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Subcritical crack growth and in vitro lifetime prediction of resin composites with different filler distributions

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ABSTRACT

Objectives. Verify the influence of different filler distributions on the subcritical crack growth (SCG) susceptibility, Weibull parameters (m and σ_0) and longevity estimated by the strength–probability–time (SPT) diagram of experimental resin composites.
Methods. Four composites were prepared, each one containing 59 vol% of glass powder with different filler sizes ($d_{50} = 0.5$; 0.9; 1.2 and 1.9 μm) and distributions. Granulometric analyses of glass powders were done by a laser diffraction particle size analyzer (Sald-7001, Shimadzu, USA). SCG parameters (n and σ_{f0}) were determined by dynamic fatigue (10^{-2} to 10^2 MPa/s) using a biaxial flexural device (12 × 1.2 mm; $n = 10$). Twenty extra specimens of each composite were tested at 10^0 MPa/s to determine m and σ_0. Specimens were stored in water at 37 °C for 24 h. Fracture surfaces were analyzed under SEM.
Results. In general, the composites with broader filler distribution (C0.5 and C1.9) presented better results in terms of SCG susceptibility and longevity. C0.5 and C1.9 presented higher n values (respectively, 31.2 ± 6.2^a and 34.7 ± 7.4^a). C1.2 (166.42 ± 0.01^a) showed the highest and C0.5 (158.40 ± 0.02^d) the lowest σ_{f0} value (in MPa). Weibull parameters did not vary significantly (m: 6.6 to 10.6 and σ_0:170.6 to 176.4 MPa). Predicted reductions in failure stress ($P_f = 5\%$) for a lifetime of 10 years were approximately 45% for C0.5 and C1.9 and 65% for C0.9 and C1.2. Crack propagation occurred through the polymeric matrix around the fillers and all the fracture surfaces showed brittle fracture features.
Significance. Composites with broader granulometric distribution showed higher resistance to SCG and, consequently, higher longevity in vitro.

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1. Introduction

Bulk fracture is one of the main causes of resin composite restoration failure in the clinic [1–6]. Resin composites are frequently evaluated in relation to their mechanical strength, which can be related to static loads that cause fracture of the restoration. However, such tests do not provide much information about failure mechanisms that occur during clinical use, which are related to the complex fatigue scenario found in the oral cavity, i.e., low cyclic loads and high humidity. In addition to strength tests, the association of other methods, such as fatigue tests, methods for determining subcritical crack growth (SCG) parameters, fractographic and Weibull analyses are necessary to understand the causes of failure [7–13].

Based on results of different mechanical tests and qualitative analyses, it seems that in the oral environment, repeated mechanical loads even at physiological levels can contribute to decrease the longevity of composite restorations due to the subcritical growth of flaws present in the material which eventually result in catastrophic failure [14]. The growth of a crack in a material depends on the material's behavior (plastic or elastic), temperature, strain rate and the stress state at the crack tip. The principles of the Linear-Elastic Fracture Mechanics (LEFM) are used to understand fracture events that occur without being preceded by extensive plastic deformation. The type of deformation observed during fracture is directly related to the material's properties and geometric factors, such as specimen thickness [15].

Plastic deformation occurs in brittle and ductile fractures. However, in brittle materials the crack needs minimal plastic deformation to propagate, while in ductile fracture this deformation is necessarily greater. Although the polymeric matrix of resin composites presents viscoelastic properties, fractographic analyses revealed features of brittle failure such as SCG [16–20]. This finding makes possible the application of the LEFM principles to study the failure mechanisms of resin composites.

The strength of many brittle materials is environment- and time-dependent, i.e., it decreases due to SCG in the presence of a reactive medium, such as water, and increases with the stress rate. This time-dependent strength is exponentially related to the material's susceptibility to SCG [21]. The susceptibility of a material to SCG, expressed by the stress corrosion coefficient (*n*), and the material's resistance in the initial stages of mechanical loading (σ_{f0}) can be determined by direct and indirect in vitro methods [22].

Direct methods are very complex, use large specimens and simulate large flaws, which do not correspond to the small flaws usually found in dental restorations [23]. For those reasons, indirect methods, as the dynamic fatigue test, are often used to obtain the SCG parameters based on mathematical relationships among fracture resistance values obtained at different stress rates [23–27]. High values of *n* (>100) indicate a significant resistance against SCG and low values (5–30) indicate a high susceptibility to this phenomenon [23,28]. In the scientific literature, it is possible to find *n* values ranging from 7 to 34 for resin composites, 15–28 for feldspathic porcelains and glass-ceramics and 60–95 for high-density alumina [25,29–38].

Weibull analysis associates the failure probability (P_f) of a material with the corresponding stress values (σ_f), resulting in an approach that takes into account the population of defects in a specimen [39,40]. The association between Weibull statistics and the results of a time-dependent analysis, as the dynamic fatigue, allows the estimation of a material's failure probability over its lifetime of for a given stress level [23]. This information is displayed in a strength–probability–time (SPT) diagram [41]. Such diagram is commonly used for dental ceramics to predict the maximum stress at which the material should survive for both a given time span and percentage probability of failure [22,42]. For resin composites, however, this analytical tool has not been explored in depth. SPT diagrams of commercial resin composites were previously constructed based on the compressive strengths obtained at crosshead speeds of 1, 5 and 10 mm min^{-1} and showed that for a lifetime of 10^2 s, with a failure probability of 38%, the working stress should not exceed 150 MPa, whereas for a lifetime of 10^3 s, with the same probability of failure, the applied stress should not exceed 100 MPa [41].

The mechanical behavior of resin composites is determined by the degree of conversion of the polymeric matrix, the inorganic fraction characteristics and the bonding between the fillers and the matrix [25,43–47]. In general, the fracture strength of composites increases with higher inorganic content [46–50]. Studies with experimental composites showed an inverse relationship between fracture strength and mean particle size. However, they did not explore the possible mechanisms responsible for that [45,51–53]. Besides, long term analyses as the SPT diagram using a clinically relevant failure probability of 5% are important to gather information about the lifetime of these materials [22]. The present study sought to verify the influence of different filler sizes and distributions of experimental resin composites in the SCG (*n* and σ_{f0}) and Weibull (*m* and σ_0) parameters and the longevity estimated by the SPT diagram. Such information could contribute to the understanding of the underlying mechanisms leading to bulk fractures found in clinical practice, as well as to the development of resin composite improved formulations. The null hypothesis was that there is no relationship between the filler size and/or distribution and the evaluated properties and parameters.

2. Materials and methods

2.1. Composite characterization

Four experimental resin composites were prepared (Table 1). All composites presented 22 wt% (41 vol%) of organic matrix and 78 wt% (59 vol%) of inorganic content. The inorganic fraction was constituted by 67 wt% (49 vol%) of glass particles and 11 wt% (10 vol%) of pyrogenic silica (Aerosil OX-50, Degussa Corporation, New Jersey, USA). All fillers were equally silanized using a solution containing hydrolyzed gamma-methacrylyloxypropyltrimethoxysilane (gamma-MPS, Dynasylan Memo, Degussa Corporation) by FGM Produtos Odontológicos (Joinville, SC, Brazil). Thermogravimetric analyses (TGA) showed that the content of gamma-MPS was 5.6% (w/w) for all the fillers. The organic matrix was the same