

The Study of Water Sorption and Solubility of Differently formulated resin composite restorative materials. A Comparative Study in vitro

*Aya Alaa Elden**, *Zainab M. Soliman*** and *Omaima H. Ghallab****

Abstract:

Objective: In vitro study to evaluate the water sorption and solubility behavior of four types of resin composite materials of different filler loading.

Materials and methods: Four types of dental resin composites were used: Grandio, Grandio Flow, Xtra- fill and Xtra Base composites. Eighty discs shaped specimens were prepared of 15 mm diameter and 1 mm thickness. The discs were placed in a desiccator and weighed followed by placement in distilled water and weighed to obtain water sorption weight and finally placed again in desiccator to obtain the final weight, Water sorption and solubility values were calculated by mathematical equations from the different weights. Data was collected and statistically analyzed to compare different materials behavior.

Results: No significant differences in water sorption and solubility testes were observed.

Conclusion: Water sorption and solubility are two parameters that are affected by the type and chemical composition of the material.

Keywords: composites, water sorption, solubility.

* Master's Degree student of Operative Dentistry, Faculty of Dentistry, Ain Shams University.

** Lecturer of Operative Dentistry, Faculty of Dentistry, Ain Shams University.

*** Professor of operative dentistry department Faculty of Dentistry, Ain Shams University

Introduction:

Composite restorations are extensively used types of restoration ¹. Though they have many advantages, polymerization shrinkage is considered a major disadvantage which might lead to microleakage, debonding, secondary caries and postoperative sensitivity ². Bulk fill composite material had been developed as low shrinkage stress material ³. It can be placed in thicker increment up to 4mm. Two types of bulk fill composite have been introduced in the market flowable type and higher viscosity type bulk fill composite ⁴.

Water sorption and solubility of resin based materials are important characteristic of composite resin, since they influence the mechanical properties of the dental materials and longevity of composite restorations ⁵. In a wet oral environment, composites absorb water and release unreacted monomers and inorganic ions. The water uptake had harmful effects on the mechanical/physical properties of the material due to hydrolytic breakdown of the bond between the silane-filler particles, filler-matrix debonding or hydrolytic degradation of the filler. In the same time, residual monomers and other components eluted from composites into the oral environment, weaken their mechanical properties ⁶. On the other hand, the absorbed water may cause a hygroscopic expansion of the composite that could compensate the polymerization shrinkage and improve the restoration seal ⁷. Moreover, water sorption and solubility affect the strength, abrasion resistance, volume, and color stability of resin composites.

In order to maintain the composite restoration material in the oral cavity for a long duration, it is mandatory to study their water sorption and solubility properties to be able to predict their behavior in the oral cavity.

Materials and methods: Four types of dental composites were used: Grandio as Conventional restorative composite, Grandio Flow as Conventional flowable composite, Xtra-Fill as Bulk fill restorative composite and Xtra Base as Bulk fill flowable composite.

Specimen preparation:

A total of eighty discs shaped specimens was prepared. Specimens were equally divided into four groups according to type of dental resin composite (20 specimen each). Disc shaped resin composite specimens were prepared using a split circular copper mold of 15 mm diameter and 1 mm thickness according to dimensions specified by (ISO FDIS 4049:2009). The mold was placed over a glass slab covered with celluloid sheet. The composite material was packed against the celluloid sheet then another celluloid sheet was placed on the surface of composite followed by microscopic glass slide as shown in figure (1). To obtain a flat surface free of voids and extrude excess material, a weight of 1 kg was placed over the glass slide for 5 seconds as shown in figure (2).

After weight removal, each specimen was light cured using LED light curing unit (Guilin Woodpecker Instrument Co.,Ltd. Guangxi, 541004 P.R. China) as shown in figure (3), the intensity of light cure was periodically checked using a radiometer (Model 100 curing radiometer, Kerr, USA). Curing was done in five overlapping sections to cover all specimen diameter. Specimen curing started at the center by placing light cure tip in contact with glass slide and curing through the slide. This was followed by four overlapping curing each of 20 seconds according to the manufacturer's instructions as shown in figure (4). Glass slide and celluloid sheet were removed. The mold was disassembled and specimen was removed. Excess material flashes were removed carefully using Bard Parker blade number 11.

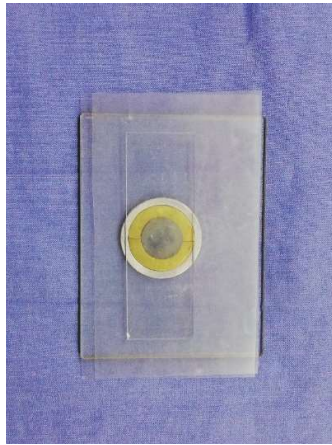


Figure (1)



Figure (2)

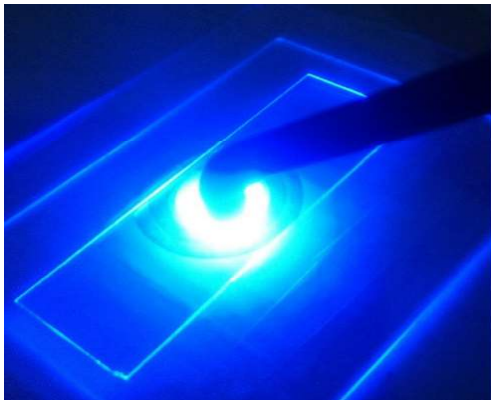


Figure (3)

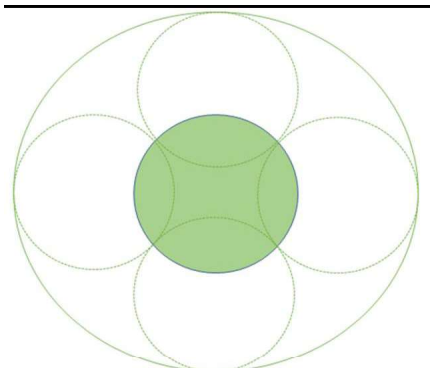


Figure (4)

Testing procedure:

The thickness and diameter of each specimen were measured before testing using a digital micrometer caliper (precision measuring, china), with an accuracy of 0.01mm. Three measurements were taken and the mean of each diameter and thickness was recorded. The average volume (V) of samples was calculated in cubic millimeters (mm³).

The sorption and solubility tests were performed in compliance with the ISO 4049:2009 standards. The specimens were inserted into a desiccator containing anhydrous self-indicating silica. The temperature of the incubator (incubator 20 litre, coated, analogue, 50 HZ. 2015) was adjusted first at $37 \pm 1^\circ\text{C}$ for certain time then the desiccator was transferred to the incubator and maintained at this temperature for 22 h. The samples in desiccator were left at room temperature for 2 additional hours before any measures were done. Specimens were then weighed to an accuracy of 0.001 g, using a high precision electronic balance with three digits (model BS150, ST instruments, Taiwan). This cycle was repeated until the mass loss of each specimen was lower than 0.1 mg in two successful days. For each sample, three measurements were taken & mean of weight was recorded as M1 which represents the initial mass of the specimen and was expressed in micrograms (μg).

Each specimen was immersed in 10 ml distilled water inside a glass jar to insure complete immersion of specimens in water. The glass jar was completely sealed and kept in the incubator for 1 week at $37 \pm 1^\circ\text{C}$. After completing the storage period, the specimens were removed, gently dried with absorbent paper for 15 seconds. After 1 minute from removal from water three measurements were taken and the mean of weight of specimens was taken to obtain the mass M2.

The specimens were reinserted in the desiccator and placed again in the incubator for another cycle until they reached a constant weight in two successful days. Three measurements were taken and the mean of weight was recorded as M3.

The values for water sorption (Wsp) and solubility (Wsl), expressed in micrograms per cubic millimeter, were calculated using the following formulae:

$$Wsp = (M2 - M3) / V$$

$$Wsl = (M1 - M3) / V$$

Statistical analysis:

Numerical data were explored for normality by checking the distribution of data and using tests of normality (Kolmogorov-Smirnov and Shapiro-Wilk tests). All data showed non-normal (parametric) distribution. Data were presented as mean, median, standard deviation (SD), minimum, maximum and 95% Confidence Interval (95% CI) for the mean values.

Kruskal-Wallis test was used to compare between the four groups. Dunn's test was used for pair-wise comparisons when Kruskal-Wallis test is significant.

Results:

For water sorption results: Results showed that: Grandio Flow showed the highest median and (range) of water sorption followed by Xtra Base followed by then Xtra Fill. The lowest median water sorption was found with Grandio. However, there was no statistically significant difference between median water sorption values in the four groups (P -value = 0.327).

Table (1): Descriptive statistics and results of Kruskal-Wallis test for comparison between water sorption values ($\mu\text{g}/\text{mm}^3$) in the four groups

Group	Mean	SD	Median	Minimum	Maximum	95% CI		P-value
						Lower bound	Upper bound	
Grandio	32.2	10.8	29	18.5	55.5	25.9	38.4	0.327
Xtra Fill	28.2	7.2	29.5	12.4	36.5	24.2	32.2	
Grandio Flow	37.1	15.9	34.8	16.3	71.1	28.3	45.9	
Xtra Base	40.8	20.9	32.4	12.7	78.2	29.7	51.9	

*: Significant at $P \leq 0.05$

For solubility results: Grandio Flow showed the highest median and (range) of solubility followed by Xtra Base then Grandio. The lowest median solubility was found with Xtra Fill. However, there was no statistically significant difference between median solubility values in the four groups (P -value = 0.091).

Table (2): Descriptive statistics and results of Kruskal-Wallis test for comparison between solubility values ($\mu\text{g}/\text{mm}^3$) in the four groups

Group	Mean	SD	Median	Minimum	Maximum	95% CI		P-value
						Lower bound	Upper bound	
Grandio	15.3	9.9	12.9	3.9	32	9.6	21	0.091
Xtra Fill	15.4	9.1	12.1	4.1	30.7	10.3	20.4	
Grandio Flow	23.5	9.7	26.4	5.6	44.7	18.1	28.9	
Xtra Base	22.1	14.3	19.7	4.2	52.9	14.5	29.7	

*: Significant at $P \leq 0.05$

Discussion:

As dental composites are used in humid oral environment, these materials are exposed to saliva and water continuously. They are subjected to water sorption and desorption cycles during function that had a negative effect on resin composite stability and mechanical properties.

In this study, four types of composite differ from each other in filler loading and types of monomer in the resin matrix were compared. In this test the data had non-parametric distribution, so we use the median values as our references and to represent the results and statistical data.

No statistically significant difference between median water sorption values in the four groups used in this study. The differences in water sorption values might be attributed to variability in polymers regarding microstructural and molecular aspects. Water sorption is influenced by the polarity of the molecular structure, the presence of hydroxyl groups capable of forming hydrogen bonds with water, and the degree of cross linking in the continuous matrix⁸. The uptake of water may result in an expansion of the gap between polymer chains, depending on the degree of cross-link density in its structure. Water sorption values had a negative correlation with the amount of filler content which is in line with previous studies⁹. When the weight percentage of filler decrease, polymer matrix increased so water sorption increase. Another important factor is the chemistry of the monomers. Water uptake is greater for composite resins containing higher concentrations of hydrophilic TEGMDA monomer due to its hydrophilic ether linkages.¹⁰ These two factors could explain the high water sorption results of Grandio Flow as the material has low filler loading and high resin matrix content, in addition to its higher content of TEGDMA.

Xtra Base had higher water sorption results than Xtra Fill and Grandio composites. This could be explained by the amount of filler loading (75%) and resin matrix of the material which is lower than that of Grandio (87%) and Xtra-Fill (86%). On the other hand, Alshali R., et. Al.⁹, Satterthwaite, et. Al.¹¹ and Porto, et. Al.¹² found that Xtra base had low water sorption values due to its Bis-EMA hydrophobic monomer.

Xtra Fill had polymerization modulator chemical groups or plasticizers in their resin matrix that reduce polymerization shrinkage stress¹³. This chemical alteration may affect the quality of polymer network of bulk fill material and its resistance to moisture compared to conventional composite¹⁴. And this could explain the low values of Xtra Fill composite material.

The solubility behavior of dental materials is affected by the type and surface area of the filler used, the particular silane treatment, the difference in the monomer structure, and the degree of cross linking¹⁵. The molecular size of the monomer is another important factor in the dissociation of residual monomers, as the smaller molecules will decompose faster. Lower weight monomers

can be decomposed in greater amounts than higher weight monomers¹⁶.

TEGDMA is a low molecular weight monomer, which shows high mobility and decomposes more quickly than larger molecules, such as Bis-GMA¹⁷. This might explain the highest solubility values of Grandio flow that has high content of TEGDMA monomer.

On the other hand, the high solubility values of Xtra Base could be attributed to the lower filler loading (75%) of the material compared to other tested material. This is against Satterthwaite study¹¹ that resulted low values of Xtra base due to its Bis-EMA hydrophobic monomer. The high filler loading of Grandio composite might explain its low solubility value compared to other composites. Also, the presence of less hydrophilic Bis-GMA and UDMA monomer may have added value in the low solubility results of the material^{6,18,19}.

Xtra Fill composite showed significantly the least solubility values. These results may be attributed to higher amount of Bis-GMA, in addition to the presence of micro-sized filler particle sizes (2-4 μm)²⁰. This filler particle size had lower filler-matrix interface surface area which allowed better light transmission and less light scattering through material resulting in higher degree of conversion. Bulk fill composite may contain polymerization modulator chemical groups or plasticizers in their resin matrix to reduce polymerization shrinkage stress. This chemical alteration may affect the quality of polymer network of bulk fill material and its resistance to moisture compared to conventional composite¹³.

Conclusions:

Under the limitations of this study, the following conclusions could be suggested:

The resin matrix and filler loading of composite material might have an effect on water sorption and solubility of the material.

Bulk fill materials showed lower water

sorption and solubility results which may be attributed to their polymerization mechanism

References:

1. Rees JS, Jagger DC, Williams DR, Brown G, Duguid W. A reappraisal of the incremental packing technique for light cured composite resins. *J Oral Rehabil.* Jan 2004;31(1):81-84.
2. Tantbirojn D, Versluis A, Pintado MR, DeLong R, Douglas WH. Tooth deformation patterns in molars after composite restoration. *Dent Mater.* Jul 2004;20(6):535-542.
3. Leprince JG, Palin WM, Vanacker J, Sabbagh J, Devaux J, Leloup G. Physico-mechanical characteristics of commercially available bulk-fill composites. *J Dent.* Aug 2014;42(8):993-1000.
4. Finan L, Palin WM, Moskwa N, McGinley EL, Fleming GJ. The influence of irradiation potential on the degree of conversion and mechanical properties of two bulk-fill flowable RBC base materials. *Dent Mater.* Aug 2013;29(8):906-912.
5. Berger SB, Palialol ARM, Cavalli V, Giannini M. Characterization of water sorption, solubility and filler particles of light-cured composite resins. *Brazilian dental journal.* 2009;20(4):314-318.
6. Sideridou ID, Karabela MM, Vouvoudi EC. Physical properties of current dental nanohybrid and nanofill light-cured resin composites. *Dental Materials.* 2011;27(6):598-607.
7. Ferracane JL. Hygroscopic and hydrolytic effects in dental polymer networks. *Dental Materials.* 2006;22(3):211-222.
8. Mortier E, Gerdolle DA, Jacquot B, Panighi MM. Importance of water sorption and solubility studies for couple bonding agent--resin-based filling material. *Oper Dent.* Nov-Dec 2004;29(6):669-676.

- 9.** Alshali RZ, Salim NA, Satterthwaite JD, Silikas N. Long-term sorption and solubility of bulk-fill and conventional resin-composites in water and artificial saliva. *J Dent.* Dec 2015;43(12):1511-1518.
- 10.** Hermes C, Wall B, McEntire J. Dimensional stability of dental restorative materials and cements over four years. *General dentistry.* 2003;51(6):518-523.
- 11.** Satterthwaite J, Alshali R, Silikas N. Water sorption of flowable bulk fill versus conventional resin composites. *Dental Materials.* 2013;29:e57.
- 12.** de Moraes Porto ICC, de Aguiar FHB, Brandt WC, Liporoni PCS. Mechanical and physical properties of silorane and methacrylate-based composites. *Journal of dentistry.* 2013;41(8):732-739.
- 13.** Le Bell AM, Tanner J, Lassila LV, Kangasniemi I, Vallittu PK. Depth of light-initiated polymerization of glass fiber-reinforced composite in a simulated root canal. *Int J Prosthodont.* Jul-Aug 2003;16(4):403-408.
- 14.** Misilli T, Gonulol N. Water sorption and solubility of bulk-fill composites polymerized with a third generation LED LCU. *Braz Oral Res.* Sep 28 2017;31:e80.
- 15.** Örtengren U, Wellendorf H, Karlsson S, Ruyter I. Water sorption and solubility of dental composites and identification of monomers released in an aqueous environment. *Journal of oral rehabilitation.* 2001;28(12):1106-1115.
- 16.** Ferracane J. Elution of leachable components from composites. *Journal of oral rehabilitation.* 1994;21(4):441-452.
- 17.** Tanaka K, Taira M, Shintani H, Wakasa K, Yamaki M. Residual monomers (TEGDMA and Bis-GMA) of a set visible-light-cured dental composite resin when immersed in water. *Journal of oral rehabilitation.* 1991;18(4):353-362.
- 18.** Zankuli MA, Devlin H, Silikas N. Water sorption and solubility of core build-up materials. *Dent Mater.* Dec 2014;30(12):e324-329.
- 19.** Vasiliki Maselia DDS MMKP, Konstantinos Kyritsis. Water Sorption and Solubility of Light-Cured Resin-Based Dental Restorative Nanocomposites. 2012.
- 20.** Moraes LGP, Rocha RSF, Menegazzo LM, Araújo EBd, Yukimito K, Moraes JCS. Infrared spectroscopy: a tool for determination of the degree of conversion in dental composites. *Journal of Applied Oral Science.* 2008;16(2):145-149.

