



Enhanced photocatalytic efficiency of Mg-doped ZnO thin films: a comparative study of methanol and ethanol-based sol-gel synthesis

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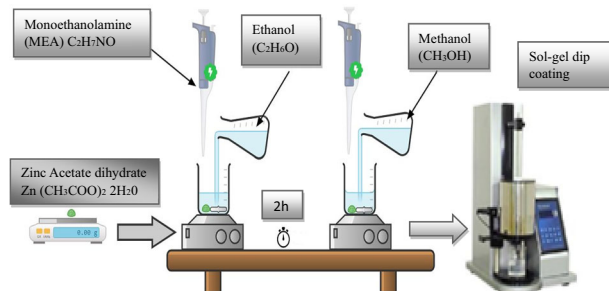
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Abstract

This study provides a comprehensive comparison of the structural, optical, and electrical properties of Mg-doped ZnO thin films synthesized using methanol and ethanol as solvents. X-ray diffraction analysis confirmed the hexagonal wurtzite structure for both cases, with a stronger c-axis orientation in ethanol-derived films. Notably, magnesium incorporation led to an increase in crystallite size, reaching 23 nm in methanol-based films. Morphological analysis using scanning electron microscopy (SEM) revealed dense, homogeneous, and smooth surfaces for both solvent-derived films. Optical characterization indicated a reduction in transmittance for ZnO films synthesized in methanol compared to ethanol-based films, alongside an increase in the energy band gap (ranging from 3.23 to 3.34 eV) as Mg concentration increased. Photoluminescence spectroscopy identified UV emission and defect-related peaks in the visible region at room temperature. Magnesium incorporation was further verified by energy-dispersive spectroscopy (EDS) and Fourier-transform infrared spectroscopy (FTIR). Electrical and photocurrent measurements demonstrated n-type semiconducting behavior with enhanced photoresponse sensitivity. The Seebeck coefficient showed a decreasing trend with increasing Mg doping, with methanol-derived films exhibiting a higher coefficient ($-525 \mu\text{V/K}$) compared to ethanol-derived films ($-400 \mu\text{V/K}$). Despite this, methanol-based films exhibited a higher carrier concentration at the same Mg doping level. Furthermore, photocatalytic performance was optimized with 9% Mg-doped ZnO films synthesized in methanol, leading to enhanced degradation efficiency under solar irradiation. These findings emphasize the combined influence of Mg doping and solvent choice on the functional properties of ZnO thin films, providing valuable insights for their potential applications in environmental remediation and energy-related technologies.

Graphical Abstract

This study conducts a comparative analysis of Mg-doped ZnO thin films synthesized using methanol and ethanol as solvents. The photocatalytic efficiency of the films is assessed through the degradation of methyl blue and methyl orange, serving as model pollutants. The results reveal that methanol-derived films exhibit superior photocatalytic performance, emphasizing their potential for advanced environmental remediation applications.



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Highlight

- This study examines the photocatalytic performance of Mg-doped ZnO thin films synthesized using ethanol and methanol solvents, demonstrating the superior efficiency of methanol-based films in enhancing photocatalytic activity.
- Our findings indicate that methanol-derived Mg-doped ZnO thin films exhibit enhanced photocatalytic performance, effectively degrading methyl blue and methyl orange pollutants, with promising applications in wastewater treatment.
- The results highlight that both Mg doping concentration and solvent choice significantly influence the material properties, contributing to improved environmental sustainability.
- A detailed investigation of charge carrier transport mechanisms reveals their crucial role in photocatalytic activation and overall material efficiency.

1 Introduction

Environmental pollution has emerged as a critical global challenge, with dye wastewater from the textile industry posing severe ecological risks [1]. Among various remediation strategies, photocatalysis has garnered significant attention for its potential applications in environmental conservation, including water and air purification, hazardous waste management, and sustainable energy production through environmentally friendly approaches [2]. In this context, materials with piezoelectric and pyroelectric properties—especially those exhibiting a wurtzite crystal structure and strong electromechanical characteristics—are of great importance in materials science [3].

Several wide-bandgap inorganic semiconductors, such as TiO_2 , WO_3 , and ZrO_2 , have demonstrated effectiveness in light-driven catalytic redox reactions for dye degradation [4, 5]. Among these, zinc oxide (ZnO) stands out due to its versatile nanostructural properties, possessing a wide bandgap ($E_g = 3.37$ eV) and a high free exciton binding energy of 60 meV [6]. ZnO is an n-type semiconductor that offers several advantages over alternative materials, including non-toxicity, cost-effectiveness, chemical stability, and excellent optical transparency in the visible and near-infrared spectrum [7, 8]. These properties enable ZnO's use in a wide range of applications, including light-emitting diodes, laser diodes, ultraviolet photodetectors, solar cell windows, transducers, and gas sensors [9–12]. Additionally, ZnO has found applications in biomedical fields, serving as a bactericide and photocatalyst, contributing to infection prevention, and enhancing the biocompatibility of medical implants [13].

ZnO's remarkable photocatalytic potential has been widely explored for the degradation of various dyes, including methylene blue (MB), methyl orange (MO), rhodamine B, acid red 14, eosin Y, and other organic pollutants, demonstrating its effectiveness in environmental remediation [14–16]. The synthesis of ZnO nanoparticles can be achieved through multiple techniques, including

alkali precipitation [17], hydrothermal synthesis [18], organo-zinc hydrolysis [19], spray pyrolysis [20], microwave irradiation [21], and plasma heat decomposition [22]. The morphology, size, and crystallinity of the synthesized ZnO nanostructures depend largely on the chosen synthesis method [23].

Among these synthesis approaches, the sol-gel method is particularly favorable for producing thin films due to its ability to control the composition and structure of the final material. Furthermore, ZnO's photocatalytic properties can be significantly enhanced through selective metal doping, which plays a crucial role in modifying its luminescence and optical characteristics [24]. This strategy has been employed to tune the bandgap, introduce localized defect states, and alter the surface morphology, improving ZnO's overall photocatalytic performance. Several metal dopants, such as Al, Co, Mn, Ag, and Fe, have been explored for this purpose [25]. Among these, magnesium (Mg) doping has gained special interest due to its impact on ZnO's optical and luminescent properties [26].

In addition to doping, various processing parameters influence ZnO thin film quality, including solution pH, deposition time, bath temperature, and solvent properties. López et al. [27] reported that solvent characteristics significantly affect ZnO crystal formation. Similarly, Anastasiadis et al. [28] documented the development of ZnO nanostructures with varying morphologies depending on the choice of solvent. Yin et al. [29] demonstrated that the solvent's boiling point substantially impacts ZnO's surface structure.

Huang et al. [30] studied the structural and optical properties of Mg-doped ZnO (MZO) thin films deposited via the sol-gel method, revealing that increasing Mg concentration leads to the formation of a secondary MgO phase and an increase in the ZnO bandgap. Arshad et al. [31] synthesized MZO thin films via a wet chemical route and reported that Mg doping inhibited particle growth, increased the optical bandgap, and enhanced visible photoluminescence (PL) emission. Additionally, Karthick et al.

[32] investigated the impact of Mg doping on ZnO thin films produced by sputtering, attributing the observed blue shift in PL emission to bandgap modulation and a reduction in defect centers. Surface morphology was also notably altered upon Mg doping [32].

Moreover, Madahi et al. [33] demonstrated that Mg-doped ZnO thin films exhibited significant antibacterial properties, further enhanced by increasing Mg concentration. Rana et al. [34] fabricated multilayer Mg:ZnO/ZnO thin films for UV photodetectors and reported that the $\text{Mg}_{0.02}\text{Zn}_{0.98}\text{O}/\text{ZnO}$ structure exhibited the highest response under 365 nm UV light at a 5 V bias. Studies on ZnO-based thermoelectric properties indicate that Tambasov et al. achieved a maximum Seebeck coefficient of $-60 \mu\text{V/K}$ at 300 K [35], while Saini et al. found that pulsed laser-deposited Al-doped ZnO thin films on SrTiO_3 substrates exhibited a Seebeck coefficient of $-65 \mu\text{V/K}$ for 2% Al doping at 300 K [36–40].

This study presents an in-depth investigation into the solvent-mediated effects of methanol and ethanol on the structural, optical, morphological, and electrical properties of undoped and Mg-doped ZnO thin films. The research examines the correlation between Mg doping concentration, defect states, and photocatalytic efficiency. Additionally, the photocatalytic activity of these films is evaluated for the degradation of MB and MO under ultraviolet (UV) and solar irradiation.

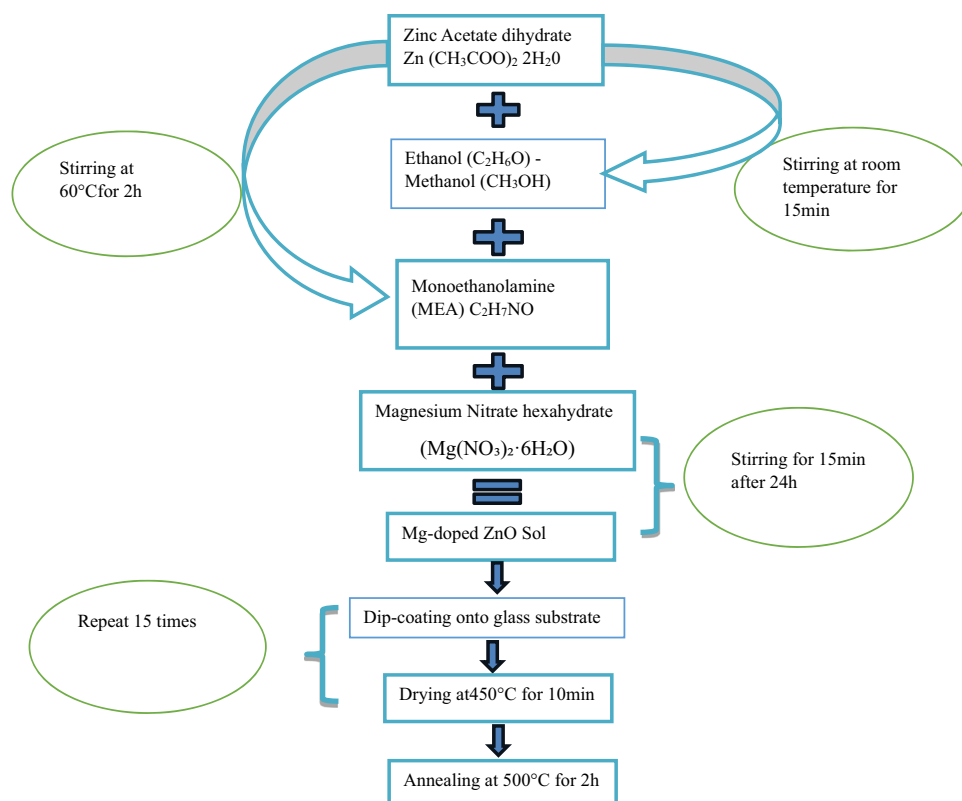
The primary objectives of this study are to assess the impact of solvent choice on the photocatalytic efficiency of ZnO thin films, examine the correlation between defect density and photocatalytic performance, investigate the influence of crystallinity on photocatalytic activity, explore charge carrier transport mechanisms and their role in photocatalytic activation, and develop innovative materials with enhanced photocatalytic properties for environmental applications. By elucidating the mechanisms underlying Mg doping effects, optimizing solvent conditions, and identifying the most effective solvent for pollutant degradation, this study aims to contribute to environmental sustainability and the advancement of efficient photocatalytic systems.

2 Experimental sections

2.1 Film preparation

In this study, magnesium (Mg)-doped and undoped zinc oxide (ZnO) thin films were fabricated on glass substrates ($2.5 \times 2.5 \text{ cm}$) using the sol-gel dip-coating method. The overall deposition process is illustrated in Fig. 1. To formulate the precursor solutions, zinc acetate dihydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$) served as the primary zinc source, while magnesium nitrate hexahydrate ($\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) acted as the dopant precursor. These compounds were

Fig. 1 Flowchart of sol-gel process for preparing Mg-doped ZnO thin films



dissolved in absolute ethanol ($\text{C}_2\text{H}_6\text{O}$, 99% purity) and methanol (CH_3OH , 99% purity) as organic solvents. Additionally, monoethanolamine (MEA) was introduced as a stabilizing agent to enhance solution uniformity and stability.

Each precursor solution was prepared by dissolving zinc acetate and magnesium nitrate in 30 mL of the respective solvents at room temperature, ensuring a zinc acetate concentration of 0.4 mol/L. The Mg:Zn atomic ratios were adjusted to obtain five distinct films: undoped ZnO, 3% Mg:ZnO, 5% Mg:ZnO, 7% Mg:ZnO, and 9% Mg:ZnO. The MEA-to-zinc acetate molar ratio was maintained at 1:1 for optimal stabilization. The solutions underwent magnetic stirring at 60 °C for 120 min to achieve a clear and homogeneous mixture. Subsequently, they were left to age at room temperature for 48 h to enhance solvation and stability.

Before deposition, the glass substrates were thoroughly cleaned using an ultrasonic bath in acetone, methanol, and distilled water for 10 min to remove contaminants. The dip-coating process involved immersing the substrates in the prepared Mg-doped ZnO solutions, followed by spin-coating at 3000 rpm for 30 s. The coated substrates were then preheated at 400 °C for 10 min, with this cycle repeated 15 times to ensure uniform film thickness. Deposition parameters such as coating repetitions and solution concentration influenced the final thickness of the films.

To enhance the crystallinity and optimize material properties, the films underwent thermal annealing in an air atmosphere at 500 °C for 2 h using a conventional furnace.

2.2 Characterization techniques

A detailed characterization of the thin films was conducted to analyze their crystal structure, surface morphology, optical transmittance, and photoluminescence properties. Various advanced techniques were employed for this purpose. The crystal structure was examined using X-ray diffraction (XRD) with a D8 ADVANCE system, utilizing $\text{Cu K}\alpha$ radiation ($\lambda = 1.54184 \text{ \AA}$) over a scanning range of 20°–80°. Fourier-transform infrared (FTIR) spectroscopy was performed with a Perkin-Elmer spectrometer at a resolution of 8 cm^{-1} , operating in transmission mode within the spectral range of 400–4000 cm^{-1} . For these measurements, the coatings were deposited on glass substrates, then removed by scraping for analysis via the KBr disk method. The surface morphology and microstructural features were investigated using field emission scanning electron microscopy (FE-SEM) with a Thermo Scientific Quattro SEM.

The optical properties of the films were determined by measuring their transmittance using a Spectro Scan 80D spectrophotometer (UV–Vis) in the spectral range of 190–1100 nm. Photoluminescence (PL) measurements were

carried out using a Perkin Elmer LS 45 fluorescence spectrometer, with an excitation wavelength set at 290 nm. Additionally, the Seebeck coefficient was evaluated by measuring the Seebeck voltage while applying a temperature gradient ranging from 0 to 120 K.

2.3 Photodegradation of MB

The photocatalytic performance of both Mg-doped and undoped ZnO thin films was evaluated by studying the degradation of MB and MO dyes in methanol and ethanol solvents. The efficiency of the photocatalytic process was determined by measuring the absorbance intensity of the dyes at specific wavelengths: 664 nm for MB (within the spectral range of 500–750 nm) and 464 nm for MO (within 300–650 nm). The initial dye concentrations, calculated from their visible absorbance intensities, were $2.5 \times 10^{-5} \text{ mol}$ for MB and $1.78 \times 10^{-5} \text{ mol}$ for MO. A UV-VIS-NIR Lambda 19 spectrophotometer was used for all absorbance measurements. To assess photocatalytic activity under controlled conditions, the samples were irradiated with a Philips germicidal incandescent lamp G15T8 (15 W, $\lambda = 251 \text{ nm}$) at a fixed distance of 6 cm, and the degradation kinetics were monitored over time. Prior to irradiation, the dye solutions were stirred in both dark and UV-illuminated environments for 30 min to establish adsorption-desorption equilibrium. Additionally, the photocatalytic degradation of MB was investigated under natural sunlight during July in Algeria at an ambient temperature of 34 °C. A 5 mL MB solution was initially kept in the dark before being exposed to direct sunlight, and its degradation was monitored by measuring absorbance intensity variations at 30-minute intervals [41].

2.4 UV light response tests

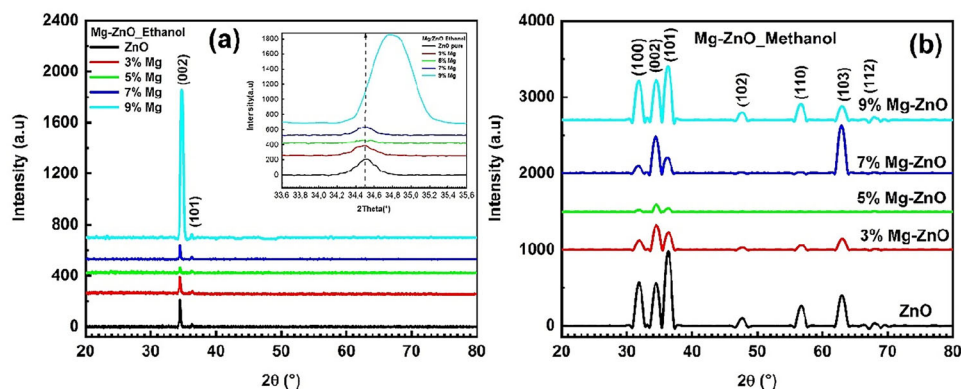
Upon exposure to 365 nm ultraviolet (UV) light, the UV photodetector was subjected to a controlled UV power level, restricted to a minimum of 90 mW/cm^2 using a UV power management system. The sample surface was illuminated perpendicularly within a light-shielded experimental enclosure to minimize external interference. The photocurrent signal was then recorded using a precision source/measurement unit. Real-time measurements were conducted using a two-point probe setup, with an applied input voltage fixed at 5 V [42].

3 Results and discussion

3.1 Structure analysis

Figure 2 presents the X-ray diffraction (XRD) analysis of ZnO thin films prepared using ethanol and methanol as

Fig. 2 XRD patterns of undoped and Mg-doped ZnO thin films using: **a** ethanol and **b** methanol solvents



solvents. In ethanol-based samples (ZnO, 3%, 5%, and 7% Mg-doped ZnO), diffraction peaks corresponding to the (002) and (001) crystal planes were observed at 2θ values of 34.5° and 36.4° , respectively. Notably, the 9% Mg-doped ZnO sample exhibited a slight peak shift to 34.7° for the (002) plane, as illustrated in the inset of Fig. 2a. The identified diffraction peaks were validated against the standard JCPDS Card No. 036-1451 [43], confirming a polycrystalline nature with a hexagonal wurtzite crystal structure. The films displayed a preferred orientation along the c-axis, dominated by the (002) peak [33, 44].

These findings align well with previous studies employing various solvents, including ethanol, methanol, propanol, and 2-methoxyethanol [45–47]. Yang et al. [1] demonstrated that higher solvent polarity enhances the preferred orientation along the (002) direction. This effect is attributed to variations in film growth kinetics influenced by solvent polarity. ZnO, in its wurtzite phase, possesses a polar crystal structure composed of alternating Zn^{2+} and O^{2-} layers along the c-axis. As solvent polarity increases, crystal formation along the (002) plane is promoted due to strong interactions between the polar solvent and the polar surface, which reduce surface energy [48].

Figure 2b further confirms the polycrystalline ZnO structure, characterized by a hexagonal wurtzite phase (ICSD card number 01-075-0576). The diffraction peaks correspond to the (100), (002), (101), (102), (110), (103), and (112) planes at 2θ values of 31.82° , 34.51° , 36.31° , 47.66° , 56.61° , 62.96° , and 67.95° , respectively [49]. The intensity of the (002) peak generally decreased with increasing Mg doping concentration in ethanol-based samples. However, the 9% Mg-doped ZnO sample exhibited an increase in peak intensity, suggesting enhanced crystallinity and reduced structural defects. This trend is consistent with prior findings by Xue et al. and Zhang et al. [22, 50].

The (002) peak position exhibited a shift toward lower angles with increasing Mg doping levels in 3%, 5%, and 7% Mg-doped ZnO films, but an unexpected shift toward higher angles was observed in the 9% Mg-doped sample [51]. This

anomaly is attributed to the substitution of Zn^{2+} ions by Mg^{2+} ions, leading to an increase in the c-axis lattice constant [32, 52–54].

Examining the XRD patterns of methanol-based samples revealed a preferential c-axis orientation, with the (002) peak appearing at 34.51° for ZnO:Mg 3% and at 34.50° for ZnO:Mg 5% films [49]. Interestingly, the 7% Mg-doped ZnO sample exhibited superior crystallinity, characterized by a pronounced (103) peak at 62.96° . The relative intensity of the primary XRD peaks fluctuated with increasing Mg doping concentration, but no secondary phase peaks were detected. This confirms the successful incorporation of Mg ions into the ZnO lattice and the preservation of the hexagonal wurtzite phase, as reported by Kayani et al. [55].

These findings underscore the successful doping of Mg into ZnO thin films and highlight the significant impact of solvent choice on their structural properties.

The X-ray diffraction pattern is utilized to determine structural parameters such as lattice parameters, c/a ratio, and crystallite size (D) for both solvents (ethanol-methanol). The crystallite size is calculated employing the Debye-Scherrer formula [56]:

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (1)$$

The dislocation density is obtained from Eq. (2) [57]:

$$\delta = \frac{1}{d^2} \quad (2)$$

where D is the crystallite size $\lambda = 0.15406$ nm is the X-ray wavelength, K is the constant of 0.9, θ is the diffraction angle and β is the full at half maximum (FWHM) [32].

The micro-strain exhibits variations with increasing Mg doping content, as shown in Tables 1 and 2. The lattice constants a and c of the wurtzite-structured ZnO are determined using Bragg's law [55]:

$$2d \sin \theta = n\lambda \quad (3)$$

Table 1 The comparison of inter planer distance, grain size, lattice constants, dislocation density and lattice ratio among Mg: ZnO films using ethanol as a solvent

Samples	(hkl)	2 θ (°)	FWHM	a(Å)	c(Å)	c/a	D (nm)	δ	d(hkl)
Pure	002	34.467	0.5077	3.0022	5.2000	1.7320	17	0.0034	2.6000
3% Mg	002	34.449	0.5082	3.0037	5.2026	1.7320	17	0.0034	2.6013
5% Mg	002	34.460	0.4898	3.0027	5.2009	1.7320	18	0.0031	2.6004
7% Mg	002	34.456	0.4621	3.0027	5.2016	1.7320	19	0.0028	2.6008
9% Mg	002	34.793	0.3884	2.9749	5.1527	1.7320	22	0.0019	2.5763

Table 2 The comparison of inter planer distance, grain size, lattice constants, dislocation density and lattice ratio among Mg: ZnO films using methanol as a solvent

Samples	(hkl)	2 θ (°)	FWHM	a(Å)	c(Å)	c/a	D (nm)	δ	d(hkl)
Pure	101	36.326	0.9980	2.8533	4.9421	1.7320	9	0.0130	2.4710
3%Mg	002	34.511	0.7535	2.9985	5.1935	1.7320	12	0.0075	2.5967
5% Mg	002	34.507	0.6643	2.9987	5.1940	1.7320	13	0.0058	2.5970
7% Mg	103	62.966	0.4261	1.7031	2.9499	1.7320	23	0.0019	1.4749
9% Mg	101	36.311	0.7653	2.8544	4.9441	1.7320	11	0.0076	2.4720

The hexagonal structure lattice constants are given by [12]:

$$\frac{1}{d_{hkl}^2} = \frac{4}{3} \left(\frac{h^2 + hk + k^2}{a^2} \right) + \frac{1}{c^2} \quad (4)$$

A comparative analysis of ZnO thin films synthesized using ethanol and methanol as solvents revealed notable differences in structural properties. Films derived from ethanol exhibited a slight increase in lattice constants compared to those fabricated with methanol. This expansion may be attributed to the presence of free charge carriers or oxygen vacancies, which can enhance lattice constants, as observed by Mahajan et al. [46].

The incorporation of Mg into the ZnO lattice plays a crucial role in this phenomenon. Since the ionic radius of Mg²⁺ (0.57 Å) is smaller than that of Zn²⁺ (0.60 Å), Mg²⁺ ions may hinder the aggregation and growth of ZnO nanocrystals [58]. This suggests that ethanol facilitates the more efficient integration of Zn into the film, resulting in increased film thickness. The Full Width at Half Maximum (FWHM) of the Zn signal is directly correlated with the film's thickness [48]. The results indicate a significant reduction in FWHM values for ZnO thin films synthesized with ethanol compared to those obtained using methanol. This reduction suggests improved crystallinity and reduced lattice strain, leading to enhanced film stability and uniformity.

These findings emphasize the solvent's substantial impact on the characteristics of ZnO thin films. Notably, the data indicate that films deposited using ethanol exhibit a stronger tendency toward crystallization compared to those synthesized with methanol. This variation in crystallinity highlights the solvent's influence on crystal formation.

Potter et al. [59] attributed these differences to the interaction between the precursor and the solvent during

film formation. They proposed that as the solvent evaporates, precursor clusters diffuse toward the substrate and adsorb onto its surface, thereby regulating nucleation and growth. The choice of solvent influences the development of these clusters, ultimately affecting the film's morphology and topography.

The findings reveal a distinct correlation between the average crystallite size and the solvent used in the synthesis of ZnO thin films [48]. A detailed analysis of Mg-doped ZnO thin films prepared using ethanol and methanol solvents demonstrated unique crystalline growth patterns. Increasing the Mg concentration led to an enlargement of crystallite size, ranging from 17 to 22 nm for ethanol-based samples (9%), with a maximum of 23 nm for methanol-based samples.

In methanol-based Mg-doped ZnO thin films, crystallite size initially increased, with a notable enhancement observed in the 7% Mg-doped sample. This improvement is attributed to enhanced charge transport and recombination kinetics. Higher crystallinity reduces grain boundary scattering, promoting efficient charge carrier mobility and extending carrier lifetimes. Consequently, this lowers recombination rates and enhances photocatalytic performance. However, a significant decrease in crystallite size to 11 nm was observed in methanol-based samples at 9% Mg doping. This reduction is attributed to excessive Mg incorporation disrupting ZnO lattice growth, introducing strain, and inhibiting grain coalescence. The smaller ionic radius of Mg²⁺ reduces crystallite size but simultaneously increases macrostrain, leading to a decline in overall crystal quality [60].

3.2 Morphological analysis

Field emission scanning electron microscopy (FE-SEM) revealed significant morphological differences between

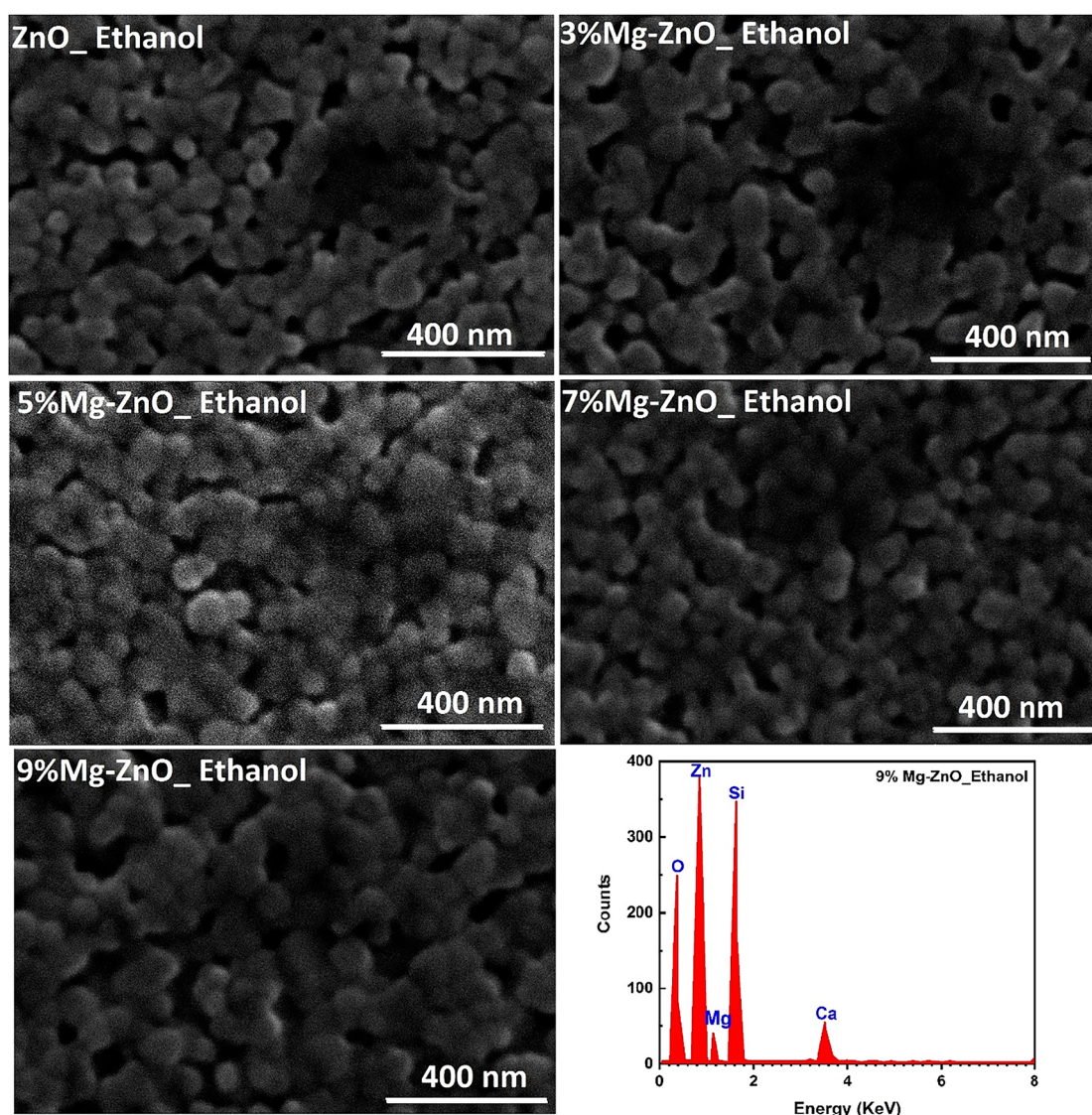


Fig. 3 FE-SEM image for undoped and doped ZnO as a function of Mg content and EDX spectra of 9% Mg-ZnO using ethanol as solvent

ZnO thin films synthesized using ethanol and methanol as solvents. As illustrated in Fig. 3, ethanol-based films exhibited nanorod structures approximately 400 nm in length, composed of oval ZnO grains with smooth, homogeneous surfaces that were dense and strongly adherent to the substrate [61]. The introduction of Mg doping influenced grain uniformity and pore size, where undoped ZnO displayed small interconnected pores that expanded at 3% and 9% doping levels [58]. Additionally, particle size increased with doping concentration, likely due to the presence of structural defects [62].

In contrast, Fig. 4 shows methanol-based films with smooth, uniform surfaces consisting of aggregated nanospheres, devoid of apparent fissures [63]. As the Mg doping concentration increased, the grains became larger, forming more porous structures that effectively covered the substrate

surface [64]. Doping also enhanced nanoparticle coverage across the thin film surfaces [65].

The micrographs of films prepared using methanol indicated improved grain growth. This behavior is attributed to the lower viscosity of methanol, which facilitates more efficient grain development. Methanol and ethanol possess viscosity values of 0.594 mPa·s and 1.2 mPa·s, respectively. As noted by Wang et al., a solution with lower viscosity enables a more rapid delivery of precursors to the substrate, thereby accelerating the growth mechanisms [66].

A study was conducted to analyze the chemical composition of various samples using energy dispersive X-ray spectroscopy (EDX). The atomic concentrations of the primary elements (O, Zn, and Mg), derived from the EDX spectra, are presented in Table 3. The weight percentages of Mg in the samples increased with the doping concentration

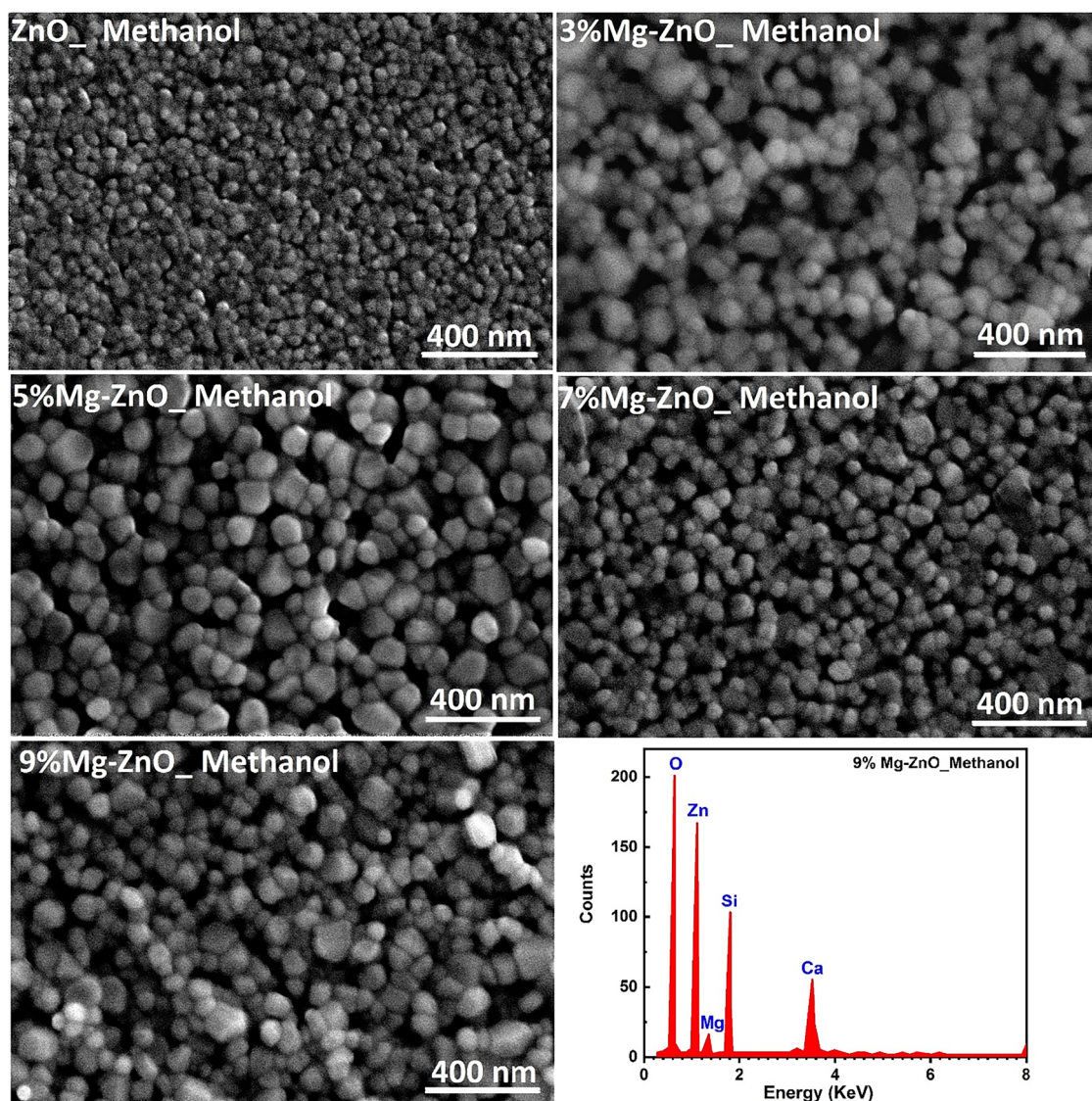


Fig. 4 FE-SEM image for undoped and doped ZnO as a function of Mg content and EDX spectra of 9% Mg-ZnO using methanol as solvent

for both ethanol- and methanol-based films. These findings align with the results obtained from XRD measurements [67].

The EDX characterization confirms the successful incorporation of magnesium into the ZnO films. The detection of O K, Mg K, Si K, and Zn K peaks in the spectrum validates the presence of oxygen, magnesium, silicon, and zinc, while also indicating the absence of foreign contaminants in the samples. The presence of Mg, Zn, and O further verifies that Mg atoms were well integrated into the ZnO lattice. This is corroborated by Table 3, which shows an increase in the intensity of Mg-related peaks.

Additionally, characteristic peaks corresponding to Zn, O, Si, and Ca were identified at 1.10 keV, 0.63 keV, 1.80 keV, and 3.50 keV, respectively, for methanol-based films, whereas ethanol-based films exhibited these peaks at

Table 3 The elemental composition of undoped ZnO and Mg-doped ZnO films prepared from methanol and ethanol as solvents

phases	Weight. %					
	Methanol			Ethanol		
	Zn	O	Mg	Zn	O	Mg
Pure	51.1	30.8	0	39.6	30.4	0
3% Mg	65.6	23.0	1.5	26.0	39.0	2.0
5% Mg	60.4	27.3	0.5	6.3	49.4	2.0
7% Mg	68.8	22.6	1.4	32.2	35.1	2.2
9% Mg	43.8	33.3	1.7	30.9	35.1	3.2

0.84 keV, 0.38 keV, 1.62 keV, and 3.51 keV. The presence of elements such as Si and Ca can be attributed to the soda-lime glass substrate, as also reported by Baig et al. [24].

Moreover, the intensity of Mg peaks increased progressively with higher doping concentrations, following an upward trend [67]. These results are consistent with previous studies, confirming the effective incorporation of Mg into ZnO films synthesized using both ethanol and methanol solvents.

Figure 5 illustrates the evolution of particle size for undoped and Mg-doped ZnO as a function of particle number when synthesized using ethanol as a solvent. The particle size increased with Mg doping concentration, ranging from 65 nm for pure ZnO to 83 nm for 9% Mg doping.

Conversely, methanol-based films exhibited varied nanoparticle diameters, with the smallest particle size recorded at 50 nm for pure ZnO and the highest frequency observed for 5% Mg-doped ZnO. The average particle sizes for 3%, 7%, and 9% Mg-doped ZnO were 80 nm, 66 nm, and 86 nm, respectively [63].

These variations are attributed to interactions between Mg and ZnO during film synthesis, which significantly influence the material's microstructure and growth mechanisms [68].

3.3 Fourier transform infrared (FTIR) analysis

In the characterization of thin films, Fourier-transform infrared (FTIR) spectroscopy has been instrumental in analyzing undoped and Mg-doped ZnO thin films synthesized using ethanol and methanol as solvents, as shown in Fig. 6 [69].

Notably, distinct absorption peaks were detected at 3440 cm^{-1} , 1635 cm^{-1} , 570 cm^{-1} , and 530 cm^{-1} for both solvents. The well-defined peaks at 570 cm^{-1} and 530 cm^{-1} are specifically attributed to ZnO vibrations, confirming the structural formation of the ZnO lattice [70]. A decrease in the intensity of these peaks was observed with increasing Mg concentration in methanol-based samples, whereas ethanol-based films exhibited an initial decline followed by a significant increase at 9% Mg doping. This variation indicates the incorporation of Mg into the ZnO lattice, altering the host structure [71].

Additionally, the peak at 1635 cm^{-1} corresponds to the bending vibrations of the H-O-H bond, signifying the presence of hydroxyl groups from adsorbed water molecules [72]. The broad peak near 3440 cm^{-1} is attributed to the O-H stretching mode, primarily caused by residual water molecules within the ZnO films [73].

A notable distinction in the FTIR spectra was the presence of a 960 cm^{-1} peak, attributed to C-O stretching vibrations of residual solvent (or other organic species), which appeared exclusively in ethanol-based ZnO films. This suggests unique vibrational modes and molecular structures differing from those observed in methanol-based samples [74].

3.4 Optical properties

The UV-VIS transmittance spectra of ZnO thin films prepared with ethanol and methanol solvents exhibit diverse characteristics. Films produced with ethanol, Fig. 7a, demonstrate high transparency (around 75–90%) between 400 and 1100 nm, indicating suitability for optical applications, with smooth and uniform surfaces inferred from interference fringes [75]. The optical transmittance changes with Mg doping concentration due to alterations in band-gap, trap states, and crystallographic defects. A pronounced absorption region is observed at higher photon energies, indicative of inter-band electronic transitions [76]. Alternatively, methanol-based films Fig. 7b exhibit good transparency in the visible spectrum (about 75%), with diminished optical transmission at higher photon energies. This study reveals a significant decrease in optical transmittance for methanol-derived ZnO films compared to their ethanol-based counterparts. The superior transmittance exhibited by ethanol-based films corroborates the solvents' pivotal role in enhancing optical properties, consistent with the findings of Potter et al. which highlighted solvent-dependent variations in ZnO film transmittance, underscoring the critical impact of solvent selection on optical characteristics [77].

All thin films have a heterogeneous absorption edge between 380 nm and 400 nm, which can infer the correlation between $(\alpha h\nu)^2$ and $(h\nu)$ to subsequently determine the energy gap (Eg) using, for instance, the Tauc plot model [78, 79].

$$\alpha h\nu = A(h\nu - E_g)^n \quad (5)$$

In this equation, $h\nu$ represents the photon energy, E_g denotes the optical gap energy, A is a constant, α signifies the absorption coefficient, and n is equal to $1/2$, as ZnO possesses a direct band gap [80].

The corresponding curves for the samples are displayed in Fig. 8, where the optical band gap (E_g) is determined by extrapolating the linear segment of the curve to $\alpha h\nu = 0$ [50]. This approach involves plotting $\alpha h\nu$ against photon energy and extending the linear portion of the curve to $\alpha h\nu = 0$.

As shown in Fig. 8 and Table 4, the results indicate that the energy band gap increases with magnesium concentration and is further enhanced when methanol is used as the solvent during sample preparation. The band gap values range from 3.23 to 3.34 eV for methanol-based films. However, in ethanol-based samples, a distinct trend is observed: the 3%, 5%, and 7% Mg-doped ZnO thin films exhibit a significant decrease in E_g values compared to undoped ZnO, while the 3% Mg-doped ZnO sample shows a pronounced increase.

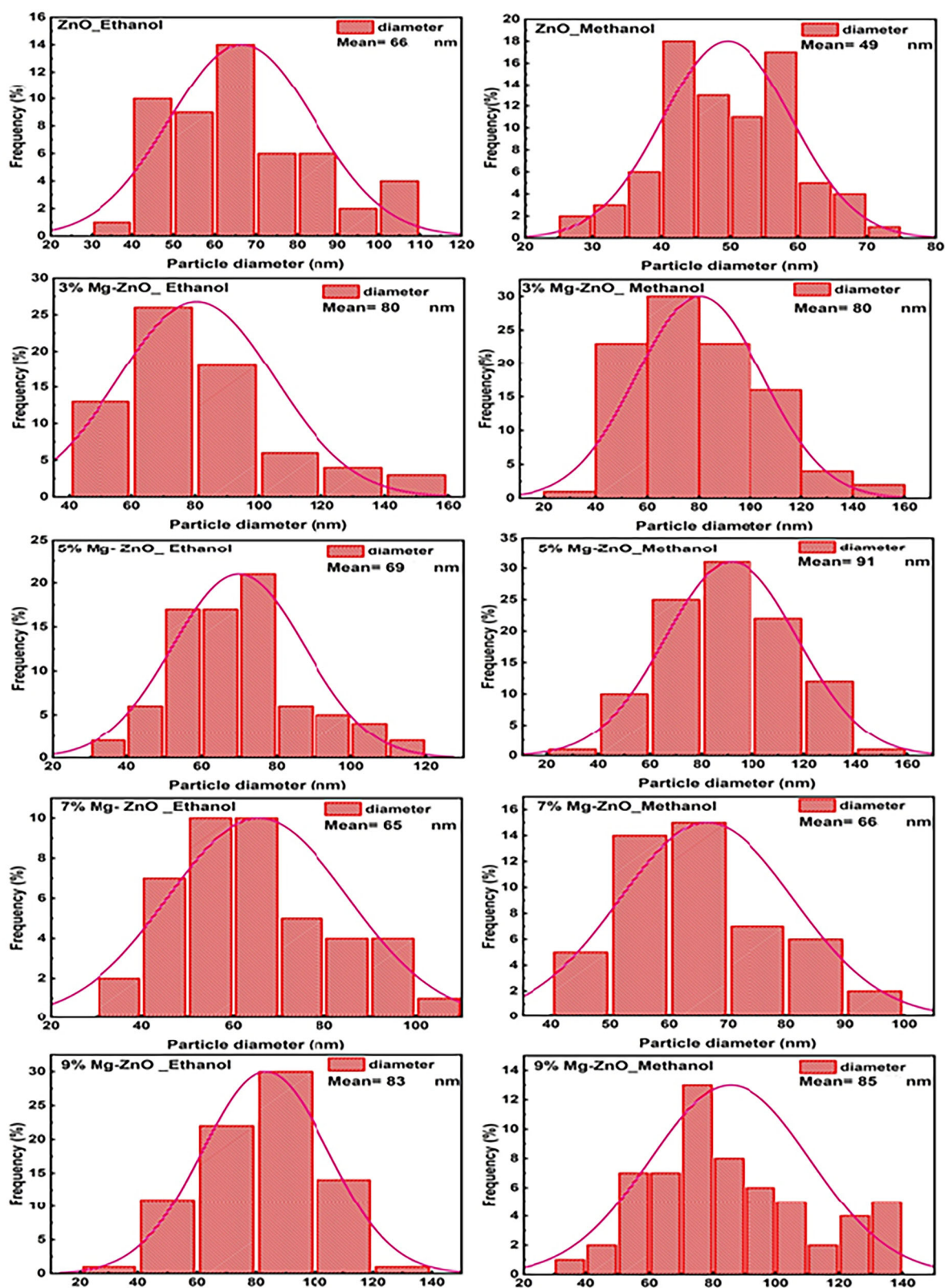


Fig. 5 Evolution of the particle size of magnesium-doped zinc oxide as a function of particle number for both solvents (ethanol—methanol)

This phenomenon suggests that Mg doping significantly influences the optical properties of ZnO. The broadening or

blue shift of the E_g is likely due to the Burstein-Moss effect, which occurs as a result of an elevated charge carrier

Fig. 6 FTIR spectra of undoped ZnO and Mg-doped ZnO thin films using: **a** ethanol and **b** methanol solvents deposited onto glass substrate

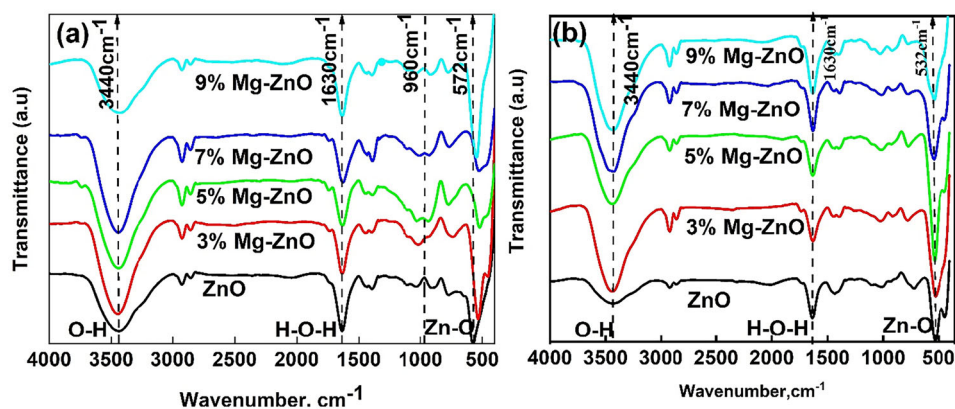


Fig. 7 Transmission spectra of ZnO and MZO thin films deposited on glass substrate at 500 °C. Measured (full circles) and calculated (solid lines) transmittance spectra of films using: **a** ethanol and **b** methanol as solvents

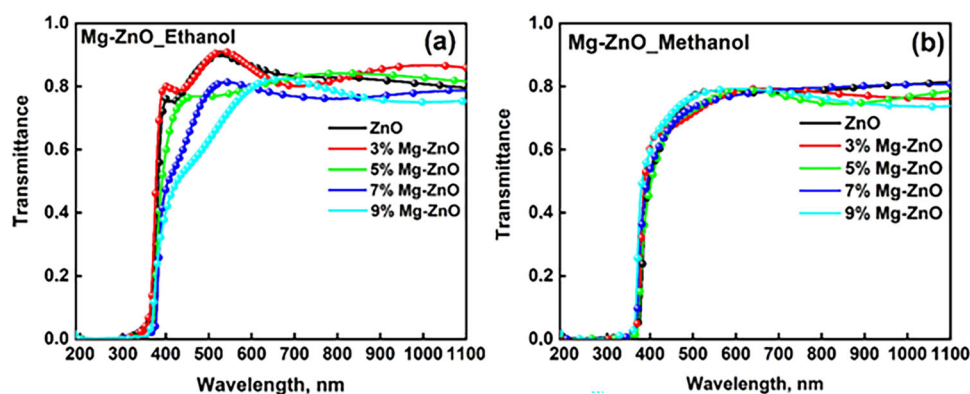


Fig. 8 $(\alpha h\nu)^2$ versus photon energy ($h\nu$) of undoped and Mg-doped ZnO thin films prepared using: **(a)** ethanol and **(b)** methanol solvents and their corresponding band gaps

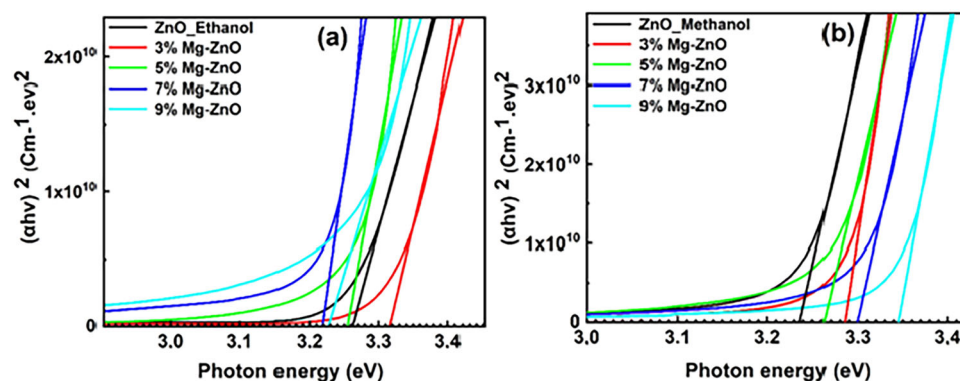


Table 4 Dispersion parameters of the films prepared using ethanol and methanol as solvents extracted by fitting the experimental data

Samples	ethanol				methanol			
	Eg (eV)	Thickness (nm)	n at 598 nm	n _∞	Eg (eV)	Thickness (nm)	n at 598 nm	n _∞
Pure	3.31	293	1.71	1.84	3.24	274	1.74	1.90
3% Mg	3.36	278	1.89	1.92	3.35	193	1.77	1.82
5% Mg	3.27	220	1.76	1.91	3.15	351	1.75	1.76
7% Mg	3.24	278	1.75	1.89	3.29	307	1.70	1.74
9% Mg	3.27	322	1.79	1.85	3.38	249	1.71	1.75

density induced by increased Mg doping [81]. As the charge carrier density rises, the lowest energy states at the bottom of the conduction band become occupied. According to the

Pauli exclusion principle, these states cannot accommodate multiple electrons, necessitating additional energy for electrons in the valence band to transition to higher-energy

levels in the conduction band. As a consequence, the optical band gap energy of ZnO films increases with Mg doping [34].

Statistical analysis reveals a strong correlation between magnesium concentration and solvent composition, both of which significantly influence the optical band gap energy (E_g). This synergistic effect suggests that by carefully adjusting magnesium doping levels and solvent characteristics, the optical properties of ZnO materials can be effectively tailored to meet specific application requirements.

A comparative assessment revealed analogous outcomes between the alternative method and Tauc plot analysis. The dispersion parameters of the samples—specifically d , E_g , n at 598 nm, and n_∞ were estimated using a single oscillator fit, as described by Wemple and DiDomenico [34]. The corresponding data is summarized in Table 4 for ZnO thin films synthesized using ethanol and methanol as solvents [82].

The solid curves in Fig. 7 represent the curve fitting, while the symbols correspond to the experimental data. The strong correlation between the model and experimental findings confirms the successful integration of Mg ions into the ZnO host lattice. Similar results were previously reported by Huang et al. [30] and Yusuf et al. [83].

It is well known that impurity incorporation can reduce the band gap, while higher doping concentrations introduce defect states in the valence and conduction bands, leading to

a further reduction in E_g . The optical band gap (E_g) of thin films synthesized using ethanol (as shown in Table 4) exhibits a decreasing trend with increasing Mg content, ranging from 3.31 eV (undoped ZnO) to 3.24 eV (7% Mg-doped ZnO). However, an increase to 3.36 eV is observed at 3% Mg doping, suggesting that defects and doping levels significantly impact the energy band structure [78, 84].

Conversely, thin films synthesized with methanol exhibit an increase in E_g with Mg doping, reaching 3.38 eV at 9% Mg concentration. The refractive index (n), known to be sensitive to structural defects such as voids, dopants, and inclusions, provides key insights into the material's microstructure. As presented in Table 4, the determined refractive index values for both ethanol- and methanol-based ZnO films are lower than those of bulk ZnO crystals (at $k = 598$ nm) [85].

3.5 Photoluminescence spectra analysis

Figure 9 presents the photoluminescence (PL) spectra of Mg-doped ZnO thin films prepared using different solvents and annealed at 500 °C for 2 h. The PL spectra provide insights into optically active defects and relaxation pathways of excited states [30]. Measurements were conducted in the 350–550 nm range for all samples. In ethanol-based ZnO films, Fig. 9a, a strong ultraviolet (UV) emission peak appeared between 382 nm and 389 nm for Mg doping

Fig. 9 Photoluminescence spectra of ZnO and MZO films and the Gaussian deconvolution of the PL intensity using **a** ethanol and **b** methanol as solvents

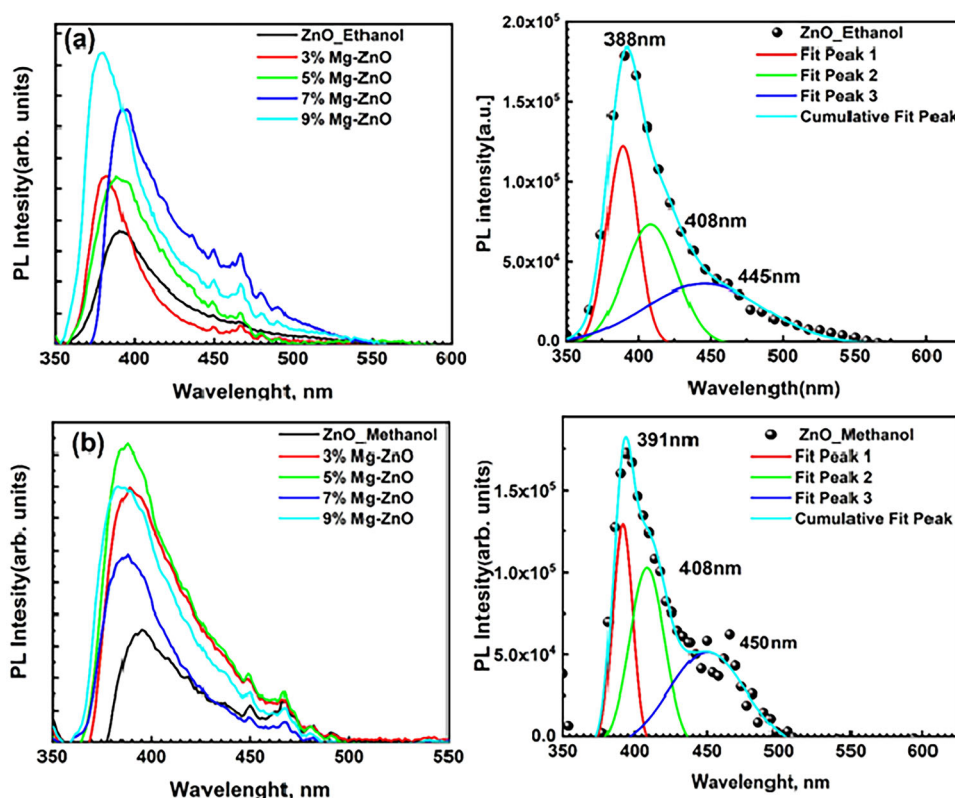
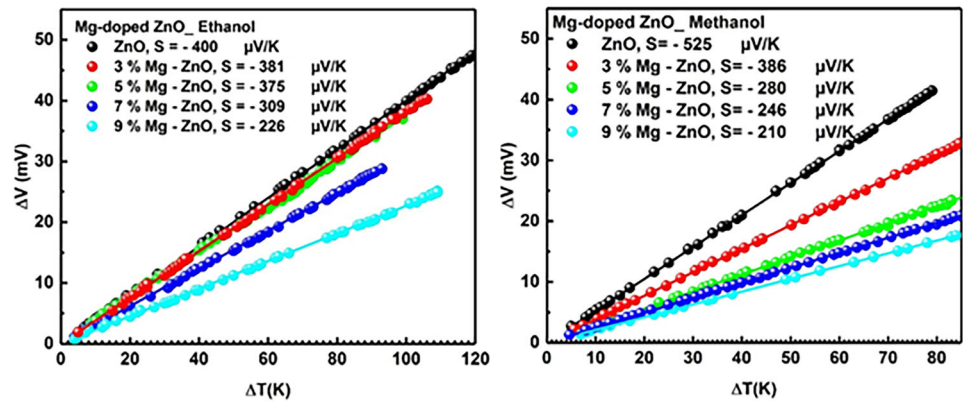


Fig. 10 Seebeck coefficients for ZnO and Mg-doped ZnO thin films with varying Mg concentrations for both solvents (ethanol- methanol)



concentrations ranging from 3% to 9%, compared to undoped ZnO thin films [43]. Additionally, a visible emission peak was observed between 408 nm and 422 nm for 3%, 5%, and 9% Mg doping, while undoped ZnO and 7% Mg-doped ZnO thin films exhibited a blue emission peak at 449 nm [43]. Conversely, methanol-based ZnO films, Fig. 9b, displayed a violet emission peak in the visible region between 390 nm and 399 nm for all samples, along with a blue emission peak at 450 nm for undoped ZnO [86]. These visible emissions are attributed to deep-level defects such as zinc interstitials (Zn_i), zinc vacancies (V_{Zn}), and oxygen vacancies (V_O) [87]. The weak blue emission at 449 nm likely originates from transitions between intrinsic defect states, particularly interstitial zinc and zinc vacancies near the valence band [30, 88]. The emission intensity analysis revealed that in ethanol-based samples, UV emission intensity increased with Mg concentration, reaching its peak at 9% Mg doping [30]. In contrast, methanol-based films exhibited heightened visible emission intensity, which peaked at 5% Mg doping, followed by a decrease at 7% and 9% doping levels [43]. The UV-to-visible emission intensity ratio (UV/Vis) increased with Mg concentration, suggesting a reduction in defect density and enhanced crystal quality for both solvents [86]. This improvement in crystallinity and defect suppression implies better charge carrier transport and lower recombination rates, which are crucial for photocatalytic activation [87].

3.6 Seebeck coefficient

Figure 10 illustrates the variation in the Seebeck coefficient at different Mg doping concentrations for ZnO thin films synthesized using ethanol and methanol as solvents. The recorded Seebeck coefficient (S) values, calculated via Eq. (6), for ethanol-based films are -400 , -381 , -375 , -309 , and -226 $\mu\text{V/K}$ for ZnO Pure, 3% Mg-doped ZnO (MZO), 5% MZO, 7% MZO, and 9% MZO, respectively.

Meanwhile, for methanol-based films, the values are -525 , -386 , -280 , -246 , and -210 $\mu\text{V/K}$ for the same doping concentrations [89]. The negative values of $|S|$ confirm the n-type semiconducting nature of both pure and Mg-doped ZnO films. As the Mg concentration increases, the absolute value of the Seebeck coefficient decreases with rising temperature. This behavior is attributed to the increase in free electron concentration, which enhances the carrier density within the ZnO thin films. Given the inverse relationship between the Seebeck coefficient and carrier concentration, the peak Seebeck coefficient at 120 K for ethanol-based films is recorded at $-400\mu\text{V/K}$ for undoped ZnO, whereas for methanol-based films, the maximum value is $-525\mu\text{V/K}$ at 80 K. However, at 9% Mg doping, the Seebeck coefficient drops to $-226\mu\text{V/K}$ and $-210\mu\text{V/K}$ for ethanol and methanol-based films, respectively, indicating a significant decline at higher doping levels [90]. This reduction in the Seebeck coefficient is accompanied by an increase in electrical conductivity due to the greater availability of free electrons. Simultaneously, the thermal conductivity decreases, which can be linked to the improved preservation of the organized pore architecture in mesoporous ZnO relative to its undoped counterpart [90].

The Seebeck coefficient (measured in $\mu\text{V/K}$) is derived from the following connection

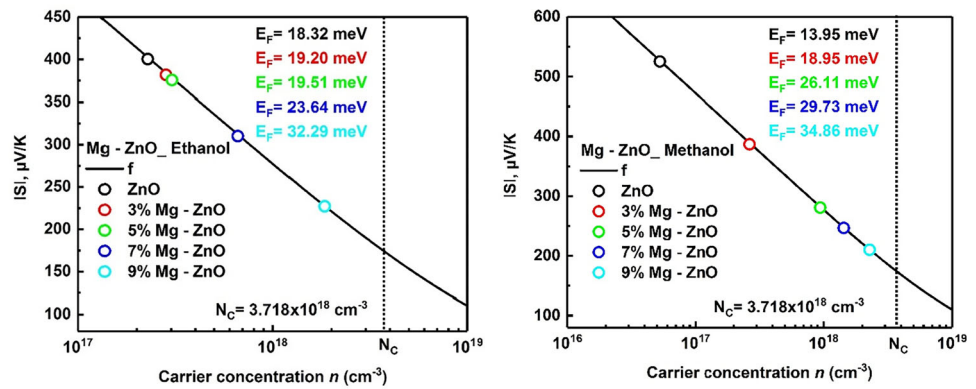
$$S = -\frac{\Delta v}{\Delta T} \quad (6)$$

where Δv is the offset voltage recorded when the temperature differential ΔT exists between the two tip contacts on the sample ($V_{\text{hot tip}} - V_{\text{cold tip}}$). The progression of the thermoelectric coefficient is directly proportional to the temperature gradient between the tips [91].

The Fermi energy (E_F) has been determined using the following equation [79]:

$$E_F = \frac{\pi^2 K_B^2 T}{3|e||s|} \quad (7)$$

Fig. 11 Seebeck coefficients plotted against carrier concentrations for ZnO and Mg-doped ZnO thin films for both solvents (Ethanol- Methanol)



where e represents the electron charge, k_B denotes the Boltzmann constant, and T signifies the absolute temperature.

Figure 11 presents the variation in Fermi energy values with different Mg doping concentrations for ZnO thin films synthesized using ethanol and methanol as solvents. The results indicate that the Fermi level increases as the magnesium doping concentration rises for both solvents. This trend suggests an enhancement in the crystal quality and a more uniform distribution of impurities within the Mg-doped ZnO thin films. The observed increase in the Fermi level implies that a higher energy input is required for electron transport in these doped films, which can be attributed to the increased carrier concentration and the modification of the electronic structure due to Mg incorporation. These findings further support the idea that controlled Mg doping can effectively influence the electronic properties of ZnO thin films, making them suitable for various optoelectronic and thermoelectric applications.

Carrier concentration was calculated using Eq. (8), while the effective density of states in the conduction band was determined using Eq. (9) [91]:

when $|S| > 75 \mu\text{V/K}$: [91]

$$m_s^* = \frac{h^2}{2K_B T} \left\{ \frac{3n}{16\sqrt{\pi}} \left(\exp \left[\frac{|S|}{(K_B/e)} - 2 \right] - 0.17 \right) \right\}^{2/3} \quad (8)$$

$$N_C = 2 \left(\frac{2\pi m_s^* K_B T}{h^2} \right)^{3/2} \quad (9)$$

In this context, e signifies the charge of the carrier, k_B represents Boltzmann's constant, T indicates the absolute temperature, h denotes the Planck constant, m_s^* (where $m_s^* = 0.28 m_0$) refers to the effective Seebeck mass of ZnO, and n symbolizes the carrier concentration [92].

Figure 11 illustrates the carrier concentration findings of the ZnO and MZO thin films. The carrier concentration values increased with the doping level, with values of

2.27×10^{17} , 2.82×10^{17} , 3.03×10^{17} , 6.62×10^{17} , and $1.84 \times 10^{18} \text{ cm}^{-3}$ for undoped ZnO, 3% MZO, 5% MZO, 7% MZO, and 9% MZO films, respectively, deposited using ethanol as solvent. The corresponding values obtained using methanol as solvent were 5.26×10^{16} , 2.65×10^{17} , 9.43×10^{17} , 1.43×10^{18} , and $2.29 \times 10^{18} \text{ cm}^{-3}$ for undoped ZnO, 3% MZO, 5% MZO, 7% MZO, and 9% MZO films. The increase in carrier concentration due to doping is attributed to the increased free carrier density in ZnO, which enhances electrical conductivity. The calculated effective density of ZnO, amounting to $3.71 \times 10^{18} \text{ cm}^{-3}$, indicates that the ZnO and MZO layers exhibit non-degenerate behavior. This indicates that the material maintains its ideal thermoelectric characteristics, making it appropriate for many applications in thermoelectric devices. [42] The observations indicate that the carrier concentration of both methanol and ethanol rises with increased magnesium doping. At the maximum magnesium concentration 9% Mg-doped ZnO, methanol shows a markedly elevated carrier concentration ($2.29 \times 10^{18} \text{ cm}^{-3}$) in contrast to ethanol ($1.84 \times 10^{18} \text{ cm}^{-3}$). This study indicates that films deposited from methanolic solutions have enhanced carrier concentration characteristics at increased magnesium doping levels.

3.7 UV photodetector

Figure 12 illustrates the time-dependent photoresponse of Mg-doped ZnO (MZO) thin films synthesized using ethanol and methanol as solvents. Zou et al. [93] demonstrated that ZnO thin film UV photodetectors exhibit varied photoresponses when exposed to UV light, depending on the solvent used. The photoresponse measurement was conducted by modulating the UV light source ($10 \mu\text{W/cm}^2$) at a 5 V bias, and the resulting photocurrent was recorded as a function of time (I - t) with 30-s intervals for switching UV illumination on and off [94]. This analysis was aimed at examining the rise and decay characteristics of the photocurrent.

Fig. 12 Photocurrent as a function of time (Lt) for films deposited using (a) ethanol and (b) methanol as solvents, during the activation and deactivation of UV light at 30-s intervals

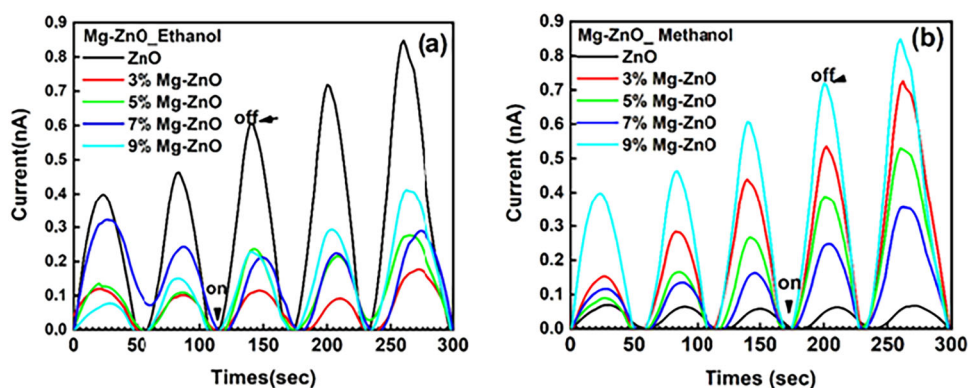
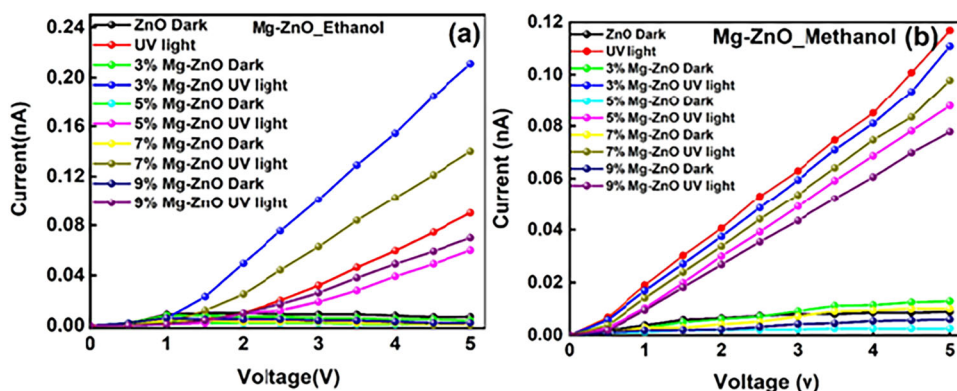


Fig. 13 The I-V curves of ZnO and MZO samples measured in the dark and under UV illumination utilizing: a ethanol and b methanol as solvents



For all thin films, the photocurrent increased sharply upon light exposure due to the generation of electron-hole pairs, which enhanced the electrical conductivity of the semiconductor material when light energy exceeded the band gap energy [45]. Once the light was turned off, the photocurrent returned to its initial state within approximately 135 s due to charge carrier recombination [95]. The presence of defects or trap states in ZnO played a significant role in trapping charge carriers and affecting the recombination process [96].

As shown in Fig. 12a, ZnO thin films synthesized with ethanol exhibited a decreasing trend in photocurrent with increasing Mg doping, with the highest peak at 0.84 nA for undoped ZnO and a decline to 0.17 nA for 3% Mg-doped ZnO. Conversely, as seen in Fig. 12b, methanol-based ZnO films showed an increasing photocurrent trend with Mg doping, reaching a maximum of 0.84 nA for 9% Mg-doped ZnO, while the lowest photocurrent value was 0.06 nA for pure ZnO. The enhanced photoresponse in MZO thin films synthesized using both solvents can be attributed to improved crystallinity and conductivity at optimal doping concentrations [97]. Unpaired photo-generated electrons and available holes facilitated photoconduction under an external driving force [44]. Additionally, the photocurrent under UV illumination (365 nm) was consistently higher than the dark current for both solvents.

Mg doping played a crucial role in introducing electron donors to enhance photodetector performance. Moreover, Mg incorporation reduced lattice defects, as confirmed by XRD analysis, minimizing mid-gap or trap states within the bandgap region [98]. These defect states could hinder electrical excitation from the valence band to the conduction band, leading to prolonged rise and decay times, particularly for undoped ZnO samples [99].

Density functional theory (DFT) simulations offer an alternative approach to investigating the electronic structure and can provide significant insights into the modifications induced by Mg doping using different solvents. These mathematical analyses enable a deeper understanding of variations in band alignment, defect formation energies, and charge carrier mobility, which in turn elucidate solvent-dependent doping effects. Specifically, DFT simulations can validate the role of methanol in stabilizing Mg incorporation and reducing deep-level trap states, aligning with experimental findings [100].

Figure 13 presents the typical I-V characteristics of ZnO and MZO thin films in darkness and under UV light exposure (365 nm, 5 V) at room temperature, using methanol and ethanol as solvents. The linear and symmetric I-V curves confirm the effective ohmic contact between the Mg:ZnO layer and Ag electrodes [101], which is crucial for improved photosensitivity [102]. This ohmic behavior is

Table 5 Photodetector performance of ZnO and MZO films deposited from ethanol and methanol, at 5 V bias

Samples	Ethanol			Methanol		
	I_{dark} (nA)	I_{photo} (nA)	Sensitivity	I_{dark} (nA)	I_{photo} (nA)	Sensitivity
Pure ZnO	0.007	0.09	11.9	0.008	0.116	13.5
3% Mg	0.004	0.21	51.8	0.012	0.110	8.6
5% Mg	0.001	0.06	59	0.002	0.087	42.5
7% Mg	0.001	0.14	139	0.010	0.097	8.7
9% Mg	0.002	0.07	34	0.006	0.077	11.8

attributed to the good contact properties of the Mg:ZnO film. The slope of the I-V curve increases with forward bias, likely due to heavy doping [103–105].

The dark current (I_d) remains consistently low and stable (<0.007 nA for ethanol-based films and <0.012 nA for methanol-based films) even at a 5 V bias. This phenomenon is primarily due to electron trapping by adsorbed oxygen molecules [95–106]. Under UV illumination, both pure and Mg-doped ZnO samples exhibit an increase in photocurrent as a result of electron-hole pair generation. The highest photocurrent (0.211 nA at 5 V) is observed for ethanol-based samples with 3% Mg doping, whereas undoped ZnO synthesized with ethanol exhibits a higher photocurrent (0.116 nA) compared to methanol-derived films [107].

The higher number of oxygen vacancy defects in methanol-based films lead to increased recombination, thereby reducing photocurrent. In contrast, ethanol-based samples exhibit fewer oxygen vacancy defects, resulting in an expanded depletion zone, enhanced electric field, and improved carrier separation, which collectively contribute to higher photocurrent. This enhancement is particularly evident at 3% and 7% Mg doping levels [103]. The reduction in deep-level trap states further optimizes charge carrier dynamics. Methanol-derived films, however, contain a greater density of oxygen vacancies, which act as recombination centers, negatively affecting photocatalytic activity. These findings highlight the critical role of solvent selection in tailoring defect-related properties for optimized photocatalytic performance.

The sensitivity values of undoped ZnO and Mg-doped ZnO thin films prepared from both solvents are presented in Table 5. The photosensitivity of the device exhibits a significant enhancement under white light illumination with bias, demonstrating ohmic behavior, as indicated by the notable increase in the $(I_{\text{light}} - I_{\text{dark}})/I_{\text{dark}}$ ratio [105]. Notably, 5% Mg-doped ZnO layers synthesized using methanol show heightened sensitivity, albeit with a reduced current of 0.087 nA. Conversely, 7% Mg-doped ZnO layers prepared with ethanol exhibit superior sensitivity compared to both pure ZnO films and methanol-derived ZnO films, with a moderate current of 0.14 nA. These findings are consistent with the observations of Yen-Lin Chu et al. [108], who reported that Ga-doped ZnO thin films

synthesized through straightforward chemical techniques display enhanced photoresponsivity and stability under 380 nm UV irradiation. The incorporation of Mg into the ZnO lattice plays a crucial role in modulating the preferred crystal orientation, surface morphology, and oxygen vacancy density, which influences oxygen adsorption and enhances the suitability of MZO thin films for ultraviolet light detection [109, 110]. Furthermore, ZnO films fabricated using ethanol as a solvent exhibit a significantly higher photocurrent response sensitivity to UV light intensity, reaching 139%, compared to those prepared using methanol solvent [111].

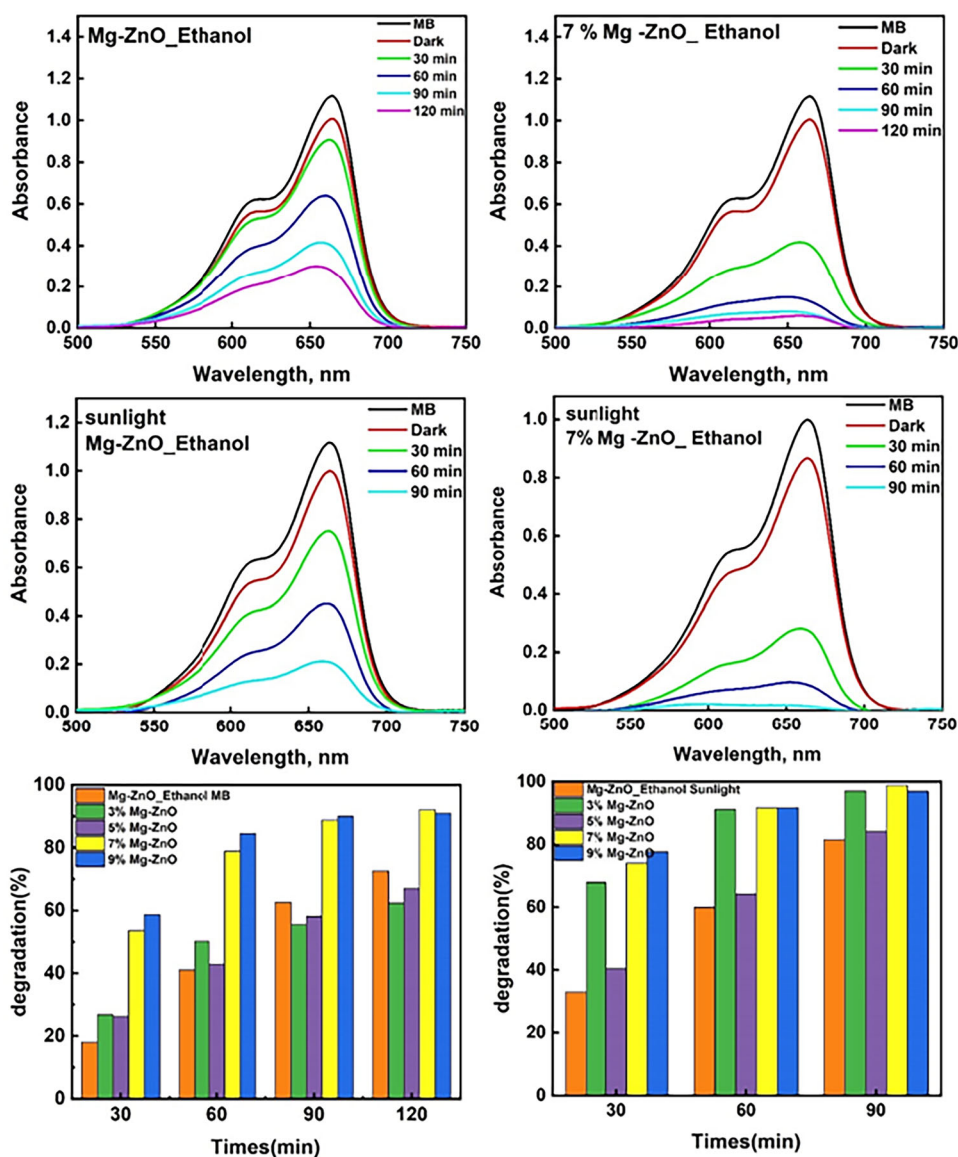
3.8 The photocatalytic performance of the Mg-doped ZnO thin films

The successful fabrication of undoped and Mg-doped ZnO thin films enables an in-depth investigation of their photocatalytic activity. The photocatalytic degradation of model contaminants, MB and MO, was carried out at ambient temperature [43]. The results, illustrated in Figs. 14, 15, indicate that all ZnO thin films exhibit significant photocatalytic activity, achieving nearly complete decolorization of MB in both solvents under UV light and sunlight exposure.

However, the inclusion of Mg-doped ZnO (MZO) thin films enhances photocatalytic activity, leading to a faster decrease in the peak intensity of the MB absorption spectrum. Specifically, 9% Mg-doped ZnO demonstrates the highest photocatalytic activity for MB degradation in methanol solvent under UV light irradiation within 120 minutes and under sunlight irradiation within 60 min. Similarly, for the ethanol solvent, 7% Mg-doped ZnO exhibits the most effective catalytic activity within 120 min under UV irradiation. Notably, the photocatalytic efficiency improves further under sunlight exposure, achieving superior degradation outcomes within 90 min for 7% Mg-doped ZnO. Additionally, replacing MB with MO dye results in observable variations in photocatalytic performance, as depicted in Fig. 16.

The progressive decline in the intensity of primary absorption peaks under prolonged UV-visible light exposure highlights the photocatalytic activity of both undoped

Fig. 14 Absorption spectra of MB aqueous solution and its degradation as a function of irradiation time with sunlight and UV illumination for ZnO and MZO thin films synthesized utilizing ethanol solvents



and Mg-doped ZnO thin films prepared using methanol and ethanol as solvents. Notably, 3% Mg-doped ZnO achieves complete degradation of MO within 180 min in ethanol, whereas 9% Mg-doped ZnO demonstrates superior performance, fully degrading the MO solution in just 120 minutes in methanol. This enhanced photocatalytic efficiency resulting from Mg doping can be attributed to several factors.

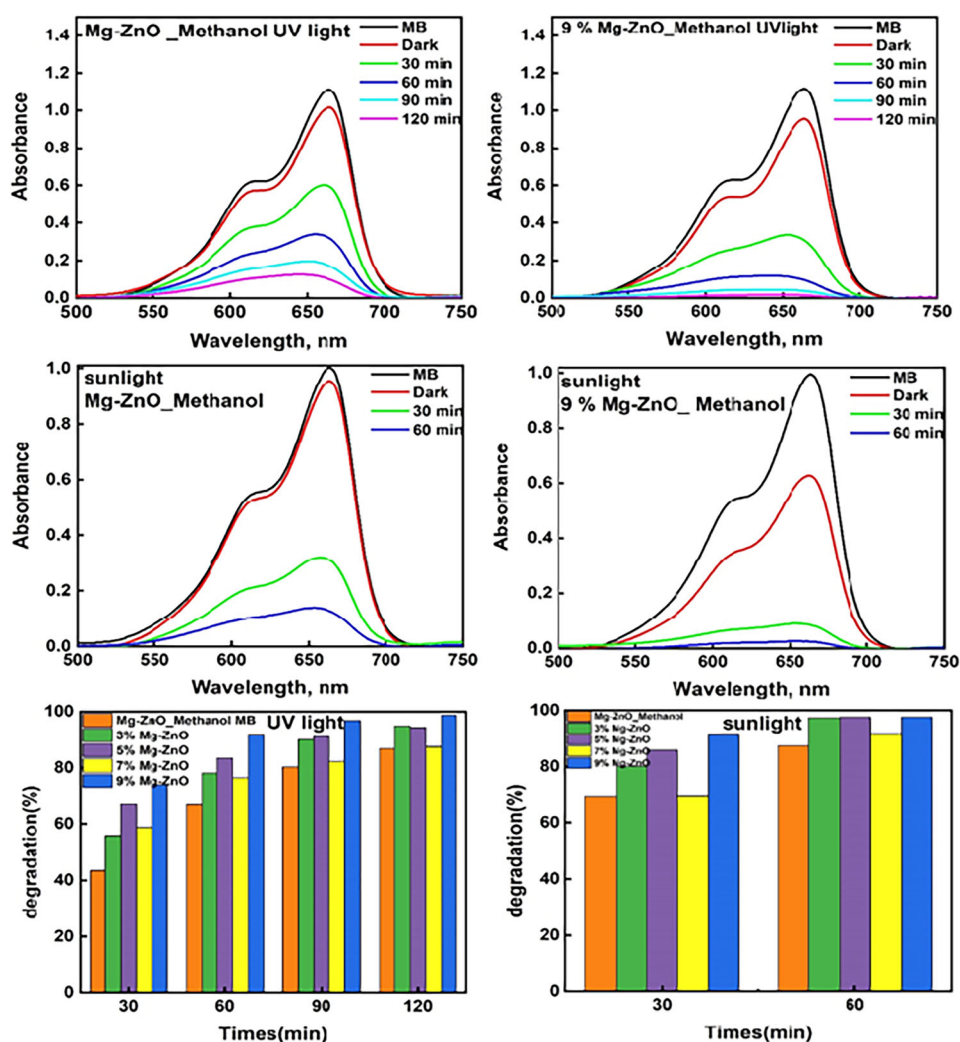
Key parameters such as surface defects, surface area, and surface reactivity significantly influence photocatalytic performance [112]. Additionally, characteristics like morphology, specific surface area, defects, and surface hydroxyl groups play a crucial role in determining the overall efficiency of semiconductor photocatalysts [113, 114]. As the catalyst concentration increases, more active ZnO-Mg sites become available to absorb photons and generate electron-

hole pairs, thereby accelerating the photodegradation process. Etacheri et al. found that Mg doping enhanced the photocatalytic efficiency of ZnO, likely due to improved structural properties and more effective electron-hole separation [115, 116].

Upon UV irradiation, ZnO-Mg absorbs photons, exciting electrons to transition from the valence band (VB) to the conduction band (CB), leading to the formation of electron-hole pairs. The improved photocatalytic activity is attributed to the efficient transfer of these charge carriers and the suppression of recombination, ensuring a higher degradation rate of organic pollutants [98]. The degradation efficiency (%) was ascertained using the formula (10) [117]:

$$D\% = \frac{C_0 - C_t}{C_0} \times 100 \quad (10)$$

Fig. 15 Absorption spectra of MB aqueous solution and its degradation as a function of irradiation time with sunlight and UV illumination for ZnO and MZO thin films synthesized utilizing methanol solvents



where C is the initial concentration of MB and MO solution, C_t is the concentration of MB and MO at time t , and the photocatalytic activity is measured under sunlight and UV irradiation.

The correlation between degradation efficiency and time is summarized in Table 6. The highest degradation efficiency for MB was observed with 7% Mg-doped ZnO films prepared using ethanol as solvent (92.1%) and 9% Mg-doped ZnO for deposition from methanol (98.8%) within 120 minutes [118]. When MO replaced MB in, an even greater degradation efficiency of 99.9% was achieved within the same duration for 9% Mg-doped ZnO.

In contrast, when using ethanol as the deposition solvent, the degradation efficiency decreased to 97.5%, requiring an extended reaction time of 180 minutes for 3% Mg-doped ZnO. Notably, the photocatalytic performance significantly improved under sunlight exposure, particularly when using methanol as a deposition solvent, where 9% Mg-doped ZnO

reached an absorption rate of 97.7% within just 60 minutes. Similarly, using ethanol as deposiotion solvent, 7% Mg-doped ZnO demonstrated a high degradation efficiency of 98.8% within 90 minutes. These results highlight the crucial role of solvent selection and light exposure conditions in optimizing photocatalytic activity.

The photocatalytic degradation kinetics of ZnO and MZO thin films were examined employing the Langmuir-Hinshelwood (L-H) model [119, 120]. According to the L-H model, the reaction rate (R) is proportional to the fraction of the surface encompassed by the reactant (θ) and can be articulated as:

$$R = -\frac{dc}{dt} = krk\theta = krkC/(1 + kC) \quad (11)$$

Here, C represents the reactant concentration, kr denotes the reaction rate constant, k signifies the adsorption equilibrium constant, and θ indicates the fraction of the surface occupied by reactants. When the initial concentration of the

Fig. 16 Influence of Mg doping on the photocatalytic degradation of aqueous MO solution when exposed to UV light for films prepared using both solvents (ethanol-methanol)

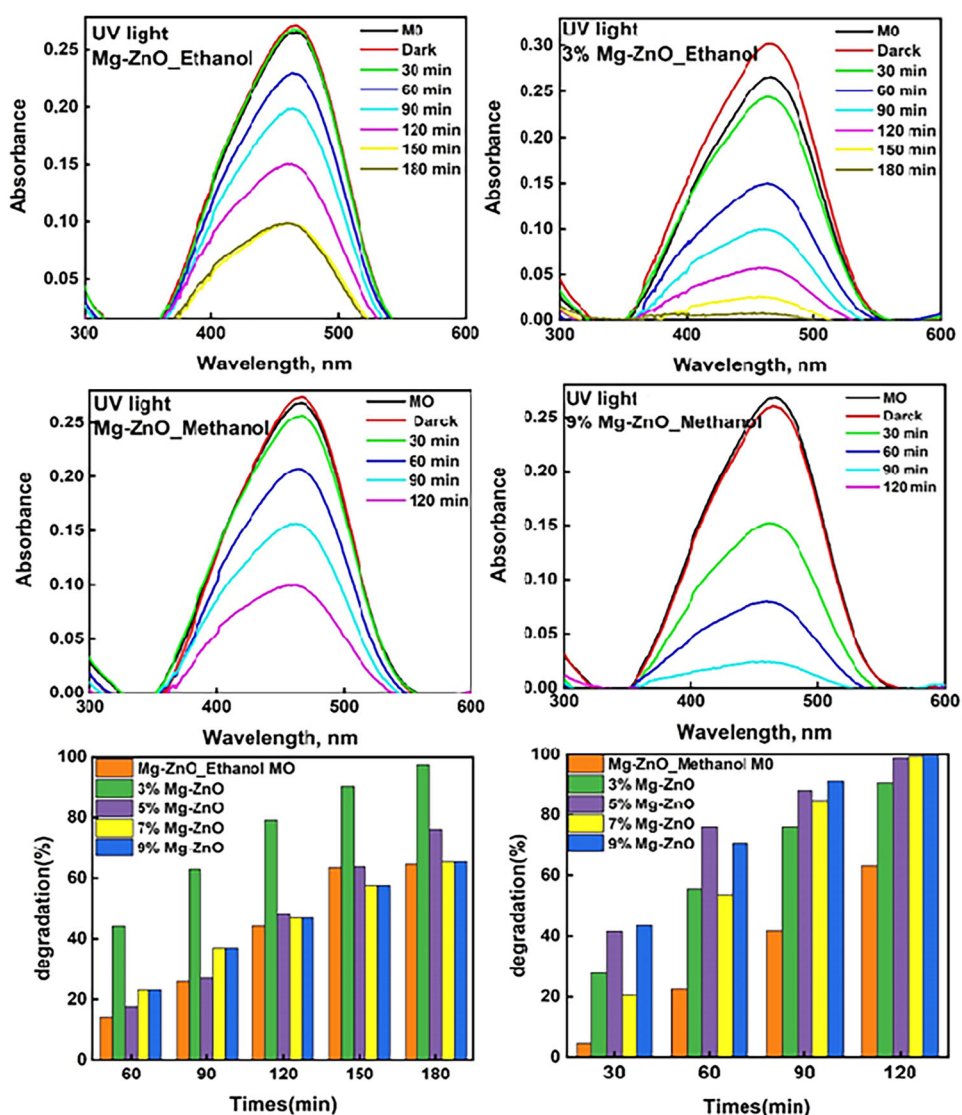


Table 6 Reaction rate constants (k) and degradation efficiency (D) values for the kinetic data fit by the first-order model for MB and MO dye solutions with ZnO and Mg-doped ZnO thin films under UV light and sunlight irradiation (k_n , $n = \% \text{ Mg loading}$)

	Ethanol			Methanol		
	Times (min)	D (%)	k (min^{-1})	Times (min)	D (%)	k (min^{-1})
MB UV light	120	92.1	$k_7 = 0.023 \pm 0.002$	120	98.8	$k_9 = 0.028 \pm 0.002$
MO UV light	180	97.5	$k_3 = 0.016 \pm 0.001$	120	99.9	$k_9 = 0.040 \pm 0.008$
MB Sunlight	90	98.8	$k_7 = 0.049 \pm 0.004$	60	97.7	$k_9 = 0.06 \pm 0.01$

organic compound is minimal ($KC \ll 1$) the equation simplifies to a pseudo-first-order reaction:

$$\frac{dc}{dt} = krkc \quad (12)$$

Integrating this equation yields:

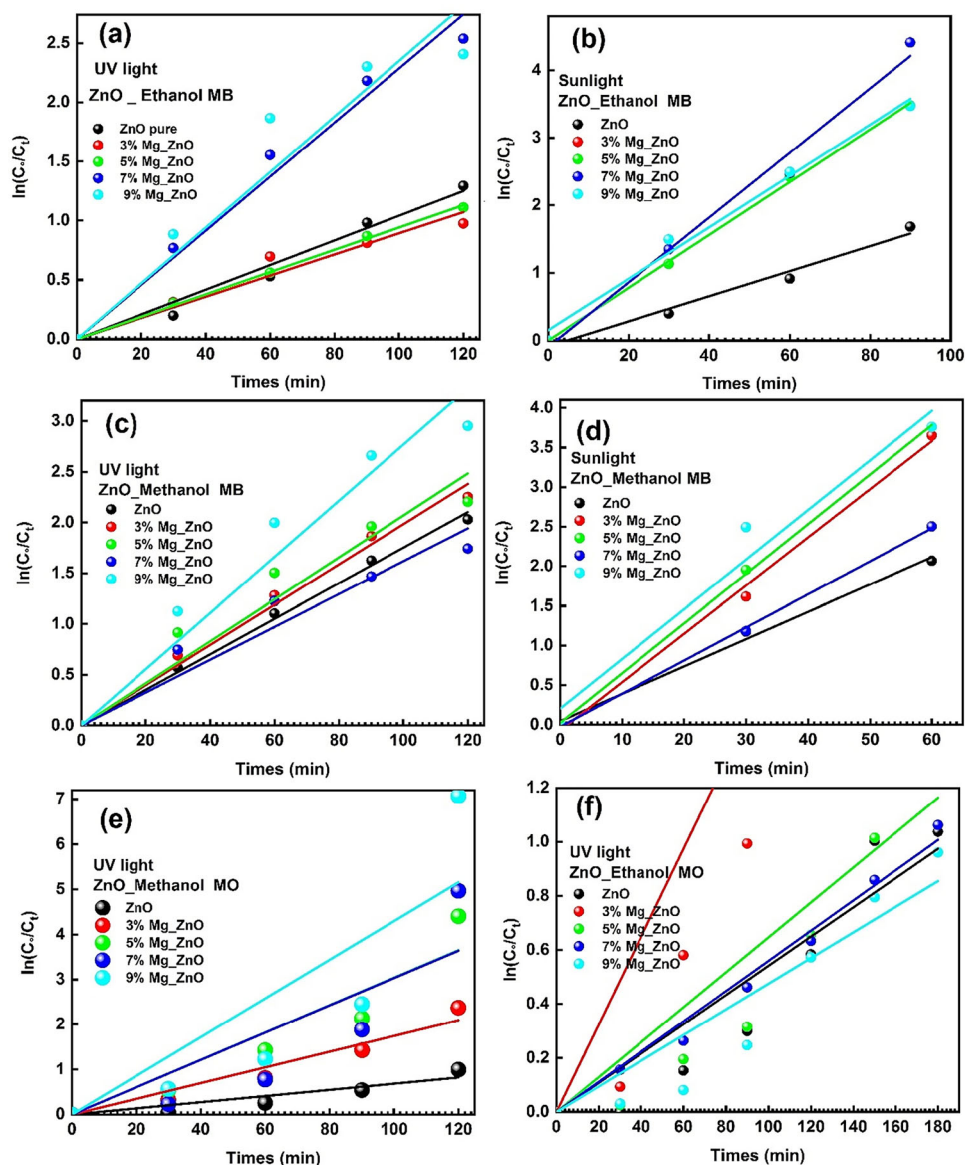
$$\ln(C_0/C_t) = kt \quad (13)$$

where, k signifies the rate constant for a first-order reaction.

Figure 17 illustrates the relationship between $\ln(C_0/C_t)$ and time, demonstrating the photocatalytic degradation kinetics of MB and MO dye solutions in ethanol and methanol solvents. The degradation rate constant (k) is determined from the slope of these fitted curves, providing insight into the efficiency of pure ZnO and Mg-doped ZnO thin films (3%, 5%, 7%, and 9% Mg content) under both UV illumination and sunlight exposure.

The first-order rate constants (k), summarized in Table 6, confirm the enhanced photocatalytic performance of

Fig. 17 **a** Variation of $\ln(C_0/C_t)$ as functions of reaction time for MB degradation using Ethanol solvent under UV Light, **b** variation of $\ln(C_0/C_t)$ as functions of reaction time for MB degradation using Ethanol solvent under sunlight irradiation, **c** variation of $\ln(C_0/C_t)$ as functions of reaction time for MB degradation using Methanol solvent under UV Light, **d** variation of $\ln(C_0/C_t)$ as functions of reaction time for MB degradation using Methanol solvent under sunlight irradiation, **e** variation of $\ln(C_0/C_t)$ as functions of reaction time for MO degradation using Methanol solvent **f** variation of $\ln(C_0/C_t)$ as functions of reaction time for MO degradation using Ethanol solvent



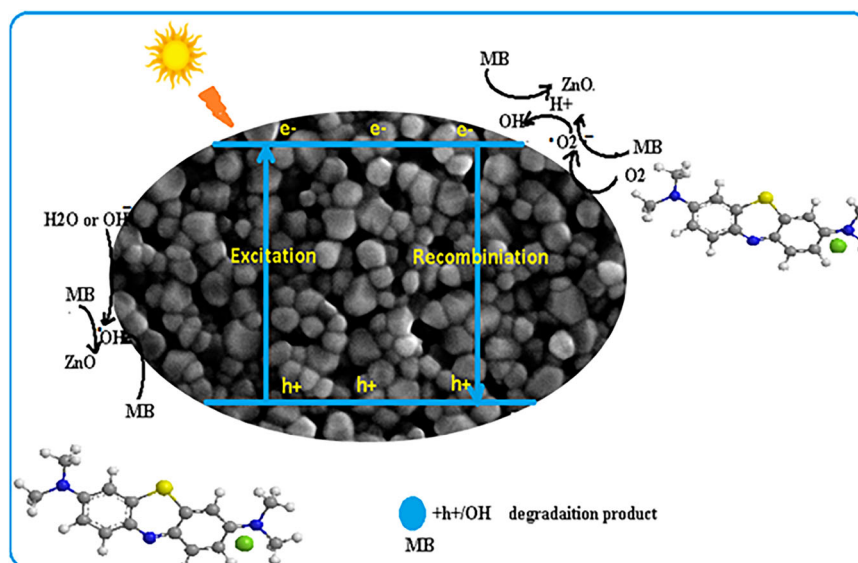
Mg-doped ZnO compared to undoped ZnO. The results highlight the influence of Mg concentration and solvent type on photocatalytic activity, with MZO thin films demonstrating superior efficiency under both UV and sunlight irradiation [2].

Among all conditions, the highest first-order rate constant (k) was found to be $k_9 = 0.06 \text{ min}^{-1}$ for 9% Mg-doped ZnO prepared in methanol solvent with aqueous MB dye solution under sunlight exposure. This is followed by $k_9 = 0.04 \text{ min}^{-1}$ for MO dye solution and $k_9 = 0.028 \text{ min}^{-1}$ for MB dye solution under UV illumination. When ethanol is used as the deposition solvent, a decrease in k values is observed, with $k_7 = 0.049 \text{ min}^{-1}$ for 7% Mg-doped ZnO in MB dye solution under sunlight, and $k_7 = 0.023 \text{ min}^{-1}$ and $k_3 = 0.016 \text{ min}^{-1}$ under UV exposure for MB and MO dye solutions, respectively.

These results emphasize that photocatalytic performance is influenced by factors such as pollutant type, solvent selection, and Mg doping level. The most efficient degradation occurs with 9% Mg-doped ZnO thin films deposited from methanol solvent under sunlight, demonstrating the crucial role of Mg incorporation in enhancing photocatalytic activity.

Photodegradation involves the oxidation and reduction of dye molecules, driven by electrons generated within the semiconductor under light exposure [121]. The enhanced photocatalytic efficiency of ZnO upon Mg doping can be attributed to several factors. The increase in band-gap energy due to Mg incorporation raises the redox potential of photogenerated electron-hole pairs, thereby improving photocatalytic performance. Additionally, Mg doping extends the recombination time of

Fig. 18 Degradation mechanism of MB solution by ZnO thin film under sunlight irradiation



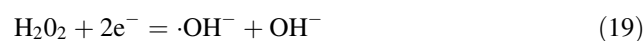
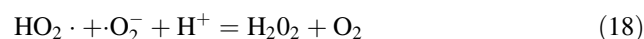
these charge carriers, promoting the formation of hydroxyl radicals, which play a key role in the degradation process [25].

Methanol-based Mg-doped ZnO thin films exhibit superior photocatalytic activity compared to ethanol-based films, primarily due to their higher crystallinity, lower defect density, and improved charge carrier separation. Furthermore, methanol facilitates the development of larger crystallites with elevated surface energy, hence improving light absorption and charge transfer efficiency. These structural, optical, and electrical improvements collectively contribute to the superior photocatalytic performance observed in methanol-based films.

Figure 18 illustrates the photocatalytic degradation mechanism of ZnO thin films. Under light exposure, electrons (e^-) are excited from the valence band to the conduction band, leaving behind holes (h^+) in the valence band. These charge carriers interact with water molecules and oxygen in the environment, generating hydroxyl ($\text{OH}^\cdot$) and superoxide ($\text{O}_2^\cdot$) radicals, which oxidize and break down organic dye molecules [2, 43].

The solvent choice significantly influences the interaction of Mg ions with the ZnO lattice, affecting defect formation and charge carrier dynamics. Methanol, with its superior solvation capacity and lower steric hindrance, enhances the dispersion of Mg ions, reducing defect clustering and promoting uniform lattice substitution. In contrast, ethanol's bulkier molecular structure may limit Mg incorporation, leading to localized strain and additional defect states. These variations impact charge carrier lifetime, with methanol-based films exhibiting lower recombination rates and higher charge mobility—key factors in optimizing photocatalysis.

The photocatalytic discoloration of dyes is presumed to occur through the following mechanism [117]:



ZnO nanostructures have recently drawn significant attention in photocatalytic applications due to their unique structural and electronic properties. These nanostructures can adopt diverse morphologies, ranging from one-dimensional (1D) forms like nanowires, nanorods, and nanotubes to two-dimensional (2D) and three-dimensional (3D) configurations such as nanodisks, nanoflowers, nanocubes, and nano-urchins. Each morphology offers distinct surface characteristics, polar facets, and oxygen vacancy distributions, which directly impact their photocatalytic efficiency [122].

Methanol-based ZnO films exhibit superior crystallinity and fewer structural defects, enhancing their stability and resistance to photo-corrosion. The reduction in deep-level trap formation minimizes unwanted degradation pathways, ensuring sustained photocatalytic performance. In contrast, ZnO films synthesized using ethanol tend to have a higher

defect density, making them more susceptible to degradation over time due to increased grain boundary oxidation. These findings highlight methanol as a more suitable solvent for producing durable and efficient photocatalysts.

The choice of solvent plays a crucial role in regulating the nucleation and growth dynamics of Mg-doped ZnO thin films, directly impacting their band structure and optical properties. Methanol, with its higher volatility and polarity, promotes the formation of compact and uniform thin films with minimal grain boundary defects. This results in an upward shift of the conduction band minimum and a reduction in non-radiative recombination sites, thereby improving optical absorption and widening the bandgap. Conversely, ZnO films synthesized using ethanol often exhibit increased surface roughness, which can affect light absorption and overall photocatalytic performance.

4 Conclusion

This study provides an in-depth comparative analysis of Mg-doped ZnO thin films synthesized using methanol and ethanol solvents. The findings confirm that by adjusting the precursor solvent and Mg doping concentration, the optical, structural, morphological, and electrical properties of ZnO can be fine-tuned for diverse optoelectronic applications. Both ethanol- and methanol-derived films exhibit high crystallinity, with XRD analysis revealing a hexagonal wurtzite structure. Notably, 9% Mg-doped ZnO films synthesized with ethanol exhibit an increased crystallite size of 22 nm, while 7% Mg-doped ZnO films prepared with methanol display an even larger crystallite size of 23 nm.

FESEM analysis shows a uniform grain distribution, with methanol-based films demonstrating larger grain sizes (92 nm). EDS and FTIR spectroscopy confirm the successful incorporation of magnesium. UV-Vis measurements reveal an average transmittance of 75–90%, with ethanol-based films achieving the highest transparency (90%). The optical bandgap increases with Mg concentration, reaching 3.34 eV for methanol-based films. Photoluminescence studies indicate enhanced UV emission intensity with increasing Mg doping.

In terms of photocatalytic activity, methanol-derived ZnO films outperform their ethanol-based counterparts, attributed to their higher carrier concentration at increased Mg levels. Photocurrent measurements further demonstrate enhanced photoresponse sensitivity, with ethanol-based films showing exceptional sensitivity, particularly for the 7% Mg-doped ZnO sample, which reaches a sensitivity value of 139. Analytical evaluations reveal that 9% Mg-doped ZnO films synthesized via the methanol-assisted sol-gel process and tested with methylene blue as a pollutant exhibit superior photocatalytic efficiency

under solar radiation compared to ethanol-based ZnO films.

These results highlight the beneficial effects of Mg doping and solvent selection in developing highly efficient, cost-effective ZnO photocatalysts for environmental remediation. The enhanced photocatalytic activity of these films is primarily attributed to the suppression of electron-hole recombination, facilitated by charge trapping sites and increased surface area due to pore formation induced by Mg doping. Overall, this study underscores the crucial role of solvent choice in optimizing the quality and functional properties of ZnO thin films, paving the way for their application in photocatalysis and optoelectronic devices.

Data availability

No datasets were generated or analyzed during the current study

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Author contributions H.F collected and analyzed the literature, wrote much of sections “Introduction” and wrote the remaining portions, as well as the perspective and prospective components. All authors reviewed the manuscript. A.D : Review and correct the article S.I: advices and guidances

Compliance with ethical standards

Conflict of interest The authors declare no competing interests.

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