Modification in properties of Ni-NWs meshes by Ar+ ions beam irradiation

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Abstract

Influence of 30 kilo-electron-volt (keV) Argon (Ar+) ions on optical and electrical properties of nickel nanowires (Ni-NWs) meshes is being reported. Ni-NWs are being exposed to 30 keV Argon (Ar+) ions at various beam fluences. These fluences of Ar+ ions are $7 \times 10^{14}$ ions cm$^{-2}$, $3 \times 10^{15}$ ions cm$^{-2}$ and $3 \times 10^{16}$ ions cm$^{-2}$. After irradiation, Ni-NWs meshes were analyzed through transmission electron microscopy technique (TEM). The structural analysis has been done through X-ray diffraction technique. It is found from TEM results that atoms are sputtered out from surfaces of Ni-NWs due to collision cascade effect persistently and lead to reduce the diameters or thicknesses of Ni-NWs. X-ray diffraction results reveal that crystalline quality is reduced under Ar+ ions irradiation which may be due to defects induced in Ni-NWs as a result of collision cascade effect. The Ni-NWs meshes are characterized optically and electrically through UV–VIS spectroscopy and four probe techniques. The optical transparencies of Ni-NWs meshes are increasing with increase in beam fluence of Ar+ ions. The electrical conductivity of the mesh is decreased continuously with increment in beam fluence of Ar+ ions which might be due to production of defects in Ni-NWs. The tuning of optical transparency and electrical conductivity of Ni-NWs meshes is required for their application as successful transparent electrodes in optoelectronic nanodevices.

1. Introduction

Transparent Electrodes (TEs) are key components of optoelectronic devices such as light emitting diodes (LEDs), solar cells, electronic displays etc. The important features of TEs are high values of optical transparencies and electrical conductivities which makes TEs capable to be exploited as components of optoelectronic devices [1]. Commercially, indium tin oxides (ITO) are most excellent TEs but due to some challenges of TEs such as high production costs, brittleness of ITO and dearth of indium are major reasons for their demise. Besides, it is also not easy to protect the underlying substrates of organic material during sputter deposition [1–4]. In fact, these negative aspects of ITOs make it indispensable for scientific community to seek out alternative materials to compete with ITO and replace it from market.

Many nanomaterials come on frontage to compete with ITO based TEs which includes meshes of carbon nanotubes (CNTs), metallic nano-grid and metallic nano-wires (MNWs) meshes and thin layers of graphene [5–7]. However, due to excellent physical properties, high aspect ratio and mechanical flexibility of MNWs
meses based TEs; TEs based on MNWs are proven as an excellent replacement of ITO. Another positive characteristic of MNWs meshes based TCEs is the simplicity of synthesis procedures [8]. Simple solution process techniques such as spin coating [9, 10], rod coating [11, 12], drop casting [13], spray coating [14–17] can be employed to turn out thin films of meshes of MNWs. From aforementioned deposition techniques, drop casting is rather simple, easy and economical which needs no costly equipments. Moreover, consumption of material is less while depositing thin film by drop casting technique and managing of colloidal solution is also easy [10, 18]. The main challenges of MNWs meshes for their integration in optoelectronic devices as TEs are enhancement in electrical conductivity and optical transparency [18].

In recent years, ions beam irradiation technique has been frequently employed to modify the properties of various nano-scale materials such as NTs/NWs [19]. Ahmad et al used keV Ar+ ions beam irradiation technique to modify the properties of carbon nano-tubes (CNTs) [20]. Similarly, in one of our previous report, welded Ni-NWs were exposed to MeV H+ ions. It was found that electrical conductivities and optical transparencies of Ni-NWs meshes are enhanced after ion beam irradiation [21]. In present report, meshes of Ni-NWs are produced on glass substrates by drop casting method in form of thin film. Thereafter, these meshes are exposed to various fluencys of beam of keV Ar+ ions. Diameters or thicknesses of Ni-NWs are reduced by exposing Ni-NWs to keV Ar+ ions. Optical transparencies of Ni-NWs meshes is increased which may be attributed to thinning of Ni-NWs under the influence of Ar+ ions beam irradiation which consequently increases the gaps between individual Ni-NWs in the mesh. This increment in gaps between Ni-NWs would result in enhancement in optical transparencies of Ni-NWs meshes. However, the electrical conductivities of the Ni-NWs meshes is decreased continuously with increase in beam fluence of Ar+ ions due to production of defects in Ni-NWs. For the first time, this novel technique is exploited to tune the optical and electrical properties of Ni-NWs meshes.

2. Experimental section

Ni-NWs with diameters ranging from 200–300 nm and lengths approximated 100 to 200 μm were supplied from PlasmaChem (Germany). Initially, the Ni-NWs were in the form of wool type fibre. The wool type fibres of Ni-NWs were converted into aqueous dispersion in ethanol. 5 mg of Ni-NWs wool fibres was dispersed in 1 ml of iso-propanol solution. Thereafter, glass substrates and copper grids were coated with Ni-NWs solution using a drop casting method.

For structural, optical and electrical characterization of samples, the coated glass substrate was then cut into small pieces for Ar+ ions irradiation. For TEM characterization, samples of Ni-NWs dispersed in to copper grids placed on glass substrate were used. Thereafter, the finally prepared samples of Ni-NWs were irradiated with 30 keV Ar+ ions beam an ion implanter with different fluencies (7 × 1014 ions cm−2, 3 × 1015 ions/cm2 and 3 × 1016 ions cm−2) at room temperature. The diameter of beam was ~4 mm and irradiation time was ~2 h for each sample. Ar+ ion beam irradiation energy, current and substrate temperature were 30 keV, 0.05 μA and room temperature respectively. The prepared samples of Ni-NWs were analysed structurally through X-ray diffraction (XRD) technique using BRUKER AXS (Germany) D8 Advance X-ray diffractometer with monochromatic CuKα radiation (λ = 1.5418 Å). The 2Θ scanning range was selected to be 40–80° to cover all diffraction lines of Ni-NWs, while the morphology was investigated by transmission electron microscopy (TEM) technique using FEI Tecnai G2 20 Field Emission Gun (FEG) with a resolution of approximately 0.2 nm. Ni-NWs meshes are characterized electrically using Keithley Source meter Model 2400 at room temperature before and after each irradiation. For optical characterization, Perkin Elmer Lambda 950 UV–vis–NIR spectrophotometer was used, with uncoated glass slide as reference slide. After irradiation, to see the implantation of Ar+ ions within lattices of Ni-NWs, a computer simulation program Stopping Range of Ions in Matter (SRIM) was used [22].

3. Results and discussions

The Ni-NWs were analyzed through transmission electron microscopy (TEM) technique before and after being exposed to Ar+ ions. TEM images of un-irradiated Ni-NWs are shown in figure 1(a). Un-irradiated Ni-NWs are having diameters approx. 200–300 nm and it is seen in figure 1(a). However, after being irradiated at a dose 7 × 1014 ions cm−2 of Ar+ ions, the diameters of Ni-NWs is shortened to 100–120 nm and displayed in figure 1(b). Similarly, diameters of Ni-NWs is reduced up to 60–80 nm after exposure to Ar+ ions at a fluence 3 × 1015 ions cm−2 and this shortening of diameters is shown in figure 1(c). After irradiation at a dose ~3 × 1016 ions cm−2, diameters of Ni-NWs are further reduced up to 25–40 nm and shown in TEM image of figure 1(d). Some of Ni-NWs with smaller diameters are also highlighted by yellow lines in respective TEM figures at all beam fluencies.
The diameters or thicknesses of Ni-NWs might be reduced because of sputter-induced atomic ejections from surfaces of Ni-NWs after being exposed to Ar\(^+\) ions. Due to continuous striking of Ar\(^+\) ions on surfaces of NWs induces collision cascade effect and this effect leads to sputter/eject atoms from surface of Ni-NWs and consequently Ni-NWs becomes thin [22].

In fact, there are two modes of collisions of energetic ions with lattices of Ni-NWs, either elastic or inelastic mode. Elastic mode of interaction between an energetic ion and Ni-NWs will produce the series of impacts with atoms in lattices and finally releases its kinetic energy due to these impacts. As a result of series of impacts, a series of recoiling atoms is generated in the NWs and this process will keep on in the similar way. This process is called collision cascade effect. As a result of collision cascade effect, some recoiling atoms gain sufficient amount of energy that is required to eject the atom out from NW’s lattice and may cause it to move along surfaces of NW. The penetration depth of each recoiling atom in the NW is different which depends on kinetic energies of recoiling atoms [23]. If the atoms that ejected out from surfaces of NWs are large in number, then diameters of NWs would be reduced. If energetic ions which are interacting with NWs have energy in keV range, then nuclear collision would be dominant and atoms would be ejected out due to collision cascade effects [24, 25]. The ejection of atoms from surfaces of NWs is also verified through thinning of diameters of NWs after exposure of NWs to low energy (keV) Ar\(^+\) ions in TEM images of figures 1(a)–(d). Schematic diagram of thinning of Ni-NWs under the influence of keV Ar\(^+\) ions irradiation is also shown in model diagram of figures 2(a)–(c).

Analysis of structural alterations in Ni-NWs after exposure to different fluencies of Ar\(^+\) ions has been done by X-ray diffraction (XRD). The main purpose of analyzing samples through XRD is to see structural stability of Ni-NWs under beam irradiation. It is found from XRD results in figure 3, un-irradiated Ni-NWs reveal 2\(\theta\) peaks which corresponds to planes (111) and (200) that are associated with cubic structure of Ni-NWs. After irradiating at various fluencies of Ar\(^+\) ions such as \(3 \times 10^{15}\) ions cm\(^{-2}\) and \(3 \times 10^{16}\) ions cm\(^{-2}\), the crystalline structure would remain unaffected. Moreover, spectra in figure 3 are showing the reduction in intensities of XRD peaks with increment in dose of Ar\(^+\) ions. The defects or vacancies generated by Ar\(^+\) ions irradiation would decrease the intensities of XRD peaks. These defects are assembled together in form of amorphous regions and
reduce the crystallinity of NWs and might be generated due to collision cascade effect. As it is mentioned in TEM figures of respective irradiated Ni-NWs meshes (figures 1(b)–(d)), the diameters of Ni-NWs is reduced in comparison with un-irradiated Ni-NWs. The thinning of Ni-NWs diameters confirms the atomic removal from surfaces of NWs. Therefore, TEM results are confirmed by XRD results.
The value of grain size (i.e. crystallite size) of Ni-NWs is estimated using X-ray diffraction data. In this method, it is realized that sharp XRD peaks are might be due to large size grains. Scherrer’s formula was employed to calculate the crystallite size or grain size $D$ [26]:

$$D = \frac{K \lambda}{\beta \cos \theta}$$  \hspace{1cm} (1)

where $\beta$ is in radians and indicates the broadening of diffraction line measured at half of its maximum intensity (FWHM), $\lambda = 1.5418$ Å shows the wavelength of X-rays, $\theta$ is the angle of diffraction and $K$ is the shape factor. For polycrystalline film, value of $K$ is 0.9 [27]. In figure 3(b), the variation in sizes of grains (average) versus fluence of $Ar^+$ ions is shown. Before and after irradiation, the values of average grain sizes of Ni-NWs are 4.56 nm, 4.44 nm and 4.40 nm respectively. In XRD results of figure 3(a), sharp diffraction peaks are the indication of large crystallite size. After ion beam irradiation of Ni-NWs, the values of crystallite sizes have been reduced from 4.44 nm to 4.28 nm with increments in beam fluencies from $3 \times 10^{15}$ ions cm$^{-2}$ to $3 \times 10^{16}$ ions cm$^{-2}$ (see in figure 3(b)).

The crystallite sizes have might be reduced due to loss in crystallinity of NWs [23]. The loss in crystallinity is observed in Ni-NWs after irradiating with low energy $Ar^+$ ions at high beam fluencies whereas Ni-NWs are highly crystalline in nature after irradiating at low beam fluencies [23].

The optical properties of nanoscale materials are different than bulk materials and originate from surface plasmonic resonance effect. When the electromagnetic field will interact with conduction band electrons of Ni-NWs, electric field (oscillating) will produce. This electric field will produce excitation in conduction band electrons at surfaces of Ni-NWs and cause them to displace with respect to nuclei in form of electronic cloud. The coulomb force of attraction will originate between nuclei of materials and electronic cloud will cause electronic cloud to oscillate with respect to nuclei. The effect of oscillations of electronic cloud or conduction band electrons collectively on NWs’ surfaces is called surface plasmonic resonance effect [18, 27]. In this band, transmittance of light is reduced and scattering and absorption of light in UV region is increased. A very interesting aspect of surface plasmonic band of MNWs is the exploitation of this band in tunable and controllable optoelectronic properties. However, increment has been observed in values of the optical transmittance of Ni-NWs meshes with beam fluence of $Ar^+$ ions and this trend can also be observed in figure 4. This increment in transmittance is associated with increase in spaces between Ni-NWs. This increase in spaces between NWS was also confirmed through TEM results of figures 1(a)–(d).
The spaces between Ni-NWs are increased due to thinning of Ni-NWs or removal of atoms from surface of Ni-NWs as an outcome of collision cascade effect. Optical characterization of Ni-NWs meshes is a powerful tool which gives information about optical transmittance, absorption edge, extinction coefficient, and absorption coefficient of nanomaterials and so on. For polycrystalline films, information about the absorption coefficient is imperative. The value of absorption coefficient of Ni-NWs meshes is less which makes it appropriate nanoscale material for application in optoelectronic devices as transparent conducting electrode.

Before and after irradiating Ni-NWs meshes with Ar$^+$ ions at various beam fluences $7 \times 10^{14}$ ions cm$^{-2}$, $3 \times 10^{15}$ ions cm$^{-2}$, and $3 \times 10^{16}$ ions cm$^{-2}$, the absorption coefficients, $\alpha$, is measured by using the values of transmittance, $T$, of Ni-NWs meshes using formula [29]:

$$\alpha = \frac{1}{d} \ln \left( \frac{1}{T} \right) \quad (2)$$

$d$ is the thickness of thin film of Ni-NWs mesh in above relation.

Before and after irradiating Ni-NWs meshes with Ar$^+$ ions at fluencies $7 \times 10^{14}$ ions cm$^{-2}$, $3 \times 10^{15}$ ions cm$^{-2}$ and $3 \times 10^{16}$ ions cm$^{-2}$, absorption spectra is shown in figures 5(a)–(d). It is found that Ar$^+$ ions beam irradiation is key parameter which affects the absorbance of film.

The co-efficient of extinction ‘$k$’ could be measured by formula [30]:

$$k = \frac{\alpha \lambda}{4\pi} \quad (3)$$

‘$\lambda$’ indicates wavelength and ‘$\alpha$’ is absorption co-efficient in the above relation. In figure 6, plots of extinction co-efficient versus wavelength for Ni-NWs meshes are given. It is observed from figure 6 that value of co-efficient of extinction is greatest at wavelength around 400 nm. However, a monotonous decrease is found in value of extinction co-efficient decreases with increase in wavelength above 300 nm; this might be due to decrease in absorption co-efficient.

After TEM and XRD analysis of samples, conductivity of Ni-NWs meshes is recorded using four probe technique before and after Ar$^+$ ions irradiation. At first beam fluence of Ar$^+$ ions $\sim 7 \times 10^{14}$ ions cm$^{-2}$, electrical conductivity was decreased in comparison with un-irradiated samples. Upon increasing beam fluence of Ar$^+$ ions upto $3 \times 10^{15}$ ions cm$^{-2}$, the electrical conductivity of Ni-NWs meshes is further decreased. This decrease is might be due to sputtered atoms of Ni agglomerated in form of amorphous regions [18, 31]. In TEM results (see in figure 1), Ni-NWs with reduced diameters after Ar$^+$ ions beam irradiation are confirming that atoms are sputtered /ejected from surface of Ni-NWs due to collision cascade effect. Similarly, XRD spectra in figure 3(a) are showing the reduction in intensities of XRD peaks after Ar$^+$ ions beam irradiation which is the indication of existence of defects due to collision cascade effect [18, 31, 32]. The relative conductivity of Ni-NWs network is decreased to a relative value of 0.5 at a dose $\sim 7 \times 10^{14}$ ions cm$^{-2}$. The variation in the conductivity of carbon nanotubes and silver nanowires networks after ion irradiation has already been reported in one of our previous research work [10, 14]. With further increase in Ar$^+$ ions beam fluence, i.e., at a dose $\sim 3 \times 10^{16}$ ions cm$^{-2}$, Ni-NWs network becomes less conductive.

Figure 5. (a)–(d) Plots of absorption co-efficient versus wavelength for Ni-NWs networks.
Another reason for reduction in electrical conductivity is might be increase in contact resistance on junction points between NWs; hence a resistant path is offered to electrons at junction locations. A plot of conductivity versus irradiation dose of Ar\textsuperscript{+} ions is shown in figure 7 to observe the variation of electrical conductivity in Ni-NWs with irradiation fluence of Ar\textsuperscript{+} ions. SRIM (Stopping Range of Ions in Matter) computer simulation software has been used to evaluate the variation of conductivity with Ar\textsuperscript{+} ion irradiation in Ni-NWs. The calculations were done for a Ni layer with parameters: i) 30 keV (Energy of Ar\textsuperscript{+} ions), ii) 1500 ions, iii) incident angle of 0°. The graphs of ions paths or trajectories (depth versus Y-axis) and events of impacts have been plotted while covering a full cascade of damage. Vacancy created by an incident Ar\textsuperscript{+} ion in the ion path is indicated by a red dot which means that a Ni atom is removed from its lattice site (figure 8(a)).

Moreover, table 1 is given to represent the simulation results for the interaction of Ar\textsuperscript{+} ion in Ni. The loss of energy due to Ar\textsuperscript{+} ions and recoils is shown in table 1 for energy of Ar\textsuperscript{+} ions 30 keV and incident angle was 0°. Loss of energy to target electrons is described by term ionization.

The energy transferred directly from Ar\textsuperscript{+} ions to Ni atom is represented by data related to ’Ions’ in the table 1. Similarly, the energy transferred from recoiling Ni atoms to their electrons is represented by data related to ’Recoils’ in the table 1. It is observed from computer simulation ‘SRIM’ that defects are generated in Ni layer due to hitting of Ar\textsuperscript{+} ions in form of vacancies.
It is observed from computer simulation that conductivity of Ni-NWs would be reduced due to defects induced in Ni-NWs. Due to production of these defects, the contact resistance is increased and conducting path length will also be increased that cause the reduction in conductivity. From XRD and computer simulation results, it is seen that defects are generated and lead to reduce the conductivity and grain size. In case of Ni-NWs, defects induced due to $\text{Ar}^+$ ions beam irradiation are more in comparison with ion beam irradiation induced heat or ionization. This means that if path length is decreased by ion induced defects or vacancies, conductivity will decrease. Similarly, in high fluence case (fluence of $3 \times 10^{16}$ ions cm$^{-2}$), both effects are introduced, i.e., defects increase as well as path length decrease due to loss in crystallinity of nanowires. Ultimately electrical conductivity decreases.

4. Conclusions

Optical transparencies are modified by $\text{Ar}^+$ ions beam irradiation-induced thinning of Ni-NWs. Atomic ejections induced by $\text{Ar}^+$ ions beam irradiation from surfaces of Ni-NWs would result in reducing the thicknesses of Ni-NWs. Spaces in individual Ni-NWs would be increased due to reduction in thicknesses of Ni-NWs in the mesh. $\text{Ar}^+$ ions beam irradiation is found to be useful method to reduce diameters of Ni-NWs via sputtering of atoms from Ni-NWs surfaces that may be exploited to tune optical properties of Ni-NWs meshes for optoelectronic applications. XRD analysis revealed preserved crystalline structure of Ni-NWs during ion beam irradiation with a 30 keV $\text{Ar}^+$ ions. Electrical conductivity is decreased with increase in beam fluence of $\text{Ar}^+$ ions which might be happened due to loss of crystalline structure of Ni-NWs or increase in contact resistance between Ni-NWs. Before and after $\text{Ar}^+$ ions irradiation, TEM analysis of Ni-NWs is completely different from each other and diameters of Ni-NWs are diminishing sequentially with boost up of beam fluencies of $\text{Ar}^+$ ions.

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Conflict of Interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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