

Cigarette smoke: effects on water sorption and solubility of restorative dental composites

Paula Mathias, DDS, MS, PhD ▪ Sara Ramos B. Santos ▪ Thaiane R. Aguiar, DDS, MS, PhD ▪ Poliana Ramos B. Santos, DDS
Andrea N. Cavalcanti, DDS, MS, PhD

Although scientific evidence has shown the effects of tobacco on changes in the color of composite resins, the association between tobacco exposure and the physical properties of composite resins has not been thoroughly investigated. The purpose of this study was to evaluate the effects of cigarette smoke products on water sorption and solubility of microfilled, microhybrid, and nanofilled composite resins (Durafill VS, Filtek Z250, and Filtek Z350 XT, respectively). Ten discs were prepared of each material and divided into 2 groups ($n = 5$), according to cigarette smoke exposure. Specimens were first desiccated until a constant mass was obtained (M_1). Then half of the samples were immersed in deionized water while the other half were exposed daily to tobacco smoke, then washed and stored in deionized water. After 21 days, the resin discs were measured (M_2) and placed in desiccators until constant mass was achieved (M_3). Water sorption and

solubility were calculated and the data was statistically analyzed. Water sorption revealed significant differences among the composite resins. The Filtek Z350 XT exhibited the highest water sorption, followed by Durafill VS and Filtek Z250. Cigarette smoke significantly increased water sorption for all products, but only the solubility of Durafill VS showed a significant difference. Filtek Z250 demonstrated significantly lower solubility than Durafill VS, and Filtek Z350 XT had intermediate values. These results indicated that water sorption and solubility varied among the products, and tobacco smoke may alter the physical properties of resin-based materials.

Received: June 18, 2013

Accepted: September 25, 2013

Key words: cigarette smoke, solubility, water sorption, composite resin

Esthetics, color stability, and good physicochemical performance are essential characteristics for a dental restorative composite.^{1,2} The clinical behavior of a composite resin is multifactorial and has been associated with intrinsic characteristics such as organic composition, type of fillers, coupling agent used, conversion degree, surface roughness, water sorption, and solubility parameters.³⁻⁶ In addition, positive correlations between composite resin and extrinsic stains (such as coffee, tea, orange juice, carbonated drinks, and cigarette smoke) have recently been reported in the dental literature.^{1,2,7-9}

According to Mackay, the use of tobacco is epidemic and expanding, and even though the harmful effects of tobacco have been well-documented, new risks continue to appear.¹⁰ In restorative dentistry, a new study by Huang et al suggested that smoking can increase the development of caries, and Takeuchi et al reported that byproducts of cigarette smoke were observed in composite resin and dental structures.^{11,12} Moreover, Almeida e Silva et al found that contamination by cigarette smoke significantly decreased the bond strength between dentin and composite resin following exposure to 30 cigarettes a day for 17 days.¹³ However, no association between the physical properties of composite resins

and cigarette smoke—which may provide important implications for the clinical longevity of restorative materials—has been investigated until now.

The water sorption and solubility properties of resin-based materials have an appreciable effect on the durability of dental restoratives.² In the oral environment, these materials may uptake water and suffer chemical degradation from components present in saliva, beverages, and foods.¹⁴ These processes may promote a plasticizing effect on restorative structures by releasing intrinsic

components such as residual monomers and filler particles, negatively affecting the functional lifetime of restorations.¹⁵⁻¹⁷

Water sorption and solubility studies have been carried out after immersion in various solutions such as distilled/deionized water, ethanol, and acidic drinks.^{2,16-19} However, studies evaluating the effect of tobacco smoke on the water sorption/solubility properties of restorative composite resin have not been reported. The objective of this study was to determine the water sorption and solubility characteristics of various types of restorative dental composite

Table 1. Specifications of the composite resins.

Composite resin (manufacturer)	Resin type	Shade color	Filler content (wt%)	Filler type/size	Organic matrix (lot number)
Filtek Z350 XT (3M ESPE)	Nanofilled	A3B	78.5	Zirconia/silica clusters 0.6-1.4 μm ; silica 5-20nm	Bis-GMA, UDMA, Bis-EMA, TEGDMA, (202748, 182351, 182963)
Filtek Z250 (3M ESPE)	Microhybrid	A3	84.5	Zirconia/silica 0.01-3.5 μm	Bis-GMA, UDMA, Bis-EMA, TEGDMA (75553BR, 41393BR)
Durafill VS (Heraeus Kulzer)	Microfilled	A3	50.5	Prepolymerized silica 10-20 μm ; silica 0.02-2 μm	UDMA (010214, 010213)

Abbreviations: Bis-EMA, bisphenol A polyethylene glycol diether dimethacrylate; Bis-GMA, bisphenol A diglycidyl ether dimethacrylate; TEGDMA, triethylene glycol dimethacrylate, UDMA, diurethane dimethacrylate.

Table 2. Mean (\pm SD) of water sorption ($\mu\text{g}/\text{mm}^3$) of the composite resins after immersion in either deionized water or cigarette smoke during 21 days.

Composite resin	Water sorption		Tukey
	Water (control group)	Cigarette smoke	
Filtek Z350 XT	17.30 (0.14)	18.24 (0.44)	A
Filtek Z250	11.42 (0.80)	13.06 (0.52)	C
Durafill VS	14.24 (0.45)	16.01 (0.56)	B
Tukey	b	a	

Means followed by distinct letters represent statistically significant differences (2-way ANOVA and Tukey, $\alpha = 5\%$). Lower-case letters compare treatments.

Table 3. Median levels of solubility ($\mu\text{g}/\text{mm}^3$) of composite resins after immersion in deionized water or exposure to cigarette smoke during 21 days.

Composite resin	Solubility	
	Water (control)	Cigarette smoke
Filtek Z350 XT	-2.45 ^{ABa}	-2.29 ^{ABa}
Filtek Z250	-3.21 ^{Ba}	-4.22 ^{Ba}
Durafill VS	6.21 ^{Aa}	4.86 ^{Ab}

Means followed by distinct letters represent statistically significant differences (Kruskal-Wallis and Mann-Whitney U test, $P < 0.05$). Upper-case letters compare composite resin, lower-case letters compare treatments.

resins, and to evaluate the effect of cigarette smoke on these characteristics. The null hypotheses proposed were that among the composite resins tested there would be no differences in water sorption and solubility, and that cigarette smoke would have no significant effect on the water sorption/solubility behavior of the resins.

Materials and methods

Experimental design

The water sorption and solubility tests were performed according to ISO 4049-2000 standard specification, except for cigarette smoke treatment and storage time. Three different dental composites—Filtek Z350 XT (3M ESPE), Filtek Z250 (3M ESPE) and Durafill VS (Heraeus Kulzer)—were used in this study. These composites were selected in accordance to the classification of dental composite resin based on filler particle size. The composition, classification, shade color, filler characteristics, and lot numbers of the composite resins are listed in Table 1.

Samples preparation

Ten disc-shaped specimens of each composite resin were prepared (15 ± 0.1 mm diameter \times 1 ± 0.1 mm depth), and a total of 30 discs were obtained ($n = 5$). The experimental groups were assigned according to the composite resin and cigarette smoke treatments. The composite resin was inserted in the metal mold in a single increment and covered with a mylar strip and glass slide. Special care was taken to allow for extrusion of excess material and the prevention of air bubbles.

All specimens were irradiated with a light-emitting diode curing unit (Radium-cal, SDI [North America], Inc.) at an intensity of $1200 \text{ mW}/\text{cm}^2$. The light curing was performed for 15 seconds on the middle of the sample through the mylar strip and addition polymerization was applied at 4 equally peripheral points on the circumference for 10 seconds each, resulting in a total curing time of 55 seconds. The output irradiance was measured with a radiometer unit.

Water sorption/solubility and smoke exposure

The polymerized discs were stored in a desiccator filled with anhydrous calcium sulphate (CaSO_4) at 37°C for 24 hours. Their masses were measured using an electronic balance with 1.0 mg precision (AUD 220D, Shimadzu Corp). This cycle was repeated until a constant mass was obtained (M_1 , when the mass loss of each specimen was ≤ 0.1 mg within a period of 24 h). To calculate the volume (V) of each specimen (mm^3), the diameter and thickness was individually measured using a digital electronic caliper (Mitutoyo American Corporation), rounded to the nearest 0.01 mm.

After M_1 was achieved, each composite resin group of 10 discs was divided into 2 subgroups. One set of specimens were placed in deionized water (control group) and the other set was submitted to cigarette smoke treatment (10 cigarettes for an 8 minute period/twice a day/21 days) in a smoking machine designed according to previous studies.⁷⁻⁹ Lit cigarettes (Hollywood Original Blend, Souza Cruz) which contained an elevated amount of tar

(10 mg), were placed in the first chamber of the smoking machine, receiving external ventilation from an air pump. The apparatus was hermetically sealed but had a special orifice to release the smoke/air stream from the first chamber through a tar filter barrier to a second chamber that contained the dry resin composite specimens. Using this mechanism, composite resin specimens were in contact with pigments and components of the cigarette smoke during the experiment. After each cycle of smoke exposure, specimens were washed and stored in deionized water. The deionized water was renewed daily for all groups.

After 21 days, specimens were gently wiped free of excess water with absorbent paper and weighed (M_2). The specimens were dried in the same manner as previously described until a constant mass was achieved (M_3). The values for water sorption (WS) and solubility (SL) were calculated as:

$$\text{WS} = (M_2 - M_3) / V$$

$$\text{SL} = (M_1 - M_3) / V$$

M_1 is the initial dry constant mass (μg) prior to immersion in water, M_2 is the mass of the specimen (μg) after immersion in deionized water or submitted to cigarette smoke simulation, M_3 is the mass of the reconditioned specimen (μg), and V is the specimen volume in mm^3 .

Statistical analysis

Water sorption and solubility release were expressed in $\mu\text{g}/\text{mm}^3$. The data of the water sorption behavior were compared by 2-way ANOVA and Tukey tests. The

nonparametric solubility values were analyzed by Kruskal-Wallis and Mann-Whitney U tests. The analysis of variance was determined via the SAS 9.1 statistical package (SAS Institute). The nonparametric analysis was performed via Bioestat 5.0 software (The Mamiraua Institute), both with a 95% confidence level.

Results

The mean values and standard deviations of water sorption and median levels of solubility behavior are summarized in Tables 2 and 3, respectively. According to the statistical analysis of water sorption, significant statistical differences were observed among composite resins, regardless of the experimental condition ($P < 0.001$). However, no significant interactions between the main factors (composite resin and experimental condition) were observed ($P = 0.22$). The composite resin Filtek Z350 XT showed the highest water sorption values, followed by Durafill VS and Filtek Z250. In addition, cigarette smoke significantly increased the water sorption independent of the composite resin ($P < 0.001$).

For solubility analysis, there were statistically significant differences among composite resins in the control groups and in the experimental groups exposed to cigarette smoke ($P = 0.005$ and $P = 0.002$, respectively). The Durafill VS microfilled composite showed higher solubility values than Filtek Z250 in both conditions, whereas the resin, Filtek Z350 XT, exhibited intermediate values. When comparing the effect of treatments, only the solubility value of Durafill VS composite showed a significant difference after exposure to cigarette smoke ($P < 0.05$).

Discussion

Differences in water sorption and solubility among restorative dental composites may be due to the fillers or the organic matrix content of each material. The molecular mobility during the polymer formation and the presence of hydrophilic groups may determine the properties of resin-based materials.^{6,20,21} The hydrophilic character of these monomers tends to facilitate the water uptake into polymer matrix and the difference in their water sorption was related. Triethylene glycol dimethacrylate (TEGDMA) was considered the most hydrophilic monomer ($69.51 \mu\text{g}/\text{mm}^3$),

followed by bisphenol A glycidyl methacrylate (Bis-GMA, $33.49 \mu\text{g}/\text{mm}^3$), urethane dimethacrylate (UDMA, $29.46 \mu\text{g}/\text{mm}^3$), and ethoxylated bisphenol-A-dimethacrylate (Bis-EMA, $20.10 \mu\text{g}/\text{mm}^3$).⁶ Moreover, different monomers can be combined and may result in better mechanical performance.²¹

In both experimental conditions analyzed in this study, the microfill composite (Durafill VS) showed an intermediate mean value of water sorption. This could be attributed to the UDMA monomer, which may hinder the water uptake in the polymers chains.² In contrast, Filtek Z350 XT and Filtek Z250 exhibited significant differences in water sorption patterns despite sharing similar monomers. Thus, the differences in water sorption may also be affected by concentration, type, size, and distribution of filler particle in the polymer matrix and silane content.^{2-4,22}

Filtek Z350 XT contains nanometric filler particles arranged in clusters (known as *nanoclusters*).^{3,23} According to Rastelli et al, the nanoclusters were similar in size to the individual filler particles contained in the microhybrid composite Filtek Z250.⁴ However, these clusters of nanofillers increase the area of surface exposed to water, especially in the interface between the filler particles and the organic matrix, which supplies a route for diffusion of water molecules into the polymer.^{2,20}

Lower filler content may increase the available free volume for water uptake and provide a higher water sorption in reduced time, whereas large filler loads exhibit smaller external contact area to volume ratios, restricting the amount of water at the filler-matrix interface.² Thus, it is speculated that the lower filler percentage (50.5 wt%) of Durafill VS and the smaller filler size (0.6-1.4 μm) of Filtek Z350 XT may affect water sorption behavior. Additionally, water sorption values obtained in this study for Filtek Z350 XT and Durafill VS when stored in water (17.30 and $14.24 \mu\text{g}/\text{mm}^3$, respectively), were in agreement with findings reported by Rahim et al (16.13 and $14.24 \mu\text{g}/\text{mm}^3$, respectively).²

Unpolymerized monomers and filler particles can leach out of polymers when immersed in water, saliva, and acidic beverages, promoting polymer degradation and reducing the lifetime of dental restorations.¹⁴⁻¹⁷ The resistance to dissolution has

been considered an important property of resin-based materials, and studies have revealed that solubility behavior is significantly influenced by the degree of monomer conversion.²⁴ Thus, higher levels of monomer conversion have been associated with lower mean solubility values.^{6,24} In the present study, Durafill VS demonstrated the highest solubility behavior. This composite contains UDMA monomers of high molecular weight, along with 2 amino groups (-NH), which form intermolecular hydrogen bonds, thus increasing the monomer viscosity.²⁴ Although the elevated viscosity of monomers may reduce molecular mobility, high UDMA monomer conversions (69.6% and 63.52%) were obtained in other studies.^{4,6}

In contrast, Filtek Z250 showed an approximate 70% to 71% degree of conversion.²⁵ Pfeifer et al reported good results when different monomers were mixed, especially composites formulated with Bis-GMA/TEGDMA/UDMA.²¹ These combinations are present in Filtek Z350 XT and Filtek Z250 and may contribute to the lower solubility of these compounds compared to Durafill VS. In addition, the negative solubility values observed for the nanofilled and microhybrid composites could be due to the fact that their water uptake was higher than their solubility.^{17,19} Although the ISO 4049-2000 standard specification for water sorption and solubility tests was followed, the authors believe that longer storage of specimens in the desiccator may result in positive values for all composites tested. Thus, the first hypothesis tested in which there were no significant differences in water sorption and solubility among dental composites was rejected.

In an attempt to evaluate the effect of cigarette smoke during water sorption and solubility, the composite resins were exposed to cigarette smoke during the stored phases (10 cigarettes for an 8 minute period/twice a day/21 days). Similar regimens were used in previous studies that evaluated the effect of cigarette exposure on the color of composite resins.⁷⁻⁹ In these studies, the cigarette smoke promoted an irreversible change in color of the composite resins, despite repolishing attempts.⁷ In addition, chemical elements from cigarette smoke, such as cadmium, arsenic, and lead have been observed in the composite resins after microanalytic study.¹²

In this study, water sorption was significantly increased for all the composites after cigarette smoke exposure. The solubilities of Filtek Z350 XT and Filtek Z250 did not show significant differences between experimental conditions. However, Durafill VS revealed a lower solubility when exposed to cigarette smoke (-22%). Therefore, the second null hypothesis was not accepted.

The authors of this study speculate that cigarette smoke products can permeate into the composite resin and promote physicochemical alterations in their structure. After cigarette smoking simulation, tar particles were visible on the composite resin. Durafill VS showed higher staining than microhybrid and nanocomposite composites, and exhibited a significant solubility decrease when compared to the control group. It is theorized that tar particles impregnated in the resin matrix can affect the release of intrinsic resin components, such as residual monomers and filler particles, thus reducing the solubility, especially for microfilled composites. To the authors' knowledge, this is the first study that analyzed the water sorption/solubility of composite resins after cigarette smoke exposure. Further studies are needed to investigate the interactions between cigarette smoke compounds and resin-based materials, as well as the effects on the physicomechanical properties of the composites.

Conclusion

Water sorption and solubility behavior are material-dependent. In the experimental conditions analyzed in this study, cigarette smoke affected the water sorption and solubility of the dental composites, which can lead to greater polymer degradation and possibly reduce the durability of restorative procedures.

Author information

Dr. Mathias is an associate professor, Department of Clinical Dentistry, School of Dentistry, Federal University of Bahia, Salvador, Brazil, where SRB Santos and Dr. PRB Santos are graduate students, and Dr. Cavalcanti is an assistant professor. Dr. Aguiar is a postdoctoral research

candidate, Department of Restorative Dentistry, College of Dentistry, University of Illinois at Chicago.

Acknowledgments

This investigation was supported by the FAPESB (APP022/2011) and PIBIC-UFBA programs, Brazil.

References

1. Nasim I, Neelakantan P, Sujeer R, Subbarao CV. Color stability of microfilled, microhybrid and nanocomposite resins—an in vitro study. *J Dent.* 2010;38(Suppl 2):e137-e142.
2. Rahim TN, Mohamad D, Md Akil H, Ab Rahman I. Water sorption characteristics of restorative dental composites immersed in acidic drinks. *Dent Mater.* 2012;28(6):e63-e70.
3. Curtis AR, Palin WM, Fleming GJ, Shortall AC, Marquis PM. The mechanical properties of nanofilled resin-based composites: characterizing discrete filler particles and agglomerates using a micromanipulation technique. *Dent Mater.* 2009;25(2):180-187.
4. Rastelli AN, Jacomassi DP, Faloni AP, et al. The filler content of the dental composite resins and their influence on different properties. *Microsc Res Tech.* 2012;75(6):758-765.
5. da Silva EM, Poskus LT, Guimaraes JG. Influence of light-polymerization modes on the degree of conversion and mechanical properties of resin composites: a comparative analysis between a hybrid and a nano-filled composite. *Oper Dent.* 2008;33(3):287-293.
6. Sideridou I, Tserki V, Papanastasiou G. Study of water sorption, solubility and modulus of elasticity of light-cured dimethacrylate-based dental resins. *Biomaterials.* 2003;24(4):655-665.
7. Mathias P, Silva LD, Saraiva Lde O, et al. Effect of surface sealant and repolishing procedures on the color of composite resin exposed to cigarette smoke. *Gen Dent.* 2010;58(4):331-335.
8. Mathias P, Costa L, Saraiva LO, Rossi TA, Cavalcanti AN, da Rocha Nogueira-Filho G. Morphologic texture characterization allied to cigarette smoke increase pigmentation in composite resin restorations. *J Esthet Restor Dent.* 2010;22(4):252-259.
9. Mathias P, Rossi TA, Cavalcanti AN, Lima MJ, Fontes CM, Nogueira-Filho G da R. Cigarette smoke combined with staining beverages decreases luminosity and increases pigmentation in composite resin restorations. *Compend Contin Educ Dent.* 2011;32(2):66-70.
10. Mackay J. Implementing tobacco control policies. *Br Med Bull.* 2012;102:5-16.
11. Huang R, Li M, Gregory RL. Effect of nicotine on growth and metabolism of *Streptococcus mutans*. *Eur J Oral Sci.* 2012;120(4):319-325.
12. Takeuchi CY, Correa-Afonso AM, Pedrazzi H, Dinelli W, Palma-Dibb RG. Deposition of lead and cadmium released by cigarette smoke in dental structures and resin composite. *Microsc Res Tech.* 2011;74(3):287-291.
13. Almeida e Silva JS, de Araujo EM Jr, Araujo E. Cigarette smoke affects bonding to dentin. *Gen Dent.* 2010;58(4):326-330.

14. Dos Santos PA, Garcia PP, De Oliveira AL, Chinelatti MA, Palma-Dibb RG. Chemical and morphological features of dental composite resin: influence of light curing units and immersion media. *Microsc Res Tech.* 2010;73(3):176-181.
15. Soderholm KJ. Degradation of glass filler in experimental composites. *J Dent Res.* 1981;60(11):1867-1875.
16. Ito S, Hoshino T, Iijima M, Tsukamoto N, Pashley DH, Saito T. Water sorption/solubility of self-etching dentin bonding agents. *Dent Mater.* 2010;26(7):617-626.
17. Malacarne J, Carvalho RM, de Goes MF, et al. Water sorption/solubility of dental adhesive resins. *Dent Mater.* 2006;22(10):973-980.
18. Bail M, Malacarne-Zanon J, Silva SM, et al. Effect of air-drying on the solvent evaporation, degree of conversion and water sorption/solubility of dental adhesive models. *J Mater Sci Mater Med.* 2012;23(3):629-638.
19. Lopes LG, Jardim Filho Ada V, de Souza JB, Rabelo D, Franco EB, de Freitas GC. Influence of pulse-delay curing on sorption and solubility of a composite resin. *J Appl Oral Sci.* 2009;17(1):27-31.
20. Ferracane JL. Hygroscopic and hydrolytic effects in dental polymer networks. *Dent Mater.* 2006;22(3):211-222.
21. Pfeifer CS, Silva LR, Kawano Y, Braga RR. Bis-GMA copolymerizations: influence on conversion, flexural properties, fracture toughness and susceptibility to ethanol degradation of experimental composites. *Dent Mater.* 2009;25(9):1136-1141.
22. Polydorou O, Konig A, Hellwig E, Kummerer K. Long-term release of monomers from modern dental-composite materials. *Eur J Oral Sci.* 2009;117(1):68-75.
23. Chen MH. Update on dental nanocomposites. *J Dent Res.* 2010;89(6):549-560.
24. Goncalves L, Filho JD, Guimaraes JG, Poskus LT, Silva EM. Solubility, salivary sorption and degree of conversion of dimethacrylate-based polymeric matrixes. *J Biomed Mater Res B Appl Biomater.* 2008;85(2):320-325.
25. Porto IC, Soares LE, Martin AA, Cavalli V, Liporoni PC. Influence of the photoinitiator system and light photo-activation units on the degree of conversion of dental composites. *Braz Oral Res.* 2010;24(4):475-481.

Manufacturers

Heraeus Kulzer, South Bend, IN
800.435.1785, www.heraeus-dental-us.com
Mitutoyo American Corporation, Aurora, IL
888.648.8869, www.mitutoyo.com
SAS Institute, Cary, NC
800.727.0025, www.sas.com
SDI (North America), Inc., Bensenville, IL
800.228.5166, www.sdi.com.au
Shimadzu Corporation, Kyoto, Japan
81.75.823.1111, www.shimadzu.com
The Mamiraua Institute, Manaus, Brazil
55.97.3343.9700, www.mamiraua.org.br
3M ESPE, St. Paul, MN
888.364.3577, solutions.3m.com