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Preparation by sol-gel method and evaluation of lanthanum-containing filling glass ionomer cement powder

Xiao-ming Tu, Bao-hui Su*, Jun-guo Ran

College of Materials Science and Engineering, Sichuan University, 24N,Yihuan Rd, Chengdu, 610065 PR China

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ABSTRACT

To improve the mechanical properties of filling glass ionomer cements (GICs), lanthanum substitution for calcium in glass composition was investigated based on formula $2.3SiO_2-2.2Al_2O_3-0.5CaO-0.5CaF_2-0.08P_2O_5$ (BF). Compared with the commercial GIC, a series of lanthanum-containing glass powders were prepared via Sol-gel method. Compressive strength and Vickers hardness of experimental GICs were tested. XRD, SEM and EDAX were used to evaluate GIC powders and samples. In vitro biocompatibility was evaluated by methyl-tetrazolium (MTT) assay as well as the net setting time was tested. Incorporation of lanthanum in the glass powder could significantly improve GIC's mechanical properties. The all-lanthanum substitution (1La) GIC showed the highest Compressive strength (146.25 MPa), 47.7% and 41.98% higher than commercial and 0La (BF) group, and Vickers hardness (49.25 MPa), 39.2% and 28.02% higher ($P<0.05$), which was possibly due to the new stronger glass network forming, and leading to the improvement of curing reaction.1La and 0La GICs showed fine biocompatibility in initial cultivation stage. The net setting time of experimental GICs met the clinical requirements. The results demonstrated that lanthanum-containing filling GIC powder could be prepared successfully by Sol-gel method as well as could improve the GIC's mechanical strength effectively.

1. Introduction

Glass ionomer cement (GIC) has been successfully and widely applied in dentistry for several decades. Nevertheless, relatively insufficient mechanical properties have limited filling GIC's clinical applications especially for the restoration of posterior dentition [1]. The GIC conventional melt-quench preparation method involves some drawbacks such as uncontrolled fluorine losses leading to the variable compositions between batches and higher energy consumption, while the sol-gel method has more advantageous–lower synthesis temperatures to realize more stable compositions and better chemical homogeneity [2–4]. In recent times, there are some reports of using sol-gel method to prepare luting GIC glass powder [5,6]. Nevertheless, the preparation of filling glass powder by sol-gel method has not been reported yet. H.A.Abo-Mosallam et al. [7] showed that the influence of partial substitution of lanthanum for calcium in glass compositions by melt-quench method on glass transition temperature, setting time and crystallization. Other papers had reported the good biocompatibility of lanthanum and its addition could improve the chemical durability of some bio-glasses [8,9]. The purpose of this study is to investigate the influence on the mechanical strength of lanthanum-containing filling GIC prepared by Sol-gel route.

2. Experimental

A glass composition based on $2.3SiO_2-2.2Al_2O_3-0.5CaO-0.5CaF_2-0.08P_2O_5$ in molar ratio was selected as the basic formula (BF) [9]. The calcium element in the BF composition was set as 1, and derived from calcium nitrate(x). The lanthanum element was supplied with lanthanum nitrate (y) ($x+y=1$). Five different lanthanum groups (0La (BF), 0.25La, 0.5La, 0.75La, 1La) were prepared by the Sol-gel method. All the raw materials were analytical reagent.

Firstly, tetraethyl orthosilicate (TEOS) was hydrolyzed in ethanol under continuous stirring at room temperature for 1 h. Aluminum nitrate, calcium nitrate, lanthanum nitrate and ammonium dihydrogen phosphate were previously dissolved in distilled water. Then, they were added drop-wise to the hydrolyzed TEOS solution, as well as fluorosilicic acid was added into it. The mixed solution was then heated with continuous stirring to 80 °C for 4 h until gelation occurred. The gel was dried at 80 °C for 24 h, and then heated at 750 °C for 2 h using an electrical tube furnace to obtain the glass particles. The glass particles were pulverized and then passed through a 500 mesh sieve.

* Corresponding author.
E-mail address: sbh1004@163.com (B.-h. Su).

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X-ray diffraction (XRD, X-Pert TRO MPD, Philips) patterns of 6 glass powders were compared. Particle size distribution of glass particles was determined with laser diffraction (Mastersizer, Malvern). The glass powders were sputter coated with gold and examined by scanning electron microscope (SEM, S-3400 N, Hitachi) coupled with EDAX analysis (Ametek).

Cements were prepared by mixing with commercial liquid which is an aqueous solution including 45–50% (m/m) of polyacrylic acid and 5–10% (m/m) of tartaric acid (Dental Materials of Shanghai), at a powder to liquid (P/L) ratio of 1:1 by weight. The samples were kept in an incubator at relative humidity of approximately 95% and 37 °C for 24 h. Cylindrical specimens (ϕ4.0 mm, height 6.0 mm) were made to evaluate the Compressive strength (CS) using a universal testing machine (AG-IC 50kN, Shimadzu) at a crosshead speed of 0.5 mm/min, and the Vickers hardness (VH) (ϕ6.0 mm, thickness 3 mm) by the Digital Vickers hardness tester (HVS-50, Shanghai material test factory), the diamond indenter with 9.8 N load and a dwell time of 10 s. The morphology of fractured specimens was observed by SEM.

The net setting times (Ts) of the GICs were conducted by the method outlined in ISO 9917–1:2007.

In vitro biocompatibility of the prepared GICs was evaluated by methyl-tetrazolium (MTT) test using mice fibroblast cells (L929) with the blank control group setting [10–12]. An inverted fluorescence microscope (IX71, Olympus) was used to observe cell morphology. The optical density was measured at 570 nm (OD570) using an automated plate reader (Microplate Reader 3550, Bio-Rad).

Statistical differences of the mechanical properties and MTT data among groups were tested with one-way ANOVA and Tukey post-hoc test with a significance at $p=0.05$.

3. Results and discussion

The XRD patterns in Fig. 1A revealed similar amorphous phase with no any crystallization peak of 6 GIC powders. It indicated that lanthanum-containing-GIC powders were successfully prepared

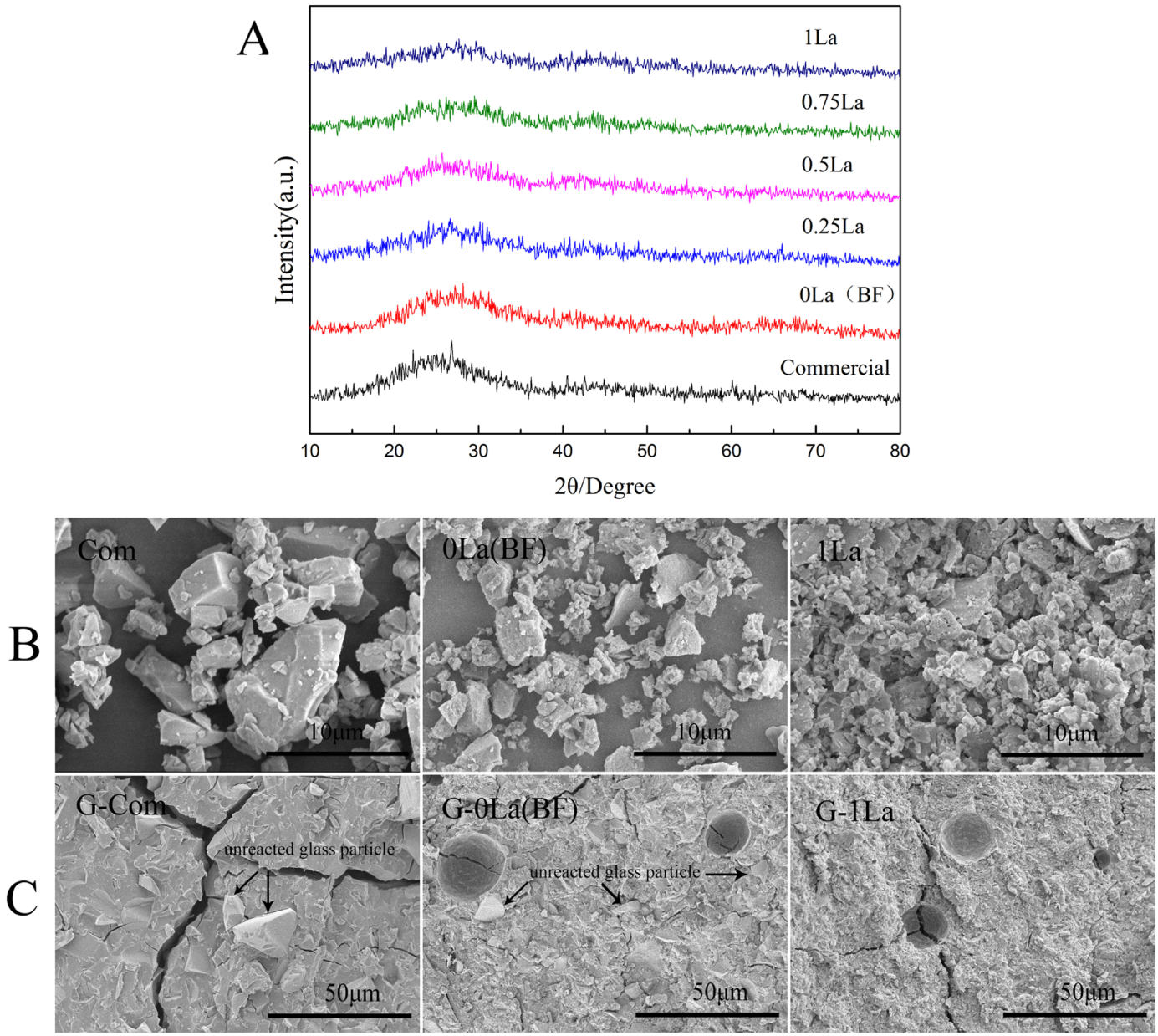

Fig. 1. XRD analysis of glass powders (A), Morphology of glass powders (B) and GIC fractured surfaces (C).

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