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Fracture toughness and cyclic fatigue resistance of resin composites with different filler size distributions

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ABSTRACT

Objectives. To verify the influence of filler size distributions on fracture toughness (K_{Ic}), initial fracture strength (IFS) and cyclic fatigue resistance (CFR) of experimental resin composites.
Methods. Four composites were prepared with same inorganic content (78 wt%), in which 67 wt% was constituted by glass particles with d_{50} of 0.5; 0.9; 1.2; 1.9 μm K_{Ic} of the composites was determined by the single-edge notched beam (SENB) method. To evaluate the IFS and the CFR a biaxial bending test configuration was used. The CFR was determined under cyclic loading for 10^5 cycles using the 'staircase' approach. The fracture surfaces of IFS and CFR specimens were analyzed under scanning electron microscope (SEM).
Results. There was a positive linear correlation between d_{50} vs. K_{Ic} and statistical difference was found only between C0.5 (1.24 ± 0.10 MPa $m^{0.5}$) and C1.9 (1.41 ± 0.17 MPa $m^{0.5}$). There were no statistical differences among IFS means, which ranged from 155.4 ± 18.8 MPa (C0.9) to 170.7 ± 23.1 MPa (C1.2). C0.5 (93.0 ± 18.6^a MPa) showed the highest and C0.9 the lowest CFR (82.5 ± 8.0^c MPa). There was no correlation between CFR with d_{50} values or with K_{Ic} means. SEM images showed the morphology with brittle fracture patterns for the surfaces of IFS specimens and a more smooth fracture surface for CFR specimens.
Significance. Resin composites showed different failure mechanisms for quasi-static and fatigue loading. For K_{Ic} and IFS, composites with larger filler size distributions showed better results due to crack deflection; while under cyclic loading, viscous behavior was predominant and composites with smaller particles showed higher fatigue resistance.

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1. Introduction

Mechanical properties express the response of materials to external loading, manifested by their ability to sustain reversible or irreversible deformation, or to fracture [1]. Bulk fracture is one major cause of failure of resin composite restorations [2–7], and the repeated application of mechanical loads in the oral cavity, even at levels considered physiological, may reduce the longevity of the restoration due to slow crack growth (SCG) degradation, originated from material intrinsic defects [8]. Therefore, knowing the material's behavior in terms of fracture toughness and fatigue resistance is of utmost importance to evaluate its clinical performance [9].

The critical conditions for catastrophic crack propagation are evaluated using the Linear Elastic Fracture Mechanics (LEFM) concept. Crack growth can be studied based on the stress state at the crack tip using the stress intensity factor (K_I) or based on the release of elastic strain energy using the strain energy release rate (G_I). The critical values of both parameters (K_{Ic} and G_{Ic}) correspond to the final moment of the stable crack growth, i.e., the limit values reached immediately before the catastrophic crack growth under the influence of an external load [10–12]. For plane strain conditions, these parameters are related by:

$$K_{Ic} = \sqrt{\frac{G_{Ic}E}{(1-\upsilon^2)}} \quad (1)$$

where E is the elastic modulus and υ is the Poisson's ratio [1,11,13]. These critical values describe the last events of the failure process, but do not characterize the early stages of SCG [10].

Resin composites show an increase in stress intensity factor with the size of the crack, which follows an R-curve behavior [14–17]. That occurs because of stress reducing toughening mechanisms at the tip of a growing crack [17,18]. Some of these mechanisms are related to the characteristics of the dispersed phase of resin composites and have been identified as deflection, bridging, and branching of a progressing crack, or blunting of crack tips [19,20].

Cyclic masticatory loads initiate the process of failure by fatigue. Fatigue is a progressive reduction in strength as a result of slow crack growth in combination to hydrolytic degradation and viscoelastic relaxation of the polymer network, specifically for polymeric materials, such as resin composites [10,21]. Thus, cyclic fatigue tests conducted in humid environments are another important tool for assessing the life expectancy of a material under stress, under clinically relevant loading conditions [22]. For brittle materials, such as resin composites, the usual methods to evaluate the material's resistance to fatigue include testing cylindrical specimens in compression or beam specimens in flexure. Data are presented as *s-n* diagrams, where *s* is the mean or maximum stress in a cycle and *n* is the number of cycles until failure. Therefore, fatigue strength can be estimated by the number of cycles to failure. However, such evaluation is more time-consuming and there is a large variation in number of cycles to failure, even under similar experimental conditions [10,23–25]. For that reason, the 'staircase' method can be an alternative method for studying fatigue [21,26–28].

In this method, the maximum stress of each cycle of the first specimen in each group corresponds to a 50% probability of failure, i.e. 50% of fracture stress obtained in the pilot study under the same experimental conditions. Furthermore, there is a reduction in the number of specimens per group to determine the fatigue limit, since the tests are conducted sequentially with the maximum stress applied to each test being increased or decreased at a fixed stress increment, depending on whether the previous test resulted in failure or survival of the specimen to a given number of cycles. Moreover, with this method it is possible to calculate the standard deviation of the fatigue limit [21,29,30].

Based on the above, the aim of this study was to verify the influence of different filler size distributions on fracture toughness (K_{Ic}), initial fracture strength (IFS) and cyclic fatigue resistance (CFR) of experimental resin composites and analyze by the fractographic features the failure mechanism presents on these materials when they are submitted to different loading conditions.

2. Material and methods

2.1. *Formulation and characterization of the experimental composites*

Four experimental resin composites, all presenting the same organic matrix (22 wt% and 41 vol%) and inorganic content (78 wt% and 59 vol%), were prepared. The organic matrix was the same for all composites, consisting of 59 wt% of BisGMA (bisphenol A glycidyl dimethacrylate, Degussa Corporation, NJ, USA), 39 wt% of TEGDMA (triethyleneglycol dimethacrylate, Degussa Corporation), 0.5 wt% of camphorquinone (Sigma Aldrich, St. Louis, USA), 1.0 wt% of the co-initiator ethyl N,N-dimetil-4-aminobenzoate (EDMAB, Sigma Aldrich,) and 0.1 wt% of the inhibitor butyl-hydroxytoluene (BHT, Sigma Aldrich). 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). All fillers were silanized using a solution containing hydrolyzed gamma-methacryloxypropyltrimethoxysilane (gamma-MPS, Dynasylan Memo, Degussa Corporation). Filler size distribution for each composite, as well as their d_{25}, d_{50}, d_{90} values, the minimum and maximum filler size and the specific surface areas of the silanized glasses, and the degree of conversion of each composite are shown in Table 1. Each composite was labeled based on its d_{50} grain size value.

For microstructural characterization, disk-shaped specimens (2.0 mm thick and 5.0 mm in diameter) of each composite were built using a stainless steel split mold and light activated under 1200 mW/cm^2 (Radii-cal, SDI, Victoria, Australia) for 40 s. Immediately after light curing, the upper surface of each specimen was polished using 400 grit sandpaper. Half of each surface was etched by 5% hydrofluoric acid gel (Vita Ceramics Etch, VITA Zahnfabrik, Bad Säckingen, Germany) for 15 s. The etched and non-etched surfaces were gold-sputtered (model SCD 050, Bal-Tec Coating System, Balzers, Liechtenstein) and analyzed in a scanning electron