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Nanocrystalline Nickel Oxide (NiO) Thin Films Grown on Quartz Substrates: Influence Of Annealing Temperatures

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In the present investigation, nanocrystalline NiO thin films were prepared by thermal oxidation annealing of DC magnetron sputtered Ni thin films on quartz substrates. The effect of annealing temperature on the films structural, morphological and optical properties was investigated. The XRD analysis shows that all prepared films were of NiO with cubic structure and (200) orientation. The thickness of NiO films was in range of 40–100 nm. The average crystallite size is found to increase from 16 to 36 nm and the optical band gap energy decreases from 3.62 to 3.38 eV by increasing the annealing temperature from 400 °C to 600 °C. The AFM and SEM results show that the annealing temperature effectively influences the surface morphology of the films.

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1. Introduction

Nickel oxide (NiO) is a semi-transparent, stable wide direct band-gap material (3.56 eV), which exhibits p-type semiconducting behavior with weak absorption bands [1–8]. NiO thin films are attractive materials which can be used as antiferromagnetic layers, as active electrodes in electrochromic devices, p-type layers for UV detectors and functional sensor layer for chemical sensors [1–5]. NiO thin films can be fabricated by different physical and chemical deposition methods, such as sputtering, pulsed laser deposition, thermal evaporation, spray pyrolysis, electron beam evaporation, dip coating, spin coating and electro deposition [1–8]. In the present work, DC magnetron sputtering was employed to prepare nickel (Ni) films. Then, for preparation of NiO films, the Ni films were annealed in oxygen atmosphere at different annealing temperatures. The focus of the paper is to investigate the effect of annealing temperature on structural, morphological and optical properties of NiO thin films.

2. Experimental details

In the first step Ni thin films were deposited on 1×1 cm^2 quartz substrates by DC magnetron sputtering method at the same deposition conditions. Prior to films deposition the substrates were cleaned ultrasonically in acetone and ethanol for 15 min. The metal nickel with purity of 99.999% was used as target and quartz was used for substrates. The base pressure was 5.8×10^{-5} mbar and after the introduction of the sputtering gas (Ar 99.999%) into the chamber the deposition pressure had been reaching 7×10^{-3} mbar. The deposition time for all films was 20 min. The target to substrate distance was kept at 4.2 cm. The deposition was done at room temperature. In the second step, the Ni films were annealed in oxygen atmosphere for 240 min at different temperatures, namely 400 °C, 500 °C and 600 °C. The thickness of NiO films was in range of 40–100 nm. The crystal structure of the films was characterized by means of X-ray diffraction (XRD) analysis (XRD, Philips, PW-1800, with Co K$_\alpha$ radiation with $\lambda = 0.17890$ nm). The diffraction patterns of films were recorded by varying diffraction angle 2θ in the range of 10° to 70° in steps of 0.02° and time per step of 1 s. The surface morphology of the deposited films was examined by atomic force microscopy (AFM, Park Scientific Instrument, Auto Probe CP, USA) and scanning electron microscopy (SEM, Hitachi S-4160). The optical transmittance spectra of the films were measured by a spectrophotometer (CARY 500 Scan) in the range of 200–1100 nm.

3. Results and discussion

The XRD spectra of the NiO thin films prepared at different annealing temperatures are shown in Fig. 1.

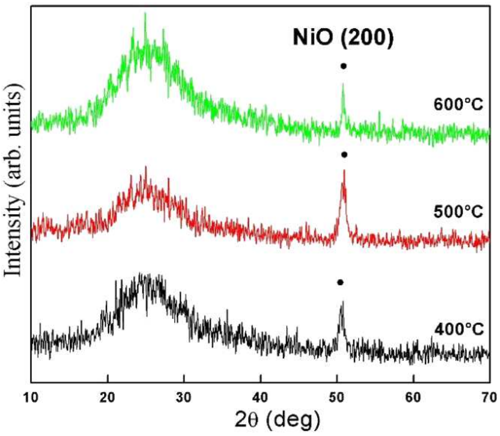

Fig. 1. XRD patterns of NiO thin films prepared at different annealing temperatures.

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We can observed a broad peak at $2\theta = 24°$ that belongs to quartz substrate and another peak at $2\theta = 50.7°$, corresponding to the (200) crystal plane of the cubic NiO phase (JCPDS 78-0643).

The XRD patterns indicate that the NiO thin films have a single crystalline cubic structure. Additionally the average crystallite size D and microstrain ε of the NiO films have been also calculated according to Debye-Scherer equation [9] and the results are presented in Table I. According to data from Table I we can observe that the average crystallite size had increased with increase of annealing temperature, which reveals a fine nanocrystalline grain structure. Similar results, showing influence of annealing, were also obtained in [9–11] for silver, molybdenum oxide and zirconium oxide thin films. As the annealing temperature was increased the XRD peaks became sharper, which was due to the increase of particle size and due to the enhancement of the crystallinity. The full width at half maximum (FWHM) was found to decrease with annealing temperature, which may be due to the decrease in the concentration of lattice imperfections.

TABLE I

Summary of structural data obtained for NiO thin films.

Anneal. temp. [°C]	Phase composit.	Miler indices (hkl)	FWHM [rad]	Average crystallite size D [nm]	Microstrain
400	NiO	(200)	10.8×10^{-3}	16	5.79×10^{-3}
500	NiO	(200)	9.7×10^{-3}	18	5.20×10^{-3}
600	NiO	(200)	4.9×10^{-3}	36	2.61×10^{-3}

Surface morphology of the films was examined by AFM analysis. Figure 2 gives the 3D images of the AFM scans taken on the scale of 2×2 μm^2. The results show that the annealing temperature has effectively influenced the surface morphology of the films. The films are dense and contain regular surface grains. The root mean square roughness (RMS) of films increases from 1.22 nm to 4.76 nm with the increase of annealing temperature from 400 to 600 °C.

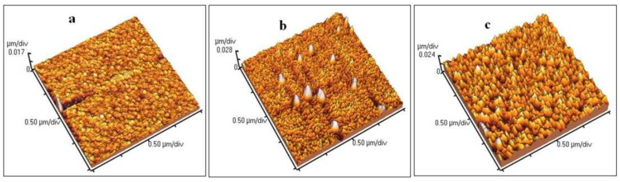

Fig. 2. 3D AFM images of NiO thin films prepared at different annealing temperatures: (a) 400 °C, (b) 500 °C and (c) 600 °C.

The morphological characterization was also performed using SEM analysis. Figure 3 shows the SEM images of the NiO thin films annealed at different temperatures. We can observe that the surface of the films prepared at annealing temperature of 400 °C (Fig. 3a) consists of regular and small grains and the increase of annealing temperature up to 600 °C (Fig. 3b and c) has led to agglomeration of grains and to an increase of the particles sizes.

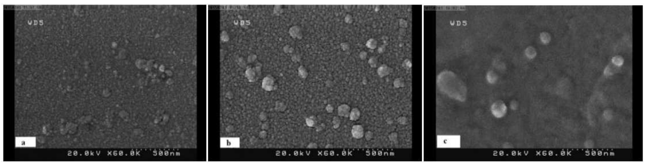

Fig. 3. SEM micrographs of NiO films for different annealing temperatures: (a) 400 °C, (b) 500 °C and (c) 600 °C.

The transmittance spectra of the NiO thin films at wavelengths in range of 200–1100 nm are shown in Fig. 4. The optical transmittance of prepared films shows that the transmittance increases at elevated annealing temperatures due to improving crystalline microstructure, which results in lesser defect scattering. The optical absorption coefficient α was evaluated from the optical transmittance T using the relation $\alpha = -\ln T/d$, where d is the thickness of films. The optical band gap E_g of the NiO films prepared at different annealing temperatures was calculated from plot of $(\alpha h\nu)^2$ versus photon energy $h\nu$ by extrapolating the linear portion to zero absorption ($\alpha = 0$) [12].

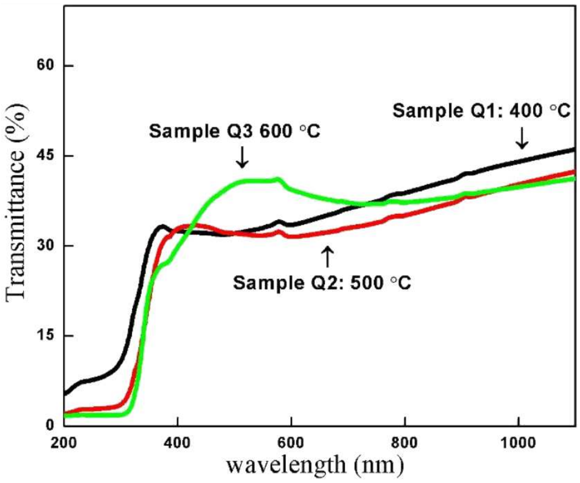

Fig. 4. Transmittance spectra of the NiO thin films prepared at different annealing temperatures.

The plots of $(\alpha h\nu)^2$ versus photon energy $h\nu$ for NiO films prepared at different annealing temperatures of 400, 500 and 600 °C are shown in Fig. 5, curves a, b and c, respectively. The deduced E_g of NiO films was found to be 3.62, 3.41 and 3.38 eV for the films annealed at 400, 500 and 600 °C, respectively. These E_g values are in good agreement with the values reported in [4, 7, 8, 13] for NiO thin films. The decrease of E_g with annealing temperature may be attributed to the increase of crystallite size and decrease of defect sites concentration [13, 14].

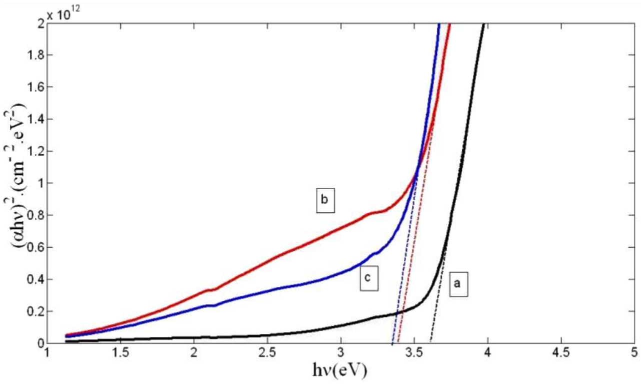

Fig. 5. The plot of $(\alpha h\nu)^2$ versus photon energy $h\nu$ for the NiO films prepared at different annealing temperatures: (a) 400 °C, (b) 500 °C and (c) 600 °C.

4. Conclusions

The nanocrystalline NiO thin films with cubic structure with (200) orientation were successfully prepared on quartz substrates by thermal annealing of DC magnetron sputtered Ni films. The average crystallite size was in range of 16–36 nm. The SEM and AFM results exhibit that the surface morphology of the NiO thin films is influenced by annealing temperature. The RMS roughness of films has increased from 1.22 nm to 4.76 nm by increasing the annealing temperature from 400 °C to 600 °C. The optical studies exhibit that the transmittance of the films increases with annealing temperature and that the optical band gap was in range of 3.38–3.62 eV.

Acknowledgments

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