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Correlation of shear and dielectric ion viscosity of dental resins – Influence of composition, temperature and filler content

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ABSTRACT

Objective. Shear viscosity and ion viscosity of uncured visible light-curing (VLC) resins and resin based composites (RBC) are correlated with respect to the resin composition, temperature and filler content to check where Dielectric Analysis (DEA) investigations of VLC RBC generate similar results as viscosity measurements.

Methods. Mixtures of bisphenol A glycidyl methacrylate (Bis-GMA) and triethylene glycol dimethacrylate (TEGDMA) as well as the pure resins were investigated and compared with two commercial VLC dental resins and RBCs (VOCO, Arabesk Top and Grandio). Shear viscosity data was obtained using a Haake Mars III, Thermo Scientific. Ion viscosity measurements performed by a dielectric cure analyzer (DEA 231/1 Epsilon with Mini IDEX-Sensor, Netzsch-Gerätebau).

Results. Shear viscosity depends reciprocally on the mobility of molecules, whereas the ion viscosity also depends on the ion concentration as it is affected by both ion concentration and mixture viscosity. Except of pure TEGDMA, shear and ion viscosities depend on the resin composition qualitatively in a similar manner. Furthermore, shear and ion viscosities of the commercial VLC dental resins and composites exhibited the same temperature dependency regardless of filler content. Application of typical rheological models (Kitano and Quemada) revealed that ion viscosity measurements can be described with respect to filler contents of up to 30 vol.%.

Significance. Rheological behavior of a VLC RBC can be characterized by DEA under the condition that the ion concentration is kept constant. Both methods address the same physical phenomenon – motion of molecules. The proposed relations allows for calculating the viscosity of any Bis-GMA-TEGDMA mixture on the base of the viscosities of the pure components. This study demonstrated the applicability of DEA investigations of VLC RBCs with respect to quality assurance purposes.

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1. Introduction

Visible light curing resin based composites (VLC RBC) for esthetic tooth-colored dental restorations have successfully replaced amalgam fillings or gold inlays [1–3]. They consist of an organic light-curable resin matrix, inorganic solid filler powder having micro and/or nano size and functional additives [4]. The flow properties of pure VLC resins, semi-filled so called flowables and highly filled composites play an important role in clinical handling as well as manufacturing [3–6].

First, the differences in a monomer mobility, and thus viscosity, affect strongly the curing reaction of the pure resin. Viscosity of visible light curing (VLC) monomer resins varies from 0.01 up to 800 Pa s at room temperature, and is typically Newtonian [3,7,8].

Second, besides resin composition the filler content, its particle size, size distribution, shape and tendency to agglomeration affect the flow characteristics of composites. VLC dental composites are reported to behave pseudo-plastic [3,8–10] and/or thixotropic [3,9]. Furthermore, close to the maximum filler content the viscosity increases dramatically and the filler particles will not be covered completely with resin leading to unfavorable kneading properties, and thus undesired gap formation at the interface of the tooth restoration causing secondary caries.

Despite of a certain viscosity required by clinicians in order to have specific shaping and manipulation properties of the composite [3,5,6,9,11], quality, biocompatibility and longevity of composite restorations are also governed by the curing behavior, e.g. curing kinetics, degree of cure (DC), depth of cure (DoC), curing shrinkage and relaxation properties [12,13]. While DoC, shrinkage and mechanical properties of composites are strongly affected by the filler shape, size, size distribution and content, the curing kinetics, DC, relaxation and post-curing properties are governed mainly by the VLC resin system itself (base and diluent monomers, initiators, accelerators and daylight stabilizers) [4,14].

Rheometry is a widely used testing method to investigate both the flow properties during processing of the uncured composites as well as their curing properties [3,5,6,11,15–17]. The curing process is mainly investigated in an oscillatory mode [3,8,10,18]. However, it is difficult to design the experiments similar to the dental application process concerning sample mass, layer thickness and illumination intensity. Furthermore, after gelation the dynamic (oscillatory) tests soon reach their resolution limit [10,18,19].

A lot of effort has been spent to fit rheological data of particle filled composites with models intercepting various parameters (especially filler content) [20–25]. Nevertheless, examination of validity of these models on experimental data for highly filled composites revealed surprisingly ambiguous results. Contrary to the fact that several models fitted relatively well the flow data for lower filler contents, for higher filler contents irregular filler particle shape, agglomeration and size distribution are highly affecting the flow behavior [20–24]. The predicted values of the maximum volume fraction for the same powder depend on the model employed [21].

Dielectric Analysis (DEA) is another method to gain information about the flow and curing properties of resins and composites. It is widely used in composites manufacturing in automotive and aircraft industry [26–31], but a DEA setup allowing for analyzing the light-curing process of dental composites at high time resolution has been introduced quite recently [32–37]. Further, the DEA method is advantageous in monitoring the polymerization of VLC composites even after the glass transition [38–40].

This study correlates and compares DEA and rheological data of various uncured VCL dental resins and composites differing in their filler content. The aim is to investigate the influence of temperature, resin composition and filler content, and thus only the measurements of initial ion viscosities and shear viscosities prior to curing are presented and discussed.

2. Materials and methods

2.1. Materials

Experimental mixtures of bisphenol A glycidyl methacrylate (Bis-GMA) and triethylene glycol dimethacrylate (TEGDMA) in ratios 0:100, 20:80, 40:60, 60:40, 80:20 in wt.% were supplied by Voco GmbH, Cuxhaven, Germany, and investigated prior to curing using both DEA and rheometry at clinically relevant temperatures of 23 °C (extraoral material preparation) and 36 °C (body temperature). The experimental mixtures contained small amounts of storage additives (Bis-GMA contained <1 wt.% Hydochinonmonomethylether and <0.1 wt.% Hydrochinone, TEGDMA contained 0.008–0.012 wt.% Hydochinonmonomethylether), light sensitive initiator system consisting of camphorquinone (CQ, 0.3 wt.%) and ethyl 4-(dimethylamino)benzoate (DABE, 0.5 wt.%) and stabilizer butylated hydroxytoluene (BHT, 0.1 wt.%).

Two commercial VLC dental resins (supplied by Voco GmbH, Cuxhaven, Germany) Arabesk Top (shade A1, Batch no. 1307385) and Grandio (shade I, Batch no. 1316541) were also investigated prior to curing during DEA and rheometry at clinically relevant temperatures of 23 and 36 °C. To account for the effects of the filler content on the ion viscosity, various filler contents were realized by diluting the commercial composites in their corresponding resins (also supplied by VOCO), Table 1. The two components were stirred until a homogeneous paste was achieved. The stirring process was repeated prior to every measurement to compensate for sedimentation processes. The Arabesk Top resin consists of a monomer mixture of Bis-GMA, UDMA and TEGDMA and is polymerized by a CQ/DABE system. The Grandio resin consists of a monomer

Table 1 – Commercial composites diluted in pure resins.

Material	Filler volume fraction v_F
Arabesk Top (Bis-GMA + UDMA + TEGDMA)	0.60
	0.30
	0.15
	Pure resin
Grandio (Bis-GMA + TEGDMA)	0.71
	0.60
	0.30
	0.15
	Pure resin