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Original research article

Nonmetal—Metal transition in carbon films embedded by Ni nanoparticles: The temperature coefficient of resistivity (TCR), Raman spectra and surface morphology

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ABSTRACT

In this paper, carbon films embedded by Ni nanoparticles were deposited on glass substrates. Films using four mosaic targets made of carbon and nickel with Ni surface areas 1.78, 3.21, 3.92 and 4.64% are made. With increasing Ni content in these films a nonmetal–metal transition especially above room temperature region is observed. This transition is explainable by electrical measurements in temperature range 15–500 K. With increasing Ni surface areas from 1.87 to 4.64%, Ni content of films increases from59.68 to 92.89%. The electrical conductivity of dielectric films was explained by tunneling effect. Activation tunneling energy that was obtained from temperature dependence of electrical resistivity correlates both with G and D peaks of Raman spectra and depend on Ni content of films. With increasing Ni surface areas from1.78 to 3.92% due to increasing graphen like carbon, a nonmetal–metal and from 3.92 to 4.64% due to increasing amorphous carbon interconnections of Ni nanoparticles, a peculiar effect of metallic to nonmetallic state occurs. The RMS roughness of films in 3.92% has minimum value about 0.54 nm.

1. Introduction

Nanostructured materials recently have attracted attention due to their many technological applications in chemist, physics, material, pharmaceutical, agriculture [1–5]. Nonmetal–metal transition can be arising from variation of chemical composition composites of dielectric and metal [6,7]. Two fundamentally different classes of theories have been used to explain the conductivity transition in these systems [8]. In the first class, theories such as percolation have been applied to mixtures which are inhomogeneous on the macroscopic scale. In the second class, quantum theories of localization by various models such as Anderson localization, variable rang hopping and the scaling theory of localization of non-interacting electrons has been developed [8]. Tunneling and percolation in metallic and dielectric composite materials films with thickness 2–5 mm was investigated by Toker et al. They argued that all-connected tunneling network can be reduced to a well-defined percolation network [9]. In this work, we prepared Ni nanoparticle in carbon films with various Ni content by RF magnetron sputtering. Our specific deposition conditions including room temperature and non-wet chemical deposition are prerequisites for applications in optical and electronic devices. In the previous reports we the dependence of the structural properties

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on the deposition times, deposition rate, and annealing temperature in carbon films embedded by nickel nanoparticles investigated [10–17]. In this paper, we study variation of electrical resistivity versus Ni content obtained from different mosaic targets for films with same deposition time. With variation of Ni content, nonmetal-metal transition is observed and is explainable by the temperature coefficient of resistivity (TCR), Raman spectra studies, activation tunneling energy and AFM images.

2. Experimental details

The amorphous carbon films doping by nickel nanoparticles were deposited by RF magnetron sputtering using four mosaic targets on glass substrates. The substrates were first ultrasonically cleaned with ethanol for 20 min followed by deionized water. Before deposition of films, the mosaic targets were pre-sputtered with Ar ions plasma at a substrate bias about of−250 V for 2 min in order to remove oxide layers and adsorbed impurities on the surfaces. Stripes of Ni are attached on the pure graphite target with a length about of 40 mm. The Ni content in the grown films can be controlled by variation of number of the Ni stripes. The number of stripes is varied as 10, 18, 22, and 26 attached to the graphite race track, that corresponds to approximately 1.78, 3.21, 3.92 and 4.64% in area, respectively. The films deposited have Ni atomic ratios of 59.68, 86.47, 89.91 and 92.89 wt.%, respectively. The background pressure of the sputtering chamber was 2×10^{-5} mbar, and the working pressure was 2×10^{-2} mbar. The films deposited at same deposition time 3 min and in constant RF power regime 400 W. The angle of incident ions with target surface was 90°, and the substrate-target distance was 60 mm. Atomic forces microscopic (AFM) analysis on non-contact mode was used to obtain surface morphology properties. The Root Mean Square (RMS) roughness was obtained from AFM data (WSxM software-2007). The field emission scanning electronic microscopy (FESEM) images were used for the morphological characterization. Energy dispersive X-ray studies (EDAX) were carried out with AMETEK EDAX analyzer. Raman studies were performed using a He–Ne laser 633 nm Thermo Nicolet FTRAMAN 960 spectrometer. Using the four points contact method the direct currents electrical conductivity of films were measured by cooling samples in a continues He flow in a cryogenic units (optical low temperature model CCS 450 USA) in a thermostatic chamber. The high voltage power supply (ORTEC 456, USA, 0–3 kV), Metrix VX102A and Keithley 196 DMM electrometers at temperatures range 15–500 K, were used for voltage and current measurements, accordingly.

3. Results and discussion

Fig. 1(a) shows Ni concentration of films with Ni surface areas1.78, 3.21, 3.92 and 4.64% on mosaic targets. With increasing surface of Ni stripes on graphite target, Ni concentration of films is increased. Three conduction regions A (dielectric), B (metallic), and C (dielectric) are distinguished. In the region A, the Ni surface area is less than 3.21%, the electrical resistivity is large and the temperature coefficient of electrical resistivity is negative. In the region B, the Ni surface area is about3.92%, the electrical resistivity is small and the temperature coefficient of electrical resistivity is positive. In the region C, with increasing Ni surface area over 4% a change from metal to nonmetal one is acquired. The electrical conduction in composite systems comprising of metallic and dielectric component is determined by two mechanisms; percolation in a continuous conducting network and tunneling between isolated conducting particles [18]. For high metallic content in granular materials, a continuous network by the percolating metallic particles is formed. Fig. 1(b) shows variation of electrical resistivity of films with temperature in range 15–50 K. The electrical resistivity of films in region A and C with increasing temperature decreases showing the semiconducting behavior, and in region B it specially above room temperature is nearly an increasing function of temperature which showing the metallic behavior. Fig. 1(a) shows that with increasing Ni surface area from 1.78 to 4.64%, Ni concentrations of films increase. As we expect and shown in Fig. 1(b), with increasing metal concentrations the resistivity of films is decrease and for films deposited at a transition from nonmetal to metal occurs which it is inconsistent with an increasing function with temperature. Due to disappearing of metaling paths in films deposited at 4.64% in spite of increasing metal concentrations, the resistivity of films increases. It is due to a transition from metal to nonmetal which is inconsistent with a decreasing function of films with temperature. The films in regions A and C have a nonmetal behavior however the films in regions B have a metal behavior. In manuscript we revised regions in Fig. 1(a), films deposited at 1.78 and 3.21% are in region A, films deposited at 3.92% are in region B and films deposited at 4.64% are in region C. Fig. 1(c) shows the typical mosaic target consist of pure graphite target and 10 Ni stripes. The values of Ni content in the deposited films can be controlled by variation of number of Ni stripes on graphite target. The number of these stripes is varied as 10, 18, 22, and 26 attached to surface of the graphite target race track. The values of estimated areas of Ni stripes on graphite targets surface corresponds to approximately 1.78, 3.21, 3.92 and 4.64% in total area of graphite surface, respectively.

Fig. 2(a) and (b) show variation of resistivity R(T)/R(300 K) with temperature and values of the temperature coefficients of resistivity (TCR) with Ni surface areas, respectively. The values of resistivity of films which normalized to room temperature and TCR of the films at 3.21, 3.92 and 4.64% is not large, we show that even such minor differences provide useful information, and they can be clearly correlated with relevant structural data. The films deposited at Ni surface area 3.92% show positive TCR of 0.0013 K^{-1}, which implies a degenerate semiconductor or nearly metallic type of conductivity. It means that in these films there must exist at least a single percolation conduction path formed by particles with metallic character. As shown in Fig. 2(b), composite films prepared at Ni surface area 1.78, 3.21 and 4.64% showed a slightly negative TCR of about −0.00471, −0.00118 and −0.00157 K^{-1}, respectively. The films deposited at Ni surface areas 1.78, 3.21 and 4.64%, as shown in Fig. 2(a), have a decreasing function with temperature; therefore they are semiconductor and it expect that they have