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Synthesis and characterization of hafnium oxide films for thermo and photoluminescence applications

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ABSTRACT

Hafnium oxide (HfO₂) films were deposited by the ultrasonic spray pyrolysis process. The films were synthesized from hafnium chloride as raw material in deionized water as solvent and were deposited on corning glass substrates at temperatures from 300 to 600 °C. For substrate temperatures lower than 400 °C the deposited films were amorphous, while for substrate temperatures higher than 450 °C, the monoclinic phase of HfO₂ appeared. Scanning electron microscopy showed that the film's surface resulted rough with semi-spherical promontories. The films showed a chemical composition close to HfO₂, with an Hf/O ratio of about 0.5. UV radiation was used in order to achieve the thermoluminescent characterization of the films; the 240nm wavelength induced the best response. In addition, preliminary photoluminescence spectra, as a function of the deposition temperatures, are shown.

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1. Introduction

The measurement of ionizing radiation is, at this moment, a great challenge due to the risk associated with the exposure to this kind of radiation as well as solar radiation in medical and scientific equipments. Many measurement programs on ultraviolet radiation (UVR) have been implemented due to the spoiling of the ozone layer. However, little efforts have been dedicated to generate new materials that could be used as meters of ultraviolet radiation. UVR measurement using thermoluminescence (TL) materials has been suggested in the past by several authors (Driscoll, 1966; Chang and Su, 1993; Colyott et al., 1999; Azorin et al., 1998). The advantages of this technique are the easy readout of the samples and the small size of these kinds of dosimeters. In this sense, the hafnium oxide (HfO₂) is a material with a wide range of possible technological applications because of its chemical and physical properties such as high melting point, high chemical stability, and hardness near to diamond in its tetragonal phase. These properties make HfO₂ an attractive compound to be used as gas sensors and along with many electronic and optical applications (Capone et al., 1998; Wilk et al., 2001; Niimistö et al., 2004; Zukic et al., 1990; Edlou et al., 1993).

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The large energy gap and low phonon frequencies of the HfO₂ makes it appropriate as host lattice for being doped with rare earth activators (Mignotte, 2001; Zhao and Vanderbilt, 2002). In the recent years the study of luminescent materials based on HfO₂ has been intensified. Some groups have studied the optical properties of doped and undoped HfO₂ (Lange et al., 2006; Ito et al., 2005). Hafnium oxide films have been deposited by a variety of techniques; these include atomic layer epitaxy (Ritala et al., 1994), chemical vapor deposition (Balog et al., 1977; Reicher et al., 2000), electron beam evaporation (Cho et al., 2002). Compared to powdered materials, luminescent coatings offer advantages such as good adhesion to the substrates, have no outgassing problems, better thermal stability, possess uniform properties across the covered area and higher resolution and contrast with lesser materials. Ultrasonic spray pyrolysis represents an alternative processing method that has been employed for deposition of a wide variety of thin films, coatings and several types of powder production. Among them we can highlight those with luminescent properties such as Al₂O₃, ZrO₂ and ZnAl₂O₄, doped with rare earth or transition elements (Mn) (Esparza-García et al., 2002; García-Hipólito et al., 2002, 2003).

In this work, preliminary thermoluminescence (TL) and photoluminescence (PL) features of non-doped HfO₂ coatings, synthesized by the ultrasonic spray pyrolysis technique, are studied. Also, the surface microstructure characteristics, the crystalline structure, are shown.