

Research Article

Synthesis of Bioactive Glass by Microwave Energy Irradiation and Its In-Vitro Biocompatibility

Swapan Kumar Sarkar and Byong Taek Lee

Department of Biomedical Engineering and Materials, Soonchunhyang University, 366-1, Ssangyuoung-dong, Cheonan, Chungnam 330-090, Korea
Address correspondence to Byong Taek Lee, lbt@sch.ac.kr

Received 14 January 2011; Accepted 3 February 2011

Abstract Bioactive glasses were synthesized in an inexpensive hydrothermal chemical route by the use of microwave energy irradiation. Compositional changes were carried out to synthesis bioactive glass with different Si content. Densification behavior was investigated by microwave sintering process from 700 °C to 1000 °C temperature. The powders were characterized for their physical and morphological characteristics like degree of crystallinity, crystallization temperature etc. Optimization of the heat treatment of the obtained amorphous powder was carried out. Biocompatibility test of the synthesized materials was carried out by in-vitro investigation by apatite formation behavior investigation by inserting the bioactive glass in simulated body fluid. Mechanical properties like density, compressive strength, hardness etc. were investigated for the dense bioactive glasses depending on the Si content. Apatite formation extent was investigated in SBF in 1 to 3 days.

Keywords bioactive glass; microwave energy assisted synthesis; SBF

1 Introduction

Bioactive materials for bone tissue regeneration and healing are now a day a promising area of research. Different kinds of bioactive materials spanning biopolymers, ceramics, glass ceramics, and glasses are being used for their unique features. The main constituent materials of bone tissue are hydroxyapatite nano crystal as the inorganic part and collagen protein as the organic part. Bone tissue regeneration is approached by providing the regeneration site with sufficient constituent materials so that the healing site gets suitable chemical environment for easy regeneration of the hard tissue. Usually calcium phosphate based ceramic materials like hydroxyapatite, TCP or BCP; which are close to the mineral constituent of bone are used, but lacked in quick resorption for rapid healing. Calcium

silicate based glass materials have been used for decades for damaged bone tissue healing. Bioactive glasses are a class of unique synthetic materials that react in the presence of body fluids, whereby they enhance the body's ability to regenerate tissue and heal it. This is accomplished by attracting essential biological elements produced by the body for healing and holding them in the defect site while natural healing occurs. These glasses are highly bioactive and their bioactivity has been demonstrated both in vitro and in vivo, with the formation of a strong bonding with the neighboring bone. Bioactive materials elicit a specific biological response at the interface of the material leading to the formation of a natural bond [3] and development of new mineralized bone tissue.

A few methods have been developed to prepare the bioglasses. Melting method is the traditional one for glass preparation [1,6]. It is simple and suitable for massive production. However, during the high temperature stage, the volatile component P_2O_5 tends to escape. In recent years, sol-gel method has attracted many studies [5]. The sol-gel process has the advantage of low reaction temperature. Compared with the melting method, the sol-gel derived glass powder has homogenous composition in particles. A group of sol-gel derived bioglasses in the system of SiO_2 -CaO- P_2O_5 has been widely studied, such as 55S, 58S, 60S, etc. Their excellent bioactivity offers great potential for hard tissue surgery [5]. But the precursor material for synthesis by sol-gel is expensive. For low cost bio-active glass we need a less expensive chemical route for synthesis. Recently ultrasonic assisted synthesis and microwave assisted synthesis is gaining attention as they can help to reaction in a short time and can modify the reaction environment to produce nano phase powders [2,4].

In this work we investigated the synthesis of bio active glass by the microwave energy assisted hydrothermal method coupled with ultrasonic energy assistance for a rapid and low cost powder synthesis method.

2 Materials and methods

The raw materials were calcium nitrate tetra hydrate for Ca source, diammonium hydrogen phosphate for P source and sodium silicate solution (as 37% SiO_2 in NaOH solution) for Si source. The precursors were dissolved in de-ionized water and transferred to the ultrasonic bath. The irradiation time was varied to obtain the optimum synthesis condition. Microwave operation was performed in a second batch of powders after the ultrasonic irradiation. The obtained amorphous powder was washed in de-ionized water and filtered. After drying for 24 hours in oven at 80°C the powders were calcined at 700°C temperatures for the development of bioglass. The synthesis procedure is shown in Figure 1. Both pressureless and microwave sintering was used for making dense compacts and the resulting variation of the microstructure was investigated. Phase analysis and microstructure were investigated by XRD and SEM. The biocompatibility of the powder was investigated by immersing the dense bioglass body to SBF solution for 1 day and 3 day and the resulting apatite formation was investigated.

3 Results and discussion

Bioactive materials can elicit a specific biological response at the tissue/materials interface which results in the formation of a bond between the tissues and the materials. When bioactive materials are implanted into human body, it will interact with the surrounding bone or other tissue to some extent. An ion-exchange reaction between the bioactive implant and surrounding body fluids results in the formation of a biologically active carbonate-containing hydroxyapatite layer on the implant that is chemically and crystallographically equivalent to the mineral phase in bone, which promotes bonding between the natural tissues and the material. In the ultrasonic and microwave assisted methods the as received powders appeared to be amorphous. Calcination of the powders are carried out at 700°C and found that the powders remain mostly amorphous except a slight appearance of CaSiO_3 . Figure 2 showed the XRD profiles of the synthesized bio-glass powders calcined at 700°C synthesized at different synthesis condition. All of the powders retained their amorphous phase and tinny peak or CaSiO_3 was observed irrespective of the synthesis rout.

Figure 3 shows the fracture surface of pressureless and microwave sintered bio-glass sintered at 1000°C and 1100°C . We can see a lot of micro-pores on the surface which showed pore densification. At 1100°C the densification is better than 1000°C as shown in Figure 3 (b). However, the microwave sintered compacts showed better sintering behavior compared to the pressureless sintered bio-glass as depicted in Figures 3(c), 3(d). EDS profile was taken from the bio-glass compacts to investigate the

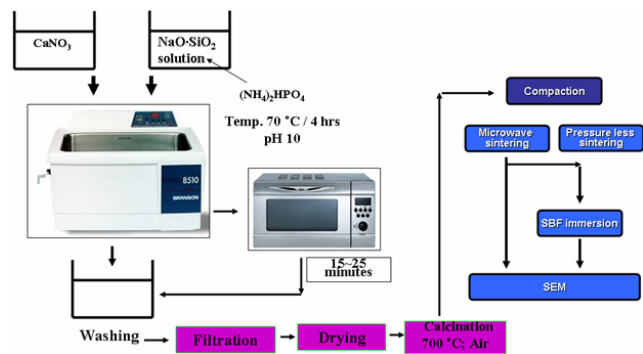


Figure 1: Schematic diagram of bio-glass synthesis and characterization.

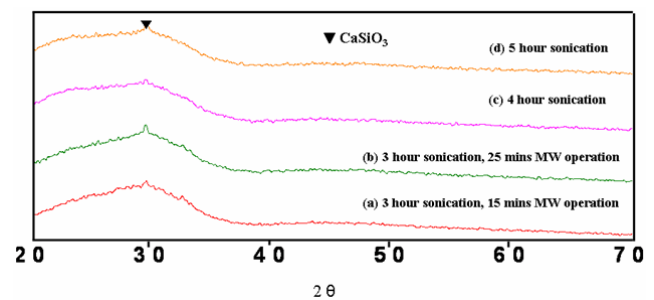


Figure 2: XRD profiles of bio-glass powder calcined at 700°C obtained from different synthesis conditions.

elemental presence and showed that it is consistent with the bio-glass composition.

To investigate the biological response on the bio-glass after implantation and formation of apatite we immersed the sintered bio-glass compacts in simulated body fluid (SBF) for 1 and 3 days and investigated by SEM. After one day the apatite formation was not prolific and only appeared covering the surface very thinly. This is was true for both pressure-less and microwave sintered bio-glass compacts. However, after 3 days of SBF immersion the compact surface was fully covered with the biological apatite in both pressure-less sintered and microwave sintered compacts. As shown in Figure 4. However, the formed apatite microstructure was different depending on the sintering method. From Figures 4(a), 4(b) we can see that at 1000°C sintering temperature apatite formation was prolific and surface growth sites were evident. In case of sintering at 1100°C the surface showed more intense apatite formation, whether sintered by pressure-less sintering or microwave sintering. For pressureless sintered bio-glass compacts the growth sites were around $1\ \mu\text{m}$ and very fine nano-fiber like offshoots were directed in all direction from the surface. In the microwave sintered compact the growth sites were homogenized as intense apatite formation occurred and the grains developed during sintering were smaller. The

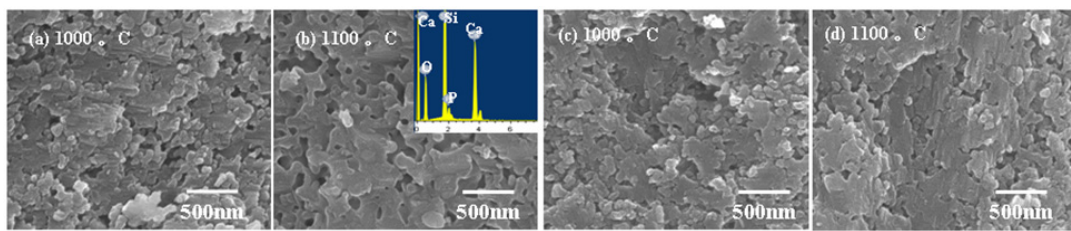


Figure 3: Fracture surface of pressureless sintered bio-glass compacts (a, b) and of microwave sintered bio-glass compacts (c, d).

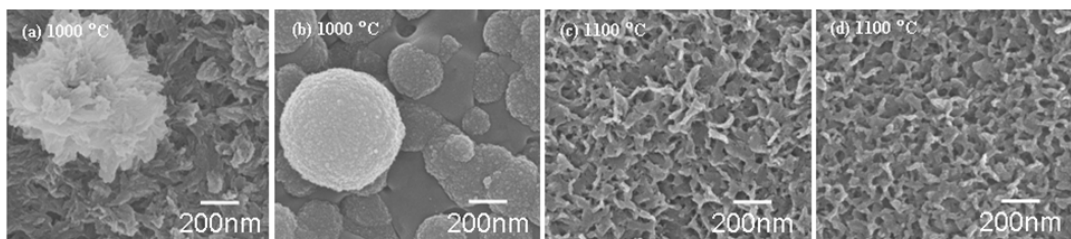


Figure 4: SEM of pressureless sintered bio-glass (a, c) and microwave sintered bio-glass (b, d) after 1 day and 3 day of SBF immersion, respectively.

nano-fiber like offshoot was finer than that of the pressureless sintered counterpart. But the densification was clearly higher in this case. This implies that the microwave sintered compacts would show better apatite densification.

However, both the apatite surfaces, showed cracks. This was because of the thin film nature of the apatite for a short formation time and during sample handling like blotting, drying etc. However, these cracks are expected to be eliminated with higher thickness of apatite for prolonged SBF immersion mimicking the actual in-vivo condition.

4 Conclusions

Bio-active glass powder was synthesized by microwave energy assisted method coupled with ultrasonic energy assistance. The powder produced was amorphous, which after calcining at 700 °C retained its glass phase. Dense compacts were made by sintering at 1000 °C and 1100 °C by conventional pressureless sintering furnace and microwave furnace. The SBF response was good and after 3 days of immersion vigorous apatite formation was observed on surface of the compacts.

References

- [1] K. Franks, I. Abrahams, G. Georgiou, and J. C. Knowles, *Investigation of thermal parameters and crystallisation in a ternary CaO-Na₂O-P₂O₅-based glass system*, Biomater, 22 (2001), 497–501.
- [2] J.-K. Han, H.-Y. Song, F. Saito, and B.-T. Lee, *Synthesis of high purity nano-sized hydroxyapatite powder by microwave-hydrothermal method*, Materials Chemistry and Physics, 99 (2006), 235–239.
- [3] L. L. Hench, R. J. Splinter, W. C. Allen, and T. K. Greenlee, *Bonding mechanisms at the interface of ceramic prosthetic materials*, J Biomed Mater Res, 5 (1971), 117–141.
- [4] W. Kim and F. Saito, *Sonochemical synthesis of hydroxyapatite from H₃PO₄ solution with Ca(OH)₂*, Ultrasonics Sonochemistry, 8 (2001), 85–88.
- [5] A. Rámila and M. Vallet-Regí, *Static and dynamic in vitro study of a sol-gel glass bioactivity*, Biomaterials, 22 (2001), 2301–2306.
- [6] J. D. Santos, P. L. Silva, J. C. Knowles, S. Talal, and F. J. Montero, *Reinforcement of hydroxyapatite by adding P₂O₅-CaO glasses with Na₂O, K₂O and MgO*, J Mater Sci Mater Med, 7 (1996), 187–189.