J. Phys. D: Appl. Phys. **41** (2008) 015104 (7pp) doi:10.1088/0022-3727/41/1/015104

Characterization of luminescent samarium doped HfO_2 coatings synthesized by spray pyrolysis technique

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Received 14 June 2007, in final form 2 October 2007
Published 12 December 2007
Online at stacks.iop.org/JPhysD/41/015104

Abstract
Trivalent samarium (Sm^{3+}) doped hafnium oxide (HfO_2) films were deposited using the spray pyrolysis deposition technique. The films were deposited on Corning glass substrates at temperatures ranging from 300 to 550 °C using chlorides as raw materials. Films, mostly amorphous, were obtained when deposition temperatures were below 350 °C. However, for temperatures higher than 400 °C, the films became polycrystalline, presenting the HfO_2 monoclinic phase. Scanning electron microscopy of the films revealed a rough surface morphology with spherical particles. Also, electron energy dispersive analysis was performed on these films. The photoluminescence and cathodoluminescence characteristics of the $HfO_2:SmCl_3$ films, measured at room temperature, exhibited four main bands centred at 570, 610, 652 and 716 nm, which are due to the well-known intra-4f transitions of the Sm^{3+} ion. It was found that the overall emission intensity rose as the deposition temperature was increased. Furthermore, a concentration quenching of the luminescence intensity was also observed.

1. Introduction

Transition metal oxides, such as hafnium oxide (HfO_2), have attracted significant attention because of their potential applications. HfO_2 can be used as a gas sensor [1] and waveguide [2], as well as a protective coating, due to its thermal stability and hardness [3, 4]. Up to now, one of the most important applications of HfO_2 is probably in the microelectronic industry. HfO_2 is studied as a high-*k* gate dielectric layer with a relatively high index of refraction and a wide energy band gap. These properties make HfO_2 a good candidate for applications in metal-oxide-semiconductor devices for the next generation [5, 6]. The wide band gap of HfO_2 makes it transparent over a wide spectroscopic range: from the ultraviolet to the mid-infrared [7]. Furthermore, HfO_2 has been employed in optical coating applications such as high-reflectivity mirrors, anti-reflection coatings, filters and beamsplitters [8, 9]. In addition, the large energy band gap and the low phonon frequencies make HfO_2 an appropriate host matrix for being doped with rare earth activators [10, 11]. Efficient luminescent materials have wide applications in electroluminescent flat panel displays; colour plasma display panels, scintillators, cathode ray tubes, fluorescent lamps, lasers, etc. In recent years the study of luminescent materials based on HfO_2 has been intensified. Some groups have studied the optical properties of doped and undoped HfO_2 [12–14]. Specifically, some studies on rare earth and Mn doped HfO_2 films have been recently reported [12, 15–17]. One of the most important properties of rare earth ions doped compounds is the very narrow absorption and emission band arising from the parity-forbidden intraconfigurational 4f–4f transitions. As a result, trivalent rare earth ions possess very good luminescent characteristics (high colour purity). In particular, luminescence of samarium ions in solids has attracted much attention in recent years to produce reddish-orange emitting phosphors for colour display panels [18, 19],

