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The Influence of the Substrate Temperature on the Properties of Solar Cell Related Thin Films

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1. Introduction

Polycrystalline films are generally considered to consist of crystallites joined together by the grain boundaries. The grain boundary regions are disordered regions, characterized by the presence of a large number of defect states due to incomplete atomic bonding or departure from stoichiometry for compound semiconductors. These states, known as trap states, act as effective carrier traps and become charged after trapping [1]. The density of defects and impurities in the grain boundaries is larger than that within the grains, so as the orientations of the grains change, the density of traps also changes [2]. The density of trap states depends critically on the deposition parameters [1] including the substrate temperature.

By increasing the substrate temperature the grain size increases, grain boundaries become narrower and their number decreases, the height of the potential barrier between grains decreases, and some impurities go out from the grain boundaries and become effectively incorporated in the lattice and other impurities migrate to the grain boundaries. Evaporation of some elements changes stoichiometry and may create other defects. These changes produce changes in the structure and phase of the films. As a result, the electrical, optical and electronic properties will change too. The presence of some of these changes in a film depends on the deposition technique followed in producing the film, raw materials used and deposition conditions.

There are different deposition techniques to prepare thin films in which the deposition temperature is one of the main parameters that should be controlled to get high quality films. These methods include thermal evaporation [3-7], spray pyrolysis (SP) [8-27], chemical bath deposition (CBD) [28-29], dc magnetron sputtering [30] etc.

In the following sections we will discuss the effect of the substrate temperature on the structural, morphological, electrical and optical properties of thin films deposited by different techniques. A review of experimental results obtained by different authors will be performed with discussions and comparisons between different results.

2. Structural properties

There is agreement between authors that the increase in the substrate temperature improves the crystallinity of the films and encourages the change from amorphous to polycrystalline

structure and increases the grain size. X-ray diffraction (XRD) is the suitable tool to reveal these changes. For polycrystalline films, the variations of the intensity of Bragg peaks and their width at half maximum (FWHM) with substrate temperature are evidences on the changes in grain size. The narrowing of the lines of crystal growth at the higher substrate temperature (the decrease in FWHM) means that the grain size had increased. The shifts of the positions of the peaks refer to changes in lattice spacing and then lattice parameters. The appearance of some lines and disappearance of others with substrate temperature may mean a phase transition and/or the appearance or disappearance of other phases of the compounds under study or the presence of some elements. In this section different experimental results will be discussed to show the different effects of the substrate temperature on the structure of thin films through XRD diffractograms.

A lot of experimental results are found about the change from amorphous to polycrystalline structure with substrate temperature. For films prepared by the spray pyrolysis (SP) technique, a lot of workers [8, 12, 14, 18] found that CdS films prepared at substrate temperatures more than or around 200 °C are polycrystalline. Our CdS:In thin films [8] were prepared at $T_s = 350\text{--}490$ °C and they are polycrystalline. Bilgin et al. [12] prepared CdS thin films by the SP technique at substrate temperatures 473–623 K and found them to be polycrystalline. But we reported that [15] SnO₂:F thin films were amorphous at temperatures lower than 360 °C. Gordillo et al. [22] found that SnO₂ films deposited at temperatures lower than 300 °C grow with an amorphous structure, but those deposited at $T_s = 430$ °C present a polycrystalline structure. Rozati [2] found that increasing the substrate temperature causes the SnO₂ thin films to exhibit a strong orientation along (200).

Films prepared by chemical path deposition (CBD) which is a low temperature technique are in most cases partially or totally amorphous [28]. Liu et al. [28] prepared CdS films by this technique at deposition temperatures in the range 55–85 °C and found that all of the produced films have some amorphous component and an improved crystallinity with the increase of deposition temperature was obtained.

Numerous experimental data showed that the orientations of crystal growth and preferential orientation are sensitive to the substrate temperature. For ZnO spray-deposited thin films of the hexagonal wurtzite-type, Hichou et al. [10] found that the intensity of the diffraction peaks is strongly dependent on the substrate temperature, where they got maximum intensity at $T_s = 450$ °C. They found that the [002] direction is the main orientation and it is normal to the substrate plane. For these films some orientations of crystal growth appeared and others disappeared with the variation in the substrate temperature. For CdS thin films prepared by SP technique Acosta et al. [14] found that the intensity of the (002) line increases with temperature, while the (101) peak tends to disappear, which is exactly opposite to what we have in our diffractograms for CdS:In thin films [8]. But we also showed that the preferential orientation of the crystal growth is very sensitive to the substrate temperature. At $T_s = 350$ °C the preferential orientation in our diffractogram [8] is the H(002)/C(111)- The peaks of these two lines are very close to each other, so it is difficult to distinguish them-, and at $T_s = 460$ °C it is the H(101), but at $T_s = 490$ °C it is the H(112)/C(311)- also it is difficult to distinguish the peaks of these two lines.

As we see the orientations of crystal growth and the preferential orientations for a certain compound are different for different authors [8, 14]. In some cases [10] the preferential orientation does not change with the substrate temperature. The preferential orientation in Ashour's [12] diffractograms for CdS thin films which showed just the hexagonal phase is

the (101) which was not affected by the substrate temperature but all of the other lines are affected by the substrate temperature. Ashour [12], Pence et al. [13] and Bilgin et al. [18] did not find an influence of the substrate temperature on the preferential orientation for CdS films prepared by the SP technique. On the other hand, Abduev et al. [30] found that the position of the preferential orientation (002) of the hexagonal ZnO:Ga films of thickness 300 nm prepared by dc magnetron sputtering was shifted from 34.25° to 34.41° when the substrate temperature was increased from 50 to 300 °C.

A lot of authors observed a phase transition from cubic to hexagonal phase with the increase in substrate temperature [8, 14]. For spray-deposited CdS:In thin films our XRD diffractograms [8] showed a mixed (cubic and hexagonal phase) at $T_s = 350^\circ\text{C}$ which was converted to only hexagonal phase at $T_s = 490^\circ\text{C}$. Also for CdS:In thin films prepared by the spray pyrolysis technique Acosta et al. [14] found that X-ray diffractograms of the samples prepared with In/Cd = 0.1 in the solution, the intensity of the (002) peak shows a noticeable increase while the (101) tends to disappear for higher T_s . They [14] say that these variations in peak intensity might be related with phase transition from a cubic to a hexagonal structure. For films prepared by CBD the phase change was observed too, where Liu et al. [28] found that the phase of CdS films was ambiguous, at low deposition temperatures. That is, it couldn't be distinguished (cubic or hexagonal) because the positions of the (002) and (110) lines of the hexagonal structure are similar to the (111) and (220) lines of cubic one, making it difficult to conclude whether the film is purely hexagonal or purely cubic or a mixture of the two phases. But the phase was predominantly hexagonal at higher temperatures, where the presence of the lines (102) and (203) of the hexagonal phase are evidences.

On the other hand, Ashour [12] observed spray-deposited CdS thin films with just one phase (wurtzite) in the temperature range 200-400°C. Their [12] XRD diffractograms showed a preferential orientation (002) along the c-axis direction perpendicular to the substrate plane. Also Bilgin et al. [18] observed just the hexagonal phase for CdS thin films prepared by ultrasonic spray pyrolysis (USP) technique onto glass substrates at different temperatures ranging from 473 to 623K in 50K steps.

We conclude that authors who got a preferential orientation that is independent on the substrate temperature, got just one phase (hexagonal), while those who got a change in the preferential orientation with substrate temperature have a phase transition (from cubic to hexagonal). From these results it is confirmed that increasing the deposition temperature promotes phase transformation from cubic to hexagonal and improves the crystallinity in CdS films. Fig.1 displays the XRD diffractograms of spray-deposited SnO₂:F thin films taken at different substrate temperatures by Yadav et al.[31].

The grain size of the polycrystalline films greatly depends on the substrate temperature during deposition [1]. The grain size was found to increase with the substrate temperature for thin films prepared by different deposition techniques [8, 18, 30]. Acosta et al. [14] found that grain size increases with the substrate temperature and presents a smaller dispersion as T_s is increased. This increase is evident from the decrease in FWHM that they observed in their XRD diffractograms. For spray deposited CdS:In thin films, we [8] got an increase in grain size from 10 to 33 nm for the change in the substrate temperatures from 350 to 490 °C, which was calculated by using XRD diffractograms and Sherrer's formula. Bilgin et al. [18] obtained an increase of the grain size of the CdS films from 126 to 336 Å with increasing substrate temperature from 473 to 623K, showing the improvement in the crystallinity of the

films. Abduev et al. [30] got an increase from 32 to 36 nm for substrate temperature change from 50 to 300 °C and a decrease in FWHM from 0.27° to 0.24° for the same change in substrate temperature. This change was accompanied by a change in the lattice parameter c which decreased from $c = 5.232$ Å for the film deposited at 50 °C to $c = 5.208$ Å for the film deposited at the substrate temperature of 300 °C.

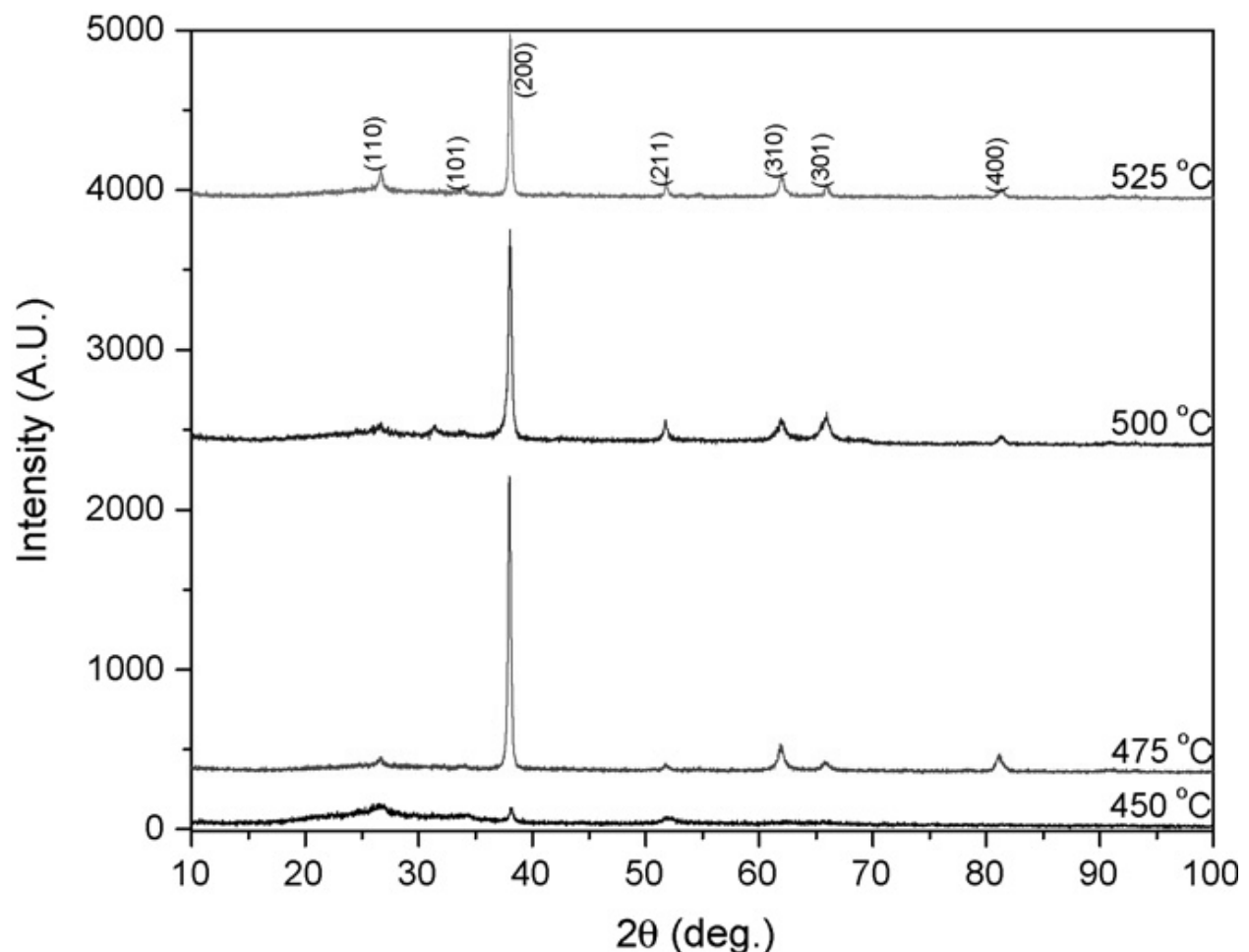


Fig. 1. XRD patterns of spray deposited SnO₂:F thin films at various substrate temperatures. Reprinted with permission from Yadav et al. [31]; Copyright © 2009, Elsevier.

Different authors plotted the relation between grain size and the substrate temperature [12, 18]. Bilgin et al. [18] obtained a non-linear relation with the curve concaves down and the grain size increases then becomes constant after a certain value of T_s . Ashour [12] and Patil et al [32] got increasing in grain size with increasing the substrate temperature where the curve concaves up which means that the grain size did not reach a certain size after which there is no increase.

Stress is also varying with substrate temperature as seen by Abduev et al. [30] who found that for gallium doped ZnO films, the film stress had varied with increasing the substrate temperature from -1.915 GPa (the compression condition) at the room substrate temperature to 0.174 GPa (the tension condition) at the substrate temperature $T = 300$ °C.

In thin film solar cells it is found that the substrate temperature is also an effective parameter on the structure and the grain size. For CdS/CdTe thin film solar cells deposited

on SnO₂-coated Corning 7059 borosilicate glass, or (100) Si wafers, the substrate temperature caused an increase in the grain size of the CdTe layer as found by Al-Jassim et al. [33]. Also for CdS/CdTe thin films Dhere et al. [34] used AFM measurements and showed that there was no CdTe grain growth, for samples deposited at different substrate temperatures after CdCl₂ heat-treatment, but samples deposited at lower temperatures have smaller grains and consequently higher grain boundary volume.

Substrate temperature enhances the interdiffusion in the interface region in CdS/CdTe polycrystalline thin films. Al-Jassim et al. [33] found that at deposition temperatures below 450 °C, only small amounts (~1%) of sulfur were detected in the CdTe films in the vicinity of the interface. On the other hand at deposition temperature of 625 °C, sulfur levels exceeding 10% in CdTe films were detected. This clearly indicates that CdTe devices deposited at high temperatures have an alloyed (CdS_xTe_{1-x}) active region. Dhere et al. [34] found that compositional analysis by small-area, energy dispersive X-ray analysis (EDS) revealed significant sulfur diffusion into the CdTe film. The amount of sulfur was below detection limit (<0.1 at.%) at the lowest deposition temperature, and increased with increasing deposition temperature.

3. Film morphology

Substrate temperature is an effective parameter in determining the shape and size of grains, surface roughness, porosity and density of voids as found by different authors.

For CdS:In thin films prepared by the SP technique at different substrate temperatures we [8] observed different surface morphologies. At 350 °C long rods or chains were observed, which are related to complex compounds. At $T_s = 460$ °C we got open cubes and at $T_s = 490$ °C spherical grains were observed. For SnO₂:F thin films prepared by the SP technique different morphologies were observed for films prepared at different substrate temperature [15] too. El Hichou et al. [10] observed a change in surface morphology for ZnO spray deposited thin films with the substrate temperature, where they have the larger grains in the film deposited at $T_s = 450$ °C. The film deposited at the smallest substrate temperature $T_s = 350$ °C had shown porous structure but films deposited at $T_s > 350$ °C had a close-packed morphology.

For spray-deposited CdS:In films Acosta et al. [14] got AFM images which are shown in Figs. 2. Besides the grains size and topology details, it can be observed that grains present regular shape and surfaces for T_s values ranging from 300 °C (Fig.2a) to 400 °C (Fig.2b). In samples obtained at $T_s = 425$ °C (Fig.2c) and 450 °C (Fig.2d) respectively, aggregates of small grains covering grains with bigger sizes are found everywhere. Noting that these results are related with the XRD diffractograms in that reference. Since the substrate temperature is the only parameter that changes, Acosta et al. [14] say that the changes observed in surface morphology might have to do with particular specific thermodynamic parameters during the pyrolysis and nucleation processes.

Besides increasing the grain size, the increase in the substrate temperature decreases the density of voids. Fig.3 illustrates the SEM micrographs of the surfaces of the CdS films deposited by CBD at 55 °C, 65 °C, 75 °C and 85 °C taken by Liu et al. [28]. These micrographs show that increasing the deposition temperature results in an increase in grain size and consequently a decrease in voids. When the deposition temperature is 55 °C, CdS particles of 50 nm dot the surface of the glass substrate attributing to the controlled

nucleation process associated with the low deposition rate. CdS films deposited at 65 °C have spherical particles of about 100 nm in size. The voids with different sizes ranging from 50 nm to 300 nm are still observed, indicating low packing density of the film. The surface of the CdS films deposited at 75 °C is compact and smooth, showing a granular structure with well-defined grain boundaries. It indicates that the increase of the bath temperature is an effective method to diminish voids on the CdS films. But it is noticed that CdS film deposited at higher temperature 85 °C displays a rather rough, inhomogeneous surface with overgrowth grains.

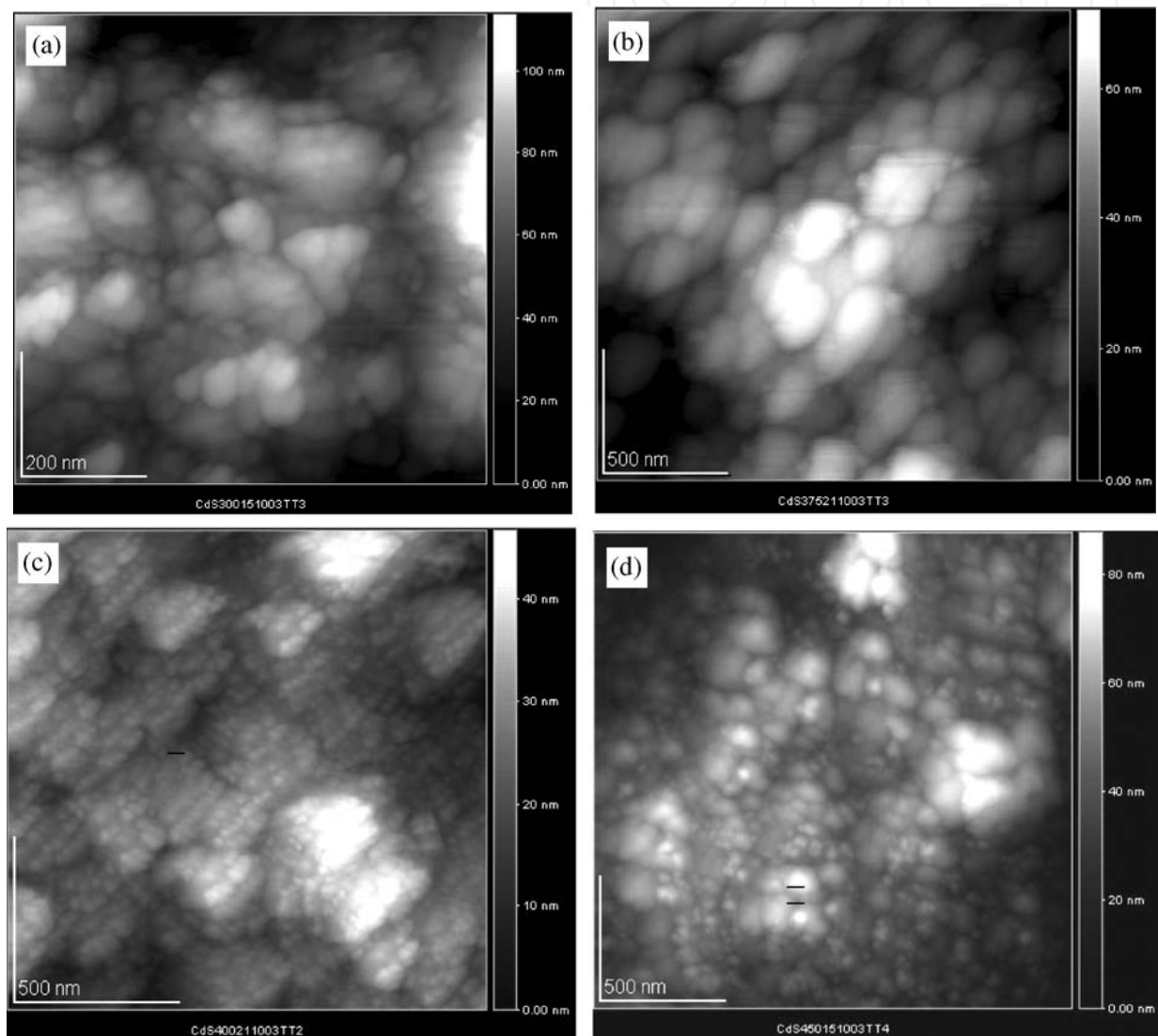


Fig. 2. AFM micrographs of CdS:In deposited by SP technique for different substrate temperatures. a) $T_s = 300\text{ }^{\circ}\text{C}$: The grain size ranges from 50 to 75 nm. b) $T_s = 375\text{ }^{\circ}\text{C}$: The grain size varies between 75 and 225 nm. c) $T_s = 400\text{ }^{\circ}\text{C}$: The grain size between 45 and 60 nm and the size of grain agglomerates is between 170 and 350 nm. d) $T_s = 450\text{ }^{\circ}\text{C}$: The grain size varies between 25 and 65 nm and the size of agglomerates is between 180 and 400 nm, respectively. Reprinted with permission from Acosta et al. [14]; Copyright © 2004, Elsevier.

Other authors showed that roughness increases with the substrate temperature such as Haug et al. [35] who found that the CdTe layers show a higher roughness with increasing substrate temperature, but they are less compact. Atomic Force Microscopy analysis showed that the root mean square (RMS) surface roughness ranges from 100 nm for 500 °C films to 550 nm for 600 °C films. On the other hand some authors found a decrease in roughness with the substrate temperature [14, 36]. For CdS:In thin films prepared by the SP technique Acosta et al. [14] found that as T_s begins to increase, the surface shows a decrease in roughness in zones surrounding pore-like configuration. Also surface roughness was found to decrease with substrate temperature by Abduev et al. [30] for ZnO thin films prepared by magnetron sputtering. Li Zhang et al. [36] also found that surface roughness decreases with the substrate temperature.

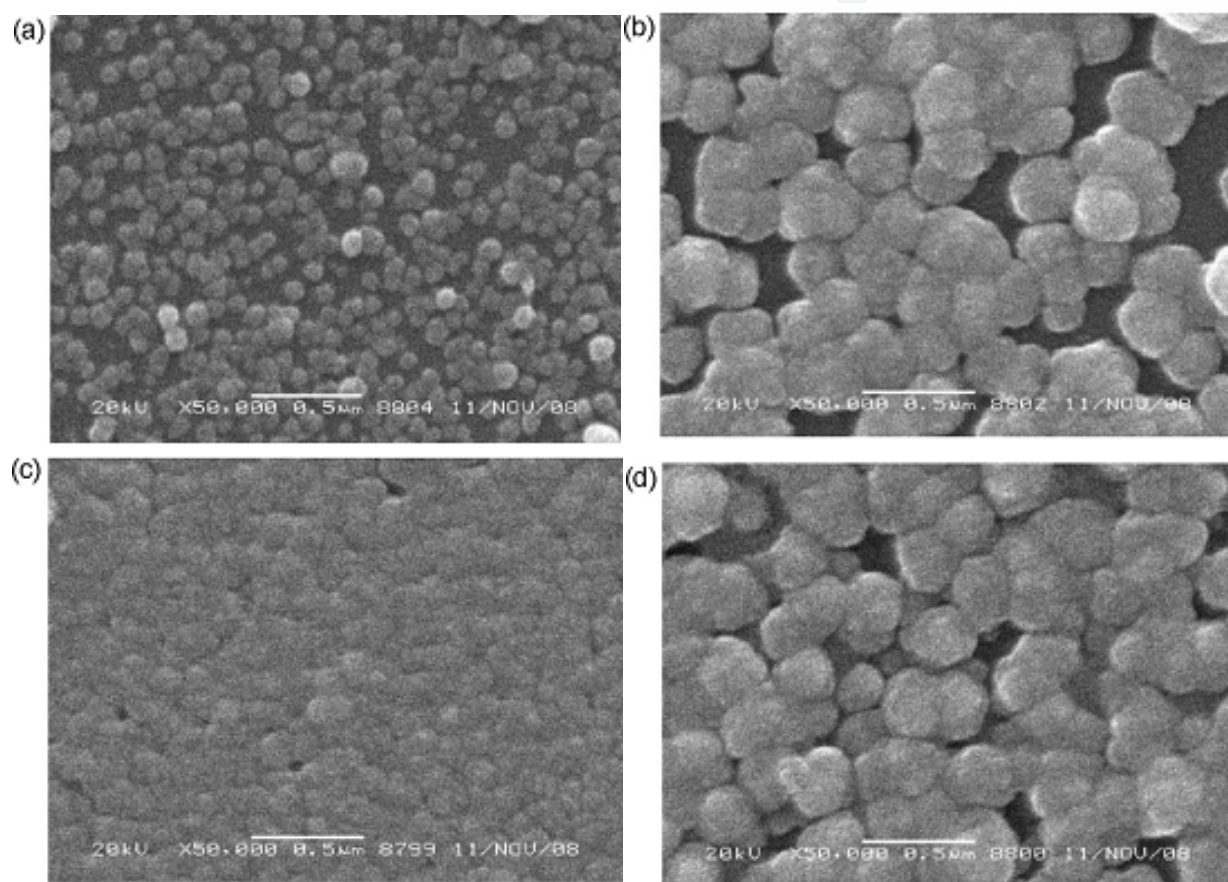


Fig. 3. SEM micrographs of CdS films grown at different temperatures by CBD: a) 55 °C. b) 65 °C. c) 75 °C. d) 85 °C. Reprinted with permission from Liu et al. [28]; Copyright © 2010, Elsevier.

4. Electrical properties

Changes in the structural and morphological properties of the films with the substrate temperature have their effect on the electrical properties of the films. These changes include the phase change, enlargement of grains, diminishing of grain boundaries, motion of impurities from or to the grain boundary region and evaporation of some elements during deposition process. For undoped compound semiconductors, it is found that stoichiometry increases with the substrate temperature due to the reduction in the density of defect states.

These variations will change the number of charge carriers which directly affect the resistivity of the films. They also affect the mechanism of carrier transport and then the linearity of current-voltage characteristics. We will discuss some of the experimental results which include some of these changes. The occurrence of all of these changes or some of them simultaneously has a net effect on the electrical properties as will be seen in the experimental results obtained by different authors.

I-V plots are usually used to investigate the electrical conduction mechanisms and to determine the resistivity of the films. Linear I-V plots mean that the ohmic conduction mechanism is predominant (i.e. electronic conduction through grains). Nonlinear I-V plots mean that other conduction mechanisms are found which are non- electronic and the conduction through the grain boundaries is predominant. It is known that the trap states, which act as effective carrier traps and become charged after trapping result in the appearance of a potential barrier which impedes the flow of majority carriers from one grain to another and affects the electrical conductivity of the films [1]. Three possible mechanisms may govern the grain-to-grain carrier transport through the potential barrier mentioned above:

- i. over-the-barrier thermionic emission of carriers having sufficient energy to surmount the barrier;
- ii. quantum mechanical tunneling from grain to grain through the barrier by carriers having energy less than the barrier height; and
- iii. hopping through the localized states.

The relative magnitudes of the barrier height and the width of the barrier will depend critically on the crystallite size and carrier concentration. Depending on the above, with respect to the energy of carriers, one of the above processes will be operative in charge transport in polycrystalline semiconductor films. The thermionic process is limited by the height of the barrier. The thermionic emission will be temperature dependent, with activation of the order of the barrier height, while the tunneling process would be almost independent of temperature. For films with a barrier height larger than what could be surmounted by the carriers with the energy at lower temperatures, tunneling seems to be the dominant mechanism of charge transport in the films. The films grown at lower temperatures will have a smaller crystallite size, and as such the grain boundary region will be substantially larger than the grains. The grain boundary regions being disordered and highly resistive, the film will look like a conglomeration of crystallites embedded in the amorphous matrix [1].

Linear I-V characteristics were recorded by us at all deposition temperatures under study for CdS:In [8], SnO₂:F thin films prepared by the SP technique on glass substrates [15, 25-26], undoped ZnO thin films [37-38], Al-doped ZnO thin films [39] and CdTe thin films prepared by vacuum evaporation [40]. Bilgin et al. [18] have linear I-V curve for a CdS thin film prepared by ultrasonic spray pyrolysis (USP) technique at 523 K (250 °C) in the voltage range 0-100 V. This means that this film has not got trapped structure and so, the ohmic conduction mechanism is dominant for this film in the whole voltage range. For the sample obtained at 473 K they [18] found four regions with different slopes where the drawing was performed on log-log scale. The ohmic conduction is dominant in the 0.1-8V voltage range where the slope is 1.11. The slope is 2.24 in the second region which is called space charge limited (SCL) region. The existence of this region shows that CdS films have shallow trapped structure. Then, the trap filled limited (TFL) region comes as the third region. This

implies that all traps are filled. The last region with a slope of 2.42 shows trap free region. The other two films prepared by Bilgin et al. [18] at 573 and 623K have deep trapped structure. The mechanism in the sample obtained at 623K is more complex. There are three deep trap levels with different energies for this film. The voltage ranges of these three regions are 16–24, 40–56 and 80–100V, respectively.

Numerous experimental results showed that the resistivity of semiconducting thin films decreases with the deposition temperature [8, 12, 15, 23–24]. For spray-deposited CdS:In thin films we [8] got a decrease of the room temperature resistivity in the dark from $1.5 \times 10^8 \Omega \cdot \text{cm}$ at $T_s = 380^\circ \text{C}$ to $1.2 \times 10^6 \Omega \cdot \text{cm}$ at $T_s = 490^\circ \text{C}$ and we explained this by the encouragement of crystal growth at higher substrate temperature as concluded from the XRD diffractograms. For CdS thin films prepared by SP technique Ashour [12] got a decrease of room temperature resistivity (10^5 – 10^3) with substrate temperature in the range 200–400 °C and related it to the growth of the grain size and the improvement in film stoichiometry. For SnO₂:F thin films prepared by SP technique [15] we found a rapid decrease of the resistivity with the substrate temperature. The same behavior was also observed by Shanthi et al. [23] for undoped SnO₂ films prepared by the spray pyrolysis, where they recorded a gradual decrease in the resistivity with the deposition temperature in the range 340–540 °C. Also the same behavior was observed by Zaouk et al. [24] for fluorine-doped tin oxide thin films prepared by electrostatic spray pyrolysis at substrate temperatures in the range 400–550 °C.

Other authors found a decrease in resistivity until a certain temperature and then it increases again [15, 18–20, 30]. Abduiev et al. [30] found that for ZnO thin films prepared by dc magnetron sputtering, the growth temperature dependence of resistivity is nonmonotonic. They found that the lowest resistivity ($3.8 \times 10^{-4} \Omega \cdot \text{cm}$) is attained at the substrate temperature of 250 °C; then, it increases insignificantly. On the other hand some authors found an increase in resistivity followed by a decrease [28, 36]. Liu et al. [28] measured electrical resistivity for CdS thin films prepared by CBD and found that it arises to $5 \times 10^5 \Omega \cdot \text{cm}$ level for temperature 55–75 °C, then it decreases to $7.5 \times 10^4 \Omega \cdot \text{cm}$ for 80 °C and $8.5 \times 10^3 \Omega \cdot \text{cm}$ for 85 °C sharply. They explained this variation as can be due to the cubic-hexagonal transformation in agreement with structural and optical analysis. Li Zhang et al. [36] also observed an increase in the resistivity with substrate temperature followed by a decrease for Cu(In, Ga)Se₂ films prepared by the three-stage co-evaporation process.

From the results of Hall coefficients measurements taken by Liu et al. [28] for CdS thin films prepared by CBD it is found that the CdS films are of n-type conductivity. It is also found that mobility increases from $3.228 \times 10^{-1} \text{ cm}^2/(\text{V} \cdot \text{s})$ to $6.517 \text{ cm}^2/(\text{V} \cdot \text{s})$ with the increase of deposition temperature from 55 °C to 75 °C tardily which can be understood by considering the increase of the grain sizes and decrease of the grain boundaries. However, the mobility increases to $6.513 \times 10^1 \text{ cm}^2/(\text{V} \cdot \text{s})$ at 80 °C and $1.183 \times 10^2 \text{ cm}^2/(\text{V} \cdot \text{s})$ at 85 °C promptly in contrast with the behavior of resistivity. This behavior can be attributed to the transition from the cubic to the hexagonal phase again, besides the improvement of crystallinity.

The investigation of Hall parameters by Abduiev et al. [30] showed that the charge carrier mobility continuously grows with increasing the substrate temperature, and the free carrier concentration has the peak ($1.27 \times 10^{21} \text{ cm}^{-3}$) at the substrate temperature of 250 °C. Such a character of the temperature dependence of the free carrier concentration in doped ZnO films is caused by the fact that in zinc oxide there are always intrinsic donor defects in the

bulk and at the surface grains in addition to the impurity donors introduced in the ZnO lattice (the substitutional impurity). The multiple experimental and theoretical data indicate that oxygen vacancies play an important role in the conductivity of transparent conducting films. It is observed that the behavior of the resistivity to a large extent is reflected by the carrier density and only little by the mobility; low resistivity corresponds to high carrier density and vice versa.

Abduev et al. [30] explains this behavior by: At low film growth temperatures ($T \leq 150^\circ\text{C}$), the main contribution to the charge carrier concentration is made by intrinsic defects and the efficiency of the Ga incorporation in the ZnO lattice is low, which is confirmed by the small values of Hall mobility in these films. With increasing substrate temperature, the efficiency of impurity atom incorporation into the crystal lattice increases and the concentration of intrinsic defects inside ZnO grains decreases, which is confirmed by the data of the X-ray diffraction analysis and by a substantial increase in the Hall mobility values at a deposition temperature of 200°C . The increase in Hall mobility at $T \geq 200^\circ\text{C}$ is caused also by the lowering of potential barriers for free carriers on the grain boundaries due to the intensification of the process of oxygen thermal desorption from the grain surface during the film growth in vacuum.

For thin film solar cells, the performance is dependent on the substrate temperature. That is, the short-circuit current density J_{sc} , open circuit voltage V_{OC} , Fill factor FF and efficiency η are all dependent on the substrate temperature.

Li Zhang et al. [36] showed that for CIGS solar cells the cell performance increases with the increase in the growth temperature. It is noticed that the cell efficiency increases with increase in the growth temperature. When the substrate temperature is 380°C the efficiency is very low. The best efficiency at 550°C is related to the better structural and electrical properties. It is noticed that the effects of the substrate temperature on fill factor (FF) and open circuit voltage (V_{OC}) shows similar trends with cell efficiency. That means the dependence of cell efficiency on the substrate temperature is dominated by the value of FF and V_{OC} . That can be explained by the improvement of carrier concentration and resistivity of CIGS films dominated by Na incorporation diffused from the glass substrate which is expected to be temperature dependent.

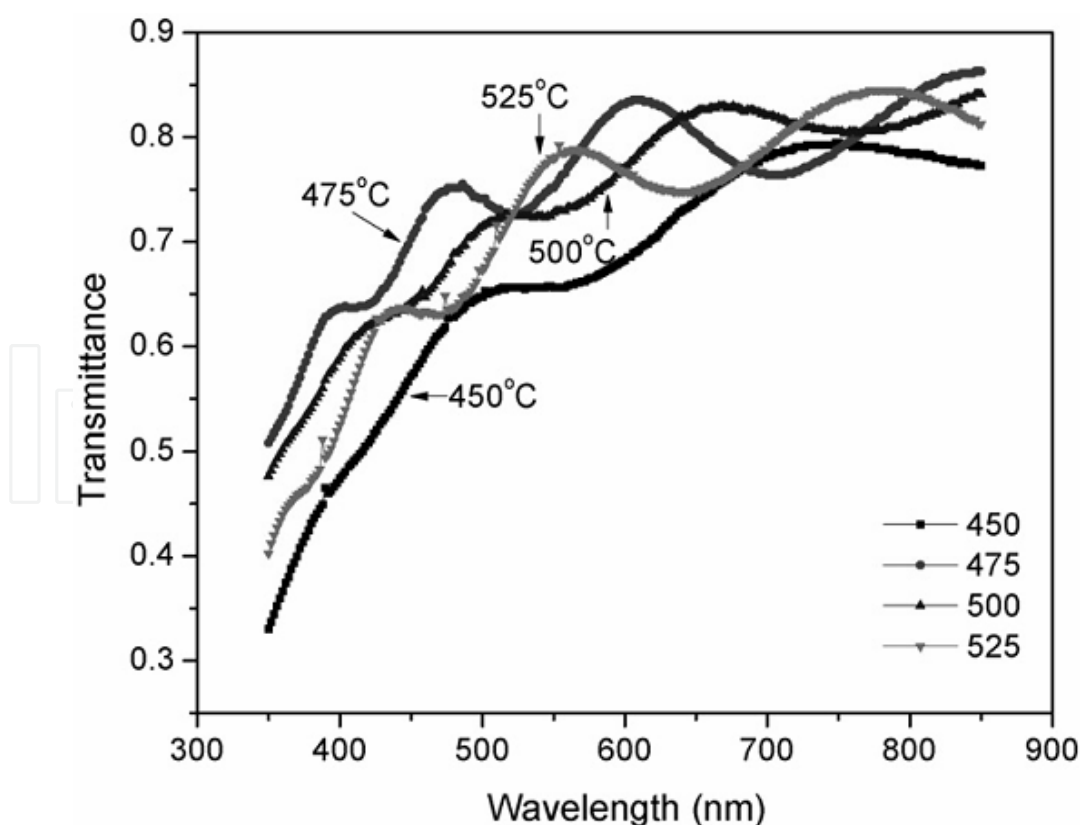
Julio et al. [27] investigated the electrical and photovoltaic properties of ZnO/CdTe heterojunctions where ZnO was prepared by the SP technique on CdTe single crystal under the effect of varying the substrate temperature and post deposition temperature for annealing in H_2 . For substrate temperatures in the range $T_s = 430\text{--}490^\circ\text{C}$ for the spray-pyrolysis deposition the optimum behavior was obtained for $T_s = 460^\circ\text{C}$. They [27] found that as the substrate temperature is increased from 430 to 460°C the dark J-V characteristics improved considerably and shifted towards higher bias voltages, remaining almost parallel to one another and exhibiting a strong reduction in J_0 with increasing T_s . The reverse current characteristics show similar improvement. Under simulated illumination, large values of short-circuit current were observed: typically of the order of 20 mA/cm^2 for illumination of 87 mW/cm^2 for T_s less than 470°C . The solar conversion efficiency increased markedly with increasing T_s up to 460°C , primarily because of an increase in V_{OC} and a fill factor which can be correlated with the decrease in J_0 . The improvement in junction characteristics observed with increasing substrate temperature up to 460°C according to Julio et al. [27] may have several explanations: The density of the interface states may depend on the orientation of the film; preferential orientation increases as a characteristic temperature is reached.

5. Optical properties

Since the substrate temperature affects the structural properties of the films including lattice parameters and phase, and the electrical properties including the density of charge carriers and density of traps, the optical properties will change.

The absorption coefficient is dependent on the conductivity which is a function of the density of charge carriers. The change in the absorption coefficient will change the transmittance of the films. Some authors found that the transmittance of thin films increases with the substrate temperature [12, 31]; other workers found a decrease in the transmittance with substrate temperature [14] and others found no change in the transmittance of thin films with the substrate temperature [8].

The increase of transmittance with substrate temperature was recorded by Ashour [12] who found an increase of the transmittance with substrate temperature in the range 200-400 °C for undoped spray pyrolyzed CdS thin films of thickness 500 nm. He attributed this improvement in transmittance with substrate temperature to either the decrease in thickness or the improvement in perfection and stoichiometry of the films. Yadav et al. [31] found an increase in transmission with the increase in the substrate temperature for SnO₂:F thin films prepared by the spray pyrolysis technique on glass substrates at substrate temperatures 450-525 °C (Fig.5a). At lower temperatures, i.e. at 450 °C, relatively lower transmission is due to the formation of whitish milky films due to incomplete decomposition of the sprayed droplets.



(a)

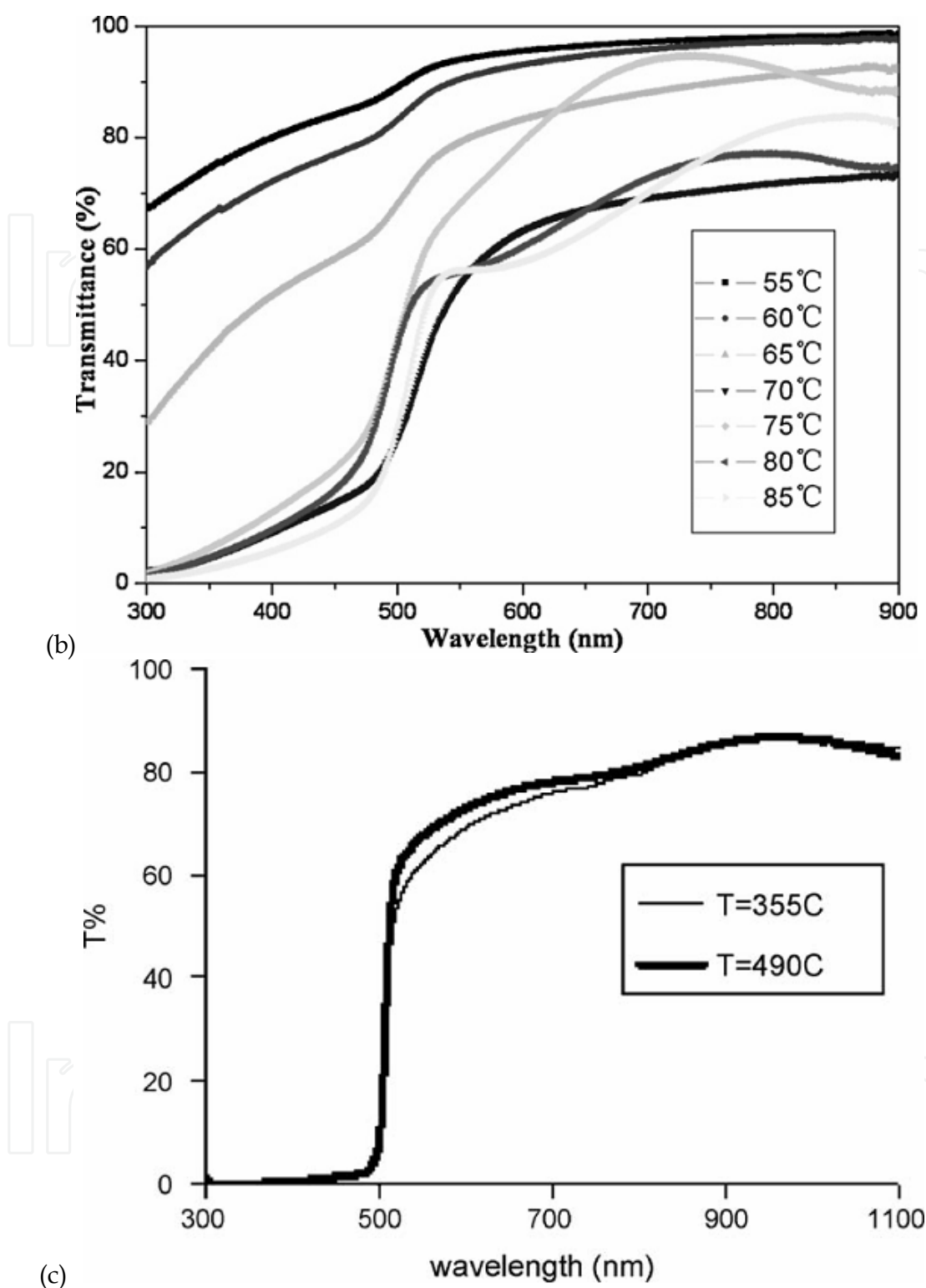


Fig. 4. The optical transmittance of thin films at different substrate temperatures against the wavelength of incident radiation. a) SnO₂:F thin films. Reprinted with permission from Yadav et al. [31]; Copyright © 2009, Elsevier. b) CdS films. Reprinted with permission from Liu et al. [28]; Copyright © 2010, Elsevier. c) CdS:In thin films [8]. permission from [9], S. A. Studenikin et al. Journal of Applied physics, 84 (4) (1998), 2287-2294. Copyright [1998], American Institute of Physics.

The decrease in transmittance with substrate temperature was observed by Acosta et al. [14] for CdS:In thin films prepared by the spray pyrolysis technique, but at the same time they have a variable thickness with substrate temperature (decrease then increase) which may be the main reason of the decrease in transmittance. Also the decrease of transmittance with substrate temperature was recorded by Liu et al. [28] for CdS films prepared by CBD (Fig.5b). It can be observed that the transmittance of the film decreases rapidly with the increase of the deposition temperature from 55 °C up to 70 °C, which is caused by reducing voids and increasing film thickness mainly. For higher deposition temperatures, the transmittance initially increases to 84% for the film deposited at 75 °C due to less light scattering by its smoothest surface. It decreases to about 68% at deposition temperatures above 80 °C, which may be due to either more light scattering on their rough surfaces or the transition of the CdS phase from the cubic to hexagonal structure [28]. Another observation about these transmission spectra is that the absorption edge shifts towards higher wavelength side, suggesting a reduction in the bandgap value, and it becomes steeper with deposition temperature rising.

No dependence of transmittance on the substrate temperature was recorded by us [8] for CdS:In thin films prepared by the spray pyrolysis technique (Fig.5c). We think that the transmittance of our films was independent of the substrate temperature due to the way of spraying that we used. We sprayed for 10 s, waited 1–3 min and then sprayed again. The preparation of a set of films by this method takes a long period of time depending on the required thickness of the films (around 4 h for films of thickness around 1 µm). Ashour [12] did not mention the deposition time that he used or the way he followed in spraying, while Acosta et al. [14] produced their films with a deposition time of 5 min in all cases. From our trials we found that using longer deposition times results in less transparent films, and the short period of spraying results in highly transparent films. Also for transparent conducting gallium_doped ZnO films prepared by magnetron sputtering on glass substrates at $T_s = 100\text{--}300$ °C, Abduev et al. [30] have high transmittance which is approximately independent on the substrate temperature, but they observed a shift of the absorption edge in spectra to shorter wavelengths.

The dependence of the bandgap energy on substrate temperature was recorded by different workers [8, 12, 18, 30]. One reason of this dependence is that stress is greatest in films deposited at low temperatures, which results in wider bandgap than bulk. So the increase in substrate temperature reduces stress and then reduces the bandgap energy. Another reason is the increase in interplanar distances or equivalently the lattice parameter with the substrate temperature which appears as a shift in the XRD diffractogram towards smaller angles. It is well known that the lattice parameter and energy gap have opposite behavior [41]. Other reasons include the change in the density of charge carriers with the substrate temperature and the movement of dopants from grain boundaries to the grains to be effectively incorporated in the crystal lattice.

A slight increase in the optical bandgap energy with substrate temperature was observed by different authors [8, 12, 18, 42]. For spray-deposited CdS:In thin films we [8] found that E_g slightly increases with the substrate temperature. This increase can be related to the phase change from mixed (cubic and hexagonal) to hexagonal phase as seen in XRD diffractograms in reference [8]. We found that the $E_g = 2.42$ eV for a film deposited at 355 °C and $E_g = 2.44$ eV for a film deposited at 490 °C. Bilgin et al. [18] observed slight increase of

E_g for CdS films prepared by ultrasonic spray pyrolysis (USP) technique onto glass substrates at different temperatures ranging from 473 to 623K. Ashour [12] got $E_g = 2.39 - 2.42$ eV for CdS films prepared by chemical spray-pyrolysis technique on glass at substrate temperatures in the range 200-400 °C. Melsheimer and Ziegler [42] observed this increase of E_g with substrate temperature for tin dioxide thin films prepared by the spray pyrolysis technique. Values of $E_g = 2.51-3.05$ eV were obtained for amorphous and partially polycrystalline thin films prepared at $T_s = 340-410$ °C, and $E_g = 3.35-3.43$ eV for polycrystalline tin dioxide thin films produced at $T_s = 420-500$ °C.

An increase followed by a decrease in bandgap energy with substrate temperature was observed by Abduev et al. [30] for ZnO:Ga thin films prepared by dc magnetron sputtering (from 3.52 to 3.72 eV when T_s increases to 250 °C) then a decrease to 3.65 eV at 300 °C. This result was consistent with their electrical properties. Other authors got a decrease then an increase in the bandgap energy with substrate temperature. For spray-deposited indium doped CdS thin films on glass substrates, Acosta et al. [14] got a decrease in the bandgap energy with substrate temperature from 300-425 °C then it increased at $T_s = 450$ °C. They interpreted the increase observed in E_g by saying that it might be related with the variations in size and morphology of grains.

The decrease of bandgap energy with substrate temperature was observed by some authors such as Liu et al. [28] who observed this for CdS films prepared by CBD. But it is important to notice that the thickness of their films is not constant, which means that the decrease in bandgap is also related to the increase in film thickness not only to the increase in substrate temperature.

Urbach tail width E_e which is known to be constant or weakly dependent on temperature and is often interpreted as the width of the tail of localized states in the band gap [43] was also found to be randomly affected by the substrate temperature as shown by Bilgin et al. [18] for CdS thin films prepared by USP technique, where it has values in the range 122-188 meV for substrate temperatures in the range 473-623 K. But Melsheimer and Ziegler [42] observed a decrease of E_e with substrate temperature for spray-deposited tin dioxide thin films. For amorphous and partially polycrystalline films prepared at $T_s = 340-410$ °C, it decreased from 530 to 350 meV. For polycrystalline films prepared at $T_s = 420-500$ °C it decreased from 240-200 meV.

Photoluminescence and cathodoluminescence always used to explore defects and traps. But the density of trap states depends critically on the deposition parameters [1] and hence on the substrate temperature. Changes in phase, bandgap and density of traps will be reflected on the photoluminescence and cathodoluminescence spectra. It is found that the luminescence intensity depends strongly on the deposition temperature [10].

Fig.6 displays the photoluminescence (PL) spectra for a set of ZnO films deposited by the SP technique by Studenikin et al. [9] at different substrate temperatures and annealed identically in forming gas at 750 °C for 40 min. As we said before, the photoluminescence intensity depends strongly on the substrate temperature. Fig.7 shows the relation between the PL intensity and the substrate temperature for the green peak in the same reference [9]. As the figure shows, the maximum PL intensity is at $T_s = 200$ °C. They attributed the green PL to oxygen deficiency. This means that much lower temperatures could be used to produce oxygen-deficient ZnO in a reducing atmosphere. Stoichiometry increases with temperature so the green peak becomes smaller with temperature due to the decrease of oxygen deficiency.

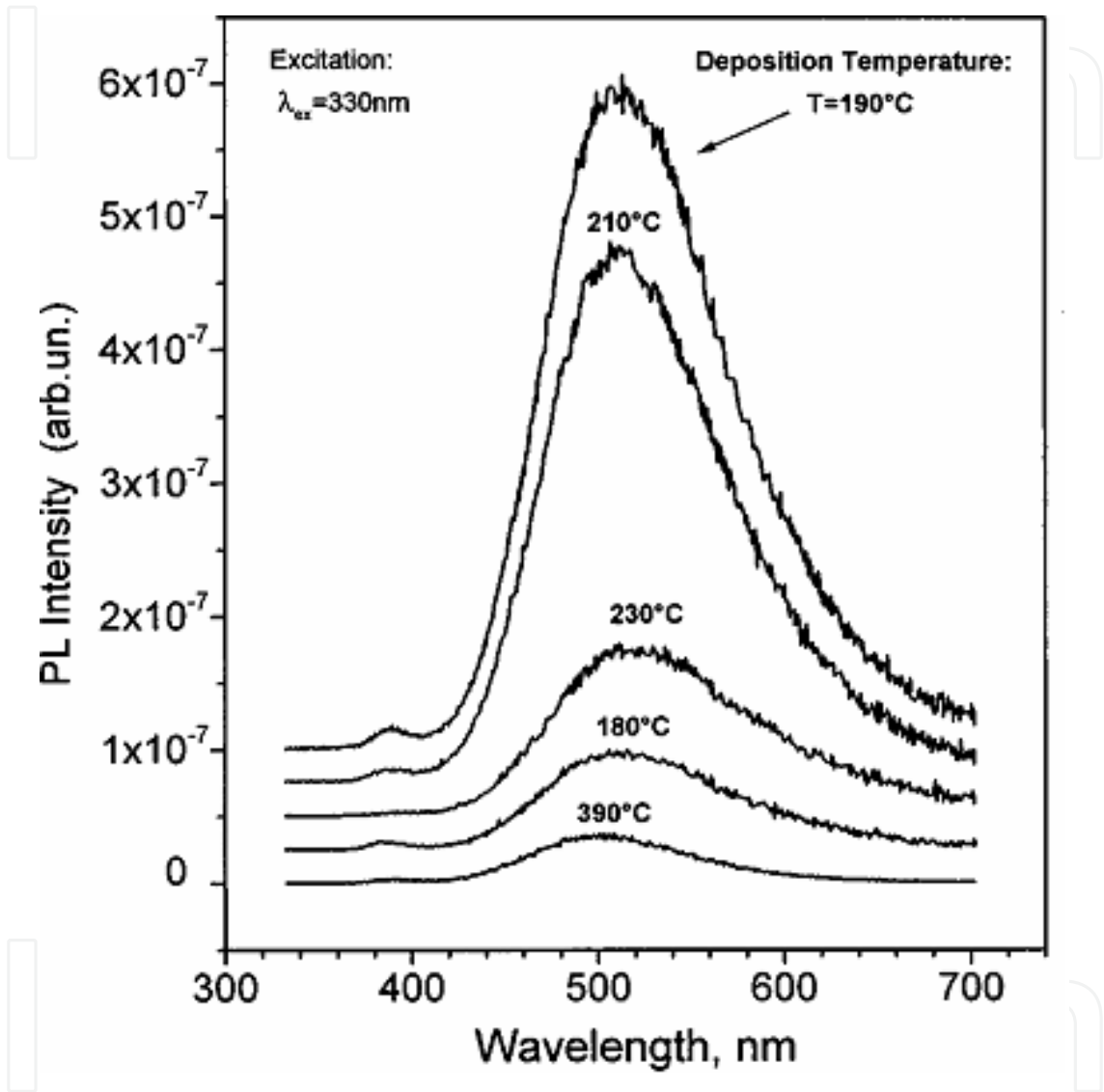


Fig. 5. Photoluminescence spectra of undoped ZnO films grown at different temperatures and annealed in one process in forming gas at 750 °C during 40 minutes. Reprinted with permission from [9], S. A. Studenikin et al. Journal of Applied physics, 84 (4), 2287-2294(1998). Copyright [1998], American Institute of Physics.

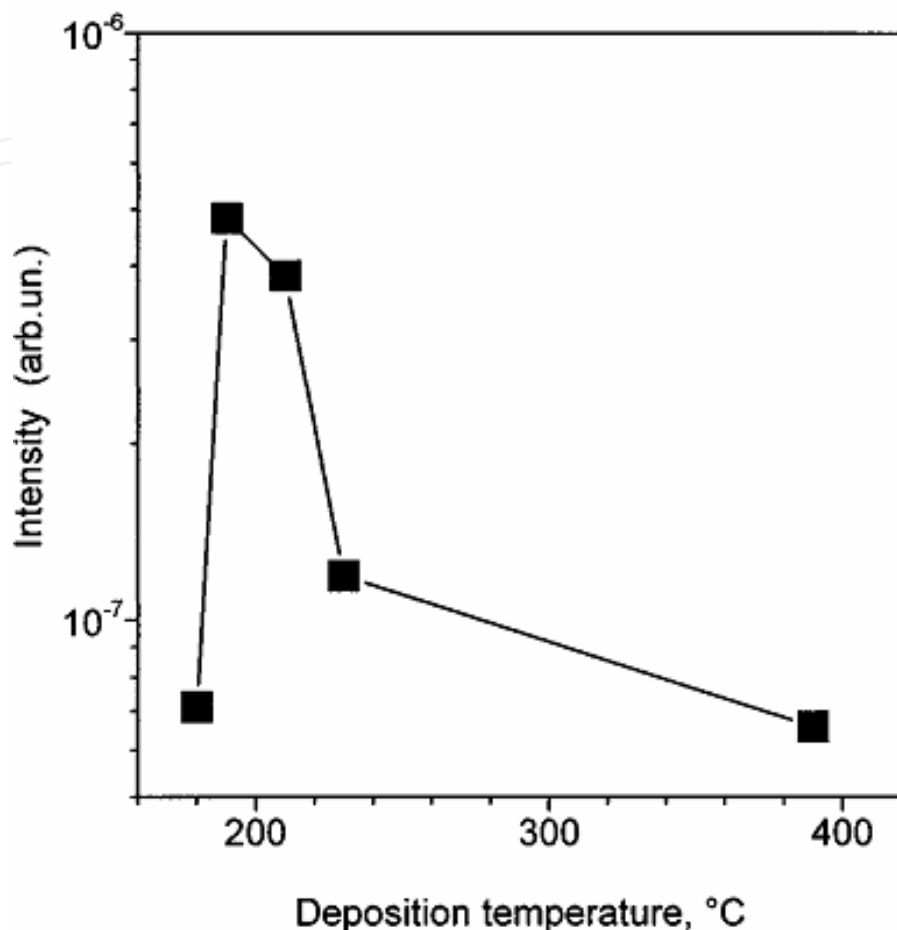


Fig. 6. Intensity of the green photoluminescence of undoped ZnO films as a function of deposition temperature. Reprinted with permission from [9], S. A. Studenikin et al. Journal of Applied physics, 84 (4), 2287-2294(1998). Copyright [1998], American Institute of Physics.

Fig.7 shows the cathodoluminescence spectra for ZnO films prepared by the SP technique at different substrate temperatures taken by El Hichou et. al. [10]. They found that when the substrate temperature increases, the surface of the films is entirely covered by grains and condensed. They observed that extinction of the blue-green emission (centred around 510 nm) is at substrate temperature of 350 and 400 °C, whereas the near UV emission at 382 nm becomes more dominant than other transitions (blue-green and red emissions) at 450 °C. The blue-green emission (510 nm) appears above substrate temperature 450 °C but the red emission (640 nm) appears at different substrate temperature. At $T_s = 500$ °C, the UV transition shifts to higher wavelength and becomes comparable in cathodoluminescence intensity with blue-green emission. The maximum value of cathodoluminescence intensity for three bands is obtained at $T_s = 450$ °C [10].

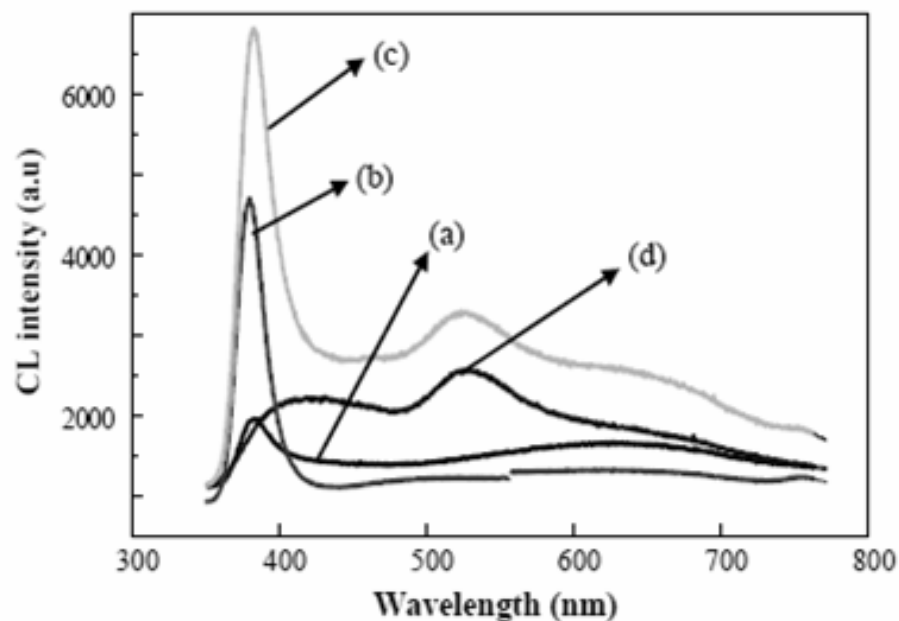


Fig. 7. Cathodoluminescence spectra of ZnO sprayed at flow rate $f = 5$ ml/min at different substrate temperatures: a) $T_s = 350$ °C, b) $T_s = 400$ °C, c) $T_s = 450$ °C, and d) $T_s = 500$ °C. Reprinted with permission from El Hichou et al. [10]; Copyright © 2005, Elsevier.

6. Conclusions

Experimental results show that there are influences of the substrate temperature on the properties of semiconducting thin films which are related to solar cells. Change of state from amorphous to polycrystalline, phase change from cubic to hexagonal, increase in grain size and decrease in the number and width of grain boundaries were observed with the increase of the substrate temperature. Morphological changes such as shape of grains, surface roughness, porosity and density of voids were also observed by different authors. The electrical properties were also found to change due to the changes in the density of charge carriers and density of traps with substrate temperature, beside changes in the structural and morphological properties. The optical properties are also sensitive to these changes, and so the transmittance, optical bandgap, width of Urbach tail, photoluminescence and cathodoluminescence were found to change with changes in the substrate temperatures too.

7. References

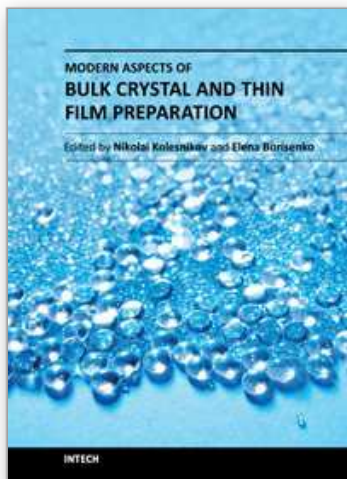
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