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Impedance changes during setting of amorphous calcium phosphate composites

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ABSTRACT

Objectives. To investigate the electrical properties of experimental light-curable composite materials based on amorphous calcium phosphate (ACP) with the admixture of silanized barium glass and silica fillers.

Methods. Short-term setting was investigated by impedance measurements at a frequency of 1 kHz, while for the long-term setting the impedance spectra were measured consecutively over a frequency range of 0.05 Hz to 1 MHz for 24 h. The analysis of electrical resistivity changes during curing allowed the extraction of relevant kinetic parameters. The impedance results were correlated to the degree of conversion assessed by Raman spectroscopy, water content determined by gravimetry, light transmittance measured by CCD spectrometer and microstructural features observed by scanning electron microscopy.

Results. ACP-based composites have shown higher immediate degree of conversion and less post-cure polymerization than the control composites, but lower polymerization rate. The polymerization rate assessed by impedance measurements correlated well with the light transmittance. The differences in the electrical conductivity values observed among the materials were correlated to the amount of water introduced into composites by the ACP filler. High correlation was found between the degree of conversion and electrical resistivity. Equivalent circuit modeling revealed two electrical contributions for the ACP-based composites and a single contribution for the control composites.

Significance. The impedance spectroscopy has proven a valuable method for gaining insight into various features of ACP-based composites. Better understanding of the properties of ACP-based composites should further the development of these promising bioactive materials.

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1. Introduction

Incorporation of amorphous calcium phosphate (ACP) fillers in methacrylate resins results in bioactive, ion-releasing composite materials. Their bioactivity is due to the thermodynamic instability of ACP, which in aqueous media spontaneously converts into hydroxyapatite and concomitantly releases calcium and phosphate ions [1]. When embedded in the appropriate methacrylate resin, ACP provides a sustained release of calcium and phosphate ions in concentrations sufficient for hydroxyapatite precipitation in dental hard tissues adjacent to the restoration [2,3]. The remineralizing ability of ACP-based composites has been demonstrated in vitro [3], in situ [4] and in animal model in vivo [5]. These findings indicate the potential of ACP-based composites for preventing secondary caries, which is considered the main cause for failure of contemporary composite restorations [6].

The major drawbacks of ACP-based composites are poor mechanical properties and high shrinkage stress in comparison to commercial composites [7]. To address these issues, ACP-based formulations were introduced with conventional silanized fillers [8,9]. Since the ACP-based composites attain sufficient ion release with 40 wt% of ACP [7], their filler load can be increased by the addition of inert fillers similar to those contained in conventional composites. In this way, a “hybrid” material is composed, which consists of a bioactive part (ACP fillers) and a reinforcing part (inert fillers). When exposed to water, ACP undergoes a dissolution-reprecipitation process converting into hydroxyapatite and releasing ions [10], while the inert fillers remain bonded to the methacrylate network and reinforce the restoration. The admixture of inert fillers (barium glass and silica) has been shown to improve the mechanical properties, without compromising the ion release [8,9]. By fine-tuning the composition of such “hybrid” ACP-based composites, their flexural strength may be enhanced to match the values of commercial resin-modified glass ionomer cements [11] and possibly even commercial composites [12].

Various properties of ACP-based composites have been investigated, but there are no studies examining their electrical properties by means of impedance spectroscopy (IS). Generally, IS is rarely used in investigations of dental

composites—only several papers are available, dealing with composite degradation [13], water sorption [14,15], microleakage [16] and polymerization kinetics [17–19]. In particular, Steinhaus et al. [19] have described the kinetics of composite polymerization from the changes of the electrical resistivity as a function of time, $\rho(t)$, using the relation: $\rho(t) = \rho_{\text{init}} + B \times \Delta t$; where ρ_{init} is the initial resistivity measured before the curing and Δt is the time period since the start of curing. In the linear range of the resistivity vs. time curve, the resistivity is assumed to be proportional to the rheological viscosity and molecular mass of the growing polymeric chains. Therefore, slope B can be used as a measure of reaction rate during an early stage of polymerization, while polymeric chains grow linearly. Another kinetic parameter in the model is the reaction time constant, τ , contained in the relation: $\Delta\rho(t) = \Delta\rho_{\text{max}} \times e^{-(\Delta t/\tau)}$ where $\Delta\rho(t)$ is the change of resistivity as a function of time and $\Delta\rho_{\text{max}}$ is the total change of resistivity during curing.

All above-mentioned IS studies are performed at a fixed frequency and there are no reports of the electrical properties of dental composites measured in a wide frequency range. This study employs IS to investigate the structural changes occurring during the short-term setting (during light-curing) and long-term setting (up to 24 h post-cure) of five ACP-based and two control composites. The short-term setting is analyzed in terms of impedance (electrical conductivity) changes at a fixed frequency as a function of time, while for the long-term setting the impedance spectra were measured consecutively over a frequency range of 0.05 Hz to 1 MHz for 24 h. Furthermore, the results obtained by IS are correlated to the degree of conversion, water content, light transmittance and microstructural features.

2. Materials and methods

2.1. Composite materials

The composition of composite materials is given in Table 1. The synthesis of zirconia-hybridized ACP (Zr-ACP) fillers followed the procedure by Skrtic et al. [20]. The resin was blended with fillers in lightproof containers using a dual asymmet-

Table 1 – Composition of the experimental composite materials.

Material	Filler (wt%)	Resin (wt%)	Filler load (vol%)
ACP-based materials	ACP40	40% Zr-ACP	60 wt%
	ACP40-Ba10	40% Zr-ACP, 10% Ba-fillers	50 wt%
	ACP40-Si10	40% Zr-ACP, 10% Si-fillers	50 wt%
	ACP40-Ba5Si5	40% Zr-ACP, 5% Ba-fillers, 5% Si-fillers	50 wt%
	ACP40-Ba9Si1	40% Zr-ACP, 9% Ba-fillers, 1% Si-fillers	50 wt%
control materials	Ba40	40% Ba-fillers	60 wt%
	Ba40Si10	40% Ba-fillers, 10% Si-fillers	50 wt%

Barium-fillers (Ba): SiO₂ 55.0%, BaO 25.0%, B₂O₃ 10.0%, Al₂O₃ 10.0%, F 2.00%, particle size (d50/d99 [μm]) 0.77/2.28, silanization 6 wt%, product name/manufacturer: GM39923/Schott, Germany.

Silica-fillers (Si): SiO₂ ≥ 99.8, primary particle size: 12 nm, silanization 4–6 wt%, product name/manufacturer: Aerosil DT/Evonik Degussa, Germany.

EBPADMA: ethoxylated bisphenol A dimethacrylate, Esstech, PA, USA; TEGDMA: tri-ethylene glycol dimethacrylate, Esstech; HEMA: 2-hydroxyethyl methacrylate, Esstech; CQ: camphorquinone, Aldrich, WI, USA; 4E: ethyl-4-(dimethylamino) benzoate, Aldrich.

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