

Optical Investigation of In₂O₃/ Quartz Nano-films Deposited at Different Laser Energies

Dua'a R. T. Alrayyes^a, Ali A. Alwahiba^a, Makram A. Fakhri^{*a}, Motahher A. Qaeed^b, and Subash C.B. Gopinath^{c,d,e}

^aLaser and Optoelectronic Department, University of technology-Iraq, Baghdad, Iraq.

^bPhysics Department, Faculty of Science, University of Jeddah, Jeddah, Saudi Arabia.

^cInstitute of Nano Electronic Engineering, Universiti Malaysia Perlis (UniMAP), 01000 Kangar, Perlis, Malaysia.

^dFaculty of Chemical Engineering & Technology, Universiti Malaysia Perlis (UniMAP), 02600 Arau, Perlis, Malaysia.

^eMicro System Technology, Centre of Excellence (CoE), Universiti Malaysia Perlis (UniMAP), Pauh Campus, 02600 Arau, Perlis, Malaysia.

*Corresponding author. E-mail: mokaram_76@yahoo.com, & makram.a.fakhri@uotechnology.edu.iq

ABSTRACT

The study conducted in this research focuses on the deposition of In₂O₃ thin films onto quartz substrates using the pulsed laser deposition technique (PLD). The importance of this study lies in the exploration of the optical properties of these thin films under varying laser energies (1200 mJ, 1400mJ, 1600mJ, 1800 mJ, and 2000 mJ), and the subsequent determination of their energy band gaps. In the pursuit of enhancing our understanding of In₂O₃ thin films, this research exhibits a direct relationship between the laser energy and laser band gap. It was discovered that the energy band gap of thin In₂O₃ films increased due to the increase in laser energy. This phenomenon resulted in an overall blue shift from the fundamental energy gap of In₂O₃ thin films. The significance of this study extends to potential applications in various fields. In₂O₃ thin films with tunable energy band gaps can find applications in optoelectronics, photovoltaic and other emerging technologies. This research not only contributes to the fundamental understandings of thin film properties but also offers a pathway for tailoring their characteristics for specific applications, thus advancing the field of materials science and engineering.

Keywords: Deposition, Indium Oxide, Nano-films, Optical characterization, Quartz

1. INTRODUCTION

Indium Oxide (In₂O₃) is a well-known n-type, yellow-colored powder and a transparent semiconductor with a wide band gap of 3.75 eV, making it an attractive material for a range of optoelectronic applications such as solar cells, detectors, sensors, and light-emitting diodes due to its several advantages high electrical conductivity, high transparency, essential limitations, such as a modest maximum sensitivity, extremely high operating temperatures (200°C - 600°C), stable and durable material, high surface area-to-volume ratio and excellent biocompatibility [1-4].

In₂O₃ exhibits different crystalline structures, or phases, depending on its synthesis and processing conditions [5]. The most commonly observed phases are the Cubic (bixbyite) phase: which is the stable phase of In₂O₃ at high temperatures [6]. It belongs to the cubic crystal system with a space group of Ia-3, and it has a relatively high symmetry [7]. Monoclinic phase: At lower temperatures, In₂O₃ can undergo a phase transition and transform into a monoclinic structure [8]. The choice of synthesis method and processing conditions can be used to control the crystalline structure of In₂O₃ and tailor its properties for specific applications [9]. In₂O₃ can be easily synthesized using various methods, including sol-gel, hydrothermal, and co-precipitation method [10, 11]. The material characteristics are heavily influenced by synthesis techniques corresponding to the process parameters of deposition [12,

13] there are many techniques and procedures for producing a thin film [14, 15]: chemical spray pyrolysis (CSP), chemical vapor deposition (CVD), sputtering technique, sol-gel method, thermal pyrolysis deposition (TPD), thermal evaporation in vacuum deposition. (TEVD) and pulse laser deposition (PLD) [16-22]. Overall, PLD using Nd:YAG lasers and other laser sources offers an efficient and precise method for the preparation of indium oxide thin films with tunable properties, which can be utilized in various technological applications such as the transparent conductive films, solar cells, gas sensors, photodetectors, transparent electrodes, semiconductor devices, catalysis, optical coatings and the emerging technologies. In₂O₃ has been extensively used in biosensors for the detection of various biomolecules, including glucose, cholesterol, and DNA. In₂O₃-based biosensors have been reported to exhibit excellent sensitivity, selectivity, and stability, making them ideal for use in clinical and diagnostic applications [23-28]. In₂O₃ has gained significant attention in sensing applications, particularly in gas sensors and biosensors, due to its advantageous properties. Recent studies have highlighted the unique features of In₂O₃ and its potential for use in various sensing platforms. In gas sensing applications, In₂O₃ has demonstrated exceptional performance as a sensing material. Its high surface area and good conductivity make it highly sensitive to target gases. In addition, In₂O₃ exhibits excellent selectivity, stability, and response time, making it suitable for the detection of a wide range of gases. In₂O₃ have made significant contributions to the field by developing methods to tune the energy band

gaps of In_2O_3 . The key contributions and developments in the area of doping techniques, alloying, nano structuring, strain engineering, surface functionalization, hybrid materials, thin film deposition techniques, computational modeling and optical engineering.

The research on In_2O_3 thin films with tunable energy band gaps is a cornerstone in the development of materials for optoelectronics, photovoltaics, and emerging technologies. The ability to customize these films' properties is a powerful tool for enhancing the efficiency and performance of a wide range of devices and technologies, contributing to energy savings, cost-effectiveness, and sustainability.

2. EXPERIMENTAL WORK

Figure 1 illustrates the whole experimental work.

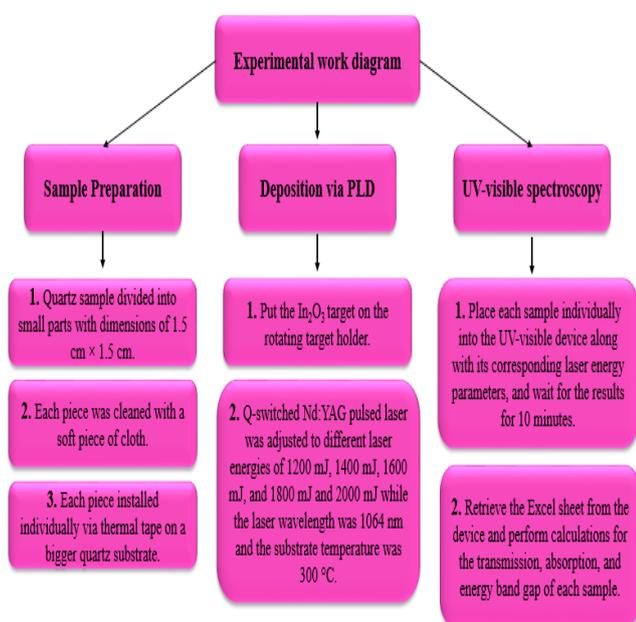


Figure 1. Experimental work diagram.

2.1 Quartz Sample Preparation

The quartz samples were divided into small parts with dimensions of approximately $1.5 \text{ cm} \times 1.5 \text{ cm}$ and it was cleaned with a soft piece of cloth (the same that was used to clean the sunglasses). Each piece was installed individually via thermal tape on a bigger quartz substrate. The quartz material was picked out because of its high bearing temperatures without being broken or damaged; it also allows a wide range of wavelengths compared to the other materials. Figure 2. Showed the piece of quartz that was used.

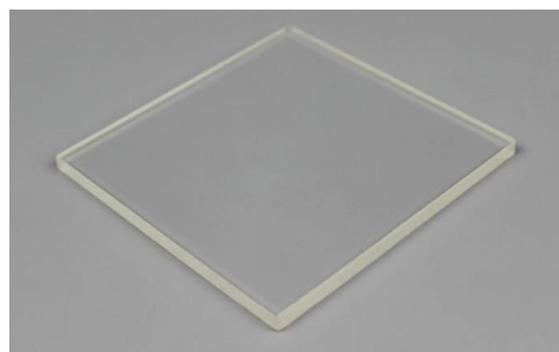


Figure 2. Quartz sample.

2.2 Pulsed Laser Deposition (PLD)

PLD is a powerful technique for depositing In_2O_3 thin films on quartz substrates. It offers excellent control over stoichiometry, high film quality, uniformity, and the ability to tailor film properties, making it well-suited for various optoelectronics and photovoltaic applications where the quality and performance of the thin films are crucial. The In_2O_3 target was bought readily from the nano laboratory at the University of Technology with a diameter of 2 cm and a thickness of 1 cm. Q-switched Nd:YAG pulsed laser was adjusted to different laser energies of 1200 mJ, 1400 mJ, 1600 mJ, and 1800 mJ and 2000 mJ while the laser wavelength was 1064 nm and the substrate temperature was $300 \text{ }^\circ\text{C}$. Figure 3 illustrates the setup for the PLD technique. The parameters and values that utilized in this technique were discussed in table 1.

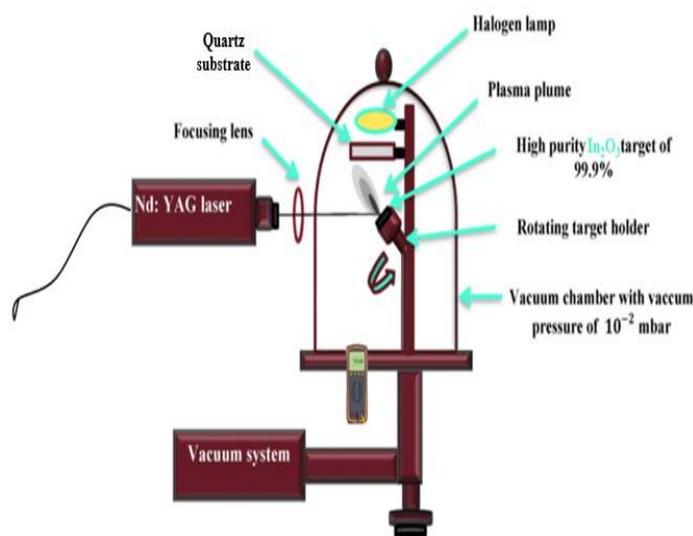


Figure 3. The pulsed laser deposition setup (PLD).

Table 1 The parameters used in the PLD technique

Laser parameters	Values
Laser Wavelength	1064 nm
Pulse Duration	7 ns
Pulse Energy	1200 mJ 1400 mJ 1600 mJ 1800 mJ 2000 mJ
Substrate Temperature	300°C
Frequency	3 Hz
Repetition Rate	300 pulses
Spot Size	2 mm
Focal length of lens	12 cm
Power Supply	220 V
Substrate	quartz

2.3 UV-Visible Spectroscopy

This test was used on quartz samples at the University of Technology laboratory to investigate the optical absorption, transmission, and energy gap of the In₂O₃ deposited over quartz by PLD. UV-Visible spectrophotometer (Metertech SP8001, Taiwan) was used to investigate the transmittance (T%) spectra. A blank quartz slide was put in the first section as a reference, and the film was placed in the second section. The spectral range was chosen to be within 200-1000 nm. The absorbance (A_b) was calculated from (T%) as follows [29-32]:

$$A_b = \log_{10} * 1 / (T\% * 10^{-2}) \quad (1)$$

The energy band gap (E_g) was estimated from the Tauc plot (hν) versus (αhν)^{0.5}, where (h) is the plank's constant (4.1356x10⁻¹⁵ evs). The UV-Vis beam frequency (ν) and the absorption coefficient (α) were calculated from [33-38]

$$\nu = C/\lambda \quad (2)$$

$$\alpha = (2.303A_b)/t \quad (3)$$

Where (C) is the speed of light (3x10⁸ m/s) [39-41].

3. RESULT AND DISCUSSION

Figure 4 (a,b,c) illustrated the transmission, absorption and the energy gap of the grown In₂O₃ thin film over the quartz substrate at different laser energies ranging from 1200 mJ - 2000 mJ [42-44]. As the laser energy increase the transmission decrease while the absorption increased due to the increased of the deposition rate which leads to increase the thin film thickness, the highest transmission observed at 1200 mJ with the lowest absorption [45-47]. Table 2 shows the energy band gap values at each laser energy, as the particle size reduced at 1600 mJ the energy gap increased due to the quantum confinement, as the laser energy increased the higher energy band gap observed with a total blue shift [48-50].

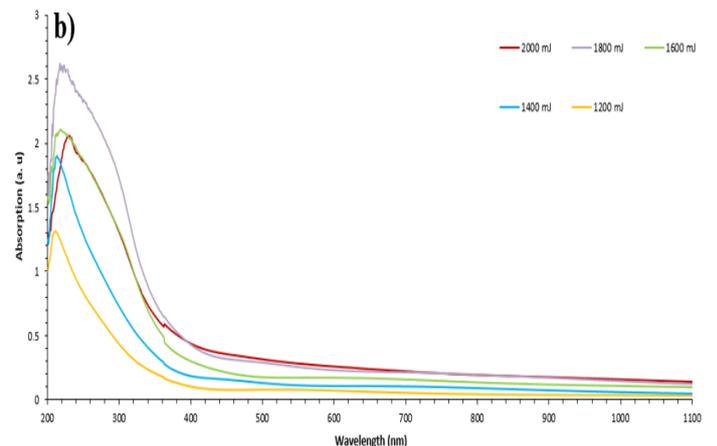
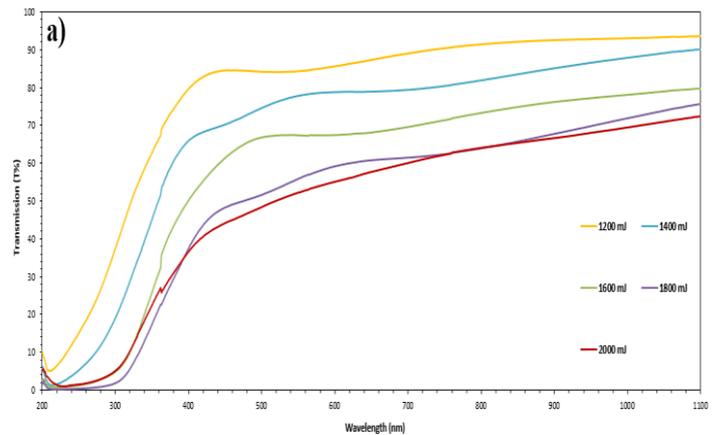
The relationship between laser energy and particle size, with the resulting quantum confinement effect, allows for the precise adjustment of the energy band gap in In₂O₃ thin films.

This control is vital for tailoring the films' characteristics to meet the unique demands of optoelectronics, photovoltaics, and other applications, where the band gap plays a crucial role in device performance.

The relationship between laser energy and the optical properties of In₂O₃ thin films is complex and crucial for tailoring the film's characteristics for various applications. The variation in laser energy can impact the transmission, absorption and energy band gap of the thin films.

The lower laser energies result in higher transmission, wider band gaps, and better transparency, while higher laser energies lead to increase the absorption, narrower band gaps, and reduced transparency due to thicker film deposition.

The optical properties of In₂O₃ thin films, including their energy band gap, are highly relevant to a wide range of applications. The observed blue shift in the band gap due to tunable deposition conditions allows for the customization of the optical properties, which is valuable in optoelectronics, photovoltaics, and emerging technologies, where precise control over light-matter interactions is essential.



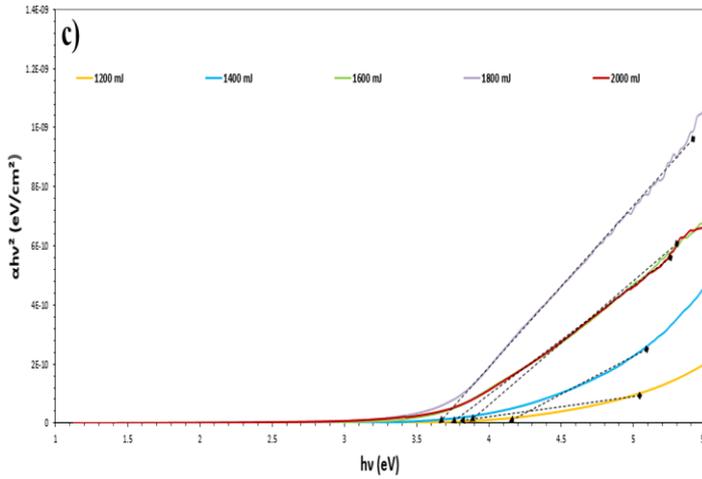


Figure 4. a) The Transmission b) absorption c) energy gap of In₂O₃/quartz at different laser energies with 1064 nm laser wavelength and 300°C substrate temperature.

Table 2 The energy band gap values at different laser energies

Laser Energy (mJ)	Energy Band Gap (eV)
1200	3.89
1400	3.81
1600	3.75
1800	3.83
2000	3.91

Figure 5 present the FESEM image of the Nano In₂O₃ grown film at the optimum condition laser energy 1800 mJ. The Nano In₂O₃ granes covered the PSi layer. As shown in the table 3 increasing in the particle size with the laser energies increasing as a result of laser fluency effects.

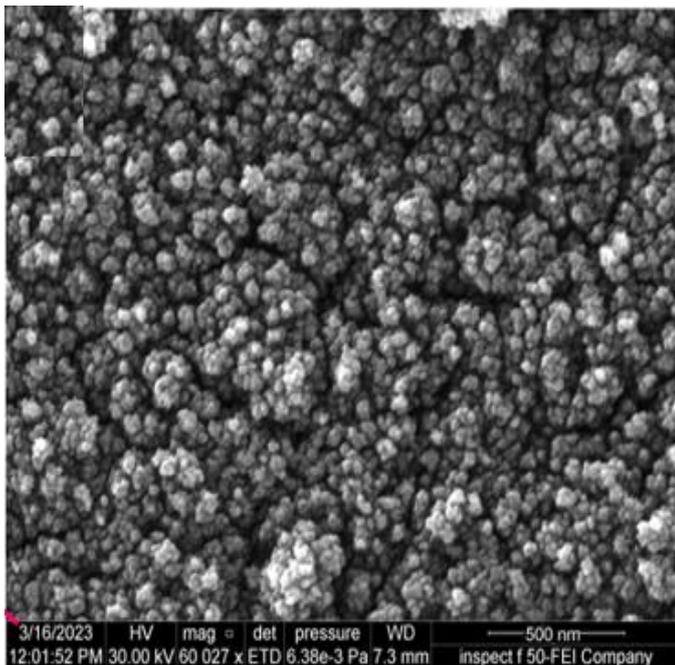


Figure 5. FESEM images of the Nano In₂O₃ grown thin film at 1800 mJ laser energy with Magnified of 120,000.

Figures 6 present the grown of Nano In₂O₃ films over PSi layer at the optimum condition laser energy 1800 mJ. The present fig shows high distribution and soft structure. The surface roughness play an important role in the gas sensor performance. A rougher surface provides greater surface area for gas molecules to adsorb on and can enhance the sensitivity of the gas sensor response.

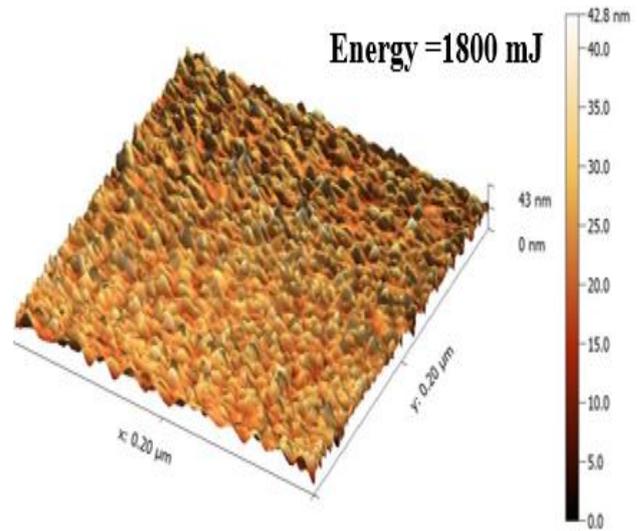


Figure 6. 3D AFM of the grown Nano In₂O₃/PSi deposited at 1800 mJ laser energy.

4. CONCLUSION

UV-visible spectrophotometer investigated the optical properties, of In₂O₃ thin films at different laser energies (1200 mJ, 1400mJ, 1600mJ, 1800 mJ, and 2000 mJ) and measured the energy band gaps for the five samples (3.89, 3.81, 3.75, 3.83, 3.91 eV) respectively. As the laser energy increased the higher energy band gap was observed with an overall blue shift from the fundamental energy gap of In₂O₃ thin films. The optimum energy band gab was obtained at 1600 mJ which is closer to the energy gap of In₂O₃ thin films. The key findings and contributions:

- The study focused on In₂O₃ thin films deposited using PLD and their optical properties.
- It demonstrated that variations in laser energy can control the particle size and consequently, the energy band gap of the thin films.
- Specifically, higher laser energy led to smaller particle sizes and a blue shift in the band gap.
- The ability to tailor the band gap is significant for applications in optoelectronics and photovoltaics, allowing for improved performance and efficiency.
- The research highlighted the importance of quantum confinement in achieving tunable energy gaps in thin films.

Further research directions and the potential areas of investigation:

- 1- Multifunctional thin films.
- 2- Material integration.
- 3- Stability and longevity.
- 4- Nanoscale engineering.
- 5- Incorporating dopants.
- 6- Alternative deposition techniques.
- 7- Environmental impact.

It can also continue expanding our understanding and harness the full potential of these versatile materials.

REFERENCES

- [1] Pawar, R. C., Mali, S. S., Devan, R. S., & Pawar, S. H. *Materials Science and Engineering: B*, vol. 250 (2020) pp. 114295.
- [2] Salim, E. T., Khalef, W. K., Fakhri, M. A., Fadhil, R. B., Azzahrani, A. S., Ibrahim, R. K., Ismail, R. A., *Scientific Reports*, vol. 13 (2023) p. 11514.
- [3] Fakhri, M. A., Salim, E. T., Sulaiman, G. M., Albukhaty, S., Ali, H. S., Salim, Z. T., Gopinath, S. C. B., Hashim, U., Al-aqbi, Z. T., *Plasmonics* (2023) <https://doi.org/10.1007/s11468-023-01961-3>.
- [4] Adam, H., Gopinath, S. C. B., Md Arshad, M. K., Sasidharan, S., Wu, Y. S. *Journal of the Taiwan Institute of Chemical Engineers*, vol. 146 (2023) p. 104814.
- [5] Ismail, R. A., Salim, E.T., Alwazny, M.S., *Plasmonics*, vol. 18, issue 2 (2023) pp. 561-576.
- [6] Kim, K. H., Kim, Y. K., Kim, K. T., and Kim, J. M., *Nanotechnology*, vol. 30 (2019) p. 355101.
- [7] Salim, E. T., Fakhri, M. A., Kadhim, A. C., Azzahrani, A. S., and Alsultany, F. H., *Frontiers in Optics + Laser Science 2022 (FIO, LS) Technical Digest Series (Optica Publishing Group, (2022), paper JW5A.75. <https://doi.org/10.1364/FIO.2022.JW5A.75>*.
- [8] Y. Zhang, Y. Zhang, H. Liu, and W. Wang, *Al-Ani, Biotechnology Reports*, vol. 26 (2020) pp. e00422.
- [9] Fakhri, M. A., Razzaq, M. J. A., Jabbar, H. D., Salim, E. T., Alsultany, F. H., Hashim, U., *Optical Materials* vol. 137 (2023) p. 113593.
- [10] Salim, E. T., Shafeeq, S. R., M. J. A. Razzaq, Fakhri, M. A., Gopinath, S. C. B., *Surfaces and Interfaces*, vol. 36 (2023) p. 102618.
- [11] J. Yan, L. He, X. Cui, Q. Chen, and H. Luo, *Journal of Materials Science: Materials in Electronics*, vol. 30, issue 19 (2019) pp. 17405-17424.
- [12] Fakhri, M.A., Alwahib, A.A., Alhasan, S.F.H., ...Abdulwahab, A.W., Hashim, U., *Journal of Optics (India)* (2023) <https://doi.org/10.1007/s12596-023-01173-2>.
- [13] A. S. A. Al-Hadi, S. A. Hussain, S. H. HasanAl-Ani, *Biotechnology Reports*, vol. 26 (2020) pp. e00422.
- [14] Salim, E.T., Saimon, J.A., Abood, M.K., ...Alsultany, F.H., *International Journal of Nanoelectronics and Materials*, vol. 16, issue 1 (2023) pp. 21-32.
- [15] Iqbal, S., Shah, M. A. B. S., Qamar, M. M. and Khan, M. R., *Journal of Materials Science: Materials in Electronics*, vol. 31, issue 3 (2020) pp. 1413-1428.
- [16] Jabbar, H. D., Fakhri, M. A., Razzaq, M. J. A., Dahham, O. S., Salim, E. T., Alsultany, F. H., *Journal of Renewable Materials* vol. 11, issue 3, (2023) 1101-1122. <https://doi.org/10.32604/jrm.2023.023698>.
- [17] Singh, S. P., Pandey, P., Kumar, A., and Singh, V. K., *Journal of Sol-Gel Science and Technology*, vol. 93, issue 1 (2020) pp. 1-19.
- [18] Mohammed, R. S., Fakhri, M. A., Alsultany, F. H. and Hashim, U., *International Journal of Nanoelectronics and Materials*, vol. 15 (Special Issue) (2022) pp. 93-102.
- [19] Kim, J., Lee, H., Park, J., and Lee, J. H., *ACS Applied Materials and Interfaces*, vol. 12, issue 17 (2020) pp. 19825-19832.
- [20] Khalaf, R. M., and Fakhri, M. A., *International Journal of Nanoelectronics and Materials*, vol. 15 (Special Issue) (2022) pp. 103-110.
- [21] Resen, D. A., Mohammed, M. F., and Fakhri, M. A., *International Journal of Nanoelectronics and Materials*, vol. 15 (Special Issue) (2022) pp. 165-181.
- [22] S. Palanisamy, S. Thanikaivelan, and S. Ponnusamy, in *Nanobiosensors*, Elsevier, (2020) pp. 139-150.
- [23] Shafeeq, S. R., Salim, E. T. and Razzaq, M. J. A., *International Journal of Nanoelectronics and Materials* vol. 15 (Special Issue) (2022) pp. 1-12.
- [24] Abdulrahman, T. E., Mahdi, R. O. and Salim, E. T., *International Journal of Nanoelectronics and Materials*, vol. 15 (Special Issue) (2022) pp. 13-25.
- [25] Barfidokht, N., Hassan, M. Z., Ahmad, F., and Yaacob, M. H., *Journal of Materials Science: Materials in Electronics*, vol. 31, issue 6 (2020) pp. 3752-3774.
- [26] Fadhil, R. B., Salim, E. T., Khalef, W. K. and Alsultany, F. H., *International Journal of Nanoelectronics and Materials*, vol. 15 (Special Issue) (2022) pp. 49-58.
- [27] Song, H., Joo, H. J., Yoon, S., and Han, S. W., *Biosensors and Bioelectronics*, vol. 169 (2020) pp. 112574.
- [28] Muhsin, M. S., Saimon, J. A. and Salim, E. T., *International Journal of Nanoelectronics and Materials*, vol. 15 (Special Issue) December (2022) pp. 111-118.
- [29] Kudhur, A. Y., Salim, E. T., Kara, I., Mahdi, R. O. and Alsultany, F. H., *International Journal of Nanoelectronics and Materials*, vol. 15 (Special Issue) (2022) pp. 131-137.
- [30] Zhang, Y., Xu, X., Wei, Z., Zhang, H., and Liu, J., *Materials Today Chemistry*, vol. 17 (2020) pp. 100326.
- [31] Fakhri, M. A., Alwahib, A. A., Salim, E. T., Ismail, R. A., Amir, H. A. A. A., Ibrahim, R. K., Alhasan, S. F. H., Alsultany, F. H., Salim, Z. T., Gopinath, S. C. B., *Silicon*, (2023) <https://doi.org/10.1007/s12633-023-02528-x>.
- [32] Jabbar, H. D., Fakhri, M. A., Razzaq, M. J. A., *Silicon*, vol. 14, issue 18 (2022) pp. 12837-12853.
- [33] S. Shinde, S. R. Sangle, S. S. Mali, and S. H. Pawar, *Biosensors and Bioelectronics*, vol. 176 (2021) pp. 112915.
- [34] Mohamed, F. A., Salim, E. T., Hassan, A. I., *Digest Journal of Nanomaterials and Biostructures*, vol. 17, issue 3 (2022) pp. 1029 – 1043.
- [35] Fadhil, R. B, Salim, E. T., Khalef, W. K., *Egyptian Journal of Chemistry*, vol. 66, issue 4 (2023) pp. 63-70.

- [36] Chaitanya, K. V., Kumar, K. N., Kadam, K. D., and Swamy, K. M. K., *Journal of Materials Science: Materials in Electronics*, vol. 32, issue 3 (2021) pp. 2435-2446.
- [37] Abdulrahman, T. E., Salim, E. T., Mahdi, R. O and Wahid, M. H. A., *Adv. Nat. Sci: Nanosci. Nanotechnol.* Vol. 13 (2022) p. 045006.
- [38] Salim, E. T., Yahya, A. M., and Abdulwahab, A. W., *AIP Conference Proceedings*, vol. 2660 (2022) pp. 020130.
- [39] Hu, C., Liu, Y., and Li, Y., *Biosensors and Bioelectronics*, vol. 171 (2021) pp. 112703.
- [40] Fakhri M.A., Jabbar H.D., Alsultany F.H., Salim E.T., Hashim U., *AIP Conference Proceedings* vol. 2660 (2022) p. 020124.
- [41] Mohammed, R. S., and Fakhri M.A., *AIP Conference Proceedings*, vol. 2660 (2022) p. 020133.
- [42] J. Shao, Z. Huang, Q. Wang, and H. Liu, *Journal of Physics: Conference Series*, vol. 1333, issue 1 (2019) p. 012007.
- [43] Fakhri M.A., Alwahib, A. A., Salim, E. T., Amir, H. A. A A., Alsultany, F. H and Hashim, U., *Physica Scripta*, vol. 97, issue 11 (2022) 115813.
- [44] Amir, H. A. A A., Fakhri M.A., Alwahib, A. A., Salim, E. T., Alsultany, F. H and Hashim, U., *Materials Science in Semiconductor Processing*, vol. 150 (2022) p. 106911.
- [45] Varma, H. K., Mehta, S. K., Patil, P. S., Kondawar, S. B., and Dhopte, S. K. *Journal of Alloys and Compounds*, vol. 775 (2019) pp. 1018-1026.
- [46] Jasim, F. Z., Razzak, M. J. A., Ahmed, H. M., *Optoelectronics and Advanced Materials- Rapid Communications*, vol. 8, issue 1-2, (2014) pp 7-9.
- [47] Evan T. Salim, Ali A. Taha, Sura A. Abdullatef, and Mohammed M. Farhan, *AIP Conference Proceedings* vol. 2400 (2022) p. 030015.
- [48] Mohamed, F. A., Hassan, A. I., Salim, Evan T. *International Journal of Nanoelectronics and Materials*, vil. 15, issue 4 (2022) pp. 281-292.
- [49] Kim, Y. H., Kim, M. J., Kim, J. H., Park, J. H., Lee, J. Y. and Son, J. G., *Journal of the Korean Physical Society*, vol. 74, issue 12 (2019) pp. 1192-1198.
- [50] Amir, H. A. A. A., Fakhri, M. A., Alwahib, A. A., Salim, E. T., Alsultany, F. H., Hashim, U., *Sensors and Actuators B: Chemical*, vol. 367 (2022) p. 132163.