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Influence of discharge voltage on the sensitivity of the resultant sputtered NiO thin films toward hydrogen gas

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ABSTRACT

Nickel oxide thin films were deposited on glass substrates as the main gas sensor for H_2 by the DC sputtering technique at various discharge voltages within the range of 1.8–2.5 kV. Their structural, optical and gas sensing properties were investigated by XRD, AFM, SEM, ultraviolet visible spectroscopy and home-made gas sensing measurement units. A diffraction peak in the direction of NiO (200) was observed for the sputtered films, thereby indicating that these films were polycrystalline in nature. The optical band gap of the films decreased from 3.8 to 3.5 eV when the thickness of the films was increased from 83.5 to 164.4 nm in relation to an increase in the sputtering discharge voltage from 1.8 to 2.5 kV, respectively. The gas sensitivity performance of the NiO films that were formed was studied and the electrical responses of the NiO-based sensors toward different H_2 concentrations were also considered. The sensitivity of the gas sensor increased with the working temperature and H_2 gas concentration. The thickness of the NiO thin films was also an important parameter in determining the properties of the NiO films as H_2 sensors. It was shown in this study that NiO films have the capability to detect H_2 concentrations below 3% in wet air, a feature that allows this material to be used directly for the monitoring of the environment.

1. Introduction

In recent years, NiO thin films have attracted particular interest due to their potential for use in a wide variety of applications, for instance, as electrodes for super capacitors [1] or as electro-chromic coatings [2]. Furthermore, they are considered to be semiconductor models with p-type conductivity films due to their wide band-gap energy range of from 3.6 to 4.0 eV [3]. Recent works have shown that NiO is also a promising functional material for applications using thin NiO films in resistive type gas sensors [4,5]. The most attractive features of NiO are: (i) its excellent durability and electrochemical stability, (ii) low material cost, (iii) promising ion storage in terms of cyclic stability, (iv) large spin optical density, and (v) the possibility of manufacturing it by a variety of techniques.

NiO films can be prepared by physical and chemical methods such as spray pyrolysis, electron beam evaporation, pulsed laser deposition, plasma-enhanced chemical vapour deposition, and reactive sputtering. Among these methods, sputtering has been the most widely used one. An important advantage of sputter deposition is that even materials with very high melting points can be easily sputtered, while the evaporation of these materials in a resistance evaporator is problematic or impossible. Sputter-deposited films have a composition that is close to that of the source material. Sputtered films typically have a better adhesion to the substrate than evaporated films. With sputtering, the films from a target can be deposited in virtually any direction. The target contains a large amount of material and is maintenance-free, making this technique a suitable one for ultrahigh vacuum applications [6–12].

Metal oxide conductometric gas sensors detect gases by measuring the variation in the conductance or resistance of the metal oxide material. When the metal oxide is exposed to a gas atmosphere at a moderate temperature, two things can occur, namely gas adsorption due to the high reactivity of the metal oxide surface, or reaction of the gas molecules with the surface species that have already been iono-sorbed at the metal oxide surface [13]. Gas sensitivity is strongly related to the adsorption/processes occurring at the gas surface interface. It has been shown that for the effective operation of chemical sensors, the gas sensing materials should respond specifically to adsorption/desorption parameters for oxygen and the detecting gas [14].

Many researches have been carried out on the dependence of film properties on sputtering parameters such as sputtering pressure,

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Fig. 1. DC-sputtering device set up for preparing NiO thin films.

sputtering power and deposition time [15–18]. This paper investigated the structural and optical properties, and the sensitivity of deposited NiO films towards H_2 gas as a function of the sputtering discharge voltage.

2. Experimental procedure

The films were prepared by the DC sputtering technique with nickel oxide as the target (99.9) in a homemade glow discharge plasma system. Fig. 1 shows the photo of the sputtering chamber and the associated DC power supply. The sputtering chamber was evacuated to less than 5×10^{-5} mbar, and then, argon gas was introduced into the chamber at a specified pressure through a throttle at a sputtering gas pressure of 6×10^{-2} mbar, while the discharge current was 15–30 mA and the voltage was 1.8–2.5 kV. Glass substrates, with dimensions of (4×1.5×0.15) cm and an optical transmission of about 95%, were used for depositing the films. The optical spectra of absorption and transmission were measured using a spectrophotometer (UV-Vis-2601, Biotech Engineering Management CO. LTD). X-ray diffraction (XRD) was used to characterize the structure of the films on a diffractometer (Shimadzu 6000) with Cu Kα (λ=0.15418 nm) radiation. The thickness of the prepared films was measured using an FT-650 film thickness measurement probe. The atomic force microscope (AFM) from Angstrom Inc. (AA3000), and a Japanese-type scanning electron microscope (SEM) (TESCAN), were used to analyse the morphological structure of the deposited films. The resistance (current variations) of the NiO films that were exposed to various environmental gases at 200–350 °C was measured inside the homemade gas sensing set-up and the bias voltage was supplied by a power supply of 4.8 V, while the flow rate of the carrier gas was calculated by using a flow meter.

Fig. 2 shows gas sensor of NiO thin film. The resistance measurements of the NiO samples were carried out in the dark, so that the photon excitation of the carriers would not be significant. H_2 was mixed with air before it arrived at the chamber inlet. The gas flow was controlled by two calibrated flow meters, and the gas was fed into an injection point located below the sample holder. After introducing the H_2 gas into the chamber, the film resistance (current) versus time was recorded by a PC–interfaced digital multimeter (UNI-UT81B) at various operating temperatures. The sensitivity of the gas sensor is defined as the capability of the sensor to respond to the presence of a given gas concentration. Mathematically, the sensitivity, S, is defined by the formula S=Rg–Rn/Rn for the reducing gas, and S=Rn–Rg/Rg for the oxidizing gas, where Rg and Rn are the resistances of the sensor after and before the gas is passed and achieves saturation [19].

Fig. 2. Shows gas sensor of NiO thin film.

3. Results and discussion

3.1. Structural analysis of NiO sputtered films

The crystal structures of the as-deposited NiO films were identified by X-ray diffraction (XRD). The reflection peak indicated that the films deposited on the glass substrates were polycrystalline in nature with an fcc structure (JCPDS card no. 78-0643). Fig. 3 displays the X-ray diffraction patterns for the nickel oxide films prepared at 2.1 kV and 2.5 kV at a pressure of 0.06 mbar in argon, with a thickness ranging between 128.3 and 164.4 nm. It was found that when the sputtering voltage (film thickness) was at 1.8 kV (83.5 nm) and 2 kV (102.4 nm), there was only one peak package in the diffraction pattern, without the appearance of an obvious crystallization peak, thus indicating that the NiO thin films under these sputtering voltages and related thicknesses had an amorphous structure. When the sputtering voltage was at 2.1 kV and 2.5 kV with a Bragg angle of 2θ=43.10°, the widened dispersion peak appeared in correspondence with the crystal plane (200). The increase in the plane intensity for the sputtered films could be attributed to the increase in the crystalline grain size caused by an increase in the thickness of the film and the substrate (anode electrode) temperature [20,21]. The crystallite size of the films was calculated from the (200) orientation by using the Scherrer formula [20–22]:

$$L = K\lambda/\beta\cos\theta \quad (1)$$

where L is the mean crystallite size, K is the correction factor, λ is the wavelength of the incident beam, and β is the full width at half maximum corresponding to the Bragg diffraction angle, θ. Here, the values of K and λ were taken as 0.94 and 0.1541 nm, respectively. The crystallite size was measured in nanoscale and was estimated to be

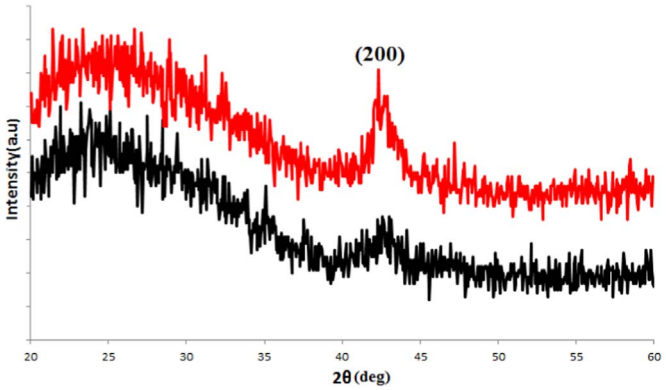

Fig. 3. X-ray diffraction profiles of NiO films at sputtering discharge voltages 2.1 and 2.5 kV.