



The preparation of dental glass-ceramic composites with controlled fraction of leucite crystals

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Abstract

This work is dealing with synthesis of leucite powder, which can be used for the preparation of dental glass-ceramic composites by subsequent thermal treatment. Newly developed procedure is based on preparation of dental raw material as a mixture of two separate compounds: the crystalline leucite powder prepared at relatively low temperature and a commercial matrix powder.

Hydrothermal synthesis of tetragonal leucite particles ($KAlSi_3O_8$) with the average size of about 3 μm was developed in our laboratory. The leucite dental raw material was prepared by mixing of 20 wt.% of synthetic tetragonal leucite with commercial matrix. Dental composites were prepared from the dental raw material by uniaxial pressing and firing up to 960°C. Dilatometric measurements confirmed that the coefficient of thermal expansion increased by 32% when 20 wt.% of the tetragonal leucite was added into the basic matrix. In addition, it was showed that the synthesized leucite powder was suitable for the preparation of leucite composites with controlled coefficient of thermal expansion. High value of the thermal expansion coefficient enables application of prepared composite in metal-ceramics restorations.

Keywords: leucite, analcime, hydrothermal synthesis, dental materials

1. Introduction

Today, ceramics are one of the most common materials used in dentistry. Due to its esthetics, biocompatibility, strength, optical properties etc. ceramics fulfill all the requirements of a dental restoration. Dental ceramics can be distinguished in accordance with many criteria. In dependence on chemical composition and microstructure (resulting in different mechanical and optical properties) dental ceramics can be divided into three main classes: glass-ceramics (Vitadur[®] alpha, IPS Empress[®], Paradig[™] C), infiltrated (VITA In-Ceram) and polycrystalline (Lava[™]) ceramics.

Leucite glass-ceramic composites considered as aluminosilicate, consisting of amorphous and crystalline parts, were introduced into the dentistry as a material having higher coefficient of thermal expansion

compared to the ceramics without leucite crystals [1]. This has enabled better fusion of ceramics with metal reinforcement. Afterwards, it was suggested that leucite presented in glassy matrix could increase the hardness [2,3] and fracture toughness [4] of the composite. Rasmussen et al. [5] demonstrated stress induced phase transformation from metastable cubic form of leucite to tetragonal one. Further they proposed that the martensitic transformation could be responsible for increasing the fracture toughness by mechanisms similar to ZTC (Zirconium Toughened Ceramics) [6,7]. Nowadays, leucite glass-ceramics are used both for metal and all ceramics restoration.

Preparation procedures of leucite glass-ceramics were firstly made by a classical technology of dental porcelain based on a crystallisation from a melt. In this way, it was difficult to control final microstructure of a material, especially leucite particles size and their homogenous distribution in a glassy matrix [8,9]. At pres-

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ent, preparation of composite material, in which leucite and glassy matrix are synthesized separately, seems to be promising. This technology ensures reproducible control of leucite glass-ceramics microstructure, which is required to improve its fracture toughness and persistence.

To show that hydrothermally synthesized leucite powder is a suitable material for the preparation of leucite dental ceramics, following research was undertaken. Especially, this work was aimed at the preparation of dental glass-ceramic composites with controlled both the composition and the value of (its) thermal expansion coefficient by the addition of tetragonal leucite particles into a commercial leucite-free matrix.

II. Experimental

Hydrothermal synthesis of tetragonal leucite powder was carried out in two step-process: i) synthesis of analcime by hydrothermal treatment of an aluminosilicate solution and ii) transferring of analcime to leucite by subsequent ion-exchange reaction. Starting sols were obtained by mixing aluminate and silicate solutions, which were prepared separately by dissolving amorphous SiO_2 (Lach Ner s.r.o., Czech Republic) and aluminium powder (Lachema, Czech Republic) in 4M sodium hydroxide solution (NaOH, Penta Chrudim Czech Republic). After two hours of hydrothermal treatment, analcime was identified as a product of the reaction. Tetragonal leucite was synthesized from analcime by ion-exchange of Na^+ ions for K^+ ions in 4M solution of KCl [10,11]. The reactions were carried out in teflon-lined stainless steel autoclaves in an oven at 200°C . The products of the syntheses were washed in boiling distilled water, vacuum filtered and dried in an oven at 100°C .

Commercial leucite-free matrix, Vitadur® alpha, was used for preparation of the dental raw material consisted of 20 wt.% of the hydrothermally prepared tetragonal leucite and 80 wt.% of the matrix. The raw materials were mixed together for 2 hours by the addition of about 5 wt.% of a commercial deflocculant (Modeling fluid VITA). A cylindrical stainless steel mold and hydraulic press (30 MPa pressure) were used to form the porcelain discs (30 mm in diameter and 5.5 mm thick). The discs were then fired in electric furnace (Clasic, Revnice, Czech Republic) at 960°C for 7 min following the commercial heating/cooling schedule. Pure matrix was prepared as a reference sample under the same conditions as the composites.

Philips X'Pert PRO θ - θ powder diffractometer was used for X-ray powder diffraction analysis. The results for each specimen were analyzed using computer X'Pert High Score program. Average particle size was determined by optical (Jenapol, Zeiss, Germany) and electron (LEO VP 1450 instrument, Carl Zeiss AG, Oberkochen, Germany) microscope and laser method (FRITSCH Particle Sizer Analysette 22). The dilatometer (Adamel Lhomargy, Division d'Instruments s.a.) was used to measure the coefficient of thermal expansion (CTE) of samples. The specimens were heated with a constant heating rate $5^\circ\text{C}/\text{min}$ up to 700°C .

III. Results and Discussion

Powdered synthetic analcime and tetragonal leucite were identified using X-ray diffraction analysis. It was confirmed that the peak positions and intensities of hydrothermally synthesized products well corresponded to XRD databased cards of analcime and tetragonal leucite, Fig. 1.

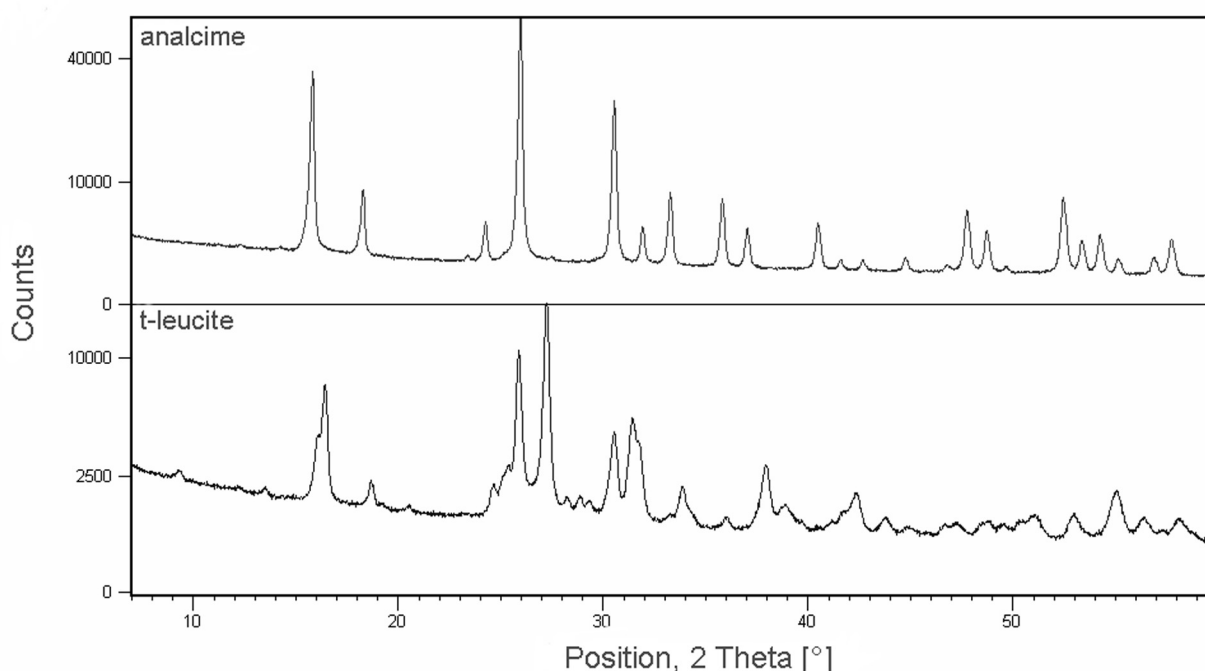


Figure 1. XRD patterns of synthetic analcime and tetragonal leucite

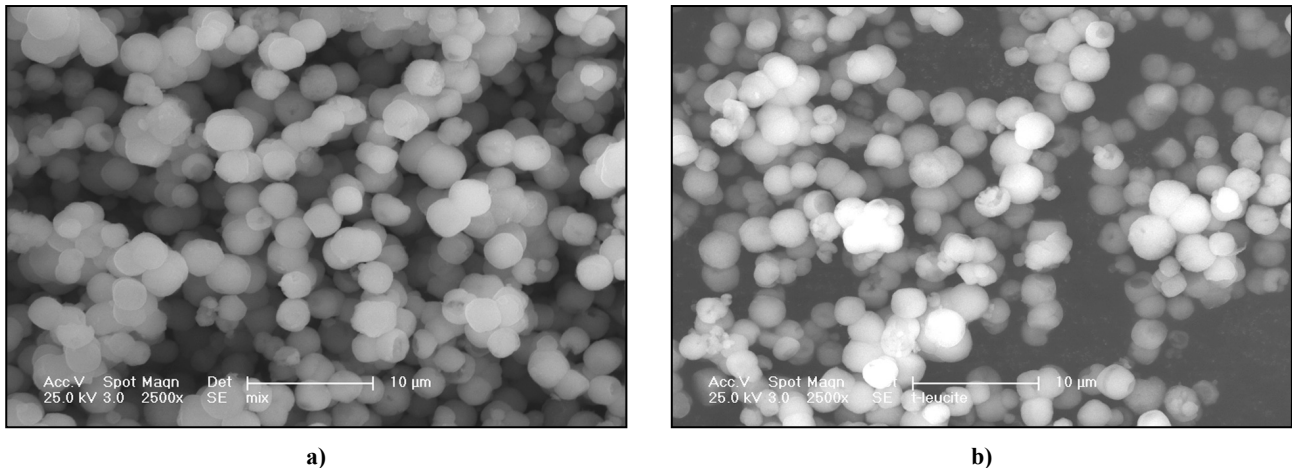


Figure 2. SEM images of hydrothermally synthesized analcime (a) and tetragonal leucite (b), at magnification 2500×

SEM studies showed that the particle size does not change within the ion exchange process from analcime to tetragonal leucite. The final tetragonal leucite grains are rounded polycrystalline particles having the average particle size of about 3 µm, Fig. 2. Rietveld refinement confirmed that these coarse particles consist of much smaller crystallites. The calculated size of crystallites was in the range 30–70 nm. XRD patterns of the pure matrix and composite consisting of 20 wt.% of tetragonal leucite and 80 wt.% of matrix, heat treated at 960°C, are presented in Fig. 3. It was proved that tetragonal leucite did not appear during the heating process in pure matrix. This demonstrated that the only crystalline phase in our composite originated from the hydrothermally synthesized leucite powder. Based on the previous results, it is possible to control the amount of tetragonal leucite easily using different ratio of leucite and matrix.

The dilatometric measurements confirmed the possibility to increase the coefficient of thermal expansion of the composite, which is important for metal-ceramics restorations. By the addition of 20 wt.% of tetragonal leucite CTE increased by 32 %, Fig. 4. In this way, it is possible to control the CTE of the composite using different ratio of leucite and the matrix.

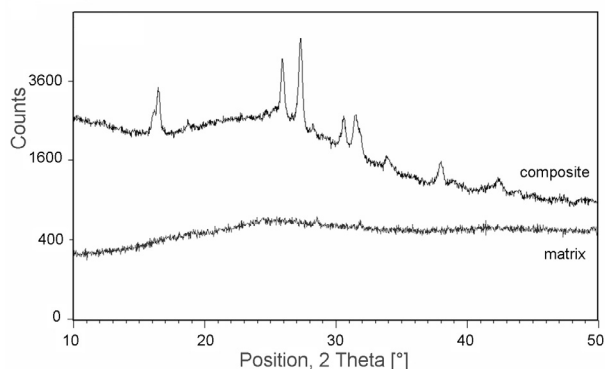


Figure 3. XRD patterns of pure matrix Vitadur® alpha and the composite consists of 20 wt. % of t-leucite and 80 wt. % of matrix after the heat treatment

IV. Conclusions

Presented work is dealing with the preparation of leucite glass-ceramic composites using leucite powder synthesized at low temperature. It was proved that the hydrothermally synthesized tetragonal leucite is suitable dental component for preparation of leucite dental composites. Different ratio of tetragonal leucite and matrix powders enables controlling of the CTE of the final composite. Dilatometric measurements showed that the coefficient of thermal expansion increased by 32 % when 20 wt.% of tetragonal leucite was added into matrix.

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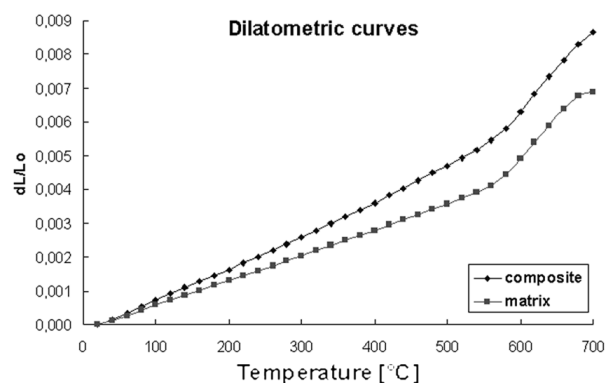


Figure 4. Dilatometric measurements of the composite and pure matrix

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