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Enhancement of the densification and mechanical properties of aluminum-doped zinc oxide ceramics by hot isostatic pressing

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ABSTRACT

The characteristics of sputtering targets used for transparent conductive oxide (TCO) films significantly affect the performances of TCO films produced by sputtering. The purpose of this study was to investigate the effect of hot isostatic pressing (HIP) on the densification, microstructure, electrical properties, and mechanical properties of Al-doped ZnO (AZO) and ZnO ceramic targets. The results showed that HIP treatment obviously improves the relative density, hardness, transverse rupture strength (TRS), and resistivity of AZO ceramics. AZO ceramics with relative densities of higher than 99.9% can be obtained after HIP at 1000 °C or 1250 °C. After HIP at 1250 °C, the resistivities of AZO ceramics are reduced from $6.0 \times 10^{-2} \pm 1.1 \times 10^{-2}$ Ω cm to $6.4 \times 10^{-4} \pm 2.9 \times 10^{-4}$ Ω cm. Furthermore, the hardness and TRS of AZO ceramics are raised from HV 261 ± 6 to HV 322 ± 5 and from 118 ± 3 MPa to 260 ± 3 MPa, respectively, after 1250 °C HIP. In contrast, for ZnO ceramics, the HIP process does not enhance the relative density or hardness, though the TRS and resistivity are improved.

1. Introduction

Transparent conductive oxide (TCO) films are widely used in the fields of liquid crystal displays, touch panels, solar cells, and other optoelectronic devices as the transparent conductive electrode. Aluminum-doped zinc oxide (AZO) film is both highly conductive and transparent and is a potential substitute for the conventional tin-doped indium oxide (ITO) film [1]. Due to a shortage of indium, the material cost of AZO is much lower than that of ITO. In addition, ZnO-based materials were also extensively used in other applications such as piezoelectricity and surface hydrophobicity [2–4]. Magnetron sputtering is a versatile technique for the deposition of TCO films, and the influences on the film properties of sputtering parameters, including deposition power, working pressure, sputtering atmosphere, and substrate temperature, have been clarified [5–9]. Recently, the importance of the sputtering targets used in producing TCO films has been gradually revealed, and the processes and properties of sputtering targets, particularly those of AZO, have been extensively investigated [10–22]. Numerous studies [10,11,22–26] have demonstrated that the properties of TCO targets affect not only the performances of TCO films but also the stability of the sputtering process. Therefore, in addition to the various sputtering parameters, the characteristics of sputtering targets are also critical factors for the optimization of sputtered films.

To date, several studies have indicated the importance of the effects of the sintered density [23,24], microstructural uniformity [25], stoichiometry [22,26], and electrical properties [10,11] of the oxide sputtering targets on the film properties and the stability of sputtering. An ITO target with a higher sintered density can be used to produce an ITO film with lower resistivity, though the visible transmittance of an ITO film does not apparently change with the ITO target [23]. Moreover, when an ITO target with a higher sintered density and more uniform SnO_2 distribution is used, the nodule and arcing phenomena are inhibited [24,25]. On the other hand, an AZO target with lower resistivity can be used to produce a more homogeneous film having lower resistivity [10,11]. Minami et al. [10] also indicated that using an AZO target with lower resistivity can decrease the arcing counts. Furthermore, the direct-current (DC) deposition rate of an AZO film is higher when the resistivity of an AZO target is lower [10].

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In general, the mechanical properties of oxide sputtering targets have rarely been investigated or discussed because the sputtering target does not directly sustain a mechanical loading. However, the mechanical properties of the oxide sputtering target are also important because the sputtering target must be mechanically machined to an accurate target size after sintering. The oxide target is intrinsically brittle and prone to cracking during mechanical machining. A sputtering target with a low mechanical strength could also result in the arcing phenomenon and nodule generation upon ion bombardment. Only Sun et al. [12] and Liu et al. [17] have examined the mechanical properties of AZO and gallium-doped zinc oxide (GZO) targets, respectively. Sun et al. [12] showed that the bending strength of an AZO target with a relative density of 99.8% is 148 MPa. Liu et al. [17] demonstrated that the bending strength and Vickers hardness of a GZO target with an addition of 1 wt% Ga_2O_3 are 91.79 MPa and ~HV 350, respectively.

The above review clearly demonstrates the complexity of the characteristics and industrial production of oxide sputtering targets. A feasible method for improving the mechanical properties of oxide targets is hot isostatic pressing (HIP), which can effectively reduce the porosity while retaining a fine grain structure. Reducing the porosity could also improve the conductivity of TCO targets. Thus, this study investigated the influences of HIP treatment on the sintered density, microstructure, mechanical properties, and electrical performances of AZO ceramic targets. Pure ZnO targets were also studied for comparison.

2. Material and methods

AZO and ZnO ceramic targets were prepared from ZnO and Al_2O_3 powders with median sizes of 0.4 μm and 0.2 μm, respectively. To produce AZO targets, 2 wt% Al_2O_3 powder was mixed with 98 wt% ZnO powder. AZO and ZnO ceramic slurries with 30 vol% solid contents were prepared. The detailed procedures and process parameters of the two ceramic slurries were reported in a previous study [13]. These two slurries were spray-dried in 140 °C air using a rotary spray dryer (L-8, Ohkawara Kakohki Co., Yokohama, Japan) to produce free-flowing granulated powders. The AZO and ZnO granules were uniaxially pressed at a pressure of 150 MPa into green disks with a diameter of 12.5 mm and a thickness of 7 mm. For transverse rupture strength (TRS) test, the two granules were also compacted into TRS specimens with dimensions of $55 \times 10 \times 5$ mm^3. The green densities of these green specimens were about 61%.

The green specimens were first heated at 5 °C/min to 600 °C and soaked for 30 min to remove organic additives. The debound specimens were subsequently heated at 10 °C/min to 1300 °C and sintered for three hours. For both the debinding and the sintering, the atmosphere was air. A HIP equipment made by Avure Technologies was used to further densify the two ceramics. The as-sintered specimens were HIPed at 1000 °C or 1250 °C for three hours to improve the sintered densities and various properties. The pressurized gas was argon, and the gas pressure used was 150 MPa. The designations of as-sintered, 1000 °C-HIPed, and 1250 °C-HIPed specimens were sintering, HIP-1000, and HIP-1250, respectively, in this study.

The sintered densities of as-sintered and HIPed ceramics were calculated using the Archimedes method. A scanning electron microscope (JSM-6360, JEOL, Tokyo, Japan) and an X-ray diffractometer (D8, Bruker, Karlsruhe, Germany) with Cu Kα radiation were used to investigate the microstructure and crystal structure, respectively. For observation of the microstructure, the fracture surfaces of the two ceramics, which provide valuable information on the three-dimensional distributions of pores and grains, were thermally etched and then analyzed. The average grain sizes of the two ceramics were estimated according to quantitative metallography [27]. The resistivities of the ceramics were also analyzed with the four-point probe method using a source meter (2400, Keithely Instruments Inc., OH, USA). Hardness was determined for both ceramics with a loading of 500 gf ($HV_{0.5}$) using a Vickers hardness tester (Micro Wizhard, Mitutoyo Co., Tokyo, Japan). TRS was measured by three-point bending tests using a material testing device (MTS 858, MTS Systems Corporation, MN, USA).

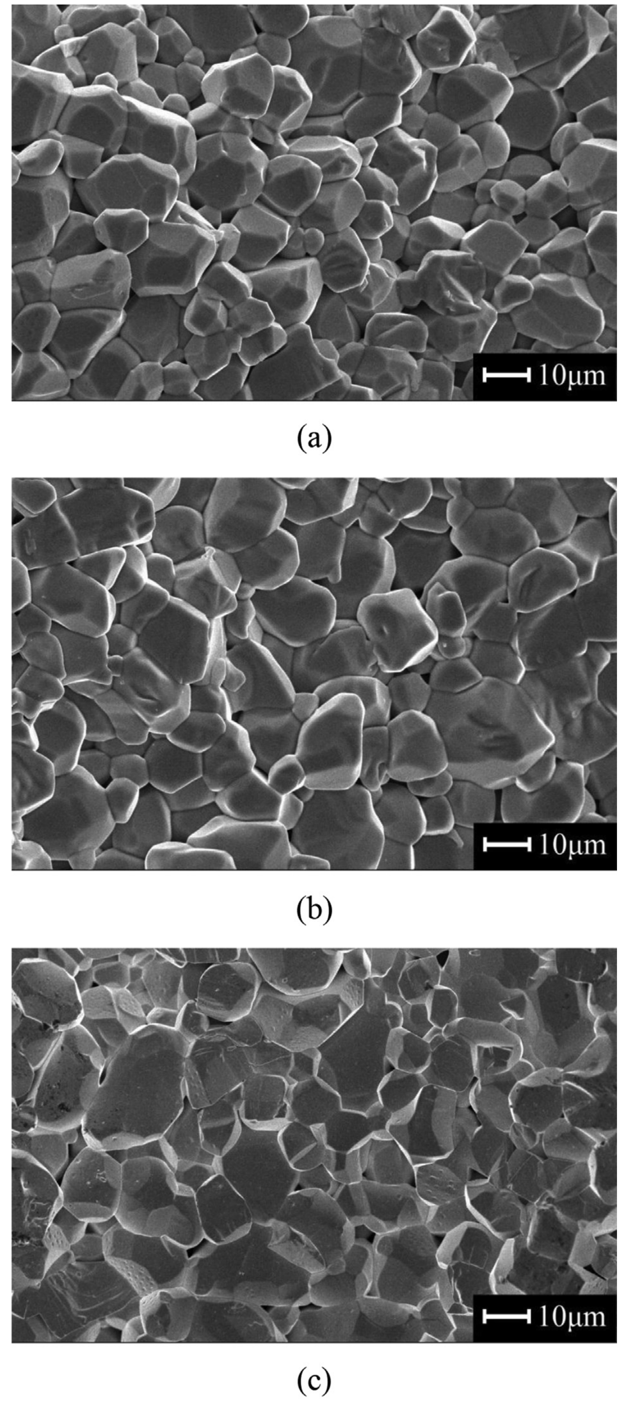

Fig. 1. The microstructures of ZnO ceramics after different processes. (a) 1300 °C pressureless sintering, (b) 1000 °C HIP, and (c) 1250 °C HIP.