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Letter

Polarization-assisted surface engineering for low temperature degradation-proof in yttria-stabilized zirconia ceramics

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ABSTRACT

This study demonstrates that electrical polarization inhibited low-temperature degradation (LTD) of yttria-stabilized zirconia (YSZ). Electrical polarizations, which were confirmed by the thermally stimulated depolarization current (TSDC) measurements, were induced by an applied dc voltage. The induced polarizations caused hydrophilic surfaces. The more hydrophilic the surfaces became, the more resistant they became to degradation. We propose a mechanism of LTD inhibition in which the electric repulsive force induced by the polarizations prevented the invasion of water into the inner part of the YSZ.

1. Introduction

Owing to the defect structure introduced through substitution of Zr^{4+} with aliovalent cations such as Y^{3+} (Fig. 1(a)), tetragonal zirconia polycrystalline (TZP) ceramics gained both high mechanical toughness (Arrow 1 in Fig. 1) and superior O^{2-} ionic conductivity (Arrow 2 in Fig. 1), which currently contribute to biomedical applications of orthopedic hip joint balls and dental crowns [1,2] (Arrow 4 in Fig. 1), and high temperature fuel cells and O_2 gas sensors, respectively. The defect structure, however, concurrently brings the critical difficulty of low temperature degradation (LTD) to TZP according to the increased interaction of oxygen vacancies ($V_O^{\bullet\bullet}$) with water molecules [3–6] (Arrow 3 in Fig. 1), giving rise to complete destruction of TZP devices even at room temperature under water environment such as in vivo (Arrow LTD in Fig. 1). Surface engineering of TZP has been thus a crucial issue in biomedical materials science for these decades since the finding of LTD in the 1980s [3,4].

Polarization-assisted surface engineering (PASE), which utilizes the spontaneous surface electric filed on polarized materials, has been experimentally proved to have physical, chemical and biological effects on spatial manipulation of surrounding constituents both in vivo and in vitro [7–10]. In a series of polarization studies on bioceramic electrets, we effectively applied polarization to TZP for enhancement of biomedical function. Acceleration of apatite crystal growth in simulated body fluid [11] and improvement of cell proliferation [12] were represented. Combination of polarization and alkaline treatment also improved bioactivity in vitro and resistance to LTD [13]. This report demonstrates the effective LTD-proof of PASE through the arrows P1 to P2.

2. Experimental

Yttria doped (3 mol%) zirconia (YSZ) powders (TZ-3YB; Tosoh Corp., Japan) were pressed uniaxially into disks and sintered at 1450 °C for 2 h. The obtained disks were 10 mm in diameter and 1 mm in thickness. The prepared YSZ disks were divided into 6 groups, each with 6 disks. Each group was polarized under different conditions. The polarization procedure is illustrated in Fig. 2(a). Each disk was sandwiched between a pair of platinum films. A DC voltage was applied between the opposite electrodes at an elevated temperature of 200 °C for 30 min. The voltage was applied until the specimen was cooled to room temperature. The 6 groups were polarized under different DC voltages (0 V, 100 V, 500 V, 700 V and 1000 V). The surfaces of the polarized disk which were attached to negative and positive electrodes are named as P- and N-surfaces (Positively and Negatively charged surfaces), respectively. After the polarization procedure, contact angle measurements were performed using a commercial contact angle meter (DM-301; Kyowa Interface Science Co. Ltd., Japan) with drops of de-ionized water placed on the disk surfaces. The contact angle was measured at least 10 times using the same sample in ambient air, and the average value was used. Accelerated degradation tests were performed to evaluate LTD inhibition by the electrical polarization. The prepared disks were annealed in water at 120 °C to accelerate degradation. Phase transformation, tetragonal to monoclinic, was evaluated using X-ray diffraction (D8 advance; BrukerAXS). The volume fractions of monoclinic phases were calculated using the Garvie–Nicholson formula [14] and calibration method described by Toraya et al. [15]. In addition to the accelerated degradation tests, thermally stimulated depolarized currents (TSDC) measurement was performed (Fig. 2(b)). The measurement was performed using a sample which was polarized under applied voltage of 1000 V. After the polarization procedure, thin film Au electrodes were fabricated on the top and bottom surfaces of the polarized disk using vapor

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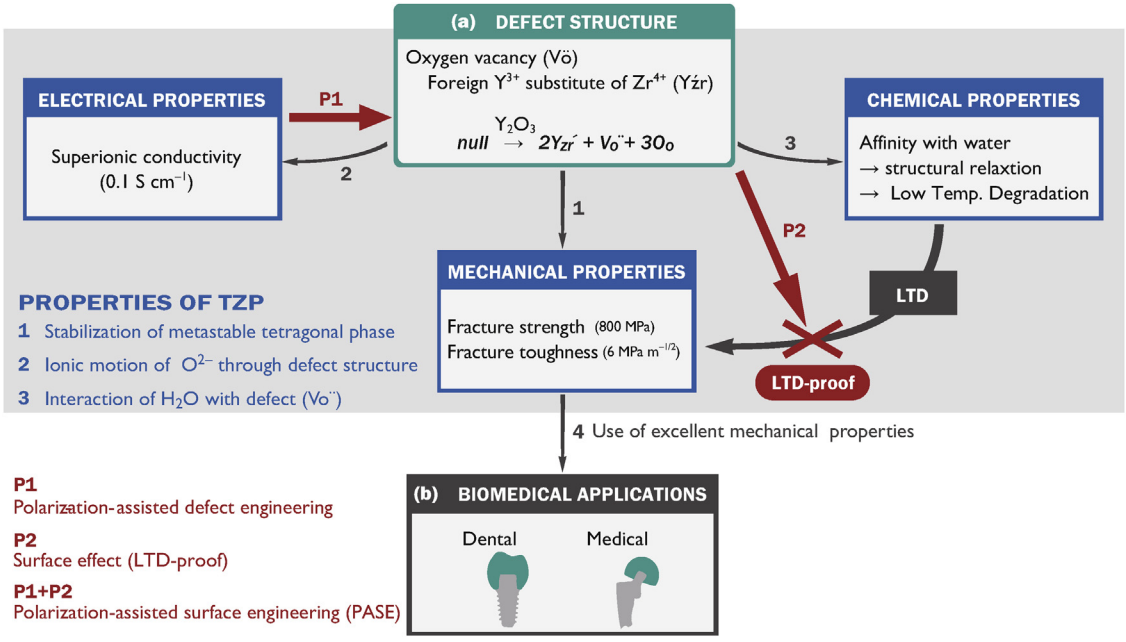

Fig. 1. Defect structure induced by aliovalent substitution derives three kinds of properties: (1) mechanical, (2) electrical, and (3) chemical. The chemical property, affinity with water, causes low temperature degradation (LTD). Application of electrical property affects the defect structure, i.e., induced electrical polarization engineers the defect structure of TZP (P1), and the engineered defect structure provides LTD-proof surface (P2).

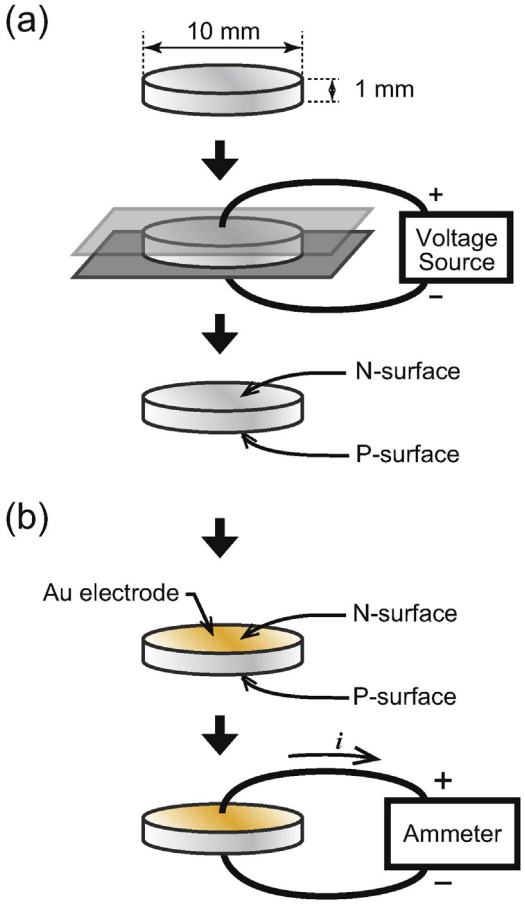

Fig. 2. Schematic illustrations of (a) polarization procedure and (b) thermally stimulated depolarization current (TSDC) measurements.

deposition method. The Au electrodes were connected to an ammeter (6514; Keithley Instruments, Inc.), and the disk was heated at a constant rate (5 °C min^{-1}). The current during the heating were measured using the ammeter.

3. Results and discussion

Fig. 3(a) shows monoclinic fractions on the surfaces after the accelerated test. The horizontal axis depicts the voltage applied in the polarization procedure. The monoclinic fractions decreased with increased applied voltage. Although monoclinic fractions of non-polarized surfaces were about 0.6, monoclinic fractions of the N-surface for applied voltage over 700 V were less than 0.3. The phase transformation from tetragonal to monoclinic phase was decelerated in the surfaces polarized at high voltage.

The LTD occurs in wet conditions; therefore, interaction between water and the YSZ surface is expected to play a large role in the progress of the transformation. The surface wettability on the polarized surfaces was characterized using contact angle measurements. Fig. 3(b) depicts the contact angle of water as a function of the applied voltage in the polarization procedure. The contact angles decreased as the applied voltages increased, indicating that the surfaces became hydrophilic after polarization procedure. Although, at lower polarization voltages, the contact angles on the N-surfaces decreased more than did those on the P-surfaces, no difference was found between the N-surface and the P-surface for surfaces polarized at 1000 V. This result, the decrease of the contact angles on N and P-surfaces, could be explained by electrowetting effect [16,17]. Electric charges on the surface result in decreased contact angles because of the electrostatic attractive force formed by the surface charges, and the effect does not depend on the charge polarity. The higher polarization voltage induces the larger surface charge. Therefore, the surfaces polarized at a higher voltage had the lower contact angles.

Fig. 3(a) and (b) shows quite similar appearance. The relation between the measured contact angles and monoclinic phase fractions is shown in Fig. 4. Horizontal and vertical axes respectively show contact angles and monoclinic fractions. A correlation exists