MECHANICAL PROPERTIES OF HYDROXYLAPATITE SPECIMENS OBTAINED BY COLD ISOSTATIC PRESSING

G. Ya. Akimov, V. M. Timchenko, and P. A. Arsen'ev


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The authors investigate how the density and mechanical properties of green and sintered hydroxylapatite specimens depend on the pressure of cold isostatic pressing (CIP). It is shown that as the CIP pressure grows, the density, the ultimate bending strength, and the fracture toughness in green specimens increase to reach 2.70 g/cm$^3$, 50 MPa, and 0.8 MPa $\cdot$ m$^{1/2}$, respectively, at 2.0 GPa. The maximum value of the ultimate bending strength of the sintered material is 120 MPa at a density of 3.02 g/cm$^3$. A qualitative explanation of the observed phenomena is suggested.

Calcium hydroxylapatite (HA), whose chemical formula is $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$, is the principal inorganic component of bone tissue, which makes HA-based materials and other calcium phosphates bioactive, i.e., capable of chemical interaction and bone substitution. Therefore, HA-based ceramics can be distinguished among other materials used in stomatology and orthopedics.

Other investigators before us have made experimental attempts to obtain HA-based ceramics with high physical and mechanical properties using hot isostatic pressing [1], thermoplastic casting followed by sintering [2], and explosive compacting [3]. In this work we tried to solve the problem using cold isostatic pressing (CIP) followed by free sintering.

CIP allows us to obtain a high density uniformly distributed over a specimen. As the CIP pressure in the molds increases, the grains obviously become closer, which leads to their plastic deformation and destruction. With further growth of the pressure separate particles that formed the grains lose their shape and break down. This process may be accompanied by the appearance of dislocations and their multiplication [4]. The resultant high-density compact consists of activated particles or grains. We can then sinter CIP-treated specimens at lower temperatures and in less time. The density of ceramics obtained in this way is usually very high and close to the theoretical value, which provides high mechanical characteristics.

The results presented in this paper concern the effect of CIP pressure on the physical and mechanical properties of green compacts (obtained after CIP) and sintered HA-based ceramics.

Specimens were prepared from HA powders synthesized at the Moscow Institute for Power Engineering (MEI). We investigated powders of different compositions, namely, a pure HA and an HA with approximately 1% of $\text{Al}_2\text{O}_3$ and $\text{SiO}_2$ admixtures. The initial components were mixed by joint grinding in a planetary mill in ethanol with $\text{Al}_2\text{O}_3$ grinding bodies. After drying, the powders were uniaxially pressed in a steel mold to form specimens of size $6 \times 6 \times 42$ mm, which were then subjected to CIP at pressures up to 2.0 GPa and sintered at 1370$^\circ$C for 6 h. We have determined the density, strength, and fracture toughness of the green and sintered materials. The density was found by hydrostatic weighing, the strength was tested by three-point bending of a specimen, and the fracture toughness by three-point bending of a notched specimen. The mechanical testing was carried out at room temperature with loading of a specimen at a rate of 0.5 mm/min. The strength data represent the values averaged over 2–4 measurements.

The density and mechanical properties of the green compact are given in Fig. 1. In both batches the density of the specimen is identical (see Fig. 1a), but by introducing a small amount of an impurity we noticeably changed the mechanical properties (see Figs. 1b, c).

The nature of the dependence of the green density on the CIP pressure and the analysis of the strength and fracture toughness behavior imply that green compacting in CIP occurs in three stages. The first stage occurs at pressures less than 0.6 GPa. In this pressure range the density grows rapidly and reaches 2.30 g/cm$^3$ at $p = 0.6$ GPa, while the strength
and fracture toughness increase monotonically. It should be noted that 2.30 g/cm³ constitutes 73% of the theoretical density (3.15 g/cm³). Furthermore, we know that a volume is closely packed with spheres to 73 – 74%, i.e., this is the extend to which hexagonal close-packed and cubic face-centered crystal lattices are filled with atoms. This means that at this stage the compacting mainly occurs due to a close and dense arrangement of the grains that form the dense frame. As the density and the number of contacts between individual grains increase, the strength and fracture toughness in this pressure range grow practically linearly.

The next compacting stage occurs within $P = 0.6 - 1.3$ GPa. The density grows much more slowly because the grains have already been arranged closely. It seems that at this stage compacting is possible because the grains are deformed and start to fracture. Nonetheless, a growing fracture is arrested by fractures formed earlier and therefore the minimum strength corresponds to the maximum fracture toughness. As the pressure grows further, the grains are broken into particles, which increases the strength and decreases the fracture toughness.

Specimens of pure HA are destroyed because the elastic and mechanical properties along different crystallographic directions are different (the effect is known as compression anisotropy of the first kind) [5]. Specimens with $\text{Al}_2\text{O}_3$ and $\text{SiO}_2$ impurities are destroyed due to the different properties of the impurity phases and the HA (compression anisotropy of the second kind). It is obvious that the second-kind effect is manifested much more strongly than the first-kind one and therefore specimens of pure HA lose much less strength than HA specimens with impurities, and no fracture toughness maximum is observed at all. The fact that the strength maximum in HA specimens with $\text{SiO}_2$ and $\text{Al}_2\text{O}_3$ impurities is shifted to higher pressures can be explained by the fact that the grain frame contains stronger components, which break at higher pressures.

The third compacting stage occurs up to a pressure of 2.0 GPa. It is characterized by rapid growth of the density accompanied by sharply increasing strength and fracture toughness. By analogy with the first stage we ascribe these effects to a closer arrangement of granules and separate particles. The volume of such a polydisperse system is packed to a very high degree, which explains the rapid compacting process. The substantial gain in strength is due to the higher density and smaller size of fractured particles.

It is possible that by using higher CIP pressures we will observe a fourth stage analogous to the second one, in which particles and agglomerates will be arranged still closer and be deformed with further growth of the strength and fracture toughness. Clearly, this picture of the processes occurring in CIP is rather hypothetical and requires a detailed structural investigation.

Figure 2 presents the measured characteristics of sintered ceramics. The density of the sintered material of pure HA decreases linearly with increase in the CIP pressure. The sintered specimens were partially fractured and therefore the relationship between the pressure and the strength is represented by two curves (the dashed line in Fig. 2b corresponds to fractured specimens). The strength of specimens of pure