
Effect of Pulse Energy on the Optical Properties of ZnS Thin Films Prepared Using Pulse Laser Technique

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Abstract: In these work four samples of ZnS thin films deposited on glass substrate using pulse laser deposition method with different pulse energies, the effect of the laser pulse energy on the optical properties of the four ZnS thin films fabricated was studied. Q-Switched Nd: YAG laser with the fundamental wavelength 1064 nm, laser Pulse energies of (125, 150, 175, and 200) mj with fixed number of pulses of 20, and pulse repetition rate of 5 Hz were used. The target to the substrate distance and angle were kept fixed. The film thicknesses were measured using FESEM measurement tool. The thickness of the deposited ZnS thin films was found to be linearly dependent on the pulse energy used. The transmission spectra in the tested region (532 to 915) nm were found to be in the range from 0.41 to 0.59% depending on the ZnS thin film thickness, and for each ZnS thin film the transmission spectrum is unique. The refractive indices of all samples were determined; and for each sample and it were found to change with wavelength, the highest refractive index of 5.6 at 915 nm was obtained for the sample of the smallest thickness 0.49 microns. Transmission spectra, absorption coefficients and the refractive indices they were in good agreement with the literature.

Keywords: Laser Pulse Energy, Optical Properties, ZnS Thin Films, Pulse Laser Deposition (PLD)

1. Introduction

Zinc Sulphide (ZnS) is a wide gap and direct transition semiconductor that belong to group II-VI semiconductors [1, 2]. ZnS thin films are believed to be one of the most promising materials for blue light emitting diodes, and in electroluminescent displays [3]. As a result, ZnS is an important material used as an antireflection coating in heterojunction solar cells [4]. And in Infrared windows [4]. There are many challenges to produce this material in thin film structure. There exist several methods to produce thin films from this material such as sol-gel, radiofrequency sputtering [5], pulse laser deposition [6], and so on.

Pulse laser deposition (PLD) has been successfully used to deposit an extraordinary wide range [7]. In thin films prepared by PLD technique a variety of deposition parameters

corresponds to the laser properties such as laser pulse energy, pulse repetition rate, etc., in addition to the rest of the other deposition condition such as target – substrate distance and angle, substrate temperature, ambient gas and pressure are all fundamental deposition conditions that determines the properties of the fabricated films [8]. The PLD is widely used technique for the fabrication of thin films because of its numerous advantages such as its simplicity, etc [9]. The PLD technique enables the deposition of many complex materials over a wide range of background gas composition and pressures [8]. E. Ma´rquez et al. in (2014) fabricated ZnS thin films on glass substrate via thermal evaporation technique and studied their structural and optical properties [10]. In (2014) K. R. Murali deposited ZnS films by the pulse plating technique at different duty cycles in the range of 6 – 50% and at a constant current density of 10 mA cm⁻² and he studied their Optical constants (refractive index, n,

extinction co-efficient, k , dielectric constant) of the films in the wavelength range 300-1850 nm by using spectrophotometric measurement [1]. In this work four samples of ZnS thin film were fabricated using Q-switched Nd:YAG pulsed laser deposition with the wavelength of 1064 nm, 10 pulses with pulse repetition rate of 2 Hz, the target to substrate distance and angle were fixed to 2 cm and 45°, respectively. Varied pulse energy of (100,150, 180, 200 and 250) mj were used. The film thickness was measured by Field Emission scanning Electron Microscope (FESEM) measurement tool, and the transmission spectrum at certain wavelengths for each film was recorded. ZnS thin films transmission data and the measured film thicknesses were used to calculate their optical properties.

2. Materials, Tools and Methods

2.1. Materials

The material used in this work were ZnS of 99.9% purity and refractive index of 1.46 it was prepared in disc form by mixing 50:50 ratio of the ZnS powder and Potassium bromide (IR spectroscopy grade). The powder materials were pressed using hand pressing machine to form solid disks as target for ablation.

2.2. Tools and Equipments

Different tools and equipments were used to complete this work. They were described below with their specifications and needs:

2.2.1. Pressing Machine

A hand press machine manufactured by Shimadzu (Japan) was used in this work [11]. The machine is used to press the ZnS powder in disc form (after mixing with Potassium Bromide) for target ablation.

2.2.2. Q- Switched Nd: YAG Laser

Q-switched Nd: YAG model OW D1 was used to deposit

ZnS thin film.

2.2.3. Scanning Electron Microscope (FESEM)

Scanning electron microscopy type field emission model TESCAN MIRA3 was used to measure the films thickness. MIRA3 is a high performance SEM system which gives high resolution and low-noise imaging. MIRA3 offers all the advantages that come with the latest technologies and developments in SEM; delivering faster image acquisition, an ultra-fast scanning system, dynamic and static compensation and built-in scripting for user-defined applications [6, 11].

2.2.4. Different Light Sources

Different monochromatic light sources were used to record the transmission spectra for the fabricated films [12-14].

Table 1. The monochromatic light sources used in this work.

Light Source	Wavelength (nm)
Diode Laser	532
He-Ne Laser	632.8
Diode laser	660
Omega XP Laser (red probe)	675
Omega XP Laser (IR probe)	820
Omega XP Laser (IR probe)	915

2.2.5. The Photodetector

A silicon pin photodiode was used in this work for detecting the transmitted intensity of each light source after passed through the fabricated ZnS films.

2.3. The Method

The procedure to fabricate ZnS thin films and study the effect of the pulse energy on their properties was done as follows:

The experimental setup used to produce ZnS thin films was arranged as shown in figure 1.

Different disks of ZnS (as targets) were prepared by the press machine.

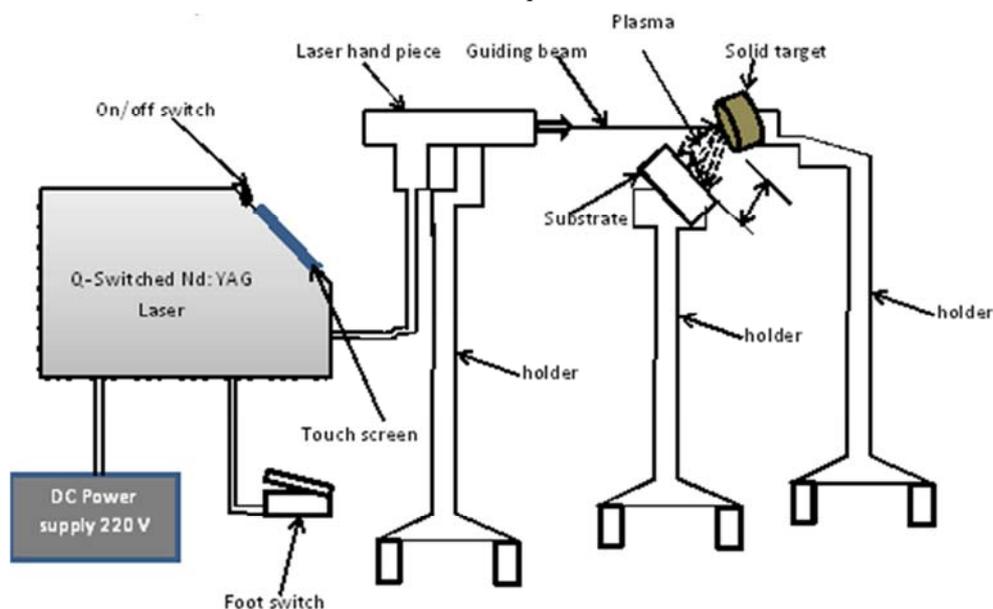


Figure 1. Schematic diagram of the experimental setup for fabrication of ZnS thin films.

The distance and the angle between the target and the glass substrate were fixed to 3 cm, and 45°, respectively.

The glass substrates were cut into the dimensions suitable for SEM imaging 2X2 cm, and then washed with distilled water and cleaned with alcohol.

The Q-Switched Nd: YAG laser machine was switched on and 10 pulses with energy of 100 mj and a R. R. of 2 Hz was used to deposit ZnS thin film on the glass substrate.

The above step was repeated four times with varied pulse energy of (125, 150, 175 and 200) mj with fixed repetition rate and number of pulses.

The fabricated ZnS thin films were examined using FESEM to measure their thicknesses.

The relation between the laser pulse energy and the ZnS thin film thickness was plotted.

The transmission spectra of the fabricated ZnS films were recorded using different monochromatic light sources.

Thicknesses of the ZnS films and the transmission data were used to calculate the optical properties for each film.

The refractive index of each thin film was calculated using the measured reflectivity R and the glass refractive index μ_s according to: [15, 16].

$$\mu = \left(\frac{\mu_s [1 + \sqrt{R}]}{1 - \sqrt{R}} \right)^{\frac{1}{2}} \tag{1}$$

$$\mu_s = \frac{1}{T_s} \left(\frac{1}{T_s^2} - 1 \right)^{\frac{1}{2}}$$

where T_s represents the transmission of the glass substrate.

The absorption coefficients were deduced from the measured value of reflectivity R, the transmittance T, refractive index μ_s , and thickness t according to [15, 16]:

$$\alpha = \frac{1}{t} \mu \frac{(1 - R)^2}{T} \tag{2}$$

3. Results and Discussion

The results of this work are here were composed of two parts; the first part is: Pulse energy deposition parameter of the PLD on the thickness of the fabricated ZnS thin films and second part is: The effect of the pulse energy on the optical properties of the produced ZnS thin films.

3.1. Pulse Energy Effect on the Thickness of the ZnS Thin Films

Four ZnS thin films using 20 pulses with pulse repetition rate of 5 Hz and varying pulse energies of (125, 150, 175, and 200) mj were deposited on glass substrates. Then the four deposited ZnS thin film samples were imaged using FESEM machine and their thicknesses were measured and tabulated in table 2 with the corresponding pulse energy used. Figure 2 shows the morphology of the ZnS thin film deposited using laser pulse energy of 125 mj and figure 3 shows the FESEM thickness measurement of the produced ZnS thin film.

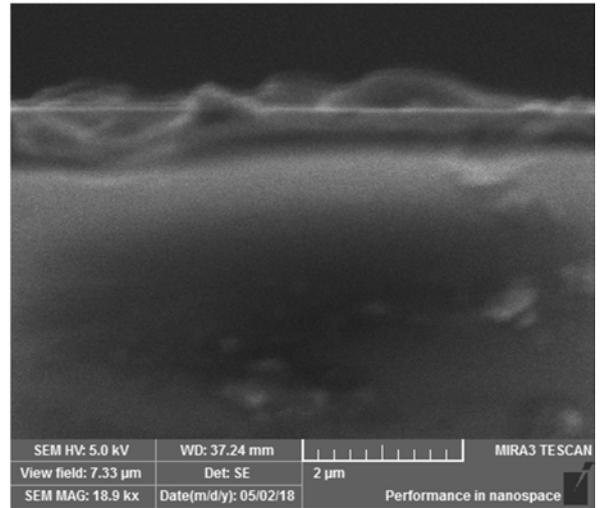


Figure 2. ZnS thin film deposited on glass substrate with laser energy of 125 mj and repetition rate of 5 Hz.

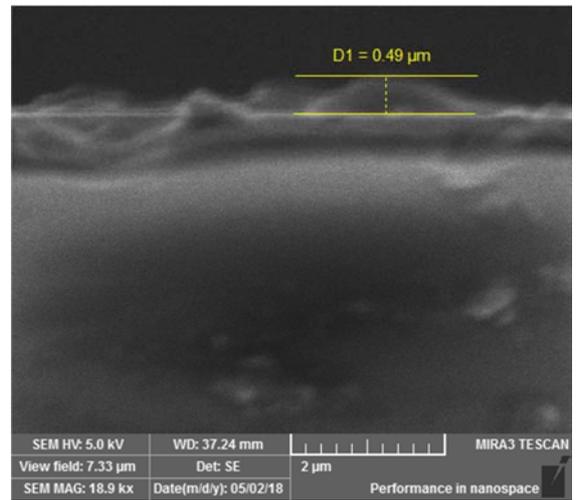


Figure 3. Thickness measurement of the ZnS thin film deposited on glass substrate with laser energy of 125mj and repetition rate of 5 Hz.

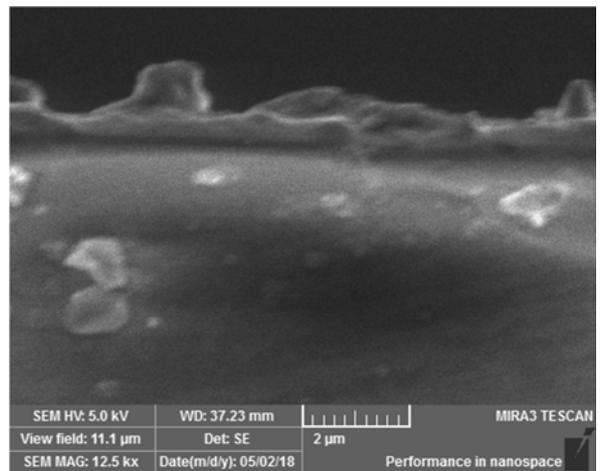


Figure 4. ZnS thin film deposited on glass substrate with laser energy of 200 mj and repetition rate of 5 Hz.

The FESEM image together with the thickness measurement

shown in figures 2 and 3, respectively, illustrate that the ZnS thin film has a thickness of 0.49 μm and it's clear that the fabricated film is dense and has smooth film morphology.

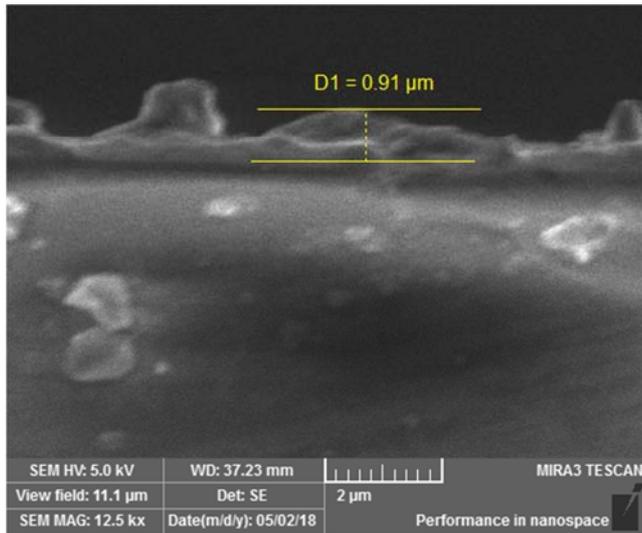


Figure 5. Thickness of ZnS thin film deposited on glass substrate with laser energy of 200 mj and repetition rate of 5 Hz.

Figure 4 shows the ZnS thin film that was achieved laser pulse energy of 200 mj, while the number of pulses, the pulse repetition rate and other deposition parameters were the same. The film thickness that results when the pulse energy was 200 mj is shown in figure 5.

Also figure 4 proved this film is dense and with non-smooth morphology compared to the first film in figure 2 and this is due to the increasing the pulse energy.

Table 2. Thicknesses of the five fabricated ZnS thin films versus laser pulse energy.

Pulse energy in (mj) with R. R = 2 Hz	ZnS thin film thickness in (μm)
125	0.49
150	0.52
175	0.58
200	0.91

The results obtained showed that nanometric ZnS thin films of the order of 100 nanometers are easy fabricated using PLD.

The relation between the ZnS thin film thickness and the pulse energy is plotted together with its fitting in figure 6, and it was showed that the thickness of the ZnS is linearly dependent on the laser pulse energy used for deposition.

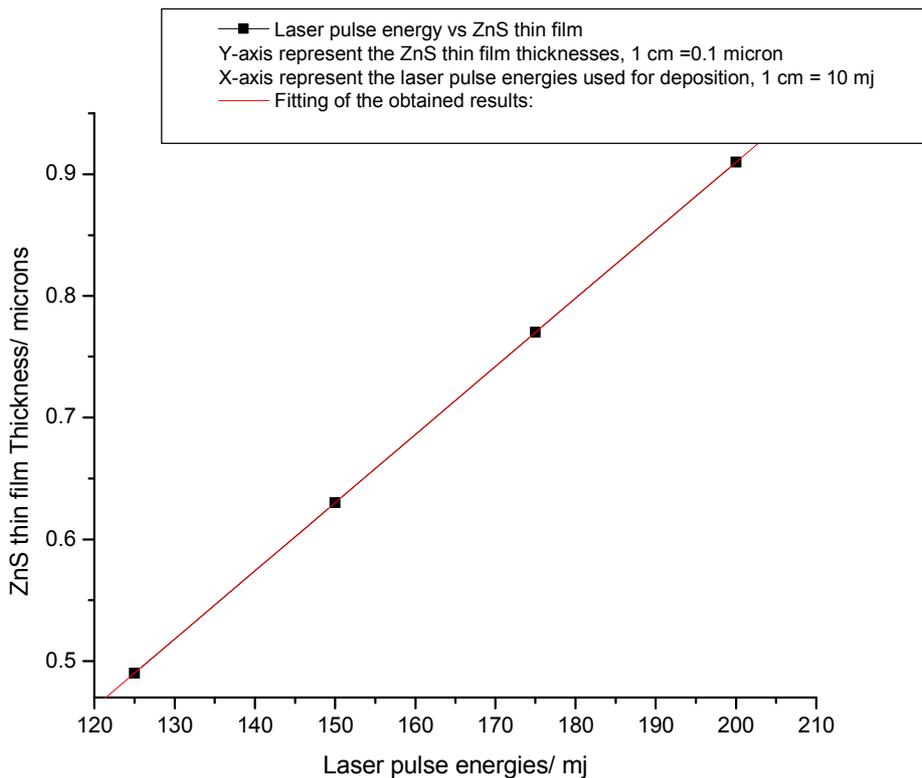


Figure 6. The ZnS thin film thicknesses versus pulse energy used for deposition.

From figure 6 it is clear that increasing the pulse energy results in an increment of the ZnS thin film thickness.

3.2. Optical Properties of ZnS

Then the transmission intensities of different monochromatic light sources were detected before and after deposition for the four ZnS thin film samples, and then the

transmitted intensities were used to calculate the transmission percentage ($T\% = I_t/I_0$) at a certain monochromatic light wavelengths. The calculated values are plotted for the the four ZnS thin film samples S_1, S_2, S_3, S_4 in figure 7 as a function of wavelengths.

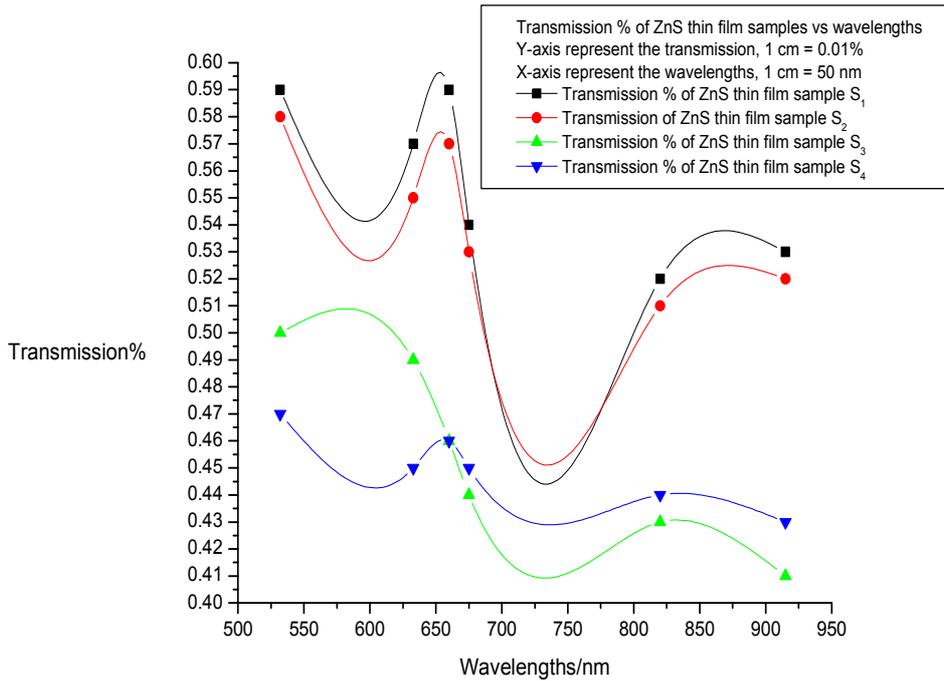


Figure 7. Transmission spectrum of the four ZnS thin film samples.

Figure 7 showed that the thickness of the thin film affected its transmission, comparing between the transmission of the four samples of ZnS thin films, samples S₁, S₂, S₃, and S₄ together with the results shown in figure 6 illustrates that the large thickness of the thin film and thus the higher the pulse energy used for deposition gives the lower transmission of the film.

The transmission spectra of all ZnS samples are in good

agreement with the work of K. R. Murali in the region from 532 nm to 915 nm.

The relation between the calculated refractive indices for the four samples of ZnS thin films deposited on glass substrates using pulsed laser technique with fixed pulse repetition rates and varied pulse energy and were done according to equation (1), and are plotted as a function of wavelengths as shown in figure 8.

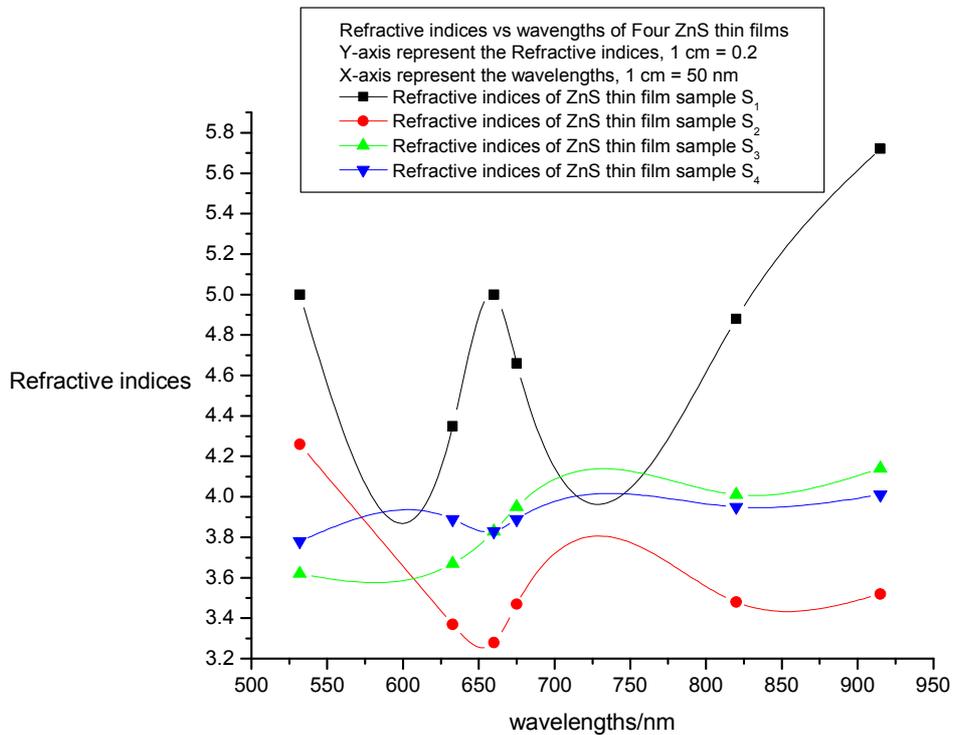


Figure 8. The refractive index of the four samples of ZnS thin films versus wavelengths.

The refractive index of any material in thin film profile is usually deviates from that of the bulk of the same material [15]. This is due to the void fraction typical of the thin film microstructure. Figure 8 showed that ZnS thin film sample S_1 which of smallest thickness among the other has highest refractive indices from 532 to 915 nm this result suggest using ZnS thin films in applications that require high index materials, and also the results in figure 8 proved that

increasing the thickness of the thin film the refractive index decrease and when the thickness of the thin films is in the range of 1 micron or above the refractive index of the film deviates infinitesimally from that of the bulk material.

The absorption coefficients calculated for the four samples using equation (2) and were plotted versus wavelengths in figure 9 below:

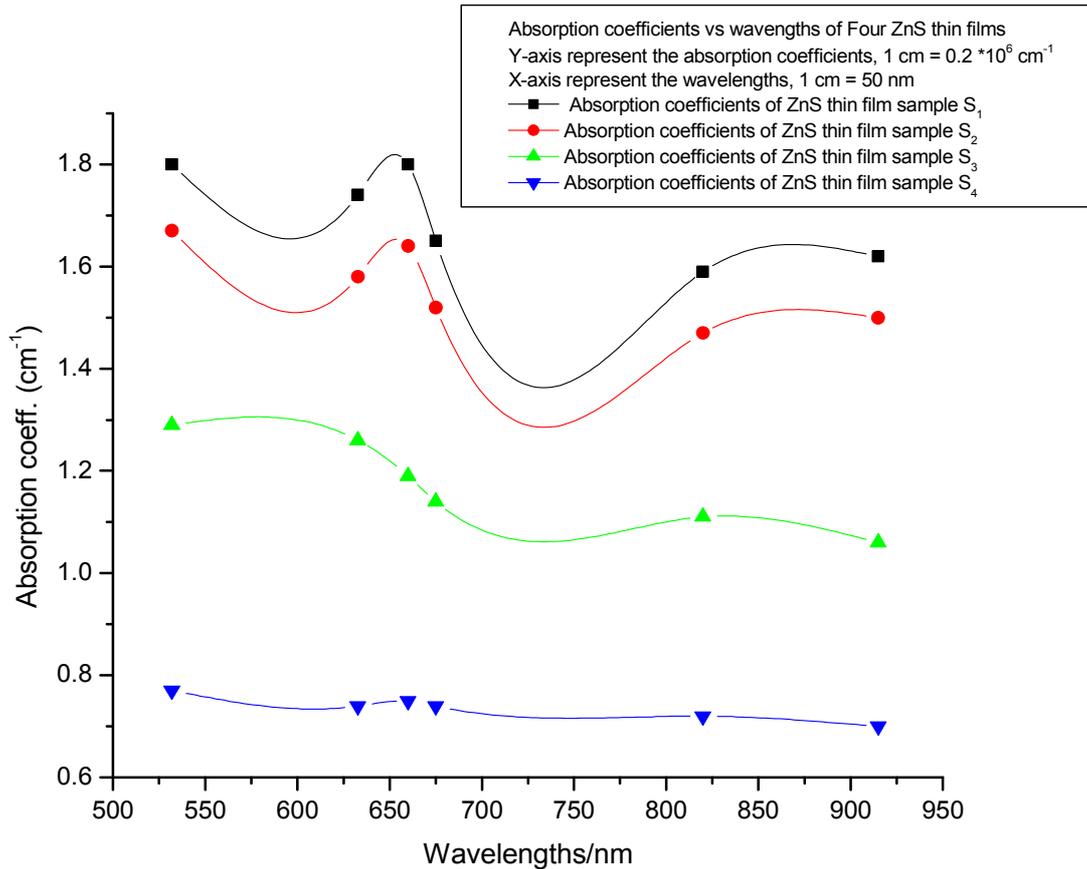


Figure 9. Absorption coefficients versus wavelengths for four samples of ZnS thin films.

Figures 8 and 9 support the idea of using such film as an optical filter or as a reflector in specific wavelengths deduced from the transmission spectrum, since the four samples has different absorption coefficients, the absorption coefficients of the ZnS thin films for all samples showed in figure 9 exhibits similar phenomenon with the wavelengths. The refractive indices and absorption coefficients of the pulsed laser deposited ZnS thin films varies with thickness and for each thickness the transmission is unique, therefore this two optical properties are functions in the film thickness and therefore on the repetition rate used for the deposition of such films. Optical measurement constitutes the most important means of determining the band structure of the materials. And the optical constants of thin films provide us with information concerning microscopic characteristics of the material and its determination is very important for using it in any of such devices [6, 8].

4. Conclusions

In this study, the effect of laser pulse energy on the thickness and optical properties of ZnS thin films deposited on glass substrate by pulse laser deposition method has been investigated. The ZnS thin deposited films thicknesses were measured using FESEM measurement tool. Transmission spectra of the deposited ZnS thin films at certain wavelengths were recorded. The measured ZnS thin film thicknesses and the transmission data were used to deduce the refractive indices and the absorption coefficients of each thin film. The transmission, refractive indices and absorption coefficients values obtained are in good agreement with the literature. In conclusion ZnS thin films deposited by pulse laser deposition method can be used to produce optical components in the range from visible to IR regions by controlling its thickness via controlling the laser pulse energy, the results also showed

that the ZnS thin films are suitable for applications requiring high refractive index material.

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