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Water sorption and solubility of core build-up materials

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ABSTRACT

Objectives. To investigate the variation in water sorption and solubility across a range of different core build-up materials.

Methods. Five materials were tested, four of which are resin-based materials (Grandio Core, Core.X Flow, Bright Flow Core, Speedee) and one resin-modified glass ionomer (Fuji II LC). All specimens ($n = 10$) were immersed in 10 ml distilled water in individual glass containers and weighed at one week, 14 and 28 days. After a total immersion time of 28 days, 7 specimens were dried to a constant mass, in a desiccator for 28 days. Three samples of each material were not dried, but were left in distilled water for 1 year, to determine the long-term water sorption properties. Specimens were weighed at monthly intervals until 6 months and then at the 9th and 12th months. Each specimen was measured using a digital electronic caliper (Mitutoyo Corporation, Japan).

Results. After 28 days immersion, the change in water sorption and solubility of the materials ranged from 12.9 to 67.1 μg/mm^3 ($P < 0.001$) and 0.9–6.4 μg/mm^3 respectively ($P < 0.001$). Except for Fuji II LC, an independent *T*-test showed significantly higher water sorption and solubility for the other materials after 1-year total immersion in water compared to 1 month ($P < 0.05$). Using repeated measures ANOVA, all materials showed mass changes over time (1 month) ($P < 0.001$).

Significance. Grandio Core had the lowest water sorption and solubility among the tested materials. According to the ISO 4049 standards, all the tested materials showed acceptable water sorption and solubility, apart from the water sorption behavior of Fuji II LC.

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1. Introduction

Water sorption and solubility can affect the mechanical strength, color stability, and abrasion resistance of resin-composites [1]. Moreover, resin-composites have shown reduced strength and longevity, as a result of extensive water sorption and solubility behavior [2,3].

The resin composite's polymer network will absorb a certain amount of water and release monomers and ions to the surrounding environment [4–7]. Water most likely permeates through the network due to its porosity and intermolecular

spaces, so the water gained is highly dependent on the density of the polymer network.

Dental composite restorations in a patient's mouth are continuously exposed to a wet environment. The water absorbed and consequent hygroscopic expansion of a composite may compensate for the polymerization shrinkage-stress. Endodontically treated posterior teeth as well as anterior teeth with extensive coronal loss, may need to receive crowns after being built-up with core materials [8,9]. If a final restoration is a resin based direct restoration, it will be exposed to a wet environment and will absorb water. Water sorption for a core material may not be avoided even when the final restoration is a crown, as after the cementation of a crown, water may still reach the core material from diffusion through the cement [10]. Thus, evaluating the water sorption of core materials is critical for an understanding of their long term success.

In situations where there has been extensive coronal destruction, marginal leakage under crowns may result in the core build-up margins coming in contact with moisture. Marginal leakage will occur with poor adaptation of a casting to the margins, or lack of integrity of the chosen luting cement. Also, a core material may come in contact with moisture if it is used as an interim restoration. Moisture in the oral environment may result in dimensional changes, swelling and hygroscopic expansion. This can lead to micro cracks of the resin based core materials and subsequently a failure of the restoration [11]. There are no long term studies that have investigated water sorption of core build-up materials.

Using resin based core build-up materials may contribute to the failure of the overlying restorations. The fracture resistance of zirconia copings has been shown to be affected by the water absorption of the luting agent [12]. It is well known that the hygroscopic expansion of resin modified glass ionomers and compomers can lead to the failure of all ceramic crowns, when these materials are used for either core build up or adhesive bonding [11].

Significant differences between dental resin-based materials, regarding their water sorption and solubility have been shown [13,14]. 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 month.
2. There is no difference in the water sorption and solubility of the tested materials between 1 month total immersion and 1 year total immersion.

2. Materials and methods

The tested materials and relevant information are shown in Table 1. The specimens were prepared according to ISO 4949:2009 and the manufacturer's instructions. Specimens were prepared in cylindrical molds (16.0 mm diameter × 1.0 mm thickness) at (23 ± 1) °C and the material packed was slightly overfilled into a brass ring mold set on a piece of transparent polyester film on a glass microscopic slide. It was then covered with another piece of polyester film while being

Table 1 – All tested materials, their composition and manufacturers information.

Material	Composition	Manufacturer
Grandio Core	*Dual cure composite:* UDMA, BIS-GMA. Fillers (77% wt)	Voco, Germany
Core.X Flow	*Dual cure composite:* UDMA, Di&tri-functional methacrylates. Fillers (69% wt)	DENTSPLY, Caulk, USA
Bright Flow Core	*Dual cure composite:* Methacrylate polymers (48%). Fillers (52% wt)	DMPLTD, Greece
Speedee	*Dual cure composite:* Self-adhesive resin hydrophilic based monomer	DENTSPLY, Caulk, USA
Fuji II LC	*Light cured resin reinforced glass-ionomer cement:* Powder: fluoroaluminosilicate glass, polycyclic acid Liquid: water, polyacrylic acid, HEMA	GC, UK Ltd.

pressed by another glass slide. The specimens were then light cured by an Optilux curing unit (Optilux 501, USA) with an irradiance of 620 mw/cm^2. Five overlapping sections (20 s) on each side of the specimen were irradiated. A 1000 grit silicon carbide paper was used to remove any excess flash, to finish the specimens and to obtain uniform thickness. The thickness and diameter of each specimen were measured at 4 and 2 points respectively, using a digital electronic caliper (Mitutoyo Corporation, Japan). Mean values were used to calculate the volume of each specimen in mm^3. The specimens were then incubated in a lightproof desiccator with anhydrous self-indicating silica gel at (37 ± 1) °C. After 22 h, the specimens were transferred to another desiccator at (23 ± 1) °C for 2 h and then weighed to an accuracy of ± 0.1 mg using a calibrated electronic analytical balance with precision of 0.01 mg (Ohaus Analytical Plus, Ohaus Corporation, USA). This cycle was repeated until the mass change of each specimen was not more than ± 0.1 mg in any 24 h period to ensure completion of polymerization and dehydration. This constant mass (m_1) was the initial mass of the specimen.

Specimens of each material ($n = 10$) were immersed in 10 ml distilled water in individual glass containers for a total immersion time of 28 days. They were weighed every day for the first week, then after 14 days and finally after 28 days. Specimens were gently dried on filter paper until free from visible moisture, waved in air for 15 s and weighed 1 min later to ± 0.01 mg and returned to the glass containers filled with distilled water. The recorded mass was denoted as m_2 (t, time).

After a total immersion time of 28 days, not all 10 specimens were dried to a constant mass. Seven specimens of each material were dried to constant mass (m_3) in the desiccators for 28 days using the cycle described above. Three samples of each material were not dried and were left in distilled water for a total immersion time of 1 year just to determine whether the water sorption and solubility will be different from the one 1 month group after long term water immersion in water of 1 year. These groups (3 samples for each material) were

Table 2 – Mean (SD) values of water sorption and solubility for all groups, for a total immersion period of 28 days as well as 1 Year. Within each column different superscripts indicate statistical significant difference (P < 0.001). Within the same row of the same property, different superscript numbers indicates statistical significant difference for each material.

Materials	Water sorption (μg/mm^3) after 28 days	Water sorption (μg/mm^3) after 1 year	Water solubility (μg/mm^3) after 28 days	Water solubility (μg/mm^3) after 1 year
Grandio Core	12.9^{a,1} (0.5)	14.5^{a,2} (0.6)	0.9^{a,1} (0.05)	1.07^{a,2} (0.1)
Core.X Flow	13.6^{a,b,1} (0.8)	16.9^{a,b,2} (1.3)	1.3^{b,2} (0.1)	1.5^{b,2} (0.1)
Bright Flow Core	15.0^{b,1} (0.9)	19.5^{b,c,2} (0.2)	1.8^{c,1} (0.1)	2.5^{c,2} (0.1)
Speedee	18.2^{c,1} (0.8)	22.9^{c,2} (0.8)	3.5 ^{c,1} (0.1)	4.0 ^{d,2} (0.2)
Fuji II LC	67.1^{d,1} (0.5)	72.2^{d,1} (7.8)	6.4 ^{d,1} (0.5)	7.1^{e,1} (0.2)

measured monthly up to 6 months and then at 9 and 12 months. After a total of 1 year immersion, these groups were, dried to constant mass (m_3) for 120 days as previously described. Water sorption and solubility were calculated for tested materials after 1 month and after 1 year. Sorption and desorption cycle mass change percentage was calculated for both groups (1 month group and 1 year group). Water sorption, solubility and mass change percentages were calculated by the following equations:

    1. Mass change percentages

(A) Sorption mass change percentages

$$Mg\% = \frac{m_{2(t)} - m_1}{m_1} \times 100$$

(B) Desorption mass change percentages

$$Mg\% = \frac{m_{3(t)} - m_1}{m_1} \times 100$$

    2. The water sorption (W_{SP}) and solubility (W_{SL}) in μg/mm^3 were calculated by:

(A) $W_{SP} = m_2 - m_3/v$
(B) $W_{SL} = m_1 - m_3/v$

2.1.   Statistical analysis

One-way ANOVA followed by either Tukey or Dunnett's (depends on whether equal variances are assumed or not on each ANOVA test) post hoc tests were used to detect statistical differences of the ($P < 0.05$) water sorption and solubility of the tested materials after 1 month and after 1 year. The independent T test was used to detect any differences between the water sorption and solubility of the 1 month group and the 1 year 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 ($P < 0.05$).

3.   Results

3.1.   Water sorption (W_{SP}) and solubility (W_{SL})

Water sorption and solubility of the tested core build-up materials are presented in Table 2 and Figs. 1 and 2. The tested materials were significantly different from each other with regard to both their water sorption and solubility. Grandio Core

had the smallest water sorption (12.9 μg/mm^3) and solubility (0.9 μg/mm^3) among the tested materials, while Fuji II LC had the highest (67.1 μg/mm^3) after 28 days as well as after 1 year (72.2 μg/mm^3). The water sorption of Bright Flow Core after 1 month was not significantly different from Core.X Flow ($P = 0.07$) but was different from Speedee ($P < 0.001$). P values for Grandio Core, Core ($P = 0.002$), Core.X Flow (0.001), Bright Flow Core ($P < 0.001$), Speedee ($P = 0.001$) indicated that water sorption of the 1 year groups was significantly different from that of
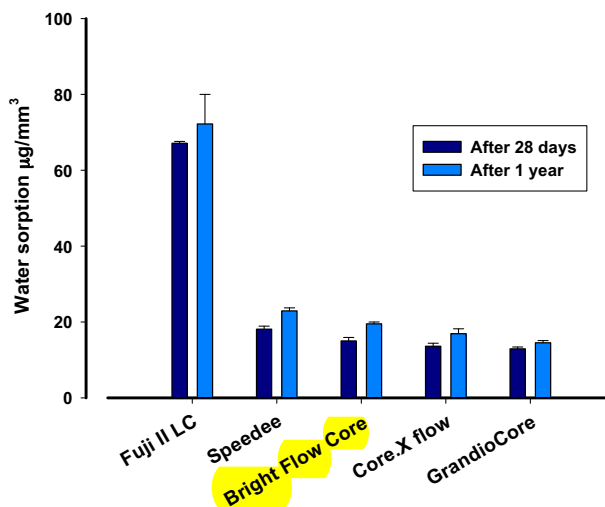
Fig. 1 – Mean (SD) values of water sorption (μg/mm^3) for all groups after 28 days versus 1 year.
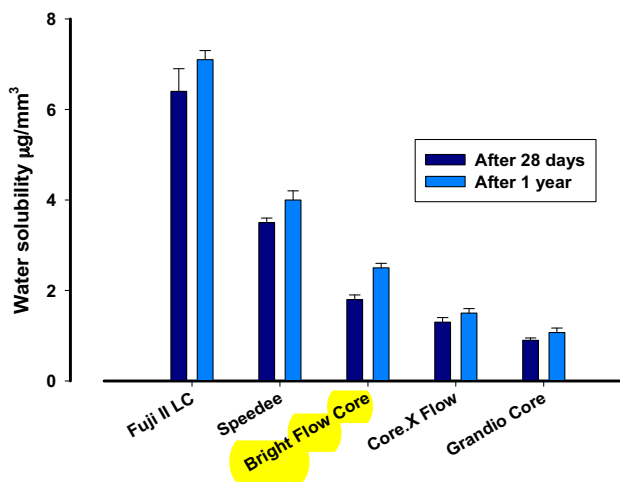
Fig. 2 – Mean (SD) values of water solubility (μg/mm^3) for all groups after 28 days versus 1 year.

the 1 month groups. After 1 year, all the tested resin-composite materials showed significantly higher solubility compared to their solubility after 1 month (Grandio Core $P = 0.01$, Core.X Flow $P = 0.001$, Bright Flow Core $P < 0.001$ and Speedee $P = 0.001$). Water sorption ($P = 0.113$) and solubility ($P = 0.06$) of Fuji II LC was not different from that of the 1 month group ($P = 0.113$ >0.05). Regression analysis of the sorption and solubility of Grandio Core, Core.X Flow and Bright Flow Core versus filler % were as following: $r^2 = 0.99$ for W_{Sp} after 1 month and $r^2 = 0.97$ after 1 year. For W_{SL} after 1 month $r^2 = 0.98$ and $r^2 = 0.99$ after 1 year.

3.2. Mass change percentages

Using ANOVA repeated measures, all materials showed significant sorption mass changes percentages over time (1 month) ($P < 0.001$).

The mass change percentages for the tested core build-up materials at different times as well as the water sorption/adsorption mass percentage for each material both after 1 month and 1 year are shown in Figs. 3–6.

4. Discussion

The current study compared the water absorption behavior of several commercial resin-based materials of varying composition. This was done at two different times, over a period of 1 month, and using a different sample over 1 year. All the materials showed a continuous increase in the amount of water gain all through the first 28 days, but the maximum water gained for all the tested materials was during the first week. This was in agreement with several studies in terms of the maximum amount of water sorption gained within the first week [15,16].

The tested materials showed significantly different water sorption behavior. After 1 month, apart from Fuji II LC, which had higher water sorption values than those stated by the ISO 4049 standards, all the tested materials showed accept-
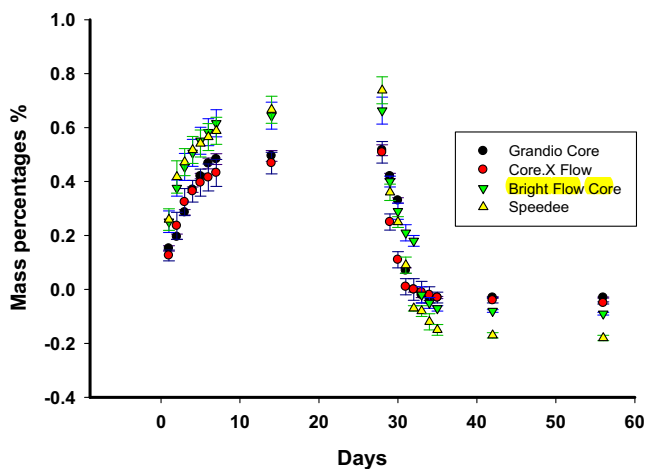

Fig. 4 – **Water sorption/desorption cycle of Fuji II LC after28 days of total immersion.**

able water sorption behavior, below 40 µg/mm^3 (the maximum water sorption stated by the ISO 4049) [17]. All materials showed significantly different water solubility from each other after a total immersion of 28 days. All the materials showed acceptable solubility behaviors below 7.5 µg/mm^3, which is the maximum water solubility stated by ISO 4049. To sum up, the first null hypothesis of no difference in the water sorption and solubility of the tested core materials after 1 month was rejected.

Comparing the sorption of 1 month to 1 year, it can be concluded that most of the water gain (mass gain) was achieved in the first month. After the first month, the water gain increased at a much lower rate, and all materials reached a plateau within the first 6 months, this is in agreement with one study in which all resin-composite materials reached a plateau within the 150 days [7]. However, this small increase in the sorption and solubility of the 1 year groups was significantly different from the sorption and solubility of the same

Fig. 3 – **Mass changes % (water sorption/desorption cycles) of the tested materials after 28 days. The mass change in Fuji II LC (Fig. 4) was far higher than the rest of the materials.**
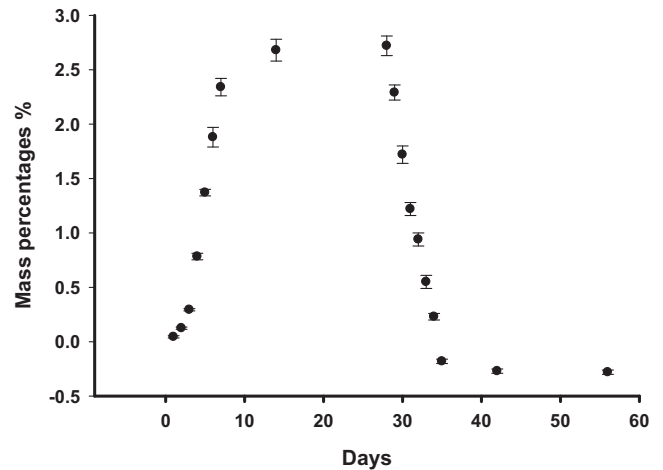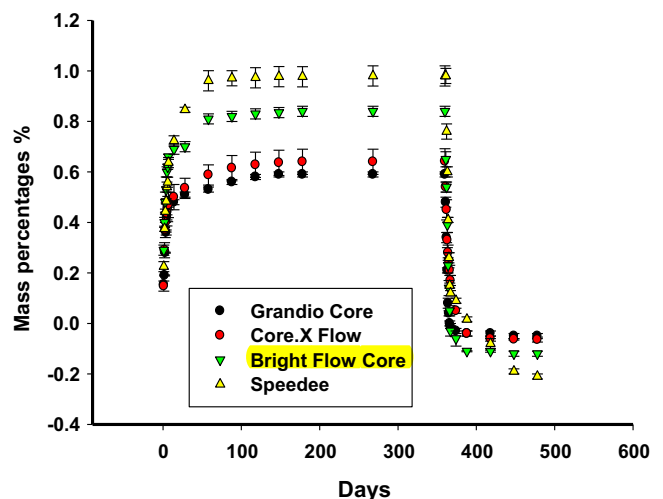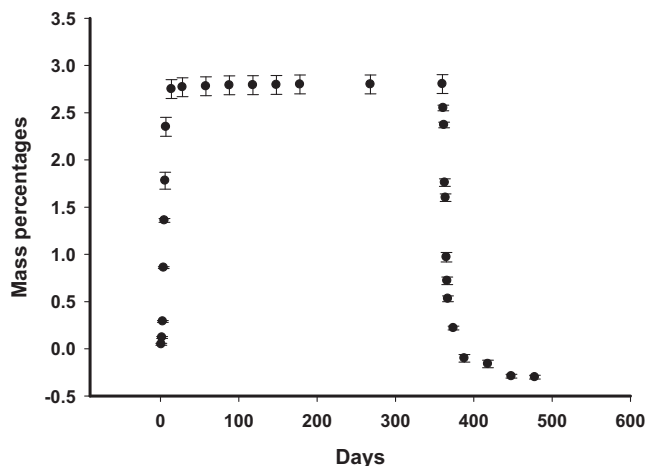

Fig. 5 – **Mass changes % (water sorption/desorption cycles) of the tested materials after 1 year of total immersion. The mass change in Fuji II LC is shown in Fig. 6.**

Fig. 6 – Water sorption/desorption cycle of Fuji II LC after 1 year of total immersion.

materials after 1 month for all the materials. Also, water solubility of all materials was significantly different from their solubility at 1 month. Therefore, the second null hypothesizes was rejected. However, Fuji II LC's water sorption and solubility after 1 year were not significantly increased. Fuji II LC had a far higher water sorption and solubility than the other tested materials after 1 month which made the further increase insignificant; even after 1 year Fuji II LC is still far higher than the rest of the materials. Fuji II LC showed higher sorption and solubility than the rest of the tested resin based materials this is in agreement with a previous study tested the water sorption and solubility of resin based materials where Fuji II LC had higher values than the rest of the materials after 1 week storage in water [18].

The differences between materials may be attributed to their composition. Hydrophilicity of the monomers used in resin based materials can play a major role in the water sorption of resin-composites. The resin-composites tested showed lower water sorption values than the light-cured reinforced glass ionomer (Fuji II LC). This may be attributed to the fact that the matrix in the tested resin composites is more hydrophobic than the one used in Fuji II LC (HEMA) [18]. Moreover, the water uptake in resin composites is only limited to the matrix, while in the glass ionomer it affects the entire hydrogel structure, which leads to a more rapid water uptake.

The amount of water gained in a composite material depends on the content of its hydrophilic monomers [19]. Speedee is a hydrophilic material based on an embrace monomer which is believed to be more hydrophilic than other monomers such as urethane dimethacrylate (UDMA) and Bis-GMA, which means it absorbs more water. This may be the reason why Speedee had a significantly higher amount of water sorption than Grandio Core, Core.X Flow and Bright Flow Core. However, Speedee had a significantly lower amount of water sorption than the more hydrophilic monomers Fuji II LC.

The presence of fillers also seems to have an impact on the sorption and solubility of the tested materials. The difference in their filler loading percentages may explain some of the differences. There was a strong inverse correlation between

the first three materials with the lowest water sorption and solubility and their filler loading. The difference in the composition of the tested materials may explain the differences between them and also why three materials had less water sorption and solubility than the other two materials.

In the present study the solubility behavior was measured for 2 groups, 1 month, and 1 year. The solubility for each material in both groups was very similar to their sorption behavior. The solubility of the 1 year group was slightly higher than the solubility of the 1 month groups. That may be explained by the fact that exposing resin based materials to water for a long time may lead to the degradation of the resin matrix and debonding of the filler matrix interlayer [20,21]. Similarly, in this study the 1 year group was subjected to a continuous attack of water for 1 year which may have resulted in a more extensive degradation of the materials where all these degradation products were eluted and consequently resulted in increased solubility compared to the 1 month group.

The solubility of all tested materials (except Fuji II LC) was lower than $7.5\,\mu g/mm^3$. This means that those materials showed acceptable solubility behavior (according to ISO 4049). It was reported that solubility can be expected to be correlated with the water sorption of a material. This is because unreacted components can only be leached out when water penetrates the material [22]. This is in accordance with the current study as all the materials had the same ranking with respect to their sorption and the water solubility. The density of crosslinked dimethacrylates can vary. The extent of this will determine the amount of residual monomers that can leach out [14]. It is likely that the difference in solubility between the materials can be due to the cross links exists in each of the materials. This is because the solvent permeability of a polymer can be decreased by the existence of cross links between the polymer chains as they decrease the free hole-free volume and reduce the swell of the polymer chain [2,23]. Therefore, the more cross-linked the polymer network, the less the water sorption and solubility of the resin based material [21]. In this study Grandio Core had a significantly lower water sorption and solubility than the other materials that may be due to a more cross-linked polymer network. Filler content is believed to affect the solubility of resin-composites [24] that is in agreement with our study as there was a strong positive correlation between filler loading and sorption and solubility for Grandio Core, Core.X Flow and Bright Flow Core.

5.   Conclusion

All tested core build-up materials absorbed water and showed significantly differing water sorption behavior. Grandio Core had the lowest water sorption and solubility among the tested materials. According to the ISO standards, all the tested materials showed acceptable water sorption and solubility apart from the water sorption behavior of Fuji II LC.

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