Reduced electrical performance of Zn enriched ZnTe nanoinclusion semiconductors thin films for buffer layer in solar cells

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Reduced Electrical Performance of Zn Enriched ZnTe Nanoinclusion Semiconductors thin films for Buffer Layer in Solar Cells

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Abstract

Closed space sublimation (CSS) technique was employed to deposit thin films of Zinc Telluride (ZnTe) on glass substrate under high vacuum. Two sets of ZnTe thin films and Zn enriched ZnTe thin films were prepared for comparative study. The enrichment for Zn onto the as-deposited ZnTe thin films by the novel manner of layer by layer deposition with subsequent annealing. X-ray diffraction (XRD) studies revealed before and after enrichment of Zn the preferred orientation is [111] having cubic phase. The lattice constant was found to be increased and crystallite size decreased 28 nm to 24 nm after the enrichment of Zn. Morphological study was carried out through scanning electron microscope (SEM). For Zn enriched samples the average grain size is smaller as compared to ZnTe thin films. The local compositions of Zn and Te were confirmed by energy dispersive X-rays (EDX) from 51 atomic % of as deposited ZnTe thin films to 68 atomic % in Zn enriched ZnTe thin films. The Zn enriched samples have slight decrease in optical transmission in UV-VIS-NIR range as compare to the as-deposited ZnTe thin films. Due to the deposition of Zn there is a very small change in optical band gap energy. Four-probe technique was used to study electrical properties of as-deposited and Zn-enriched ZnTe thin films. These results shows that the as-deposited samples has resistivity of 10^6 Ω-cm. For Zn enrichment samples resistivity increases from 10^6 Ω-cm to 10^8 Ω-cm. Which shows that Zn-enriched samples are not suitable for back contact of II-VI solar cells. X-rays Photoelectron Spectroscopy (XPS) was used to confirm the elemental compositions and its bonding strength

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before and after enrichment of Zn.

Keywords: Layer by Layer deposition, ZnTe thin films, Resistivity, Zn enrichment, XPS

1. Introduction

Zinc telluride (ZnTe) has been found a potential candidate for several applications in optoelectronic [1] due to its tunable energy band gap property. ZnTe thin films has been used in CdTe/CdS hetrojunction solar cells as a back contact due to its band gap offset 0.1 eV [2-6]. Historically ZnTe have been deposited through DC and RF sputtering [3, 4], brush plated [7], closed space sublimation [8, 9], electro-deposition [10], two source evaporation [11] etc. The advantage of CSS technique among other competitive deposition techniques is its efficient use of material due to small substrate-source separation. The physical properties of thin film, thus deposited, can be in-situ controlled. In II-VI solar cell, doping plays a vital role. Copper (Cu) and silver (Ag) used as a dopant to enhance its conductivity also improves the electrical properties on ZnTe [1, 12]. There is need of a low resistive back contact in solar cells for cadmium tellurice (CdTe). For thin film solar cells in order to enhance the performance of hetrostructure, ZnTe is an ideal intermediate layer between CdTe active layer and metallic back contact. The work function of CdTe is 5.78 eV. It is the sum of band gap (1.5 eV) and electron affinity (4.28 eV) of CdTe. It means for this purpose one needs a heavily doped p-type material. [13]. ZnTe is a suitable material which make a buffer layer with CdTe for metallic back contact. ZnTe is a p-type material and due to its self-compensating properties its type of conductivity could not be changed easily [1, 4, and 11]. The main problem with ZnTe is its high resistivity, which can be reduced with metal doping to some extent as discussed earlier.

In this research work we tried to investigate the enrichment effects instead of doping, for this purpose Zn enriched ZnTe thin films have been prepared for comparative study. The close spaced sublimation (CSS) technique was used to sublimate as deposited ZnTe thin films and a novel method of layer by layer deposition have been employed for Zn enriched ZnTe thin films that has not been reported to best of our knowledge. The structure variation with morphology, electron transport, composition and optical analysis was carried out and results was compared for ZnTe, Zn enrichment samples. SEM, XRD, EDX, Spectrophotometer, Hall measurement system and XPS cylindrical analyzer was use to study the effect of enrichment of Zn.

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2. *Sample Synthesis*

High purity zinc telluride powder (99.99%) was used as a source material on to the substrate glass slide. The substrate was cleaned for 20 min at 60 °C in IPA using sonication. The amount of source material was 20 mg for sublimation in a graphite boat. The optimized distance between source-substrate was 5 mm in order to get good quality of film. The chamber pressure was lowered to x 10^{-5} mbar by the combination of rotary and diffusion pumps. Heating the material directly, the source and substrate lamps of power rating of 1000 W and 500W were used. After achieving the required vacuum, temperatures were monitor by using two K-type thermocouples along with temperature controllers. The temperatures of substrate and source were gradually increased up to 470 ± 5 °C and 350 ± 5 °C respectively. The lamps were switched off after deposition of ZnTe thin films for 5 min deposition time but pumping continued till room temperature (RT) achieved. Once the RT achieved, the vacuum pumps along with gauges were switched off and vacuum was broken to get ZnTe thin films out of it.

In next phase, same process was repeated but source material was Zn instead of ZnTe. Zn was evaporated on to the as-deposited ZnTe thin films for 5 min deposition time under high vacuum, named as a 2nd layer. Zn doesn't have a good sticking with glass substrate therefore it is not possible to deposit the Zn as 1st layer and it was sandwiched between two layers of ZnTe thin films [1]. Finally, same process was done to sublimate a 3rd layer of ZnTe on to the 2nd layer for 1 min, 3 min, 5 min and 7 min deposition times under same conditions for deposition as followed in earlier layers shown in Fig. 1. For 3rd layer of ZnTe the substrate temperature was same as the source temperature of 2nd layer (i.e. 425 °C). So that the re-evaporation of Zn can be avoided. The post annealing was carried out for improvement of crystallinity of prepared ZnTe thin films samples for 1 hour at 350 ± 5 °C after deposition.

Finally the structural, compositional, optical, and electrical characterizations of thin films and the effect of annealing was studied as described below.

3. *Results and Discussion*

3.1 Structural Study

XRD results of Zn-enriched ZnTe thin films samples are shown in Fig. 2. XRD results of the as-deposited ZnTe thin films showed the films are polycrystalline. The preferred orientation is to be (111) with a cubic phase. The peak at 2Θ = 27.2770 has a full-width half-maximum (FWHM) of

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0.3542, which matches with ICDD reference card no. 00-089-3054 corresponds to a lattice parameter of 6.0980 Å.

Scherrer's formula was used to calculate the the crystallite size (D) from XRD obtained data sheet [2]. The as-prepared ZnTe thin films have average crystallite size of 28 nm. The 3rd layer of ZnTe that was deposited for 1 min, the strongest diffraction peak appears at 2Θ = 25.2828 having a FWHM of 0.2755. Which matches with reference card No.00-015-0746, the lattice constant for this film is 6.1026 Å and the average crystallite size of 38 nm. Also the 3rd layer of ZnTe deposited for 3 min, 5 min and 7 min we get the values of 2Θ of 25.3525, 25.1594, 25.3705 and the values of FWHM of 0.2362, 0.5510 and 0.3936 for the strongest peak. Deposition for 3 min, film the ICDD reference card No. that matches is 01-071-8947, the same card matches for 7 min deposition with a difference of 0.01 in the value of 2Θ, with lattice constant of 6.0790 Å. The 5 min deposition matches with ICDD reference card No. 01-071-5965 with a difference of 0.01 in the value of 2Θ. The obtained values of crystallite size these samples are 46 nm, 17 nm and 24 nm, respectively. The enrichment samples have cubic phase for all samples. The XRD data sheet shows that after enrichment the number of different planes is reduced. The already deposited ZnTe thin film has seven observed peaks. The 1 min deposition of a 3rd layer of ZnTe has six peaks observed in XRD data sheet. The 3 min and 5 min deposition has four peaks and the 7 min deposition of 3rd layer of ZnTe has three peaks.

It is found that 2Θ increases with increasing of crystallite size for the already deposited films. The FWHM has reduced which shows the improvement in crystallinity. A number of peaks decreases as enrichment times increases which support the above argument. The reduction of lattice constant, suggests that the ZnTe thin films, might be subject to a tensile stress after the enrichment process, due to the planes of Zn and ZnTe thin films. A color change from brown to dull brown was also observed after the Zn-enrichment process, which might be due to the dissipation of Te or increasing Zn concentration. These results are in agreement with those of E. Bacaksiz *et al*. [3]. With increase in Zn concentration the preferred orientations shift towards higher angular positions. It might be due to the bond length of ZnTe lattice by Te or Zn interstitials. After enrichment of Zn the lattice constant also decreased, which is good agreement with previous reports [4].

3.2 Surface study

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Fig. 3 shows the effect on morphology of ZnTe thin films due to Zn enrichment. As-deposited ZnTe thin film have smooth surface, the grains are fine having different sizes as shown in Fig. 3 (a). Due to enrichment of Zn on ZnTe thin film there are many changes in the microstructure. One change is the larger grains. For the samples with Zn enrichment for 1 min, 3 min and 5 min the SEM images are shown in Fig. 3 (b), (c) and (d) respectively. The images show that after Zn enrichment there is re-crystallization indicating merging of grains. The average grain size was significantly found to be reduced in Zn-enriched ZnTe thin films. In Zn-enriched ZnTe thin films the grains are non-homogeneous grains and there are some discontinuities. Separation of grains might be the reason of this effect. In addition, the grains have rod like shape and had also observed the similar results in CdTe [14]. Due to this the coalescence of small grains occurs which results in grain growth and the grains also reorient. This coalescence of grains has very important role in compound semiconductors [14]. Due to temperature increase in annealing and annealing time the Zn and Te impurity atoms become more mobile therefore the coalescence of small grains occur to form larger grains.

The elemental composition of the films obtained from EDX analysis as shown in Table 1. The atomic % of Zn also increased with increase in enrichment. Also with increase in annealing temperature and annealing time Te content in ZnTe thin films also decreased.

3.3 Optical study

Spectrophotometer (LAMDA 950) in transmittance mode was used at room temperature to measure the optical properties of the films and the Swanpoul model [15] was used to measure the energy of the band gap. Refractive index (n) was calculated using the relation

$$n = \frac{[N+(N^2-4s^2)^{1/2}]}{2} \qquad (1)$$

'N' is the number of oscillation whereas s is the refractive index of the glass

$$N = 1 + s^2 + 4s\left(\frac{T_M - T_m}{T_M T_m}\right) \qquad (2)$$

T_M and T_m is the transmittance maximum and minimum respectively. Thickness was calculated by the expression

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$$\mathbf{d} = (\frac{\mathbf{1}}{\mathbf{4n}})\,[\frac{\boldsymbol{\lambda}\mathbf{M}\,\boldsymbol{\lambda}\mathbf{m}}{\boldsymbol{\lambda}\mathbf{M} - \boldsymbol{\lambda}\mathbf{m}}] \quad (3)$$

In Eq. 3. λ_M was the wavelength maximum cross ponding to maximum transmittance and λ_m is minimum.

Taue relation was used for energy band gap calculations as shown in Eq. 4.

$$(\boldsymbol{\alpha}.\mathbf{h}\boldsymbol{\upsilon}) = \mathbf{A}(\mathbf{h}\boldsymbol{\upsilon} - \mathbf{Eg})^{1/2} \quad (4)$$

In Eq. 4. A is the absorption coefficient, A is constant, Where A is constant, hν is photon energy and Eg is energy band gap of the material. The relation for absorption coefficient ‘α’ can be calculated by

$$\boldsymbol{\alpha} = (\frac{1}{d})\,\boldsymbol{ln}(\frac{1}{T}) \quad (5)$$

These parameters have usual meaning as described earlier.

The already deposited ZnTe thin film showed greater than 80 % transmission. Fig. 4 shows the transmission spectra of already deposited ZnTe thin film and Zn-enriched ZnTe films. The in-set of Fig. 4 showed the small variation of band gap calculations from Taue relation. As concentration of Zn is increases, the transmission decreases. The reason might be increase in optical scattering due to crystal structure distortion. The energy band gap decreases by a small amount due to diffusion of Zn as shown in Fig. 5. The grain size changes may also be related to this. As already discussed, due to annealing process diffusion of Te in to the ZnTe thin films occurs also observed similar effects [16]. The effect of thickness also shown in Fig. 5. Which confirmed the slight change in thickness of the films.

3.4 Electrical study

Van der Pauw technique at room temperature was used to measure the electrical properties of ZnTe thin films. The already deposited ZnTe film has resistivity of 10^6 Ω-cm. As we increase the Te concentration in the ZnTe thin films, the resistivity of the films decreases to 10^4 Ω-cm. As Zn concentration increase resistivity of samples increases upto 10^8 Ω-cm. The reason is the enhancement in micro-structural defects and discontinuities at grain boundaries as shown in Fig. 6. This increase in resistivity due to enhancement of Zn in ZnTe might be due to reduction in

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optical band gap. [15-17]. However larger defects at grain boundaries due to change in stiochiometry may also be a reason of the reduction in resistivity.

3.5 X-rays Photoelectron Spectroscopy (XPS) study

The adsorbed contamination and elemental information of Zn-enriched ZnTe thin films can be studied using X-rays Photo-electron Spectroscopy (XPS). XPS has been recognized over the past few decades as a surface characterization technique. It is useful to find the information about elemental compositions, chemical states and thickness etc. compositional study is extensively used in thin films. The survey scans and oxygen (O-1s) was shown in Fig. 7 (a-d). The pass energy of 100 eV was applied for ZnTe and Zn-enriched ZnTe thin films on glass substrates. The XPS peaks clearly showed the presence of O, C, Zn and Te in these films. The binding energies of these photoelectron were standardized with C 1s peak at 284.5 eV for charge referencing. The carbon line is on 288 eV indicates the presences of CO_2 from environment. It might be due to carbonate or as an adsorbate [18-20]. The carbide entities were not found as we don't have any peak around 284.5 eV which might be the surface charge calibrant. The other elements may exist in low level and contained in the baseline. The peaks of XPS data were de-convoluted by using CASAXPS software. A series of high resolution spectra were carried out on Zn, Te, O and C in order to get elemental compositions as shown in Fig. 7 & 8. Lorentzian-Gaussian peaks were nicely fitted to Zn, Te, O and C data points. Shirley type (non-linear) background was also added in all fitted graphs.

The contaminant elements like carbon and oxygen were on the surface and variation in concentration of oxygen was observed, so the existence of formation of auxiliary oxides [20]. Oxygen peaks were de-convoluted at 529.05 and 532.86 eV for as-deposited ZnTe thin films as shown in Fig. 7 (c). The peak at 530.0 eV was due to 1s spectrum to O^{2-} on cubic structure of Zn^{+2} [21]. The Zn enriched ZnTe thin films had the peak at 532.38 eV as shown in Fig. 7 (d). Small peak (not shown) at 531.6 eV was due to the variation in concentration of oxygen vacancies associated to the deficient region [22]. The small peak at 532.86 eV was due to attribution of dissociated oxygen or chemisorption, it may also be due to the OH-species on surface. Small peaks around the binding energies of 537.5 eV was due to the O-C contaminations [23]. The intensity of region is higher than the other two regions which cross ponds to the strong bonding between them.

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Symmetric peaks were observed for Zn core level spectra of 2p in Fig. 7 (a) for as-deposited ZnTe thin films and Fig. 8 (b) for Zn-enriched ZnTe thin films. As-deposited ZnTe thin films had peaks at 1023.1 eV and 1046 eV for Zn $2p_{3/2}$ and $2p_{1/2}$ respectively. The noise level for as-deposited ZnTe thin films was high in Zn-2p spectra due to the fact that in as-deposited ZnTe thin films, Te is in large concentration. Te-3d spectra of Zn-enriched ZnTe also have high noise level as after Zn enrichment it might be due to the fact of larger concentration of Zn in ZnTe thin films [19-21]. The binding energy (B.E.) of Zn-enriched ZnTe thin films was higher as compared with as-deposited ZnTe thin films. The peak appeared at 1024.82 eV for $2p_{3/2}$ and the peak at 1047.98 eV was $2p_{1/2}$. The spin orbit splitting (ΔE_{Zn} = 23.1 eV) resulted oxidation state of Zn is +2. Another peak around 1021.50 eV was observed in some samples due to metallic species (not shown).

Te-3d spectra for as-deposited and Zn-enriched ZnTe thin films were shown in Fig. 8 (c) & (d) respectively. The peak at the B.E. of 571.15 eV cross ponds to Te^{2-} whereas the peak at 574.47 eV related to TeO_2 in Fig. 8 (c) of as-deposited ZnTe thin films along with ½ spin orbit components at higher B.E. The noise level is good in as-deposited ZnTe thin films but for Zn enriched ZnTe thin films noise level increased in Te spectra which might be due to less concentration after Zn enrichment and disturbance in structure.

Conclusions

The enrichment of Zn on to ZnTe thin films was reported using CSS technique. The variation in structure, surface morphology, electron transport and optical properties of ZnTe thin film having different concentrations of Zn were studied. As-deposited ZnTe thin films have 51 atomic % which further increased to 68 atomic % on the cost of reducing Te contents up to 32 atomic % from 49 atomic % confirmed in EDX. The Zn diffusion effects were analyzed successfully. XRD results shows that the as-deposited ZnTe and Zn -enriched thin films have a polycrystalline cubic structure with preferred orientation of [111]. XRD results showed the crystallinity of samples is affected by enrichment. In addition, the lattice constant was observed to 6.0980 A to 6.1026 A in Zn enriched Znte thin films which shows that bond length between Zn changes. Relative to enriched films the as-deposited ZnTe films were more tightly packed and larger. The crystallite size also reduced form 28 nm to 24 nm. There are smaller grains and discontinuities' between grains in Zn-enriched ZnTe thin films. In Zn-enrichment samples the optical energy band gap and transmission decreases as Zn concentrations increase which shows that Zn is a

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good reflector. The band gap of as-deposited ZnTe thin films was 2.16 eV which further reduced to 2.14 eV after Zn enrichment of 7 min. As Zn concentration increases to 68 atomic %, the electrical resistivity increases which may be the effect of discontinuities. EDX results also show a stoichiometry change after the enrichment process. XPS supported the argument of increased contents of Zn in ZnTe thin films. The increase in resistivity of Zn-enriched ZnTe thin films suggests that these thin films could not be used in solar cells as a back contact due to its high resistivity but Zn enriched ZnTe thin films are still good for optical properties.

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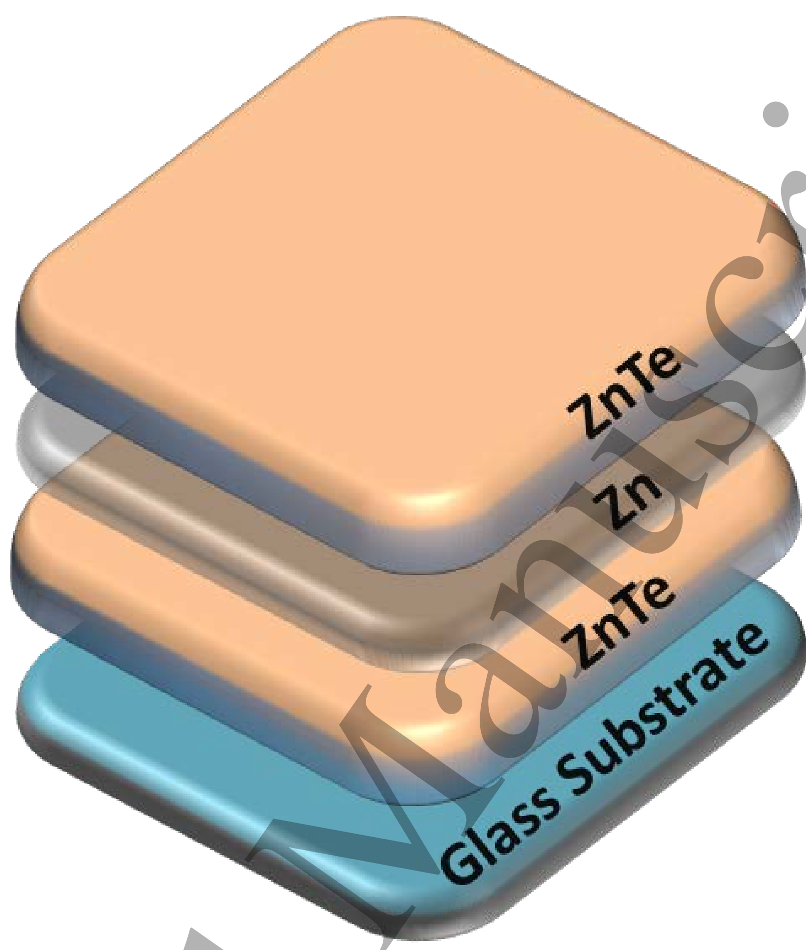

List of Figures

Fig. 1.

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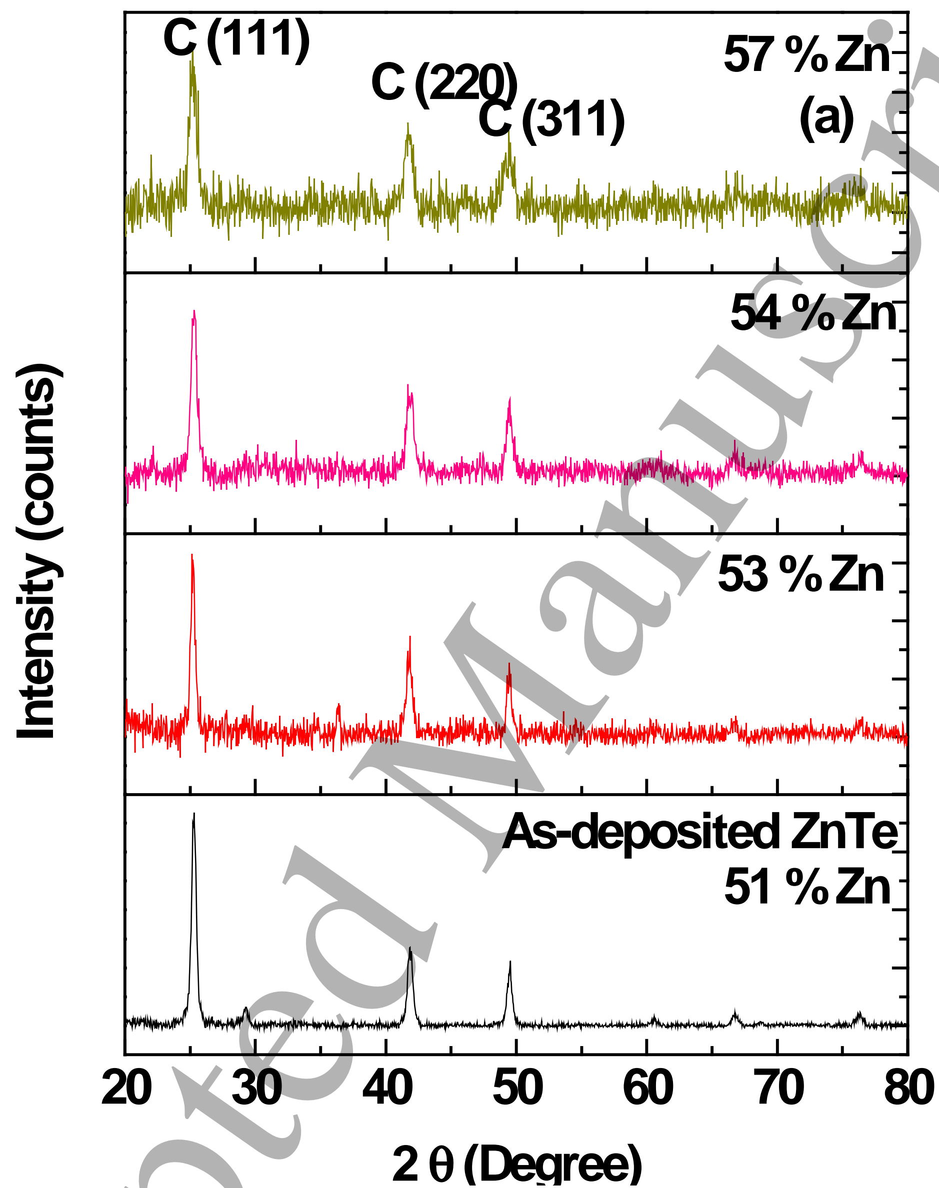

Fig. 2.

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Fig. 3.

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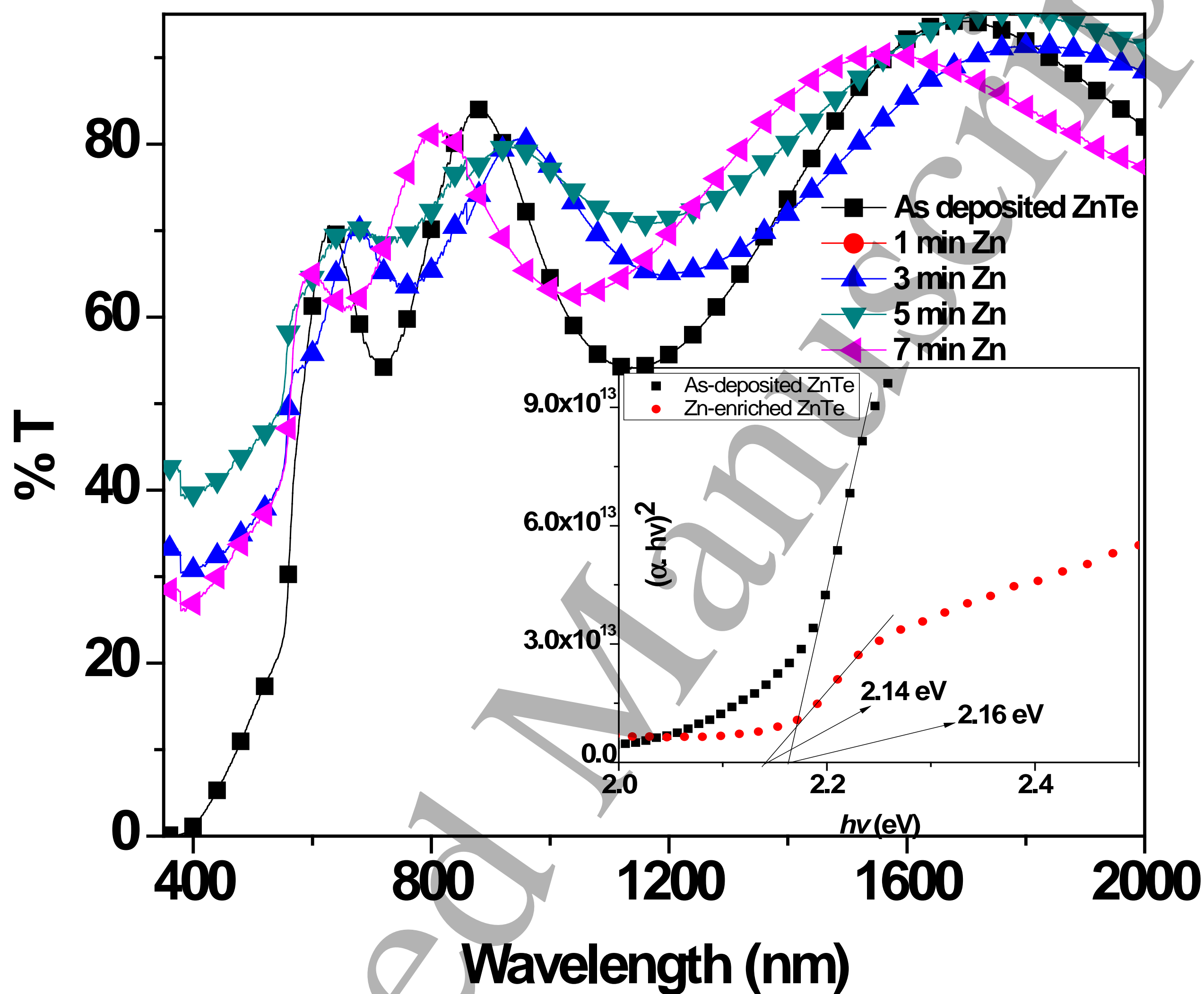

Fig. 4.

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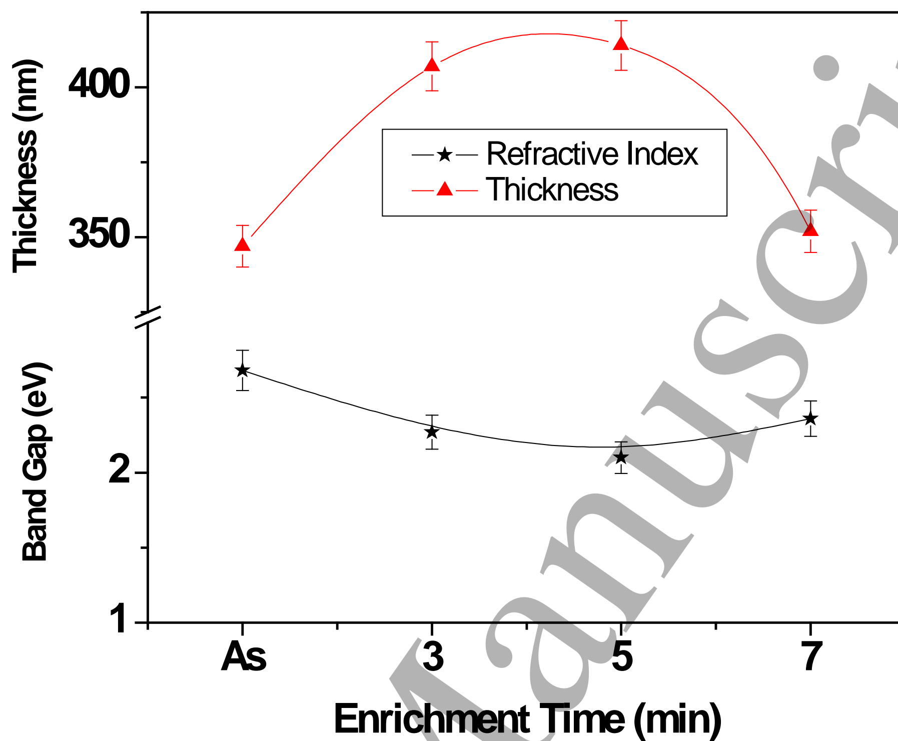

Fig. 5.

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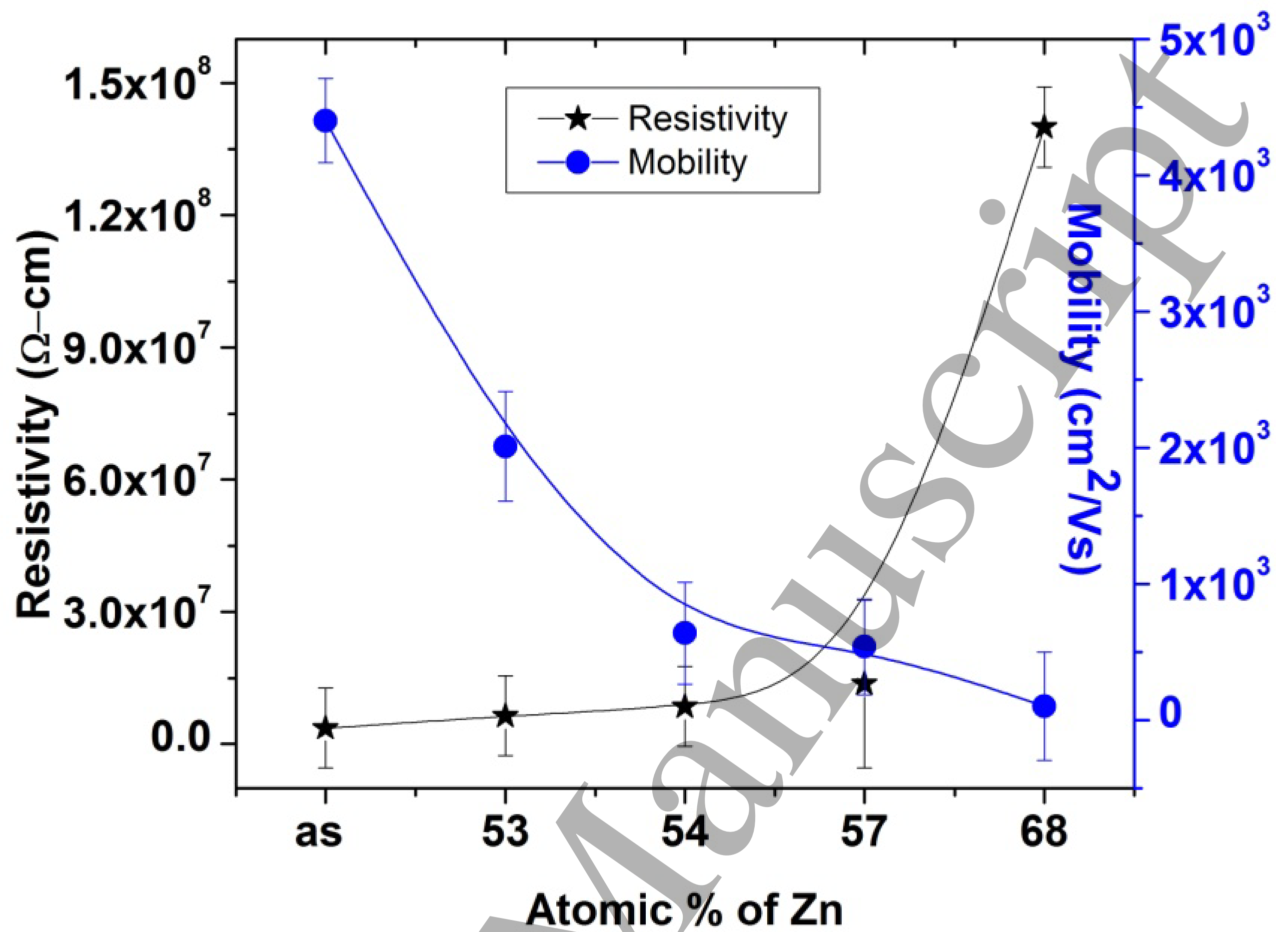

Fig. 6.

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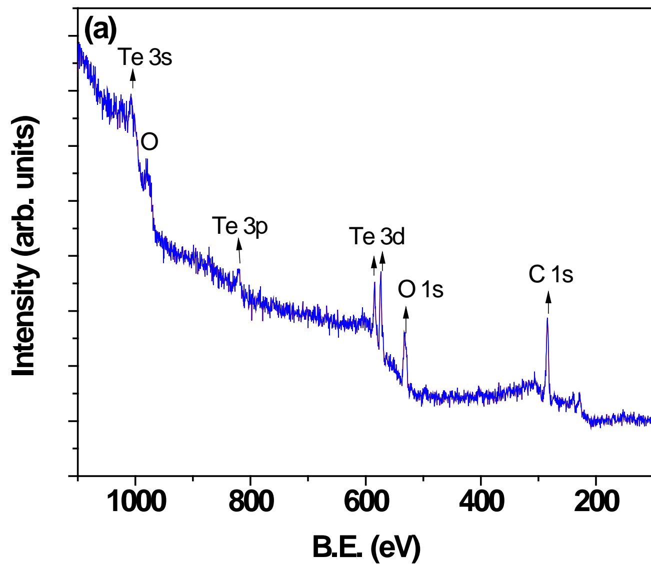

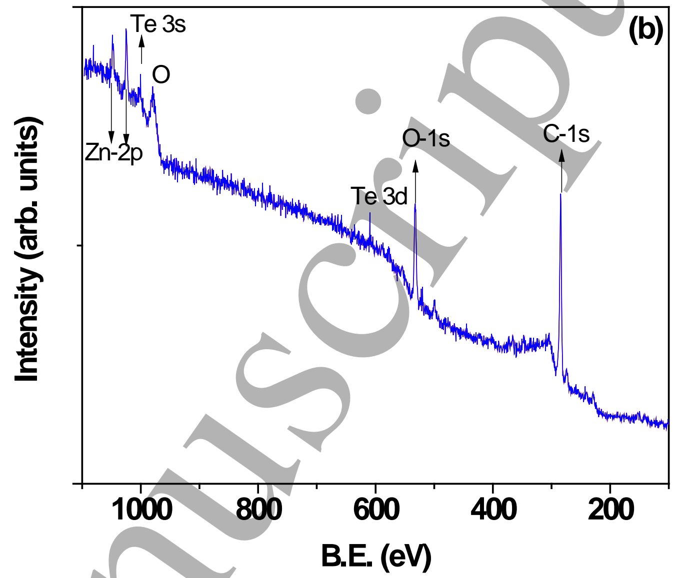

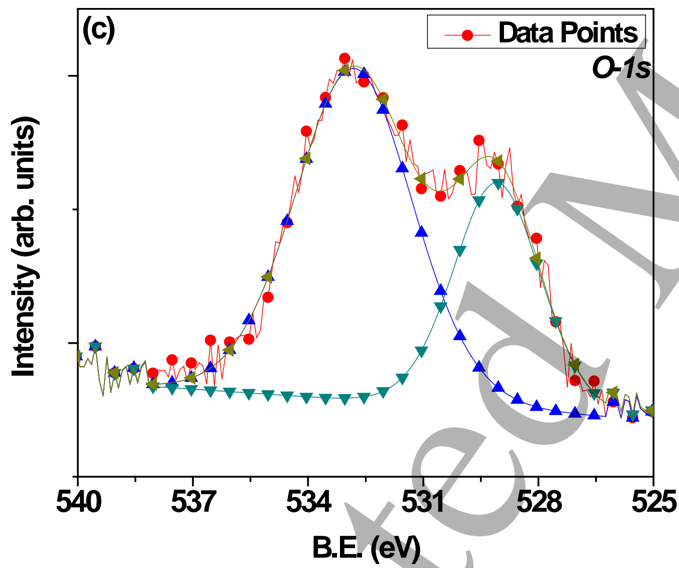

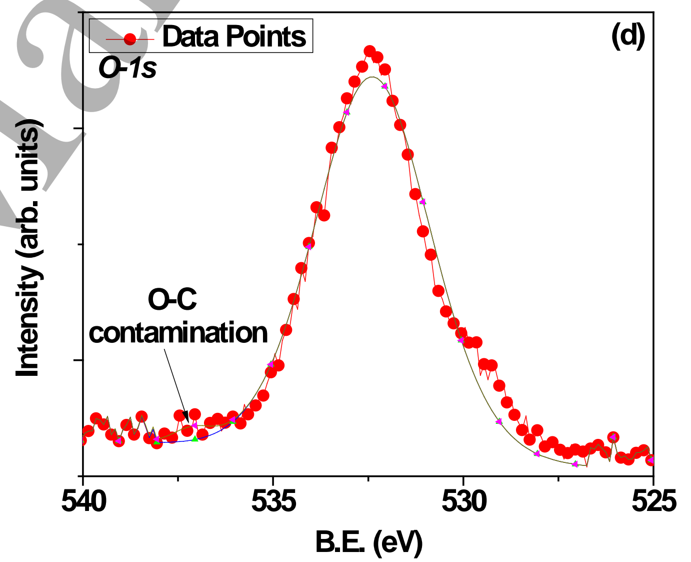

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Fig. 7.

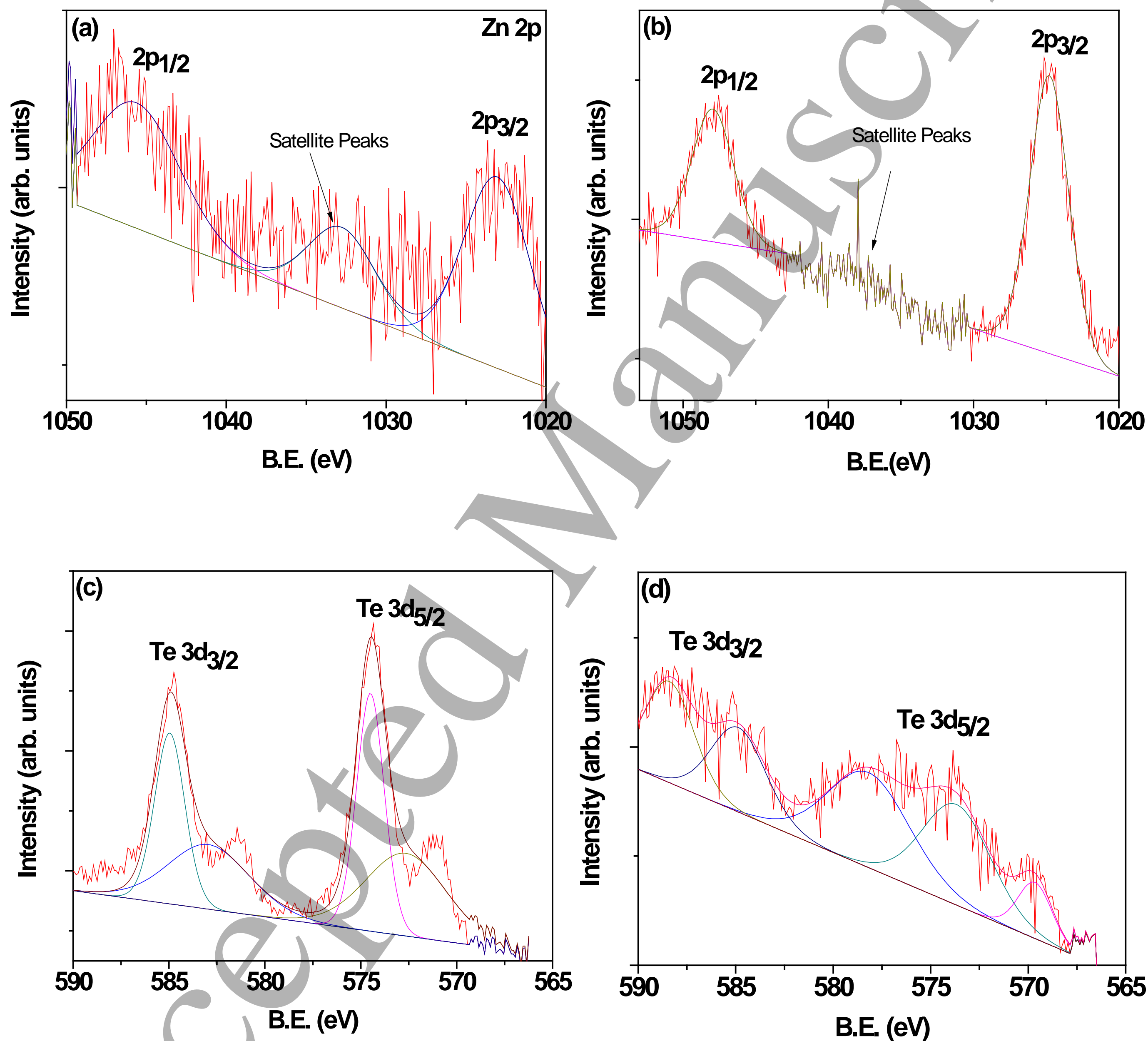

Fig. 8.

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Figures Caption

Figure 1 Schematics of Layer by Layer deposition of Zn-enriched ZnTe thin films

Figure 2 X-ray diffraction patterns of (a) Zn-enriched ZnTe thin films

Figure 3 SEM micrographs of (a) as-deposited ZnTe thin film (b) 3 min deposition of ZnTe (d) 7 min deposition of ZnTe thin film

Figure 4 Transmission spectra of as-deposited ZnTe thin film and Zn-enriched thin films (in-set Band gaps of as-deposited and 7 min Zn-enriched ZnTe thin films).

Figure 5 Variation of band gap and thickness with Zn enrichment time.

Figure 6 Variation in resistivity and mobility vs. atomic %.

Figure 7 (a) Survey Scan of as-deposited ZnTe (b) Survey Scan of Zn-enriched ZnTe (c) Oxygen scan for as-deposited ZnTe (d) Oxygen scan for Zn-enriched ZnTe

Figure 8 (a) Zn-2p Scan of as-deposited ZnTe (b) Zn-2p of Zn-enriched ZnTe (c) Te-3d scan for as-deposited ZnTe (d) Te-3d scan for Zn-enriched ZnTe

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Table 1 EDX analysis of as-deposited ZnTe thin film with Zn enriched ZnTe thin films

Zn-enrichment	Zn (Atomic %)	Te (Atomic %)
As-deposited ZnTe thin film	51	49
1 min Zn	53	47
3 min Zn	54	46
5 min Zn	57	43
7 min Zn	68	32

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