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Research Article

Temperature-Dependent Optical Properties of Spray-Pyrolyzed Cadmium Oxide Thin Films

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Abstract: In present study, Cadmium Oxide (CdO) films have been prepared using spray pyrolysis deposition (SPD) at different substrate temperatures (350, 450, and 550 °C) with glass substrate. Thickness of these films prepared was approximately 400 ± 10 nm. XRD diffraction analysis revealed the CdO were polycrystalline and cubic, exhibiting grain sizes ranging from 198.5 to 229.8 nm. The effect of varying substrate temperatures on the optical properties of CdO was analyzed by examining transmittance and absorbance spectra within the 300 to 900 nm range. The transmittance of CdO increases with increasing the substrate temperature. The energy gap for direct transitions was determined to range between 2.4 and 2.5 electron volts. It was found that varying the substrate temperature affects CdO films, increasing its transmittance and energy gap, and decreasing its reflectivity, absorbance coefficient, extinction coefficient, real parts and imaginary parts of the dielectric constant, and refractive index. Based on these observed properties, thin films fabricated in this way can be used in sensors and catalysts, or in solar cells.

Keywords: Cadmium Oxide (CdO), Optical Properties, Spray Pyrolysis Deposition (SPD), Energy Gap, X-ray Diffraction.

1. INTRODUCTION

Currently, cadmium oxide (CdO) films have become increasingly important in the photovoltaic cell industry, mainly due to their exceptional optical transparency and excellent electrical conductivity across visible and ultra-visible light spectra, and having excellent gas sensing properties [1]. CdO is classified as a transparent conductive oxide due to its remarkable physical properties, including its high optical transmittance within the visible spectrum and excellent charge carrier mobility [2]. Conductive CdO films are gaining increasing recognition for their significant importance due to their diverse industrial and technological applications, these applications include is diodes, solar cells, phototransistors, and gas sensors [3].

CdO is classified in groups II-VI of the periodic table; having a cubic crystal structure characterized by XRD [4]. It is an n-type semiconductor characterized by direct energy band gap ranging from 2.2 - 2.5 electron volts [5]. Researchers have sought to discover new deposition techniques to improve upon existing methods, and it is observed that the physical properties of the films vary depending on the deposition method [6]. In this context, CdO has garnered significant attention due to its electrical, optical, and chemical properties, leading to its uses in advanced applications [7]. Various methods have been used to synthesize CdO thin films, such as chemical vapor deposition (CVD) [8, 9], chemical spray deposition [10], magnetic spray deposition [5], sol-gel deposition [11], and chemical spray decomposition (CSP) [12].

In the present study we have synthesized thin films of CdO using spray pyrolysis deposition technique and their optical properties are measured at different substrate temperatures (350, 450, and

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550 °C). The structural and optical properties of the as prepared films were thoroughly investigated by X-ray diffraction (XRD) and UV-Vis spectroscopy. In future, we can explore the potentials of this material as a gas sensor, catalyst, use in solar cells, and assess the feasibility of applying these findings in industrial applications.

2. MATERIALS AND METHODS

Cadmium acetate $Cd(CH_3COO)_2.2H_2O$ with molecular weight (266.52 g/mol), supplied by the British company (BHD) with a purity of 99%, was used to prepare the solution used to produce the CdO thin film. The solution was prepared at a molar concentration of (0.1 mol/L) by adding (2.6652 g) of it to (100 ml) of distilled water and gradually dissolving it by a magnetic stirrer for (15-20) minutes according to the Equation (1) [13]. This resulted in a clear, homogeneous, and colorless solution.

$$M = \left(\frac{W_t}{M_{wt}}\right)\left(\frac{1000}{V}\right) \quad (1)$$

where M is the molar concentration, W is the weight of the solute, M_{Wt} is the molecular weight of the substance, and V is the volume of distilled water in which the solute was dissolved.

After preparation of the solution Spray Pyrolysis Deposition (SPD) method was used to deposit CdO thin films at different substrate temperatures. Table 1 shows the detailed conditions used for the synthesis of the CdO thin films.

After spraying process is complete, the gases evaporate due to heat, leaving a cadmium oxide on the substrate according to the Equation of reaction [4]:

$$(CH_3COO)_2Cd.2H_2O + H_2O \rightarrow CdO + 2H_2O\uparrow + 2CH_3COOH\uparrow$$

The thin film thickness (t) was calculated using helium-neon (He-Ne) laser interferometry method, it was calculated from the following Equation (2) [14]:

$$t = \frac{\Delta L}{L} * \frac{\lambda}{2} \quad (2)$$

where λ is the wavelength of the laser light used and its value (632.8 nm), L is the width of the bright fringe (nm), and ΔL is the width of the dark fringe (nm).

Figure 1 depicts the diagram of workflow and measurements adopted during the preparation and analysis of the CdO thin films.

Table 1. Standard conditions used for the synthesis of the CdO thin films.

Parameters of spray	
Distance between substrate and atomizer	25 cm
Liquid flow rate	1 ml min^{-1}
Utilized air compressor	1 bar
Time for spraying	20 min
Interval between sprays	5 min

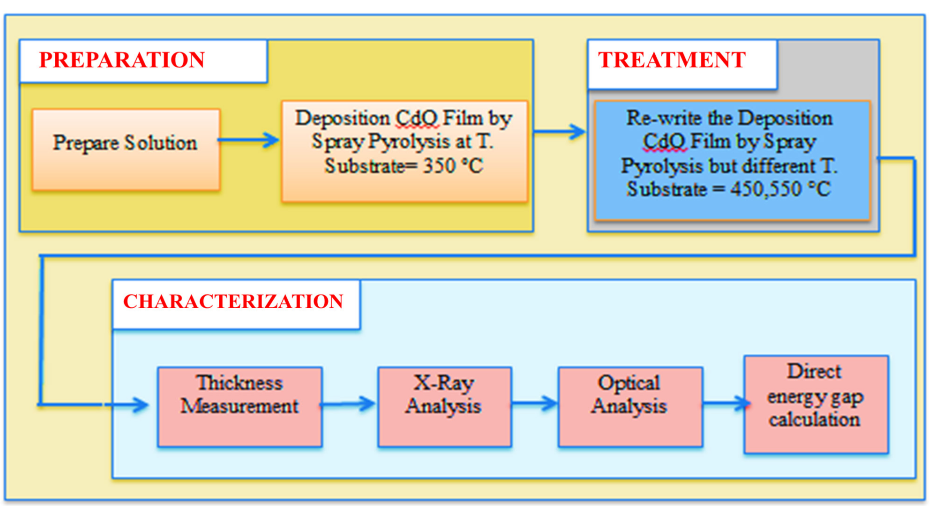

Fig. 1. Diagram the workflow and measurements used in the preparation and analysis of CdO thin films.

3. RESULTS AND DISCUSSION

The structural analysis and crystallinity of CdO thin films were investigated by X-ray diffraction which is crucial for understanding the relation between the optical properties and structural variations of these films. The XRD diffraction spectra shown in Figure 2, confirm that the CdO thin films are polycrystalline and have a cubic symmetry [9]; which is consistent with standard diffraction data (JCPDS card 05-0640). Figure 2 also represents that the main peaks (111), (200), and (220), which are the dominant diffraction peaks, indicate characteristic reflections of CdO. The highest intensity was recorded for (111) peak, which is the preferred direction for crystal growth [9].

The average grain size (G) was calculated from (111) plane by applying Scherrer Equation (3) [9]:

$$G = \frac{0.9\lambda}{\beta \cos\theta} \tag{3}$$

where λ represents the wavelength of the X-rays, β is the full width at half maximum (FWHM) of the diffraction peak and θ is the angle of the diffraction peak.

Table 2 shows that the average grain size increases with increasing substrate temperature, ranging from 198.5 to 229.8 nm. This increase in average crystal size is attributed to increased crystal disorder resulting from the higher substrate temperature.

The optical properties of the CdO thin films play a vital role in their evaluation and the determination of their potential applications. This includes examining key optical properties and determining the optical band gap of the CdO thin films at different substrate temperatures.

3.1. Transmittance (*T*)

Figure 3 illustrates the change in transmittance values with wavelength for CdO films prepared at different substrate temperatures. These results show that transmittance is increased with increasing wavelength; however, with increase in the substrate temperature, we observe that the behavior of the transmittance spectrum curve does not change. It is evident that the value of transmittance increases with increasing the substrate temperature; this can be attributed that by increasing the substrate temperature structural defects are reduced, which are attenuating regions for the incident radiation, and thus changes values of absorbance coefficient with increase in crystallinity of thin films. This is consistent with what was stated in the published research [15].

3.2. Absorbance (*A*)

Figure 4 demonstrates the change in absorbance values with wavelength for CdO films prepared at different substrate temperature. The maximum absorbance value is at short wavelengths, after

Table 2. Average grain size of the CdO thin films at different substrate temperature.

Temperature °C	350 °C	450 °C	550 °C
G(nm) (111)	198.5	226.1	229.8

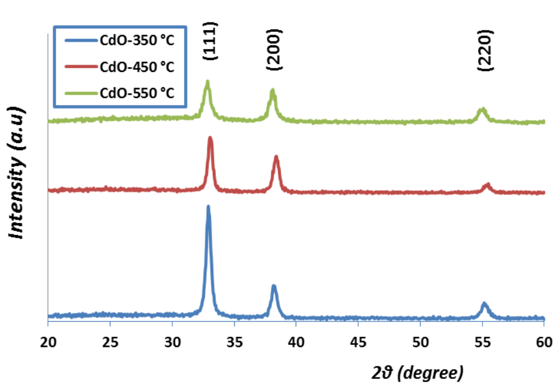

Fig. 2. XRD diffraction of CdO thin film with different substrate temperatures.

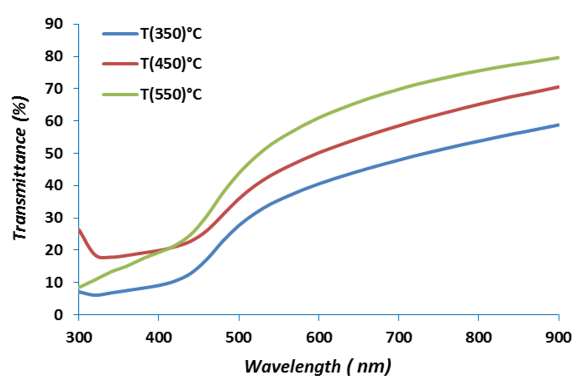

Fig. 3. Relationship between Transmittance (T) with wavelength (λ) for different substrate temperatures.

which it rapidly decreases in the (300-500) nm range, and then gradually decreases in the (500-900) nm range, reaching its lowest value at (900) nm. This decrease in the absorbance can be ascribed that the energy of the photons is less than energy gap value, making them unable to excite electrons and transfer them from the valence band to the conduction band [16]. In addition, from Figure 4 it observed that by increasing the substrate temperature does not affect the behavior of the absorbance spectrum curve; however, it is found that to have a decreasing trend in the absorbance value as the different substrate temperature is increased, and this can be due a change in energy gap values.

3.3. Reflectivity (*R*)

The reflectivity was calculated from the fallowing Equation (4) [17]:

$$R + T + A = 1 \tag{4}$$

Figure 5 shows the reflectivity decreases with increasing wavelength, as well as with increase in the substrate temperature. Because due to change in surface roughness with increasing temperature, it is known that roughness is inversely proportional to grain size. This behavior is consistent with the transmittance results and absorbance according to Equation (4).

3.4. Absorbance Coefficient (α)

The absorbance coefficient (α) is calculated from Equation (5) by employing Beer-Lambert Law [18]:

$$\alpha = \frac{2.303 \times A}{t} \tag{5}$$

where *A* is the absorbance of the material and *t* is the thickness of the film.

Figure 6 demonstrates that absorbance coefficient of the CdO films decreases with both increasing wavelength and increasing substrate temperature. The temperature-induced decrease can be attributed to the thermal remediation of structural defects. Deviations from the ideal stoichiometric and crystalline structure of a semiconductor typically generate localized energy levels within the band gap (above the valence band and below the conduction band), which supply free charge carriers (electrons or holes). Higher substrate temperatures reduce these structural deviations, thereby eliminating some of these localized states and decreasing the carrier concentration. Consequently, both free-carrier absorbance and defect-mediated transitions are suppressed, leading to a lower absorbance coefficient [19]. These results show that CdO the films have high absorbance coefficient $(\alpha > 10^4\ cm^{-1})$, this suggests the existence of direct electronic transitions [20].

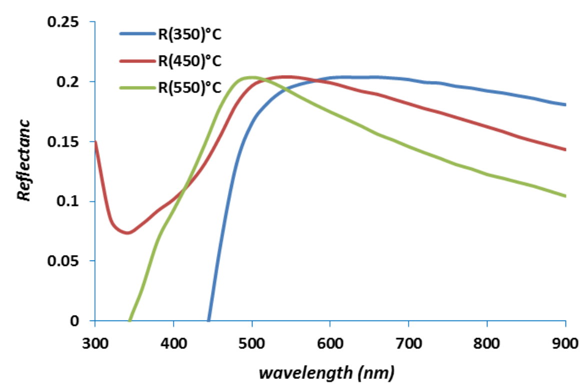

Fig. 5. Relationship between Reflectance (R) with wavelength (λ) for different substrate temperatures.

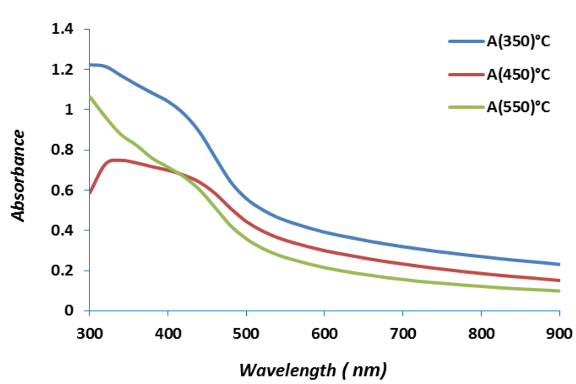

Fig. 4. Relationship between Absorbance (*A*) with wavelength (λ) for different substrate temperatures.

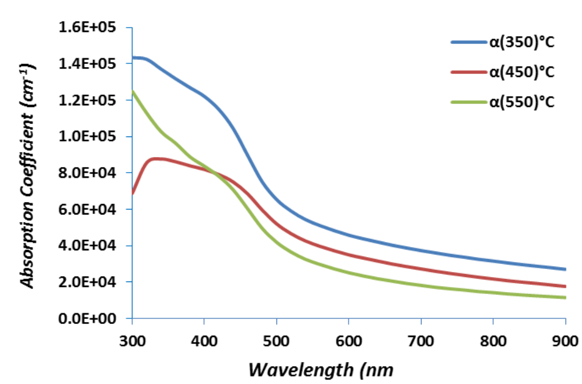

Fig. 6. Relationship between Absorbance Coefficients (α) with wavelength (λ) for different substrate temperatures.

3.5. Optical Energy Gap (E_g)

Figure 7 displays the variations in the optical energy gap (E_g) of the CdO films prepared at different substrate temperatures. The data reveals that the energy gap increases with rising substrate temperature. This broadening of the band gap can be attributed to the thermal reduction of structural defects and the subsequent elimination of localized states lying within the forbidden region between the valence and conduction bands. As these mid-gap defect states are minimized at higher temperatures, the fundamental absorbance edge sharpens and shifts to higher energies, resulting in an increased effective energy gap.

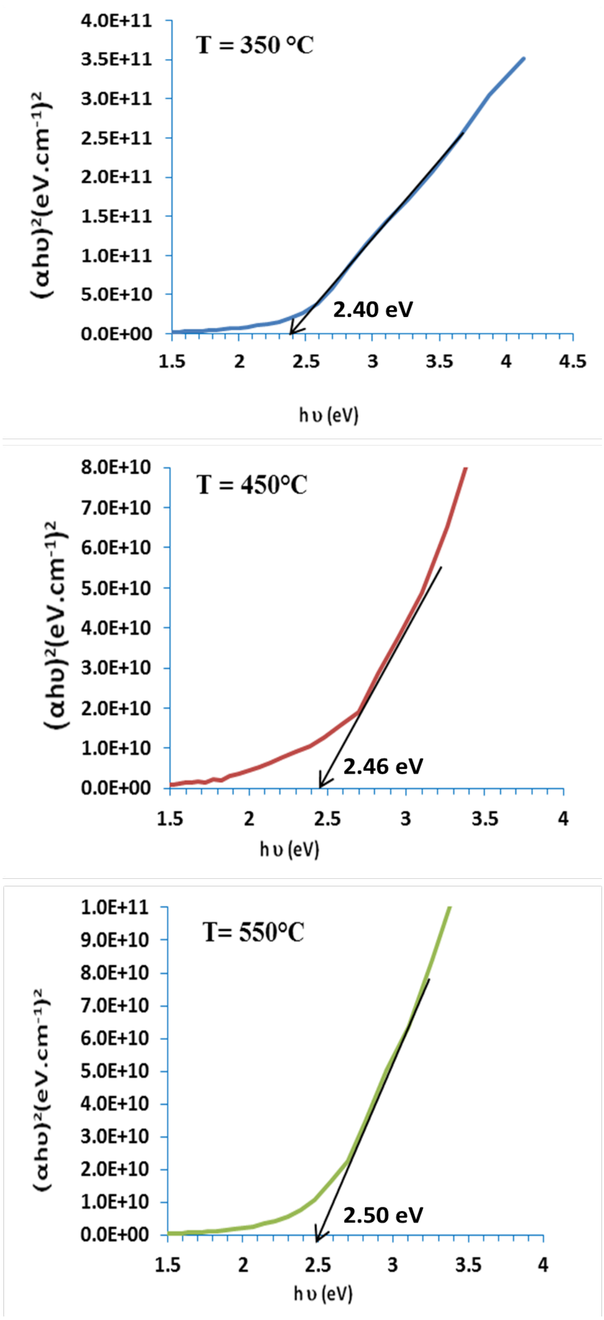

Fig. 7. Relationship between $(\alpha h\nu)^2$ and photon energy $(h\nu)$ for different substrate temperatures.

3.6. Extinction Coefficient (K_o)

Extinction Coefficient (K_o) is derived from Equation (6) [21]:

$$K_o = \frac{\alpha \lambda}{4\pi} \tag{6}$$

where λ is the wavelength of the incident beam.

Figure 8 shows the variation in extinction coefficient with changing wavelength for CdO films prepared at different substrate temperatures. The extinction coefficient decreases as the wavelength increases, exhibiting a sharp decline near λ = 500 nm. This trend is attributed to decreased absorbance at longer wavelengths, which occurs when the incident photon energy drops below the optical bandgap energy. Furthermore, by changing the substrate temperature does not alter the overall profile of the extinction coefficient curve; however, the magnitude of the extinction coefficient decreases as the substrate temperature rises.

3.7. Refractive Index (n)

Refractive Index (n) is derived from Equation (7) [22]:

$$n = \left(\frac{1+R}{1-R}\right) \pm \sqrt{\frac{4R}{(1-R)^2} - K^2} \tag{7}$$

where K is extinction coefficient.

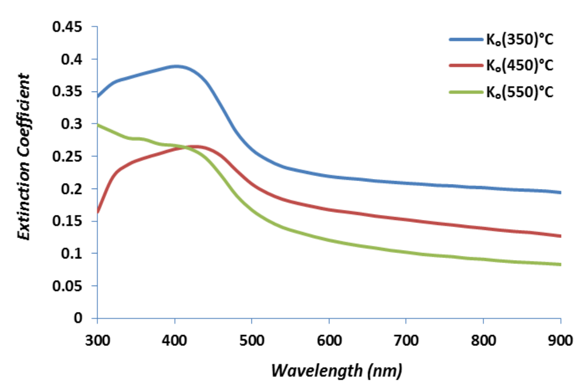

Fig. 8. Relationship between the Extinction Coefficient (K_o) with wavelength (λ) for different substrate temperatures.

Figure 9 depicts the change in refractive index values with varying wavelength for CdO films having substrate temperatures. The refractive index reaches its highest peak value in the blue/violet spectral region (450 nm), then begins to decrease with increase in the wavelength. This decrease in refractive index can be attributed to two interpretations: the effect of voids causing a reduction in density [23], and the effect of abnormal scattering, where the film shows abnormal scattering in the range of 450-900 nm, where *n* decreases with decreasing photon energy with increasing wavelength [24]. We find that the behavior of the refractive index curve does not change, but the refractive index value decreases with increasing the substrate temperature.

3.8. Dielectric Constant

The dielectric constant with real (ε_r) and imaginary (ε_i) parts has been calculated from Equations (8) and (9) [25].

$$\varepsilon_r = n^2 - K^2 \tag{8}$$

$$\varepsilon_i = 2n\,K \tag{9}$$

where ε_r is the real part and ε_i is the imaginary part of dielectric constants.

Figure 10 and Figure 11 present the wavelength dependence of the real (ε_r) and imaginary (ε_i) parts of the dielectric constant for the CdO films with varying substrate temperatures. For all films the dielectric constants increase with increasing the wavelength up to ($\lambda \approx 500nm$), and then begin to decrease.

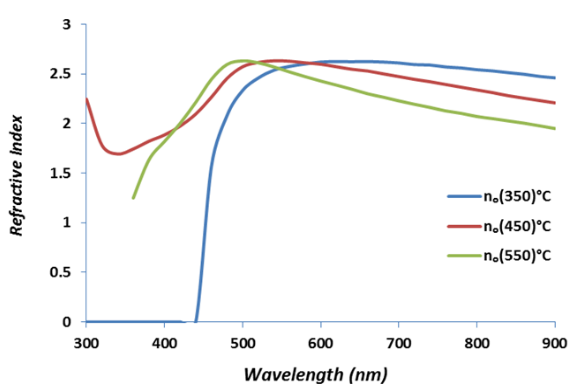

Fig. 9. Relationship between refractive index (*n*) with wavelength (λ) for different substrate temperatures.

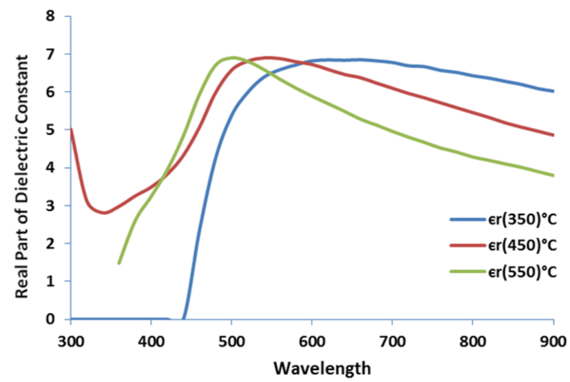

Fig. 10. Variation of real part of dielectric constant (ε_r) with wavelength (λ) for different sabstrate temperatures.

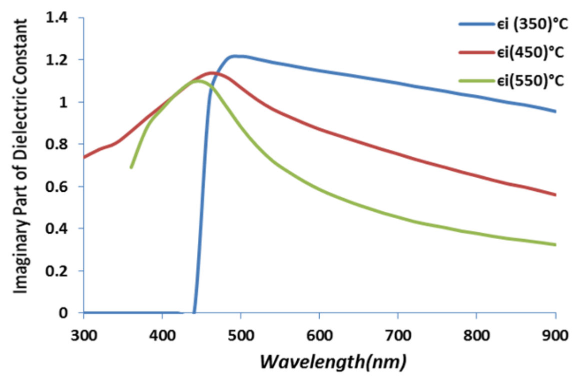

Fig. 11. Variation of imaginary part of dielectric constant (ε_i) with wavelength (λ) for different sabstrate temperatures.

4. CONCLUSIONS

The present study demonstrates that structural properties investigated by X-ray diffraction analysis confirmed the CdO films were polycrystalline and have a cubic structure, exhibiting grain sizes ranging from 198.5 to 229.8 nm. The optical properties of CdO thin films are significantly optimized by thermal treatment, achieving a high optical transmittance more than 80% for film synthesized at a substrate temperature of 550 °C. A systematic increase in the transmittance is observed at rising substrate temperatures. Furthermore, the CdO thin films exhibited a high absorbance coefficient ($\alpha > 10^4\ cm^{-1}$) within the 300-900 nm spectral range, which strongly indicates direct electronic transitions and confirms a direct optical band gap, consistent with this structural improvement, the optical energy gap (E_g) shifts toward higher energies with increasing substrate temperatures. This thermal enhancement corresponds directly to a uniform decrease in the values of the extinction coefficient, refractive index, and both the real part (ε_r) and imaginary part (ε_i) of the dielectric constants across the investigated 300-900 nm wavelength range.

5. ACKNOWLEDGMENTS

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6. ETHICAL STATEMENT

This study did not involve any human participants or live animal testing. Therefore, ethical approval was not required for this research.

7. CONFLICT OF INTEREST

The authors declare no conflict of interest.

8. FUNDING

The authors received no financial support for the research authorship and or publication of this article.

9. AUTHORSHIP CONTRIBTION STATEMENT

All authors contributed to this work. Helen J. Abdalhussain, Rasool O. Saleem and Aws A. Mahdi produced the idea and method of work. Helen J. Abdalhussain, Rasool O. Saleem conducted the practical application, calculations, and data analysis. Rasool O. Saleem, Aws A. Mahdi worked on writing the theoretical part. All authors contributed to writing the summary and conclusion.

10. DECLARATION

All authors of this study, declare that the results of this study are original. The same material is neither published nor under consideration for publication elsewhere. In case the article is accepted for publication, its copyright will be assigned to the Pakistan Academy of Sciences. All results were interpreted solely by the authors; Grammarly and Google Translate were used for English language refinement.

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