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Isostatic compaction behavior of yttria-stabilized tetragonal zirconia polycrystal powder granules

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ABSTRACT

The compaction behavior of a granulated powder of nanocrystalline 3 mol% yttria-stabilized tetragonal ZrO_2 polycrystals (3Y-TZP) was investigated by cold isostatic pressing. An elastomer gel was used as the solid pressure medium to form the compacts, in a process referred to as elastomer gel-cold isostatic pressing (E-CIP). The relative density (D_{re}) of the compact produced by isostatic pressing at a pressure (P) of 1522 MPa is 0.617, which is close to the upper limit of random packing (0.63). A curve fitted to the ln [1/(1–D_{re})] vs $P^{1/2}$ plot shows two linear regions, with a point of intersection at a transition pressure (P_c) of 520 MPa. A coral-like morphology of approximately spherical aggregates remains unchanged in E-CIP compacts formed at below the P_c. Many isolated aggregates are observed in E-CIP compacts formed at above the P_c. Thus, the transition in the fitting curve indicates the destruction of strong bonds between aggregates.

1. Introduction

Yttria-stabilized tetragonal zirconia polycrystals (Y-TZP) have been studied for >20 years, and are one of the most highly anticipated structural ceramics [1]. The nanocrystallization of Y-TZP enhances its strength, toughness, and high-speed superplasticity required for product molding. Microwave sintering (MWS) [2,3] and spark plasma sintering (SPS) [4,5] are effective methods for the densification of nanocrystalline Y-TZP powder compacts at low temperature in a short time duration. In SPS, heat is transferred mainly by thermal conduction from a surrounding resistance-heated die (e.g. a graphite die) to the nonconductive ceramic powder compact. Thus, SPS is an external heating method. In MWS, heat transfer occurs by the interaction between an irradiating electromagnetic wave and the dielectric powder compact. MWS is an internal heating method, and the powder compact is heated in an extremely short period of time. In MWS, the powder compact is densified at lower temperatures than those resulting from other sintering methods such as SPS or conventional sintering (e.g. heating in an electric furnace), as a result of the "microwave effect" (also called the non-thermal effect)" [6,7]. While the rapid heating induced by MWS is advantageous in industrial application, it can also lead to thermal cracking. To prevent thermal cracking, it is necessary to obtain homogeneously densified green compacts.

Perhaps the most effective method for homogeneously densifying powder compacts is cold isostatic pressing (CIP). A liquid is generally used as the pressure medium in CIP. Liquids have a lower incompressibility than solids under high hydrostatic pressure conditions. For example, the volume of water at room temperature (298 K) reportedly decreases by 4% at hydrostatic pressures up to 100 MPa [8]. However, under relatively low hydrostatic pressure conditions, the volumetric decrease of liquids can be ignored. Under these conditions, a liquid pressure medium shows adequate incompressibility and also excellent pressure transmissibility. Unless the viscosity is high, the shear stress of the liquid is extremely small (almost zero). Thus, any change in normal pressure at any point in an enclosed liquid is transmitted instantly to all points in the liquid, without decreasing the pressure (so-called Pascal's law). However, the high pressure transmission property can cause leakage of the liquid pressure medium from the (supposedly) sealed container. It is technically difficult to achieve secure sealing under high hydrostatic pressure conditions.

Isostatic compaction can also be achieved using a solid pressure medium such as rubber, and this is known as rubber isostatic pressing (RIP). Natural or silicone rubber is commonly used for RIP [9,10], because of its excellent incompressibility (Poisson's ratio: $\nu \approx 0.5$ [11,12]) and elasticity. In addition, an applied external shear stress is shared by part of the solid pressure medium to some extent, i.e. the shear stress is not propagated immediately to the entire enclosed solid pressure medium. Thus, it is relatively easy to ensure sealing of the container, even under high hydrostatic pressure conditions. However, the pressure transmissibility of a solid pressure medium is generally inferior to that of a liquid pressure medium. Thus, compared to CIP, it is more difficult to obtain highly dense and homogeneous green compacts by RIP. The properties of the rubber are of paramount importance for obtaining high quality green compacts.

Silicone rubber with a Shore-A hardness (type A) of 40 or more is usually used for RIP [10]. An elastomer consisting of urethane or silicone

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gel with an extremely low Shore-C (type C) hardness of <30 (corresponding to a Shore-A hardness of <17), and a low Young's modulus of <0.75 MPa, was recently obtained by lowering the crosslinking density. Material characteristics such as a high incompressibility modulus, small shear modulus, and high phase stability under high hydrostatic pressure conditions (i.e. several GPa) are not unknown. However, the above reported urethane or silicone gel has excellent potential as a solid pressure medium.

The objective of the current study is to fabricate homogeneous high-density Y-TZP green compacts by isostatic pressing. A urethane elastomer gel is used as a pressure medium, so the process is referred to as elastomer gel-cold isostatic pressing (E-CIP) (the elastomer gel used in this study is not synonymous with a rubber, so "E-CIP" is used instead of "RIP"). The compaction mechanism at hydrostatic pressures ranging from 66 to 1522 MPa is discussed.

2. Materials and methods

3Y-TZP powder (TZ-3Y; 3 mol% yttria-stabilized zirconia powder, Tosoh Co. Ltd., Minato-ku, Tokyo, Japan) was used in E-CIP experiments in this study. In general, most ceramic powders have particle sizes in the range of micrometer or sub-micrometer. The friction force (F_f) of a powder is proportional to its cohesive force (F_c), and increases linearly with the particle diameter (d_p). The inertial force (F_i) of a powder is proportional to the particle mass, and increases in proportion to the cube of d_p (i.e. d_p^3). To obtain sufficient powder flow-ability, it is necessary to reduce the F_f/F_c value by increasing the d_p [13]. The present 3Y-TZP powder had also been subjected to size enlargement treatment by agglomeration (granulation) by the manufacturer, but details of this method have not been reported. The chemical composition of the 3Y-TZP powder including impurities (as specified by the supplier) is 0.005 wt% Al_2O_3, 0.002 wt% SiO_2, 0.002 wt% Fe_2O_3, 0.020 wt% Na_2O, and 5.22 wt% Y_2O_3 [= $(ZrO_2)_{97.1}(Y_2O_3)_{2.9}$]. In addition, the specific surface area is 16±3 m^2/g, and the crystallite size is 26 nm.

Prior to E-CIP, the 3Y-TZP powder was pre-formed in a closed graphite die with inner and outer diameters of 15 and 45 mm, respectively, and a height of 40 mm. After evacuating the chamber to a pressure of 10 Pa, a constant uniaxial compression pressure of 50 MPa was applied to a pair of graphite rams with diameters of 15 mm and lengths of 30 mm. The powder compact mass was adjusted so that the height of the pre-formed compact reached 5 mm (i.e. 2.3×10^{-3} kg). The pre-formed compacts (15 mm in diameter and 5 mm in height) were covered with a urethane gel (Hyper Gel Firmness 30, Exseal Co. Ltd., Mino-city, Gifu, Japan), as shown in Figs. 1(a) and (b). The rubber hardness of the urethane gel is Shore-C 30, which corresponds to Shore-A 17. The urethane gel has a specific gravity is 1.08, elastic modulus in compression of 74.7 MPa, compression strength (at 10%) of 6.8 MPa, and visible light transmission of 93.4% (as specified by the supplier). Five 20-mm-diameter discs of the urethane gel were punched out from a 500 mm × 500 mm 5-mm-thick sheet. A ring-shaped urethane gel was prepared by punching out a 15-mm-diameter concentric disc from one of the 20-mm-diameter discs (Fig. 1(b) 1). After covering the pre-formed compacts, the outer surfaces of the urethane gel were lubricated using boron nitride (BN) spray. Because the urethane gel itself is strongly adhesive, part of the two upper gel discs was cut into a circular shape, to provide an evacuation route for air (see Fig. 1(a), (b) 1, and (b) 2).

E-CIP experiments were conducted using a cemented carbide die and rams (Fujilloy F10 and F20, respectively, Fujilloy Co. Ltd., Ōta-ku, Tokyo, Japan). Cemented carbide and alumina spacers (Fujilloy F10 and FCA10, respectively, Fujilloy Co. Ltd., Ōta-ku, Tokyo, Japan) were inserted between the ram and the cross-head. After evacuating the chamber to a pressure of 10 Pa, a compression pressure was applied at 10 MPa/s to up to the desired pressure of between 66 and 1522 MPa, and was then held at this pressure for 30 s. After the E-CIP experiments, no changes in the elastomer gel characteristics (e.g. size, color, and elasticity) were observed. Powder entrainment into the elastomer gel did not occur.

The densities of the obtained powder compacts as measured by the Archimedes' method were similar to those estimated by dividing the mass by the volume. A theoretical density of 6.06 g/cm^3 was used to estimate the relative density, to facilitate comparison with other reported data [14,15]. Microstructures of the as-received powder and powder compacts were examined using a field-emission scanning electron microscope (FE-SEM; S-4200, Hitachi High-Technologies Co., Minato-ku, Tokyo). To prepare FE-SEM specimens from the E-CIP powder compacts, plate samples with a vertical length of about 15 mm (corresponding to the compact diameter), horizontal length of 0.5 mm, and height of about 5 mm (corresponding to the compact height) were cut out from the E-CIP powder compacts by using a diamond wire profile saw (Millennium Series Model 228080, DWT Inc., Colorado Springs, CO) in dry condition. The plate samples were carefully fractured ensuring no damage to the fracture surface, and were then subjected to gold evaporation treatment. FE-SEM images were obtained from the central portion of the fracture surfaces.

3. Results and discussion

The change in density (D) as a function of applied compression pressure (P) is shown in Fig. 2. Previously reported densities of the same TZ-3Y powder compacts obtained by CIP at up to 500 MPa are also shown for comparison. Some differences exist in the pre-forming conditions. Sasazaki et al. [16] used a smaller pre-forming pressure of 10 MPa than was used in the current study (50 MPa). In addition, the pre-formed compacts used by Sasazaki et al. [16] had a smaller diameter (13 mm) and larger height (10 mm) than those in the current study (15 mm diameter and 5 mm height). Groot Zevert et al. [14] provided no detailed information on their powder compacts (i.e. the size and presence or absence of pre-forming treatment). Although some differences exist in the pre-forming conditions, the D–P curves for CIP are similar to the D–P curve for E-CIP. In particular, the CIP data from Groot Zevert et al. [14] lie on the D–P curve for E-CIP. These results indicate no apparent difference in the powder compacts obtained by CIP and E-CIP. Even at pressures above 500 MPa (i.e. the pressure range above the upper limit for conventional CIP), the densities increase monotonically with increasing E-CIP pressure, and the data points lie on a single smooth D–P curve. The maximum relative density of 0.617 can be obtained by E-CIP at 1522 MPa. This value is close to 0.63, which is the upper limit for the random packing of uniform rigid spherical particles [17].

Many attempts have been made to express the relationship between relative density (D_{re}) and compression pressure (P) for various powders. An empirical equation has been presented by Kawakita et al. [18]:

$$\frac{D_{td}}{D_{td}-D_{re}} = \frac{A_1 B_1 P}{1 + B_1 P} \tag{1}$$

where D_{td} is a tapped density, and A_1 and B_1 are constants. The main features of Eq. (1) are as follows. When $P[D_{td}/(D_{td} - D_{re})]$ is plotted as a function of P, almost all data points lie on a straight fitting line, and the results are independent of the compression pressure range, and thus, the compaction process. Therefore, it is difficult to establish the mathematical meaning of the constant values of A_1 and B_1. In addition, we could not determine the transition of the compaction mechanism (if it occurs) from a single straight line. An alternative expression is based on the consideration that a change in porosity (ε) is caused by a reaction with compression pressure. Thus, the compaction process is expressed by a differential equation:

$$-\frac{d\varepsilon}{dP} = K\varepsilon^m(1-\varepsilon)^n P^l \tag{2}$$