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Effect of Annealing Temperatures on Structural and Morphological Properties of Copper doped Nickel Oxide Thin Films Prepared by RF Magnetron Sputtering

E. Feyzi[1], F. Hajakbari[2*], A. Hojabri[2]

[1]Department of Physics, Karaj Branch, Islamic Azad University, Karaj, Iran.
[2]Advanced Materials Engineering Research Center, Karaj Branch, Islamic Azad University, Karaj, Iran.
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Abstract

Nickel oxide (NiO) thin film is metal oxide that has attracted much attention in recent years due to its environment friendliness. In addition, by doping impurities such as copper (Cu) in NiO films the properties of prepared films can be changed. In this study, Cu:NiO thin films were deposited on silicon substrates by RF reactive magnetron sputtering. Then the deposited films were annealed at different temperatures of 200-600 °C. X-ray diffraction (XRD) results showed that all the prepared films were amorphous and the structural properties of films didn't vary by annealing temperature. Also, atomic force microscopy (AFM) images demonstrated that the surface morphology of the films was affected by annealing temperature. The root mean square (Rms) roughness and average roughness (Ra) values were obtained from AFM observations. FTIR analysis shows the peaks in the wavenumber of 528 cm^{-1} and 742 cm^{-1} which are related to Ni-O stretching mode and Cu-O bond bending vibrations.

Keywords: Sputtering, Cu, Nickel Oxide, Thin Films, FTIR.

1. Introduction

Nickel oxide (NiO) thin film is metal oxide that has attracted much attention in recent years due to its environment friendliness [1, 2]. Similar to all metallic oxides semiconductors thin films, NiO is achieving considerable attention as a promising photo-catalyst to replace powder catalysts, which are difficult in recycling [2, 3]. NiO thin films optical band gap is in range 3.5 to 4 eV and can change drastically by doping impurities such as Cu, Ag, Zn, K and so on [3-6]. Some properties of NiO thin films are unique catalytic, optical and electrical properties, stability at high temperature, durability, excellent chemical stability [3]. Several physical and chemical methods were applied to prepare NiO thin films including, DC and RF sputtering [4, 5], pulsed laser deposition [6], chemical vapor deposition [7], and so on. All these techniques offer various advantages depending on the application on interest. Among different methods employed for the preparation of thin films, the reactive sputtering is attractive due to high deposition rate, achieve the high quality thin films in large area substrates and proper control on the chemical composition and film thickness [4, 5]. Therefore, in the present work, the Cu doped NiO thin films were prepared by RF reactive sputtering. The focus of this study is to investigate the effects of annealing temperature on the structural and morphological properties of the prepared films.

*Corresponding author
Email address: fatemeh.hajakbari@kiau.ac.ir

2. Materials and Methods

In this work, in the first step, the Cu:NiO thin films were deposited onto silicon substrates by RF reactive magnetron sputtering using a Ni-Cu target (99.999% purity). The Ni-Cu composite target comprises of a Ni target (size: 76.2 mm in diameter, thickness of 1.5 mm) with four number of bonded Cu square chips (size: 10 mm×10 mm). The purity of Cu chips was 99.99%. The substrates were cleaned ultrasonically in ethanol and acetone for 20 min before introducing them into the deposition chamber. The distance between the target and the substrates was 70 mm. Before the sputtering, the base pressure of the vacuum chamber was lower than 4.5×10^{-3} mbar. During the deposition, the pressure was 4.3×10^{-2} mbar and the substrates were heated from room temperature (RT). The plasma power was kept at 60 w. The working plasma gas has a composition of Ar and O_2 by ratio; $\frac{O_2}{O_2+Ar} = 0.1$. The thickness of the Cu:NiO thin films was about 60 nm that was measured by thickness monitor. In the second step, the prepared films were annealed in the oxygen atmosphere at temperature of 200-600 °C for 60 min. The samples prepared at different temperatures of RT, 200, 400 and 600 °C were named as S1, S2, S3 and S4 respectively. The thicknesses of annealed films were determined by surface profilometer. Finally, the effect of annealing temperatures on the structural and morphological properties was studied by different analysis.

The crystal structure of the prepared thin films was evaluated by X-ray diffraction (XRD; Philips 3710) analysis with Cu Kα (λ, kαl = 1.54 A°) radiation.
For the surface morphological studies of films, atomic force microscopy (AFM, Park Scientific Instruments Auto Probe cp) in contact mode was employed. The scanning area of AFM images for all samples was 2 μm×2 μm. In order to know the chemical bonds present in the film, the prepared films were analyzed using Fourier-transform infrared spectrophotometry (FTIR; Perkin Elmer Spectrum 100) in the wavenumber range of 400-4000 cm^{-1}.

3. Results and Discussions

Fig. 1. shows the typical XRD spectra of the Cu:NiO films before and after annealing at temperature of 600 °C as a function of 2Θ, which was changed from 20° to 80° with step 0.02°. The peaks obtained at 2θ=32.96° and 2θ=68.92° are belong to silicon substrates and there were no significant peaks in the XRD patterns of the prepared films indicating that they may possibly be in the amorphous state.

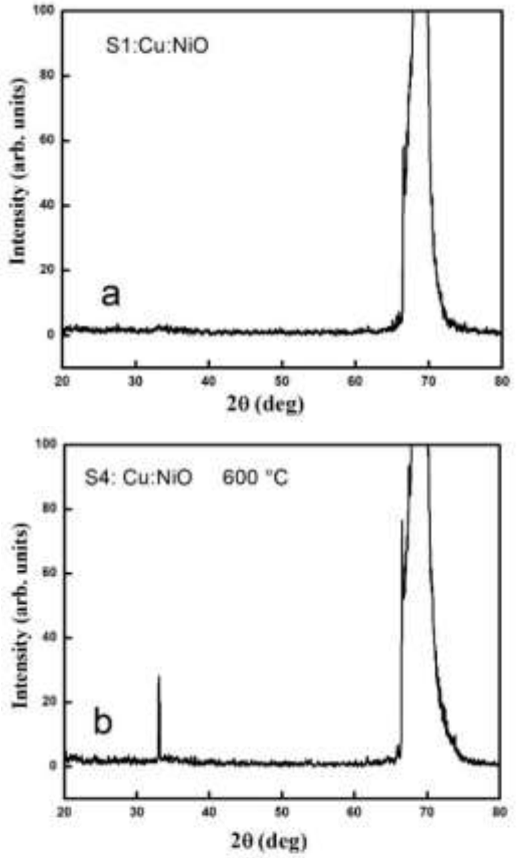

Fig. 1. Typical XRD spectra of Cu:NiO thin films grown on silicon substrates. a: before annealing and b: after annealing at temperature of 600 °C.

It is seen from the XRD patterns that all these films are amorphous and the structure of films didn't vary by annealing temperature. Similar behavior in XRD pattern was observed in ref [4].
The formation of Cu:NiO films was also confirmed from FTIR analysis. Fig. 2. shows the FTIR spectrum of Cu:NiO film grown on silicon substrate. As can be seen from Fig. 2 the absorption at 529 cm^{-1} indicated to Ni-O stretching mode which was further confirmed by the other authors [8]. The other peak at 742 cm^{-1} is associated to the Cu-O bond bending vibrations which is in agreement with reported literature [8].

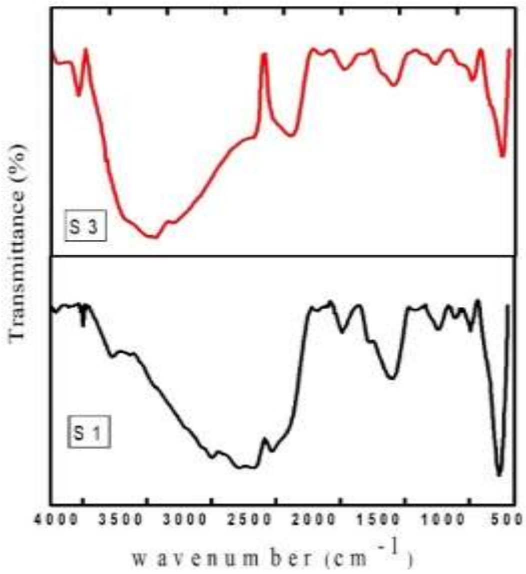

Fig. 2. FTIR analysis of Cu:NiO thin films grown on silicon substrates before (S1) and after annealing in temperature of 400 °C (S3).

Fig. 3. exhibits the 2D and 3D AFM images of Cu:NiO thin films, which were grown at different annealing temperature. It can be seen that the morphology of the films depends on annealing temperatures [9,10].
The annealed films exhibit well developed and uniformly distributed surface grains with more even surface morphology. This suggests that the heat treatment facilitates the process of grain-growth. The root mean square (Rms) roughness and average roughness (Ra) values were obtained from AFM observations and the plot of surface roughness variations for all samples were exhibited in Fig. 4. We can observe that the roughness of films decreases by annealing temperature enhancement up to 400 °C and then increased at annealing temperature of 600 °C. In this study the optimum annealing temperature for Cu:NiO films with low surface roughness was 400 °C.

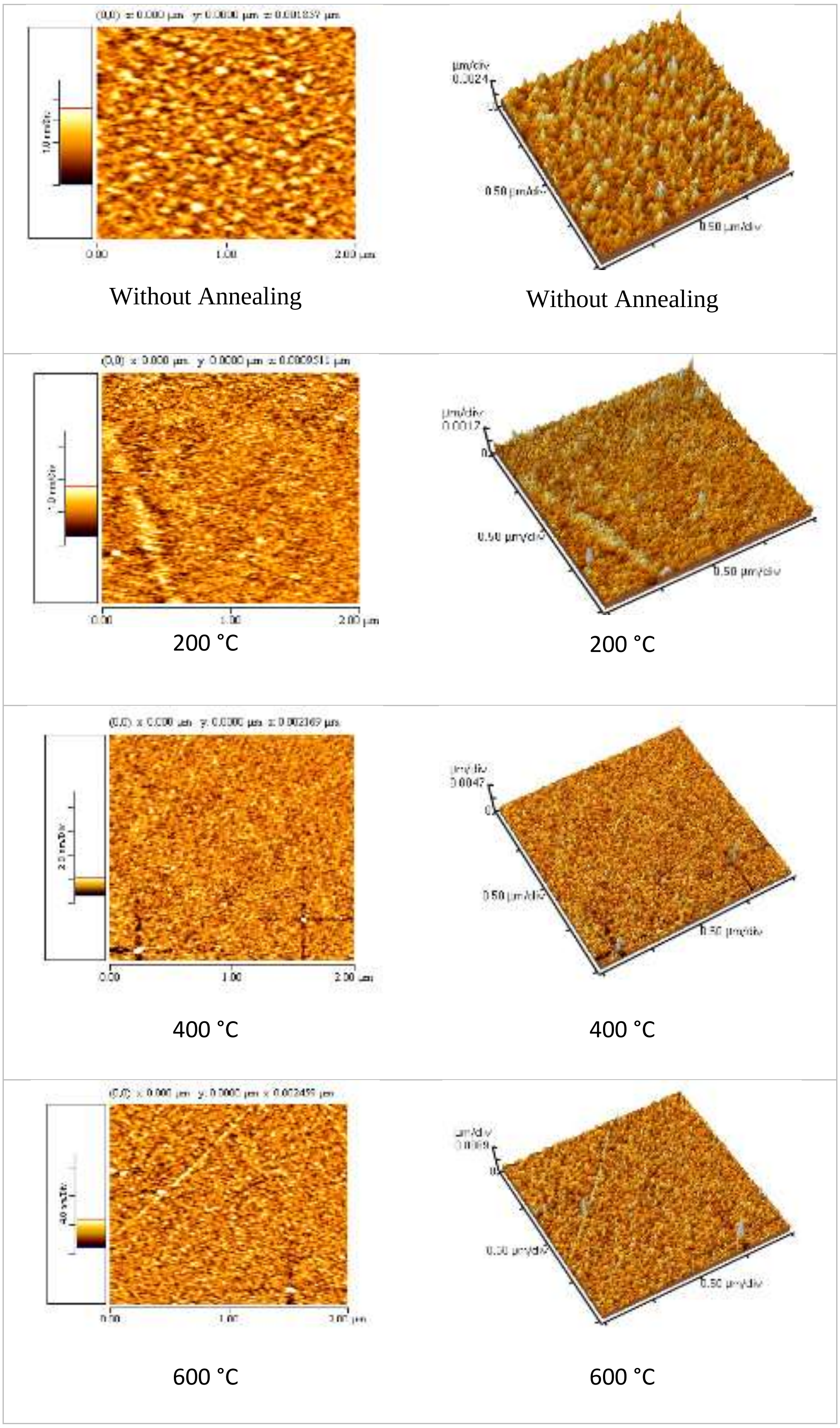

Fig. 3. AFM images of Cu:NiO thin films grown on silicon substrates before and after annealing at different annealing temperatures.

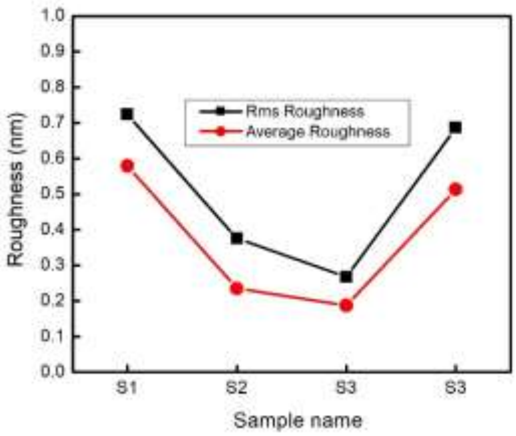

Fig. 4. Variation of surface roughness versus annealing temperature. S1: without annealing, S2: 200 °C, S3: 400 °C and S4: 600 °C.

4. Conclusion

Cu doped NiO thin films were successfully grown by the RF magnetron sputtering method onto silicon substrates. Then the deposited films were annealed at different temperatures of 200- 600 °C.

1. As a result, it is found that the morphological properties of the films were dependent on the annealing temperature and the optimum annealing temperature for Cu:NiO films with low surface roughness was 400 °C.
2. Also, all the prepared films were amorphous and the structural properties didn't change by annealing.
3. FTIR analysis confirmed the formation of Cu:NiO films.
4. It is expected that the prepared films in this work can be used in the optoelectronic devices.

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