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Research Paper

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Effect of Ni doping on the photovoltaic conversion efficiency of ZnO nanostructured dye sensitized solar cells

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Abstract— In this work, pure and Ni-doped ZnO nanostructures with dopant concentrations (2%, 4%, 6%) of Ni are successfully prepared via flame synthesis. The as-synthesized nanostructures are characterized using TEM, XRD, and FTIR spectrophotometer. Further the application of Ni doped ZnO nanostructures in dye sensitized solar cells (DSSCs) is investigated. The photovoltaic performance of DSSCs with photoanode of Ni doped ZnO nanostructures show the increase in efficiency from 3.76 % (ZnO) to 5.01 % and 5. 65 % with the dopant (Ni^{2+}) concentration 2 % and 4% respectively. With the further increase in dopant concentration (6%) decrease in efficiency (3.53%) is observed, which may be due to increase in particle size and Burstein-Moss effect.

Keywords— ZnO, flame synthesis, DSSC

I. INTRODUCTION

In the last two decades, semiconductor nanostructures are of fundamental importance because of their potential applications in sensors, solar cells, optoelectronic and optical devices [1-4]. Many of the oxide materials like ZnO, SnO_2, MoO_2, NiO and TiO_2 have been investigated for dye sensitized solar cells. Among them ZnO is an important functional photoelectrode material for dye sensitized solar cells because of its direct energy band gap 3.37 eV, exciton binding energy (60 meV) and availability in different morphological structures [5-8]. Bulk ZnO is an insulator but shows semiconductor behavior at nanoscale. This semiconductor behavior of ZnO is attributed due to deviation in stoichiometry. It is the well known fact that the efficiency of DSSCs depends upon photoanode, dye, electrolyte and counter electrode. Photoanode material plays an important role in DSSCs as it serves as electron-transport layer. Doped ZnO nanostructured electrode in dye sensitized solar cells shows relatively low resistivity, high transmittance, and chemical stability than the undoped ZnO nanostructured electrodes [9-12]. In recent years, many researchers have reported their work for improved efficiency of DSSCs by using various dopant such as Sn [13], B [14], Al [15-16], Cu [17], Eu [18], Ga [19], La [20] in ZnO. Ni doped ZnO nanoparticles also show some interesting magnetic, optical and electric properties [21-22]. Ni^{2+} is one of the most efficient doping element to tune the optical and electrical properties of ZnO due to its chemical stability while replacing Zn^{2+} ion. In the present work, Ni doped ZnO nanostructures are prepared by using flame synthesis and as-prepared nanostructures are characterized by using Transmission electron microscopy (TEM), X-ray diffraction (XRD), Fourier transform infrared spectrometer (FTIR). Further, these nanostructures are used as photoelectrode material of the dye sensitized solar cells to study the effect of Ni doping on the photovoltaic conversion efficiency (PCE) of ZnO nanostructured DSSCs.

II. EXPERIMENT

Preparation of ZnO and Ni doped ZnO

The flame synthesis technique also known as solution combustion technique is used to prepare ZnO and Ni doped ZnO nanostructures. A stoichiometric mixture (molar ratio 1: 1.66) of zinc nitrate solution and urea solution are heated in muffle furnace, foamed and ignited to burn with an incandescent flame yielding ZnO nanostructures. The reaction mechanism for solution combustion synthesis of ZnO nanostructures is :

$$3\ Zn\ (NO_3)_2\ 6H_2O\ (aq) + 5\ NH_2CONH_2\ \ (aq)$$
$$\downarrow 500^0C$$
$$3ZnO\ (s) + 5CO_2\ (g) + 8\ N_2\ (g) + 28\ H_2O\ (g)$$

Ni (2%, 4%, 6%) doped ZnO nanostructures are prepared by adding appropriate amount of nickel nitrate into the mixture of zinc nitrate and urea solution.

Characterization techniques

The as-prepared samples are characterised using TEM, XRD, and FTIR at room temperature. For TEM characterizations, the nanostructures are sonicated for 8 hours in ethanol. In

order to study the effect of ZnO and Ni doped ZnO nanostructures as photoelectrode material in the dye sensitized solar cells, dye extracted from Syzygium Cumini (Jamun), Iodine/Tri Iodide as electrolyte is used. Carbon coated ITO glass is used as counter electrode.

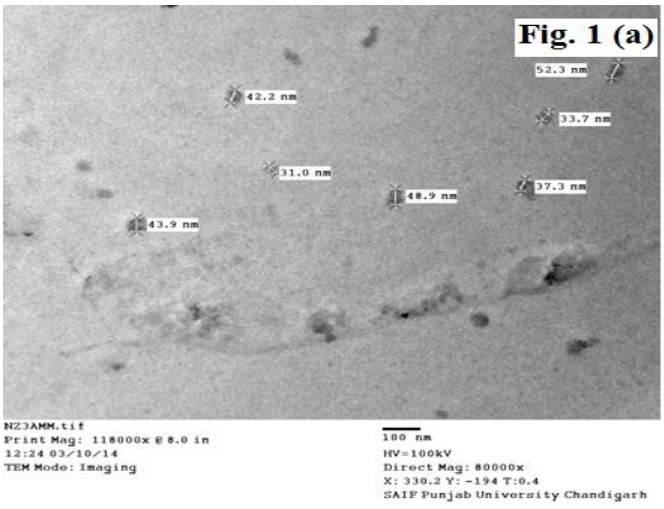
III. RESULTS AND DISCUSSION

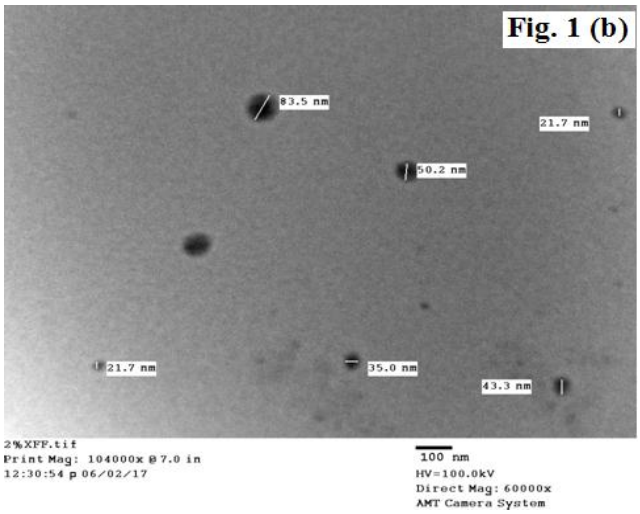
TEM characterizations

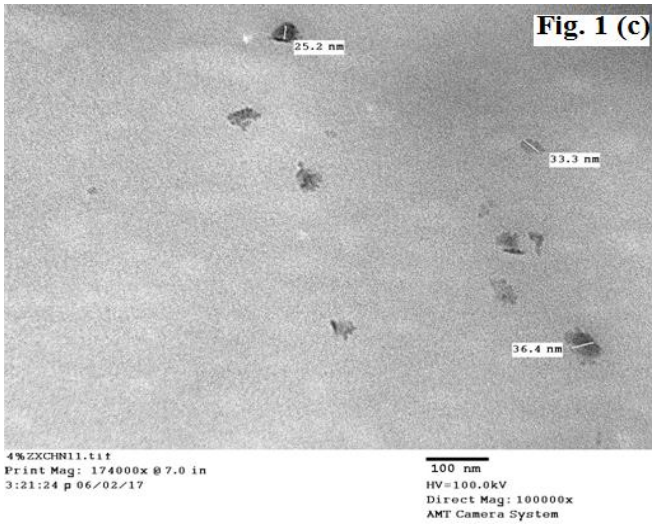
The crystallite size confirmed by TEM observations is shown in Fig. 1. The slight decrease in particle size is observed with the increase in doping from 2% to 4% which may be due to creation of defects such as oxygen/Zn vacancies and interstitials. The nucleation in doped ZnO is suppressed by these defects and formation of Zn-Ni bonding structure (substitution of Zn in ZnO by larger ionic radius i.e. Ni^{2+} ions), which subsequently limits the growth of ZnO nanostructures. The increase in particle size is observed when the doping of Ni^{2+} is further increased from 4% to 6%. With the increase in dopant concentration mismatch between the ionic radii of zinc and nickel ions produces distortion around the Ni^{+2} which may increase the particle size**.**

X-ray diffraction (XRD) analysis

Fig.2 shows the XRD spectra of ZnO and Ni doped ZnO nanostructures. It is observed from the spectra that with 4% and 6% Ni doping, a secondary NiO phase appears in addition to the peaks corresponding to ZnO whereas no such phase appear for Ni^{2+} (2%) ZnO nanostructures. XRD investigations show the hexagonal wurtzite structure of ZnO and shows that the Ni^{2+} ions are well incorporated into the crystal lattice of ZnO without altering the overall crystal structure. The substitution limit for doping Ni^{2+} ions into ZnO has also been confirmed by Anbuselvan et al [22] i.e. 3%.







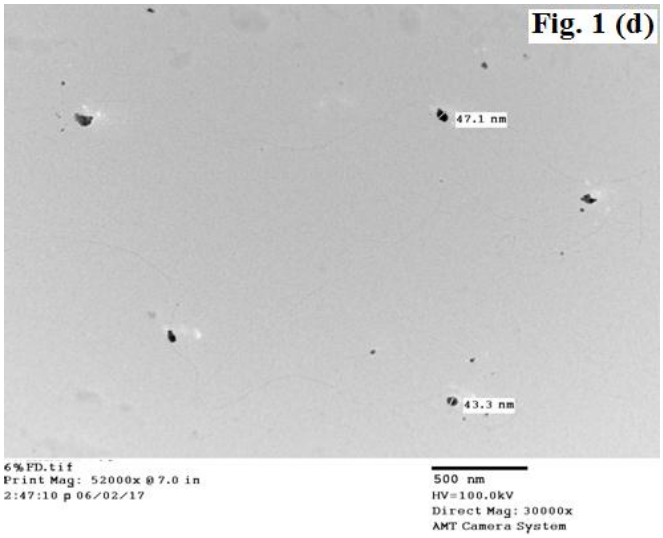
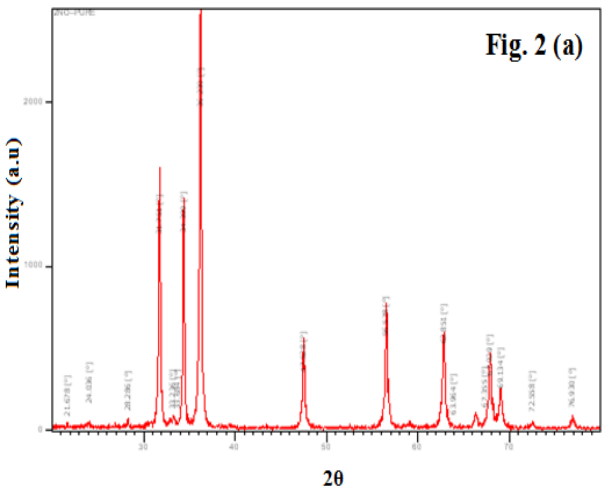
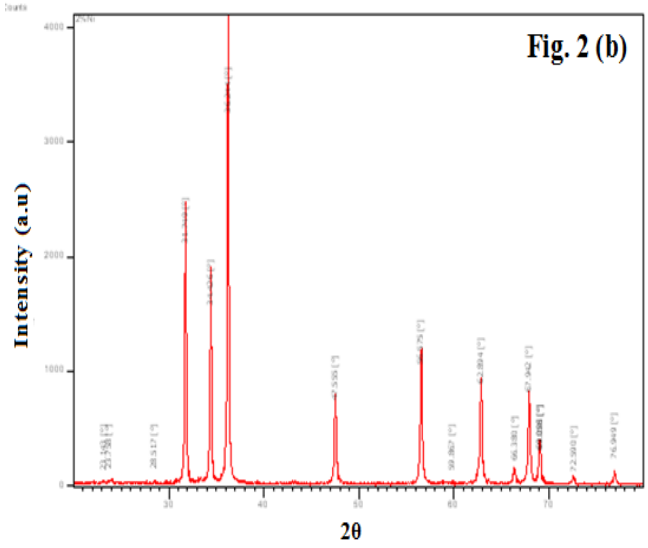
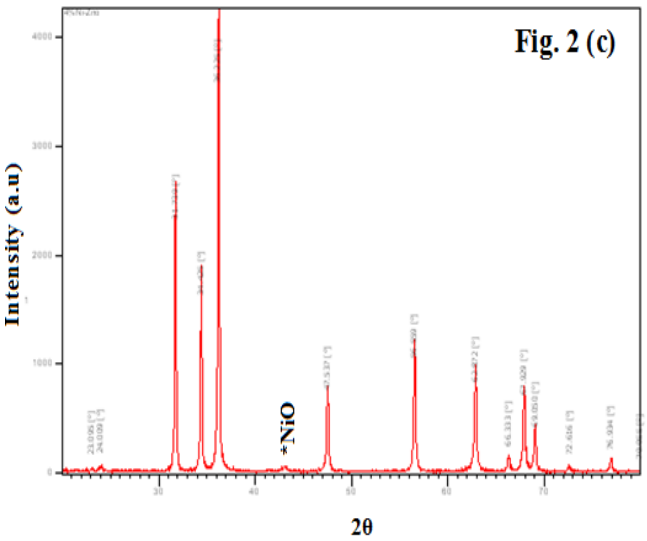


Fig. 1: TEM spectra of (a) ZnO nanostructures (b) Ni (2%) doped ZnO nanostructures (c) Ni (4%) doped ZnO nanostructures (d) Ni (6%) doped ZnO nanostructures







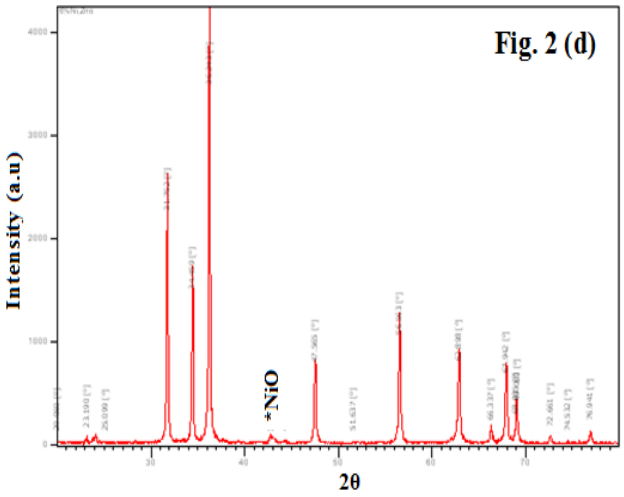


Fig. 2: (a) ZnO nanostructures (b) Ni (2%) doped ZnO nanostructures (c) Ni (4%) doped ZnO nanostructures (d) Ni (6%) doped ZnO nanostructures

Fourier transform infrared spectroscopy (FTIR) analysis

Fig.3 shows the FTIR spectra for ZnO and Ni doped ZnO nanostructures recorded in the range of 3750-400 cm^{-1}. The doping of Ni broadened the shoulder of ZnO band at 480cm^{-1} attached to the ZnO peaks which may assign as Ni-O stretching mode. A strong absorption peak due to presence of O-H stretching at 3331 cm^{-1} is observed for all samples. The existence of shift in the frequencies with the increase of dopant concentration (Ni^{2+} ions) is also observed which may due to the difference in the bond length (Zn-Ni) with the replacement of Zn^{2+} ion with large ionic radius Ni^{2+} ion .It also confirm the incorporation of Ni^{2+} into the ZnO lattice.

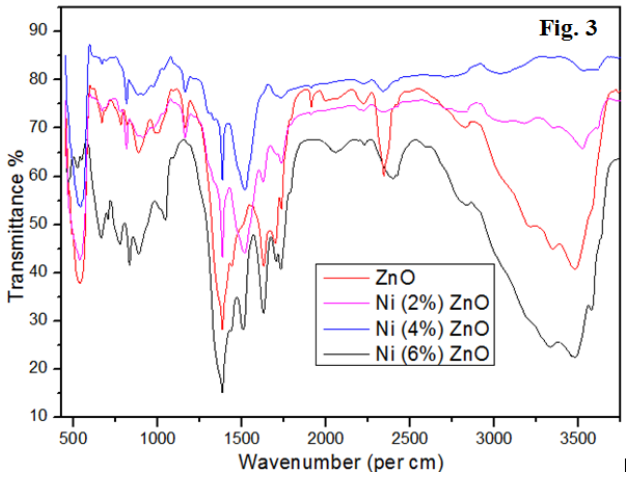
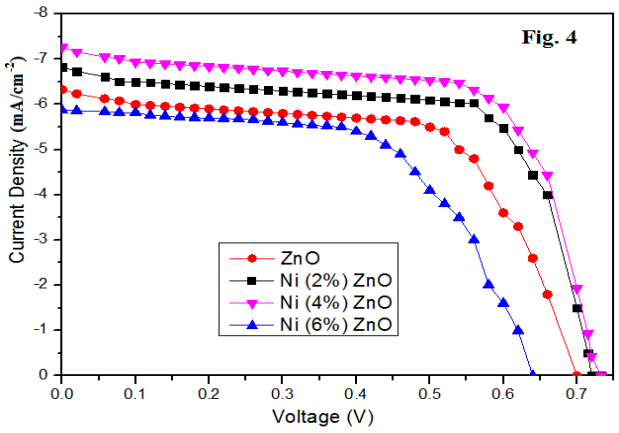


Figure 3: FTIR spectra of Undoped and Ni-doped ZnO

Photoelectrochemical properties: J-V characteristics

Fig. 4 shows the current density-voltage (J-V) characteristics of DSSCs fabricated with ZnO and Ni doped

ZnO nanostructure electrodes measured under direct sun light. For the Sun, there is an approximate conversion of 0.0079 W/m^2 per lux, so the P_{in} in our case for 80.5K lux is 0.0636 W/cm^2 [17]. The dye sensitized solar cell fabricated with Ni (2% & 4%) doped ZnO nanostructured photoanode achieves a solar to electricity conversion efficiency of 5.01 % and 5.65 % with fill factor of 0.643 , 0.69 respectively, which is larger than that measured with photoanode of pure ZnO nanostructures (3.76%).



A decrease in efficiency (3.53%) is observed when the dopant level concentration is further increase to 6 % which may be due to less dye absorption (increased particle size) and Burstein Moss effect [22 and references therein].

Table 1: Solar cell parameters of DSSC fabricated with ZnO and Ni doped ZnO nanostructures based photoanode

Dye extracted from	Photoanode	Electrolyte	Counter electrode	J_{sc} (mA/cm^{-2})	V (mV)	FF	PCE (%)
Syzygium Cumini (Jamun)	ZnO	Iodine/Tri Iodide	Carbon coated ITO	632	697	0.544	3.76
Syzygium Cumini (Jamun)	Ni (2%) ZnO	Iodine/Tri Iodide	Carbon coated ITO	688	721	0.643	5.01
Syzygium Cumini (Jamun)	Ni (4%) ZnO	Iodine/Tri Iodide	Carbon coated ITO	718	726	0.69	5.65
Syzygium Cumini (Jamun)	Ni (6%) ZnO	Iodine/Tri Iodide	Carbon coated ITO	588	642	0.598	3.53

IV. Conclusion and Future Scope

ZnO and Ni doped ZnO nanostructures are successfully synthesized by using solution combustion technique. A decrease in particle size of Ni doped ZnO nanostructure is observed with the increase in Ni dopant concentration up to 4%. However, increase in particle size is observed with further increase in Ni ion concentration i.e 6%. In X-ray diffraction pattern, a secondary NiO phase appears along with ZnO peaks for 4 % and 6% Ni doped ZnO nanostructures whereas no peak corresponding to NiO appeared in 2% Ni doped ZnO nanostructures. The efficiency of the DSSCs using these nanostructures as photoanode material observed are 3.76 %, 5.01 %, 5.65%, and 3.53 % for pure ZnO , 2 % Ni doped ZnO, 4% Ni doped ZnO and 6% Ni doped ZnO nanostructures respectively. The more dye absorption and increased light trapping due to small particle size in case of DSSCs using 2% and 4% Ni doped ZnO nanostructures as photoanode material may be one of the important factor for increase in efficiency. Further the decreased efficiency of 6% doped ZnO nanostructured DSSCs may be due to increased particle size and Burstein-Moss effect. The effect of further increase in dopant concentration (>6%) may be investigated for PCE of DSSCs to explore the effect of NiO phase in ZnO.

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