

Effect of SiO₂ Mixing on Structural Properties of ZnO Thin Films Prepared by Pulsed Laser Deposition (PLD) Technique

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Abstract

In this paper ,ZnO was mixed by various concentrations (5,10,15,20,25) w% SiO₂.The mixture was deposited on glass substrate by laser pulse deposition at room temperature. X-ray results showed that there was a decrease in the peaks of zinc oxide with gradual peaks of the zinc silicate compound at angles 2 θ (25,38,48,66) due to the increase in silicon oxide concentrations rates. The infrared spectrometer Fourier transform was used to study all prepared films and gave good results about the chemical bonds of the composite and their locations and correspond to the standard results of the zinc silicate compound . The results of the Atomic Force Microscope showed that all of prepared films were having Nano scale size and there was an increase in square root roughness index.

Keywords: pulsed laser deposition, structural properties, concentrations, chemical bonds, Nano scale, ZnO, SiO₂.

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تأثير خليط اوكسيد السليكون على الخواص التركيبية لأغشية اوكسيد الخارصين المحضرة بتقنية الترسيب بالليزر النبضي

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المخلص

في هذا البحث تم خلط اوكسيد الخارصين بتراكيز [5, 10, 15, 20, 25) w%] من اوكسيد السيليكون وتم ترسيب الخليط على ارضيات زجاجية بتقنية الليزر النبضي عند درجة حرارة الغرفة. اظهرت نتائج فحوصات الاشعة السينية ان هناك انخفاض في قمم اوكسيد الخارصين وزيادة في ظهور قمم سليكات الخارصين عند الزوايا (25°, 38°, 48°, 66°) وذلك بسبب زيادة نسبة التشويب بأوكسيد السيليكون. تم استخدام مطياف الاشعة تحت الحمراء لدراسة الاغشية المحضرة وأعطت نتائج جيدة حول الاواصر الكيميائية للمركب ومواقعها وهي تتوافق مع النتائج القياسية لمركب سيليكات الزنك. أظهرت نتائج مجهر القوة الذرية أن جميع الاغشية المحضرة كانت ذات حجم نانوي وقد وجد زيادة في معدل الجذر التربيعي وخشونة السطح.

الكلمات الدالة: الترسيب بالليزر النبضي، الخواص التركيبية، تراكيز، الاواصر الكيميائية، حجم نانوي، اوكسيد الخارصين، اوكسيد السيليكون.

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1. Introduction:

Thin film technology is one of the most important technologies that contributed to the development of the semiconductor study and its properties. Thin films are used in many different fields. They are used in the manufacturing of various components of microelectronic devices, reagents, magnetic storage equipment, inverter filters, inverter and inverter coatings. They are also used in the manufacture of micro circuits, electrical circuits for electronic microscopes and in the manufacture of capacitors and rectifiers. Because of their small size and light weight, they are used in digital computers [1]. Zn_2SiO_4 (willemite) is a long known material which still retains its place among the best inorganic phosphors [2]. Having different crystal phases and being sensitive to doping by transition metals and rare earths, it can emit light at different wavelengths in the visible and near IR range. In the past years there were successful attempts to synthesize nano-phase of Zn_2SiO_4 using both solid state techniques [3, 4] and wet chemistry aimed at the development of novel low voltage phosphors having high efficiency and chemical stability. One of the advanced methods for solid state synthesis of nanoparticles is ion implantation with subsequent annealing, which allows creation of nanoparticle-host matrix composites with high chemical stability. Many research groups have studied the ZnO nanostructure formation in SiO_2 matrices using ion beam synthesis and thermal oxidation [5].

2. Experiment part:

Zinc oxide powder was mixed with silicon oxide powder according to (5, 10, 15, 20, 25)% wt. respectively. The proportions of the powders were weighed using a sensitive electronic balance Sensitive 10^{-4} . The powders were then mixed into mixing machine Type of Spex mixer for (5) minutes and then the powders were blended by hydraulic press. And sintering the tablets in a tube electric furnace at a temperature of (1000°C) for two hours and after the process of sintering left the tablets to cool. The process of deposition was accomplished within a vacuum chamber in the laser system under pressure 10^{-3} torr and the laser energy was 1000 mJ, which is the appropriate energy for deposition. Nd: YAG laser pulse used at (1000 mJ) and frequency (6Hz) for prepared thin film.

3. Results and discussion:

A. X- ray diffraction :

X- ray diffraction used to characterize obtained films crystalline. The (d, h k l) spacing between crystal planes can be calculated using Bragg's law [6].

$$n\lambda = 2d\sin\theta \quad (1)$$

Where (θ) is diffraction angle and (λ) is the used XRD wavelength. Debay Scherrer equation, formula used to calculate crystalline size utilize the peaks broadening [7].

$$D = K\lambda / B \cos\theta \quad (2)$$

Where(λ) is the x-ray wavelength for $k\alpha$ transition from Cu target (1.5406 Å), B is full width at half maximum and (θ) is the angle of diffraction. Before the process of annealing, we observe from Fig. 1 that there are peaks showing that the original composite (ZnO) is clearly present with the presence of some unclear peaks of the silicon oxide and after annealing in Fig. 2, The peaks of the zinc silicate complex begin to appear with the reduction of the zinc oxide peaks by increasing the doping ratio with the increasing temperature of the annealing which reveals that crystallization of the membranes is improving, as it can be seen that the height of the peaks increases with the increase of the doping ratio indicating an increase in the crystallization rate. It is also possible to observe that the width of the peaks decreases with an increase in the percentage of doping, which indicates an increase in particle size, where the particle size is inversely proportional to the width of the top, this corresponds to the results in the literature [8-9] .

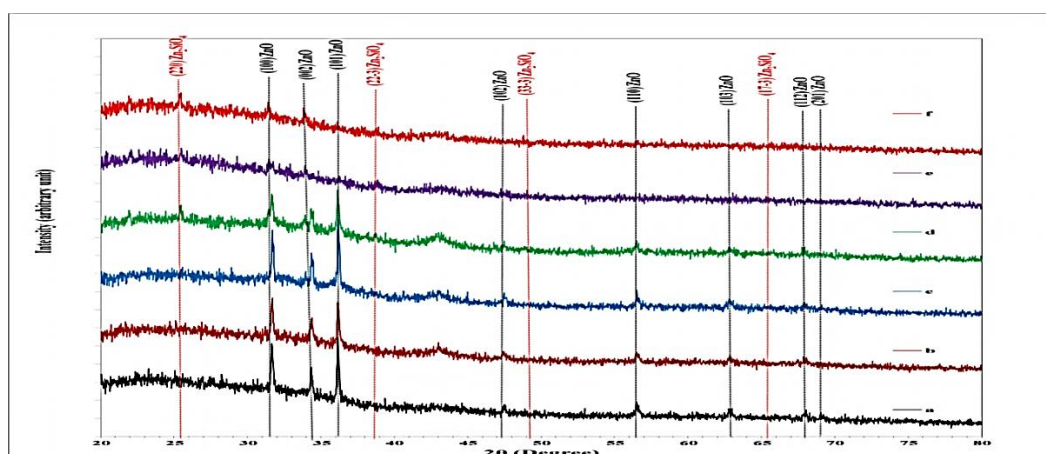


Fig. 1: XRD for ZnO with different ratio of SiO₂ before annealing

(a) Pure ZnO, (b) 5%, (c)10%, (d) 15%, (e) 20% and (f) 25%

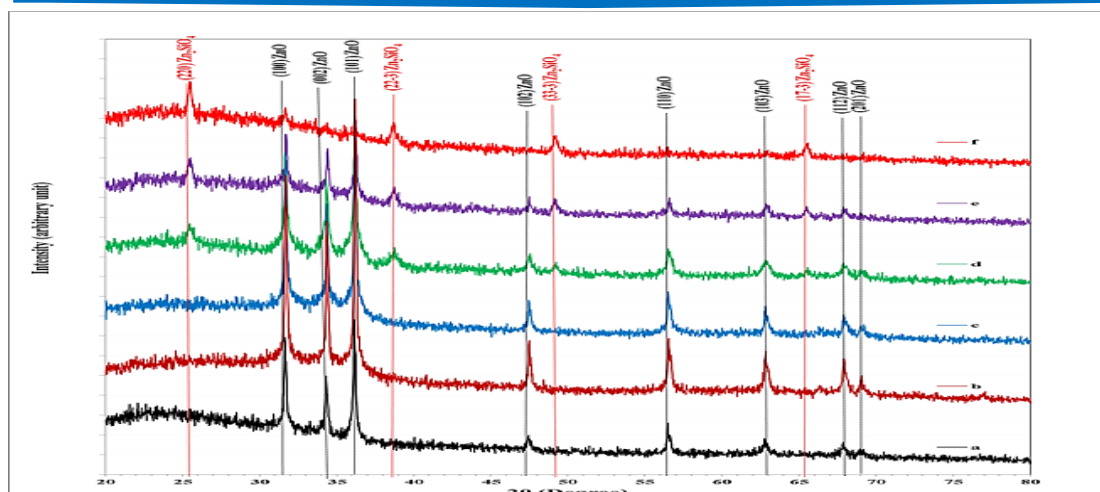


Fig. 2: XRD for ZnO with different ratio of SiO₂ after annealing

(a) Pure ZnO, (b) 5%, (c) 10%, (d) 15%, (e) 20% and (f) 25 % peaks due to the formation of Zn₂SiO₄ compound become clear for concentrations large than 15%

B. Fourier Transform Infrared Spectrometer FTIR:

Is a technique which is used to obtain an infrared spectrum of absorption or emission of a solid, liquid or gas. An FTIR spectrometer simultaneously collects high spectral resolution data over a wide spectral range. This confers a significant advantage over a dispersive spectrometer, which measures intensity over a narrow range of wavelengths at a time. The term Fourier transform infrared spectroscopy originates from the fact that a Fourier transform (a mathematical process) is required to convert the raw data into the actual spectrum [10]. The FTIR spectroscopy was employed at room temperature in the range of (400 – 4000) cm⁻¹ as shown in Fig. 3 and Table 1 where the sample inhibits the presence of composite symmetric stretching in (~880 cm⁻¹), composite stretching in (~1100cm⁻¹), Composite bending stretching in (~510 cm⁻¹). Similar study was reported by [11-12].

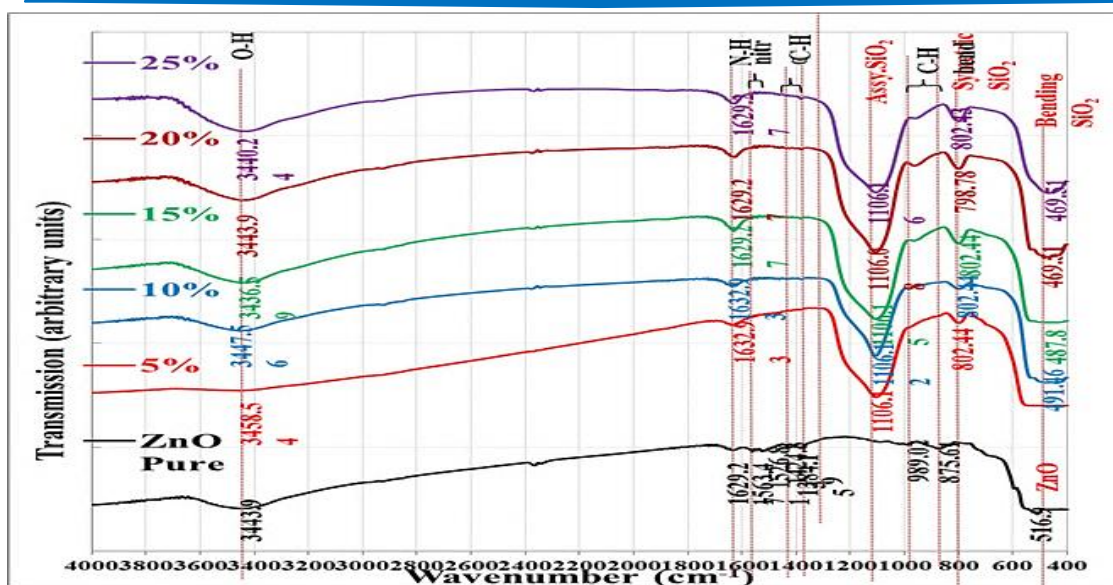


Fig. 3: FTIR for ZnO with different ratio of SiO₂ after annealing
(a) Pure ZnO, (b) 5%, (c) 10%, (d) 15%, (e) 20% and (f) 25 %

Table 1: FTIR for ZnO with different ratio of SiO₂ after annealing
(a)Pure ZnO, (b)5%, (c)10%,(d) 15%,(e) 20% and (f)25 %.

	ZnO pure	5%	10%	15%	20%	25%
SiO ₂ bending	-	-	491.46	487.80	469.51	469.51
ZnO	516.90	-	-	-	-	-
SiO ₂ Symmetric vibration	-	802.44	802.44	802.44	798.78	802.44
C-H bending	875.61	-	-	-	-	-
	989.02	-	-	-	-	-
SiO ₂ asymmetric vibration	-	1106.10	1106.12	1106.15	1106.08	1106.16
C-H	1384.15	-	-	-	-	-
	1424.39	-	-	-	-	-
Nito	1526.83	-	-	-	-	-
	1563.41	-	-	-	-	-
N-H stretch	1629.27	1632.93	1632.93	1629.27	1629.27	1629.27
O-H	3443.90	3458.54	3447.56	3436.59	3443.90	3440.24

C. Atomic Force Microscope AFM :

In this study, the use of an atomic force microscope type ((Scanning probe microscope type AA3000 equipped from (Angstrom Advanced Company), it is characterized by the ability of the high amount of analysis (0.1 - 1.0nm), high zoom power and the possibility of

runs under normal atmospheric pressure without the need to unload high as it is in electronic microscopes. this machine gives analytical images of two-dimensional (2D) and three-dimensional (3D) to the surfaces of (ZnO-SiO₂) films. Its provide us some calculations on the form of grain size, rate of surface roughness and root mean square by computer program prepared from manufacturer [13]. From Figs. (4-5) and Table 2, We note that the increase in the ratio of mixing and the temperature of the annealing lead to an increase in particle size and decrease the surface roughness and mean square root value due to good crystalline uniformity and high surface homogeneity of the particles composed of the films material. As for the fifth and sixth samples, the granular size decreases and the surface roughness And the average square root value due to the large diameter of the zinc oxide atom relative to the radius of the silicon oxide atom before annealing and after annealing, this corresponds to the results in the literature [14-15].

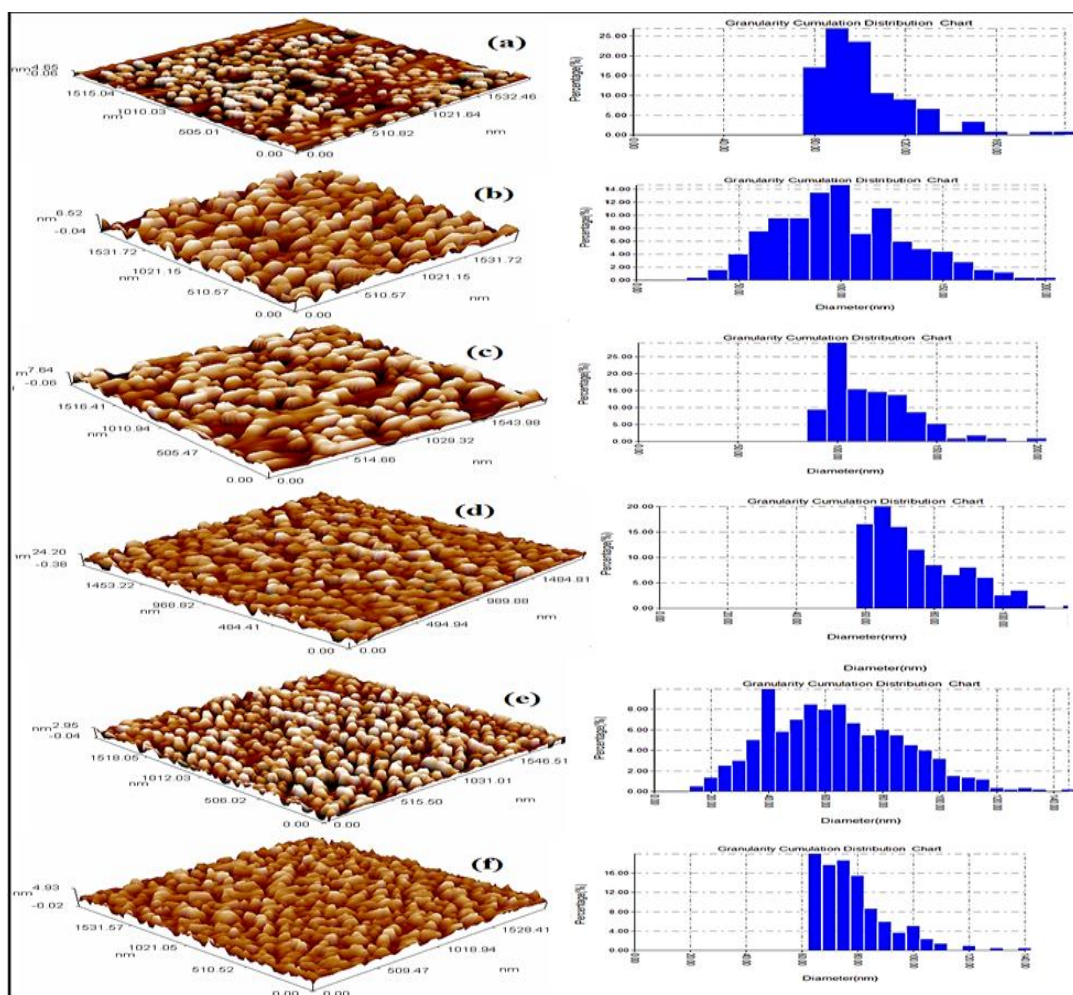


Fig. 4: AFM for ZnO with different ratio of SiO₂ Before annealing

(a) Pure ZnO, (b) 5%, (c) 10%, (d) 15%, (e) 20% and (f) 25 %

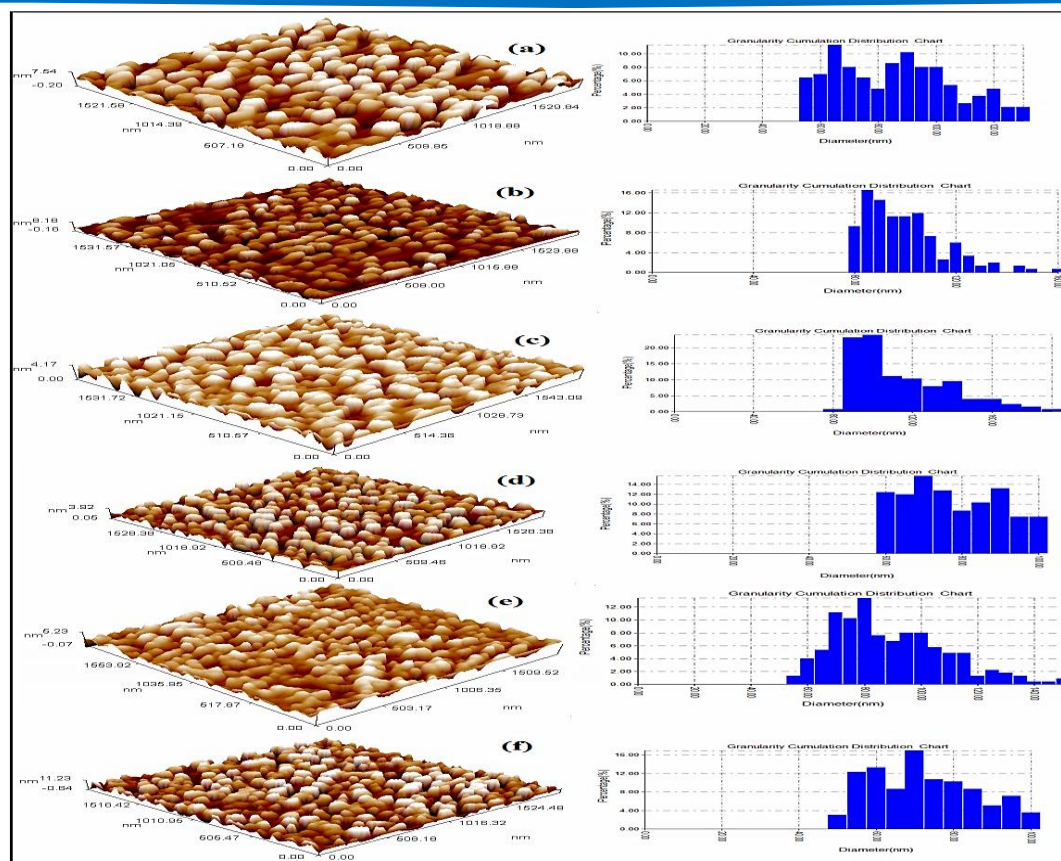


Fig. 5: AFM ZnO with different ratio of SiO₂ after annealing

(a) Pure ZnO, (b) 5%, (c)10%, (d) 15%, (e) 20% and (f) 25 %

Table 2: AFM for ZnO with different ratio of SiO₂

(a)Pure ZnO, (b)5%, (c)10%,(d) 15%,(e) 20% and (f)25 %

Annealing	SiO ₂ %	Average Diameter (nm)	Roughness Ave. (nm)	RMS roughness (nm)
Before	0	97.82	1.17	1.35
	5	95.97	1.27	1.5
	10	112.18	1.79	2.08
	15	72.61	3.69	4.59
	20	76.18	0.609	0.727
	25	61.04	0.747	0.862
After	0	83.44	1.69	1.98
	5	97.33	1.58	1.87
	10	109.73	0.681	0.833
	15	109.73	0.936	1.08
	20	86.84	0.807	0.995
	25	69.79	3	3.24

4. Conculution:

The appearance of a hexagonal structure through the compound used, as well as a clear reduction in particle size with increasing concentrations. Results showed (FTIR) there are several types of vibration in the compound silicate zinc. The particle size, square root average and coarse coefficient decrease with increasing mixing ratio.

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