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Effect of Argon:Oxygen Gas Mixing Ratio on Characteristics of Silicon Dioxide Nanoparticles Synthesized by DC Reactive Sputtering

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In this work, nanostructured SiO_2 thin films were deposited on glass substrates using DC reactive magnetron sputtering technique. Gas mixtures of argon and oxygen at different mixing ratios were used to synthesize SiO_2 nanoparticles. A transition from amorphous to partially crystalline phases was revealed as the oxygen ratio increased, indicating enhanced crystallinity under oxygen-rich conditions. The morphology of the film surface varied and the particle size decreased with higher oxygen content, suggesting improved oxidation and limited grain growth. The results confirm that the Ar:O_2 gas ratio plays a critical role in tuning the structural and optical properties of SiO_2 nanostructured thin films.

Keyword: Silicon dioxide; Gas mixture; Nanoparticles; Surface morphology
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1. Introduction

Silicon dioxide (SiO_2), commonly known as silica, is a common oxide compound made up of silicon and oxygen atoms bonded together. Within the silica molecules, every silicon atom connects with four oxygen atoms while every oxygen atom connects with two silicon atoms by Si-O bonds [1-3]. This compound is prevalent in various crystalline forms, notably quartz, and is also found in amorphous forms such as glass [4]. The basic structural unit of the silicon dioxide is a tetrahedron formed by a silicon atom and four oxygen atoms. The silicon atom sits in the center of the structure and is chemically bonded to the oxygen atoms in the four corners of the tetrahedron. The oxygen atom has two valence electrons, and thus it has a possibility to form a bond to the silicon atom of the neighboring tetrahedron. Since the oxide is amorphous, all of the oxygen atoms do not form bonds between the adjacent structural units. Consequently, depending on the bonding state of the oxygen atoms, they are referred to as bridging or non-bridging oxygen [5]. The amorphous structure forms a more open network and thus the density of the oxide is less than the crystalline form (i.e., the quartz) [6]. SiO_2 is characterized by a substantial band gap, this large band gap classifies SiO_2 as an insulator, making it an excellent electrical insulator in various applications [7]. SiO_2 nanoparticles are used in various fields, including chemical, biological, and medical applications, due to their chemical stability, optical transparency, low toxicity [8], with low refractive index [9]. It serves as a fundamental component in microelectronics [10], solar cells [11], and protective coatings [12]. SiO_2 films are widely used as low-refractive-index layers in multilayer optical devices [13,14], as passivation and protective coatings for silicon devices [15], and as scratch-resistant coatings for plastic ophthalmic lenses [16,17], and so on. The physical and optical properties of SiO_2 layer depend on the method of deposition [18-24]. In reactive sputtering used to prepare nanostructured SiO_2 thin films, compound thin films are deposited in the presence of a reactive gas. The reactive gas reacts with the sputtered material and forms a compound. This process makes it possible to deposit a wide variety of compounds (oxides, nitrides, carbides, etc.) with a wide range of properties [25]. The ability to deposit high-quality SiO_2 thin films with controlled thickness and structural properties is crucial for enhancing the performance of advanced technologies [26-28]. The key parameters influencing the film quality include oxygen partial pressure, DC power, working gas pressure, and substrate temperature, which must be optimized to achieve the desired structural, optical, and electrical properties [29-31].

In DC reactive sputtering, the gas mixing ratio significantly influences the characteristics of thin films. Adjusting the ratio of inert gas (e.g., argon) to reactive gas (e.g., oxygen, nitrogen, etc.) controls the film's composition, microstructure, and properties. For instance, increasing the reactive gas can lead to higher oxygen/nitrogen content, influencing phases (e.g., from nitride to oxynitride), crystallinity, and grain size, sometimes even causing amorphization [32-34]. This, in turn, impacts optical properties like bandgap, and electrical properties such as resistivity, which can shift from conducting to semiconducting or insulating. The gas ratio also affects deposition rates and surface morphology, making it a critical parameter for tailoring film performance in various applications [35-37].

The aim of this research is to synthesize silicon dioxide thin films using DC reactive magnetron sputtering, analyze their structural, morphological, and optical properties, and introduce the effects of gas mixing ratio on these characteristics.

2. Experimental work

A homemade dc reactive magnetron sputtering system was used to deposit SiO_2 thin films on glass substrates. A two-stage Leybold-Heraeuos rotary pump (24 m^3/h) was used to reach a base pressure of about 0.15 mbar. The targets were cleaned and dried for the deposition process. The glass substrates which are used for depositing the thin films were initially cleaned before the experiments. The target was maintained carefully on the cathode. The plasma required for sputtering was generated by the electric discharge of argon. Electrical power was provided by a high-voltage DC power supply. The operating conditions of the system were divided into two groups: constant and variable. The constant operating conditions include vacuum pressure, current limiting resistance, discharge voltage, discharge current, flow rate, deposition temperature, inter-electrode distance. The variable operating conditions were operating in deposition times of 30, 45, 60, 90 and 120 min for thin films, and gas mixing ratio (30:70), (70:30), (50:50), (10:90), (90:10). A silicon sheet with purity of 99.9% was used as a sputtered target. The discharge current was maintained at 35 mA and the inter-electrode distance was maintained at 4 cm.

The SiO_2 nanopowder was extracted from thin film samples by the conjunctional freezing-assisted ultrasonic extraction method. Full description and specifications of this method can be introduced in references [38-42]. The structural properties of the extracted nanopowders were determined by x-ray diffraction (XRD), field-emission scanning electron microscopy (FE-SEM), and energy-dispersive x-ray spectroscopy (EDX).

3. Results and Discussion

Figure (1) shows the XRD patterns of the SiO_2 film samples prepared by dc reactive magnetron sputtering using different mixing ratios of $Ar:O_2$ gases. The XRD analysis confirms that the structural nature of the SiO_2 thin films is strongly influenced by the variation in the $Ar:O_2$ gas mixture during deposition. The films deposited at higher oxygen ratios (e.g., 10:90 and 30:70) display sharper and more intense diffraction peaks, indicating the formation of partially crystalline SiO_2 phases. In contrast, films prepared under lower oxygen flow rates (e.g., 90:10 and 70:30) exhibit broad and low-intensity humps around $2\theta \approx 23°$, which are characteristic of an amorphous SiO_2 structure, that has no crystal structure, so it has no plane with specific miller indices [14,30,32]. The crystalline peaks observed at 2θ values around 36.5°, 60.1°, and 72.5° can be indexed to the (110), (113), and (020) planes of SiO_2, respectively, corresponding to known crystalline forms such as quartz or cristobalite [PDF 01-085-0794]. The appearance and growth of these peaks with increasing oxygen content suggest that higher oxygen partial pressures promote complete oxidation of sputtered Si atoms and facilitate crystallization within the SiO_2 films.

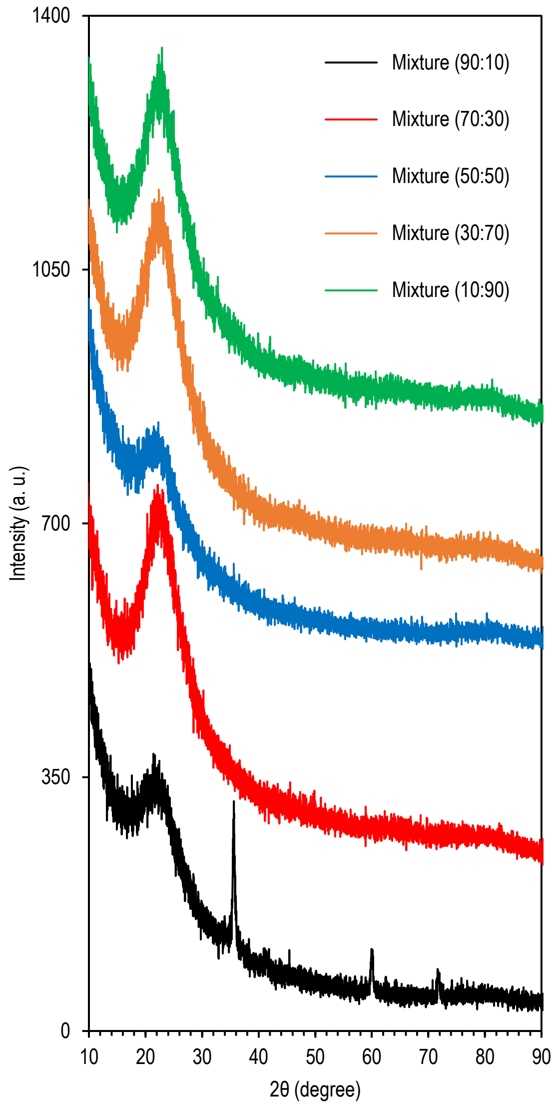
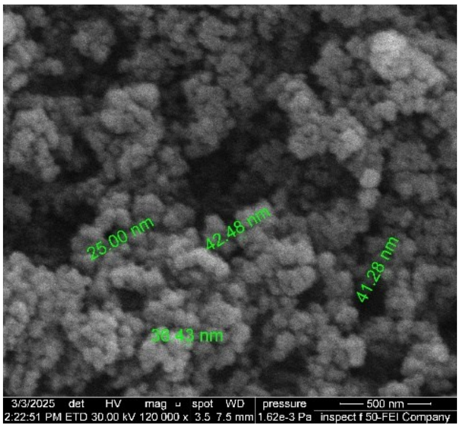


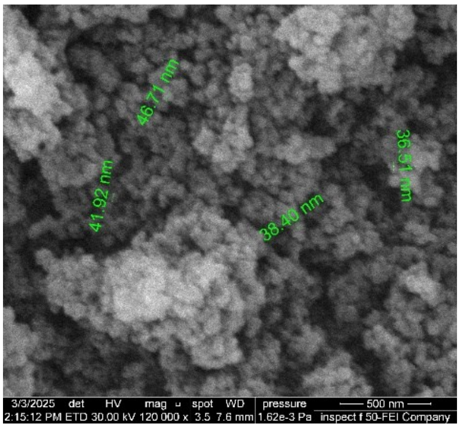
Fig. (1) XRD patterns of SiO_2 thin films prepared at different $Ar:O_2$ mixing ratios

According to the filed-emission scanning electron microscopy (FE-SEM) results shown in Fig. (2), using the 30:70 ratio, increasing the oxygen content improved oxidation with some porosity, with an average particle size of 40.88 nm. However, the high oxygen content using 10:90 ratio negatively affected the deposition quality, as the surface appeared heterogeneous with small, dispersed particles, with an average particle size of 36.29 nm. Using the 50:50 mixing ratio, a smooth, dense layer with homogeneous nanoparticles was obtained, and the average particle size was 35.4 nm, indicating that the deposition quality was optimal.

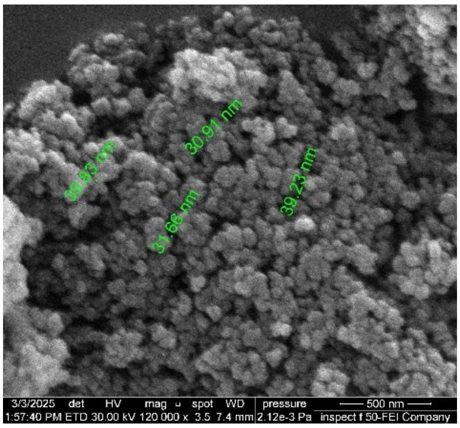
Using the 90:10 ratio, the oxygen content was low, resulting in incomplete deposition and the growth of large, heterogeneous particles with an average particle size of 36.86 nm, The surface was relatively rough. Using the 70:30 ratio, improved particle distribution and structural regularity were observed, with an average particle size of 34.07 nm, and the surface was more homogeneous.



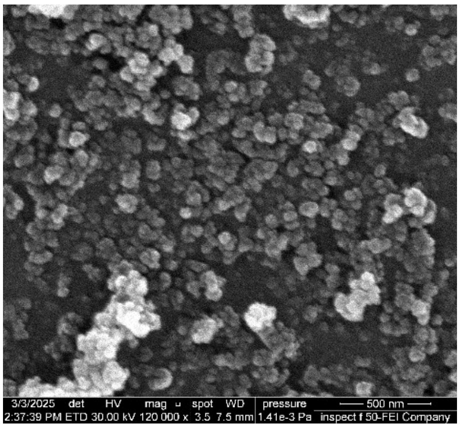
(a) Ar:O₂(10:90)



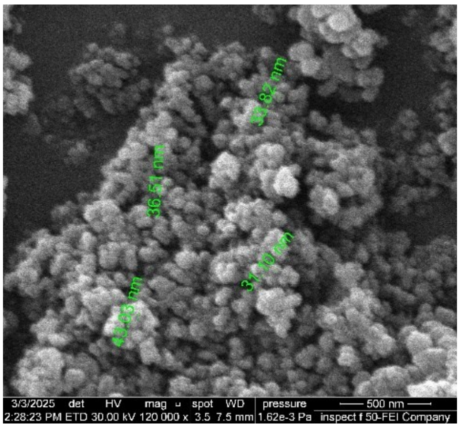
(b) Ar:O₂(30:70)



(c) Ar:O₂(50:50)



(d) Ar:O₂(90:10)



(e) Ar:O₂(70:30)

Fig. (2) FE-SEM images of SiO_2 thin films prepared at different $Ar:O_2$ mixing ratios

Figure (3) shows the contents of the composing elements for SiO_2 in the final product determined by the EDX measurements obtained for the samples prepared using different mixing ratios of $Ar:O_2$ gases. At high O_2 contents (10:90, 30:70, 50:50), the formation of stoichiometric SiO_2 was revealed. As the Ar content increases (70:30 and 90:10), a decline in oxidation efficiency was suggested and the possible formation of substoichiometric silicon oxide was confirmed. It was found that the lowest percentage of silicon to oxygen was at a gas mixing ratio of 70:30, while the highest percentage of silicon to oxygen was at a mixing ratio of 50:50 and 30:70.

4. Conclusions

The results indicate that the structural and morphological quality of SiO_2 thin films is highly sensitive to the reactive gas environment during DC magnetron sputtering. Controlled oxygen incorporation enhances film crystallinity, reduces surface roughness, and improves chemical stoichiometry, thereby enabling the fabrication of high-purity, homogeneous SiO_2 nanostructures suitable for advanced optical and electronic applications. The variation in film structure is solely due to the different $Ar:O_2$ gas ratios used during sputtering. The adjustment of the oxygen

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content effectively controls the microstructure, enabling a transition from an amorphous to a partially crystalline SiO_2 phase.

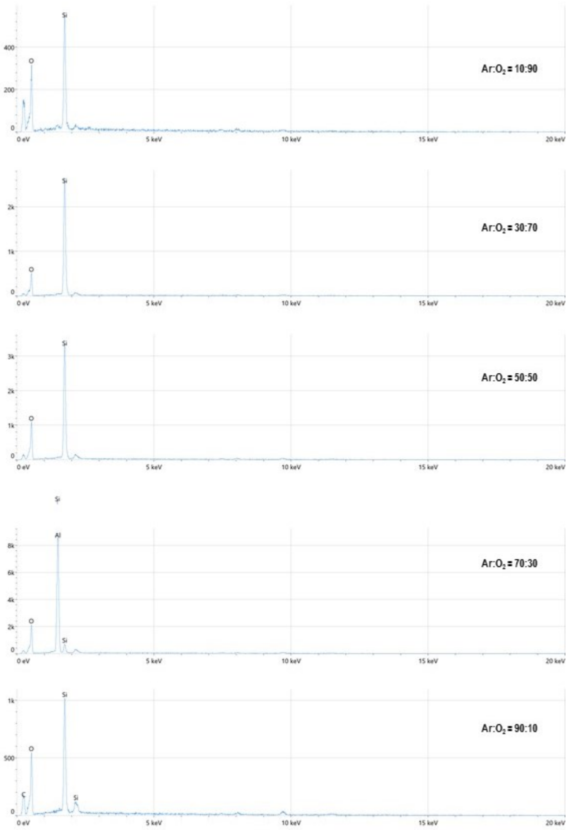

Fig. (3) EDX spectra of SiO_2 thin films prepared at different Ar:O_2 mixing ratios

References

[1] A.M. Hameed and M. A. Hameed, "Spectroscopic characteristics of highly pure metal oxide nanostructures prepared by DC reactive magnetron sputtering technique", *Emerg. Mater.*, 6(2) (2023) 627–633.

[2] K.K. Wang, P.V. Chai, and W.L. Ang, "Introduction to Nanomaterials", *Carbon Nanostructures*, F2589 (2024) 1-15.

[3] K. Pradhan et al., "Exploration of impact of ammonia concentration on the surface morphology, optical and wettability performance of SiO_2 thin film", *J. Mater. Sci. Mater. Electron.*, 36(5) (2025) 1-14.

[4] L.A.J. Garvie et al., "Bonding in alpha-quartz (SiO_2): A view of the unoccupied states", *Am. Mineral.*, 85(5-6) (2000) 732-738.

[5] N. Li and W.Y. Ching, "Structural, electronic and optical properties of a large random network model of amorphous SiO_2 glass", *J. Non. Cryst. Solids*, 383 (2014) 28-32.

[6] S. Eränen, "Silicon Dioxides", in Handbook of Silicon Based MEMS Mater. Technol., (2010) pp. 137–148.

[7] D.A.P. Wardani et al., "Functional Groups, Band Gap Energy, and Morphology Properties of Annealed Silicon Dioxide (SiO_2)", *Egypt. J. Chem.*, 66(3) (2023) 529-535.

[8] C.A. Banciu et al., "Comparative study of the hydrophobic properties of silicon dioxide particles functionalized with different agents", *J. Optoelectron. Adv. Mater.*, 25(1-2) (2023) 89-95.

[9] T. Oyama et al., "A new layer system of anti-reflective coating for cathode ray tubes", *Thin Solid Films*, 351(1-2) (1999) 235-240.

[10] V. Bhatt and S. Chandra, "Silicon dioxide films by RF sputtering for microelectronic and MEMS applications", *J. Micromech. Microeng.*, 17(5) (2007) 1066-1077.

[11] S.W. Glunz and F. Feldmann, "SiO_2 surface passivation layers – a key technology for silicon solar cells", *Sol. Ener. Mater. Sol. Cells*, 185 (2018) 260-269.

[12] T.S. Chen et al., "The effect of the native silicon dioxide interfacial layer on photovoltaic characteristics of gold/p-type amorphous boron carbon thin film alloy/silicon dioxide/n-type silicon/aluminum solar cells", *Sol. Ener. Mater. Sol. Cells*, 137 (2015) 185-192.

[13] H. Jung et al., "Growth characteristics and electrical properties of SiO_2 thin films prepared using plasma-enhanced atomic layer deposition and chemical vapor deposition with an aminosilane precursor", *J. Mater. Sci.*, 51(11) (2016) 5082-5091.

[14] M.A. Hameed and Z.M. Jabbar, "Surface Morphology and Topography of Silicon Dioxide Nanostructures Prepared by DC Reactive Sputtering", *Iraqi J. Appl. Phys. Lett.*, 7(3) (2024) 23-26.

[15] M. Yu et al., "Comparative study of the characteristics of Ni films deposited on SiO_2/Si(100) by oblique-angle sputtering and conventional sputtering", *Thin Solid Films*, 516(21) (2008) 7903-7909.

[16] E.S.M. Goh et al., "Thickness effect on the band gap and optical properties of germanium thin films", *J. Appl. Phys.*, 107(2) (2010) 1-5.

[17] Z. Falah and A.M. Rabee, "The Effects of Combined toxicity of Silver and Silicon Nanoparticles on Hematological and Biochemical Parameters in Male Albino Mice", *Iraqi J. Sci.*, 63(10) (2022) 4195-4204.

[18] M. Henini, **"Handbook of Thin-Film Deposition Processes and Techniques"**, 2^{nd} ed., William Andrew Inc. (2001), vol. 31, no. 3.

[19] V. Jokanovic et al., "Thin films of SiO2 and hydroxyapatite on titanium deposited by spray pyrolysis", *J. Mater. Sci. Mater. Med.*, 19(5) (2008) 1871-1879.

[20] W. Zhang et al., "Preparation of SiO_2 anti-

reflection coatings by sol-gel method", *Energy Procedia*, 130 (2017) 72-76.

[21] G. Wang et al., "Preparation methods and application of silicon oxide films", *Int. Conf. Mechatronics, Electron. Ind. Control Eng. MEIC 2014*, pp. 479–483.

[22] S. Chen et al., "Vanadium oxide thin films deposited on silicon dioxide buffer layers by magnetron sputtering", *Thin Solid Films*, 497(1-2) (2006) 267-269.

[23] A. Ranjgar et al., "Characterization and Optical Absorption Properties of Plasmonic Nanostructured Thin Films", *Armen. J. Phys.*, 6(4) (2013) 198-203.

[24] I. Safi, "Recent aspects concerning DC reactive magnetron sputtering of thin films: A review", *Surf. Coat. Technol.*, 127(2-3) (2000) 203-218.

[25] P. Carvalho et al., "Influence of the chemical and electronic structure on the electrical behavior of zirconium oxynitride films", *J. Appl. Phys.*, 103(10) (2008) doi: 10.1063/1.2927494.

[26] K. Pfeiffer et al., "Comparative study of ALD SiO_2 thin films for optical applications", *Opt. Mater. Exp.*, 6(2) (2016) 660.

[27] M.D. Beltrán et al., "Double laser for depth measurement of thin films of ice," *Sensors (Switzerland)*, 15(10) (2015) 25123-25138.

[28] R.H. Turki and M.A. Hameed, "Spectral and Electrical Characteristics of Nanostructured NiO/TiO_2 Heterojunction Fabricated by DC Reactive Magnetron Sputtering", *Iraqi J. Appl. Phys.*, 16(3) (2020) 39-42.

[29] K. Praweerawat, C. Muangphat, and C. Luangchaisri, "The Preparation and Characterization of SiO_2 Films by Spray Coating Technique for Radiative Cooling Glass Application", *Mater. Today Proc.*, 23 (2018) 696-702.

[30] M.A. Hameed and Z.M. Jabbar, "Optimization of Preparation Conditions to Control Structural Characteristics of Silicon Dioxide Nanostructures Prepared by Magnetron Plasma Sputtering", *Silicon*, 10(4) (2018) 1411-1418.

[31] M.O. Yusuf, "Bond Characterization in Cementitious Material Binders Using Fourier-Transform Infrared Spectroscopy", *Appl. Sci.*, 13(5) (2023) doi: 10.3390/app13053353.

[32] H.G. Fahad and O.A. Hammadi, "Characterization of Highly-Pure Silicon Dioxide Nanoparticles as Scattering Centers for Random Gain Media", *Iraqi J. Appl. Phys.*, 16(2) (2020) 37-42.

[33] N.H. Mutesher and F.J. Kadhim, "Comparative Study of Structural and Optical Properties of Silicon Dioxide Nanoparticles Prepared by DC Reactive Sputtering and Sol-Gel Route", *Iraqi J. Appl. Phys.*, 17(1) (2021) 17-20.

[34] K.A. Al-Hamdani, "Current–voltage and capacitance-voltage characteristics of Se/Si heterojunction prepared by DC planar magnetron sputtering technique", *Iraqi J. Phys.*, 8(13) (2010) 97-100.

[35] K.A. Aadim, "Control the deposition uniformity using ring cathode by DC discharge technique", *Iraqi J. Phys.*, 15(32) (2017) 57-60.

[36] D.A. Taher and M.A. Hameed, "Employment of Silicon Nitride Films Prepared by DC Reactive Sputtering Technique for Ion Release Applications", *Iraqi J. Phys.*, 21(3) (2023) 33-40.

[37] F.J. Al-Maliki, O.A. Hammadi, and E.A. Al-Oubidy, "Optimization of Rutile/Anatase Ratio in Titanium Dioxide Nanostructures prepared by DC Magnetron Sputtering Technique", *Iraqi J. Sci.*, Special Issue (2019) 91-98.

[38] A.N. Munif and F.J. Kadhim, "Structural Characteristics and Photocatalytic activity of TiO2/Si3N4 nanocomposite synthesized via plasma sputtering technique", *Iraqi J. Phys.*, 22(4) (2024) 99-106.

[39] O.A. Hammadi, "Production of Nanopowders from Physical Vapor Deposited Films on Nonmetallic Substrates by Conjunctional Freezing-Assisted Ultrasonic Extraction Method", *Proc. IMechE, Part N, J. Nanomater. Nanoeng. Nanosys.*, 232(4) (2018) 135-140.

[40] O.A. Hammadi, "Conjunctional Freezing-Assisted Ultrasonic Extraction of Silicon Dioxide Nanopowders from Thin Films Prepared by Physical Vapor Deposition Technique", *Iraqi J. Appl. Phys.*, 15(4) (2019) 23-28.

[41] O.A. Hammadi, "Effects of Extraction Parameters on Particle Size of Titanium Dioxide Nanopowders Prepared by Physical Vapor Deposition Technique", *Plasmonics*, 15(6) (2020) 1747-1754.

[42] O.A. Hammadi, "Effects of Extraction Parameters on Particle Size of Iron Oxide Nanopowders Prepared by Physical Vapor Deposition Technique", *Iraqi J. Appl. Phys.*, 20(2B) (2024) 457-460.