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Impact of Yttrium Dopant on Physical Properties of Manganese Selenide Nanoparticles synthesized via Spray Pyrolysis for Photovoltaic Applications

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Abstract

Undoped and yttrium-doped manganese selenide thin films were synthesized via spray pyrolysis deposition technique for photovoltaic purposes by utilizing manganese (II) acetate tetrahydrate, selenium (iv) oxide and yttrium. The deposited samples were characterized with instruments such as X-ray diffractometry, Uv-Vis spectrophotometry, four-point probe and SEM/EDX for structural, optical, electrical and morphological analysis. The X-ray diffraction pattern for both undoped and Y-doped MgSe samples shows the presence of crystal peaks along (111), (200), (210), (211) and (300) planes indicating polycrystalline and hexagonal structural nature. Optical analysis reveals a decrease in absorbance and an increase in transmittance as the wavelength increases for all the samples. The addition of a higher percentage (0.04 mol%) of yttrium dopant narrowed the bandgap energy (1.15 eV) of the undoped MnSe, making these materials promising for solar cell fabrication. Electrical analysis reveals that as yttrium dopant concentration in MnSe increases from 0 to 0.04 mol%, the film thickness increases from 110.0 nm to 115.13 nm with increasing resistivity and decreasing conductivity of 11.68×10^{-4} to 11.68×10^{-4} Ω.m and 8.561×10^{2} to 8.467×10^{2} $(\Omega.m)^{-1}$ respectively. The overall electrical result of both undoped and y-doped MnSe conforms to that of a typical semiconductor. For the morphology result, the addition of yttrium dopant altered the microstructure of undoped MnSe. For undoped MnSe a smooth dense layer was observed while Y-doped MnSe reveals an agglomeration with no defined shape but is also dense. EDX result confirms the growth of a novel yttrium manganese selenide (YMnSe) thin material. YMnSe films offer features like narrowed band gap energy, improved charge transport characteristics and enhanced light trapping, making them potential materials for photovoltaic applications.

Keywords: Manganese selenide; Yttrium; Spray pyrolysis; Optical features; Photovoltaic application.

I. INTRODUCTION

Over the last two decades, transition metal chalcogenides have been extensively studied due to their potential applications as semiconductors [1]. Thin films of transition metal chalcogenides are considered important technological materials because of their electrical, optical, magnetic and transport properties which have found applications in spintronics devices [2].

The transition-metal selenides possess outstanding photovoltaic and electrochemical properties when compared to transition-metal oxides because the electronegativity of oxygen is higher than that of selenium. Thus, transition-metal selenides such as CuSe, MgSe, CdSe, MnSe, and CoSe [3],[4] exhibit good optical and electrochemical properties for use as an absorber layer in solar energy collection and electrode-active materials, respectively [5].

Manganese selenide (MnSe) and its thin film are good semiconductors. MnSe thin films have attracted interest due to their semi-conductivity and magnetism properties [6]. Thin films of MnSe are usually crystallized in cubic structure (JCPDS 11-0683) with lattice constants (a = 5 .462 Å) and in hexagonal structure (JCPDS 89-4966) with lattice constants (a = 3.63 Å; c = 5.91 Å) [4]. Manganese selenide (MnSe) is a direct band gap semiconductor with a band gap value ranging from 1.13 to 1.25 eV used for fabricating diluted magnetic semiconductors [7].

The importance of manganese selenide has been put forth in supercapacitors, batteries, chemical sensing and thermoelectrics [8]. Manganese selenide is a p-type semiconductor (E_g = 2.0 eV) with rock salt, wurtzite (WZ) and zinc blende structures [9]. In manganese selenide thin films ordinary and extraordinary Hall coefficients have the same magnitudes. Electron and hole combination give positive and negative values to R_s (normal Hall effect) and R_o (extraordinary Hall effect). Theoretically, in ferromagnetic materials, an additional Hall effect known as extraordinary, spontaneous or anomalous Hall effect exists other than the Hall effect due to Lorentz force on charge carriers. In manganese selenide films, Rs and Ro depend upon Se contents in the film and substrate temperature [8].

Manganese selenide (MnSe) has been fabricated over the years with thermal evaporation at reduced pressure, spray pyrolysis, hydrothermal method, chemical vapor deposition, pulsed laser deposition, electrodeposition and colloidal methods [8]. Other methods such as molecular beam epitaxy (MBE) [10], organometallic vapour phase epitaxy [11], and brush plating [12] are used for the preparation of MnSe thin films.

Reference [10] prepared MnSe thin films using molecular beam epitaxy and studied their optical properties. The preparation of MnSe thin films using the organometallic vapour phase epitaxy technique and their structural and thermal properties have been investigated [11]. Reference [7] prepared MnSe thin films and studied their properties using X-ray diffraction and optical absorption measurements, respectively. Reference [12] obtained MnSe thin films by a brush plating method and studied their properties using X-ray diffraction, scanning electron microscopy, energy dispersive analysis by X-rays, optical absorption and Raman spectroscopic measurements, respectively.

The present study uses a spray-pyrolysis technique (SPT) which has since been used in the glass industry and in solar-cell fabrication to deposit electrically conducting electrodes [15], [16]. Through this technique, dense and porous oxide films, ceramic coatings and powders can be prepared. It is a relatively cost-effective and very simple method. Materials obtained by SPT find a wide range of applications in optoelectronic devices, anti-reflective coatings, sensors, etc. [14], [15], [16]. The spray-pyrolysis technique has several notable advantages, including the ability to change the properties of the film by changing the composition of the starting material (addition of dopants and modification of the film microstructure) and lowering production costs when large-scale production is required [17].

More recently, several efforts have been made to enhance the efficiency of semiconductors via doping, supporting and coupling these compounds. Concerning this, rare earth-doped nanoparticles have received a lot of consideration given their massive photo-catalytic activity in the degradation of organic contaminants due to the suppression of electron-hole recombination, large content of oxygen vacancies, and strong absorption of hydroxide ions on the surface of the catalyst [18]. Because of their low diffusivity, they are also known to perform useful functions such as lowering and stabilizing the dissipation factor in dielectric materials [19].

Yttrium is one of the promising dopants in the family of rare earth metals since it improves fatigue endurance, leakage current and remanent polarization [20]. It can also act as an acceptor or donor ion [3]. Another major effect of yttrium doping is the change in the electrical conductivity of doped material concerning the doping site [21].

Despite the potential advantages of doping in thin film engineering, a comprehensive understanding of the intricate interplay of yttrium doping on the properties of MnSe thin films remains elusive. This study aims to investigate the effect of yttrium doping on the structural, morphological, optical and electrical properties of yttrium-doped MnSe thin film.

II. EXPERIMENTATION

Analytical grade chemicals were utilized to grow undoped and yttrium-doped Manganese Selenide thin material. These chemicals include Manganese (II) Acetate Tetrahydrate ($Mn(CH_4COO)_2.4H_2O$), Selenium (IV) Oxide (SeO_2), hydrogen chloride (HCl) and yttrium (Y). Soda-lime glass sides were used as substrate. Before deposition, these substrates were thoroughly cleaned and activated using methanol, acetone, and deionized water. For the thin material growth, a spray pyrolysis method was adopted. It is fundamentally a chemical

growth method in which thin droplets of the supposed material are sprayed onto a pre-heated substrate [22].

A. Preparation of Solutions

A 0.01 mol solution of SeO_2 was prepared by dissolving 3.158 g with 5 ml of HCl and an additional 100 ml of deionized water. 0.01 mol solution of Mn $(CH_4COO)_2.4H_2O$ was prepared by dissolving 1.225 g of it in 500 ml of deionized water. Next, different concentrations of yttrium (used as a doping agent) pinned at 0.01, 0.02, 0.03 and 0.04 mol% were prepared from four 5ml of yttrium solutions.

B. Spray Pyrolysis Deposition Technique

The spray process involves the optimization of several process parameters. The values of the optimized process parameters are a Precursor flow rate of 0.07 ml/min, Spray-nozzle specification: outside diameter of 0.32 mm and internal diameter of 0.16 mm, Spray nozzle to substrate distance of 8 mm, Substrate temperature of 400ºC, and 3.5 kV Atomizing voltage. Electrostatic atomization is used in this technique. The precursor ion i.e. MnSe is pumped through the pump directly to the nozzle, As the mixture gets to the substrate through the nozzle and hits the surface of the substrate, evaporation of the residual solvent takes place, spreading of the droplet and then the salt decomposition, finally MnSe, is deposited on the substrate. Film uniformity is ensured by moving the nozzle (needle) and the substrate. This process is repeated with a doping agent yttrium (Y) at different dopant concentrations. Table I outlines the variation of dopant concentration at constant voltage (3.5 kV) and temperature (400^0C).

Table I. Variations of dopant concentration.

Samples	Sample Code	(Mn $(CH_4COO)_2.4H_2O$) (ml)	SeO_2 (ml)	Yttrium (ml)	Voltages (kV)	Dopant conc. (Yttrium) (mol%)
MgSe	MN	20	10		3.5	
MgSe:$Y_{0.01\%}$	MNY (0.01 mol%)	20	10	5	3.5	0.01
MgSe:$Y_{0.02\%}$	MNY (0.02 mol%)	20	10	5	3.5	0.02
MgSe:$Y_{0.03\%}$	MNY (0.03 mol%)	20	10	5	3.5	0.03
MgSe:$Y_{0.04\%}$	MNY (0.04 mol%)	20	10	5	3.5	0.04

C. Characterization Techniques

The synthesized films were characterized using the following instruments: cu-Kal (λ=.15418 Å) diffractometer, UV 1800 visible spectrophotometer, old Jandel four-point probes technique (model T345), scanning electron microscope (SEM) and energy-dispersive X-ray analysis (EDX) for structural, optical, electrical, morphology and elemental composition respectively. More so, other properties required for a proper understanding of these deposited thin materials were evaluated using appropriate mathematical equations.

III. RESULT AND DISCUSSION

A. X-ray diffraction analysis

The X-ray diffraction pattern obtained for undoped and Y-doped MnSe thin materials with different mol% concentrations (0.01 – 0.04 mol%) is shown in Fig. 1. The X-ray diffraction pattern result and the presence of multiple lattice planes (111), (200), (210), (211) and (300) confirmed that the undoped and Y-doped MnSe samples are polycrystalline and possess a hexagonal structure. The different peaks in the diffractogram were indexed and the corresponding values of interplanar spacing (d) were calculated using mathematical relations [23]. Doping MnSe films with yttrium declined the crystallinity as seen in Fig. 1. Aside from the undoped MnSe, MnSe doped with 0.03 mol% of Y gave a better crystallinity amidst other doped MnSe samples. Other structural parameters such as crystallite size (D), interplanar spacing (d) and dislocation density (δ) were obtained using (1), (2) and (3) [24] – [26]. The result of these parameters is outlined in Table II.

$$D = {k\lambda}/{\beta\cos\theta} \quad (1)$$

$$d = {\lambda}/{2\sin\theta} \quad (2)$$

$$\delta = {1}/{D^2} \quad (3)$$

Where k = 0.94, λ = wavelength of the X-ray source given as 0.154 nm, θ = the Bragg's angle/diffraction angle.

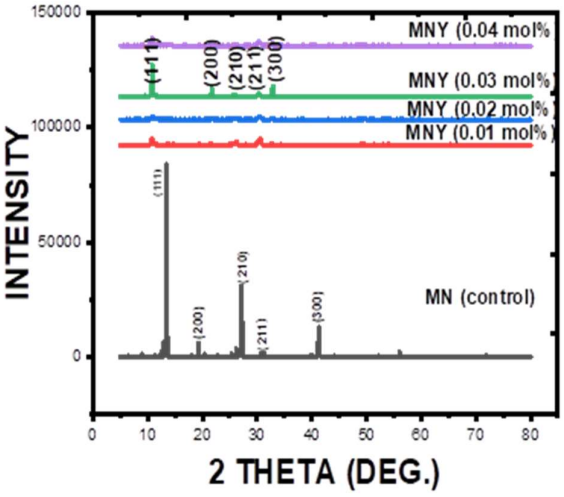


Fig. 1. X-ray diffraction (XRD) of MnSe and yttrium-doped MnSe at different molar percentage dopants (0.01 – 0.04 mol%)

Table II. Structural values for the MnSe and Yttrium-doped MnSe at different molar percent dopant (0.01 – 0.04 mol%).

Sample	2θ (degree)	Spacing, D (Å)	Lattice constant (Å)	FWHM, β	hkl	Crystallite Size, D (nm)	Dislocation density, δ (m^2)
MN-MnSe	13.3032	6.6493	11.5169	0.1851	111	0.7538	5.3351
	19.2390	4.6091	9.2182	0.2095	200	0.6710	6.7453
	27.2677	3.2674	6.5349	0.1481	210	0.9640	3.2685
	31.3507	2.8506	6.3742	0.2258	211	0.6376	7.4002
	41.4038	2.1787	5.3368	0.2249	300	0.6588	6.9599
MNY-Y/MnSe	10.9015	8.1082	14.0438	0.1851	111	0.7521	5.3589
(0.01mol%)	21.7044	4.0908	8.1816	0.2095	200	0.6736	6.6931
(0.02 mol%)	25.7580	3.4554	6.9109	0.1480	210	0.9610	3.2888
(0.03 mol%)	30.4929	2.9288	6.5490	0.2258	211	0.6363	7.4309
(0.04 mol%)	32.7917	2.7285	6.6836	0.2249	300	0.6423	7.3201

B. Optical analysis

The plot of Absorbance with respect to wavelength for undoped and yttrium-doped MnSe at different dopant concentrations is presented in Fig. 2 (a). The absorbance value for undoped MnSe was observed to have a constant value almost all through the spectrum while yttrium doped samples decrease as the wavelength tends towards the Vis-IR region. MnSe doped with 0.04 mol% of yttrium recorded the highest absorbance value all through the spectrum with a maximum of 2.25 au at the UV region, followed by undoped MnSe, MnSe doped with 0.01 mol%, 0.03 mol% and 0.02 mol%. The addition of a high concentration of yttrium was seen to increase the absorbance value of pure MnSe. Reference [27] reported a similar result for electrodeposited iron-doped ZnSe and a decreasing trend with respect to high wavelength was also reported by [6] for MnSe deposited with the chemical bath technique.

The plot of transmittance with respect to wavelength for undoped and yttrium-doped MnSe at different dopant concentrations is presented in Fig. 2(b). The result shows that the undoped MnSe recorded a very low transmittance which was greatly improved when doped with yttrium especially in the VIS-IR region of the spectrum thereby making yttrium-doped MnSe an interesting material for the production of photovoltaic devices.

The plot of reflectance with respect to wavelength for undoped and yttrium-doped MnSe at different dopant concentrations is presented in Fig. 2(c). From the reflectance plot shown in Fig. 2(c) all the deposited films exhibited very low reflectance value. More so, the reflectance increases with increased wavelength in the visible and near infra-red regions with a maximum value of > 0.3 for all the deposited films.

The variation of absorption coefficient with photon energy for undoped and yttrium-doped MnSe at different dopant concentrations is presented in Fig. 3(a). The dependence of the absorption coefficient on photon energy gives a better understanding with respect to the nature of optical shift within the conduction and valence band of a thin material [26]. From Fig. 3(a), the absorption coefficient value of all the samples increases with an increase in photon energy and the addition of yttrium to undoped MnSe was observed to improve the absorption coefficient of the material both in the low and high photon energy. The large absorption coefficient values ($< 10^6$) recorded by the undoped and Y-MnSe qualify the materials to be used for solar energy conversion (Photovoltaic) [28].

The energy band gap for undoped and MnSe doped with 0.01, 0.02, 0.03 and 0.04 mol% of yttrium were determined to be 1.25 eV, 1.35 eV, 1.57 eV, 1.10 eV and 1.15 eV respectively. These values were obtained from the plots of $(\alpha h\nu)^2$ against hv for each of the samples as seen in Fig. 3(b), and extrapolating the linear part of the plots to $(\alpha h\nu)^2 = 0$ gives us the band gaps. The energy band gap value of MnSe obtained in this study is seen to be in close agreement with the value reported by [4] for MnSe thin films synthesized by the electrodeposition method. Further insight into the energy band gap values of yttrium-doped MnSe reveals that the addition of a high concentration of yttrium dopant to MnSe narrowed its band gap thereby making YMnSe a more promising material as a solar collector [26]. A review of literature indicates no studies on the cultivation of MnSe doped with yttrium (YMnSe).

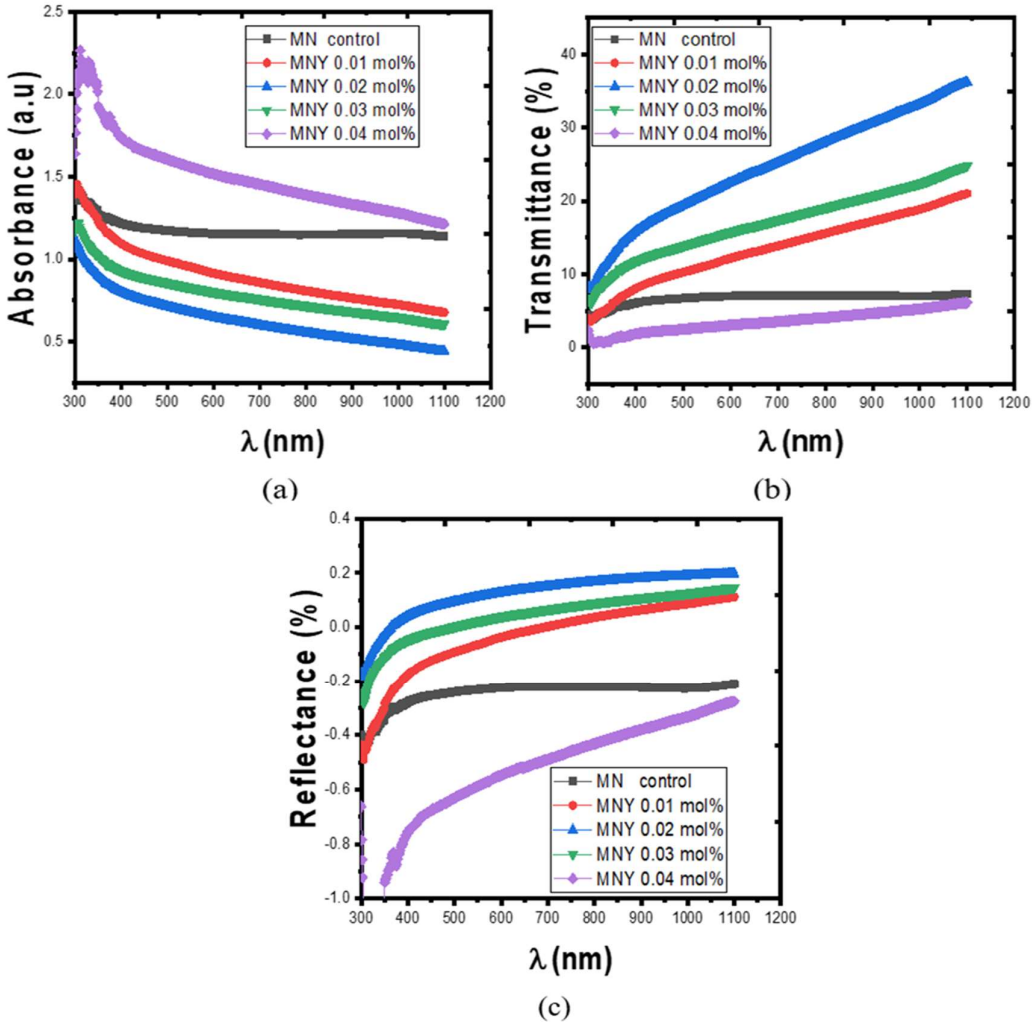


Fig. 2. Plot of (a) absorbance (b) transmittance and (c) reflectance against wavelength for undoped and yttrium-doped MnSe at different dopant concentrations.

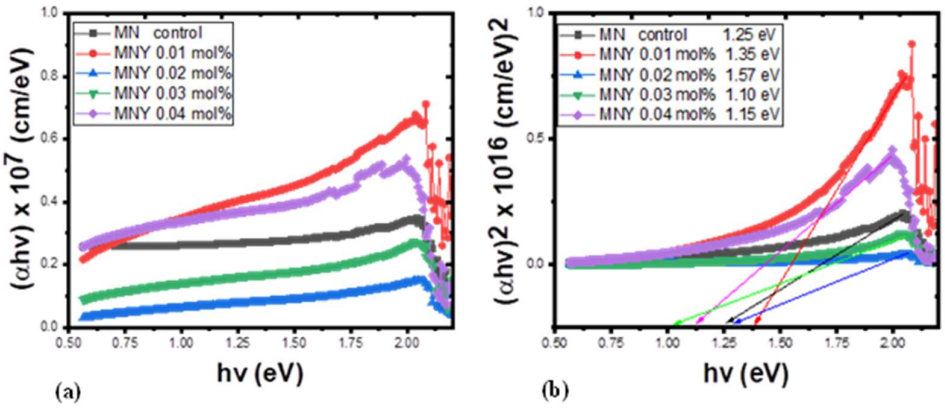


Fig. 3. (a)Variation of absorption coefficient (b) Plot of $(\alpha hv)^2$ against photon energy for undoped and yttrium doped MnSe at different dopant concentrations.

C. Electrical analysis

The thickness, resistivity and conductivity values of MnSe and Y-doped MnSe (0.01 – 0.04 mol%) are summarized in Table III.

The result reveals that the addition of yttrium strongly influenced the electrical property and thickness of undoped MnSe. As the Y concentration in MnSe increases from 0 to 0.04 mol%, the film thickness increases from 110.0 nm to 115.13 nm with increasing resistivity and decreasing conductivity of 11.68 x 10^{-4} to 11.68 x 10^{-4} Ω.m and 8.561 x 10^{2} to 8.467 x 10^{2} $(\Omega.m)^{-1}$ respectively. The overall electrical result of both undoped and y-doped MnSe conforms to that of a typical semiconductor. A similar result was obtained by [21] for yttrium-doped CuSe.

D. Morphological and elemental composition analysis

The surface morphology of these thin materials as shown in Fig. 4 was investigated using scanning electron microscopy (SEM). The images show uniform, agglomerated, well-dense, homogenous films devoid of cracks suggesting uniform deposition. From the SEM result, it was observed that the incorporation of yttrium dopant altered the morphology of the undoped MnSe.

From the EDX spectrum result in Fig. 5 the core elements manganese, selenium and yttrium were seen as basic constituents of the as-deposited thin material. This result confirms the growth of a novel yttrium manganese selenide (YMnSe) thin material.

Table III. Electrical properties of MnSe and Y-doped MnSe (at different dopant concentrations).

Sample Code	Thickness, t (nm)	Resistivity, ρ (Ω.m)	Conductivity, σ $(\Omega.m)^{-1}$
MN control	110.00	11.680 x 10^{-4}	8.561 x 10^{2}
MNY 0.01 mol%	112.32	11.721 x 10^{-4}	8.531 x 10^{2}
MNY 0.02 mol%	112.41	11.743 x 10^{-4}	8.515 x 10^{2}
MNY 0.03 mol%	113.21	11.767 x 10^{-4}	8.498 x 10^{2}
MNY 0.04 mol%	115.13	11.810 x 10^{-4}	8.467 x 10^{2}

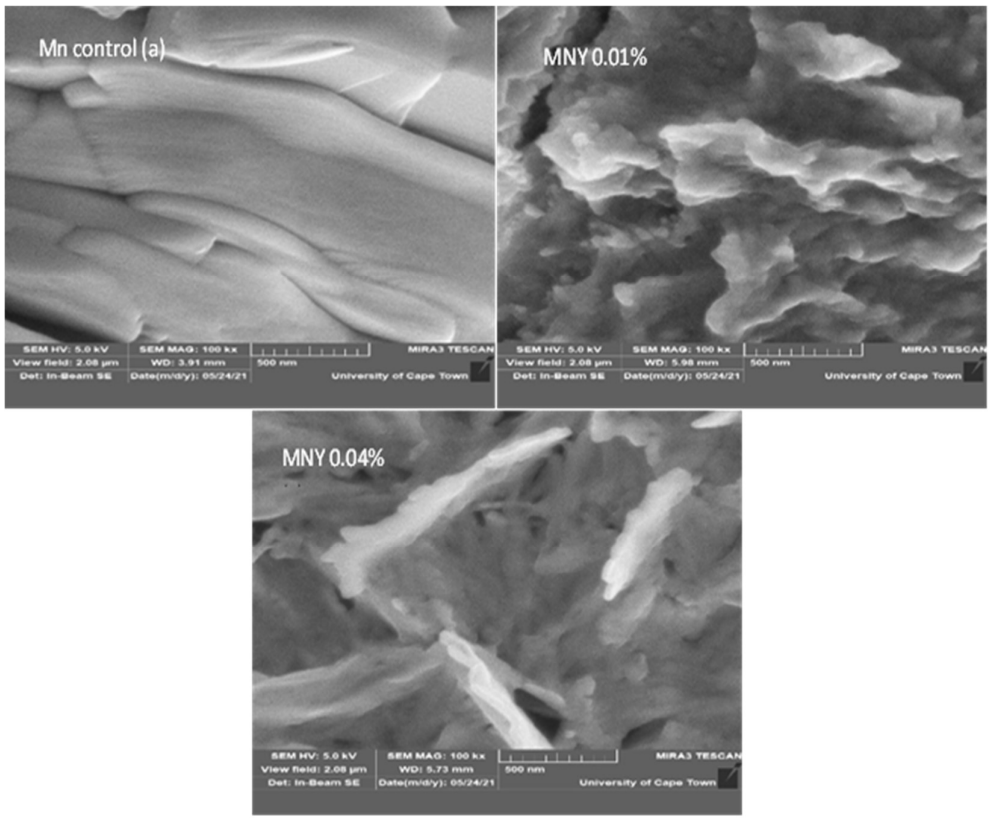
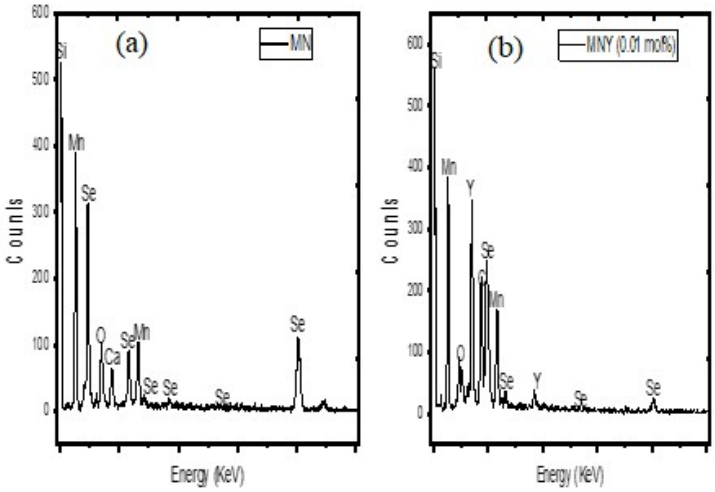


Fig. 4. Surface Morphology of (a) undoped (b) 0.01 mol% (c) 0.04 mol% yttrium-doped MnSe thin materials.



Elements	Atomic weight (%)
Mn	60.79
Se	433.12
Si	5.44
O	4.63
Ca	1.90

Elements	Atomic weight (%)
Mn	60.48
Se	38.12
Y	9.03
Si	5.65
O	4.60

Fig. 5. EDX spectrum of (a) undoped and (b) Y-doped MnSe thin material.

IV. Conclusion

The present study elucidates the deposition of undoped, and yttrium-doped manganese selenide thin films via spray pyrolysis deposition technique for photovoltaic purposes by utilizing manganese (II) acetate tetrahydrate, selenium (iv) oxide and yttrium. To study the doping effect of yttrium in MnSe, the doping percent was varied such as 0.01 mol%, 0.02 mol%, 0.03 mol% and 0.04mol%. The deposited samples were characterized with instruments such as X-ray diffractometry, UV-Vis spectrophotometry, four-point probe and SEM/EDX for structural, optical, electrical properties, microstructure and elemental composition respectively. The X-ray diffraction pattern result and the presence of multiple lattice planes (111), (200), (210), (211) and (300) confirmed that the undoped and Y-doped MnSe samples are polycrystalline and possess a hexagonal structure. Optical features evaluation via UV-Vis spectrophotometry reveals a decrease in absorbance and an increase in transmittance as the wavelength increases for all the samples. The addition of a higher percentage of yttrium dopant narrowed the bandgap energy of the undoped MnSe, making these materials promising for solar cell fabrication. Electrical property studies reveal that as yttrium dopant concentration in MnSe increases from 0 to 0.04 mol%, the film thickness increases from 110.0 nm to 115.13 nm with increasing resistivity and decreasing conductivity of 11.68 x 10^{-4} to 11.68 x 10^{-4} Ω.m and 8.561 x 10^{2} to 8.467 x 10^{2} $(\Omega.m)^{-1}$ respectively. The overall electrical result of both undoped and y-doped MnSe conforms to that of a typical semiconductor. Morphology results reveal that the addition of yttrium dopant altered the microstructure of undoped MnSe and the EDX result confirms the growth of a novel yttrium manganese selenide (YMnSe) thin material. YMnSe films offer features like narrowed band gap energy, improved charge transport characteristics and enhanced light trapping, making them potential materials for photovoltaic applications.

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