Influence of Ar gas pressure on the structural and optical properties and surface topography of Al-doped ZnO thin films sputtered by DC-Magnetron sputtering method

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Received: 6 July 2021 / Accepted: 20 September 2022 / Published online: 22 October 2022
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Abstract
In this work, Aluminum doped Zinc oxide thin films were sputtered on glass substrate by the direct current (DC) magnetron sputtering method. The influence of Ar gas pressure on the structural and optical properties was measured. The optical parameters were calculated by UV–Visible spectroscopy, the nature of transition reveals direct allowed transition for the prepared films. Also, some physical quantities such as the strength of electron–phonon interaction, dissipation factor (tanδ) in the visible region and the lattice dielectric constant were presented for these thin films. The AFM analysis extracts surface parameters of the AZO thin films that help us to quantitatively investigate the surface analysis. The band gap energy and transition index without any presumption about transition natural were calculated from Derivation Ineffective Thickness Method (DITM) and it was found that the reaction of Ar gas pressure plays an essential part in controlling the physical quantities of AZO thin films.

Keywords AL doped ZnO · Transition index · Urbach tail · Refractive index · Dielectric constant · Dissipation factor · (DITM) · The strength of electron–phonon interaction · The lattice dielectric constant

1 Introduction
Common transparent conductive oxide (TCO) thin films have been widely used in the photovoltaic application, therefor are marvelous for investigating (Kumar et al. 2018; Patel and Kim 2017; Sabeeh and Jassam 2018; Sharmin et al. 2019; Subramanyam et al. 2018; Torrisi et al. 2017; Yun et al. 2015). The zinc oxide thin films have classified in the II–VI compound semiconductors material and is composed of hexagonal wurtzite crystal structure with \(a = 3.25 \text{ Å} \) and \(c = 5.12 \text{ Å} \) where each Zn atoms is tetrahedrally coordinated to four O atoms, that the Zn d-electrons hybridize with the O p-electron, layers occupied by Zinc atom thin films. The interesting features of ZnO such as wide band gap (3.35–3.37 eV),
its large exciton binding energy of 60 meV allow a lower threshold for optical pumping at room temperature, high transmittance property alternate with layer occupied by oxygen atom, and utilized as the transparent conductive oxide (TCO) in the visible region, piezoelectric properties, high coupling factor and room temperature ferromagnetism features have attracted much regard for studying. In addition to the well-known advantages of undoped ZnO films, due to its other features such as high resistivity, instability, and low conductivity, it is not preferred, also for reducing the resistivity and modifying the electrical characteristics, the ZnO film is doped by the elements of group III such as Al, Ba, Ga, and In, it is become conducting nature owing to some free electrons was incorporated into ZnO films that these materials have been widely used for some devices such as gas sensors, transparent thin film transistors, solar cells, displays, electrochromic devices, and photocatalysis, etc. (Akkaya Arier and Uysal, 2016; Kim and Kim, 2011; Li et al. 2015; Samanta et al. 2010; Sharmin et al. 2019; Tseng 2018; Wen et al. 2019; Xia et al. 2017; Zaiour et al. 2019). These well-known advantages of ZnO have made it for a wide application, such as, enhanced photoresponse of self-powered perovskite photodetector based on ZnO nanoparticles decorated CsPbBr3 films (Li et al. 2017); efficiency enhancement of ZnO/Cu2O solar cells with well oriented and micrometer grain sized Cu2O films (Zang 2018); NH4Cl-Modified ZnO for High-Performance CsPbIBr2 Perovskite Solar Cells via Low-Temperature Process (Wang et al. 2020a, b); challenges and strategies relating to device function layers and their integration toward high-performance inorganic perovskite solar cells (Wang et al. 2020a, b).

Among these TCO materials, the Aluminum doped on ZnO (AZO) thin films semiconductors are reported because of its attractive characteristics such as low resistivity, high optical transparency in the visible region, low cost, high electrical conductivity, direct transition and strong absorbance, etc. (Chai Lin et al. 2015; Evcimen Duygulu et al., 2014; Juhnevica et al., 2015; Lee et al. 2009; Rahmance et al. 2010; Saravanakumar et al., 2014; Subramanyan et al. 2018; Wang et al. 2017; Wen et al. 2019).

In order to obtain high-quality AZO films, a variety of the deposition techniques may be used such as DC-magnetron sputtering, chemical bath deposition (CBD), sol–gel technique and pulsed laser deposition, etc. (Islam et al. 2019; Kaid and Ashour 2007; Kumar et al. 2018; Samanta et al. 2010; Sharmin et al., 2019; Sengupta et al. 2012; Terasako et al. 2007; Tseng 2018; Valentini et al. 1993; Wang et al. 2017; Wen et al. 2019).

In this paper, the AZO thin films that were deposited by DC-magnetron sputtering technique and the effects of variation of Ar gas pressure on the prepared thin films properties were investigated. This method was employed owning to its flexibility and ability for deposit films under vacuum, stoichiometry and large-area deposition, etc., (Chi et al. 2011; Fang et al. 2002a, b). In this technique the physical characteristics of AZO films such as composition, thickness and crystal phase, can be affected by the various sputtering parameters such as argon gas pressure, time of sputtering, and different concentration, etc. In this work the variation of Ar gas pressure plays an essential part for researching of the structural and optical properties of AZO films. Derivation of ineffective thickness method (DITM) was employed to extract the optical bandgap energy and the type of optical transitions in nanostructured semiconductors. In this regard, it is shown that the optical band gap is independent from thickness measuring and only requires the measurement of the absorbance spectrum of the product (Ghobadi 2016).

Numerical factors for sample surfaces as used in material surface drawings are the means of communication between fabrication and functional performance. These factors are not only used as a criterion for production and surface parameter description, but also particularly in the case of 3D factors to predict physical properties. This initial numerical
parameter as Root-mean-square deviation (S_q), Skewness of topography height distribution (S_sk), Kurtosis of topography height distribution (S_ku), and inclination angles determined from the AFM images help researchers to investigate the surface analysis with numerical data (Molamohammadi et al. 2015a, b; Talu et al. 2016).

2 Experimental details

In this paper, AZO thin films were sputtered by DC-magnetron sputtering method. A disk of AZO (purity of 99.9%) with a diameter of 100 mm was utilized as a cathode. For this study, all of films sputtered with the same condition including I = 175 mA (incoming current) and V = 300 v (incoming voltage).

Before a deposition, for removing any embedded particles on the surface of substrates, the glass substrates were completely cleaned in a bath of ultrasonic with acetone and ethyl alcohol for 18 min. Then they were rinsed in distilled water and dry in hot air before they were located into the chamber. The system of vacuum has two pumps, a mechanical rotary and the turbo pumps are used for low and high vacuum in the deposition chamber, respectively. The substrates putting up in the chamber, the chamber was vacuumed to a base pressure of $2.8 \times 10^{-5}$ mbar prior to deposition then the pressure was enhanced and high purity Argon gas (99.99%) was introduced through mass flow controller. In this study the AZO films sputtered in various Ar gas pressure which it was changed from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar.

The absorption and transmittance, XRD, FESEM and AFM measurements were evaluated with Ultra Violet Visible double beam spectrophotometer (PerkinElmer, Lambda 25-USA), diffractometer (Unisantis-XMD-300, CuK, Germany), (Tscan,Mira III,Czech Republic) and (Bruker, Icon, USA), respectively.

3 Results and discussion

3.1 Structural study

3.1.1 XRD analysis

Mainly, different structural features of AZO thin films were evaluated by X-ray diffraction (Caglar 2008; Sabeeh and Jassam 2018; Sarvankumar et al. 2014). Figure 1 illustrates the patterns of XRD for AZO films sputtered on a glass substrate by various Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar (Patel and Kim 2017; Tseng 2018). The patterns of XRD reveal only the same strong diffraction peak of (002) plane at $2\theta$ location of 34.4° that are very near to the wurtzite hexagonal ZnO crystal (34.43), also no Al2O3 phase was detected, this may be because of atoms of Al replace into the hexagonal lattice of Zinc or atoms of Al separated to the non-crystalline region in the grain boundary. (Akkaya Arier and Uysal, 2016; Dejam et al. 2019; Evcimen Duygulu et al., 2014; Kim and Kim, 2011; Lee et al. 2009; Shokri and Dejam 2019; Rahmane et al.2010; Zhang et al.2016).

To obtain the detailed structural data, the mean crystallite size for (002) plane (the highest intensity peak) was calculated from the Debye Scherrer equation (Kumar et al. 2018; Kim and Kim, 2011; Rahmane et al.2010; Sharmin et al. 2019; Saravanakumar et al. 2014).
where $k$ is the particle-shape factor (0.9), $\lambda$ stands for the X-ray wavelength (K-Alpha 1: 1.5405 Å), $\beta$ is the line broadening at half the maximum intensity (FWHM) and $\theta$ is the Bragg angle of the diffraction peak with a variation of Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar, in this study with enhancing Ar gas pressure, the mean crystallite size that indicate the longitude of the crystal in the orientation of the d-spacing increased from 10.2 to 19.9 nm and the FWHM of (002) peak decreased from 0.8160 to 0.4171 with enhancing Ar gas pressure which means the peak become more intense and sharper up to $3 \times 10^{-2}$ mbar, reducing of FWHM indicates the crystallinity of the AZO thin films is improved and the mean crystallite size becomes more extensive as the Ar gas pressure increased (See Fig. 2), the calculated results are listed in Table 1 (Chi et al. 2011; Kumar et al. 2018; Sengupta et al. 2012; Wang et al. 2017).

We can quantify the defects in the AZO thin films for the highest intensity peak (002) by computing the microstrain and dislocation density by below equations:

$$\varepsilon = \frac{\cot \theta}{4}$$  \hspace{1cm} (2)

$$\delta = \frac{1}{D^2}$$ \hspace{1cm} (3)

where $\varepsilon$ is the micro strain and $\delta$ is the dislocation density, the calculated results are low and with increasing Ar gas pressure are decreased which shows the presence of minimum defects in these films (Kumar et al. 2018), the examined results are listed in Table 1. For calculating the number of crystallite (N) per unit area, we can use the below equation:

$$N = \frac{l}{D^3}$$ \hspace{1cm} (4)

Fig. 1 Patterns of XRD for AZO films with changing of Ar gas pressure
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where \( t \) is the thickness of films and \( D \) is the mean crystallite size, with improving Ar gas pressure the number of crystallites decreased that is against the variation of the mean crystallite size. It is because with increasing the nucleation and crystal growth on the surface, the number of defects is also decreased with increasing of Ar gas so the number of atoms per unit area decreases (Kumar et al. 2018), the results are listed in Table 1.

### Table 1

| Ar gas pressure (10\(^{-2}\) mbar) | Peak position (2\(\theta\)) (°) | \( \beta \) (°) | D (nm) | d (Å) | \( \varepsilon \) | \( \delta \times 10^{16} \) | \( N \times 10^{16} \) |
|-----------------------------------|-------------------------------|----------------|--------|------|-------------|---------------------|------------------------|
| 1                                 | 34.40                         | 0.8160         | 10.2   | 2.6041 | 0.0114      | 0.961               | 34.3                   |
| 2                                 | 34.394                       | 0.6048         | 13.7   | 2.6053 | 0.0084      | 0.532               | 17.1                   |
| 3                                 | 34.388                       | 0.4171         | 19.9   | 2.6080 | 0.0058      | 0.252               | 7.2                    |

3.1.2 The FESEM analysis

Surface morphology plays an important role in investigating the change of different characteristics for any metal oxide thin films, in this paper, the FESEM study is used to realize the AZO film morphology, growth of grain and the estimation of nanoparticles size on surface of films with changing of Ar gas pressure (Kumar et al. 2018). Figure 3(a–c (200 nm)) and (d–f (1 µm)) indicate the investigation of FESEM images for various Ar gas pressure from 1 \( \times 10^{-2} \) mbar to 3 \( \times 10^{-2} \) mbar, as shown these images the mean approximate size of nanoparticles are increased with enhancing of Ar gas pressure from 35.98 to 72.93 nm, this is because When the Ar gas pressure increases further, the ion density becomes large enough for crystal growth also the thickness of films increases. Figure 3g shows the EDS spectrum of 3 \( \times 10^{-2} \) mbar sputtered AZO thin film, EDS spectrum exhibits Zn, O and Al elements. The observed values for Zn,
O and Al were 82, 15 and 3%, respectively. We conclude from the EDS result that the small quantity of dopant (Aluminum) element is successfully incorporated and substitution into ZnO lattice.

Fig. 3 The FESEM images for AZO thin films with changing of Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar. g EDS spectrum of $3 \times 10^{-2}$ mbar sputtered AZO thin film.
3.1.3 Atomic force microscopy (AFM)

The 2-D and 3-D images of surface topography for the AZO thin films were studied by the AFM analysis as shown in Fig. 4, this analysis helps us to investigate the surface characteristics with numerical data. We know this tool converts surface data into illustrative images. Calculating the three-dimensional (3-D) surface data is crucial to control the quality of thin films to engineering the surface behavior for improved optical and electrical properties; any variation between peaks and valleys is more suitable for providing information about the 3-D surface topography. Experimental information extracted from the surface description of the AFM images (according to ISO 25178–2: 2012) for the three samples with different Ar gas pressure (Molamohammadi et al. 2015a, b; Talu et al. 2016) The results of Root Mean Square (RMS) roughness parameter of films with Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar are obtained to be 22.06 to 30.18 nm, it is found that by increasing Ar gas pressure, the surface becomes rough that The rise in the RMS roughness with the formation of 3D structures is consistent with a rougher texture caused by an increase of the crystallite size as indicated by the XRD measurements. The root-mean-square deviation ($S_q$), skewness of topography height distribution ($S_{sk}$), kurtosis of topography height distribution ($S_{ku}$) calculated from the AFM images are presented in Table 2. $S_z$ ($S_z = S_p + S_v$) shows the distance between the peak and the valley which has the highest value for AZO thin films.

Fig. 4 The 3-D and 2-D images of surface topography for AZO films at various Ar gas pressure
with pressure of $2 \times 10^{-2}$ mbar so this result indicates this film has the most disorder (Table 2).

### 3.2 Optical measurements

#### 3.2.1 Determination of optical band gap and the gap type (m)

The optical transparency of film has an essential role in the characteristics of TCO thin films (Chi et al. 2011; Ghobadi 2016; Kim and Kim, 2011; Lee et al. 2009; Xia et al. 2017). In this work, the spectrums of optical transmittance were examined by UV–Visible spectroscopy in wavelengths range from 190 to 1100 nm for various Ar gas pressure. Figure 5 gives out the spectrum of average optical transmittance of the AZO films in the visible region by enhancing of Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar decreases from 87.5% to 73. %. also, the film with the least Ar gas pressure exhibits the maximum average transmittance of 87.5%, it is because of the increase in thickness of films from 365 to 570 nm. The calculated parameters are listed in Table 3. Figure 6 shows the spectrum of absorption for different Ar gas pressure, it demonstrates the absorption decreases with enhancing wavelength with sharp absorption edge (Rahmane et al. 2010; Samanta et al. 2010; Sharmin et al. 2019; Wang et al. 2017).

### Table 2 The AFM information of AZO thin films

| Ar gas pressure (mbar) | $S_q$ (nm) | $S_{sk}$ | $S_{ku}$ | $S_p$ (nm) | $S_v$ (nm) | $S_z$ (nm) | $S_a$ (nm) |
|-----------------------|------------|----------|----------|------------|------------|------------|------------|
| $1 \times 10^{-2}$    | 22.06      | 0.4563   | 4.064    | 92.46      | 64.78      | 157.2      | 16.33      |
| $2 \times 10^{-2}$    | 27.83      | 0.6032   | 4.324    | 107.8      | 111.2      | 219        | 20.63      |
| $3 \times 10^{-2}$    | 30.18      | 0.3925   | 2.955    | 101.6      | 97.63      | 199.2      | 24.02      |

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Fig. 5 The Optical transmittance of films with the Ar gas pressure changed from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar.
In the present study, for calculating the band gap energy ($E_g$), we use two techniques, the ineffective thickness method (ITM) and Derivation of ineffective thickness method (DITM) in semiconductors which are presented with Eqs. 5 and 8 respectively, for examining band gap we only need to examine the absorption instead of coefficient absorption, without requiring thickness of films, with following equations (Ghobadi 2016):

$$A(\nu)h\nu = K\left(h\nu - E_{gap}\right)^m$$

(5)

$$\ln(A(\nu)E) = \ln\left(K\left(h\nu - E_{gap}\right)^m\right)$$

(6)

$$\ln A((\nu)E) = mLn(K) + mLn(E - E_{gap})$$

(7)

And derivation of Eq. (7) gives:

$$\frac{d\{\ln[A(\nu)E]\}}{d(E)} = \frac{m}{E - E_{gap}}$$

(8)

That $A(\nu)$ is the absorption and $K$ is a constant, $m$ is the transition index where values of $m$ depend on the nature of the optical transition type that can have various measurements such as

| Ar gas pressure (mbar) | T (%) | $E_{gap}^{DITM}$ (eV) | m (DITM) | $E_{tail}$ (eV) | $\sigma \times 10^{-2}$ | $E_{el-ph}$ |
|-----------------------|------|---------------------|----------|----------------|-----------------|-------------|
| $1 \times 10^{-2}$    | 87.5 | 3.32                | 0.837    | 0.1502         | 17.211          | 3.873       |
| $2 \times 10^{-2}$    | 79   | 3.30                | 0.640    | 0.1617         | 15.987          | 4.169       |
| $3 \times 10^{-2}$    | 73   | 3.23                | 0.699    | 0.1500         | 17.234          | 3.868       |

Fig. 6 The spectrum of Absorption for films with changing of the Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar.
m = 1/2 for allowed direct, m = 3/2 for forbidden direct, m = 2 for allowed indirect and m = 3 for forbidden indirect optical transition, respectively. If the graphs of $\frac{d(Ln(A\nu E))}{dE}$ versus E are drawn, there would be a discontinuity at $E = E_{gap}$. In this study, in DITM, the evaluated band gap energy for various Ar gas pressures are from 3.23 to 3.32 eV, the increasing of band gap energy can be owing to a corresponding shift in the mean crystallite size which the mean crystallite size affects the optical characteristics obtaining higher band gap energy for thin films with smaller the mean crystallite size. Since, when the mean crystallite size is small there are less atoms and also few atomic orbitals to overlap, therefore the number of orbitals (both bonding and antibonding) are decreased resulting in higher band gap energy. Figure 7 and Fig. 8 indicate the optical band gap for ITM and DITM respectively, where the values of optical band gap were reduced by enhancing Ar gas pressure (Dejam et al. 2019; Ghobadi 2016; Ghobadi et al 2021; Kumar et al. 2018; Kim and Kim, 2011). Table 3 indicates the obtained results.

By utilizing the optical band gap energy values, examined from Fig. 8, the transition index (m) can be evaluated by the slope of the linear part of $Ln(A\nu E)$ versus $Ln(E - E_{gap})$, the graphs were indicated in Fig. 9. The calculated measurements are from 0.640 to 0.837 that they are almost about $\frac{3}{2}$ where shows direct allowed transition with changing of Ar gas pressure (Chai Lin et al.2015; Ghobadi 2016; Ghobadi et al 2021; Zhang et al.2016), the calculated results listed in Table 3.

### 3.2.2 Urbach energy: examining the crystalline quality

Urbach energy is the width of the tail of localized states as for the optical transition between localized tail states in the near to bands the Urbach tail can be obtained from the slope of straight plotting $Ln A(\nu)$ versus the incident photon energy (Belahssen et al. 2014; Dejam et al. 2019; Franz 1953; Ghobadi 2016; Ghobadi et al 2021; Hssanien and Akl 2016; Shokri and Dejam 2019).

$$LnA(\nu) = LnA_0 + \frac{hv}{E_{tail}}$$

That $A_0$ is a constant, $hv$ is the incident photon energy and $E_{tail}$ exhibits Urbach energy. Figure 10 shows the calculated of Urbach energy that the results are listed in Table 3.
The values of Urbach tail initially increased and then decreased with enhancing of Ar gas pressure. Equation 9 implies that logarithm of \( A(v) \) plotted as a function of \( E_{\text{tail}} \) can be approximated by a straight line in energies just below the fundamental absorption edge which is called the converging point. The AFM results confirm that, the \( S_z \) parameter, the distance between the peak and the valley, by increasing of Ar gas pressure also initially increased and then decreased which has the highest value for AZO thin films at pressure \( 2 \times 10^{-2} \) mbar. Indeed, decreasing of the Urbach tail can be considered a smaller number of defects where it causes the structural characteristics become more orderly in consistent with AFM results. The calculated values have been listed in Table 3 (Belahssen et al. 2014; Dejam et al. 2019; Franz 1953; Ghobadi et al. 2021; Hssanien and Akl 2016; Zhang et al. 2016). Figure 11 demonstrates the changing of band gap and Urbach tail as a function of...
Ar gas pressure that with enhancing of Ar gas pressure, the band gap is decreased and the Urbach tail initially increased and then decreased.

\( \sigma \) is the steepness parameter that characterizing the broadening of the optical absorption edges owing to electron–phonon interaction, was examined by the following equation (Bashar et al. 2020)

\[
\sigma = \frac{k_B T}{E_u}
\]  

(10)

where \( k_B \) is the Boltzmann constant and \( T \) is the absolute temperature in Kelvin, therefor the measurements of the strength of the electron–phonon interaction can be determined by the following equation:
3.2.3 Determination of refractive index and dielectric constant

3.2.3.1 Refractive index and dielectric constant in the visible region. Some optical features such as Refractive index (n) and extinction coefficient (k) play an essential part for any TCO film. The refractive index (n) was examined from below equation:

\[ E_{el-ph} = \frac{2}{3} \]

Figure 12 shows with increasing in the Urbach energy values, the steepness parameter and the strength of electron–phonon interaction decreases and increases respectively, the results indicate with increasing Ar gas pressure the localized states initially increase and then decrease which may be resulted either from an increase or decrease in the vacancies or dislocation defects of the AZO thin films (Bashar et al. 2020) The measurements values are listed in Table 3.
where R is the reflectance and k is the extinction coefficient measurements, Fig. 13 indicate the difference of the refractive index versus the wavelength where the spectral range of n is reduces with enhancing wavelength because of the increasing of transmittance that may be because of light scattering. The average measurements of refractive index examined in the Visible region; they are in the range of 1.612 to 2.001 that the refractive index with enhancing of Ar gas pressure owing to the increasing of the scattering of photon light is increased (Abdullah 2013; Dejam et al. 2019; Kumar et al. 2018). The examined data are shown in Table 4.

The extinction coefficient (k) measurement can be calculated from the below equation:

$$ k = \frac{\alpha(\lambda)\lambda}{4} $$

(13)

That $\alpha(\lambda)$ is the absorption coefficient of AZO film. Figure 14 indicates the variation of the extinction coefficient versus wavelength that the calculated results for various of Ar gas pressure is low for all wavelength range. It may be due to the improvement of crystalline structure, smooth surface, reducing of defects and the high transmittance of the sputtered AZO films (Abdullah 2013; Dejam et al. 2019; Kumar et al. 2018), also the calculated measurements are listed in Table 4.

The complex dielectric constant $\varepsilon = \varepsilon_1 + i\varepsilon_2$ can define the optical characteristics of any solid material that the dielectric feature such as real part ($\varepsilon_1$) and imaginary part ($\varepsilon_2$) of dielectric constants were calculated from the below equations:

$$ \varepsilon_1 = n^2 - k^2 $$

(14)

$$ \varepsilon_2 = 2nk $$

(15)

Figures 15 and 16 indicate the difference of the real and imaginary part of dielectric constants versus incident photon energy for AZO films by different Ar gas pressure. The

\[ n = \frac{(1 + R)}{(1 - R)} + \sqrt{\frac{4R}{(1 - R)^2}} - k^2 \]

(12)

Fig. 13 the difference of the refractive index vs the wavelength with increasing Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar.
Table 4  The information of the average of refractive index in the visible region ($n$), the average of extinction coefficient in the visible region ($k$), the real part of dielectric constant ($\varepsilon_1$), the imaginary part of dielectric constant ($\varepsilon_2$), dissipation factor ($\tan\delta$), the ratio of the free charge carrier concentration to the effective mass ($\frac{N}{m^*}$), the lattice dielectric constant ($\varepsilon_L$), refractive index ($n_{\text{edge of absorption}}$) and dielectric constant ($\varepsilon_{\text{edge of absorption}}$), for AZO films with changing of Ar gas pressure

| Ar gas pressure (mbar) | $n$   | $k$   | $\varepsilon_1$ | $\varepsilon_2$ | $\tan\delta$ | $N/m^* \times (10^{20} \text{ m}^{-3} \text{ kg}^{-1})$ | $\varepsilon_L$ | $n_{\text{edge of absorption}}$ | $\varepsilon_{\text{edge of absorption}}$ |
|-----------------------|-------|-------|------------------|------------------|--------------|-------------------------------------------------|----------------|----------------------------------|-----------------------------------|
| $1 \times 10^{-2}$    | 1.612 | 0.007 | 2.5984           | 0.025            | 0.0096       | 1.227                                           | 5.5366         | 2.31                            | 5.36                              |
| $2 \times 10^{-2}$    | 1.841 | 0.010 | 3.3891           | 0.037            | 0.010        | 1.104                                           | 6.6226         | 2.32                            | 5.38                              |
| $3 \times 10^{-2}$    | 2.001 | 0.013 | 4.0038           | 0.053            | 0.013        | 1.229                                           | 8.5195         | 2.33                            | 5.46                              |
dielectric constants measurement is most appropriate for TCO thin films that the calculated results for this study are low and the calculated real part ($\varepsilon_1$) were higher than the imaginary part ($\varepsilon_2$) measurements for these films (Abdullah 2013; Dejam et al. 2019; Kumar et al. 2018). The results summarized in Table 4.

Dissipation factor ($\tan \delta$) is an optical value which is dependent on real and imaginary part of dielectric constants. The measurements of dissipation factor of films determine by the below equation:

$$\tan \delta = \frac{\varepsilon_2}{\varepsilon_1}$$  (16)

---

**Fig. 14** the variation of the extinction coefficient versus the wavelength with increasing Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar.

**Fig. 15** the variation of the real part of dielectric constant vs incident photon energy with increasing Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar.
where $\varepsilon_2$ is the imaginary part of dielectric constant and $\varepsilon_1$ is the real part of dielectric constant, Fig. 17 indicates the variation of $\tan \delta$ versus incident photon energy for AZO films with various Ar gas pressure that calculated results are low, it is can be due to the high optical transmittance and it minimizes the waste of electrical energy as heat which with increasing Ar gas pressure increases (Abdullah 2013; Dejam et al. 2019; Kumar et al. 2018), the results listed in Table 4.

The relation between the lattice dielectric constant ($\varepsilon_L$) and the refractive index $n$ can be described with the following equation:

$$\varepsilon_1 = n^2 - k^2 = \varepsilon_L - \frac{e^2 N}{4\pi^2 c^2 \varepsilon_0 m^*}$$

(17)

where $\varepsilon_1$, $e$, $c$, $\frac{N}{m^*}$, $\varepsilon_L$ and $\varepsilon_0$ are the real part of dielectric constant, the electron charge, the speed of light, the ratio of the free charge carrier concentration to the effective mass, the

![Graph of dielectric constant vs. photon energy](image1)

**Fig. 16**  the variation of the imaginary part of dielectric constant vs incident photon energy with increasing Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar.

![Graph of tan δ vs. photon energy](image2)

**Fig. 17**  the variation of dissipation factor versus incident photon energy with increasing Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar.
lattice dielectric constant and the permittivity of free space where the slope and vertical-axis intercept of the plots of $\varepsilon_1$ versus $\lambda^2$ results in $\frac{N}{m^2}$ and $\varepsilon_L$, respectively (see Fig. 18). The results of $\frac{N}{m^2}$ has the most value for AZO thin film with pressure of $3 \times 10^{-2}$ mbar localized state is decreased and it causes to transformation localized carrier concentration to the free charge carrier concentration also the values of $\varepsilon_L$ with enhancing of Ar gas pressure is increased, the results are listed in Table 4. (Abdullah 2013; Dejam et al. 2019).

3.2.3.2 Refractive index and dielectric constant at the absorption of edge

To attain more intelligence into the optical ability of the thin films, one should try to measure some other optical parameter such as refractive index ($n$) and dielectric constant ($\varepsilon$) at the absorption of edge that can be acquired using equation, introduced by Dimitrov and Sakaa (1996a, b) and Ghabadi et al. (2021):

\[
\frac{n^2 - 1}{n^2 + 2} = 1 - \left( \frac{E_g}{20} \right)^{\frac{1}{2}}
\]

(18)

\[
\varepsilon = n^2
\]

(19)

The calculated refractive index and the dielectric constant at different Ar gas pressure from $1 \times 10^{-2}$ mbar to $3 \times 10^{-2}$ mbar is from 2.31 to 2.33 and 5.36 to 5.46 respectively that with increasing of Ar gas pressure are increased, the difference trends of these results and optical band gap are opposite, the results summarized in Table 4 (Dimitrov and Sakaa 1996a, b; Ghabadi et al. 2021).

4 Conclusions

AZO thin films were synthesized onto glass substrates using DC magnetron sputtering. The influence of the Ar gas pressure in the range of $1 \times 10^{-2}$ to $3 \times 10^{-2}$ mbar on the structural and optical characteristics of the thin films were investigated. X-ray diffraction as a function of the sputtering pressure revealed a preferential orientation in the (002) plane,
additionally, the mean crystallite size tends to increase with increasing of Ar gas pressure as FESEM analyze confirm these results. The DITM method was reported to determine the band gap energy and transition index, the band gap energy with enhancing Ar gas pressure was reduced and the obtained results indicate AZO film is a semiconductor with the direct allowed transition, all AZO thin films presented a high optical transmittance (above 73%) in the visible region. The AFM analysis shows the RMS roughness parameter with enhancing of Ar gas pressure becomes rough and the distance between peaks and valley initially increases and then decreases that the AZO film with Ar gas pressure of $3 \times 10^{-2}$ mbar has the lowest disorder.

Acknowledgements Not applicable.

Authors’ contributions Not applicable.

Funding Not applicable.

Availability of data and material Not applicable.

Code availability Not applicable.

Declarations

Conflicts of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

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