

Influence of Commercial Inert Glass Addition on the Mechanical Properties of Commercial Synthetic Hydroxyapatite

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In this study, microstructures and mechanical properties of commercial synthetic hydroxyapatite (CSHA)–commercial inert glass (CIG) composites were investigated. The goal of development of CSHA–CIG composite is to improve mechanical properties of hydroxyapatite. For this reason, CSHA powders were mixed with 5 and 10 wt% CIG separately. Pressed samples were sintered at the different temperatures in the range of 1000–1300 °C. The physical and mechanical properties were determined by measuring density, compression strength, the Vickers microhardness. Structural characterization was carried out with X-ray diffraction and scanning electron microscopy studies. The experimental results were discussed to determine optimum amount of reinforcement material and the effect of sintering temperature on the microstructure and the mechanical properties of CSHA–CIG composites. The mechanical properties of composites decreased with increasing CIG content. The highest mechanical properties and the highest density were obtained in CSHA–5 wt% CIG composite sintered at 1300 °C.

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1. Introduction

Hydroxyapatite (HA) is one of most important market valued candidate biomaterial which is still in use since decades in medicine and dentistry applications. HA with chemical formula $(\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2)$, assembling the main mineral components of bones and teeth, is among the leading biomaterials satisfying these requirements [1]. Synthetic HA has excellent biocompatibility due to its chemical stability with the mineral portions of hard tissues as well as its ability to constitute chemical bonding with surrounding tissues [2]. But still HA as a bulk material is not suitable for loaded situations due to its very brittle character [3]. For this reason, the main weakness of this material lies in its poor mechanical strength which makes it unsuitable for load-bearing applications [4, 5]. HA material must be reinforced with other materials to form a durable and more load resistible composite. For this reason, HA bioceramics are reinforced with other ceramics or metals in the form of powders, platelets, or fibers belonging to this type of material. Addition of second-phase ceramic materials (e.g., zirconia, titania, or alumina) to the HA matrix in order to obtain products with enhanced strength and toughness has been researched in recent years [4, 6].

Glass compositions would be a good option for doping apatite matrix. Goller et al. had used HA–bioglass (45S5) composites. The HA used was human teeth derived HA [7]. Batra et al. had used phosphate glass (CaO – P_2O_5 – Na_2O – CaF_2 based glasses) and density, compressive strength and hardness increased with increasing bioglass content [8]. Salman et al. had used commercial inert glass (CIG) to make HA composites. The HA

used was bovine hydroxyapatite [9]. They have shown that glass additions can work as reinforcing phase for HA bioceramic. To produce CIG no special sources are needed. Simple use of some broken window glass is far enough. But we must be very careful not to use crystal ash trays, because those glasses contain lead. Addition of lead makes easier the production of household crystals. Those are produced with simply machining and we must not forget that those are twice heavier than normal glasses (i.e. CIG). But broken boroglasses could be also easily used. According to a recent study, Valério et al. had used B_2O_3 glasses for cell culture. At the cell culture study impressive results for cell culturing were obtained [10]. Some US patents also result in boron containing bioglass composites. Using boron will decrease the melting temperature of bioglass compositions from 1450 °C to 900 °C [11, 12]. This dramatic decrease in the temperature makes the production of such components very economic. It is generally considered that the presence of boron in glass composition assists and even improves retentions of calcium, magnesium, and phosphorus [13]. According to Behnamghader et al. HA composite with 50 mol.% CaO and 50 mol.% P_2O_5 containing glass decomposes to α -tricalcium phosphate (TCP) at 1300 °C sintering temperature [14].

The aim of this study is to produce bioceramics composites from CIG and synthetic HA to improve mechanical strength of HA.

2. Materials and methods

The commercial synthetic hydroxyapatite (CSHA) used in this study was obtained from Across company

with average particle size of $6.5 \mu\text{m}$ and molecular weight of 502.31 g/mol . Table I shows the chemical analyses of CIG with average particle size of $68 \mu\text{m}$. CSHA powder was mixed with (separately) 5 and 10 wt% CIG powder for 4 h. The samples were prepared according to a British Standard for compression tests (BS 7253) [15]. The powder portions were pressed at 350 MPa between hardened steel dies. Pressed samples were sintered between 1000°C and 1300°C (with the heating rate of $+5^\circ\text{C min}^{-1}$) for 4 h. Compression strength, the Vickers microhardness as well as density were measured. Scanning electron microscopy (SEM) analyses were done after etching with 3 vol.% HF solution for 1 min. X-ray diffraction studies were also conducted. The compression tests were done with an universal testing machine (Shimadzu) at the crosshead speed of 3 mm/min . Microhardness values were determined under 200 g load. Scanning Electron Microscope (Hitachi TM-1000) was used for microscopic examinations and XRD analyses were conducted on a Bruker D8-Advanced X-ray diffractometer.

TABLE I

Chemical analyses of CIG.

Oxide	wt%
SiO_2	68.80
Na_2O	17.02
CaO	9.25
MgO	1.77
Fe_2O_2	0.084
TiO_2	0.017
Al_2O_2	2.15
Cr_2O_2	0.012
CuO	0.0036
Other	trace

3. Results

Table I shows the experimental results of density, compression strength and the Vickers microhardness of the samples sintered at different temperatures with change in reinforcement content. The Vickers microhardness values of CSHA–CIG composites increase with increasing sintering temperature, as seen in Table II. However, density and compression strength values of CSHA–CIG composites increase with increasing sintering temperature except CSHA–10 wt% CIG composites sintered at 1300°C .

Figure 1 presents the comparison of compression strengths and the Vickers microhardness of CSHA–CIG composites sintered at the different sintering temperatures. The mechanical properties of composites decreased with increasing CIG content. The highest mechanical properties and the highest density were obtained in CSHA–5 wt% CIG composite sintered at 1300°C .

The microstructures of CSHA–CIG composites sintered at different temperatures are given in Fig. 2. It is clearly seen that similar microstructures were observed

TABLE II

Influence of CIG content and sintering temperature on density, compression strength and Vickers microhardness of composites made of CIG and CSHA.

Temperature [$^\circ\text{C}$]	Density [g/cm^3]		Compression strength [MPa]		Vickers microhardness [HV]	
	5 wt%	10 wt%	5 wt%	10 wt%	5 wt%	10 wt%
1000	2.42	2.18	60	53	128	115
1100	2.65	1.54	72	66	278	202
1200	2.93	2.70	82	75	358	285
1300	2.99	1.88	95	18	382	312

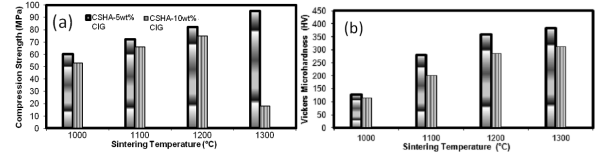


Fig. 1. (a) Compression strength, (b) microhardness of CSHA–CIG composites at different sintering temperatures.

in composites containing 5 and 10 wt% CIG sintered at 1000°C (Fig. 2a,c).

Light coloured phases are sodium calcium aluminium phosphate silicate (SCAPS) in Fig. 2b. These phases satisfy improved mechanical properties with 5 wt% CIG addition of composites sintered at 1300°C . Figure 2d shows the CSHA–10 wt% CIG composite sintered at 1300°C . If the CIG content exceeds a certain limit, the resultant composite material shows vitrification of samples and formation of porosity, which evidently resulted in dramatic reduction of the mechanical properties (Fig. 2d). However, the effect of high glass content is not detrimental at lower temperature (Fig. 2c).

Figure 3 shows the XRD patterns of the CSHA–5

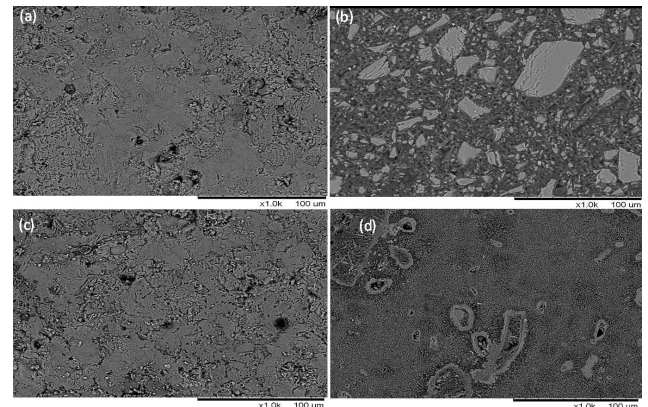


Fig. 2. Microstructures of CSHA–CIG (5 and 10 wt%) composites sintered at different temperatures. (a) CSHA–5 wt% CIG at 1000°C , (b) CSHA–5 wt% CIG at 1300°C , (c) CSHA–10 wt% CIG at 1000°C , (d) CSHA–10 wt% CIG at 1300°C .

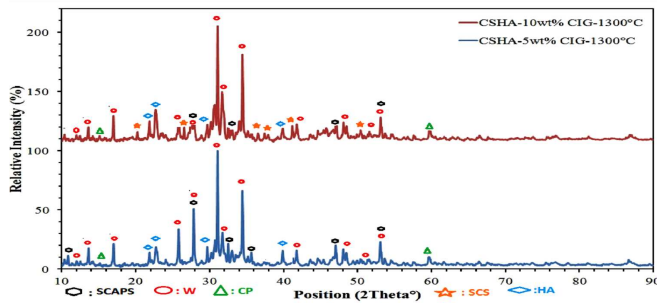


Fig. 3. X-ray diffraction studies of 5 and 10 wt% CIG added CSHA–CIG composites, sintered at 1300 °C.

and 10 wt% CIG composites sintered at 1300 °C. Both composites include whitlockite (W), calcium phosphate (CP), hydroxyapatite (HA), and SCAPS phases. CSHA–5 wt% CIG composites sintered at 1300 °C include more evident and sharper SCAPS peaks. Probably this phase causes to improve mechanical properties. However, CSHA–10 wt% CIG composite includes extra sodium calcium silicate phase. This phase causes to form glassy phase and decrease density and mechanical properties of composite.

4. Conclusions

The findings of this study are concluded as follows:

The Vickers microhardness values of CSHA–CIG composites increase with increasing sintering temperature. However, density and compression strength values of CSHA–CIG composites increase with increasing sintering temperature except CSHA–10 wt% CIG composites sintered at 1300 °C.

The mechanical properties of composites decreased with increasing CIG content.

The highest mechanical properties and the highest density were obtained in CSHA–5 wt% CIG composite sintered at 1300 °C.

Sodium calcium silicate phase and secondary porosity cause to decrease compression strength and density of CSHA–10 wt% CIG composites sintered at 1300 °C.

Biocompatibility studies are going on.

References

- [1] N. Demirkol, E.S. Kayali, M. Yetmez, F.N. Oktar, S. Agathopoulos, *Key Eng. Mater.* **484**, 204 (2011).
- [2] S. Ramesh, P. Christopher, C.Y. Tan, W.D. Teng, *Biomed. Eng.-Appl. Basis Commun.* **16**, 199 (2004).
- [3] F.N. Oktar, S. Agathopoulos, L.S. Ozyegin, O. Gunduz, N. Demirkol, Y. Bozkurt, S. Salman, *J. Mater. Sci., Mater. Med.* **18**, 2137 (2007).
- [4] V.V. Silva, F.S. Lameiras, R.Z. Domingues, *Composit. Sci. Technol.* **61**, 301 (2001).
- [5] J.W. Choi, Y.M. Kong, H.E. Kim, I.S. Lee, *J. Am. Ceram. Soc.* **81**, 1743 (1998).
- [6] E. Fidancevska, G. Ruseska, J. Bossert, Y.M. Lin, A.R. Boccaccini, *Mater. Chem. Phys.* **103**, 95 (2007).
- [7] G. Goller, F.N. Oktar, H. Demirkiran, E. Demirkesen, *Key Eng. Mater.* **240-242**, 939 (2003).
- [8] U. Batra, S. Kapoor, *Int. J. Chem. Biol. Eng.* **3**, 45 (2010).
- [9] S. Salman, F.N. Oktar, O. Gunduz, S. Agathopoulos, M.L. Öveçoğlu, E.S. Kayali, *Key Eng. Mater.* **330-332**, 189 (2007).
- [10] P. Valério, A.M. Góes, U. Karacayli, O. Gunduz, S. Salman, A. Zeki Sengil, S. Yilmaz, S. Agathopoulos, F. Nuzhet Oktar, in: *Proc. 1st Int. Conf. on Biomedical Engineering 2009*, Eds. R.M. Natal Jorge, M.A.P. Vaz, J.M.R.S. Tavares, CRC, Boca Raton 2010, p. 99.
- [11] L.L. Hench, P.J. Buscemi, *Bioglass coated metal substrate*, United States Patent, Patent date & number: Nov. 25, (1980), 4, 234, 972.
- [12] L.L. Hench, D.C. Greenspan, *Bioglass coated Al₂O₃*, United States Patent, Patent date & number: Jul. 25, (1978), 4, 103, 002.
- [13] O. Gunduz, L.S. Ozyegin, S. Dorozhkin, O. Meydanoglu, N. Eruslu, S. Kayali, S. Agathopoulos, F.N. Oktar, *Key Eng. Mater.* **396-398**, 403 (2009).
- [14] A. Behnamghader, N. Bagheri, B. Raissi, F. Moztarzadeh, *J. Mater. Sci. Mater. Med.* **19**, 197 (2008).
- [15] *British Standard Non-Metallic Materials for Surgical Implants. Part 2 Specification for ceramic materials based on alumina, BS 7253:Part 2: 1990 ISO 6474-1981.*

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