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Dielectric analysis of depth dependent curing behavior of dental resin composites[⋆]

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ABSTRACT

Objectives. The aim of this study is to investigate depth dependent changes of polymerization process and kinetics of visible light-curing (VLC) dental composites in real-time. The measured quantity – "ion viscosity" determined by dielectric analysis (DEA) – provides the depth dependent reaction rate which is correlated to the light intensity available in the corresponding depths derived from light transmission measurements.

Methods. The ion viscosity curves of two composites (VOCO Arabesk Top and Grandio) were determined during irradiation of 40 s with a light-curing unit (LCU) in specimen depths of 0.5/0.75/1.0/1.25/1.5/1.75 and 2.0 mm using a dielectric cure analyzer (NETZSCH DEA 231 with Mini IDEX sensors). The thickness dependent light transmission was measured by irradiation composite specimens of various thicknesses on top of a radiometer setup.

Results. The shape of the ion viscosity curves depends strongly on the specimen thickness above the sensor. All curves exhibit a range of linear time dependency of the ion viscosity after a certain initiation time. The determined initiation times, the slopes of the linear part of the curves, and the ion viscosities at the end of the irradiation differ significantly with depth within the specimen. The slopes of the ion viscosity curves as well as the light intensity values decrease with depth and fit to the Lambert–Beer law. The corresponding attenuation coefficients are determined for Arabesk Top OA2 to 1.39 mm^{-1} and 1.48 mm^{-1}, respectively, and for Grandio OA2 with 1.17 and 1.39 mm^{-1}, respectively. For thicknesses exceeding 1.5 mm a change in polymerization behavior is observed as the ion viscosity increases subsequent to the linear range indicating some kind of reaction acceleration.

Significance. The two VLC composites and different specimen thicknesses discriminate significantly in their ion viscosity evolution allowing for a precise characterization of the curing process even with respect to the polymerization mechanism.

[⋆] This paper is dedicated to Prof. H.-H. Kausch's 80th birthday.

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1. Introduction

An important issue in visible light-curing (VLC) composites development and research as well as in photo-chemistry in general is the question: How fast do composites cure, and how deep are they properly cured [1–6]? Since the development of VLC dental restoratives numerous studies have investigated the curing kinetics [7–11], the degree of cure (DC) [11–14] and the depth of cure (DoC) [14–21]. In order to get a better understanding of the curing process and its dependency on curing parameters such as curing time, light intensity, resin formulation, initiator system etc., various test methods have been applied in real time curing investigations. The most common techniques determine the curing-enthalpy via Differential Scanning Calorimetry (DSC) [7,22–24], the DC via Fourier Transformed Infrared Spectroscopy (FTIR) [3,4,12–14], the shrinkage strain via dilatometric devices (e.g. bonded-disk method) [9–12,25], the shrinkage stress via stress-analyzer [13,26,27] and light transmission via radiometer [6,8,28]. This time-dependent data is the base for kinetic modeling of the polymerization and curing processes.

Another method to monitor the curing behavior of resins is the dielectric analysis (DEA) which is widely used in composites manufacturing in the aircraft industry [29–34], but hardly established in dental science. In some cases it has been applied for the frequency dependent evaluation of dielectric properties in certain states of cure but without tracking the light-curing process itself in a suitable time-resolution [3,35,36].

In previous studies the authors have introduced a DEA setup allowing for analyzing the light-curing process of dental composites at high time resolution [28,37,38]. It has been shown that the ion viscosity is suitable to characterize the influence of resin compositions as well as curing conditions on the polymerization process of VLC composites. For many VLC composites the ion viscosity data shows a linear time dependency, and thus it is feasible to derive kinetic parameters [39].

Thus, in the present study the DEA is employed to investigate the curing behavior of a micro-hybrid and a nano-hybrid composite with respect to the depth within the samples. The hypothesis of this study is that in a VLC dental composite the polymerization and curing process changes significantly with increasing depth in the specimen and as a consequence the corresponding resin structure.

2. Materials and methods

For this study two VLC dental resin composites (Voco GmbH, Cuxhaven, Germany) with the initiator camphorquinone (CQ) were used varying significantly in composition (Table 1). (In the text to the composites is referred as Arabesk or Grandio.)

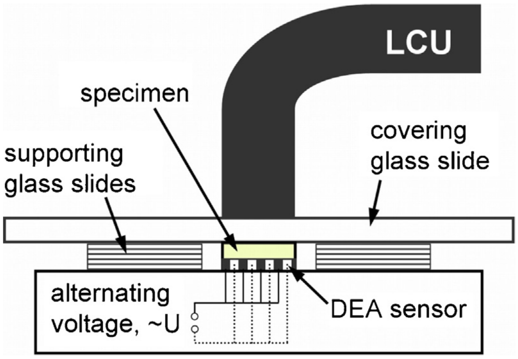

Fig. 1 – Experimental setup.

2.1. DEA curing measurements

The DEA-measurements were performed with a DiElectric cure Analyser (DEA 231/1 Epsilon, Netzsch-Gerätebau, Selb, Germany) using a measuring frequency of 1000 Hz. Fig. 1 shows the experimental setup using a sensor (Mini IDEX, Netzsch-Gerätebau) consisting of two comb-shaped interdigit electrodes (0.1 mm distance) on an insulating polyimide film. The sensor area of 5 mm × 7 mm fits well to the light guide tip cross-section (8 mm in diameter) of the light-curing unit (LCU) affirming a homogeneous irradiation of the investigated specimen sensor interface. According to the manufacturer the penetration depth of the alternating electric field of the DEA sensor corresponds to the distance of the electrodes [34]. Therefore, the curing process was mainly investigated in the bottom layer at the interface to the sensor area having a thickness of approximately 150 μm. This allowed for a stepwise thickness dependent curing monitoring of the polymerization and curing behavior within a sample.

The VLC composite specimen was put on the sensor surface and pressed with a microscope glass slide of 1 mm thickness to the desired layer thickness adjusted by the height of supporting glass slides. In this study both materials were investigated with sample thicknesses ranging from 0.5 mm up to 2.0 mm in (0.25 ± 0.05) mm steps at room temperature. All specimens were irradiated for 40 s with a LED LCU (Celalux, VOCO, Cuxhaven, Germany) having an irradiance of $880 \pm 50\,\text{mW/cm}^2$. The samples were constraint at the bottom by the sensor surface and at the top by the microscope slide but not laterally. For each depth, typically three samples ($n = 3$) were measured.

Table 1 – Composition of VLC composites according to the manufacturer's handling instructions.

Material	Arabesk Top OA2	Grandio OA2
Resin	22% (bis-GMA, UDMA, TEGDMA)	12% (bis-GMA, TEGDMA)
Inorganic filler particles	77%	87%
Additives	1%	1%