




Photovoltaic performance in CIGS solar cells: effects of using Mg- and Al-doped ZnO thin-film layers as alternative TCO and front contact layer

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Received: 12 June 2024

Accepted: 11 October 2024

Published online:
26 October 2024

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ABSTRACT

In this study, we aimed to improve the electrical, optical, and structural properties of ZnO-based layered CIGS solar cells by doping different ratios of Al and Mg. Al-doped ZnO and Mg-doped ZnO thin films were prepared using sol-gel spin-coating technique. The doping rates were 1%, 3%, and 5% for both materials. Structural, surface, and optical properties were analyzed using XRD, FESEM, AFM, and UV-Vis spectroscopy. The results revealed that Al doping increased surface roughness, while Mg doping decreased it. Al doping reduced the band gap to 3.05 eV, enhancing conductivity, whereas Mg doping increased the band gap to 3.52 eV, improving optical transparency. The ideal combination of AZ5/MZ1 layers achieved a significant efficiency increase of 26.46% in CIGS solar cells, compared to 23.24% with undoped ZnO layers. Additionally, the surface roughness values were found to be 224.3 nm for AZ5 and 69.7 nm for MZ5. This study demonstrates the potential of Al and Mg-co-doped ZnO thin films to improve the performance of solar cells and other optoelectronic devices, offering promising developments in renewable energy technologies.

1 Introduction

When the materials that make up the solar cell structure are examined, it is observed that metal oxides play an active role [1–3]. Zinc oxide (ZnO) has become a widely used material in solar cells due to its superior properties, such as wide band gap (approximately

3.37 eV) and high thermal stability [4–7]. In order to maximize the potential of ZnO in electronic and optoelectronic applications, the electrical, optical, and structural properties of the material must be modified to optimize it for the area in which it will be used. In this direction, doping ZnO with various elements is widely used as a method to improve the band

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structure, carrier density, and crystal structure of the material [8–11]. By doping elements such as aluminum (Al) and magnesium (Mg) into ZnO, the band gap can significantly improve the performance of solar cells by changing the electronic structures and optical properties of the material [12–16]. Al-doped ZnO (AZO) attracts attention as a conductive oxide with low resistivity and high transparency [17, 18]. When used as a transparent conductive layer in solar cells, AZO contributes positively to cell efficiency by increasing the transmission of light and reducing electrical losses [19–21]. Al doping increases the n-type conductivity of ZnO and therefore increases the carrier density. However, Mg doping widens the band gap of ZnO and improves its optical transparency. Mg-doped ZnO (MZO) offers higher optical transmittance, especially in the ultraviolet (UV) region, thus increasing the light absorption of solar cells [22–24]. When the studies in the literature are examined; Xian et al. synthesized and characterized 2% and 4% Ag-doped ZnO thin films on glass substrates by sol–gel method. They also analyzed the solar cell simulation with Rigorous Coupled Wave Theory (RCWT). When the test results are examined, the efficiency of 4% Ag-doped ZnO thin films increased by 2.47% compared to undoped ZnO. Bedia et al. identified AZO thin films on glass substrates by spray pyrolysis method and performed structural and optical analyses. The effect of the use of AZO thin films as contacts in HIT solar cells on solar cell efficiency was investigated. In the analysis performed with the SCAPs program, the efficiency of the reference solar cell without AZO was 16.86%, while the efficiency of the solar cell with AZO contact was 20.12%. Alam et al. deposited undoped ZnO, Al-doped ZnO, and Cu-doped ZnO thin films on FTO substrate using RF sputtering technique. After characterizing these materials, they monitored the measurements made on solar cells under both dark and illuminated conditions with Autosys DC3500 solar simulator and Keithley 2400 sourcemeter on the computer interface. They found that doping with Al or Cu improves the performance of ZnO-based solar cells. Khan et al. made a comparative analysis to improve the efficiency of perovskite solar cells using undoped ZnO and AZO in the transport layer. As a result of these experiments they conducted with SCAPS-1D simulation, they observed an increase of approximately 7% in the efficiency of the cell belonging to the layer where AZO was used. Paul et al. have investigated the performance change of CZTS solar cells with different concentrations of Mg

doping instead of intrinsic ZnO (i-ZnO). The measurements made with these materials used in the window layer were simulated by Silvaco TCAD at AM 1.5G illumination. It was observed that 6% Mg-doped ZnO had the highest efficiency. In the studies conducted in the literature, the effect of ZnO structure on solar cells has generally been investigated by doping it with Al, Mg, or another material. When compared to the studies where Al- and Mg-doped ZnO thin films were applied in solar cells, it has not been found that these two elements were doped with ZnO at different rates and compared. In our study, in order to examine the effect of the concentration of the doping materials Al and Mg, the effects on solar cell efficiency were analyzed by doping them at different rates. In addition, the simulation methods generally differ from the studies in the literature [25–29]. In this study, the effect of ZnO (AZO/MZO) material, in which Al and Mg elements are doped together, on the performance of CIGS solar cells was comprehensively examined. The effects of Al and Mg dopants on the microstructure, electrical properties, and optical behavior of ZnO were analyzed in detail. The results obtained will provide important information for increasing the efficiency of ZnO-based layered CIGS solar cells. Thus, the potential for use of Al- and Mg-doped ZnO in CIGS solar cells was evaluated and more efficient and cost-effective solutions can be offered in renewable energy technologies. In this context, it is anticipated that the results of the study will make significant contributions to improving the performance of solar cells and other optoelectronic devices.

2 Material and method

Three different solutions were prepared to produce undoped and doped ZnO thin films and these solutions were mixed in various amounts to form the doped structure. Firstly, for the preparation of ZnO solution, $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ (zinc acetate dihydrate) was dissolved in 2-methoxyethanol and monoethanolamine (MEA) was added dropwise to the solution as a stabilizer. The solution was stirred for 150 min at 70°C and finally the prepared solution was left at room temperature for two days. For Al doping, $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (aluminum nitrate nonahydrate) was dissolved in 2-methoxyethanol and MEA was added to the solution as a stabilizer. The solution was stirred for 180 min at room temperature and the final solution

was left at room temperature for two days. In Mg doping, $MgCl_2 \cdot 6H_2O$ (magnesium chloride hexahydrate) was dissolved in 2-methoxyethanol in the same way and MEA was added to the solution as a stabilizer. The solution was stirred for 90 min at $60^\circ C$ and the final solution was left for one day. Solutions were prepared as 0.5 M. In order to obtain Al- and Mg-doped ZnO thin films, the ZnO solution was mixed with 1%, 3%, and 5% Al-doped solution by volume. The resulting structures are named as in Table 1.

The series were coated with sol-gel spin-coating technique on the ITO substrates at 3000-rpm rotation speed, 30 s, and 9 layers. To complete crystallization, the coated samples were annealed in air at $500^\circ C$ and thin films were obtained. The structural, surface, and optical characterizations of the obtained thin films were examined by X-ray Diffractometer (XRD), Field Emission Scanning Electron Microscope (FESEM), Atomic Force Microscope (AFM), and UV-Vis Spectrophotometer, respectively. The solar cell design was simulated in the "OghmaNano" program. The process

flow diagram for the production of Al/Mg-doped ZnO thin films and their use as layers in CIGS solar cells is shown in Figure 1

3 Results and discussion

In this study, PANALYTICAL Impregnating X-Ray Diffraction (XRD) device was used to investigate the structural properties of doped and undoped thin films produced with different parameters. The operating conditions of the device were 45 kV voltage and 40 mA current. The scanning speed of 2 degrees/minute was selected, a $CuK\alpha$ beam with a wavelength of 1.5406 \AA was used and the samples were examined at 2θ from 30° to 70° . The obtained spectra were compared with the cards 98-003-1052 (ICDD number) for hexagonal structured ZnO and 98-064-2712 for cubic structured MgO. In the spectrum of Al-doped ZnO thin films, no peak of Al was observed, so the ICDD cards of Al were not analyzed. Comparative spectra of Al- and Mg-doped ZnO thin films are given in Figure 2.

When the comparative spectrum given in Figure 2 is examined, it is seen that all series have a polycrystalline structure. In the Z series, (010), (002), (011), (012), (110), (013), (020), (112), and (021) belonging to the hexagonal ZnO structure were observed, respectively. In AZ1, AZ3, and AZ5 series, 6 peaks belonging to the hexagonal ZnO structure (010), (002), (011), (012), (110), and (013) were observed, respectively. With Al doping, no peak belonging to Al was observed in the

Table 1 Series names of undoped, Al-, and Mg-doped thin films

Doping rate	Serial name
-	Z
1% Al	AZ1
3% Al	AZ3
5% Al	AZ5
1% Mg	MZ1
3% Mg	MZ3
5% Mg	MZ5

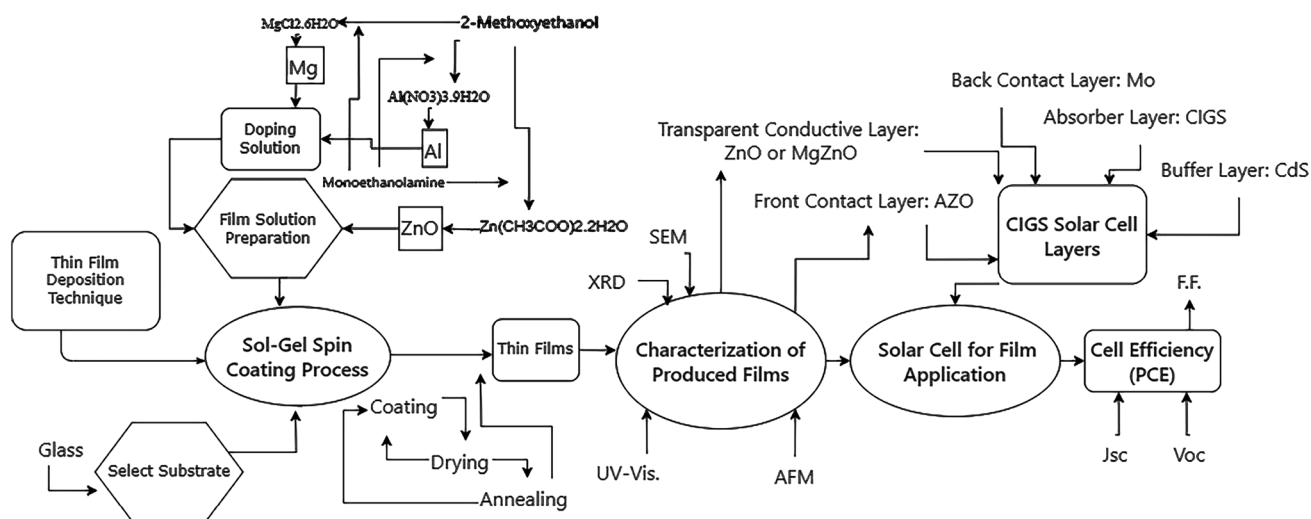
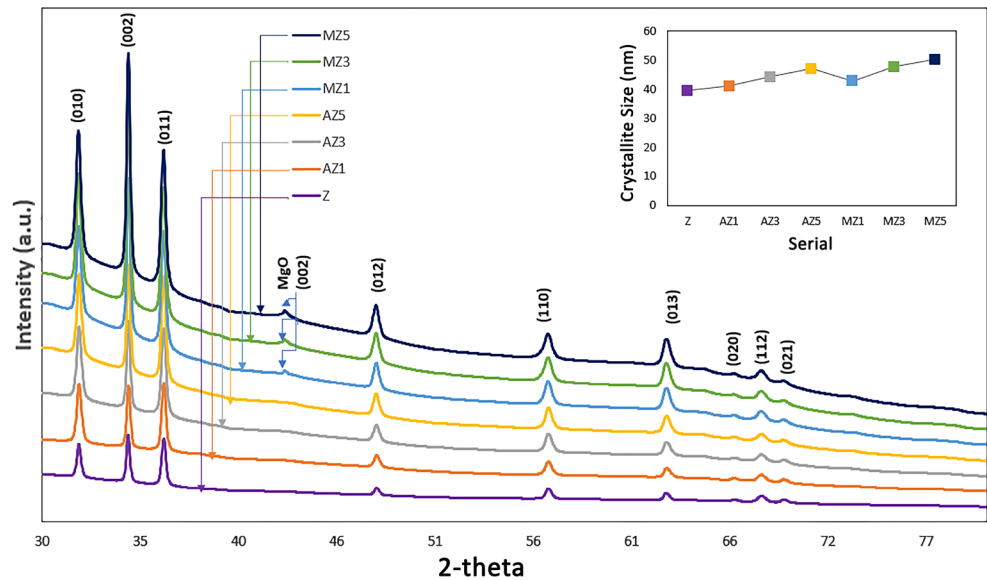


Fig. 1 Process flowchart for the production of Al/Mg-doped ZnO thin films and their use as layers in CIGS solar cells

Fig. 2 XRD spectra of Al- and Mg-doped and undoped ZnO thin films



crystal structure. In addition, as a result of doping, the number of peaks belonging to the ZnO structure and their intensities decreased. With the decrease in the intensities and numbers of peaks in the Z series with Mg doping; the (002) peak belonging to the MgO structure was formed at $2\theta \approx 42.80^\circ$ in the MZ1, MZ3, and MZ5 series. The surface properties of the obtained undoped and Al–Mg-doped ZnO thin films were analyzed by ZEISS Supra 40 VP FESEM device and Ambios Q-Scope AFM device. Figure 3a shows the surface images of Al-doped ZnO thin films, and Fig. 3b shows the surface images of Mg-doped ZnO thin films.

When the images are examined, it is seen that the undoped ZnO structure is homogeneously formed as nanoparticles on the surface in FESEM images taken with 30 kX magnification (average measured grain size value is approximately 40 nm) and there are no cracks, voids, and agglomerations on the surface that cause efficiency decrease in photovoltaic applications. It was observed that the structure maintains its homogeneity and the grain sizes increase slightly with Al and Mg doping. 3D AFM images also support that the surface is homogeneous. Surface roughness values obtained from AFM images are given in Table 2. When the average surface roughness values are examined, it is seen that Al doping increases the surface roughness of ZnO films, while 3% and 5% Mg doping decrease the surface roughness of ZnO films. The highest roughness was found in the AZ5 series, while the lowest roughness was found in the MZ5 series. Surface roughness values show that the obtained thin films can provide

high optical transmittance and can be used as transparent conductive oxide layers in photovoltaic cells. The average film thicknesses of Mg/Al-doped and undoped ZnO thin films were found between 350 and 400 nm.

The optical properties of the obtained thin films were examined in the 200–1100-nm wavelength range on the Perkin Elmer Lambda 25 UV–Vis spectrophotometer device and the graphs are given in Figure 4. The absorbance and transmittance values of ZnO thin films were analyzed to determine the band gap using the Tauc method, with the effects of Al and Mg doping on optoelectronic properties being examined. [30]

The band gap of ZnO films was determined as 3.3 eV. It was observed that Al doping decreases the band gap of ZnO, while Mg doping increases this value. The lowest band gap in AZ series was found to be 3.06 eV in AZ5 series, while the highest band gap in MZ series was found to be 3.54 eV in MZ5 series. The produced AZ series thin films can be used in various areas, such as transparent conductive electrodes, optoelectronic devices, gas sensors, photocatalytic applications, piezoelectric devices, and antistatic coatings. By changing the doping rate, it will be possible to integrate films into different applications and increase performance. The use of AZ series thin films as layers in solar cells is possible thanks to the electrical and optical properties of the films. They are used especially as transparent conductive oxide (TCO) layers in solar cells. AZ series layers increase the efficiency of cells by transmitting sunlight and providing electrical conductivity at the

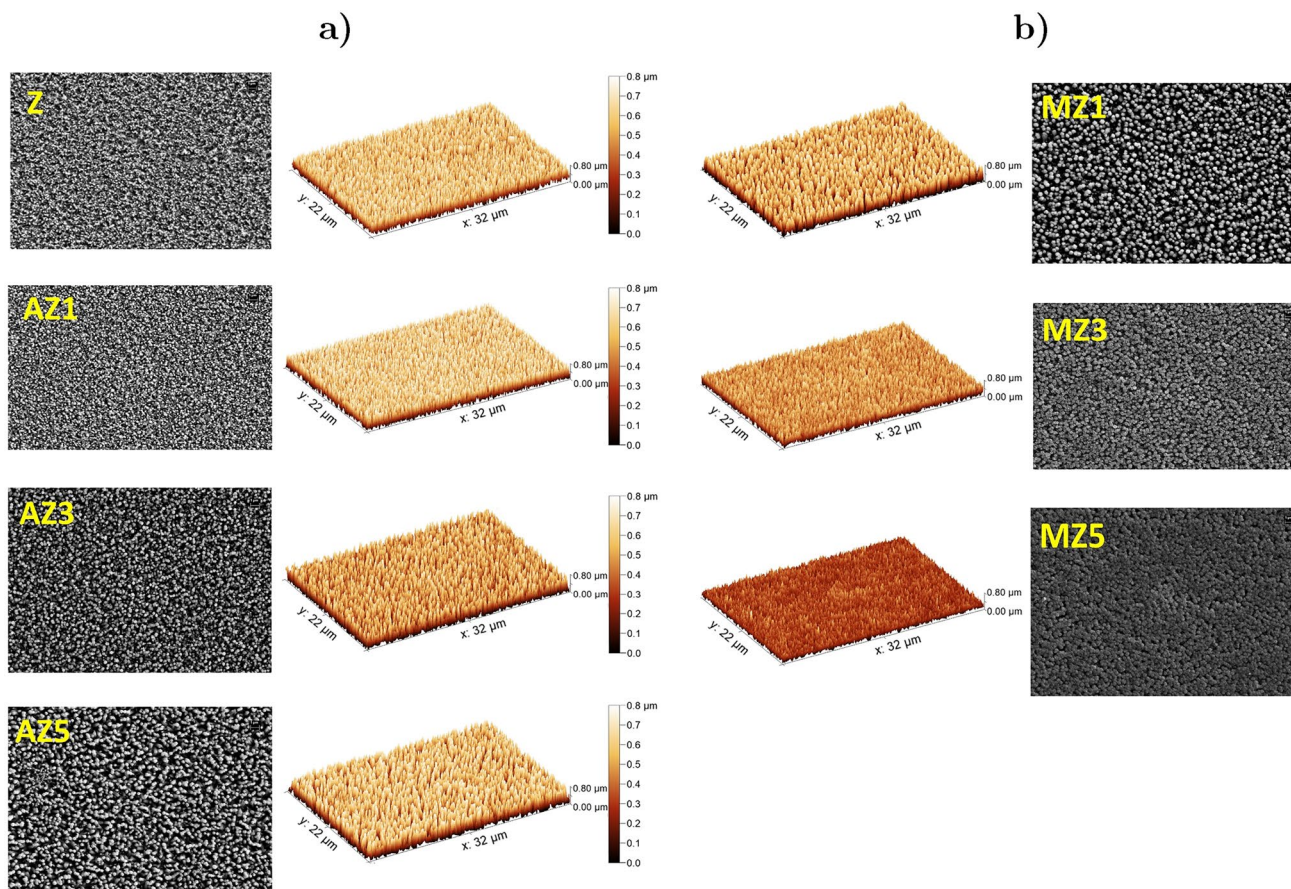


Fig. 3 SEM and AFM images of **a** Al-doped and undoped ZnO thin films and **b** Mg-doped ZnO thin films

Table 2 Roughness values of thin-film series

Serial	RMS Roughness (nm)	Mean (nm)
AZ1	206.5	174.7
AZ3	198.6	171.2
AZ5	224.3	195.2
MZ1	221,7	195,6
MZ3	146,2	121,5
MZ5	87,9	69,7
Z	184,8	155,3

same time. AZ1 films provide high transparency and sufficient conductivity, AZ3 films offer the best balance and AZ5 films provide the highest conductivity. AZ series thin films can be preferred for different solar cells. For CdTe and organic solar cells, AZ1 and AZ3 films can be considered as suitable options. For perovskite and CIGS solar cells, AZ3 and AZ5 films can meet higher conductivity requirements. The average resistivity values were found to be $3.53 \cdot 10^{-4} \Omega \cdot \text{cm}$ for

ZnO series, $6.57 \cdot 10^{-4} \Omega \cdot \text{cm}$ for AZ series, and $4.51 \cdot 10^{-3} \Omega \cdot \text{cm}$ for MZ series.

The produced MZ thin films have a wide range of applications such as solar cells, LEDs, sensors, and transparent electronics thanks to their ability to adjust optical and electrical properties. MZ series thin films can be used as both TCO and ETL layers, which increase efficiency in various solar cell technologies, especially CIGS, CdTe, and perovskite, with their adjustable band gaps and high transparency. Within the scope of this study, the use of the produced Z, AZ, and MZ series films as CIGS Solar Cell layers was studied. CIGS solar cell, Back Contact Layer; Molybdenum (Mo) (Collection, transport of electrons, mechanical support) Emission Layer; CIGS (Cu(In,Ga) Se₂) (Production of electron-hole pairs by absorbing light) Buffer Layer; Cadmium Sulfide (CdS) (For a suitable band alignment between the emission layer and the TCO layer) Transparent Conductive Layer (TCO); produced MZ series thin films (Enables light to reach the emission layer and keeps electrical conductivity

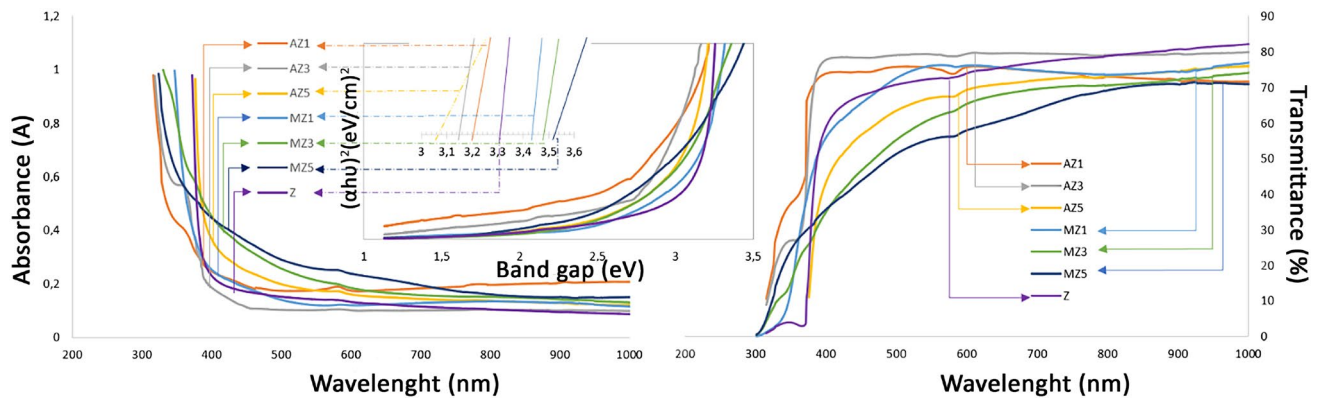


Fig. 4 Absorbance and transmittance spectra of Z, AZ, and MZ Series thin films against wavelength. The energy band gap of thin films ($T_{auc} \text{ graph } (\alpha h\nu)^2 - (h\nu)$)

high) Front Contact Layer; and produced AZ series thin films (functions as a transparent electrode, for light transmission and electrical conductivity) were formed. It was aimed to increase the performance in CIGS solar cells with the Mo/CIGS/CdS/MZ/AZ layer order. The J–V graph prepared according to the photovoltaic parameters measured when AZ series films are used as the Front Contact Layer and MZ series films are used as the TCO layer in CIGS solar cells under AM1.5G conditions (1000 W / m^2) is given in Figure 5 in comparison with the Z series.

Fill Factor values were calculated by determining the Voc values from the point where the J–V curve determined according to the series intersects the X-axis and the Jsc values from the point where it intersects the Y-axis. According to the Voc, Jsc, F.F, and Pmax values determined from the J–V graph, the calculated PCE values of the Series are shown in Fig. 6.

When the efficiency values of the CIGS solar cell formed with the Mo/CIGS/CdS/MZ/AZ layer sequence were examined, the highest efficiency was found to be 26.46% in the AZ5/MZ1 layer. The efficiency of the

Fig. 5 J–V chart prepared according to measured photovoltaic parameters when used as layers in CIGS solar cells of Z, AZ, and MZ Series

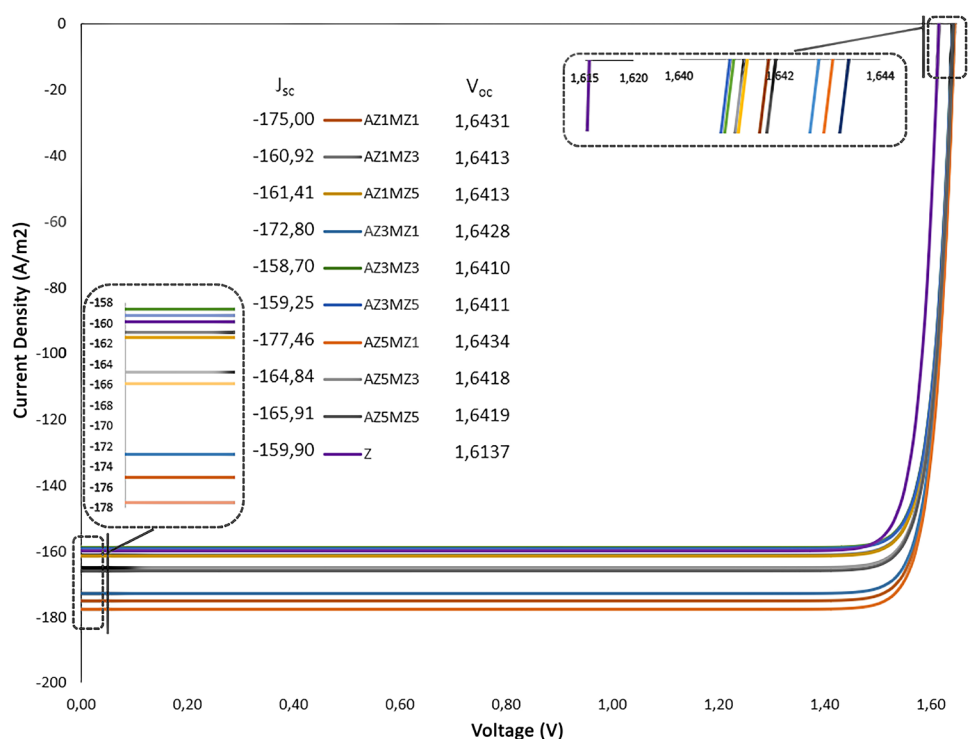
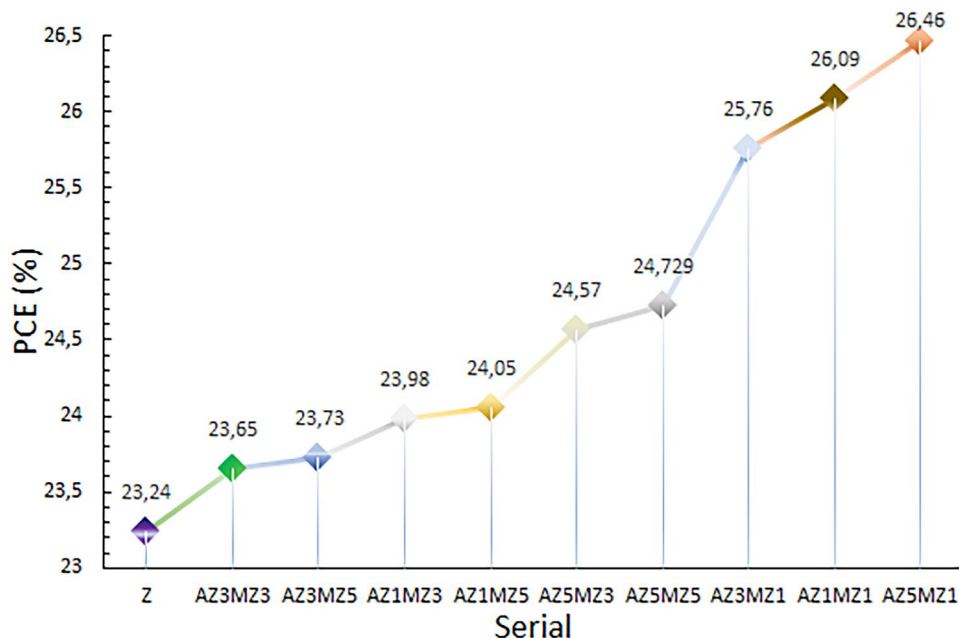


Fig. 6 PCE values of CIGS solar cells using AZ-MZ series and Z series as a layer



CIGS solar cell formed with Mo/CIGS/CdS/Z layers was determined as 23.24%. In general, the efficiency of CIGS solar cells formed with AZ5 and MZ1 layers increased compared to the Z layer. It was found that when MZ series was used as the TCO layer and AZ series as the Front Contact Layer instead of the Z layer, the efficiency of the solar cell increased by more than 3%.

4 Conclusion

The effects of Al and Mg doping on the structural, surface, and optical properties of ZnO thin films were investigated. It was observed that all series were polycrystalline and doping did not change the crystal structure. It was determined that ZnO formed nano-sized particles on the surface, Al doping increased the surface roughness, while Mg doping decreased the roughness at high rates. The band gap value of undoped ZnO thin films was 3.3 eV, while it decreased to 3.2eV, 3.15 eV, 3.05 eV with Al doping, and increased to 3.43eV, 3.48, and 3.52 eV with Mg doping, respectively. Thanks to the high optical transmittance properties of the produced thin films, their usability as TCO layers in CIGS solar cells was tested. The AZ and MZ series layers significantly improved the cell efficiency compared

to the Z series layer. The efficiency reached 26.46% with the MZ5/MZ1 layer combination, well above the 23.24% efficiency achieved with the Z series. Doping can modify the surface and optical properties of thin films, making it possible to develop more efficient and effective materials for solar energy conversion and optoelectronic applications.

Author Contributions

M.F.G. contributed to writing, experimentation, methodology, reviewing, framing, editing, visualization, and conceptualization. E.N. contributed to methodology, experimentation, review, and writing. S.T. contributed to methodology, writing, review, experimentation, and validation. F.O.G. contributed to writing, reviewing, and editing of the manuscript and validation.

Funding

This research was conducted without any funding

Data availability

All data included in this manuscript are available upon request by contacting the corresponding author

Declarations

Conflict of interest All authors declare no conflict of interest

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