DENTAL MATERIALS 32 (2016) 1555–1563

Available online at www.sciencedirect.com

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

High strength and toughness in chromatic polymer-infiltrated zirconia ceramics

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ARTICLE INFO

Article history:
Received 14 June 2016
Received in revised form
22 August 2016
Accepted 3 September 2016

Keywords:
Polymer-infiltrated ceramic
3Y-TZP
Mechanical properties
Fe_2O_3
Weibull analysis

ABSTRACT

Objective. To evaluate the microstructure and mechanical behavior of polymer-infiltrated zirconia ceramics as a function of Fe_2O_3 concentration (0–0.3 mol%).

Methods. Polymer-infiltrated zirconia ceramics with different concentrations of Fe_2O_3 were prepared by infiltration and polymerization. XRD was employed to determine phase structure. The microstructure and fracture mechanism was observed by SEM. Flexural strength and fracture toughness were measured by three-point bending method and single-edge-notched beam method, respectively. Data were analyzed by Weibull distribution. A nanoindentation system was employed to determine elastic modulus and hardness.

Results. With increasing content of Fe_2O_3, the flexural strength, fracture toughness, elastic modulus and hardness are all greatly enhanced and the chromatic behavior also improves significantly. As a tradeoff made between strength and elastic modulus, specimen containing 0.2 mol% Fe_2O_3 is found to be the better one, with flexural strength and fracture toughness values being 336.8 MPa and 3.91 MPa m$^{1/2}$, respectively. Moreover, it maintains a relatively low elastic modulus of 88.2 GPa and a moderate hardness of 4.8 GPa, close to those of natural enamel.

Significance. This polymer-infiltrated zirconia ceramic material is a dental material of biomimetic chromatic and mechanical properties.

1. Introduction

Over the past 40 years, resin-based composites have been widely applied in dental clinic and been extensively investigated by the scientific community due to their great esthetics and outstanding machinability [1–3]. But dental composites suffer from poor mechanical properties, especially the low flexural strength and fracture toughness. Taking 3M ESPE [4–8] for example, the flexural strength and fracture toughness of productions in the well-known company manufacturing dental composites vary from 64.50 MPa to 165.90 MPa and 1.01 to 1.92 MPa m$^{1/2}$, respectively, which may be similar to porcelain [9] and dental amalgam [10], but much lower than those of all-ceramics [11] and alloys [12,13]. Substantial efforts have been undertaken to improve the problem, for instance, tailoring the filler size from micrometer to nanometer, increasing

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http://dx.doi.org/10.1016/j.dental.2016.09.003
0109-5641/

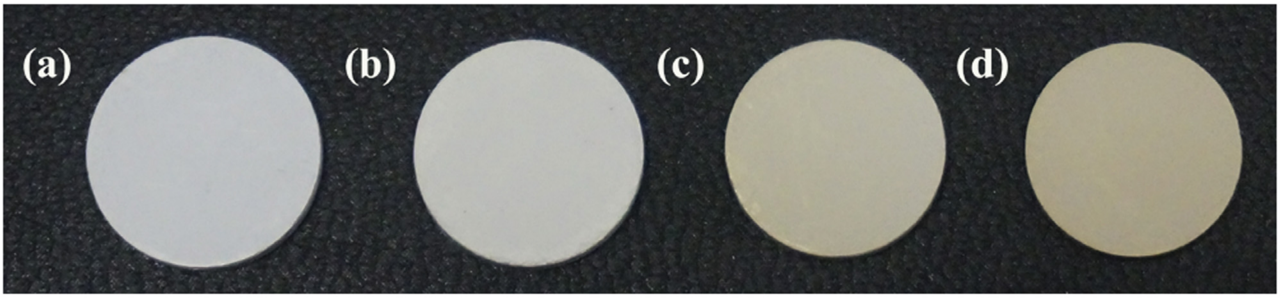

Fig. 1 – Photograph of polymer-infiltrated zirconia ceramics, (a) with 0 mol% Fe_2O_3 dopants, (b) with 0.1 mol% Fe_2O_3 dopants, (c) with 0.2 mol% Fe_2O_3 dopants, and (d) with 0.3 mol% Fe_2O_3 dopants.

filler contents or monomer molecular weight, and filling fibers or whiskers into dental resin [14–16]. But, it is seems that the problem persists, thus restricting the wide use of resin-based composites in the clinic. Because of their low strength and toughness, most of the composites are limited to the anterior area and some select posterior area.

As an attempt to overcome this problem, the polymer-infiltrated-ceramic-network material (PICN), containing primarily a porous ceramic structure with an interpenetrating resin phase, has emerged recently [17]. This novel composite combining features from both ceramics and resins is expected to exhibit high strength and toughness equivalent to ceramics, while retaining the esthetics and machinability of resin composites. However, the option of feldspar ceramic matrix for esthetic effects in the composite simply fails to make tremendous difference in mechanics. Commercial products, like Vita Enamic, have 103.36–159.88 MPa for flexural strength and 0.44–1.72 MPa $m^{1/2}$ for fracture toughness [17,18], which are similar to those of the resin-based composites.

It is well-known that zirconia-based ceramic plays a vital role in the all-ceramic system in clinic due to its excellent mechanical properties [19,20], but its application in the polymer-infiltrated-ceramic-network structure is rare, probably because of the characterized chalk color in zirconia ceramics. In order to simulating the chromaticity of natural dentin, the porous zirconia precursors are doped with different concentrations of Fe_2O_3, and it finds out that the slight content of Fe_2O_3 have positive effects on the mechanical performances of the specimens when the precursors are partly sintered at low temperature. The aim of this study is to evaluate effects of Fe_2O_3 content on the esthetic, microstructure and mechanical behaviors of polymer-infiltrated zirconia ceramics.

2. Materials and methods

2.1. *Synthesis of polymer-infiltrated zirconia ceramics*

Fe_2O_3 powder (Aldrich Chemical Co., China) was mixed with 3Y-TZP powder (Tosoh Co., Japan) at different concentrations, 0 mol%, 0.1 mol%, 0.2 mol% and 0.3 mol%, through ball-milling method. The particle size of Fe_2O_3 was 50 nm, and that of 3Y-TZP was 90 nm. The ethanol-based ball milling was for 12 h at a rate of 300 r/min. After drying, the mixtures were pressed at 4 MPa, followed by a cold isostatic pressing at 200 MPa. The initial blocks were partially sintered in air at 1150 °C, and the pre-sintered zirconia networks were conditioned with the silicone coupling agent prior to polymer infiltration. γ-MPS (Aldrich Chemical Co., China) dissolved in ethanol was employed as the silicon coupling agent. TEGDMA (Tri(ethylenglycol) dimethacrylate) (Aldrich Chemical Co., China) and Bis-GMA (Bisphenol A glycerolate dimethacrylate) (Aldrich Chemical Co., China) were uniformly blended at a ratio of 1:1 as the polymer matrix. Then, the initiation system, BPO (Benzoyl peroxide) (Aldrich Chemical Co., China), was added to the polymer matrix at a mass ratio of 1 wt.% to be blended for infiltration. Zirconia networks were immersed in the liquid polymer, forming two interpenetrating phases by capillary action. Then, the chromatic composites were obtained after polymerization by heat treatment at 70 °C for 10 h under the atmospheric pressure. Fig. 1 shows a photograph of specimens, which demonstrates the effect of Fe_2O_3 content on the chromaticity.

2.2. *Phase structure analysis*

X-ray diffraction spectroscopy (Rigaku D/MAX-2550V, Japan) was employed to analyze the phase composition. Lattice parameters of ceramic precursors were calculated based on the step scan profiles.

2.3. *Microstructure analysis*

The surface morphologies of specimens before and after infiltration, as well as the crack propagation path were examined by scanning election microscopy (Hitachi, S-2500N, Japan). The infiltrated polymer was identified by energy dispersive spectrometer (EDS). The relative density was measured by the Archimedes method. DSC reflecting enthalpy changes and TGA (Mettler Toledo STAR system) of specimens were performed to analyze effects of Fe_2O_3 on the densification and the resin content.

2.4. *Nano-indentation test*

A nano-indentation tester (MTS, XP, America) was applied to the analysis of the hardness and elastic modulus of specimens with a triangular pyramid Berkovich tip, and the indentation depth was fixed at 500 nm. 10 indentations per sample were collected randomly for each test. Values of hardness