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Fracture toughness of an α-Al_2O_3 ceramic for joint prostheses under sinter and sinter-HIP conditions

M.H. Bocanegra-Bernal [a,*], C. Domínguez-Rios [a], A. Garcia-Reyes [b], A. Aguilar-Elguezabal [a], J. Echeberria [c], A. Nevarez-Rascon [d]

[a] Centro de Investigación en Materiales Avanzados, CIMAV S.C., Miguel de Cervantes # 120 Complejo Industrial Chihuahua 31109, Mexico
[b] Interceramic, Depto. de Investigacion y Desarrollo, Av. Carlos Pacheco 7200, Chihuahua 31060, Mexico
[c] CEIT and TECNUN (University of Navarra), 20018 San Sebastian, Spain
[d] Escuela de Odontologia, Universidad Autonoma de Chihuahua, Ciudad Universitaria, Campus 1, CP 31000, Mexico

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ABSTRACT

The densification of submicron alumina powders by sinter and sinter-HIP to obtain specimens with submicron grain size has been investigated. The minimum temperature to obtain close porosity has been determined by pressureless sintering in air. Green compacts were obtained by uniaxial pressing at compaction pressures of 50–150 MPa with relative densities of 54% TD. Temperature of 1350 °C and pressures of 150 MPa were used to obtain relative densities >98% by sinter-HIP. Resulting microstructures were observed from polished surfaces by scanning electron microscopy (SEM). Submicron grain size was obtained in pressureless and sinter-HIPed samples. The cracks induced by Vickers indentations showed a more tortuous path in sinter-HIPed samples which lead to higher fracture toughness compared with only pressureless sintered samples. The maximum Vickers hardness (HV) and fracture toughness values reached 19 GPa and 5.2 MPa $m^{1/2}$, respectively, with the sinter-HIP treatment. The observed crack deflection was an important mechanism in improving fracture toughness in sinter-HIPed samples. On the other side, the grain size and remain porosity seem to be responsible to obtain this high hardness and fracture toughness.

1. Introduction

The benefit of ceramic microstructures with submicrometer grain sizes in order to obtain components with improved hardness, wear resistance, strength, good biocompatibility and excellent corrosion resistance is well known [1–6]. Alumina has extensively been used in total hip arthroplasty (THA) and total knee arthroplasty (TKA) as an alternative to replace the cobalt–chromium (CoCr) alloy articulating against UHMWPE (Ultra-high-molecular-weight polyethylene). The estimated lifetime of prosthetic devices manufactured with CoCr-UHMWPE bearings is approximately 10–15 years and approximately 25% of total hip and knee joint replacement required another surgery to substitute the joint as a consequence of premature failure of the prosthetic joints by aseptic loosening [7,8]. Considering the growing demand on performing orthopedic surgery on younger patients, the aluminium oxide ceramic implants should be designed for long-term applications and therefore should exhibit a lifetime of more than 30 years and the fracture *in vivo* is not accepted [9].

Aluminium oxide (Al_2O_3) was introduced as a material for orthopedic bearings in the 1970s and it is by far, the most widely used ceramic material in both THA and TKA. This ceramic is the cheapest and its hardness is 9 on Mohs' scale which ranks the hardness of diamond at 10 as the hardest existing material [10]. On the other hand, the biocompatibility of Al_2O_3 ceramics is related to the high chemical stability which confers resistance to corrosion and reliable *in vivo* behavior over time [11,12]. Al_2O_3 ceramic bearings today have improved mechanical and physical properties, wear characteristics, biocompatibility, and reliability when is compared with earlier bearings of the same material. Vickers hardness (HV) between 17 and 20 GPa, grain size (μm) ranges 1.8–4.5, bending strength (MPa) values of 400–580 for Alumina 1970s, 1980s, and 1990s, respectively, have been reported by Willmann [13]. Modern Al_2O_3 implants are nowadays manufactured with additions of MgO as dopant to control the grain growth and hot isostatic pressing (HIP) at temperatures of approximately 1250 °C has been used to obtain an almost fully dense material with a grain size smaller than ~2 μm as was reported by De Aza et al. [9], Hench and Wilson [14], and Ratner et al. [15].

The old generation of alumina ceramic implants was manufactured by conventional sintering which increased the severity of flaws already present in the green compact. Considering the low

* Corresponding author. Tel.: +52 614 439 11 11; fax: +52 614 439 11 12.
E-mail address: miguel.bocanegra@cimav.edu.mx (M.H. Bocanegra-Bernal).

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fracture toughness of ceramics in general, Al_2O_3 material improvements are related to their long-term reliability minimizing the presence of strength-limiting flaws in the fabricated material, improving the fracture toughness. By an appropriate selection of the particle size, purity, and composition of the initial raw materials as well as the manufacturing processes, it could be possible to obtain a ceramic with a dense microstructure and submicrometer grain size improving their mechanical properties and reliability for use in orthopedic surgery [1,11,16,17]. A number of different processing techniques such as cold isostatic pressing [2,4,18–20] and various wet shaping methods [4] can be used to manufacture defect-free green bodies with good homogeneity of compaction and that allows to attain full density at relatively low temperatures allowing the industrial production of submicrometer Al_2O_3 ceramics. An alternative to conventional sintering above mentioned has been HIP inasmuch as it is also known, pressure helps sintering and densification processes, thus allowing reducing sintering temperature. A further alternative route is sinter-HIP which involves sintering of non-encapsulated green compacts up to the closed porosity stage (approximately 92% theoretical density) and subsequently applying a high pressure for the rest of the densification period [21]. The application of an external pressure provides a slight increase in the driving force for densification. Therefore, this technique reduces two steps into one step eliminating extra heating, cooling and handling and as result, sinter-HIP is able to reduce total HIPing time to about 2/3 of that of conventional sinter + HIP [22] improving mechanical properties and guaranteeing the production of an almost fully dense material with a fine-grained microstructure. Echeberria et al. [16] reported a fracture toughness of 2.5 ± 0.1 MPa $m^{1/2}$ with Taimicron TM-DAR alumina powder of 0.2 μm particle size sinter-HIPed at 1350 °C which is lower that the reported in this work. It is important to note that the fracture toughness values of the submicron grained Al_2O_3 have been relatively independent in the range of 300 gf–2 kgf according to appreciations of Muchtar and Lim [23].

Although sinter-HIP of Al_2O_3 ceramics is familiar for us, of the many existing ceramics materials [24], this processing route is still under study for improvements in order to meet optimal schedules of cycles to obtain excellent mechanical properties and to reduce processing costs. The current research was undertaken to study the potential of a sinter-HIP-process as an alternative densification method to conventional sintering on biomedical grade alumina ceramic with an alumina content higher 99.5% according to international standards [25] and its effects on hardness and fracture toughness. According to our results, we obtained microstructures with final grain sizes lower than reported by other authors as well as higher hardness and fracture toughness values compared with others investigations.

2. Experimental procedure

High purity Baikalox SM8 (>99.99%) Al_2O_3 powders (0.05 μm primary particle size and agglomerate size d_{50} = 0.3 μm, surface area 11.12 $m^2\ g^{-1}$ and crystal structure 100% α) manufactured by Baikowski, USA) were selected as the starting material. The powders were uniaxially pressed at 50–150 MPa into discs 16 mm diameter and 5 mm height using a rigid steel die. The compaction was carried out using an Elvec Hydraulic Press at a constant strain rate. Green compacts were placed into an alumina crucible with Al_2O_3 powder bed and sintered at temperatures of 1280, 1325, 1375, 1425 and 1460 °C during 2 h in air at a heating rate of 30 K min^{-1} to determine the minimum temperature to obtain close porosity. The specimens were placed into a boron nitride crucible with Al_2O_3 powder bed to minimize possible reaction with the graphite heating element and subsequently sinter-HIPed in an ASEA-HIP equipment (QIH-6) at 1350 °C at a heating rate of 30 K min^{-1} up to 1300 °C and finally 10 K min^{-1} up to final temperature with dwell time of 50 min to the final temperature in argon atmosphere under a pressure of 150 MPa. With the purpose to verify the effectiveness of sinter-HIP process, a conventional pressureless sintering cycle in air at 1350 °C during 50 min was also carried out. The Fig. 1 shows the selected sinter-HIP schedule where t_h represents the moment the sample reached the sintering temperature, whereas t_i is the elapsed time between t_h and the moment at which the gas pressure was increased. This sinter-HIP schedule has been adapted to the powder to produce a fine-grained material with a final density of at least >98.5%. Density was measured geometrically and in a Quantachrome Multipycnometer using helium displacement gas for as-sintered and HIPed samples. The samples were ground and polished through 0.5 μm diamond paste and the microstructure was revealed by thermal etching in air for 45 min at temperature 150 °C below the sintering temperature and characterized by scanning electron microscopy (SEM: JEOL JSM 5800 LV, Japan) using an accelerating voltage of 2 kV. The average grain size was measured using the linear intercept technique using 300–400 grains for each sample.

HV measurements were carried out on sintered samples by using a Microhardness Tester FM-7. Indentations were made on polished surfaces with a load of 1 kg held for 15 s. It is important to state that if there was crack branching or if the ratio of crack length to indentation length (c/a) was <2.3, the data were rejected. 30 indents were made for each sample and the average hardness was determined. The corresponding indentations sizes and crack lengths were determined using an optical microscope Olympus PMG3 and the indentation fracture toughness (K_{IC}) was calculated applying the formula expressed by Evans and Charles [26]:

$$K_{IC} = 0.00752.P/C^{3/2} \quad (1)$$

where K_{IC} is the fracture toughness, P the load and C the crack length. The crack measurement was carried out immediately after each indentation to prevent differences in crack length as a consequence of environmental variables such as moisture and time.

3. Results and discussion

As was outlined above, the minimum temperature to obtain close porosity was determined by means of pressureless sintering in air at different temperatures (heating rate = 30 K min^{-1}). The Fig. 2 shows the relative densities obtained as a function of the temperature. In this figure can be seen that the dotted line represents the minimum density required for a closed porosity (approximately 92% corresponding to 1360 °C), previous to

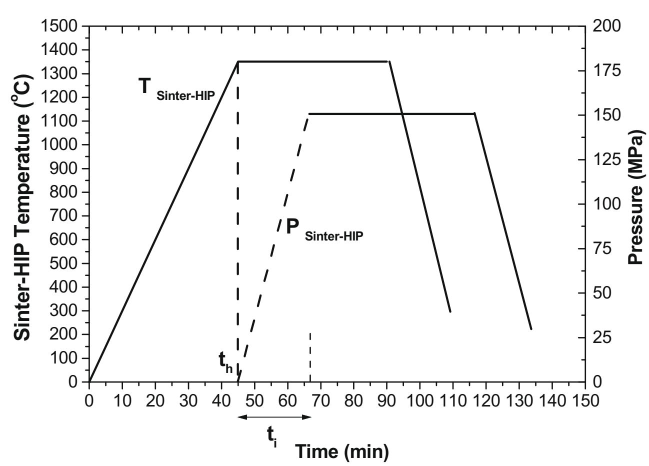

Fig. 1. Sinter-HIP schematic representation.