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Hydrogen Gas Response of $Zn_{1-x}Ag_xO_y$ and $Cu_{1-x}Zn_xO_y$ Nanostructured Films

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Detection of hydrogen gas in industry, biomedical systems and combustion systems is important for safety reasons. Silver doping in zinc oxide and zinc doping in copper oxide were investigated to obtain improved hydrogen sensing performances for sensors. Samples were grown by chemical method and studied by X-ray diffraction, SEM and sensorial techniques. For selectivity study samples were exposed to hydrogen, methane and ethanol gases. Were found growth and annealing regimes which allow us fabrication of faster and more selective gas sensors based on $Zn_{1-x}Ag_xO_y$ nanostructured films and nanocrystallite $Cu_{1-x}Zn_xO_y$ films with respect to 100 ppm H_2.

Keywords: $Cu_{1-x}Zn_xO_y$, $Zn_{1-x}Ag_xO_y$, Chemical synthesis, Nanostructures, hydrogen, oxide, film, RTA.

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1. INTRODUCTION

Metal oxide nanoscale materials have been attracting much attention due to their unique size- and morphology-dependent physical and chemical properties [1]. It is also promising in the development of gas sensors applications and it should be expected that metal oxide nanostructures exhibit better performance than the bulk based devices [1-2]. The monitoring of H_2 and other flammable gases in industry, biomedical systems, and combustion systems is becoming important factor by increasing the requirements for sustainable and renewable energy sources. Among various types of sensors for detection such gases, chemoresistive gas sensors have been intensively investigated because of their simple usability and low cost [1-2].

Zinc oxide (ZnO) is a well known *n*-type wide band gap semiconductor (3.37 eV at 300 K for bulk material). It has been studied extensively as the main material for many modern applications, especially for sensors applications. In this regard, various synthesis methods have been explored to fabricate different nanostructures like nanowires by hydrothermal method for sensor applications [2,3], by electrodeposition for light emitting devices by europium doping [4], etc.

Cupric oxide (CuO) is a typical *p*-type semiconductor with a band gap of 1.2–1.9 eV and a monoclinic crystal structure [5-9]. Additionally, compared with other metal oxide nanostructures, such as TiO_2, ZnO, and SnO_2, copper oxide nanostructures have more interesting magnetic and superhydrophobic properties [5]. In recent years, the 'mesoporous' metal oxides, containing pores [1,6], have attracted interest for the application in gas sensors because of their high surface-to-volume ratio that can lead to improve sensitivity by the increased adsorption/reaction sites. Various works have reported about the gas-sensing properties of copper oxide, especially on quasi1D structures, like nanowires growth by surface oxidation of a Cu foil for biosensor application [7], by in situ thermal oxidation in air on Cu_2O [8], by low temperature solid-phase process [9], etc. Also were studied sensing characteristics to H_2 and C_2H_5OH of CuO mesoporous films prepared via a precursor-based ink solution route [1].

Compared to *n*-type semiconducting oxide based gas sensors, p-type oxide semiconductor gas sensors exhibit not only shortcomings but also promising potentials for practical applications [10]. In this work, were studied gas response and selectivity of $Zn_{1-x}Ag_xO_y$ and $Cu_{1-x}Zn_xO_y$ nanostructured films to different gases, namely hydrogen, methane and ethanol.

2. EXPERIMENTAL

2.1 Growth of $Zn_{1-x}Ag_xO_y$ and $Cu_{1-x}Zn_xO_y$ nanostructured films synthesized by chemical solution approach

Glass substrates were used for growth of both types of metal oxides. The cleaning process were reported in our previous work [11]. For growth $Zn_{1-x}Ag_xO_y$ nanostructured films were used the aqueous zinc complex solution approach. It comprises a mixture of zinc sulfate ($Zn(SO_4) \cdot 7H_2O$), silver nitrate ($AgNO_3$) and sodium hydroxide (NaOH) mixed until complete dissolution. The concentration of the complex solution was diluted to obtain 0.05 – 0.15 M zinc concentration for deposition by adding respective quantities of deionized (DI) water. For growth $Cu_{1-x}Zn_xO_y$ nanocrystalline films were used the aqueous copper complex solution of copper sulfate ($CuSO_4 \cdot 5H_2O$) and sodium thiosulfate ($Na_2S_2O_3$). Finally nanostructured films were exposed to different durations of rapid thermal annealing at various temperatures for 60 s.

2.2 Characterization

The samples of $Zn_{1-x}Ag_xO_y$ and $Cu_{1-x}Zn_xO_y$ nanostructured films were analyzed by X-ray diffraction (XRD) using a Rigaky "DB/MAX" powder diffractometer with a nickel-filtered CuKα radiation source (λ = 1.54178 Å) and a scanning rate of 0.05 °/s in the 2θ range from 30 to 90 °. The compositional analysis was carried out using energy-dispersive X-ray spectroscopy (EDX), in combination with SEM. The different characterization techniques confirmed that the nanostructured films are crystalline material.

