Irradiation Effects on The Sensitivity of ZnO Thin Films Synthesized on Glass Substrate by Sol-gel Method

Yasir Yahya Kasim, Ghazwan Ghazi Ali* , Marwan Hafeedh Younus
Department of Physics, College of Education for Pure Science, Mosul University, Mosul, Iraq

Abstract

This work investigates the structural, optical, and surface properties of ZnO thin films prepared by sol-gel method. The effect on waveguide sensor was examined at different irradiation durations of alpha particles. The X-ray diffraction (XRD) measurements revealed that the crystalline phase of ZnO thin films does not change after irradiation and showed a hexagonal structure of wurtzite type with an orientation toward (002). Moreover, ZnO thin films absorbance was increased with increasing irradiation time, whereas the transmittance was decreased. Additionally, increasing the irradiation time of alpha particles caused an increase in the extinction coefficient and the imaginary part, while the optical energy gap of the ZnO samples was decreased. Finally, the maximum value of sensitivity was 42%, found at 6 min of irradiation duration.

Keywords: Alpha particles, ZnO thin films, Sol-gel.

تأثير التشعيع على استجابة الاغشية الرقيقة ZnO المرسبة على ارضية زجاجية بطريقة Sol-gel

ياسر يحيى قاسم[1], غزوان غازي علي*[2] , مروان حفيظ يونس فصولة[3]
قسم الفيزياء ، كلية التربية للعلوم الصرفة ، جامعة الموصل، الموصل ، العراق

الخلاصة

تم في هذا العمل دراسة الخصائص التركيبية, البصرية والسطحية للأغشية ZnO المحضرة بطريقة Sol-gel. تم فحص تأثير موجه الموجه لجسيمات الفا عند ازمنة تشعيع مختلفة. بينت قياسات الاشعة السينية X-ray بان طور التبلور للأغشية ZnO لا تتغير بعد التشعيع وتمتلك تركيبا سداسيا نوع wutizte وذات اتجاهية (002) . علاوة على ذلك , ازدادت امتصاصية اغشية ZnO مع زيادة زمن التشعيع بينما قلت النفاذية اضافة الى ذلك زيادة زمن التشعيع لجسيمات الفا تقود الى زيادة معامل الخمود وكذلك الجزء الخيالي كما تقل طاقة الفجوة البصرية للعينات ZnO . اخيرا تم ايجاد اقصى قيمة للحساسية حوالي 24.8% عند زمن تشعيع 6. min

الكلمات المفتاحية: جسيمات الفا , اوكسيد الزنك , Sol-gel

Introduction

Zinc oxide (ZnO) thin films have been extensively studied due to the high optical and electrical properties, which can be employed in different applications, such as solar cells and sensors [1,2]. Moreover, ZnO has attracted considerable attention as a promising candidate for inexpensive, light weighted, and light-absorbing mediums [3,4]. ZnO thin films can be deposited in several methods such as the evaporation method [5,6], metal organic chemical vapor deposition (MOCVD) [7],

---

*Email: dr.ghazwan39@gmail.com

sputtering [8] and spray pyrolysis [9]. Furthermore, ZnO thin films have been used as sensing materials of reactions of chemicals and gases [10]. The researchers attempted to improve the sensitivity by different methods, such as the alloying, doping, annealing, and irradiation, which increase the sensitivity of ZnO for detecting [11,12]. On the other hand, the irradiation technique has been recently used to enhance the properties of ZnO thin films [13,14,15]. The interaction between the radiation and matter, especially alpha particles, changes the optical and electrical characteristics of the materials [16,17,18]. Alpha irradiation plays a key role to enhance the sensitivity of the ZnO thin film sensor, deposited on glass substrate by sol-gel method, to detect glucose solution.

Experimental part

Sol-gel method has attracted the attention of many researchers and used in the deposition of various types of oxides due to essential for the growth along the preferred orientation and give uniform surface. The ZnO thin films were prepared using sol-gel method. Zinc hydroxide acetate (Zn (CH3COO)$_2$HO with a weight of 0.2195 g was dissolved in a mixture of 10 ml ethanol with and 90 ml deionized water in room temperature. The mixture was kept under constant stirring for 20 min using magnetic stirrer to completely dissolve. To enhance the homogeneity, 0.5 g polyvinylpyrrolidone (PVP) was added to the mixture which was then kept under constant stirring for 24 hours. Afterwards, the glass substrates were washed with distilled water and methanol and cleaned with ultrasonic waves, respectively. Finally, the ZnO thin films were deposited on 320nm-thick glass substrates and exposed to alpha particles emitted from Am-241 source with energy of 3MeV to increase its sensitivity during the glucose detection process. The energy of alpha particles was calculated based on the following equation:

$$E_{\alpha} = E_o \left[1 - \frac{x}{R}\right]^{2/3} \qquad \text{............. (1)}$$

where x represents the distance between sample and irradiated source, E_o is the maximum energy of alpha particles at x= 0 (equal to 5.485 MeV), and R is the range of alpha particles in the space and can be obtained by SRIM program [19].

Setup of glucose sensing test

Figure-1 illustrates the experimental setup of the glucose sensing test applied in the present study . The laser source emitted at 650nm wavelength and a high sensitive avalanche photo detector (EPPT-2000) with a range of 600nm to 1200nm was used. The waveguide, coated with different ZnO thin films, was used as a sensor and placed between the source and the detector of glucose solution. The irradiated waveguide sensor coated with ZnO thin films was exposed to different concentration of glucose solution. The light emitted from the laser source was passing through the surface of the irradiated waveguide during glucose detection. Then, the absorption of the power of light detected by the photo detector was increased, leading finally to increase the sensitivity of the sensors.
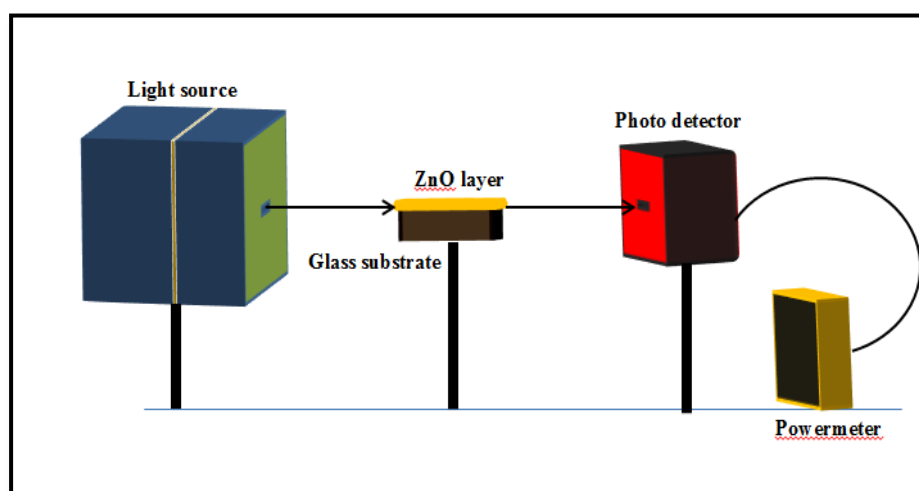

Figure 1-Sensing experimental setup

Results and Discussion
X-ray diffraction (XRD) measurement
The XRD patterns of ZnO films, with different thickness, deposited on the glass substrate before and after alpha irradiation are presented in Figure- 2. Alpha particles of 3MeV emitted from Am-241 source were irradiated on the ZnO at different time periods of 2, 4 and 6min. It can be seen from Figure- 2 that the patterns of XRD indicate a hexagonal structure and well C-axis orientation after alpha irradiation [20]. Moreover, the XRD analysis shows a strong peak of ZnO (002) at 2θ=34.24, while the other peaks are found toward the plane orientations (100), (101), (102) and (110) at 2θ =31.73^o, 2θ =36.23^o, 2θ =48.34^o and 2θ =57.32^o respectively. Furthermore, the XRD results confirmed that the peaks intensity increases with increasing irradiation time, which could be due to the breaking of bonds on the surface of ZnO thin film. The XRD analysis also revealed that the irradiated ZnO films were still in the crystalline phase and in good homogeneity. The grain size of the deposited ZnO thin films can be calculated using Scherrer equation 1 [21] :

$$D = \frac{K\lambda}{\beta cos\theta}$$ ………… …….. (2)

where D is the grain size, K is a constant which is equal to 0.89, λ is the wavelength of XRD (Cu K_α) which is 1. 54178A^o, and β is the full width at half maximum. It can be observed that the grain size increases with the increase in irradiation time, due to the defect of clusters of samples that were formed after irradiation, as well as the interface in the localization of states on the ZnO surface, as shown in Table-1.
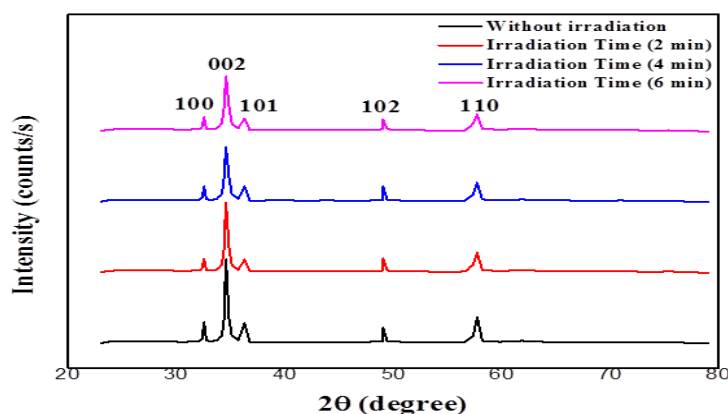

Figure 2-XRD patterns of ZnO thin films before and after irradiation at time periods of 2min, 4min , and 6min.

Table 1- ZnO grain size and FWHM as a function of irradiation time

Irradiation time (min)	Grain Size (nm)	FWHM (radian)
0	18.34	0.52
2	22.67	0.45
4	28.65	0.43
6	32.22	0.39

Optical properties
The optical properties before and after irradiation of ZnO thin films were studied. Figure- 3 shows the absorption spectra as a function of the wavelength. The results indicate that the absorbance of ZnO thin films was increased for all irradiation times. This might be due to the breaking of bonds on the ZnO surface after alpha irradiation, which leads to increase the absorbance mechanisms. Increasing of irradiation time leads to formation defects. These defects are considered as an interesting factor in

photonic devices as they play important roles in the optical properties of a material by introducing absorption bands or color centers [22]. The ZnO thin films were still in crystalline phase and did not change in crystal structure after irradiation . Furthermore, the diffraction peak of the ZnO thin film was oriented along the C-axis (002) after irradiation. Figure- 4 shows the results of transmittance against wavelength of the samples. The transmittance of ZnO thin films was decreased for all irradiation times in comparison with the non-irradiated ZnO samples. This is related to defects on the rough surface of ZnO after alpha irradiation, leading to a noticeable decrease in the transmittance [23].

Figure-5 illustrates the absorption coefficient of non-irradiated and irradiated ZnO thin films as functions of wavelength. The absorption coefficient increases as the irradiation time increases for all the samples. This could be due to the change in the crystal structure of ZnO after irradiation [24]. Moreover, all the irradiated ZnO samples exhibited high magnitudes of the absorption coefficient, which could lead to the direct transition band gap of ZnO. These results are consistent with those reported by Muhammad [25]. Figure- 6 shows the relationship between $(\alpha h\upsilon)^2$ and photon energy $(h\upsilon)$ of non-irradiated and irradiated ZnO thin films . The energy gap (E_g) can be calculated via Tauc formula:

$$\alpha h\upsilon = B(h\upsilon - Eg)^r \qquad\qquad\qquad ....(3)$$

where B is constant, α is the absorption coefficient, $h\upsilon$ is photon energy, and r takes the value of 1/2 at direct transition and 2 at indirect transition. The results show that the optical energy gap decreases with irradiation time, with values of 3.26, 3.21, 3.16 , 3.1 eV for irradiation times of 2, 4 and 6 min, respectively. This behavior can be justified by generating extra energy states after irradiation as well as interface band transition at localized states in the energy gap. The values of calculated energy gap are in agreement with those reported by Khan [26]. The results of optical properties after irradiation are attributed to the defects in ZnO thin films. Most of these defects are created by the localization of oxygen vacancies and change of electronic structure [27]. Furthermore, ionized radiation through thin films causes the loss of most of the energy, due to non-elastic collision of radiation with electrons on ZnO surface or losing of energy due to elastic collision with atomic nucleation of material [28].
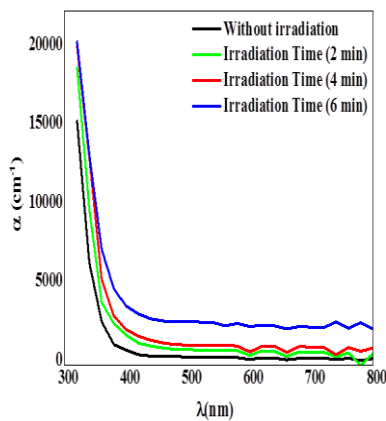

Figure 5-Absorption coefficient against wavelength for different irradiation times.
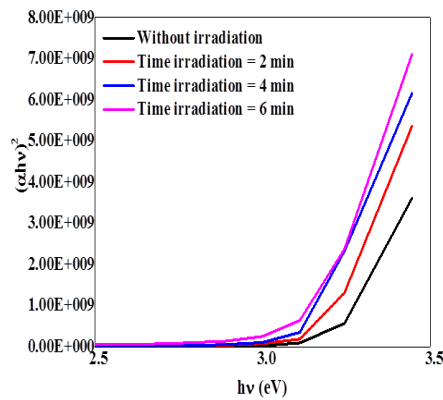
Figure 6-The $(\alpha h\upsilon)^2$ against $h\upsilon$ for different irradiation times

In addition, the extinction coefficient of ZnO thin films, which includes both the real and imaginary parts of dielectric constant, was studied in the wavelengths range of 300-800 nm. The extinction coefficient (k_o) , real part (ε_r), and imaginary part (ε_i) was decreased with increasing wavelength for all samples. The values of extinction coefficient and imaginary part of irradiated samples were larger than those of the non-irradiated samples . Moreover, the values of the real part were smaller than those before irradiation, as shown Figure-9 [29, 30].
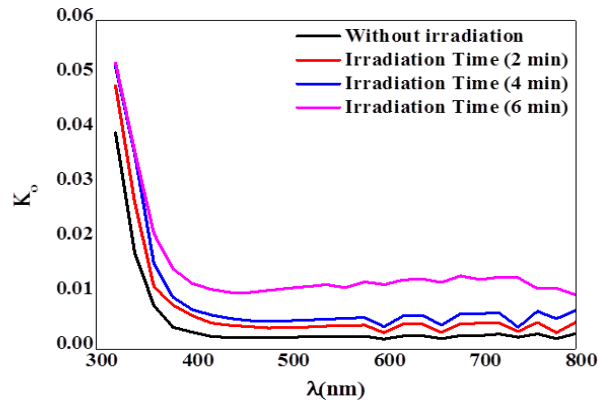
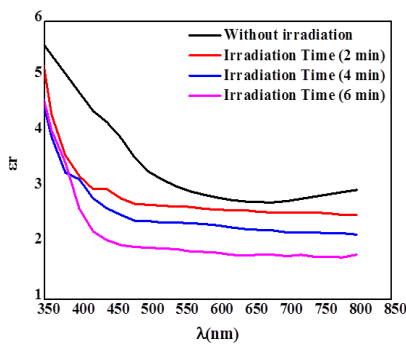**Figure 7-** Extinction coefficient versus hυ for different irradiation times

Figure 8- Real part ε_r versus hυ for different irradiation times.
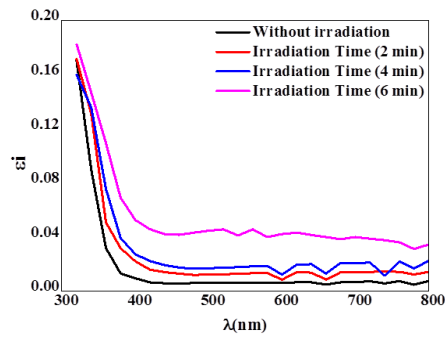

Figure 9- Imaginary part ε_i versus hυ for different irradiation times

Scanning Electron Microscopy (SEM)

Figure-10 shows the SEM images of non-irradiated and irradiated ZnO thin films for different times (2, 4, and 6 min). The images reveal that the grain size evidently increases when the irradiation time is increased [31]. This could be due to the defects formed on the ZnO surface caused by alpha particles, which lead to the formation of large-sized nuclei, especially at the longer irradiation time. Therefore, this can open the possibility for manipulating the crystallinity, shape, and size of the grains via controlling irradiation time [32].
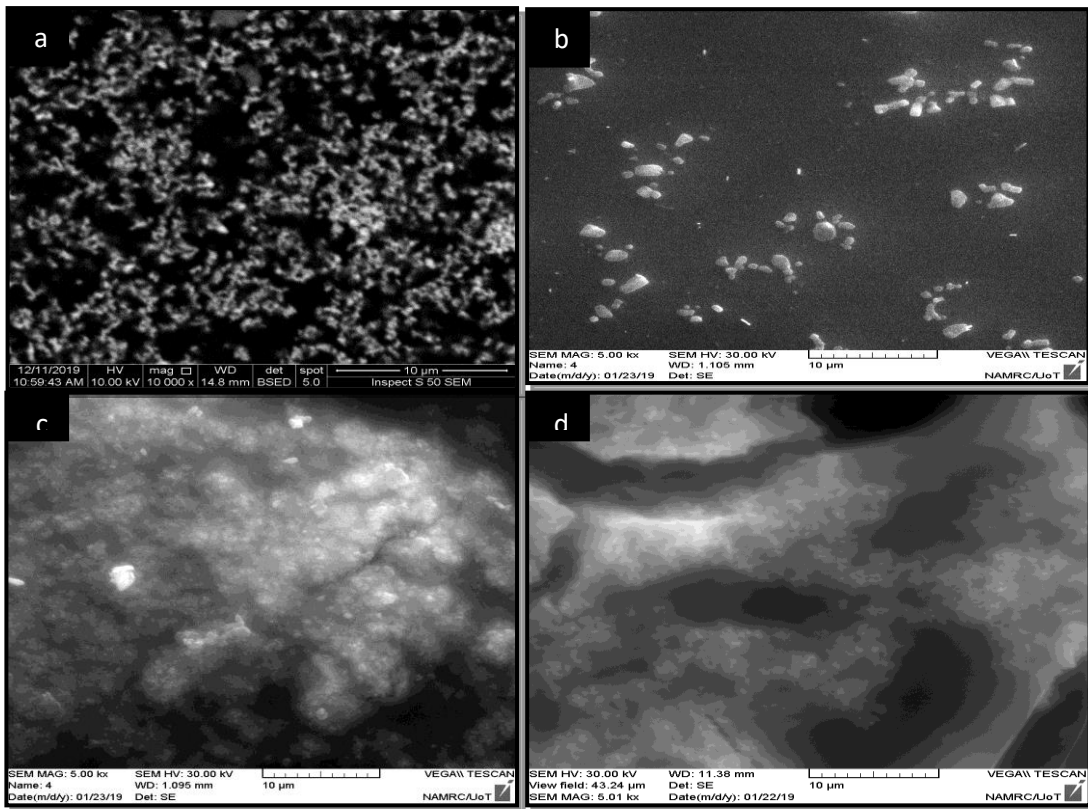
Figure 10- SEM images of ZnO thin films before and after irradiation time (a) before irradiation, (b) irradiation time 2min, (c) irradiation time 4min, and (d) irradiation time 6min.

Waveguide sensor for glucose detection

The prepared samples were used in the detection of various concentrations of glucose solution. Figure- 11 shows the normalized power of the light passing through the ZnO surface versus glucose concentration, before and after irradiation. As can be observed, the normalize power was increased as the time irradiation increased during the detection processes. This could be due to defects, such as those in color centers, on the ZnO surface after irradiation which lead to increase the absorbance process . Additionally, the response of ZnO to glucose solution, for all irradiation times, was found to be linear. The gradients of the lines were employed to give the sensitivity of ZnO. The results showed that the sensitivity of the ZnO for glucose detection increases as the irradiation time is increased. The maximum sensitivity of the ZnO is approximately 42% at 6 min irradiation time, whereas the minimum sensitivity is approximately 25 % and 14 % at 4 and 2 min, respectively. Figure- 12 shows the linear fitting of the ZnO sensitivity with various ZnO thicknesses. The R-square value for the linear line fitting is 0.98221, which indicates a good linearity. The proposed ZnO sensor can be implemented for glucose detection due to the good line fitting characteristics.
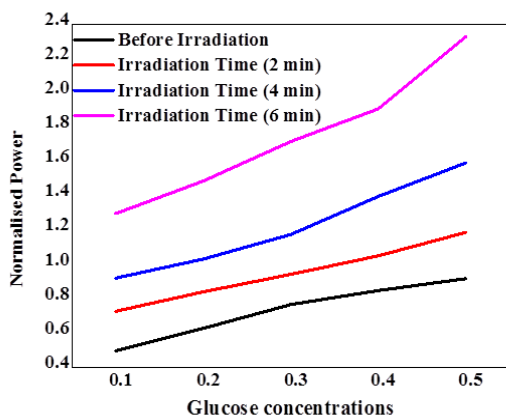

Figure 11-Normalized power versus glucose concentration of ZnO after Alpha irradiation
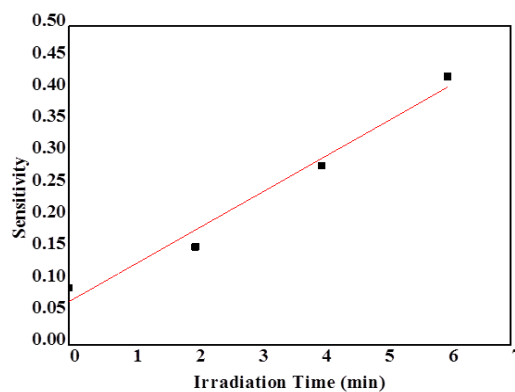
Figure 12-Sensitivity percentage versus irradiation time of ZnO waveguide sensors

Conclusions

Waveguide sensor was designed and fabricated by depositing ZnO thin films on glass substrates using sol-gel method and then irradiated by alpha particles. Structural, optical, and morphological properties of the ZnO thin films were examined and analyzed. The X-ray diffraction patterns revealed a slight shift, while the crystalline size increases, with increasing irradiation time. Moreover, the band gap slightly decreases after irradiation. Furthermore, the waveguide sensor showed an improved sensitivity to detect glucose solution with increasing irradiation time. The higher value of sensitivity was 42% dB/con at irradiation time of 6min, while the value before irradiation was found to be 9.4% dB/con. The irradiation process increased the absorbance of the sensor, which in turn increased the sensitivity of waveguide sensor. This may be used to monitor liquid leakages in different fields, such as fuel stations, wells, and oil pipes.

References
1. Khan M., Rabia Q. , Norah A. and Hayat S. **2017**. Characterizations of multilayer ZnO thin films deposited by sol-gel spin coating technique, *Results in Physics*, **7**: 651-655.
2. Mursal A., Irhamni J. , Bukhari K. and Zulkarnain J. **2018**. Structural and Optical Properties of Zinc Oxide (ZnO) based Thin Films Deposited by Sol-Gel Spin Coating Method. *J. Phys.: Conf.* Ser.1116
3. Nel J., Ahmed, M., Ahmed M. and Meyer W. **2019**. Processing of and electrical properties of ZnO thin films and nanorods for sensor applications, Proc. SPIE. 11043, Fifth Conference on Sensors
4. Ashraf R., Riaz S. and Naseem S. **2013**. Synthesis and characterization of ZnO nanoparticles ". *Advance in nano*, Seoul Korea .**4**(2): 213-219
5. Chávez-Urbiola, I. R., et al. **2019**. Development and characterization of photodiode n-ZnO/p-Si by Radio Frecuency Sputtering, a sensor with low voltage operation and its response to visible and UV light. *Thin Solid Films*, **669**: 364-370.
6. Ghazwan Gh. Ali, Abdul Kahliq A. Sulaiman, and Ivan B.Karomi . 2018. The Effect of the ZnO Thickness Layer on the Porous Silicon Properties Deposited by Chemical Vapor Deposition , AIP Conference Proceedings,**2034**: 1-7
7. Konstantaki, Maria, et al. **2012**. "An ethanol vapor detection probe based on a ZnO nanorod coated optical fiber long period grating. *Optics express,* **20**(8): 8472-8484
8. Hassan N. and Hashim M. **2012**. Catalyst-free growth of ZnO nanowires on Si (100) substrates: Effect of Substrate Position on Morphological, Structural and Optical Properties". *ECS Journal Solid State Sciences Technology*, **17**(1): 86-89 .
9. Shaveta Th.,Sharma J.,and Sanjay K .**2017**. Effect of Silver Doping on Properties of ZnO nanoparticles by chemical Precipitation method". *International Journal of Engineering Technology Science and Research,* **4**(12): 842-846.
10. Rabia Q. **2017**. Effect of gamma radiation on morphological & optical properties of ZnO nanopowder", *Results in Physics*, **7**: 807-809.
11. Najiba A. , Ramla D. , Saba J. **2014**. Effect of Gamma Irradiation on the Structural and Optical Properties of ZnO Thin Films", *Journal of Computer Engineering*, **16**(1): 11-16 .
12. Sahani R. M., Kumari C., Pandya A., and Dixit A. **2019**. Efcient Alpha Radiation Detector using Low Temperature Hydrothermally Grown ZnO:Ga Nanorod Scintillator", *Scientific Reports*, **3**(7): 1-9.
13. Chaudhary P. and Kumar V. **2019**. Preparation of ZnO thin film using sol-gel dip- coating technique and their characterization for optoelectronic applications , *WSN*, **121**: 64-71.
14. Nida K. **2019**. Investigation of Optical and Morphological Properties of Co Doped ZnO Nanomaterials", *Turkish Journal of Science and Technology*, **14**(1): 41-48.
15. Ghazwan Gh . Ali, Abdul Kahliq A.Sulamian, and Marwan Hafeedh Younus. **2019**. Study of Porous Silicon Behavior via Neutron Irradiation: Fabrication and Characterization , *Results in Physics*, **14**: 1-7.
16. Kasim U., Aisha Sh. and Alhaji Y. **2016**. Optical and Structural characterization of low dose gamma irradiated Zinc Oxide (ZnO) thin films prepared by Electrostatic spray pyrolysis , IOSR *Journal of Applied Physics*, **8**(1): 1-8.

17. Jamal R. K., Ali F. H., Hameed M. M., Aadim K. A. **2020**. Designing A Zener Diode Using Ag₂O(1-X)ZnO(X)/PSi Structures Deposited by Laser Induced Plasma Technique. *Iraqi Journal of Science*, **61**(5): 1032-1039.

18. Safa K. Mustafa, Raied K. Jamal. **2019**. Studying the Effect of Annealing on Optical and Structure Properties of ZnO Nanostructure Prepared by Laser Induced Plasma. *Iraqi Journal of Science*. **60** (10): 2168

19. Ziegler J., Biersack J., Littmark U. **2003**. "*The stopping and Range of Ions in Solids*","SRIM", Pergamon Press, Oxford.

20. Bushra K . **2015**. Effect of Gamma Ray Irradiation on Structural and Optical Properties of ZnO Thin Films, *Ibn Al-Haitham J. for Pure & Appl. Sci*, **28**(3): 7-17.

21. Ahmed M. , Taghizadeh F., Auret F. **2019**. The effect of alpha particle irradiation on electrical properties and defects of ZnO thin films prepared by sol-gel spin coating , *Materials Science in Semiconductor Processing*, **101**: 82-86.

22. Nagarani N. **2013**. Structural and optical characterization of ZnO thin films by Sol-Gel method. *Journal on Photonics and Spintronics*, **2**(2): 2324-8572.

23. Znaidi T.,Touam T., and Vrel D. **2012**. ZnO thin Films Synthesized by Sol-Gel Process for Photonic Applications. *ACTA Physics Polonica A*, **121**(1): 165-173.

24. ABDALLAH B., ISMAIL A. **2019**. Optical and structural study of low dose alpha irradiated zinc oxide (ZnO) thin film , *Journal of Optoelectronics and Advanced Materials*, **21**(5): 401-406.

25. Muhammad R. , Mukhlasur R. , Farhad J. **2019**. Structural, optical and photocatalysis properties of sol–gel deposited Al-doped ZnO thin films , *Surfaces and Interfaces* ,**16**: 120-126.

26. Khan Z., Khan M., Zulfequar M. , Khan M. **2011**. Optical and structural properties of ZnO thin films Fabricated by sol–gel method. Mater Sci. Appl. , **2**(5): 340-349.

27. Hussain H . , Basil A. , Huda S. **2013**. Nuclear radiation effect on ZnO performance, *Journal of Basra Researches* , **39**(3): 72-81.

28. Sanaz A., Majid J., Morteza S. **2019**. Preparation and characterization of gallium-doped zinc oxide/ polystyrene nanocomposite scintillator for alpha particles detection, *Applied Physics A* , **125**(6): 450-462.

29. Sahani R., Chandni K., Arun P., Ambesh D. **2019**. Efficient Alpha Radiation Detector using Low Temperature Hydrothermally Grown ZnO: Ga Nanorod Scintillator , *Scientific Reports*, **9**(1) **:**1-9.

30. Shweikani R., Ismail A., and Jerby B. **2017**. Preliminary study on the properties of Zinc Oxide(ZnO) for alpha particles detection, *International Journal of Radiation Research*, **15**(3): 301-306.

31. Rodica P. , Anca I. , Raluca G. **2015**. Study of point defects in ZnO thin films irradiated with alpha particles , International Conference on Defects in Semiconductors, Espoo, Finland.

32. Shweikani, R., Ismail , A., and Jerby, B., **2017**. Preliminary Study on the properties of Zinc Oxide (ZnO) for alpha particles detection , *International Journal of Radiation Research*,**15**(3): 301-306.