Creep-assisted slow crack growth in bio-inspired dental multilayers

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Ceramic crown structures under occlusal contact are often idealized as flat multilayered structures that are deformed under Hertzian contact loading. Previous models treated each layer as linear elastic materials and resulted in differences between the measured and predicted critical loads. This paper examines the combined effects of creep (in the adhesive and substrate layers) and creep-assisted slow crack growth (in the ceramic layer) on the contact-induced deformation of bio-inspired, functionally graded multilayer (FGM) structures and the conventional tri-layers. The time-dependent moduli of each of the layers were determined from constant load creep tests. The resulting modulus-time characteristics were modeled using Prony series. These were then incorporated into a finite element model for the computation of stress distributions in the sub-surface regions of the top ceramic layer, in which sub-surface radial cracks, are observed as the clinical failure mode. The time-dependent stresses are incorporated into a slow crack growth (SCG) model that is used to predict the critical loads of the dental multilayers under Hertzian contact loading. The predicted loading rate dependence of the critical loads is shown to be consistent with experimental results. The implications of the results are then discussed for the design of robust dental multilayers.

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1. Introduction

The study of the contact loading of dental restorations/crowns is often conducted on idealized, flat multilayered structures, that are deformed under Hertzian contact loading (Kelly, 1997; Lawn et al., 2000, 2004; Lee et al., 2002; Malament and Socransky, 1999; Rekow and Thompson, 2007; Zhang et al., 2004). This often leads to the onset of clinically-relevant sub-surface radial cracking, due to the high stress concentrations in the sub-surface regions in the top ceramic layer (Huang et al., 2007b; Shrotriya et al., 2003). Such radial cracking is also consistent with the major clinical failure mode reported by Kelly (1997).*

Abbreviations: DEJ, dento-enamel-junction; FEM, finite element method; FGM, functionally graded multilayer; SCG, slow crack growth; CASCG, creep-assisted slow crack growth

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In this paper, the viscosity of the each layer in model dental multilayers is measured using creep experiments. The measured viscous behavior is then modeled using Prony series (Bower, 2011). The Prony series fits to the experimental data are compared with those obtained from other models. A creep-assisted slow crack growth model is then developed. The model, which incorporates the combined effects of slow crack growth (in the top ceramic layers) and the viscosity of the adhesive and substrate layers, is used to predict the critical pop-in loads. The implications of the results are then discussed for the design of robust, bio-inspired, dental multilayer structures.

2. Materials and experimental methods

2.1. Fabrication of bio-inspired FGM dental structures

The bio-inspired FGM structure was fabricated using a steel plate mold with a thickness of ~4 mm and an inner diameter of ~9 mm. The substrate was a dentin-like soft material, Z100 restorative (3M ESPE Dental Products, St. Paul, MN), which is a clinically-used dental material. It was poured into the mold and then cured with UV light. The FGM was produced using nanocomposite mixtures of zirconia or alumina nanoparticles (NanoTek Instrument Inc., Dayton, OH) and an epoxy matrix, EPO-TEK 301 (Epoxy Technology Inc., Billerica, MA). After mixing, the nanocomposite material was deposited in the steel mold and then cured in a vacuum oven at a temperature of 65 °C. The deposition and curing process were then repeated to build up the multilayers.

The crown-like dental ceramic layer on top was fabricated from a medical grade 3 mol% yttria-stabilized zirconia rod (YTZP, Saint-Gobain, Colorado Springs, CO). It was pressed onto the last layer of the FGM before curing. Finally, the multilayered structures, with a diameter of ~9 mm and a thickness of ~5 mm, were cut and removed from the mold. They were then cleaned with distilled water and blow dried with compressed air.

2.2. Hertzian contact experiments

Hertzian contact experiments were performed on the fabricated dental multilayers (Fig. 1a). The tests were carried out in an Instron 8872 hydraulic mechanical tester (Instron, Canton, MA, USA). They were conducted in air at room temperature and a relative humidity of ~25%. The Hertzian contact tests were performed under load control with a hemispherical tungsten carbide indenter with a diameter of 20 mm. The tests conducted at clinically relevant loading rates between 1 N/s and 1000 N/s (Du et al., 2013). The loads and displacements were recorded by the computer attached to the Instron tester. The critical loads were then determined as the loads at which discontinuities in displacement were observed. These were also found to correspond to the onset of cracking, which could be heard clearly during the tests.

2.3. Creep experiments

The creep specimens were fabricated using the same steel mold that was described above. The nanocomposite materials for
each FGM layer were filled separately into the steel mold and cured in the vacuum oven at 65 °C. The nanocomposite layers consisted of epoxy matrix and 10 wt% zirconia, 20 wt% zirconia, 30 wt% zirconia, 40 wt% zirconia, 50 wt% zirconia, 60 wt% zirconia, 70 wt% zirconia, 40 wt% alumina and 45 wt% alumina reinforcements. The substrate material, Z100 restorative, was also molded and cured with UV light for 40 s for both sides. The cured materials were then removed from the mold, polished and cleaned with distilled water. Each specimen has a diameter of ~9 mm, a thickness of ~4 mm, and the surface roughness corresponding to 600grit sand papers.

Creep tests were carried out in a self-built creep tester (Fig. 1b). It was similar to compression test without friction (homogenous compression). A dead load of 139.2 N was applied to the specimen through a flat anvil. The applied compression stress was about 260 MPa. The deformation of the specimen was detected using a capacitance gauge (Capacitec, Ayer, MA). The resulting displacements were recorded using the LABView software package (National Instruments, Austin, TX). Each test lasted for about 24 h. For each material, between 4 and 5 specimens were tested.

3. Modeling
3.1. Constitutive equations

In an effort to develop a viscous model that can be used over a range of loading rates, a three-parameter spring-dashpot solid model (Zener model) has been used to describe the viscous behavior of dental multilayers (Huang et al., 2005, 2007a; Zhou et al., 2007). However, the model, which has only one dashpot, was found to be insufficient for the description of long-term polymer creep behavior (Huang et al., 2007a). There is, therefore, a need for an improved model for the characterization of the viscoelastic behavior of dental multilayered structures.

Since the relaxation times of polymers occur in a distribution, due to the heterogeneity of the polymeric structures (McCrum et al., 1997), Prony series use a series of relaxation times to fully describe the creep or relaxation of polymeric structures, over an extended period of time. Prony series are given by (Bower, 2011)

\[
G(t) = G_0 \left(1 - \sum_{i=1}^{N} g_i \left(1 - e^{-t/\tau_i}\right)\right)
\]

where \(G(t)\) is the time-dependent shear modulus; \(G_0\) is the instantaneous modulus; \(\tau_i\) is the relaxation time and \(g_i\) is the relaxation coefficient. When there is only one exponential term, the Prony series is equivalent to the Zener model.

3.2. Finite element simulations

The finite element simulations were carried out using the Abaqus FEA software package (Dassault Systemes Simulia Corporation, Providence, RI). The stress distributions in the dental multilayers were calculated. Axisymmetric geometries were used to simplify the problem, as shown in Fig. 2. The thickness of the bonding layer was modeled to be 100 μm. A 4-node linear axisymmetric quadrilateral element was used in the mesh. In an effort to capture the high stress concentrations, the mesh was dense in the regions near the axisymmetric axis of the model. The bottom of the substrate was fixed, while the axisymmetric boundary condition was added on the axisymmetric axis.

The simulations considered bio-inspired dental FGM multilayers, as well as structures joined by single adhesive material. The materials and the properties used in the simulation for glass-epoxy-polycarbonate and zirconia-FGM-Z100 multilayer structures are listed in Tables 1 and 2.
respectively. The Prony series parameters were obtained from the creep test experiments. The FGM was modeled as 10 sub-layers, with equal layer thicknesses. The elastic moduli for the FGM composites materials were measured using nanoindentation techniques described in a prior study (Du et al., 2013). The Poisson ratio was assumed to be 0.2 for ceramic materials and 0.4 for polymeric and composite materials.

A load of 2500 N was applied to the Hertzian indenter, which was modeled as a rigid surface. In the simulations that consider linear elastic material properties, the load was applied instantaneously. In the contrast, in the simulations that consider the viscous material properties, the load was applied gradually in a linear ramp in durations of 2.5, 25, 250 and 2500 s. They were used, respectively, to simulate loading rates of 1000, 100, 10 and 1 N/s.

### 3.3. Creep-assisted slow crack (CASCG) growth model

The tensile stress at the sub-surface center of the top ceramic layer is associated with the major clinical failure mode, the sub-surface radial crack (Kelly, 1997), in the dental multilayer. The slow crack growth (SCG) model suggests that, during contact damage in dental multilayers, sub-surface radial crack growth occurs solely as a result of slow crack growth (SCG) model into the sub-surface regions of the top ceramic layer; \( t_R \) is the rupture time, at which sub-surface radial cracks occur. When the loading rate, \( \dot{P} \), is constant, the rupture time, \( t_R \), can be expressed as \( t_R = \frac{P}{\dot{P}} \), where \( \dot{P} \) is the critical "pop-in" load.

The processes involved in the creep-assisted slow crack growth (CASCG) model are illustrated in Fig. 3. By incorporating the actual measurements of Prony series parameters (obtained from the creep test) into the finite element simulations, an expression was obtained for the time-dependent stress, \( \sigma(t) \). The critical load versus loading rate obtained from the Hertzian contact experiments was then used to fit the value of \( D \). Since \( D \) is only related to material properties and geometry, it should be consistent for different loading rates. Therefore, with \( D \) known, Eq. (2) was integrated to obtain the rupture time, \( t_R \), and then the critical load \( P_c \).

### 4. Results and discussion

#### 4.1. Viscoelastic behavior of single dental layer

The time-dependent shear moduli measured from creep tests (on the FGM dental structure) are illustrated in Fig. 4. The measured moduli appeared to have discrete values, because the measured deformation values were limited by the spatial resolution of the capacitance gauge that was used in the constant load creep test. Fig. 4a presents a comparison of epoxy nanocomposite materials with 10 wt%, 20 wt%, etc.,
and 70 wt% zirconia particles. In general, for composites with the same type of ceramic reinforcements, increased ceramic reinforcement weight fraction resulted in higher instantaneous moduli and relaxed moduli.

The measured shear moduli are presented in Fig. 4b. These are presented for epoxy nanocomposite with 40 wt% and 45 wt% alumina particles, as well as Z100 dental material, which is also a ceramic filled polymeric composite. The instantaneous modulus of Z100 was much greater than that of the alumina filled epoxy composites. However, the relaxed modulus for Z100 was smaller than those of the two epoxy/alumina composites. This indicates greater decay of the modulus of the Z100 samples over time.

The measured creep rates of the epoxy composites with similar weight percentages of zirconia and alumina fillers are compared in Fig. 4c. The alumina-reinforced composites had higher instantaneous moduli than the zirconia-reinforced composites, since alumina has a higher modulus than zirconia. However, these relaxed moduli were similar. This indicates greater modulus decay for the alumina-reinforced epoxy composites.

The time-dependence of shear modulus was modeled using several viscoelastic behavior models. Fig. 5a shows that the Zener model, with a single relaxation time, is not sufficient to capture the creep behavior of polycarbonate over a duration of 15,000 s. However, the predictions from the Prony series model, with two relaxation times, are in closer agreement with the experimental data. Furthermore, the Prony series model also exhibits good agreement with experimental data obtained for polycarbonate and epoxy (Fig. 5b). The resulting Prony series parameters for polycarbonate and epoxy are presented in Table 1.
The Prony series predictions obtained for the time-dependence shear modulus are presented in Fig. 6 for a typical single layer in the FGM, containing epoxy reinforced by 70 wt% zirconia. For Prony series with one to three exponential terms, the results show that the range of relaxation time increases with increasing number of exponential terms. Furthermore, the Prony series predictions were in closer agreement with the experimental data, as the number of exponential terms increased. Also, for a duration of \( \frac{1}{C^2} \) day \((10^5 \text{ s})\), three exponential terms were needed to model the creep behavior.

A summary of the Prony series parameters obtained for the FGM layers is presented in Table 2. The coefficients of determination, \( R^2 \), of the curve fittings were all greater than 0.85. The experimental results fell in the trend line of Prony series, as the number of exponential terms increased. Also, for a duration of \( \sim 1 \) day \((10^5 \text{ s})\), three exponential terms were needed to model the creep behavior.

4.3. Critical loads of bio-inspired dental FGM structures

In the case of the FGM structures, the computed tensile stresses associated with SCG in the subsurface regions of
the top ceramic layer are presented in Fig. 8. The tensile stresses increase with increasing Hertzian contact load. The stresses obtained from the viscoelastic models were greater than those obtained from linear elastic models. The differences between the stresses were greater at slower loading rates, where the viscous components had more time to respond. Furthermore, at slower loading rates, the FGM was less stiff. It, therefore, provided less support to the top ceramic layer than at faster loading rates. Hence, the stresses in the top ceramic layer were higher at slower loading rates. Hence, according to Eq. (2), the stress accumulated more rapidly at slower loading rates. This explains the lower critical loads that we obtained at slower loading rates.

The critical “pop-in” loads obtained for the FGM structures (under Hertzian contact loading) at different loading rates are presented in Fig. 9. The critical pop-in loads increased with increasing loading rates. For any given loading rate, the critical loads obtained for the FGM structures were greater than those obtained for the conventional tri-layer structures with single adhesive layer. The differences were greater for slower loading rates than they were for faster loading rates. For any given loading rate, the critical loads obtained from the experiments on FGM structures were lower than those predicted using linear elastic models in this study. The results also show clearly that the predictions from the CASCG model were in closer agreement with the experimental results obtained from the FGM structures.

4.4. Implications

The current results suggest that the critical loads for failure in dental multilayers can be estimated accurately when the effects of viscosity in the polymeric substrate and adhesive layers are accounted for. This is especially true when the model captures the distribution of relaxation times for polymers due to the heterogeneity of the polymer. Furthermore, failure to model the viscosity of individual layers in dental multilayers may result in non-conservative predictions of “pop-in” loads or structural life. This is especially true in the high cycle fatigue regime where the effects of viscosity are likely to be more significant. Further studies of contact-induced failure are needed to explore the effects of cyclic loading and environments that are relevant to occlusal contact. There is, also, a need to study real teeth and actual crown geometries that incorporate the combined effects of slow crack growth in the top ceramic layers and viscosity of the adhesive and substrate layers. These are clearly some challenges for future work.

5. Conclusions

This paper presents the results of a combined experimental and analytical/computational study of the effects of creep
behavior on the contact-induced fracture of bio-inspired dental FGM structures and the conventional tri-layer structures. The creep effects in the adhesive layer and substrate layer play an important role in determining the critical "pop-in" loads at various loading rates under Hertzian contact. The loading rate effects on the critical loads of dental multilayers can be explained by a combination of the adhesive and substrate layer creep and the creep-assisted slow crack growth in the ceramic layer. The adhesive and substrate layer creep are well predicted by Prony series with three exponential terms.

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