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Composite resin and water sorption test:

in vitro study

Marco Suardi

Dissertação conducente ao Grau de Mestre em

Medicina Dentária (Ciclo Integrado)

Gandra, 30 de junho de 2020

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Trabalho realizado sob a Orientação de Dr.ª Lígia Rocha

Co-orientador Professora Doutora Orlanda Torres

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Eu, **Ligía Rocha** , com a categoria profissional de Monitora Clínica do Instituto Universitário de Ciências da Saúde, tendo assumido o papel de Orientador da Dissertação intitulada **"Composite resin and water sorption test: *in vitro study"*** do Aluno do Mestrado Integrado em Medicina Dentária, **Marco Suardi**, declaro que sou de parecer favorável para que a Dissertação possa ser depositada para análise do Arguente do Júri nomeado para o efeito para Admissão a provas públicas conducentes à obtenção do Grau de Mestre.

Gandra, 30 de junho de 2020

Lígia Rocha

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Thanks.

RESUMO

As resinas compostas apresentam características higroscópicas, que podem influenciar o seu desempenho funcional e estético. Esta propriedade é determinada pela composição da matriz orgânica e inorgânica de cada tipo de resina. Portanto, os testes de sorção de água e de solubilidade são muito importantes do ponto de vista clínico. É descrito na literatura que a quantidade de absorção de água pelas resinas compostas influência as suas propriedades mecânicas.

Desenho do estudo

Numa amostra total de 15 discos (1 mm de espessura; 12 mm de diâmetro)) foram preparadas a partir de três tipos de resina composta.

Os testes de sorção de água e de solubilidade das amostras foram obtidos de acordo com as especificações das normas ISO 4049:2009, após o armazenamento em água destilada por 30 dias.

Os dados obtidos dos resultados serão analisados pelo método estatístico ANOVA.

OBJETIVOS DO TRABALHO

Este estudo de investigação pretende avaliar as propriedades mecânicas de sorção de água e o comportamento de solubilidade de três tipos de resina composta.

PALAVRA CHAVE

Adsorção; Absorção; Resinas compostas; Solubilidade.

ABSTRACT

Composite resins present hygroscopic characteristics, which may influence their functional and aesthetic performance. This property is determined by the composition of the organic and inorganic matrix of each type of resin. Therefore, water sorption and solubility tests are very important from a clinical point of view. It is described in the literature that the amount of water sorption by the composite resins influences their mechanical properties.

Study design

In a total sample of 15 discs (1mm thick; 12mm diameter) have been prepared from three types of composite resin.

The water sorption and solubility tests of the samples have been obtained according to the specifications of ISO 4049:2009 standards, after storage in distilled water for 30 days.

The data obtained from the results has been analyzed by the ANOVA statistical method.

WORK OBJECTIVES

The aim of this research study is evaluated the mechanical properties of water sorption and the solubility behavior of three types of composite resin.

KEY WORDS

Adsorption; Absorption; Composite resins; Solubility.

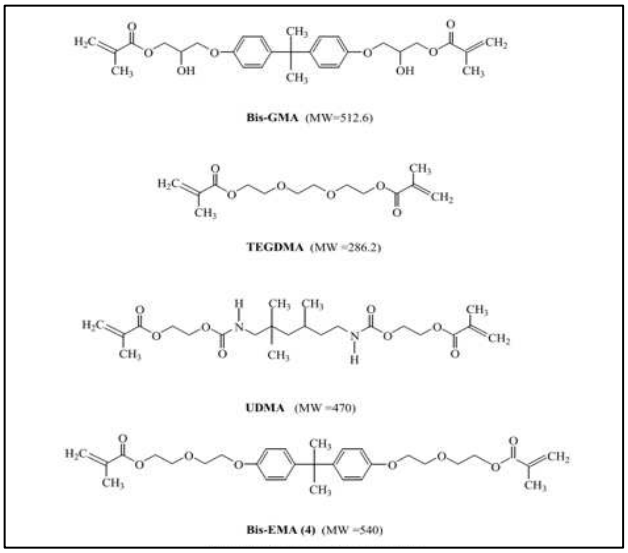
TABLE OF CONTENTS

KEY TERMS

BPA	Bisphenol A
bis-GMA	Bisphenol-A-glycidylmethacrylate
UDMA	Urethane dimethacrylate
bis-EMA	Bisphenol-A-polyethylene glycol dimethacrylate
TEGDMA	Triethylene glycol dimethacrylate
vol. %	Volume
wt. %	Weight
WSp	Water Sorption
WSl	Water solubility
S	Saturation
®	Registered brand
°C	degrees Celsius
mW	milliWatt
M1	conditioned mass, in micrograms, before immersion in water
M2	mass of the specimen, in micrograms, after immersion in water for 7 days
M3	mass of the reconditioned specimen in micrograms
V	volume of the specimen, in cubic millimeters

1. INTRODUCTION

Composite resins are now considered to be the best materials available to professionals for direct restorations. They are used for a variety of applications in dentistry. (1) These composite resins, when used correctly, provide excellent aesthetic and functional results and have many advantages, excellent aesthetic quality and the ability to adhere to enamel. (2) Over time, since their appearance until today, they have undergone a continuous improvement. Today, further progress in the filler component has led to micro-filled and nanocomposites, hybrid composites, conventional and flowable composites, for example. (3) The evolution has focused on the polymeric matrix of the material with the intent to reduce shrinkage during the polymerization phase and to increase adhesion to dental structures. (4) However, these materials are not free from defects: among these, water sorption is considered a decisive property, (1) fundamental for the success or failure of the restoration in a very complex environment such as the oral one,(4) for the ability to modify the mechanical properties of the resin itself. (2) Dental composites use a mixture of monomers that are aromatic or aliphatic dimethacrylates. Of these, bis-GMA (bisphenol-A-glycidyl-methacrylate), urethane dimethacrylate (UDMA) and bis-EMA (bisphenol-A-polyethylene glycol dimethacrylate) are the most common used. The addition of triethylene glycol dimethacrylate (TEGDMA) is used to control viscosity. (2) (Scheme 1)



Scheme 1. Structures of monomers used.

The Bis-GMA was introduced in the early 1960s. Its properties were superior to those of acrylic resins. However, it had some limitations. (1) Thus composite resins must be mixed with different monomers to optimize their properties. (2) Bis-EMA and UDMA-Bis-EMA based resins appear to be more hydrophobic and resistant to absorption and solubility than Bis-GMA based systems.(5)

The mechanical properties therefore depend on the choice of the percentages of these components. (6) Fillers play a crucial role. Most of the important properties of resin is determined by its filling content. Composite fillers are classified by material, shape and size. (4) (fig. 1)

The addition of filler particles in the resin significantly improves its properties. (7) To increase the amount of filler in the resin, it is necessary to add fillers in a different range of sizes. If a single particle size is used, there will be a space between them. The smaller particles can fill these spaces, by increasing the filler content. (4) The shape influences the filler load and the workability characteristics of the composite. The filler content is indicated as a percentage of volume (vol. %) or as a percentage of weight (wt. %)

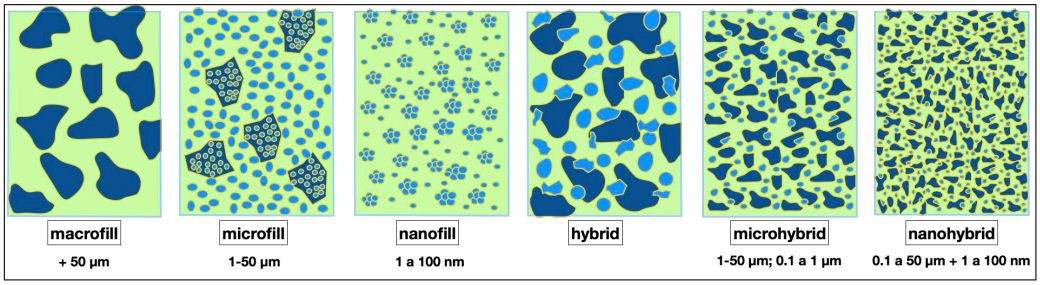


Fig.1 Effect on particle size on surface smoothness.

The percentage of weight is usually higher in value than the volume percentage.(1) Water sorption and solubility of resin composites depend on the material and are strongly influenced by filler loading and properties of polymer matrix. (5) However, attention should be focused on the composition of the resin matrix and not just in the filling system to be able to evaluate the behavior of the resins. (8) The water sorption of hybrid composites is relatively lower than micro-filled resins. ISO 4049:2009 requirements limit water sorption acceptable when WS was ≤ 40 $\mu g/mm^3$ and WL was ≤ 7.5 $\mu g/mm^3$ (2,9,10) Studies have shown that water is mainly absorbed within the resin matrix and is more influenced by the structure and quantity of this phase, so the study of water sorption and solubility of poly-dimethacrylate resins is important to understand their behavior in dental composites. (2,4,7)

Distilled water and artificial saliva are generally comparable as storage methods in terms of water sorption and antimicrobial additives have no influence.(11)

2. AIM AND HYPOTHESES

Accordingly, the present study aimed to measure the water sorption and solubility of different core build-up materials. The null hypotheses tested were:

1. There is no difference in the water sorption and solubility of the tested core materials after 1 week.

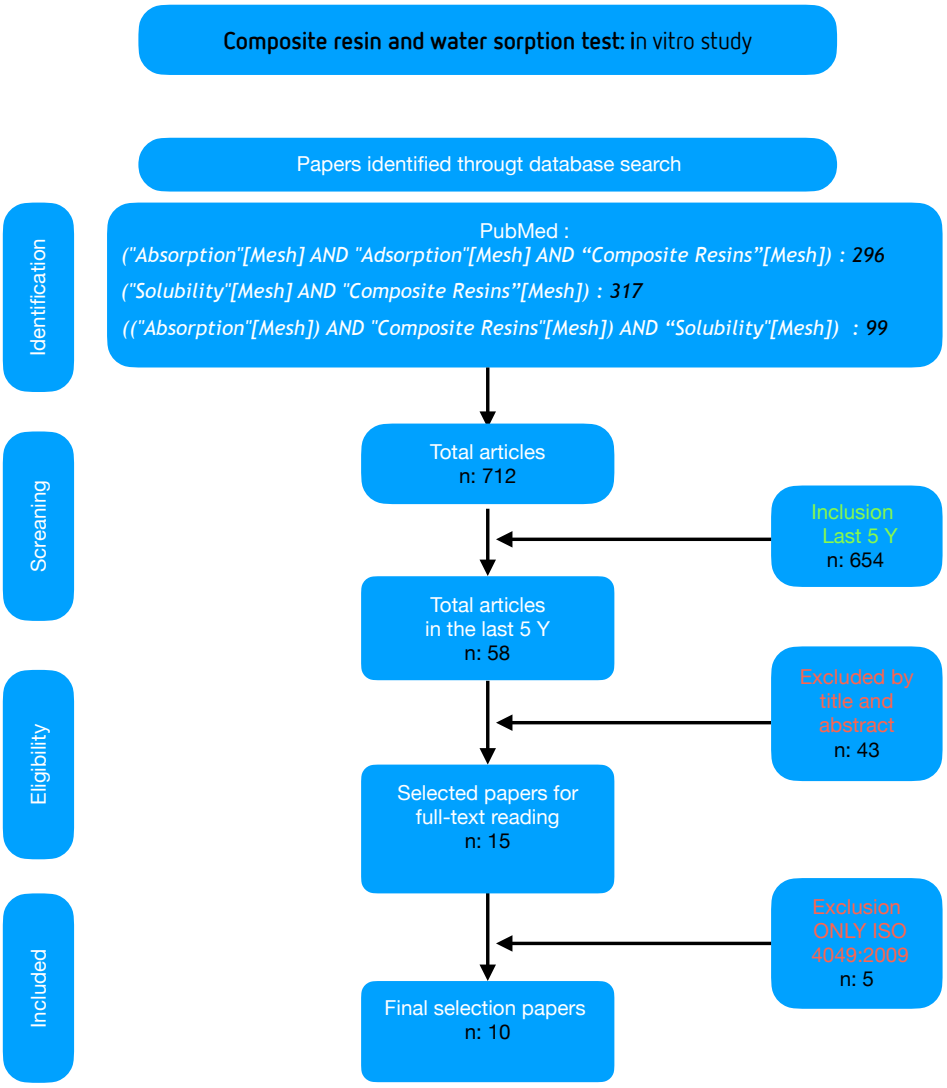
2. There is no difference in the water sorption and solubility of the tested materials between 1 week total immersion and 1 month total immersion.

3. MATERIALS AND METHODS

3.1 PAPER SELECTION

The bibliographic research was performed at PUBMED (via National Library of Medicine) using the following combination of research terms: "Adsorption"[Mesh] - AND - Absorption"[Mesh] - AND -"Composite Resins"[Mesh] - AND –

"Solubility"[Mesh]. Eligibility inclusion criteria used in article research involved: articles written in English; meta-analyses; randomized clinical trials; prospective cohort studies; integrative literature reviews on the water absorption difference of bisphenol A resins. Titles and abstracts of potentially relevant articles were independently analyzed. The total number of articles was compiled for each combination of key terms and therefore duplicates were removed using Mendeley. A preliminary evaluation of the abstracts to determine if the articles met the objective of the study. The selected articles were read and evaluated individually for the purpose of this paper. (PRISMA 2009-scheme 2)



Scheme 2. Papers identified througt database search -PRISMA 2009-

3.2 DISCS PREPARATION

In vitro scientific research work on water sorption and solubility tests for three types of Bisphenol A (BPA) resins: Brilliant ever glow® (Coltene, Switzerland), IPS Empress® Direct (Ivoclar Vivadent; Schaan, Liechtenstein), Filtek™ Supreme XTE (3M ESPE Dental Products, St. Paul, MN)(Table 1-2) The entire protocol was based on ISO 4049:2009. (10)

Table 1- All tested materials and manufacturers information.

MATERIAL	CODE	LOT NUMBER	MANUFACTURER	TYPE
Filtek™ Supreme XTE	CT enamel	NA28205	3M ESPE Dental Products, St. Paul, MN	NANOFILLER
IPS Empress® Direct	A2 enamel	Y28474	Ivoclar Vivadent;Schaan, Liechtenstein	NANO HYBRID
Brilliant ever glow®	A2 enamel	J18375	Coltene,Switzerland	SUBMICRON HYBRID

Table 2- All tested materials, their composition and manufacturers data.

MATERIAL	ORGANIC MATRIX	FILLER wt%	FILLER vol%	FILLER DIMENSION	WSP	WSL
Filtek™ Supreme XTE	bis-GMA, UDMA, TEGDMA, bis-EMA	65	46	0.6 to 20 microns		
IPS Empress® Direct	UDMA, TCDD, Bis-GMA	78.1	52/59	0.1 to 0.3 microns	19.6	<0.1
Brilliant ever glow®	Bis-GMA, Bis-EMA	79	64	0.4 to 0.7 microns	15.1	<0.1

The sampler (Styleitaliano™ products) allows to make samples of composite discs with the desired thickness in cylindrical molds: discs with 1 mm thickness x 12 mm diameter of each material (n = 5) have been created. All discs are polymerized with the same equipment high intensity LED technology (Celalux, Voco GmbH, Cuxhaven, Germany) of 1300 mW / cm^2, 450 / 480 nm for 20 seconds on each face, before each polymerization was confirmed the power of the equipment through the radiometer. (Fig. 2) Finishing and polishing was performed on all samples. (Fig. 3)

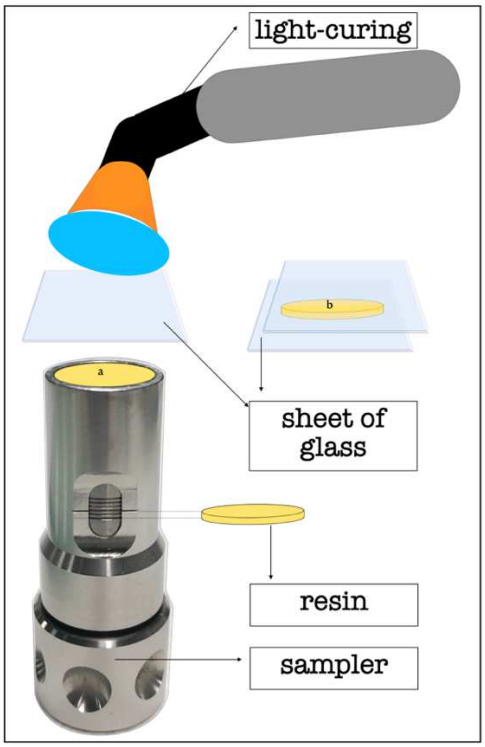


Fig. 2

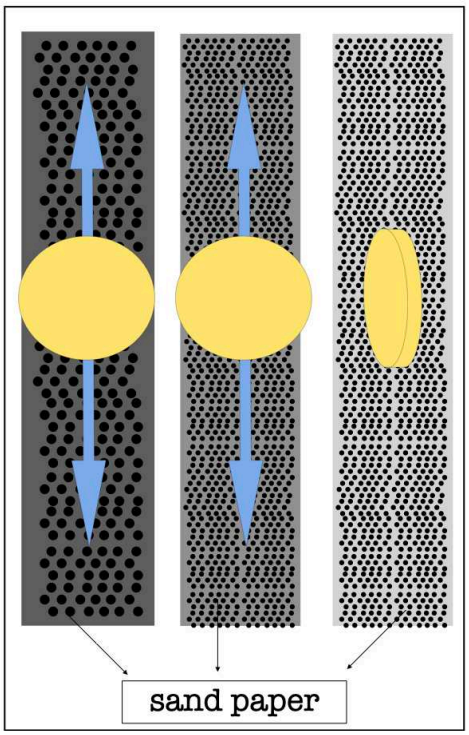


Fig. 3

Fig. 2 Production process of the discs. Sampler resin application, placement of a glass sheet and light-curing (side a), removal from the sampler and light-curing (side b).

Fig. 3 Surface polishing through a coded protocol. 5 steps each side for three different granulometry.

The thickness and diameter of the disks were measured with digital electronic caliper. (Mitutoyo Corporation, Japan) (Fig. 4) Mean values were used to calculate the volume of each specimen in mm^3.

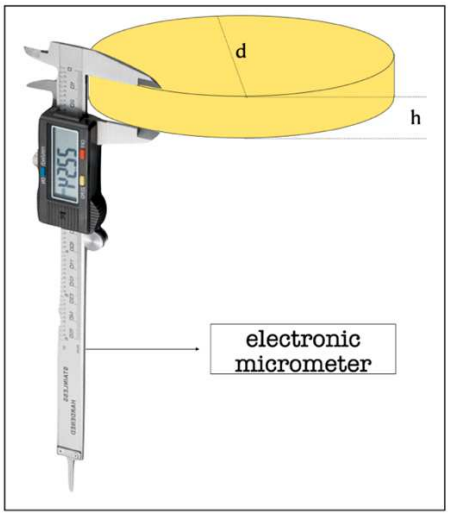


Fig. 4

Fig. 4 Thickness and diameter measurement with digital electronic caliper.

3.3 EXPERIMENTAL DESIGN

The discs were stored in glass containers protected from light with silica for 27 days in a desiccator maintained at (37 ± 1) ◦C. until a constant mass (M1) was obtained. (Fig. 5)

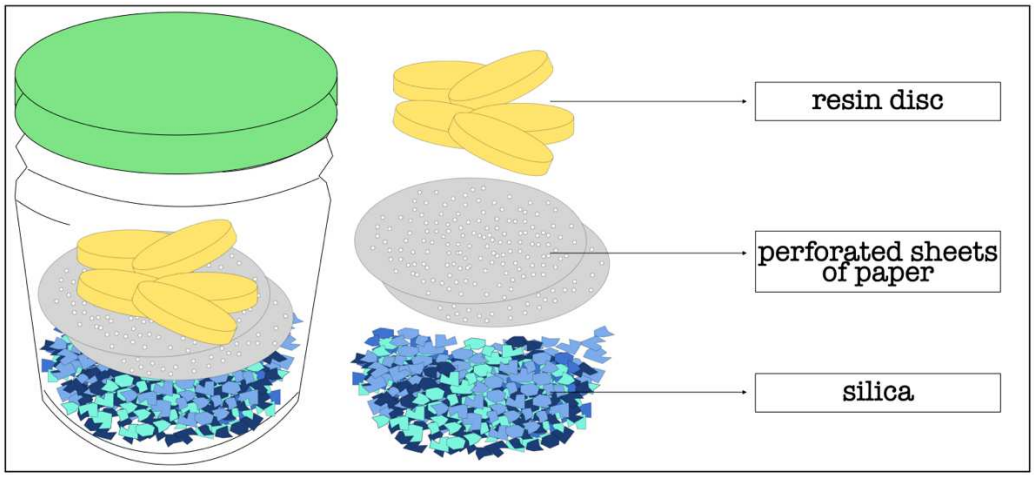


Fig. 5 Glass containers with silica layer and perforated vegetable paper to separate the resin discs placed on the surface.

Specimens were then immersed in distilled water for thirty days and weighed each day (M2). (Fig. 6)

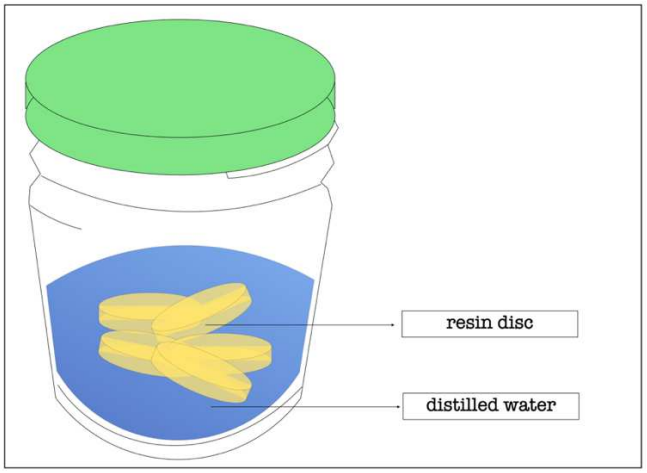


Fig. 6

Fig. 6 Glass containers with resin discs submerged in distilled water.

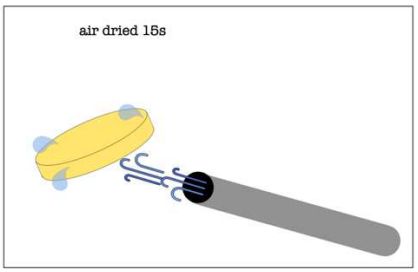


Fig. 7

Fig. 7 Once removed from the distilled water to be weighed were dried with compressed air for 15 seconds.

The specimens were removed from the distilled water and dried (Fig. 7) The weight has been measured with digital analytical balance KERN-ALJ 220-5DNM (Kern & Sohn GmbH, Balingen, Germany). At the end, we recondition the specimens in silica a desiccator-maintained oven at (37 ± 1) ◦C. until a constant mass (M3) is obtained. The design of the study is explained in the timeline (fig.8) The values of sorption and solubility were calculated from these different measurements.

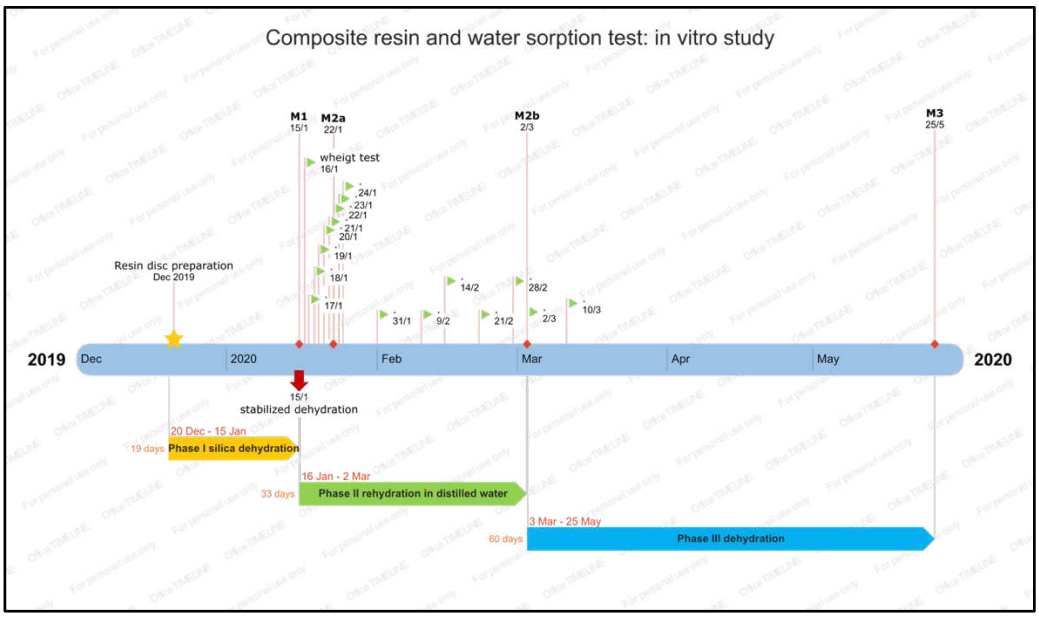


Fig. 8 Timeline of the study.

3.4 WATER SORPTION

Water sorption- Wsp :

$$W_{sp} = \frac{m2\text{-}m3}{V}$$

- M2: is the mass of the specimen, in micrograms, after immersion in water for 7 days;
- M3: is the mass of the reconditioned specimen, in micrograms;
- V: is the volume of the specimen, in cubic millimeters.

3.5 WATER SOLUBILITY

Water Solubility- Wsl:

$$W_{sl} = \frac{m1\text{-}m3}{V}$$

- M1: is the conditioned mass, in micrograms, before immersion in water;
- M3: is the mass of the reconditioned specimen in micrograms;
- V: is the volume of the specimen, in cubic millimeters.

3.6 SATURATION

Saturation- S:

$$S = \frac{m2 - m1}{V}$$

- M2: is the mass of the specimen, in micrograms, after immersion in water for 7 days;
- M1: is the conditioned mass, in micrograms, before immersion in water;
- V: is the volume of the specimen, in cubic millimeters.

3.7 STATISTICAL ANALYSIS

One-way ANOVA were used to detect statistical differences of the ($P < 0.05$) water sorption and solubility of the tested materials after 1 week and after 1 month. . The independent t-test was used to detect any differences between the water sorption and solubility of the 1-week group and the 1-month group for each material. ANOVA repeated measures was used to detect any differences in the sorption mass change percentages over time (1 month) for tested materials ($P < 0.05$).

4. RESULTS

The results of water sorption, water solubility and saturation of composites studied are presented in Table 3. According to ISO 4049:2009 (10), a resin, to be used as dental material, must have a water sorption of less than 40 $\mu g/mm^3$ and a solubility of less than 7.5 mg/mm^3.

The water sorption, solubility and saturation of the tested materials are presented in Table 3 and figures 9, 10, 11, 12. The materials tested differed significantly in both water absorption and solubility.

All materials tested showed a significant change in mass in the first week after the dive, followed by a slower increase in mass until they became balanced after 15 days. The values of water sorption for all studied composites, are within the range of the ISO's standard. Solubility values are outside the range of the ISO (4049:2009) standard (10) for IPS Empress® Direct.

Table 3– Mean (SD) values of water sorption and solubility for all groups, for a total immersion period of 7 days as well as 45 days. Within each column different superscripts indicate statistical significant difference (P < 0.001).

MATERIAL	Water sorption (μg/mm3) after 7 days	Water sorption (μg/mm3) after 45 days	Water solubility (μg/mm3)	Saturation (μg/mm3) after 7 days	Saturation (μg/mm3) after 45 days
Filtek™ Supreme XTE	26.09 (0.1)	28.23 (0.5)	5.49 (0.1)	20.60 (0.2)	22.74 (0.1)
IPS Empress® Direct	22.82 (0.2)	20.67 (0.1)	7.94 (0.5)	14.88 (0.5)	12.72 (0.2)
Brilliant ever glow®	17.10 (0.1)	17.24 (0.1)	6.25 (0.1)	10.85 (0.1)	10.99 (0.1)

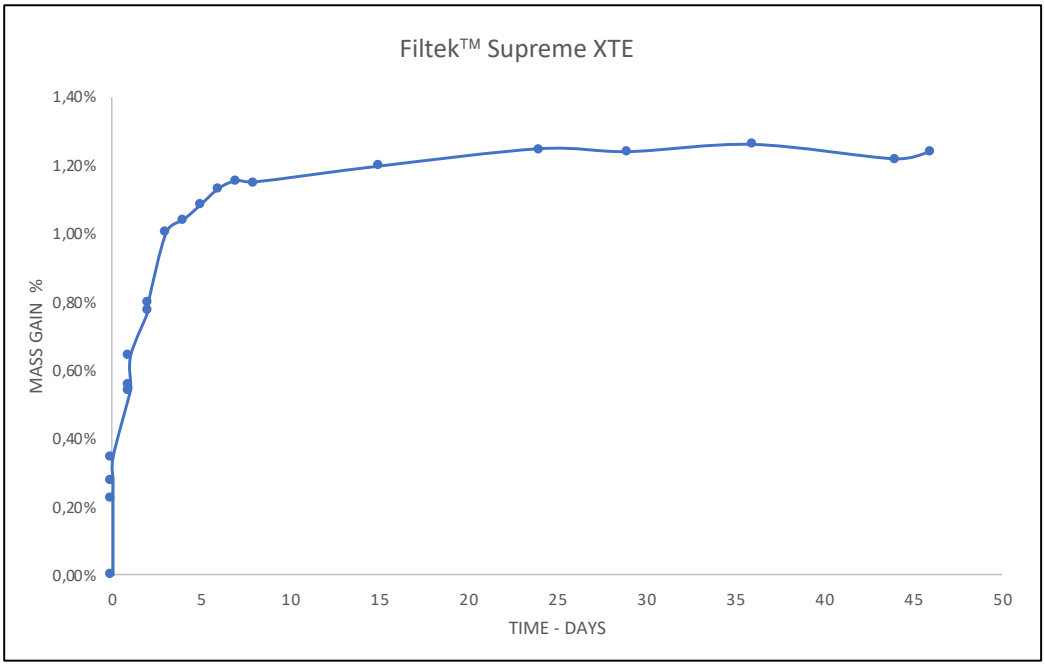


Fig.9 Mass changes % (water sorption cycles) of the Filtek™ Supreme XTE after 45 days.

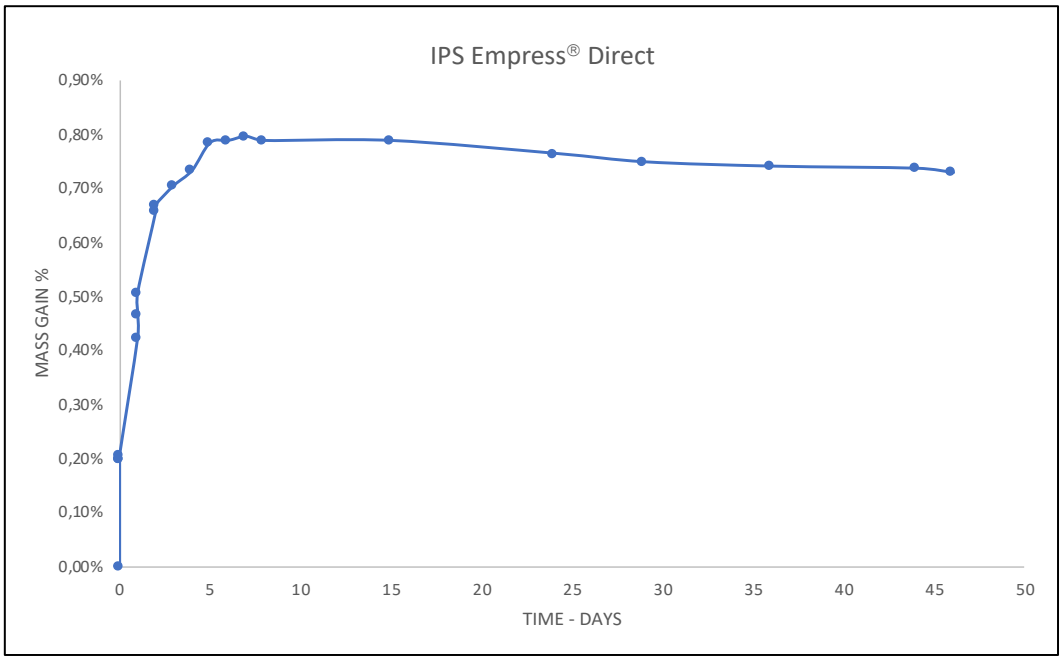


Fig.10 Mass changes % (water sorption cycles) of the IPS Empress® Direct after 45 days.

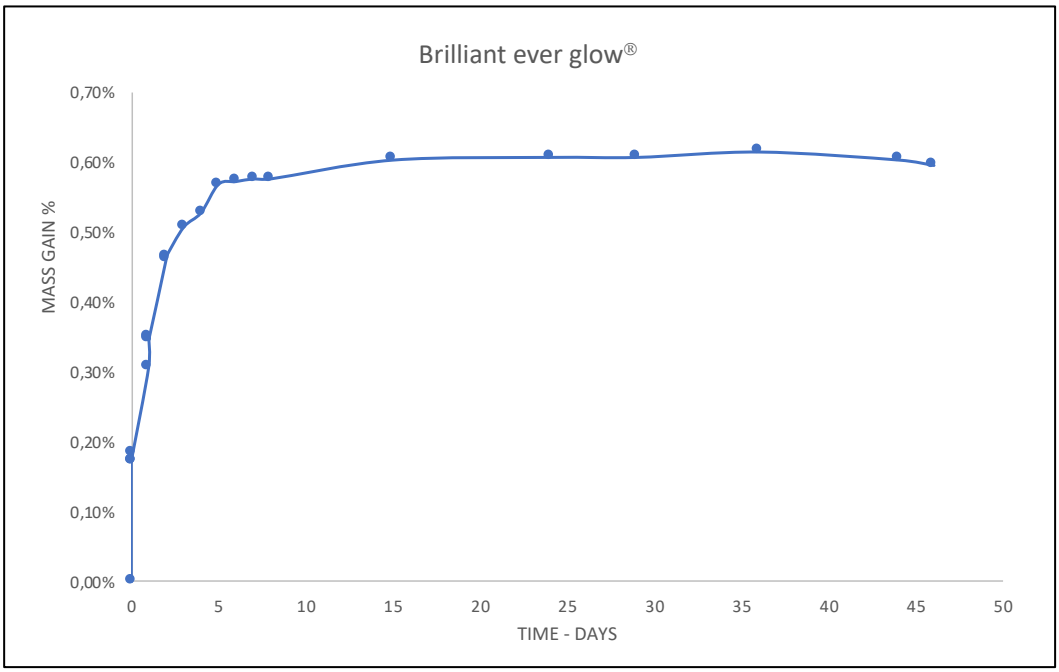


Fig. 11 Mass changes % (water sorption cycles) of the Brilliant ever glow® after 45 days.

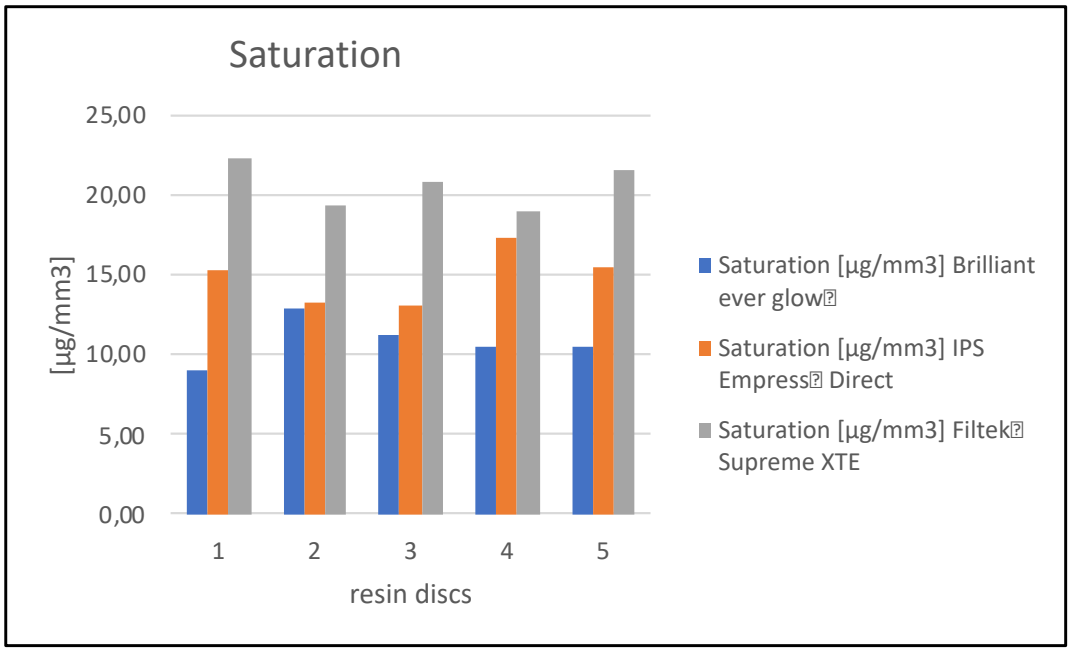


Fig. 12 Saturation (µg/mm³) of the tested materials after 45 days.

5. DISCUSSION

The current study compared the water sorption behavior of various commercial resin-based materials of different compositions. This was done for one month. All materials showed a continuous increase in the amount of water absorbed during the first 15 days, but the maximum amount of water gained for all materials tested was during the first week. This was in accordance with several studies in terms of the maximum amount of water absorption gained during the first week.(12)(12)(13) All materials tested showed significantly different behavior in water absorption. After 1 month all materials tested showed an acceptable water sorption behavior of less than 40 µg/mm³ (the maximum water absorption declared by ISO 4049:2009)(10). All materials showed significantly different solubility in water after a total immersion of 45 days. As for solubility, one material results to be outside the acceptable range of the ISO 4049:2009 protocol (10) which should be less than 7.5 µg/mm³. In summary, the first hypothesis that there is no difference in the water sorption and

solubility of the tested core materials after 1-week was rejected. In the same way, the second hypothesis that there is no difference in the water sorption and solubility of the tested materials between 1-week total immersion and 1-month total immersion was rejected. The differences between the materials can be attributed to their composition. The hydrophilicity of monomers used in resin-based materials can play an important role in the composition of resin compounds. The quantity of water gained in a composite material depends on the content of its hydrophilic monomers. Solubility of all tested materials (except IPS Empress® Direct) was less than 7.5 $\mu g/mm^3$. This means that these materials showed acceptable solubility behaviour (according to ISO 4049:2009)(10). It has been reported that solubility can be related to the water sorption of a material. This is probably due to the fact that incompletely reacted components can only leak out when water penetrates the material (14). The density of cross-linked dimethacrylates may vary. The extent of this variation will determine the amount of residual monomers that can be filter out.(12)(12) It is likely that the difference in solubility between the materials may be due to the existing cross-linkages in each of the materials. (14). The solvent permeability of a polymer may be reduced by the presence of denser cross-links between polymer chains, as they reduce the free volume and reduce the free space in the polymer chain(2). Thus, the more the polymer mesh is cross-linked, the lower the water absorption and solubility of the resin-based material (4). The values measured in this study were compared with those provided by the manufacturers. The comparison basically showed that the data are in line with what was stated. We did not find the official Filtek™ Supreme XTE official data regarding WSp and WSl (15). We compared our data with *Berger* data (16). The values they declared were: WSp 17.1 $\mu g/mm^3$ and WSl -4.0 $\mu g/mm^3$,our values are WSp 28.23 $\mu g/mm^3$ and WSl 5.49 $\mu g/mm^3$. We've had significantly higher results for both parameters. The official IPS Empress® Direct data is WSp 19.6 $\mu g/mm^3$ and WSl <0.1 $\mu g/mm^3$ (17) while in this study the WSp results were: 22.82 $\mu g/mm^3$ after 7 days, which then stabilized at 20.67 $\mu g/mm^3$ after 45 days. The WSl value was outside the ISO 4049:2009 standard(10), 7.94 $\mu g/mm^3$. This is probably due to errors in the production of resin discs where the presence of

bubbles or errors in the surface finishing of the resin has misled this data. Another reason may be a procedural error during measurements or storage of the resin. According to ISO 4049:2009 protocol if three of the values are 7,5 $\mu g/mm^3$, will have to repeat the whole test. The reported values of WSp for Brilliant ever glow® are 15.1 $\mu g/mm^3$ and WSl <0.1 $\mu g/mm^3$ (18). Our results are higher than reported by the manufacturer 17.10 $\mu g/mm^3$ after 7 days and 17.24 $\mu g/mm^3$ after 45 days. Nevertheless, they comply with the ISO 4049:2009 parameters. While WSl parameters are comparable.

6. CONCLUSION

The following conclusions may be drawn from the present work:

- Water sorption and solubility of resin-composites are material-dependent and are highly affected by the filler loading and properties of the polymeric matrix.
- All tested core build-up materials absorbed water and showed significantly differing water sorption behavior.
- Brilliant ever glow® had the lowest water sorption and solubility among the tested materials.
- According to the ISO 4049:2009 standards, all the tested materials showed acceptable water sorption and solubility apart from the water solubility behavior of IPS Empress® Direct. In which all the experimental precedent must be repeated in order to check the sorption and solubility values of Empress®Direct Resin.

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