Water sorption and water-induced molecular mobility in dental composite resins

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Water sorption in two resin composites, Kulzer's Solitaire (S) and SDI's Wave (W), and in a polyacid-modified composite resin, 3M's F2000 (compomer F), was investigated by means of equilibrium sorption isotherms (ESI) and of dynamic sorption (DS) measurements. Molecular mobility in these materials was studied by means of dielectric relaxation spectroscopy (DRS) and of thermally stimulated depolarization currents (TSDC) measurements. The results of ESI measurements show that at equilibrium, water is molecularly distributed in the materials and the effects of hydrophilic sites and clustering are negligible. Hysteresis effects in sorption–desorption cycles are larger in the resin composites than in the compomer. Equilibrium water uptakes in both ESI and DS conditions are rather low, in the range 1–2%. Diffusion coefficients of water are about 1×10^{-8} cm^2/s in the resin composites and by a factor of about 2 smaller in the compomer. Molecular mobility increases with hydration, as suggested by preliminary DRS and TSDC measurements. Detailed dielectric measurements may give important information for understanding, at the molecular level, water-induced degradation in dental materials.

1. Introduction

Restorative dental materials may absorb significant amounts of water when exposed to aqueous environments [1–3]. Generally, water uptake has been regarded as detrimental [4]. Water sorption may cause degradation of the physical properties and decrease the life of dental materials by such diverse effects as dissolution, hydrolysis, expansion, plasticization, microcrack formation and fatigue [5–7]. In fiber-reinforced thermoplastic composites for dentistry, scanning electron microscopy and mechanical tests were employed to study the hydrolytic stability of the interface and the effects of moisture on flexural properties [8, 9]. In polyacid-modified resin composites (also called compomers) it has been suggested that a second, delayed curing mechanism extending over months occurs, as water is absorbed by the material in the course of time [2].

Water sorption studies, mostly by gravimetric analysis of samples immersed in water, show significant differences among different classes of dental materials [1–3]. The uptake of water into resin composites is largely controled by diffusion, whereas the presence of hydrophilic sites, interfaces and microvoids, as well as hydrolysis, relaxation and plasticization effects [10], may become significant.

Here, we report the results of our studies of water sorption and water-induced molecular mobility (plasticization by water) in two resin composites and one polyacid-modified composite resin (compomer). The results are discussed and critically compared with those obtained by other investigators with dental materials, including also glass-ionomer cements [1, 2], despite their different nature.

In the past, less attention has been paid to water sorption into dental materials from the vapor phase. Here, we employ both equilibrium water sorption isotherm (ESI) measurements from the vapor phase at controled relative humidities [10] and dynamic water sorption (DS) measurements by immersion of the samples into water [1–3, 10]. They provide information on water uptake under the different experimental conditions, binding to hydrophilic sites, formation of water clusters, hysteresis effects and diffusion coefficients.

Degradation in polymers and in polymer-based materials is closely related to and increases with molecular mobility [11]. Thus, the investigation of effects of water sorption into dental materials on molecular mobility (plasticization effects) may significantly contribute to improving our understanding of the

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mechanisms of degradation and failure of these materials. Here, we report the results of preliminary studies of molecular mobility in the resin composites and the compomer under investigation at two levels of relative humidity by two dielectric techniques.

2. Materials and methods

Three materials were investigated in this study.

(1) SDI's Wave (W) (Southern Dental Industries, Bayswater, Australia, batch no. 70623, shade A3), which is a light cured microhybrid composite of low viscosity (flowable) and contains 50 vol % inorganic fillers (mostly strontium glass) in a resin matrix of methacrylic esters.

(2) Kulzer's Solitaire (S) (Haraeus Kulzer, Wehrheim GmbH, batch no. 24, shade A30), a light cured hybrid composite of high viscosity (condensable), which contains 55 vol % (65 wt %) polyglobular and highly porous glasses (SiO_2 and Ba–Al–B–F–Si) in a resin matrix of Bis–GA, HPMA and ETMA.

(3) 3M's F2000 (F) (3M Dental Products, St Paul, USA, batch no. 0827, shade A3), a light cured polyacid-modified resin composite (compomer), which contains 55 vol % inorganic fillers in a resin matrix of carboxyl dimethacrylate (CDMA) and glycerol dimethacrylate (GDMA).

Several samples were made of each material in order to keep only those with the best surface and side finish and with minimal air entrapment (visually unnoticed). The samples, 12 mm in diameter and 2 mm in thickness, were made by injecting or condensing the material into the respective holes of a plastic mold on top of a glass slide and a celluloid sheet. The samples polymerized under a second celluloid sheet and a glass slide with a Visilux 2 curing light (3M), were tested at the tip to have an intensity of 450 mW/cm^2. The samples were kept in tightly closed black boxes until their testing.

ESIs were measured at 25 $\pm$ 1 °C. Drying in vacuum (5×10^{-2} Torr) at 112 °C for 48 h prior to ESI measurements was adopted as the method for the determination of dry weights. After drying, the samples were allowed to equilibrate to constant weight in various dessicators with increasing relative humidity (rh), which was monitored between 8 and 100% using different saturated salt solutions [12]. The time required to reach equilibrium (sample weight change less than 10^{-4}g) increased with increasing rh from a few days to two weeks. After hydration reached full saturation ($rh = 100\%$), the samples were dehydrated by using the reverse procedure. An analytical balance (A200S, Sartorius, accuracy 10^{-4}g) was employed for weighing the samples.

Three measures of water content were used throughout this work. The initial water content (inherent water content [2]) h_i is the water content after preparation of the samples and prior to drying, and is defined as (initial weight before drying − dry weight)/dry weight. The water content h at each relative humidity is defined as (weight after saturation at the given rh − dry weight)/dry weight. Its maximum value at $rh = 100\%$ is designated as h_m. Finally, the ambient water content h_a was obtained by allowing the samples to equilibrate at ambient conditions after completing the hydration–dehydration cycle, as (weight at ambient conditions − dry weight)/dry weight. The experimental error in water content was estimated to about 0.05% (absolute values).

DS measurements were carried out at 28 $\pm$ 1 °C, by immersion of dry samples into deionized water in 50-ml Pyrex glasses. At appropriate intervals, the samples were removed from water, blotted dry, and weighed. Similar to the water content h in ESI measurements, the water uptake $(\Delta m)_t$ was calculated by

$$(\Delta m)_t = \frac{M_t - M_d}{M_d} \times 100 \qquad (1)$$

where M_t is the mass of the wet specimen at time t and M_d the mass of the dry specimen. The latter was determined by drying in vacuum at 80 °C (instead of 112 °C in ESI experiments, compare the section results) for 48 h. Following common praxis, the maximum water uptake at saturation will be designated as EWC (equilibrium water content).

Dielectric relaxation spectroscopy (DRS) and thermally stimulated depolarization currents (TSDC) techniques were employed to study molecular mobility of the samples at two levels of rh and corresponding water content h: ambient rh and $rh = 100\%$. In DRS measurements the complex dielectric permittivity $\varepsilon^* = \varepsilon' - i\varepsilon'$ was determined as a function of frequency (10^{-2}–10^6 Hz) at 25 $\pm$ 1 °C. A Schlumberger frequency response analyzer (FRA 1260) supplemented by a buffer amplifier of variable gain (Chelsea Dielectric Interface) and an LCR Meter (HP 4284A) were employed. A two-terminal parallel-plate capacitor (nickel-coated stainless steel electrodes) dielectric cell (Ando type SE-70 electrode) was used combined with an Ando type TO-19 thermostatic oven.

The method of TSDC consists of measuring the thermally activated release of stored dielectric polarization. It corresponds to measuring dielectric losses ε'' against temperature at constant low frequencies of 10^{-2}–10^{-4} Hz [13]. By this method, the sample is inserted between the plates of a capacitor, made of brass, polarized by the application of a dc field and cooled down to liquid nitrogen temperature to freeze in the polarization. The samples then short-circuited and reheated at a constant rate. A discharge current is generated, which is measured as a function of temperature with a sensitive electrometer (Keithley 610C). The TSDC spectrum thus obtained consists of several peaks whose shape and location are characteristic of the relaxation mechanisms of the sample. TSDC is characterized by high sensitivity and high resolving power. A homemade experimental apparatus for TSDC measurements in the temperature range from − 180 to 30 °C was employed. For details on the TSDC method, the apparatus used and the methods of data analysis we refer to Pissis *et al*. [13].

3. Results

The values of initial (inherent) water contents h_i determined prior to ESI measurements are listed in Table I.

Fig. 1 shows the results of ESI measurements, water content h *versus* rh for the three samples under

TABLE I Results of water ESI and DS measurements. h_i is the initial (inherent) water content, h_a the ambient water content, M_v the weight fraction of volatiles, h_m the maximum water content at 100% *rh* in sorption from the vapor, EWC the saturation water uptake in immersion, and D the diffusion coefficient of water (see Section 2. for definitions)

	ESI				DS		
Sample	h_i	M_v	h_a	h_m	h_i	EWC	$D[10^{-8}\,\mathrm{cm^2/s}]$
S	1.86%	0.85%	1.00%	2.00%	1.40%	2.03%	1.1
W	1.46%	0.84%	0.61%	1.12%	1.11%	1.01%	1.0
F	0.97%	—	1.06%	1.73%	0.75%	1.45%	0.5

investigation. h increases practically linearly with *rh* in the whole range of measurements. The values of h at 100% *rh* (h_m) are 2.00% for *S*, 1.12% for *W* and 1.73% for *F* (Table I). The values of h at decreasing *rh* are higher than those at increasing *rh*, i.e. hysteresis is observed. Hysteresis increases in the order $S < W < F$.

After completing the hydration–dehydration cycle, the samples were allowed to equilibrate at ambient conditions for several weeks. Their ambient water contents h_a were then determined (see the previous section) to 1.00% for *S*, 0.61% for *W* and 1.06% for *F* (Table I). These values are to compare with the initial (inherent) h_i values. The h_a values are significantly lower than h_i for the synthetic resins *S* and *W* and, within experimental errors, similar for the compomer *F*. The difference for the samples *S* and *W* may be attributed to the presence of volatiles, their weight fraction m_v being defined by (initial weight − final weight)/final weight and determined to 0.85% for *S* and 0.84% for *W* (Table I). However, post-curing effects and chemical reactions leading to permanent changes during drying cannot be excluded [10]. For that reason the temperature of drying was reduced to 80 °C in DS experiments.

Fig. 2 shows the results of DS measurements, water uptake $M(t)$ versus time t over a period of more than three weeks. The time interval between successive measurements was varied from a few minutes at the early stages of immersion to several hours at the final stages. The results in Fig. 2 are presented as lines connecting the experimental points. Distinct differences are observed in the behavior of the samples in Fig. 2. The water uptake is more rapid in *S* and *W*, as compared to *F*. So, the level of 80% of final (saturation) water uptake is reached for *S* and *W* after 56 h, for *F* only after 190 h. For *S* there is a decrease of water content with time between about 12.000 and 25.000 min before it stabilizes. The final (saturation) water contents (EWC values) are 2.03% for *S*, 1.01% for *W* and 1.45% for *F* (Table I).

DS measurements allow to study the kinetics of water sorption. Assuming Fickian behavior with a constant (i.e. independent of h) diffusion coefficient D, the following equation holds at relatively small values of time t, corresponding to $(\Delta m)_t/(\Delta m)_\infty > 0.6$ [14, 15]

$$\frac{(\Delta m)_t}{(\Delta m)_\infty} = \frac{4}{\sqrt{\pi}}\sqrt{\frac{tD}{d^2}} \qquad (2)$$

where $(\Delta m)_t$ is the water uptake at time t, $(\Delta m)_\infty$ the corresponding limiting value at equilibrium and d the thickness of the sample, presumed constant over the whole sorption process.

Fig. 3 shows the typical for data analysis of DS experiments plots of normalized water uptake

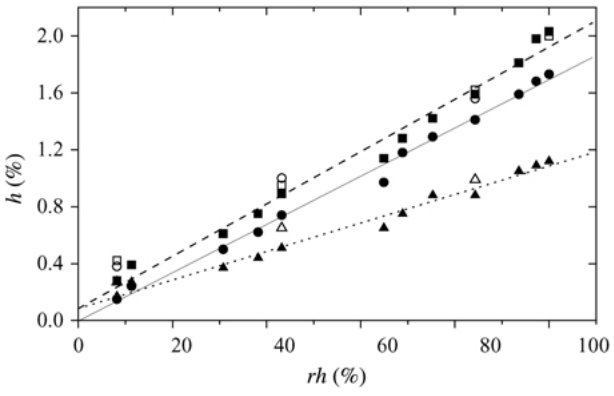

Figure 1 ESI at 25 °C of the samples *S* (squares), *W* (triangles) and *F* (circles) during absorption (full symbols) and desorption (open symbols). The lines are linear fits (Henry's law) to the absorption data.

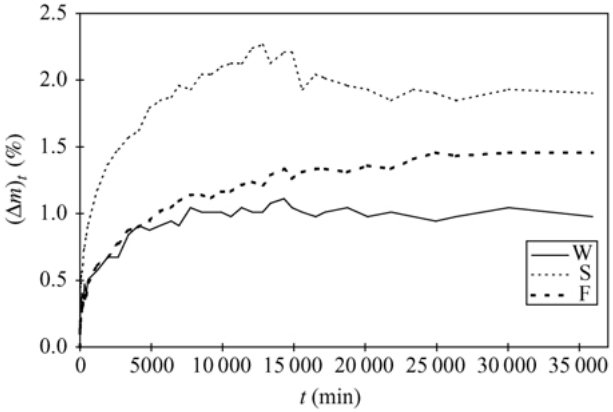

Figure 2 Water uptake $(\Delta m)_t$ versus time t in DS measurements at 28 °C for the samples indicated on the plot.

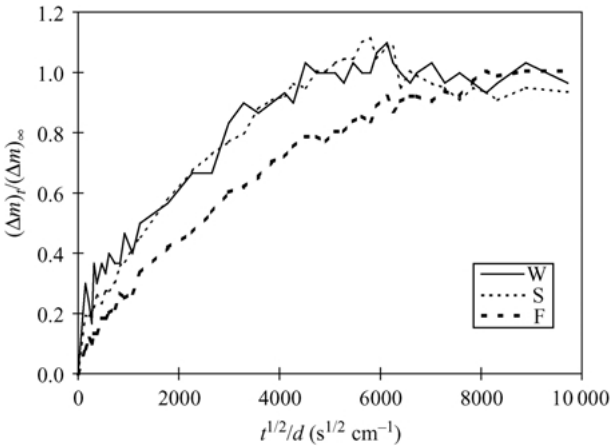

Figure 3 Normalized form of the plot shown in Fig. 2. d is the thickness of the sample.

$(\Delta m)_t/(\Delta m)_\infty$ vs. $\sqrt{t}/d$ for the samples under investigation. The initial parts of the plots can be approximated by straight lines. Their slopes give D, estimated here to $D = 1.1 \times 10^{-8}\,\mathrm{cm^2/s}$ for S, $1.0 \times 10^{-8}\,\mathrm{cm^2/s}$ for W and $0.5 \times 10^{-8}\,\mathrm{cm^2/s}$ for F (Table I). The error in D is estimated to about 10%.

Figs. 4 and 5 show the real (ε') and imaginary part (ε''), respectively, of the complex dielectric permittivity $\varepsilon^* = \varepsilon' - i\varepsilon''$, where $i = \sqrt{-1}$ [16], for the samples under investigation at 25 °C at ambient *rh* and at $rh = 100\%$. An overall increase of molecular mobility with hydration is observed, as indicated by the increase in ε' and ε''. The high values of ε' and ε'' at low frequencies, in particular in the hydrated samples, are due to ionic conductivity effects [17, 18]. A loss peak (maximum in $\varepsilon''(f)$) is observed for the sample *S* in the hydrated state.

An example of TSDC measurements is shown in Fig. 6: TSDC thermograms for sample *F* at ambient *rh* and at $rh = 100\%$. With respect to DRS measurements in Figs. 4 and 5, TSDC corresponds to measuring dielectric losses ε'' as a function of temperature at constant frequency in the range 10^{-2}–10^{-4} Hz [13]. The thermogram at ambient *rh* shows a broad and weak peak at about -100 °C and a second significantly stronger and more complex one at about 0 °C. In the thermogram of the hydrated sample, both peaks increase in magnitude and shift to lower temperatures, i.e. the corresponding relaxation mechanisms become faster and their relaxation strength increases. A third peak at temperatures lower than -170 °C is indicated by the shape of the thermogram. The main features of the TSDC thermograms in samples *S* and *W* are similar to those shown in Fig. 6 for *F*, with quantitative differences as to the temperature position and magnitude of the peaks.

4. Discussion

The linear dependence of *h* on *rh* in Fig. 1 (Henry's law) is characteristic for several glassy polymers [19, 20]. It indicates that water is mainly molecularly distributed in the sample. This point should be further followed in future work by additional tests, e.g. by measuring diffusion coefficients both during sorption and desorption and taking their ratio as an indication of the concentration dependence of the diffusion coefficient [21]. The absence of an initial steep increase of *h* followed by a plateau in the curves in Fig. 1, in particular, indicates that sorption at hydrophilic sites, present in the samples (e.g. carboxyl groups in the compomer *F*) is negligible [10, 16, 19]. This is probably due to the hydrophilic sites being involved in network formation and/or not accessible to water [11]. The absence of an upward curvature of the $h(rh)$ dependence at high *rh* values indicates that formation of water clusters around initially molecularly distributed water molecules is negligible [16, 19]. Both effects, sorption at hydrophilic sites and cluster formation, were found to be rather weak in epoxy resin systems [10, 19].

Hysteresis effects are considered as detrimental in materials science [19, 20]. Interestingly, they are smaller in the compomer (*F*) than in the resin composites (*S* and *W*). Also the concentration of volatiles was similar in the two resin composites (about 0.85%) and negligible in the compomer. These volatiles are probably unreacted monomers that dissolve in a wet environment or degradation products (hydrolization or oxidation of double bonds) [22]. Common to all materials studied here is that they contain fluoride, part of which is released by hydrolysis [23]. So fluoride release may be related to the observed hysteresis effects and to the differences observed in the behavior of the compomer on the one hand and the composites on the other hand with respect to hysteresis and to volatiles. These differences may, however, simply reflect the fact that water behaves very differently in a compomer, as compared to the composites, due to secondary glass–ionomer type reactions occuring in the former [2, 23].

The maximum equilibrium values of water content in ESI experiments h_m in Table I are in the range of values typical for glassy polymers [19, 20]. In epoxy resins, values of 2.0–2.3% have been reported [10]. It is interesting to compare these values with the EWC values measured in DS experiments (Table I). It should be stressed, however, that from the methodological point of view, the maximum water contents may be expected to be different in the two sets of experiments, although water activity is the same in the saturated water vapor and in liquid water [24]. In *F* and *W* smaller values have

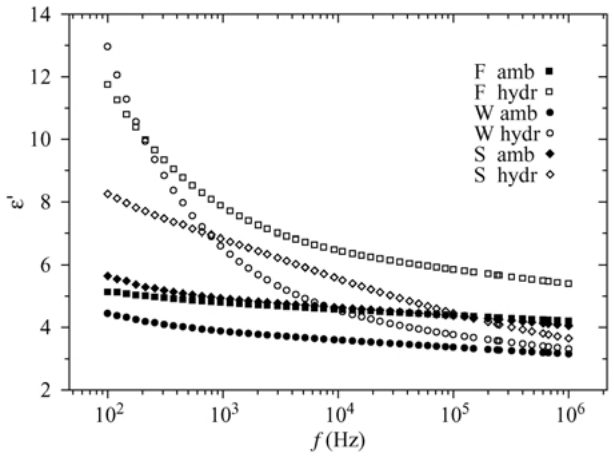

Figure 4 Frequency dependence of the real part of dielectric permittivity ε' for the samples indicated on the plot at two levels of *rh*: ambient *rh* (amb) and *rh* 100% (hydr).

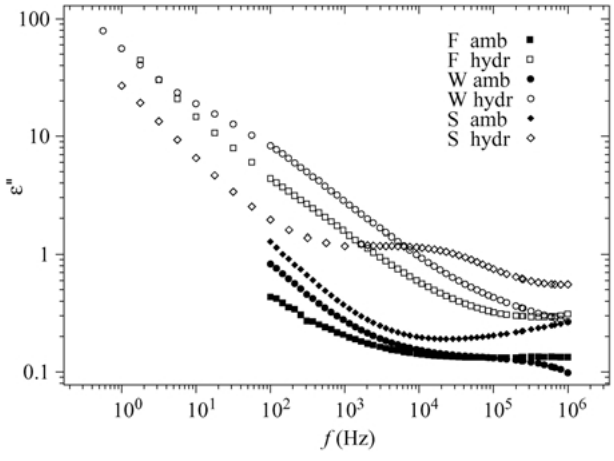

Figure 5 Frequency dependence of the imaginary part of dielectric permittivity ε'' for the samples indicated on the plot at two levels of *rh*: ambient *rh* (amb) and *rh* 100% (hydr).

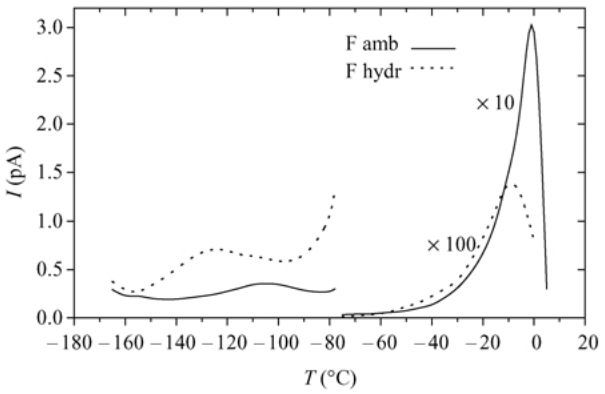

Figure 6 TSDC thermograms for sample *F* at two levels of *rh*: ambient *rh* (amb) and *rh* 100% (hydr). Please note the change of scale at −75 °C.

been measured in DS, in *S* the values are comparable to each other. Lower values may be expected in DS experiments, because of the lower temperature of drying, reflected in the lower initial water contents h_i (Table I). However, it cannot be excluded at this stage that the differences between values obtained in ESI and DS experiments for the samples *F* and *W* are, at least partly, due to the presence of water solubles, which are removed from the samples in DS experiments. This point should be further followed in future experiments. Nicholson [1] determined the content of water solubles in resin-modified glass–ionomers by two absorption cycles in DS experiments. Braden *et al*. [25] studied diffusion of water in several dental composite resins. For two of them the whole sorption – desorption process was repeated an additional two times and reversibility was observed only for one of them, whereas for the second the diffusion coefficient *D* increased in the second and in the third cycle. Iwami *et al*. [3] adjusted the final weight changes of several dental materials by water solubility. In epoxy resin systems, the final water uptakes were slightly larger in DS than in ESI experiments [10]. Braden and Clarke [26] reported that equilibrium uptake and solubility data in dental microfine composite resins obtained in DS experiments are best rationalized if expressed as volume/volume rather than weight/weight.

In both ESI and DS experiments, the final water uptakes in the two resin composites are by a factor of about 2 larger in *S* than in *W* (Table I). It is interesting to note that the two composites behave in many respects similar to each other (Table I). Also their filler contents are approximately the same, 50 vol % in *W* and 55 vol % in *S*. A possible explanation for the different water uptakes is a different quality of interfaces [8, 9], a point which can be further followed in future by studying the properties of the interfacial Maxwell–Wagner–Sillars polarization [18] by DRS and TSDC experiments.

The values of diffusion coefficient *D* in Table I are comparable to each other in the two resin composites *S* and *W* and by a factor of about 2 smaller in the compomer. This simply quantifies the features observed in Fig. 2, where *S* and *W* reach equilibrium much earlier than *F*. This is worth mentioning from the point of view of comparing different dental materials with each other, as the results may not allow calculation of diffusion coefficients [2, 3]. In the materials under investigation, here, the early stages of water uptake could be approximated by Fickian behavior. Braden *et al*. [25] and Braden and Clarke [26] reported Fickian behavior for water uptake by conventional composite and microfine composite dental materials, respectively. In general, the diffusion of water (and of other diluents) in polymers has been found to be more complex than Fickian [19, 27]. The diffusion coefficient was found, in general, to depend on diluent concentration [27]. Free volume concepts have been applied to describe the concentration dependence of *D* both above and below the glass transition temperature [28]. In composite systems, like those under investigation here, more refined models have to be employed. The results of DS measurements in epoxy resins at several temperatures were satisfactorily explained by a two-phase model, the two phases being characterized by different water uptakes and kinetics of sorption [10]. By measuring diffusion coefficients of water *D* in several dental composites during sorption and desorption, Braden *et al*. [25] obtained significantly higher values for the desorption process and explained that in terms of decrease of *D* with increasing water concentration.

The absolute values of *D* in Table I are rather high for glassy polymers [19, 20, 27], in agreement with the absence of hydrophilic effects. In epoxy resin systems values of *D* were determined by about one order of magnitude lower than in the samples under investigation here [10]. The rather high values of *D* may be related to the composite character of these materials and diffusion along the interfaces. This point should be further followed in future, also in connection to morphological characterization of the samples and to measurements at several temperatures. Also the presence of porosity in the composites may affect the diffusion coefficient, similar to the observation of increased water uptake in dental composites incorporating hydroxyapatite filler [29]. On the other hand, the compomer is known to possess hydrophilic groups (free carboxyl groups), so that water retention to these hydrophilic sites may explain the smaller (by a factor of 2) diffusion coefficient in the compomer *F* than in the composites *S* and *W*.

It is interesting to compare the values by DS experiments in Table I with the results of DS measurements in other dental materials. In some cases, this can be done only on a qualitative basis, as diffusion coefficients have not always been calculated. Iwami *et al*. [3] observed that water uptake (not normalized to dry weights) in several dental materials increases in the order resin composites < compomers < glass–ionomers. Small *et al*. [2] investigated, in comparison to each other, a resin composite, a compomer, a conventional glass–ionomer and a resin-modified glass–ionomer. They observed that the first two materials absorb more slowly and less water than the last two. Their plots show that the compomer absorb more water but less slowly than the resin composite. Nicholson [1] quantified the results of DS experiments in resin-modified glass–ionomers and studied the effects of curing time, temperature of measurements and sodium chloride concentration in water. Measured *D* values were by about one order of magnitude larger than in the samples under investigation here, in qualitative agreement with results of compara-

tive studies showing that water uptake in resin-modified glass–ionomers is much faster than in resin composites and in compomers [2, 3].

Dielectric measurements have been widely employed to study polymer–water interactions in synthetic polymers and in biopolymers [11, 13, 16, 17, 19]. From the technological point of view the main interest of these studies is to investigate and quantify degradation effects in synthetic polymers [11, 19], whereas in biological materials they may provide links between biological function and physical properties [16, 17].

The results of both DRS and TSDC measurements (Figs. 4–6) show that, in general, molecular mobility increases in the hydrated samples. This is reflected in both the magnitude of ε' and ε'' (Figs. 4 and 5) and of the loss peaks in TSDC (Fig. 6) and the shift of the TSDC peaks to lower temperatures. Such plasticization effects have been in general related to degradation in polymers [30]. Their detailed investigation in dental materials may provide the necessary information to understand water-induced degradation in this class of materials, which is necessary for designing materials with improved performance.

The results of TSDC measurements should be compared with those of similar detailed measurements in epoxy resin systems at several levels of hydration [11]. The TSDC peak at about $-100\,^{\circ}\mathrm{C}$ shifting to lower temperatures in the hydrated samples (Fig. 6) has been identified in epoxy resins as the secondary, local β relaxation attributed to the motion of the hydroxypropylether group [11]. The high-temperature TSDC peak at about $0\,^{\circ}\mathrm{C}$ is probably multiple, consisting of several contributions. The large magnitude of the peak suggests that conductivity and space charge effects, such as interfacial Maxwell–Wagner–Sillars (MWS) polarization [18], make a significant contribution to that peak. These effects increase significantly in the hydrated samples (Fig. 6), in agreement with the increase of conductivity effects in DRS measurements, reflected in the high values of ε' and ε'' at low frequencies in Figs. 4 and 5. The detailed investigation of MWS polarization by TSDC at several *rh* levels and by DRS at several temperatures should provide information on interfaces, essential for understanding degradation effects [8, 9]. It is interesting to note, with respect to the ESI and DS measurements, that the TSDC thermograms suggest a quantitatively different behavior of the compomer *F* on the one hand and of the resin composites *S* and *W* on the other hand. This point should be further followed by TSDC and DRS measurements.

5. Conclusions

1. ESI measurements indicate that in both the resin composites (S and W) and in the compomer (F) water sorption to hydrophilic sites and water clustering are negligible. The resin composites are characterized by larger hysteresis effects in sorption–desorption cycles and (probably) by larger weight fraction of volatiles, as compared to the compomer.

2. DS measurements show, in agreement with the results of ESI measurements, that the equilibrium water uptakes of the materials under investigation are low, in the range of 1–2%. Water uptake is faster in the resin composites than in the compomer. So, the former absorb 80% of the equilibrium water uptake after 56 h of immersion (at $28\,^{\circ}\mathrm{C}$), whereas the latter needs 190 h for that. In the early stages of water uptake diffusion is Fickian, the diffusion coefficient being about $1 \times 10^{-8}\,\mathrm{cm^2/s}$ for the resin composites and by a factor of 2 smaller for the compomer.

3. Preliminary dielectric measurements by means of DRS and of TSDC measurements at two levels of relative humidity show that the overall molecular mobility increases in the hydrated samples. These results should be quantified in future work to provide a basis for discussing water-induced degradation in dental materials in terms of mechanisms of molecular mobility. Dielectric measurements on samples exposed to water for long periods of time may also provide information on a hypothesized secondary, delayed water-induced curing mechanism in the compomers [2].

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