

Water sorption and solubility of hybrid and microfine resins-composite filling materials

Ahmed A. El-Hejazi, BDS, MSc, PhD

العرض من هذا البحث هو دراسة تأثير امتصاص الماء والانتحلال على خشبوات الرتنج المركب والمصلب بالضوء. تم اختبار ستة مواد للاختبار من نوعين من مواد الرتنج المركب وهي الرتنج الفائق النعومة والرتنج المركب المصنوع من Helio Progress, ClearFil, Durafil, Silux Plux, (Ful-Fil, P10). وأحرقت التجارب طبقاً للمقياس الدولي رقم ٤٠٤٩ لسنة ١٩٨٨ لمواد الرتنج المركب وأخذ في الاعتبار النسخة الأخيرة المطورة. تبين من النتائج أن أعلى درجة امتصاص كانت مادة (SLX ٣٢,٨٦ جم/سم^٣) وهي إحصائياً أعلى من بقية المواد المختبرة. وكذلك تبين أن CRF كان أقلهم (٢٨,٤٢ جم/سم^٣). بينما درجة الانتحلال تبين إن متوسط الانتحلال لمادة (DUF ٥,٧٦ جم/سم^٣) وهي إحصائياً أعلى من باقي المواد المختبرة. بينما لا يوجد اختلاف إحصائياً بين CRF, HLO, FUL, P10. الخلاصة من النتائج أن جميع المواد التي اختبرت قد احتازت اختبارات المواصفات الدولية. ولكن الرتنج الفائق النعومة DUF, HLO, SLX كانت درجة الامتصاص والتحلل فيه أكثر بكثير من الرتنج المركب المصنوع وتبين ذلك إحصائياً.

The aim of this study was to investigate the effects of water sorption and solubility of two types of light cured restorative materials (hybrid and microfine composites). Six composite materials were selected - Silux Plux (SLX), Clear Fil Photopost (CRF), Durafil (DUF), Helio Progress (HLO), P10 (P10) and Ful-Fil (FUL). The experimental methods were based closely on the procedure recommended in the ISO 4049 (1988) standard for resin-bonded filling materials, taking into account recent revisions proposed by the relevant working group. The highest mean water sorption was exhibited by SLX (32.86 g/mm³). This was significantly higher than the rest of the materials. It was also shown that CRF had the lowest mean value (8.42 g/mm³). For water solubility, the results showed that the mean value for DUF (5.76 g/mm³) was significantly higher than the rest of the materials, while CRF (0.40), HLO (0.82), FUL (0.88) and P10 (1.18) were not significantly different from one other, except P10 (1.18) which was significantly higher than CRF (0.40). All the materials passed the standard specification of ISO 4049: 1988. In general, the sorption of microfine composites (eg. SLX, HLO and DUF) was significantly greater than that of hybrid composites (FUL, P10 and CRF). The solubility of microfines was also greater than that of the hybrid composites.

Introduction

The oral environment plays an important role in the properties of the dental restorative materials.¹ Water sorption may affect dental restorative materials such as composites, by compromising their physical and mechanical properties.^{1,2,3}

The water absorbed by the polymer matrix could cause filler-matrix debonding or even hydrolytic degradation of the filler.^{3,4} Filler-matrix debonding in quartz filled as well as glass filled composites subjected to prolonged water exposure with thermocycling, have been reported.⁵ On the other hand, Calais and Söderholm⁶ concluded in their investigation on the influence of the filler type and water exposure, that water may have a more detrimental affect on the strength of the matrix at the filler-matrix interface.

The influence of water sorption on three-body wear of composite resin has been reported by Yap, Teoh and Tan.² They concluded that water sorption affects composites by increasing their wear. Øysaet and Ruyter⁷ investigated the water sorption and solubility of some posterior

composites after 3 months in water at 37°C. Their results revealed that the leaching of inorganic ions into water from the fillers varied depending on filler composition and filler treatment. Also they have shown that all fillers leach Silicon in water, with quartz composites being more stable than those based upon Barium or Strontium glasses. The degradation in properties of composites exposed to acidic media has been reported to be more extensive in Barium and especially Zinc glass composites than quartz or microfilled materials.⁸ Three factors affected the amount of leaching; the extent of the polymerization reaction; the chemistry of the solvent, and the size and chemical composition of the resin.⁹

Materials and Methods

Six composite materials, all recommended by the manufacturers for use as filling materials, were investigated (Table 1). All materials were visible-blue-light-activated, except P10 which was chemically-activated and was also described by its manufacturer as a resin-bonded ceramic. The following procedure was based closely on that recommended in the ISO 4049 (1988) standard for

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Assistant Professor, Department of Restorative Dental Sciences, College of Dentistry King Saud University, Saudi Arabia

Address reprint requests to:

Dr. Ahmed A. El-Hejazi
P. O. Box 60169 Riyadh 11545
Saudi Arabia
Fax: +966-1-467 7428
e-mail: aelhejazi@yahoo.co.uk

Table 1. Investigated dental restorative materials.

Material	Code	Filler volume (%) [*]	Batch No.	Manufacturer
Silux Plux	SLX	37.1	57020	3M Dental Products, St Paul, MN, USA
Clear fil Photopost	CRF	74.0	1621	Kuraray, Romulus, MI, USA
Durafil	DUF	37.5	168	Kulzer, GmbH Dental Division, Germany
Helio Progress	HLO	39.7	360615	Ivoclar, Liechtenstein
P10	P10	69.1	9302	3M, Dental Products, St Paul, MN, USA
Ful-Fil	FUL	52.8	9106281	Caulk, Dentsply, DeTrey, Milford, DE, USA

* Inorganic filler volume (%) as obtained from the manufacturers.

resin-bonded filling materials, taking into account recent revisions proposed by the relevant working group.

Specimen preparation

A total of 30 samples were made, consisting of five specimen discs for each material. Each specimen disc was 15 ± 1 mm in diameter and 1 ± 0.1 mm thick and was prepared using a Teflon mould. The material was prepared in accordance with the manufacturer's instructions, by filling the mould with the material using a plastic spatula to condense, and covering it with a piece of 50 ± 30 m thick polyester transparent film which was placed over the mould and finally covered by a glass slide.

By placing the exit window (5mm x 5mm) of the external energy source against the glass slide, that section of the specimen was irradiated for 60s. The exit window was then moved and another section, which overlapped the previous section of the specimen, was irradiated. This procedure was continued until the whole specimen had been irradiated. The specimens were removed from the mould and any flash if present, was removed.

Test procedure

The specimens were transferred to the desiccator containing silica gel, freshly dried for 5 hours (h) at 130°C . They were maintained in the

desiccator at $37^{\circ} \pm 1^{\circ}\text{C}$. After 24h, the specimens were removed and stored in a second desiccator which contained silica gel (freshly dried for 5h at 130°C) and stored at the lower temperature (room) of $23^{\circ} \pm 1^{\circ}\text{C}$ for 1h. The specimens were weighed using an analytical balance (Mettler Analytical Balance, Gallenkamp Mettler, E. Mettler, Zurich, Switzerland) to an accuracy of ± 0.1 mg. This cycle was repeated until a constant mass (m_1) was obtained, i.e. until the mass loss of each specimen was not more than 0.2 mg in any 24h period. The specimens were immersed in distilled water of grade 2, (ISO 3696), and maintained at 37°C for seven days. After that time, the specimens were removed, washed with water, surface water blotted away until free from visible moisture, and waved in the air for 15, then finally weighed 1 minute after being removed from the water. This mass (m_2) was recorded. The specimens were placed in the desiccator using the same cycle as described above to obtain (m_1). This cycle was repeated until constant mass (m_3) was obtained. Finally, the specimens were measured for diameter and thickness. This was done by taking three readings in the centre of the specimen to measure the thickness. The diameter was measured at four equally spaced points along the circumference. The mean values of thickness and diameter of each specimen, were used to calculate the volume (V) in cubic millimetres.

Calculations and expression of results

For water sorption, the values, W_{sp} , were calculated in micrograms per cubic millimetre for each of the specimens, by using the following equation:

$$W_{sp} = (m_2 - m_3) / V$$

where: m_2 is the mass of the conditioned specimen in micrograms, after immersion in water for seven days

m_3 is the reconditioned mass of the specimen in micrograms and V is the volume of the specimen in cubic millimetres.

For solubility, the values for solubility, W_{st} , were calculated in micrograms per cubic millimetre for each specimen using the following equation:

$$W_{st} = (m_1 - m_3) / V$$

where: m_1 is the conditioned mass in micrograms
 m_3 is the reconditioned mass of the specimen in micrograms and V is the volume of the specimen

in cubic millimetres.

The data on water sorption and solubility were subjected to Student-Newman-Keuls test at $p < 0.05$.

Results

The mean values, standard deviation, standard error and 95% confidence intervals for water sorption are presented in Table 2. The highest mean water sorption was exhibited by SLX (32.86 $\mu\text{g}/\text{mm}^3$). This was significantly higher than the rest of the materials. It was also shown that CRF had the lowest mean value (8.42 $\mu\text{g}/\text{mm}^3$) for water sorption.

Table 3 shows the mean values, standard deviation, standard error, and 95% confidence intervals ($\mu\text{g}/\text{mm}^3$) for water solubility. The mean value for DUF (5.76 $\mu\text{g}/\text{mm}^3$) was significantly higher than that for the rest of the materials, while CRF (0.40 $\mu\text{g}/\text{mm}^3$), HLO (0.82 $\mu\text{g}/\text{mm}^3$) and FUL (0.88 $\mu\text{g}/\text{mm}^3$) were not significantly different from one another, and P10 (1.18 $\mu\text{g}/\text{mm}^3$) which was significantly higher than CRF, HLO and FUL.

Table 2. Mean values ($\mu\text{g}/\text{mm}^3$), standard deviations, standard errors and 95% confidence intervals for water sorption data.

Mater	No	Mean*	SD	Error	95% Confidence interval for the mean
CRF	5	8.42 a	0.40	0.18	7.92 to 8.92
DUF	5	19.42 b	0.25	0.11	19.11 to 19.73
FUL	5	14.82 c	0.36	0.16	14.37 to 15.27
HLO	5	21.42 d	0.15	0.07	21.24 to 21.60
P10	5	11.56 e	0.29	0.13	11.20 to 11.92
SLX	5	32.86 f	0.21	0.09	32.60 to 33.12

* Different alphabets show significant differences.

Table 3. Mean values ($\mu\text{g}/\text{mm}^3$), standard deviations, standard errors and 95% confidence intervals for water solubility data.

Mater	No	Mean*	SD	Error	95% Confidence interval for the mean
CRF	5	0.40 a	0.55	0.24	0.28 to 1.08
HLO	5	0.82 ab	0.23	0.10	0.54 to 1.10
FUL	5	0.88 ab	0.38	0.17	0.40 to 1.36
P10	5	1.18 ab	0.30	0.13	0.81 to 1.55
SLX	5	4.31 c	0.29	0.13	3.96 to 4.67
DUF	5	5.76 d	0.53	0.24	5.09 to 6.42

* The mean values with the same alphabetical letter are not significantly different.

Discussion

The ISO Standard (4049:1988) method for water sorption and solubility with measurements of

mass at one week establishes minimum performance criteria. It appears from the results of water sorption and solubility that all the composite materials evaluated pass this standard. However, in this instance, the data can be examined further to illustrate significant differences between these materials.

In general, the sorption of microfine composites (e.g. SLX, HLO and DUF) was greater than that of hybrid composites (FUL, P10 and CRF). The solubility of microfines was also larger than that of the hybrid composites. The structural features which underlie these differences are as follows:

1. Greater polymer (organic) content in microfines

The interfaces between the filler particles and the polymer matrix accommodate any amounts of water that was absorbed.^{3,10} In agreement with these studies, SLX micofilled composite resin which has lower filler content (56% wt, 37.5% Vol.) showed 32.86 $\mu\text{g}/\text{mm}^3$ sorption and 4.31 $\mu\text{g}/\text{mm}^3$ solubility. While CRF hybrid composite with the higher filler content (79% wt, 74% Vol) showed the lowest amount of sorption (8.42 $\mu\text{g}/\text{mm}^3$) and solubility of (0.40 $\mu\text{g}/\text{mm}^3$). Exposure to water significantly increases three-body wear.² Therefore, the lower the amount of water absorbed, the greater is the wear resistance.

2. Variation in the extent of cure or network formation

The second possible cause for differences in water sorption and solubility relates to the degree of cure of the polymer network. Inadequate polymerization of dental composite probably increase the solubility and may also increase the water sorption, since the network will be less tightly cross-linked. Pearson and Longman,¹¹ however, concluded that inadequate polymerization of microfine composites has a greater effect on water sorption than on solubility.

The difference between the water sorption of the microfines relative to the hybrid composites is greater than the corresponding difference between the solubility data (Tables 2 and 3).

3. Variation in the susceptibility of the materials to elute

The third factor is the variation in the

susceptibility of composite materials to elute elements from filler particles and other degradation products.^{12,13} The dislodgement of filler particles has been correlated to the causes of composite wear.^{3,7} Ruyter and Øysaed,¹⁴ also showed that elements were more readily leached into water from composites containing Zinc, Barium and Strontium glass than they were from composites which incorporated particles of silica. More recently, Ferracane⁹ has concluded that elution of leachable components from composite is rapid with the majority being released within a matter of hours. Composites containing quartz filler are more stable than those based on metallo-silica glasses. Further studies are needed to investigate the effect of solubility and water absorption on longevity and discoloration of resin-composite filling materials.

Conclusions

The following conclusions were drawn:

1. All materials investigated pass the standard specification ISO 4049: 1988.
2. The water sorption in microfine composites was greater than in hybrid composites.
3. The solubility of microfines was also larger than that of the hybrid composites.

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