DENTAL MATERIALS 31 (2015) 1003–1009

Available online at www.sciencedirect.com

ScienceDirect

journal homepage: www.intl.elsevierhealth.com/journals/dema

Viscoelastic properties of uncured resin composites: Dynamic oscillatory shear test and fractional derivative model

Ljubomir M. Petrovic[a], Dusan M. Zorica[b], Igor Lj Stojanac[a], Veljko S. Krstonosic[c], Miroslav S. Hadnadjev[d], Marko B. Janev[b], Milica T. Premovic[a], Teodor M. Atanackovic[e,*]

[a] Clinic of Dentistry, Faculty of Medicine, University of Novi Sad, Serbia
[b] Mathematical Institute, Serbian Academy of Arts and Sciences, Belgrade, Serbia
[c] Department of Pharmacy, Faculty of Medicine, University of Novi Sad, Serbia
[d] Institute for Food Technology, University of Novi Sad, Serbia
[e] Department of Mechanics, Faculty of Technical Sciences, University of Novi Sad, Trg D. Obradovica 6, 21000 Novi Sad, Serbia

ARTICLE INFO

Article history:
Received 5 September 2014
Received in revised form
18 February 2015
Accepted 21 May 2015

Keywords:
Resin composites
Uncured
Flowable and universal hybrid resin
Dynamic oscillatory shear test
Viscoelasticity of fractional order

ABSTRACT

Objective. In this study we analyze viscoelastic properties of three flowable (Wave, Wave MV, Wave HV) and one universal hybrid resin (Ice) composites, prior to setting. We developed a mathematical model containing fractional derivatives in order to describe their properties.
Methods. Isothermal experimental study was conducted on a rheometer with parallel plates. In dynamic oscillatory shear test, storage and loss modulus, as well as the complex viscosity where determined. We assumed four different fractional viscoelastic models, each belonging to one particular class, derivable from distributed-order fractional constitutive equation. The restrictions following from the Second law of thermodynamics are imposed on each model. The optimal parameters corresponding to each model are obtained by minimizing the error function that takes into account storage and loss modulus, thus obtaining the best fit to the experimental data.
Results. In the frequency range considered, we obtained that for Wave HV and Wave MV there exist a critical frequency for which loss and storage modulus curves intersect, defining a boundary between two different types of behavior: one in which storage modulus is larger than loss modulus and the other in which the situation is opposite. Loss and storage modulus curves for Ice and Wave do not show this type of behavior, having either elastic, or viscous effects dominating in entire frequency range considered.
Significance. The developed models may be used to predict behavior of four tested composites in different flow conditions (different deformation speed), thus helping to estimate optimal handling characteristics for specific clinical applications.

* *Corresponding author*. Tel.: +381 632 102 81 39; fax: +381 21 458 133.
E-mail address: atanackovic@uns.ac.rs (T.M. Atanackovic).

http://dx.doi.org/10.1016/j.dental.2015.05.009
0109-5641/

1. Introduction

Contemporary restorative dentistry relies greatly on the use of resin composite materials for the restoration of lost tooth structure. However, placing dental composite is not an easy task; it requires skilful and informed clinician who is able to relate the characteristics of commercially available resin composites with specific requirements for each indication in the field of restorative dentistry. With the wide range of currently existing types of composites with different characteristic it becomes difficult for the dentist to make appropriate choice. First and widely accepted classification of composites has been based on filler particle size [1,2]. Resin composites can also be classified to give more information about their: clinical indications (anterior, posterior or universal), curing mode (self curing, light curing or dual curing), or viscosity (packable, universal, flowable) [3–5]. Viscosity is directly related to the handling characteristic of the resin composite material like mode and ease of placement, operation time, anatomical shaping, as well as the final quality of a restoration [6,7]. From a clinical point of view, uncured resin composites should flow easily into the cavities in order to achieve better adaptation to the cavity walls and yet, have high viscosity characteristics for facilitating shaping in order to recreate the anatomy of the restored teeth. Moreover, in large cavities resin composites should be placed in successive increments to ensure light-curing and prevent excessive polymerization stresses particularly on cavo-restoration interface [8,9].

All these properties are currently hard to find in one single composite, so manufacturers have developed composite materials with different viscosities which help dentists to choose the most appropriate form. Packable or condensable composites are highly viscous materials designed to imitate the handling behavior of a dental amalgam. Increased viscosity and high filler particles content of packable composites require more force needed to adapt the material in the cavity. Commercially available light-cured resin composites mainly have a generically useful viscosity which is suitable for most clinical conditions. However, they are not indicated for very small cavities or fissures and pits, because they are not able to flow in the cavity. Flowable composites are low viscosity resin-based restorative materials with reduced filler content, compared to non-flowable composites. The viscosity and flow characteristic of flowable resin composites are obvious since their handling is based on the flow of the material in the cavity. In addition, resin-composites are pseudoplastic materials meaning that they become more fluid during placement with a syringe.

Generally, paste type dental materials like resin composites are viscoelastic materials positioned intermediately between elastic, such as metals, and viscous materials like oils. They show non-Newtonian, shear-thinning flow behavior.

Lee et al. [10] using a dynamic oscillatory shear test, reported significant differences in the viscoelastic properties between flowable, universal hybrid and packable composites and even more, significant differences in the viscoelastic properties between composites in the same class. Beun et al. [11] studied viscoelastic properties of unset flowable resin composites and reported huge differences in the viscosity and flow characteristic that can have a potential influence on their handling behavior and thus on their clinical indications. Recently, Watts et al. [12] used a dynamic oscillatory rheometer to evaluate the influence of resin mixture, filler content and temperature on the viscosity of resin composites.

The aim of this study was to develop a new fractional derivative model for the assessment of viscoelastic properties related to handling characteristics of three flowable and one universal hybrid resin composite. We shall use distributed-order fractional isothermal viscoelastic model to derive several specific models, each capable to describe obtained experimental data.

2. Material and methods

Three commercial flowable composites (with three different viscosities) and one universal composite (designed for anterior and posterior restorations) were used in this study. They are listed in Table 1. It is to be noted that all tested composites were from the same manufacturer, as it was done in [13].

2.1. Rheological measurements

Rheological measurements on all four materials tested in this study were performed using an HAAKE Mars Rheometer (Thermo Fisher Scientific Process Instruments, Karlsruhe, Germany) at a temperature of $23 \pm 0.1\,^{\circ}\mathrm{C}$, as previously reported in [14].

A parallel plates viscometer module with a diameter of 20 mm was used to measure the rheological properties of the flowable resin composites tested. The gap between the plates was 1 mm. In amplitude sweep tests linear viscoelastic region was determined by recording the storage (G') and loss (G'') moduli versus shear stress (0.01–100 Pa) at constant angular velocity (6.28 rad/s). Appropriate shear stress were selected and kept constant in frequency sweep tests. Frequency sweep test was performed from 0.628 to 62.8 rad/s in order to determine the variation of the complex viscosity (η^*), storage (elastic) shear modulus (G') and loss (viscous) shear modulus (G''). Three measurements were performed for each material tested.

2.2. Material modeling and optimization procedure

Most materials used in dentistry have viscoelastic properties. Viscoelastic properties of materials are lately successfully modeled by the use of fractional derivatives, see [15–17]. In general, a viscoelastic body may be fluid or solid like. In the first case it has infinite creep strain, while in the second case the creep strain is finite. Creep strain is obtained in a creep test, when material is subject to a sudden, but later constant force on its free end.

Recall, the Riemann-Liouville fractional derivative of order $\alpha \in (0,1)$ is defined by

$$f^{(\alpha)}(t) = \frac{1}{\Gamma(1-\alpha)} \frac{\mathrm{d}}{\mathrm{d}t} \int_0^t \frac{f(\tau)}{(t-\tau)^{\alpha}}\,\mathrm{d}\tau.$$

Note $f^{(0)}(t) = f(t)$ and $\lim_{x \to 1} f^{(\alpha)}(t) = \frac{\mathrm{d}}{\mathrm{d}t} f(t)$. The first property is obvious, while the second one holds true for a broad