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CeO_2 nanocrystals and solid-phase heteroepitaxy of $CeAlO_3$ interlayer on $Al_2O_3(0001)$ substrate

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ABSTRACT

Grain growth and interfacial solid state of CeO_2 nanocrystals (NCs) layer on $Al_2O_3(0001)$ substrate were examined. CeO_2 NCs layer on $Al_2O_3(0001)$ was prepared by dipping method using CeO_2 nanocrystals colloid solution. After heat treatment at 1000 °C in air, CeO_2 NCs layer was formed on $Al_2O_3(0001)$. The CeO_2 NCs sintered to form a surface layer with an interlayer of $CeAlO_3$ after heat treatment at 950 °C in H_2/Ar, leading to dense and smooth CeO_2 NCs layer on $Al_2O_3(0001)$ substrate. $CeAlO_3$ was grown via diffusion of CeO_{2-x} (non-stoichiometric CeO_2) and Al_2O_3, suggesting solid-phase reaction heteroepitaxy mechanism on $Al_2O_3(0001)$ single crystal substrate.

1. Introduction

Cerium dioxide (CeO_2, ceria) is one of the most important materials in the current industrial field due to its versatile properties for applications such as three-way catalysts for automobile, ultraviolet shielding materials, glass polishing agents, and electrolyte materials of solid oxide fuel cells [1–5]. CeO_2 films are attractive for various electronic and optical applications, silicon-on-insulator structures, miniaturized stable capacitors, oxygen sensors and optical coating [6–11]. Also, it is a promising candidate as buffer layer to prevent the chemical reaction between superconducting metal oxides such as $YBa_2Cu_3O_{7-x}$ and Al_2O_3 substrate [12–16]. In general, CeO_2 film is prepared by dry process in vacuum including magnetron sputtering technique [17–19], pulsed laser deposition (PLD) [11,20,21] and molecular beam epitaxy (MBE) method [22,23]. We have reported a fabrication of CeO_2 NCs layer on $Al_2O_3(0001)$ using CeO_2 nanocrystals (NCs) via simple dipping process [24]. However, the prepared CeO_2 NCs layer on $Al_2O_3(0001)$ after heat treatment at 1000 °C for 3 h in air has voids among CeO_2 NCs and $Al_2O_3(0001)$ substrate. In addition, in thin films fabrication, there is a problem about phenomenon that the sample thin film might crack at the interface between the prepared thin film and the substrate after cooling, owing to the difference of the thermal expansion coefficients. Therefore, the strain from difference in the thermal expansion coefficient between the CeO_2 and Al_2O_3 should be considered when the CeO_2 films are applied to a buffer layer. In this work, we employed the process to prepare CeO_2 NCs layer on $Al_2O_3(0001)$ single crystal substrate through the heat treatment in air, followed by heating under reductive condition, and investigated grain growth and interfacial solid state. The results showed that heat treatment in reducing condition using dilute hydrogen lead to dense and smooth CeO_2 NCs layer on $Al_2O_3(0001)$ by sintering assisted with interlayer formation. An interlayer of $CeAlO_3$ was formed by solid-state reaction via diffusion of CeO_{2-x} (non-stoichiometric CeO_2) and Al_2O_3 resulting in growth orientation on substrate.

2. Experimental methods

2.1. Synthesis of CeO_2 nanocrystals

7 mmol of diammonium cerium(IV) nitrate ($(NH_4)_2Ce(NO_3)_6$, 95+%, Wako Pure Chemical Industries) and 7 mmol of potassium oleate ($C_{17}H_{33}COOK$, 19% solution, Wako Pure Chemical Industries) were dissolved with 30 mL and 20 mL distilled water, respectively. The oleate solution was added into the cerium solution at room temperature under a vigorously stirred condition, followed by

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