food-simulating solvent, showed that Concise have lower liquid sorption on polymer matrix (6.9% w/w) than Alfacom (10.9% w/w). This result is surprising since it was found that neat Bis-GMA-resin absorbs 9.77%  $\pm$  0.16% w/w ethanol/water solution 75% (v/v) and neat TEGDMA-resin absorbs 10.10%  $\pm$  0.06% w/w of this solution.<sup>20</sup> Solubility in ethanol/water solution and percentage volume increase of Concise was much lower than those of Alfacom (Table IV).

Both studied composites showed higher values for sorption, solubility, and volume increase in the case of ethanol/water solution 75% (v/v) than in water (Figs. 4 and 5). This is because ethanol/water solution is a better solvent for the polymer matrix of composites than water.

Thermogravimetric analysis of composites showed the degradation of the polymer matrix in three steps (Fig. 6 and Table V). The observed differences in degradation temperatures and the corresponding weight losses are due to the different structure of the polymer matrix of composites. The total weight loss occurred at three steps corresponds to the organic content of composites. No other weight loss was observed at higher temperatures up to 800°C. By contrast the organic content of Concise was found to be 18.0% w/w, while the manufacturer gives a content of 22.0% w/w. On the contrary, the organic content of Alfacom was found to be 20.1% w/w, while the manufacturer gives a content of 19.0% w/w.

## CONCLUSIONS

The degree of conversion of Concise (73.63%  $\pm$  4.33%) was found to be much higher than that of Alfacom (47.75%  $\pm$  1.80%). Also the sorption, solubility, and volume increase of Concise after immersion in water or ethanol/water 75% v/v solution 37°C for 30 days was lower than the corresponding ones of Alfacom. The organic content of Concise was found to be 18.0% w/w and that of Alfacom 20.1% w/w. So, the different behavior of the studied composites must be due to the differences in the chemical structure of the organic resin matrix and the type of inorganic filler.

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