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Flexural fracture mechanisms and fatigue behaviors of $Bi_4Ti_3O_{12}$-based high-temperature piezoceramics sintered at different temperatures

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ABSTRACT

In this paper, a series of Aurivillius phase $Bi_4Ti_{2.95}W_{0.05}O_{12.05}$ + 0.2 wt% Cr_2O_3 (ab. BTWC) ceramics were prepared by a solid-reaction process and sintered at different temperatures (1050 ℃~1150 ℃), their microstructures, fracture mechanisms and fatigue behaviors were investigated under three-point-bending mode. The results show that the grain size of BTWC ceramics increases with increasing the sintering temperature. The typical transgranular mode dominates the fracture behavior of the samples sintered at lower temperatures, while the intergranular fracture can be also observed in the samples sintered at higher temperatures. Besides, the storage modulus (E') and mechanical loss ($\tan_m\delta$) of BTWC ceramics have a subtle variation with the increase of sintering temperature. In addition, the high-temperature environment could not only decrease the fracture toughness and bending strength of ceramics but also change their fracture mode. On the other hand, the bending strength also decreases with the decrease of loading rates, which could be attributed to the slow crack growth referring to the dynamic fatigue behavior of brittle materials. The slow crack growth parameter (n) of BTWC ceramics shows a downtrend with increasing their sintering temperature, indicating that those high temperature sintering samples possess a higher susceptibility when subjected to the long-term stress corrosion. Furthermore, the sample sintered at 1125 ℃ exhibits an excellent fatigue resistance when subjected to the cyclic stress. The corresponding fatigue mechanism is not only related to the ferroelectric domain configuration but also involved with the pores or impurities.

1. Introduction

In the Aurivillius family (bismuth layer structured ferroelectrics, BLSFs), much attention have been focused on a compound with the prototype structure of many BLSFs, $Bi_4Ti_3O_{12}$ (BIT). It is considered to have a potential application in high-temperature piezoelectric devices and ferroelectric memory storages [1–3], due to the high Curie temperature (T_c) of ~675 ℃ and large spontaneous polarization (P_s) of ~50 $\mu C/cm^2$ along to the a-axis benefited from its coherent triple-layer structure ($m = 3$) [4]. Unfortunately, the high electrical conductivity (σ_{dc}) and low remanent polarization (P_r) associated with oxygen vacancies have appeared as obstacles for its actual applications [5]. In order to overcome this disadvantage, the ion-doping technology was widely used for tailoring the electric property of perovskite-type ferroelectrics, based on the design and controlment on the lattice defect and phase structure of matrix [6,7]. As for BIT, both a large piezoelectric constant (d_{33} = 28 pC/N) and electrical resistivity ($\rho_{dc,\ 500\,℃} = 2.5\times10^6$ Ω•cm), as well as a higher hardness ($H_v = 3.25$ GPa) and toughness ($K_{IC} = 1.97$ MPa $m^{1/2}$) have been achieved by the co-doping of W^{6+} and Cr^{3+} through our previous researching works [8,9]. These significant improvement in these aspects especially electrical properties provides W/Cr co-doped BIT ceramics with a considerable usefulness for high-temperature (~500 ℃) piezoelectric accelerometer as sensitive materials.

However, within piezoelectric transducers, piezoceramics, as sensitive elements subjected to both electric loads and stress loads, are prone to premature failure due to the inherent brittle property, stimulated crack motion and original structural defects, which also need our more attention to their mechanical properties especially fracture mechanism as for seeking a long lifetime of materials. Recently, many researches have focused their attentions on the fracture behaviors of brittle ceramics [10–12], which was further explained by some typical mechanisms including ferroelastic deformation, crack deflection and zone shielding, etc [10,13–15]. Besides, the fatigue behaviors of materials also influence the reliability of devices, and even it plays an important role in lifetime prediction and reliability analysis. Many

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researches were fond of the dynamic fatigue behaviors decided by different loading rates [16,17], while the stress cyclic fatigue behaviors was rarely reported due to the limiting experimental condition and testing time consuming [18].

As for W/Cr co-doped BIT ceramics, although some of their electrical and mechanical properties have been investigated [9,13], no research has been conducted to aim at the influence of sintering temperatures on fracture mechanism and fatigue behaviors of ceramics associated with their microstructural evolutions. In this paper, a series of W/Cr co-doped BIT ceramics with the optimal chemical composition [9,13] were fabricated by the traditional solid reaction process and sintered at different temperatures. We investigated the elastic properties, fracture toughness, bending strength and fatigue behaviors of ceramics with laying stress on the evolution in fracture mechanism of ceramics associated with their microstructures. This work could help us to further understand the correlations between microstructures and mechanical properties in high-temperature piezoceramics.

2. Experiment

2.1. Preparation of ceramics

W/Cr co-doped BIT ceramics with a chemical formula of $Bi_4Ti_{2.95}W_{0.05}O_{12.05}$ + 0.2 wt%Cr_2O_3 (abbreviated as BTWC) were fabricated by the solid reaction process, the detailed steps was described in one of our previous papers [8]. Especially, in this experiment, the calcined powders were compacted into large cuboid green bodies using different forming dies to meet different mechanical tests. After PVA was burned out, these bodies were sintered in a temperature range of 1050–1150 °C for 6 h in a sealed alumina crucible for getting testing ceramics.

2.2. Characterization of ceramics

2.2.1. Microstructures

The fracture morphology of ceramics were observed by scanning electron microscopy (SEM, JSM-610LV, JEOL, Tokyo, Japan). The grain size of ceramics was calculated by the linear intercept method from SEM images. The value of bulk density was measured by the Archimedes method in water.

2.2.2. Fracture toughness

The Single Edge-Notch Bending test (SENB) was used for determining the fracture toughness (K_{IC}) of ceramics on a dynamic thermomechanical analyser (DMA, Boss, USA) associated with a high-temperature cabinet. The dimension of testing samples were prepared and polished into L36 mm × W4 mm × B2 mm, and an automatic razor blade machine was performed to create a notch of 0.2 mm width and 2 mm depth on the testing samples. Here, Fracture toughness was measured by the three-point bending test at room temperature (20 °C) and high temperature (400 °C) under a displacement rate of 0.05 mm/min, the value of K_{IC} can be calculated by the following equations:

$$K_{IC} = \frac{3PL_0\sqrt{a}}{2BW^2} f\left(\frac{a}{W}\right) \quad (1)$$

$$f\left(\frac{a}{W}\right) = 1.93 - 3.07\left(\frac{a}{W}\right) + 14.53\left(\frac{a}{W}\right)^2 - 25.07\left(\frac{a}{W}\right)^3 + 25.80\left(\frac{a}{W}\right)^4 \quad (2)$$

where, P is the load at failure; L_0 is the sample span (30 mm); a is the pre-crack length (2 mm); B and W are the width and thickness of samples, respectively.

2.2.3. Elastic properties and fracture behaviors

The elastic properties, bending strength and fatigue behaviors of ceramics were also measured using DMA under the three-point bending mode. The dimension of all testing samples were prepared and polished into L36 mm × W4 mm × B3 mm. For elastic properties test, a sinusoidal loading (-5 N∼-45 N) was applied on the samples, both the storage modulus (E') and the mechanical loss ($tan_m\delta$) were measured at a frequency of 1 Hz. In addition, the bending strength (σ) of ceramics can be calculated by the following equations:

$$\sigma = \frac{3PL_0}{2BW^2} \quad (3)$$

where, P is the load at failure; L_0 is the sample span (30 mm); B and W are the width and thickness of samples, respectively. In order to investigate the dynamic fatigue behaviors of BTWC ceramics sintered at different temperatures, the bending strength was gained under the ramp displacement rates of 0.5 mm/min, 0.05 mm/min and 0.005 mm/min, respectively. Besides, the stress cyclic fatigue behaviors were investigated by applying a sinusoidal loading with a frequency of 50 Hz and a stress ratio (maximum stress/minimum stress) of 0.1, and the minimum load of −5 N was used for making sure that the samples neither slip nor impact in each cycle during the test. Additionally, the number of cycles at failure was auto-recorded for the samples in each loading condition.

3. Result and discussion

3.1. Microstructures and elastic properties

Fig. 1 shows the fresh fracture surfaces of BTWC ceramics sintered at different temperatures. It can be seen that the grain of these samples present a plate-like morphology, which is attributed to the prior orientated growth and the higher grain growth rate in the direction perpendicular to the c-axis of the crystal [19]. Moreover, with increasing the sintering temperature from 1050 °C to 1150 °C, the average length of such plate-like grains shows a slow increase at first, but a sharp increase in the end. The sample sintered at 1150 °C grows into the largest grains, which may be attributed to the rapid grain boundaries migration of materials in a higher temperature sintering process. In addition, more or less pores could be found in all samples, which is related to the densification effect and the way of structural elements being packed of ceramics and regarded as having a significant influence on mechanical behaviors of piezoceramics. Here, in order to further quantify the microstructural evolution of BTWC ceramics with their sintering temperatures, the grain size and bulk density of samples as a function of sintering temperatures are depicted in Fig. 2. Expectedly, a lower temperature is associated with a smaller bulk density, which could also be corroborated by the numerous pores reflected in SEM photos. Furthermore, some polyhedral grains can be also observed in BTWC ceramics, and they are indexed as $Bi_2Ti_2O_7$ phase according to X-ray diffractometer analysis, which have been proposed in our previous works [8]. Besides, the second phase is the most obvious in sample sintered at 1125 °C as it can be observed by SEM (Fig. 1(d)), which agrees well with the result of the largest content of $Bi_2Ti_2O_7$ obtained in this sintering temperature [8]. To further characterize the secondary phase grain in this experiment, the elemental composition of the grain within red cycle is illustrated by the EDS spectrum as shown in Fig. 1(f), and the analysis result referring to the atomic percentage indicates that they are not $Bi_4Ti_3O_{13}$ but close to the composition of $Bi_2Ti_2O_7$, which as the second phase tends to be formed in the solid-reaction process of Bi-Ti-O system with excess titanium and benefited from the thermal decomposition of $Bi_4Ti_3O_{12}$ during sintering. Therefore, as for the same chemical composition of W/Cr co-doped $Bi_4Ti_3O_{12}$ ceramics, the obvious occurrence of $Bi_2Ti_2O_7$ in the sample sintered at 1125 °C may be attributed to the stronger thermal decomposition effect of $Bi_4Ti_3O_{12}$ at this sintering temperature.

Besides the grain growth, the fracture mode is also varying with the sintering temperature. Firstly, the classic transgranular fracture indicated by those randomly orientated grains seems to be predominant in the samples sintered at lower temperatures. However, in the samples