Changes in expansion and mechanical strength during water storage of a traditional and three modified resin composites

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Munksgaard EC. Changes in expansion and mechanical strength during water storage of a traditional and three modified resin composites. Acta Odontol 2002;60:203–207. Oslo. ISSN 0001-6357.

Four types of resin composites (Dyract AP (DYR), Definite (DEF), Ariston pHc (ARI), and Spectrum TPH (SPE)) were tested after water storage for up to 180 days. The test parameters were flexural strength, flexural modulus, and marginal gaps at fillings in cylindrical cavities. The cavity tests were performed in two ways. One series of fillings was finished shortly after curing and the maximal gaps were followed with time during water storage. Another series was finished immediately before gap size measurement and after water storage for various periods. Absence of or reduced gap sizes at 180 days compared to 1 h or 1 day was observed for all materials and with both methods. At 180 days, DYR and ARI showed significantly smaller gap sizes than those observed with DEF and SPE. No significant differences in flexural strength were observed when means at 180 days were compared to those observed at 1 day. DEF, ARI, and SPE showed a significant increase in flexural moduli at 180 days compared to 1 day, while DYR showed none. It is concluded that fillings made of Dyract AP or Ariston pHc may show absence of or very small gap sizes between filling and cavity wall after 6 months and with no significant reduction in mechanical strength.
☐ *Flexural modulus; flexural strength; hygroscopic expansion; marginal gap; resin composite*

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Resin composites are increasingly used for restorative purposes because of good esthetics and the capability of establishing a bond to enamel and dentin. The color stability, wear, and fracture resistance of these materials have been greatly improved since their introduction about 50 years ago, but one imperfect property remains, namely gap formation caused by polymerization contraction during setting, leading to marginal discoloration and leakage (1). Leakage may induce caries and pulpal damage due to invasion of bacteria and their toxins.

Bonding, combined with an appropriate cavity design and filling technique, may reduce the gap formation caused by polymerization contraction (2), but gaps can still be present owing to insufficient bonding, or may develop later during thermal and load stress.

Modifications of the traditional resin composites have been introduced that may reduce development of, or damage caused by, marginal leakage. Examples are compomers, with fluoride release which might be beneficial (3), Ormocers, which because of their special polymer structure develop smaller gaps (4–6), and an ion-releasing product (Ariston pHc), which acts as a buffer during cariogenic processes and thus reduces or prevents dissolution of hydroxyapatite (5, 7).

In addition, over time these products may absorb relatively high amounts of water, a process that might be beneficial because the resulting hygroscopic expansion will reduce gaps that may have formed (8–11). Such water uptake may reduce the mechanical properties and thus increase wear rate (9). Detrimental water absorption has also been observed with resin-modified glass ionomer cement (11).

The aim of our study was to test and compare the hygroscopic expansion in dentin cavities of a Compomer (Dyract AP), an Ormocer (Definite), an ion-releasing resin composite (Ariston pHc), and a traditional resin composite (Spectrum TPH), and to measure changes in mechanical properties of the materials due to extended water storage. This was to test the hypothesis that resinous materials with relatively large water absorption will show reduced gap sizes around fillings in cavities as well as reduced mechanical strength.

Materials and methods

The materials used in this investigation, listed in Table 1, were all mixed and handled in accordance with the manufacturer's instructions.

A cavity test was performed as described previously (12). Briefly, extracted human teeth, stored in 0.5% chloramine T, were ground on one of the approximal sides with wet carborundum paper (#220) until a flat dentin surface appeared. A cylindrical cavity (diameter 3.1 mm and depth approximately 1.5 mm) was prepared in the dentin with a burr. The six cavities used for each experiment were filled with material (Table 1) with the use of a Hawe-Neos syringe (Hawe-Neos, Dental Gentilino, Switzerland); the cavity walls were not pretreated. The slightly overfilled cavity was covered with a transparent plastic strip and

Table 1. Materials used in the investigation

Material	Abbreviation	Manufacturer	Lot no.	Shade
Dyract AP	DYR	Dentsply DeTrey, Konstanz, Germany	0005000275	A3
Definite	DEF	Degussa AG, Hanau, Germany	231	A3
Ariston pHc	ARI	Vivadent, Schaan, Liechtenstein	A06719	—
Spectrum TPH	SPE	Dentsply DeTrey, Konstanz, Germany	0005000595	A3

polymerized for 40 s with a lamp (XL 3000, 3M, CO, USA). The specimens were then stored in water at 37°C.

In one series of experiments, designated Gap-I, excess filling material was removed after 1 h by gently grinding on wet carborundum paper (#1000) followed by polishing on linen with a slurry of Alfa Micropolish, 0.1 μm (Buehler Ltd., Chicago, IL, USA). The specimens were then inspected using a microscope (×1000) equipped with a measuring ocular and the maximal gap at the cavity margin between dentin and the polymerized material was measured. Five other groups of specimens were measured after 1, 7, 30, 90, and 180 days' water storage, and with the excess material removed immediately before inspection. Fig. 1 illustrates how the gap measurements were performed.

In another series of experiments designated Gap-II, the four groups of specimens from the Gap-I experiment which were measured after 1 h storage were further stored in water at 37°C and the width of the maximal gaps was then measured again after 1, 7, 30, 90, and 180 days.

The total number of specimens in the group designated Gap-I was $6 \times 4 \times 6 = 144$, while it was $6 \times 4 = 24$ for the group designated Gap-II.

Flexural strength was measured according to ISO 4049:2000; this testing gave in addition a measurement of the flexural modulus. Briefly, a test specimen with length = 25 mm and height = width = 2 mm was made from mold. The dimensions of the specimens were measured after storage (see below), and each specimen was then loaded in a testing machine under water at 37°C at a speed of 0.75 mm/min; there was a span of 20.00 mm between the supports. Each group consisted of six specimens and measurements were performed after storage in water at 37°C for 1 h, as well as for 1, 7, 30, 90, and 180 days. The total number of specimens was accordingly $6 \times 4 \times 6 = 144$.

The results from each group of experiments comprising six specimens were calculated as the mean and standard deviation.

Table 2. Results from the cavity test and mechanical properties as means (SD). *n* denotes number of fillings out of six showing absence of gaps. For each material and within each column, a mean marked with an asterisk is significantly different from the mean estimated at 180 days

		Gap-I		Gap-II			
Material	Time	μm (SD)	*n*	μm (SD)	*n*	Flexural strength MPa (SD)	Flexural modulus GPa (SD)
DYR	1 h	*11.3 (1.2)	0	*11.3 (1.2)	0	*89.1 (15.7)	*4.39 (0.64)
	1 d	*9.3 (3.8)	0	*10.8 (1.9)	0	116.8 (22.4)	7.55 (0.31)
	7 d	4.1 (3.7)	1	*9.3 (0.8)	0	136.9 (7.3)	7.95 (0.37)
	30 d	3.6 (3.7)	1	7.1 (1.2)	0	116.4 (16.8)	8.43 (0.75)
	90 d	0.8 (0.7)	2	4.6 (1.8)	3	118.2 (24.2)	7.99 (0.54)
	180 d	0.5 (0.5)	3	1.1 (1.6)	3	129.3 (22.3)	8.09 (0.33)
DEF	1 h	*16.2 (1.7)	0	*16.2 (1.7)	0	*78.5 (5.1)	*5.05 (0.30)
	1 d	*14.6 (2.5)	0	14.7 (0.8)	0	91.8 (12.4)	*6.98 (0.55)
	7 d	11.7 (2.4)	0	14.4 (1.1)	0	104.2 (11.9)	*7.16 (0.28)
	30 d	8.3 (4.3)	0	14.7 (1.1)	0	98.4 (12.0)	7.95 (0.20)
	90 d	8.9 (1.0)	0	13.6 (2.5)	0	98.3 (13.4)	*6.84 (0.87)
	180 d	6.8 (2.0)	0	12.4 (2.7)	0	104.3 (16.2)	8.11 (0.45)
ARI	1 h	*12.5 (0.8)	0	*12.5 (0.8)	0	91.4 (6.1)	*3.83 (0.28)
	1 d	*12.4 (1.8)	0	*10.9 (0.9)	0	107.5 (4.2)	*6.58 (0.33)
	7 d	7.2 (1.5)	0	9.0 (0.9)	0	110.6 (13.1)	*6.68 (0.31)
	30 d	4.9 (4.3)	1	7.6 (1.8)	0	91.6 (14.1)	*6.58 (0.39)
	90 d	1.0 (1.5)	3	1.6 (1.5)	2	103.6 (6.6)	7.38 (0.37)
	180 d	0.5 (1.2)	5	0.2 (0.4)	5	90.5 (9.4)	7.63 (0.42)
SPE	1 h	15.6 (1.7)	0	*15.6 (1.7)	0	*96.3 (10.1)	*5.65 (0.36)
	24 h	*16.8 (5.0)	0	*16.1 (2.0)	0	116.9 (16.5)	*8.05 (0.45)
	7 d	14.8 (4.9)	0	14.5 (1.8)	0	133.2 (11.5)	*8.82 (0.31)
	30 d	9.7 (4.4)	0	14.4 (2.4)	0	122.8 (10.2)	*8.97 (0.42)
	90 d	9.8 (2.6)	0	11.0 (2.3)	0	115.3 (8.2)	*8.61 (0.71)
	180 d	9.0 (3.3)	0	10.3 (2.0)	0	129.8 (23.4)	9.86 (0.65)

h = hours; d = days.

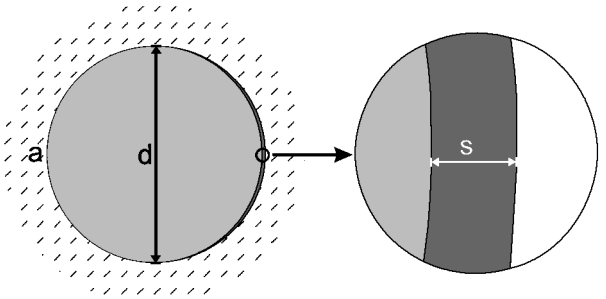

Fig. 1. Gaps at the margins are seen crescent-shaped and the width of the gap (s) at its maximum was measured (small circle that is magnified). A gap was normally seen only at one side, but occasionally an additional gap was seen at the location designated a. In those cases the width of that gap was added to s. The wall-to-wall contraction was then calculated as s/d in %; d is the diameter. In case of marginal irregularities of the edges, which seldom occur, the distance was measured to an estimated median line.

Statistical comparisons were performed as follows. For each material, the gap sizes representing Gap-1 were treated with the Kruskal-Wallis test and those representing Gap-2 with the Friedman test. In both cases, multiple comparisons versus the 180-day values were performed (13). Since eight sets of comparisons were made on the results of the Gap-1 and Gap-2 experiments, a 1% level of significance was applied.

In addition, the Kruskal-Wallis test with multiple comparisons (5% level) was performed on the $4 \times 6 \times 2 = 48$ gap sizes measured after 180 days for both the Gap-1 and Gap-2 experiments (13). The results from the measurements of the flexural strengths and moduli where analyzed by ANOVA, and the 180-day values were tested against the other values for each material with Tukey's multiple range test at 5% level of significance (14).

Results

The results are presented in Table 2. For each of the four materials, the Table gives the means and standard deviations of gap sizes for the Gap-I and Gap-II experiments as well as the flexural strengths and moduli at each measuring period. The number of cavities with absence of gaps at each measuring period is given in addition.

The statistical calculations on the Gap-I and Gap-II experiments are given in Table 3 showing in all cases a p-value greater than or equal to 3×10^{-3}. The results from the multiple comparison analysis are given in Table 2. For each material, an asterisk indicates a mean gap, which is significantly different from that measured after 180 days' water storage. The Table also indicates that for two of the materials, DYR and ARI, mean gaps around 1 μm or less were seen at the 180-day measuring period. In that period, half the cavities filled with DYR showed an absence of gaps and for ARI, 5 out of 6 cavities were without gaps, as indicated in the columns marked n. The results from the Kruskal-Wallis test performed on the gap sizes measured after 180 days' water storage in both experiments (Gap-1 and Gap-2) are given in Table 3. Multiple comparison analysis revealed that the two mean gaps for DYR at 180 days were not significantly different from those obtained with ARI, but were significantly different from those obtained with DEF and SPE, which were not significantly different. These results can be expressed: DYR = ARI ≠ DEF = SPE.

The results from the ANOVA analysis performed on the flexural strengths showed $F = 7.37$, $p = 5 \times 10^{-14}$. The results from Tukey's test on these measurements are given as asterisks in Table 2. For DYR, DEF, and SPE, the mean flexural strengths measured after 1 h were significantly different from the means at 180 days. The same type of analysis was performed on the results from the flexural moduli measurements. The analysis gave $F = 56.0$, $p = 3 \times 10^{-53}$ and the results from Tukey's test are given as asterisks in Table 2. This latter analysis revealed for DYR that only the means at 1 h were significantly different from that at 180 days. For the other materials (DEF, ARI, and SPE), significant differences from the 180-day means were seen for those measured at 1 day and 7 days or later, indicating for these materials an increasing stiffness with time.

Discussion

It is well established that water uptake by fillings of resin composites may cause a reduction in marginal gaps (15). In the above experiments, the effect of water storage on

Table 3. Statistical analysis of the results from the two cavity tests

Gap-1			Gap-2			180 days, Gap-1 + Gap-2		
Kruskal-Wallis test	H	p	Friedman test	χ^2	p	Kruskal-Wallis test	H	p
DYR	25.2	1×10^{-4}	DYR	25.6	2×10^{-5}	All materials	35.38	$<10^{-5}$
DEF	25.7	1×10^{-4}	DEF	20.8	9×10^{-4}			
ARI	28.9	2×10^{-5}	ARI	29.3	2×10^{-5}			
SPE	17.8	3×10^{-3}	SPE	27.1	5×10^{-5}			

marginal gaps was measured in cylindrical cavities filled with the four materials DYR, DEF, ARI, and SPE, and this was done in two ways. In the GAP-1 experiments, the excess filling material was removed immediately before measuring, and in the Gap-2 experiments the same set of specimens was measured after various periods in water. In the latter experiments, the grinding to remove the excess material, performed after 1 h water immersion, may cause grinding particles to be introduced in the gaps which later may inhibit subsequent closure of marginal gaps. This effect is especially apparent for the material DEF when comparing the GAP-1 results with those of GAP-2 (Table 2). As composite fillings are normally finished shortly after polymerization, the results from the Gap-II experiment might be more useful for a judgment of gap sizes at the surface of fillings performed in a clinical situation. The results from the experiment Gap-I might be useful for a judgment of gap sizes at a certain distance from the surface of the fillings at locations where grinding particles are not introduced.

In all cases (Table 2), extended water storage caused a significant reduction in the sizes of the marginal gaps, and with two of the materials, DYR and ARI, a very small or absence of marginal gaps was apparent after 180 days' water storage. This finding is supported in a study (16) of the marginal integrity of fillings in Class II cavities showing a better adaptation with Dyract AP than with Spectrum TPH. A comparison of the gap sizes obtained with DYR with that previously obtained (17) with the first compomer introduced, Dyract, showed an apparent similarity.

The experiments were performed without the use of a bonding agent. It seems likely, indeed has been shown for the present materials in an unpublished study, that use of bonding agents would have prevented or reduced the size of the gaps. In the latter case, gaps will be closed due to water storage at an earlier time period than those presented in Table 2.

The results from the mechanical tests on specimens made of the four materials immersed for various periods in water revealed (Table 2) flexural strengths for each material that were not significantly different in the range 1–180 days. Over time, resin composites will obtain an increasing flexural strength owing to the continuous although declining conversion of vinyl groups. The water uptake will tend to reduce the flexural strength, and, judging by the present experiments, it seems that these two effects have neutralized each other. The differences between the results in Table 2 and those with Dyract performed by others (9) may be explained by differences in methods such as different cross-head speeds and dry (9) instead of water-immersed specimens during testing.

For all materials except DYR, the flexural moduli estimations (Table 2) showed an increasing stiffness by time >1 h, which might be explained by the continuous although declining conversion of vinyl groups. Apparently, the water uptake might not have the same detrimental effect on the flexural modulus as on the flexural strength. Clinical observations (18–20) on the performance of a compomer used in molar cavities has not revealed detrimental results and is in agreement with those given in Table 2 showing that DYR, even after prolonged water storage, has mechanical properties which are not far from that of a traditional composite such as SPE.

It can be concluded that the hypothesis that "resinous materials with relatively large water absorption will show reduced gap sizes around fillings in cavities as well as reduced mechanical strength" may only partly be supported by the experiments. The gap sizes were reduced but the mechanical properties were either unaltered or increased in the range of 1–180 days water immersion.

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Received for publication 24 January 2002
Accepted 12 March 2002