

## Research Article

## Open Access

## Release of Organic Compounds by Composites Depending on the Source of Curing Light, Immersion Solution and PH

Karina Alessandra Grecca Pieroni<sup>1</sup>, Alexandra Mussolino Queiroz<sup>1</sup>, Marília Pacífico Lucisano<sup>1\*</sup>, Sofia Nikolaou<sup>2</sup>, Juliana Cristina Biazotto<sup>2</sup>, Marcos Samuel Azulay<sup>1</sup>, Patricia Gatón Hernandez<sup>3</sup>, Paulo Nelson-Filho<sup>1</sup> and Lea Assed Bezerra Silva<sup>1</sup>

<sup>1</sup>Department of Pediatric Dentistry, School of Dentistry of Ribeirão Preto, University of São Paulo-USP, Ribeirão Preto, SP, Brazil

<sup>2</sup>Department of Analytical Chemistry, School of Pharmaceutical Science of Ribeirão Preto, University of São Paulo-USP, Ribeirão Preto, SP, Brazil

<sup>3</sup>Catalan Society of Dentistry and Stomatology Academy of Medical Sciences and Health and Catalonia and Balears Barcelona, Espanha

### ABSTRACT

**Objectives:** This in vitro study evaluated the release of organic compounds by composite Kalore™ (GC FUJI) and Filtek™ Silorane (3M ESPE) by means of fluorescence spectrometry, varying the source of curing light, the immersion solution and the pH of these immersions.

**Methods:** 56 specimens were made for each composite, 28 polymerized with halogen light and 28 with LED light. Samples of each composite were stored in water with neutral or acidic pH and in saliva with acidic or neutral pH (n=7 specimens for each condition examined). The reading was performed between 1 and 672 hours (28 days). The results were statistically analyzed using the Tukey and ANOVA tests, adopting the significance level of 5%.

**Results:** The Kalore™ resin released more organic compounds than Filtek™ Silorane in the aqueous environment, either in the polymerization by the halogen light as by the LED light in both pHs (p<0.05). There was no significant difference in aqueous environment, only when polymerized by halogen light and acid pH (p>0.05). When immersed in artificial saliva, the Kalore™ resin also released more organic compounds than Filtek™ Silorane, but there was only statistically significant difference when polymerized by LED light, both in neutral pH and at acid pH. Both resins released greater amount of organic compounds in aqueous solution, compared to saliva, regardless of the light source and pH (p<0.05).

**Conclusion:** Even the most modern resins such as Kalore™ and Filtek™ Silorane release organic compounds and the polymerization by LED light decreases the liberation of organic compounds by Filtek™ Silorane resin.

### \*Corresponding author

Marília Pacífico Lucisano, Departamento de Clínica Infantil Faculdade de Odontologia de Ribeirão Preto, Universidade de São Paulo Avenida do Café s/n, Monte Alegre, 14040-904, Ribeirão Preto, SP, Brazil; Tel: +55-16-3315-4107; E-Mail: marilia.lucisano@forp.usp.br

**Received:** December 10, 2020; **Accepted:** December 18, 2020; **Published:** December 30, 2020

**Keywords:** Composite Resins, Organic Compounds, Fluorescence Spectrometry

### Introduction

Organic compounds (monomers), initiators and additives can be released by composite resins, even after polymerization. The liberation of these substances may occur due to the presence of residual monomers and also the degradation of the material itself. When released into the oral cavity and dental pulp, these substances can have access to other organs, causing cytotoxic, genotoxic and allergens effects and/or estrogenic activity [1-9].

Currently, composites manufacturers have pursued to overcome the effects of the composite polymerization contraction, using monomer of low contraction [10]. The Kalore™ resin (GC FUJI Kasugai, Japan) presents as the inorganic component glass Fluoro-Amino-silicate (pre-polymerized charge of silica and silicon dioxide), the camphorquinone as initiator, DX-511 (a high methacrylate molecular weight - Dupont monomer) the

UDMA and nonspecific dimethacrylate comonomers as organic matrix [10].

The Filtek™ Silorane resin (3M ESPE, Seefeld, Germany) does not present methacrylates in its composition and it has quartz and fluoride ítrium as inorganic component, and camphorquinone as initiator. Its organic matrix based on siloxane molecules and oxirane [11,12]. The physico-chemical and mechanical properties and biological effects of composites have been evaluated, however, there is only one study evaluating the behavior of organic compounds of Filtek™ Silorane resin and no study of Kalore™ resin [2,13,14].

The objective of this study was to evaluate the in vitro release of organic compounds by Kalore™ and Filtek™ Silorane resins by means of fluorescence spectrometry, varying the source of curing light, the immersion solution of the samples and the pH of the immersion solution.

## Materials and Methods

Were used the Kalore™ (GC FUJI, Kasugai, Japan) and Filtek™ Silorane (3M ESPE, Seefeld, Germany) resins, both in A3 color. For the specimens preparation was used a matrix of cylindrical polyethylene bipartite (8 mm wide and 2 mm thick). The polymerization was performed using halogen as light source (Ultralux Dhabí eletrônico Ribeirão-Preto, São Paulo, Brazil), with a wavelength of 400-500nm, or a light source based on LED (SDI Rádii-cal Bayswater, Australia) with a wavelength of 440-480nm, respecting the manufacturers recommended time.

Were prepared 56 specimens for each resin evaluated. A total of 28 samples of each resin was polymerized by halogen light and 28 with LED light. Then, these samples were stored in identified and numbered plastic bottles according to the resin and curing light source. The oxygen inside the flasks was removed with argon gas to prevent changes in the materials characteristics and were maintained far from light.

The experimental groups division was done after randomization (9.2 SAS - SAS Institute Inc. Cary, USA) (n=7) for each condition examined: composites resin - Kalore™ or Filtek™ Silorane; curing light source - LED or halogen; samples of immersion solution- water or artificial saliva and the pH of the immersion solution - neutral (pH 7.0) or acidic (pH 4.5) (hydrofluoric acid - Aldrich - Steinheim, Germany), designated as follow: Kalore + Halogen light + acid pH + artificial saliva (KHAS), Kalore + Halogen light + neutral pH + water (KHNW), Kalore + Halogen light + pH acid + artificial saliva (KHAS) Kalore + LED + neutral pH + water (KLNW), Kalore + LED + acid pH + Water (KLAW), Kalore + LED + neutral pH + artificial saliva (KLNS), Kalore + LED + acid pH + artificial saliva (KLAS), Filtek Silorane + Halogen light + neutral pH + water (SHNW), Filtek Silorane + Halogen light + acid pH + water (SHAW), Filtek Silorane + Halogen light + neutral pH + artificial saliva (SHNS), Filtek Silorane + Halogen light + pH acid + artificial saliva (SHAS), Filtek Silorane + LED + neutral pH + water (SLNW), Filtek Silorane + LED + acid pH + water (SLAW) Filtek Silorane + LED + neutral pH + artificial saliva (SLNS), Filtek Silorane + LED + pH acid + artificial saliva (SLAS).

The composition of the artificial saliva, was as follows: Phosphate Potassium diacid 0,326g, Phosphate Potassium dibasic 0,803g, Potassium Chloride 0.62g, sodium chloride 0,865g, magnesium chloride 0.125g, calcium chloride 0,072g, sodium fluoride 4,25mg, sorbitol 70% 42,7g, flavoring (minimum amount to produce flavor, scent and pleasant aspects), conservants (nipagin/nipasol) 10mL, 5g thickener and water qsp 1,0L.

To read the aspect of fluorescence emission of the organic

substances in the fluorescence spectrophotometer, encoded 15mL Falcon tubes were filled with 5 mL of deionized water (neutral or acidic) or artificial saliva (neutral or acidic). Subsequently, the samples were placed into tubes with appropriate solutions, and the tubes sealed and stored in stovens at 37°C for 1 hour. After this period for each tube, the reading was made. The spectrophotometer used was the RF-350 / PC (espectophotometer - Shimatzu 120V, 50 / 60Hz, 450VA - PCFI software - fluorescence spectroscopy for RF-531 PC version 2.04) in wavelenght of excitation 220nm and bands of emission from 250 to 425nm. The fissure of excitation for solutions in deionized water was 3nm and the fissure emission was 5nm. For artificial saliva solutions, both the fissures were 5nm. The readings to evaluate the release of organic compounds were performed after 1, 3, 24, 48, 72, 168, 216, 312, 432, 504 and 672 hours (28 days). Then, the determination of the areas was performed under the curves of organic compounds aspects for each sample, in each period of time analised (emission intensityXwavelength). Subsequently, using the Origin program 8.6, a graphic “area x time” was obtained containing the average area and the standard deviation related to 7 samples from each experimental group.

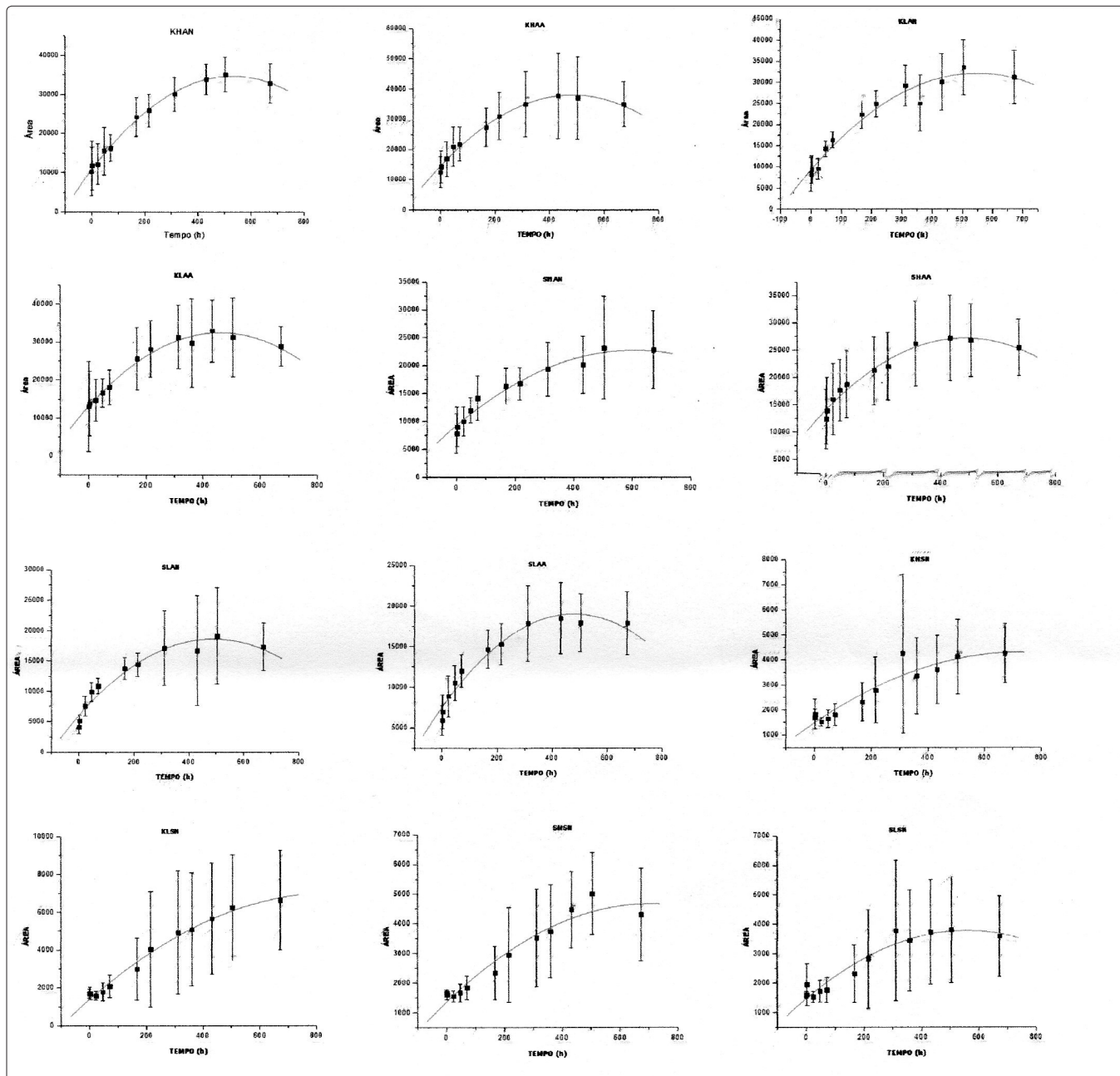
It was performed a polynomial adjustment of these graphs, resulting in a polynomial curve, representing the liberation profile of organic compounds in function of time.

The normality tests Shapiro-Wilk and ANOVA were used to compare the differences between composite resins and polymerizing sources in pairs, applying the Tukey test. Statistical analysis was performed using the program GraphPadPrism® 5.0 at 5% significance level.

## Results

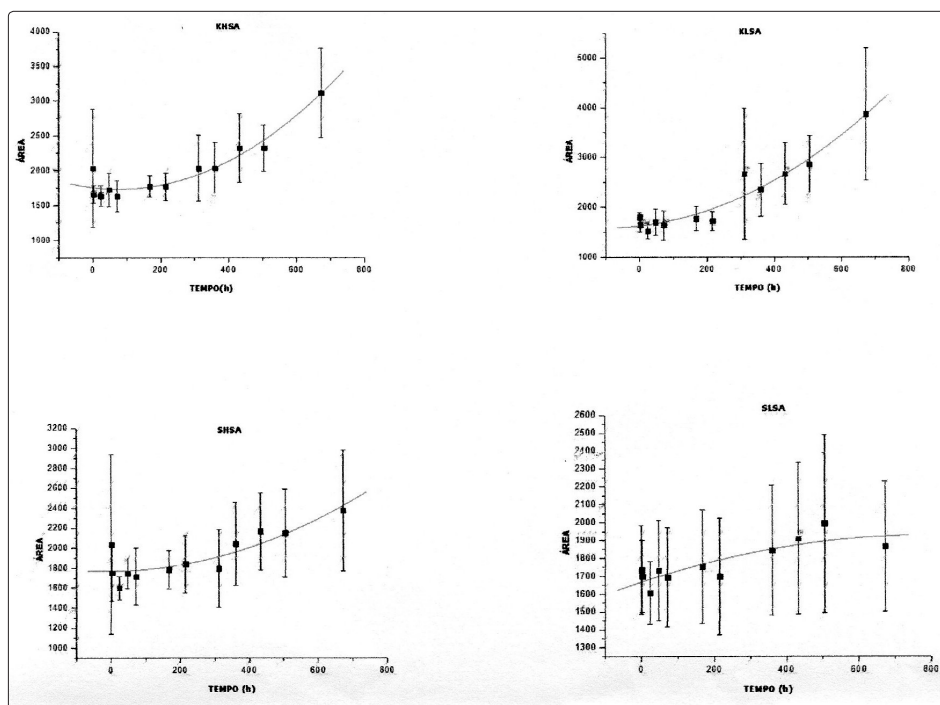
Figures 1 and 2 represent the liberation profile of organic compounds of Kalore™ and Filtek™ Silorane resins, in function of time.

The conditions Kalore/ Halogen/ neutral / water (KHNW); Kalore/ Halogen/ acid/ water (KHAW); Kalore/ LED/ neutral/ water (KLNW); Kalore/ LED/ acid/ water (KLAW); Silorane/ Halogen/ neutral/ water (SHNW); Silorane/ Halogen/ acid/ water (SHAW); Silorane/ LED/ neutral/ water (SLNW); Silorane/ LED/ acid/ water (SLAW) showed a significant initial release of organic compounds, which tends to achieve the solution equilibrium and reach a steady state. In these samples, the curves were better delineated according to the best analytical signal in the aqueous environment, although some salivary conditions with neutral pH Kalore/ Halogen/ neutral/ saliva (KHNS) (Kalore/ LED/ neutral/ saliva (KLNS) Silorane/ Halogen/ neutral/ saliva (SHNS); Silorane/ LED/ neutral/ saliva (SLNS) have also shown the same profile (Figure 1).



**Figure 1:** The liberation profile of organic compounds groups KHNW (Kalore/ Halogen/ neutral/ water): KHAW (Kalore/ /Halogen/ acid/ water), KLNW (Kalore/ LED/ neutral/ water), KLA W (Kalore/ LED/ acid/ water), SHNW (Silorane/ Halogen/ neutral/ water), SHAW (Silorane/ Halogen/ acid/ water) , SLNW (Silorane/ LED/ neutral/ water), SLAW (Silorane/ LED/ acid/ water), KHNS (Kalore/ Halogen/ neutral/ saliva), KLNS (Kalore/ LED/ neutral/ saliva), SHNS (Silorane/ Halogen/ neutral/ saliva) and SLNS (Silorane/ LED/ neutral/ saliva) from left to right, respectively

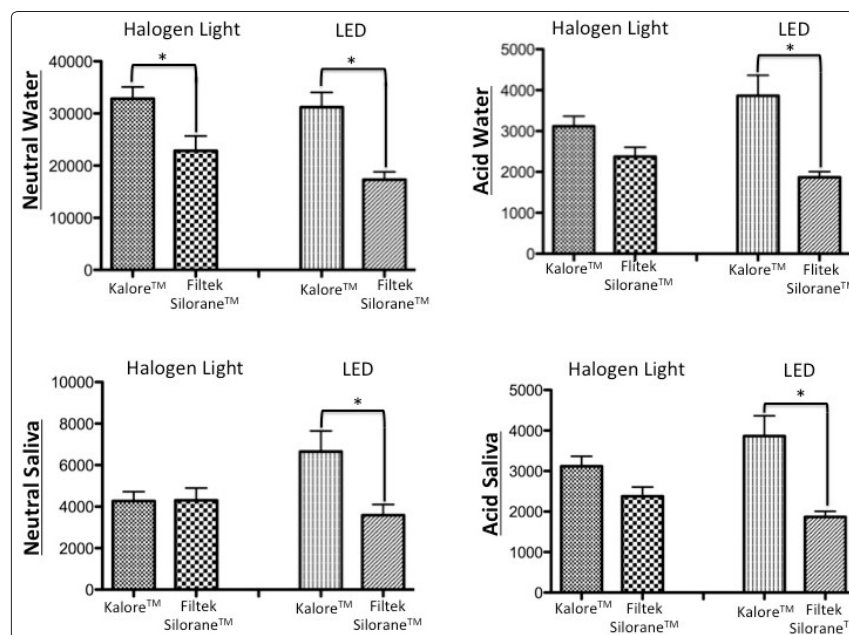
Already Kalore conditions Halogen/ acid/ saliva (KHAS); Kalore/ LED/ acid/ saliva (KLAS); Silorane/ Halogen/ acid/ saliva (SHAS) and Silorane/ LED/ acid/ saliva (SLAS) showed a slower organic compounds release, probably due to the lower solubility of the organic compound in saliva and/or the longer required time for the material imbibition (Figure 2). Samples of saliva and acidic pH showed this trend, with a curve in a less defined format, as the released amount was much lower.



**Figure 2:** Release profile of organic compounds KHNS groups (Kalore/ Halogen/ neutral/ saliva), KLAS (Kalore/ LED/ acid/ saliva), SHAS (Silorane/ Halogen/ acid/ saliva) and SLAS (Silorane/ LED/ acid/ saliva), from left to right, respectively

This liberation was higher in resin groups Kalore™ in all conditions evaluated; however, the release of these compounds was lower when polymerized by halogen light and immersion in saliva at both pHs, and in water when in acid pH. With respect to Filtek™ Silorane resin, the amount of organic compound released was lower in samples polymerized by the LED in both means of immersion, without pH influence.

The Kalore™ resin released more organic compounds than the Silorane™ resin in the aqueous environment, either in the polymerization by the halogen or the LED lamp in both pHs ( $p < 0.05$ ). This release was not statistically significant only when polymerized by halogen light in aqueous and acidic pH ( $p > 0.05$ ) environment. After immersion in artificial saliva, the Kalore™ also presented more released of organic compounds than Filtek™ Silorane resin, but there was only statistically significant difference when polymerized by LED light, both in neutral and at acidic pH (Figure 3).



**Figure 3:** Organic compounds released by Kalore™ and Filtek™ Silorane™ resin, indicating statistically significant differences (\*) when polymerized with halogen light or LED light, after 672 hours of immersion in different solutions and pHs

The curing LED light caused less release of organic compounds ( $p < 0.05$ ) by Filtek™ Silorane, independent of the pH of the

immersion environment. The Kalore™ and Filtek™ Silorane resins released more organic compounds in aqueous solution than in the saliva, in all sources of light and pH ( $p < 0.05$ ).

## Discussion

The light sources usually employed for polymerization of resinous material, are halogen lights and LED [15]. Some studies verifying that the LED light shows greater depth of cure compared to halogen light [16,17]. According to these studies, this may occur due to the LED light devices have a narrow spectral gamma, with a peak of about 470 nm, a wavelength of optimum absorption for the activation of the photoinitiator, using lower power density (intensity  $mW/cm^2 \times$  exposition time) for resinous material polymerization [18,19]. Furthermore, in this wavelength the quantity of light emitted by the LED can be the double compared to halogen light source, with an increase in the degree of conversion of the composite resins polymerized under this light source [20].

Our researches show more liberation of organic compounds by Kalore™ resin when polymerized by LED light, except when immersed in water solution and neutral pH. On the other hand, the resin Filtek™ Silorane polymerized by the LED light released smaller amount of organic compounds in both immersions (water and saliva), independent of the pH.

Considering the immersion solutions used to evaluate the release of organic resinous materials can be used water, saliva, ethanol, methanol and acetronila [21,13]. In this study, we use deionized water due to its high purity, absence of ions and organic contaminants, and the same being suitable for sensitive analytical techniques. Furthermore, the water plays a fundamental role in the oral environment as a solvent for aqueous solutions and fluids ingested by the fact that resinous material when in the oral cavity, continuously interact with this fluid [22,23]. Saliva was used to mimic the conditions of oral environment [24]. Artificial saliva prevents the possibility of obtained contaminated saliva from human donors saliva, by monomers derived from restorations that could be present in donors oral cavity, by oral care products, by products coming from tobacco or remnants from food and beverages [13].

The liberation profile curve of organic compounds in function of time for the two resins when immersed in water, was a clear delineated curve, due to the greater intensity of the analytical signal. The liberation profile curve of organic compounds presented another trend when the immersion environment was acid pH saliva, presenting a less defined format by the lower release of organic compounds.

This is probably explained by the fact that water is a less polar environment than saliva and allows their molecules to accumulate in micro-spaces, at the interface of the resin and its morphological defects without any reaction to polar groups, favoring the release of organic compounds [15].

The Kalore™ and Filtek™ Silorane resins released greater amount of organic compounds in water than in saliva, independent of the source of light or pH ( $p < 0.05$ ), probably due to the polarity of the immersion environment. Furthermore, we observe the liberation of organic compounds even after 672 hours, with both resins and in all conditions evaluated. This fact could be observed through the pattern of organic compounds release curves.

The pH of the oral environment is constantly changing that may affect the amount of organic compounds released, the wear resistance, the sorption and solubility of the resinous material [25]. However, this study did not show significant differences in

the release of compounds in relation to pH.

In 2011, Wei et al. evaluated the kinetic process of diffusion and mass change of Kalore™ resins, Filtek™ Silorane, GC Gradia anterior Direct®, GC Gradia posterior Direct® and Vértice® flow during the sorption cycles and water desorption, and found that the water absorption to Kalore™ resin was low and for Filtek™ Silorane resin was minimal. The authors suggest that, although the solubility values of Kalore™ resins, and Filtek™ Silorane were negative, does not mean that there is no organic compounds liberation by these resins, because if they had used other solvent than water, It might have occurred [23].

In this study we verified that both Kalore™ and Filtek™ Silorane resins, released compounds under all conditions evaluated by means of fluorescence spectrometry. However, in literature there is a single study of organic compounds release by Filtek™ Silorane resin, it Kopperud et al, by means of liquid chromatography and mass spectroscopy was verified the release of organic compounds of Filtek™ Silorane resin when the immersion environment was ethanol 75%; however, no organic compound was released having water as immersion environment. This difference in results may occurs due of the technique used to evaluate the release of organic compounds [2]. Fluorescence spectroscopy technique is extremely sensitive and capable of detecting easily compounds to nanomolar concentration ranges, however, the technique used by Kopperud et al. can simply not detected any released substance considering the small quantity liberated and the method was not enough sensitive [2,26].

## Conclusion

Thus, in this study it is concluded that there was organic compounds liberation of Kalore™ and Filtek™ Silorane resin, and the Filtek™ Silorane resin, when polymerized by LED light had better result than Kalore™ resin, releasing a smaller quantity of organic compounds in all analyzed conditions.

## References

1. Santerre J, Shajii L, Leung BW (2001) Relation of dental composite formulations to their degradation and the release of hydrolyzed polymeric-resin-derived products. *Crit Rev Oral Biol Med* 12:136-151.
2. Kopperud HM, Schmidt M, Kleven IS (2010) Elution of substances from a Silorane-based dental composite. *Eur J Oral Sci* 118:100-102.
3. Poplawski T, Loba K, Pawlowska E, Szczepanska J, Blasiak J (2010) Genotoxicity of urethane dimethacrylate, a tooth restoration component. *Toxicol in Vitro* 24:854-862.
4. Geurtsen W, Lehmann F, Spahl W, Leyhausen G (1998) Cytotoxicity of 35 dental resin composite monomers/additives in permanent 3T3 and three human primary fibroblast cultures. *J Biomed Mater Res* 41:474-480.
5. Schweickl H, Altmannberger I, Hanser N, Hiller KA, Bolay C et al (2005) The effect of triethylene glycol dimethacrylate on the cell cycle of mammalian cells. *Biomaterials* 26:4111-4118.
6. Schweickl H, Spagnuolo G, Schmalz G (2006) Genetic and cellular toxicology of dental resin monomers. *J Dent Res* 85:870-877.
7. Andersson J, Dahlgren U (2011) Effects on mouse immunity of long-term exposure in vivo to minute amounts of HEMA. *Eur J Oral Sci* 119:109-114.
8. Olea N, Pulgar R, Pérez P, Olea-Serrano F, Rivas A et al (1996) Sonnenschein C. Estrogenicity of resin-based composites and sealants used in dentistry. *Environ Health Perspect* 104:298-305.

9. Lewis JB, Rueggeberg FA, Lapp CA, Ergle JW, Schuster GS (1999) Identification and characterization of estrogen-like components in commercial resin-based dental restorative materials. *Clin Oral Investig* 3:107-113.
10. Kalore TM GC FUJI. Technical manual, 2009.
11. Filtek™ Silorane - 3M ESPE. Low Shrink Posterior Restorative System, 2007.
12. Castañeda ER, Silva LA, Gatón-Hernández P, Consolaro A, Rodríguez EG et al (2011) Filtek™ Silorane and Filtek™ Supreme XT resins: tissue reaction after subcutaneous implantation in isogenic mice. *Braz Dent J* 22:105-110.
13. Van Landuyt KL, Nawrot T, Geebelen B, De Munck J, Snauwaert J (2011) How much do resin-based dental materials release? A meta-analytical approach. *Dent Mat* 27:723-747.
14. Naoum SJ, Ellakwa A, Morgan L, White K, Martin FE et al (2012) Polymerization profile analysis of resin composite dental restorative materials in real time. *J Dent* 40:64-70.
15. Yoon TH, Lee YK, Lim BS, Kim CW (2002) Degree of polymerization of resin composites by different light sources. *J Oral Rehabil* 29:1165-73.
16. Aguiar FH, Georgetto MH, Soares GP, Catelan A, Dos Santos PH et al (2011) Effect of different light-curing modes on degree of conversion, staining susceptibility and stain's retention using different beverages in a nanofilled composite resin. *J Esthet Restor Dent* 23:106-114.
17. Moreira Fdo C, Antoniosi Filho NR, Souza JB, Lopes LG (2010) Sorption, solubility and residual monomers of a dental adhesive cured by different light-curing units. *Braz Dent J* 21:432-438.
18. Jeong TS, Kim YR, Kim JH, Kim HI, Kwon YH (2007) Effects of LEDs on microhardness and temperature rise of dental composite resins. *Dent Mater J* 26:838-844.
19. Da Silva EM, Poskus LT, Guimarães JGA (2008) Influence of light polymerization modes and the degree of conversion and mechanical properties of resin composite: A comparative analysis between a hybrid and nanofilled composite. *Oper Dent* 33:287-293.
20. Marson FC, Mattos R, Sensi LG (2008) Avaliação das condições de uso dos fotopolimerizadores. *Dent* 8:48-57.
21. Geurtsen W (1998) Substances released from dental resin composites and glass ionomer cements. *Eur J Oral Sci* 106:687-695.
22. Lopes, HJJ (2003) Garantia e controle da qualidade no laboratório clínico. *Controle da Qualidade no Laboratório Clínico*.
23. Wei YJ, Silikas N, Zhang ZT, Watts DC (2011) Diffusion and concurrent solubility of self-adhering and new resin-matrix composites during water sorption/desorption cycles. *Dent Mater* 27:197-205.
24. Danesh G, Hellak T, Reinhardt KJ, Végh A, Schäfer E et al (2012) Elution characteristics of residual monomers in different light- and auto-curing resins. *Exp Toxicol Pathol* 64:867-872.
25. Ortengren U, Langer S, Göransson A, Lundgren T (2004) Influence of pH and time on organic substance release from a model dental composite: a fluorescence spectrophotometry and gas chromatography/mass spectrometry analysis. *Eur J Oral Sci* 112:530-537.
26. Harris DC. *Análise Química Quantitativa*. Editora LTC, 6ª edição, 2005, cap. 18; 19 e 20.

**Copyright:** ©2020 Marília Pacífico Lucisano. This is an open-access article distributed under the terms of the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original author and source are credited.