Environmentally Benign Structural, Topographic, and Sensing Properties of Pure and Al-Doped ZnO Thin Films

Krishna Chandra Dubey, Anam Zaidi, and Ram Raseele Awasthi*

ABSTRACT: In the present research work, Zn$_{1-x}$Al$_x$O thin films with varying proportions of Al ($x = 0.00, 0.01, 0.02,$ and $0.03$) are prepared by a chemical sol–gel spin-coating technique. The crystal structural, morphological, and humidity-sensing properties of the synthesized Zn$_{1-x}$Al$_x$O thin films, with varying concentrations of Al ($x = 0.00, 0.01, 0.02,$ and $0.03$), were characterized by X-ray diffraction (XRD) and field-emission scanning electron microscopy (FE-SEM); a special humidity-controlled chamber was designed for the humidity-sensing studies. In structural and phase analyses, XRD patterns of Zn$_{1-x}$Al$_x$O thin films show a hexagonal wurtzite crystal structure. The average crystallite sizes of Zn$_{1-x}$Al$_x$O thin films were calculated and found to be $\sim 18.00, 22.50, 26.30,$ and $29.70$ nm using the X-ray diffraction (XRD) pattern. The surface morphology of Zn$_{1-x}$Al$_x$O Al ($x = 0.00, 0.01, 0.02,$ and $0.03$) thin films obtained from AFM micrographs analysis indicates the modification of the spherical grains into nanorods, which were distributed throughout the surface of the films. The SEM image of 3 wt % Al-doped ZnO nanomaterials also shows that spherical nanoparticles changed to nanorod-like structures with a high packing density. Furthermore, increasing the Al-doping concentration from 0 to 3 wt % in ZnO NPs shows lower hysteresis loss, less aging effect, and good sensitivity in the range of $9.8–16.5$ MΩ/%RH. The sensitivity of the sensing materials increased with increasing Al-doping concentration, which is very useful for humidity sensors.

1. INTRODUCTION

In the present scenario, thin films are very advantageous for various potential applications in the field of drug pharmaceutical compound, nanomedicine, optoelectronics, photocatalysts, ferroelectrics, and piezoelectrics. Nanostructures have unique physical, chemical, and biological properties at the nanoscale level compared with the corresponding particles at the microscale level. The optical bandgap engineering of synthesized ZnO nanostructures is a current area of sustained interest for developing transparent photonic devices as well as transparent conducting electrodes. The surface morphology of the transparent conducting layer of Al-doped ZnO thin films has been studied by a few researchers; it was found to have a nanowire-like structure and is used for dye-sensitized solar cell applications. The physical and chemical properties of NPs can be improved by some unequivocal modification processes. Different metal-doped ZnO tetrapods with bismuth and tin oxide hybrid nanostructures such as ZnO–Bi$_2$O$_3$ and ZnO–Zn$_2$SnO$_4$ show good performance in humidity and gas-sensing applications. Compound semiconductors having a wide optical band gap, such as MoS$_2$, ZnS, CdS, SnO$_2$, TiO$_2$, ZnO etc., with different nanostructures have attracted considerable attention from the scientific community due to their unique characteristic properties and prevalent applications in nanoscale devices. Among these, zinc oxide (ZnO) semiconductors are widely investigated nanomaterials due to their large exciton binding energy of nearly $\sim 60$ meV and direct optical bandgap energy ($\sim 3.37$ eV) at 300 K. As a result of their efficient optical properties, ZnO nanostructures have become extremely appropriate host semiconductors; codoping of rare-earth ions is the most effective approach to enhance their structural, morphological, bandgap energy, and optical properties. Many authors have also reported that ZnO thin films with different dopants, such as Ga, In, F, Al, Ca, modified the electrical conductivity and optical transparency of nanostructures for optoelectronic device applications. In the present epoch, numerous efforts have been made by physicists and researchers toward the fabrication of ZnO nanomaterials that are simple, low cost, easy to synthesize, efficient, and suitable for large-scale production for potential applications. Therefore, several techniques have been employed to fabricate ZnO nanomaterials...
als, such as the sol–gel method, solid-state reactions, coprecipitation, hydrothermal methods, pulse laser deposition, magnetic sputtering, electrospinning, electron gun evaporation, spray pyrolysis technique, etc. The abovementioned techniques are very expensive and involve high temperatures to improve the crystalline quality of the films. However, high temperatures lead to agglomeration and make the crystallite size bigger, which hinders the potential application of the fabricated sample. Therefore, to avoid conditions of high temperature as well as agglomeration, the spin-coating technique is used for the synthesis of Al-doped ZnO thin films for humidity-sensing applications. As a result, spin-coating is found to be a feasible, cost-effective, and reproducible technique. Moreover, the effect of the lattice mismatch and the extinction coefficient between the film and sample holders on the presented characteristics cannot be ignored. These parameters directly affect the physical and chemical properties of ZnO NPs already reported in the literature. Furthermore, many research groups have investigated the humidity-sensing properties of Al-doped ZnO nanomaterials and found them to possess high sensitivity, a low response time, and low power expenditure. In the present investigation, pure as well as Al-doped nanocrystalline ZnO thin films are fabricated by a simple, low-cost, easy-to-dope, and large-scale chemical solution spin-coating deposition technique. The structural and surface morphological analyses are performed by XRD and AFM/SEM techniques for Al-doped ZnO thin films. The surface morphology is evidently changed by an increased Al-doping content. The humidity and gas-sensing characteristics of Al-doped nanocrystalline ZnO thin film sensors offer good reproducibility and better stability as compared with undoped ZnO nanostructures. The sensor performance can be noticeably improved by tailoring the Al-doping concentration in ZnO thin films for their realistic humidity-sensing device applications (Table 1).

2. EXPERIMENTAL SECTION

2.1. Sample Preparation. Homogeneous precursor solutions of pure and Al-doped ZnO thin films were prepared by dissolving zinc nitrate in 20 ml of methanol at 0.5 M concentration. The stoichiometric amount of aluminum nitrate [Al(NO₃)₃·9H₂O] was dissolved in zinc nitrate solutions to obtain 1, 2, and 3 wt % Al-doped precursor solutions, respectively. Next, the obtained homogeneous precursor solutions of the 0, 1, 2, and 3 wt % Al-doped ZnO samples were aged for 3 days to obtain a viscous solution before spin-coating on glass substrates using a MATRIX instrument. The pH of the precursor solution was maintained at 9 by adding appropriate amounts of sodium hydroxide. Finally, pure and Al-doped ZnO thin films were deposited with different Al-doping concentrations by the spin-coating technique at 3000 rpm for 45 s. After the deposition of each layer, the thin film was heated to 300 °C for 30 min to evaporate the complex organic compounds present in the sample. The layer-by-layer deposition process was repeated 15 times to acquire the suitable and preferred thickness of the films. For crystallization, the prepared thin films were annealed using a SiC furnace under an open-air atmosphere at 450 °C for 2 h (Table 2).

### Table 1. Crystallite Size (D), Lattice Constant Corresponding to the Most Intense Peak, and Lattice Strain of Pure and Al-Doped ZnO Thin Films

| ZnO NPs | crystallite size [nm] by | lattice constant [nm] | lattice strain | dislocation density (δ) 10¹⁴ lines/m² |
|---------|------------------------|-----------------------|---------------|------------------------------------|
|         | XRD                    | W–H                   | a = b = c     |                                    |
| 0 wt % Al | 18.00                  | 16.59                 | 3.25          | 5.21                               | 0.00588 | 30.86 |
| 1 wt % Al | 22.50                  | 21.87                 | 3.26          | 5.23                               | 0.00587 | 19.75 |
| 2 wt % Al | 26.30                  | 25.53                 | 3.26          | 5.25                               | 0.00567 | 14.45 |
| 3 wt % Al | 29.70                  | 27.91                 | 3.27          | 5.29                               | 0.00561 | 11.33 |

2.2. Characterization. The prepared pure and Al-doped ZnO thin films were investigated using different characterization techniques. The structural and phase-purity analyses of the thin-film samples were performed by powder X-ray diffraction (PXRD) using a Rigaku Ultima IV X-ray diffractometer with Cu Kα radiation (λ = 1.5406 Å) at 2θ values ranging from 20 to 70°. The surface topography of the prepared thin films was analyzed by atomic force microscopy (JEOL-JAM). A unique humidity control chamber was used for conducting humidity-sensing studies. The variations of resistance with changes in the relative humidity were recorded using a standard hygrometer (Table 3).

### Table 2. Various Pure and Doped Nanomaterials Having Different Humidity-Sensing Properties with respect to the %RH

| materials | synthesis technique | topography | range (%RH) | references |
|-----------|---------------------|------------|-------------|------------|
| SnO₂       | magnetron sputtering| honeycomb structure | 11–96 | 45         |
| Sn-doped ZnO | sol–gel method     | spherical nanostructure | 30–70 | 46         |
| Al-doped ZnO | sol–gel method     | nanorods | 40–90 | 47         |
| Co- and In-doped ZnO | sol–gel method | finer colloidal structures | 44–70 | 48         |
| Al-doped ZnO | spin-coating method | porous nanostructures | 40–90 | 39         |
| Al and F codoped ZnO | sol–gel method | spherical nanostructure | 40–90 | 40         |

### Table 3. Undoped and Different Metal-Doped ZnO Nanomaterials Having the Maximum Sensitivity (MΩ/% RH)

| materials | synthesis technique | maximum sensitivity | MΩ/%RH | references |
|-----------|---------------------|---------------------|--------|------------|
| Al-doped ZnO | spin-coating method | 7.38                | 49     |            |
| Co-doped ZnO | sol–gel method     | 18                  | 50     |            |
| In-doped ZnO | sol–gel method     | 15                  | 51     |            |
| Al-doped ZnO | chemical solution deposition route | 11 | 52 |            |
| Al-doped ZnO | spin-coating method | 9.8–16.5           | this work |
3. DISCUSSION

Figure 1 shows the X-ray diffraction (XRD) patterns of Zn$_{1-x}$Al$_x$O with Al ($x = 0.00, 0.01, 0.02,$ and $0.03$) thin films. The XRD patterns of pure and Al-doped thin-film samples are entirely well matched, and all allocated diffraction peaks were indexed to the standard [JCPDS card No. 36-1451]. XRD results demonstrate the existence of broad diffraction peaks corresponding to different planes, (100), (002), (101), (102), (110), (103), and (112), and can be indexed to the polycrystalline hexagonal wurtzite structure of Zn$_{1-x}$Al$_x$O with the Al ($x = 0.00, 0.01, 0.02,$ and $0.03$) sample with the P6$_3$mc space group. Furthermore, no other typical diffraction peaks of impurities were observed. The XRD patterns suggest that the synthesized Al-doped ZnO (Zn$_{1-x}$Al$_x$O) thin films are highly pure. The average crystallite size ($D$) of the Zn$_{1-x}$Al$_x$O samples is calculated using Scherrer’s formula

$$D = \frac{K\lambda}{\beta\cos\theta}$$

where $K$ is a constant ($K = 0.9$), $\lambda$ is the X-ray wavelength used in XRD (Cu K$\alpha$), $\theta$ is the Bragg angle, and $\beta$ is the full-width at half-maximum (FWHM). From Scherrer’s equation, the crystallite sizes of Zn$_{1-x}$Al$_x$O with Al ($x = 0.00, 0.01, 0.02,$ and $0.03$) thin films were calculated and found to be $\sim 18.0, 22.5, 26.3,$ and $29.7$ nm corresponding to increasing Al-doping concentrations in ZnO of 0, 1, 2, and 3 wt %, respectively. The lattice parameters of Zn$_{1-x}$Al$_x$O with Al ($x = 0.01, 0.02,$ and $0.03$) thin films were found to be those of a pure ZnO unit cell: $a = b = 3.25$ Å and $c = 5.21$ Å; $a = b = 3.26$ Å and $c = 5.23$ Å; $a$
The lattice parameters of the hexagonal wurtzite structure increase gradually with increasing Al-doping concentration; this may be due to the increasing grain size of the sample. The dislocation density (δ) basically signifies the number of defects present in the ZnO nanocrystalline materials. The dislocation density was calculated along each lattice plane using the formula \( \delta = 1/D^2 \). The dislocation densities of Zn\(_{1-x}\)Al\(_x\)O with Al (x = 0.00, 0.01, 0.02, and 0.03) thin-film samples were calculated and found to be \( 3.26 \times 10^{14}, 3.27 \times 10^{14}, 14.45 \times 10^{14}, \) and \( 11.33 \times 10^{14} \) lines/m\(^2\), respectively. The dislocation density of thin films decreases with an increase in the Al-doping concentration.

3.1. Williamson–Hall Analysis. The lattice strain (ε) and crystallite size (D) are also calculated from XRD data using the Williamson–Hall (W–H) plot by the following equation

\[
\beta \cos \theta = \frac{kD}{\lambda} + 4\varepsilon \sin \theta
\]

Further, the graph plotted between \( \beta \cos \theta \) versus 4 sin \( \theta \) with a linear fit of the data (known as the Williamson–Hall plot) is depicted in Figure 2a–d. The inverse of the intercept gives the value of the crystallite size (D) and the slope of the straight lines gives the value of the lattice microstrain (ε). The crystallite sizes calculated from the W–H plots were found to be 16.59, 21.87, 25.53, and 27.91 nm of Zn\(_{1-x}\)Al\(_x\)O with Al (x = 0.00, 0.01, 0.02, and 0.03) dopant concentrations, respectively. The variations of crystallite sizes calculated from Scherrer’s formula as well as W–H plots are quite similar. The lattice microstrain strain values are clearly seen and were found to be 0.00588, 0.00587, 0.00567, and 0.00561 for Zn\(_{1-x}\)Al\(_x\)O with Al (x = 0.00, 0.01, 0.02, and 0.03) dopant concentrations, respectively. The significant change in the lattice strain value of pure (0.00588) and 1 wt % Al-doped (0.00587) samples are quite similar, whereas those of 2 wt % (0.00567) and 3 wt % (0.00561) Al-doped samples show a large difference. The decrease in microstrain with increasing Al-doping concentrations may be due to the increasing grain size of the sample. The significant variations in microstrain upon Al doping in ZnO may be ascribed to the changes in the local environment of the unit cell and can also result in the formation of the nanorods clearly seen in the AFM/SEM micrograph.

3.2. Atomic Force Microscopy Analysis. Figure 3a–d shows the two-dimensional (2D) atomic force microscopy (AFM) images of pure and 1, 2, and 3 wt % Al-doped ZnO thin films prepared by the spin-coating method. The AFM micrographs of pure and 1 wt % Al-doped ZnO thin films show good crystalline quality and uniform distribution of spherical grains with sharp grain boundaries. The AFM micrograph also reveals that spherical nanoparticles are well interconnected with each other and consistently distributed throughout the surface of the films. Increasing the Al-doping concentration from 2 to 3 wt % leads to a modification of the spherical grains into nanorod-like structures distributed throughout the surface of the films. The modification and formation of nanorods may be caused by the large decrease in lattice strain with increasing Al-doping concentration. The root mean square (RMS) roughness and average roughness (AR) of pure and 1, 2, and 3 wt % Al-doped ZnO thin films, examined by WSxM software, were found to be ~21.94, 21.76, 18.87, and 17.98, and 14.82, 14.65, 11.68, and 10.98, respectively. The RMS and AR roughnesses gradually decrease with increasing Al-doping concentrations, which indicate the modified surface topography of the films.

The particle sizes observed in AFM images (~135 to 190 nm) are larger than the crystallite size calculated from the XRD data. XRD gives the average crystallite size, whereas AFM gives the average particle size (agglomeration of crystallite). It is also observed from both the XRD and AFM analyses that the crystallite as well as the particle size increase with an increase in the Al-doping concentration. Furthermore, a high-resolution contour plot of the AFM image is shown in Figure 4a–d, which reveals the grain distributions of pure and Al-doped ZnO thin films on a glass substrate. The contour plots of pure and 1 wt % Al-doped ZnO thin films clearly reveal that the asymmetric growth of the ZnO nanostructure results in in-plane shapes with spherical bases. However, an increase in the Al-doping concentration from 2 to 3 wt % leads to out-plane shapes with cylindrical bases. Figure 5a–d shows the three-dimensional (3D) atomic force microscopy (AFM) image of
pure and 1, 2, and 3 wt % Al-doped ZnO thin films. Increasing the Al-doping concentration from 0 to 3 wt % modified the granular shape, with a decrease in the Z-height distribution, as illustrated in the micrograph. The Z-height distributions of pure and 1, 2, and 3 wt % Al-doped ZnO thin films were observed from their 3D images and were found to be 233.5, 223.1, 155.4, and 110.6 nm respectively. The modification and decrease in the Z-height distribution may be caused by the large decrease in lattice strain with an increase in the Al-doping concentration. As seen from the nanostructure shapes in the AFM micrographs, the RMS roughness and Z-height distribution are significantly affected and modified by increasing Al-doping concentrations. The surface topography can be tuned by increasing the Al-doping concentration, which plays a vital role in humidity and gas sensing. The formation of nanorods may possibly increase the conductivity, due to which the decrease in resistivity (resistance) occurs more quickly with adsorption of water vapor. AFM micrographs revealed that increasing the Al-doping concentration in the ZnO sample may enhance the formation of nanorod structures. Thus, Al doping plays a very significant role in the formation of nanorods, which increase the surface area and as a consequence increase the adsorption and condensation, which in turn enhance the humidity-sensing properties of the sample.

### 3.3. Scanning Electron Microscopy Analysis

Figure 6a,b shows the surface morphology of pure and 3 wt % Al-doped ZnO NPs. Pure ZnO nanomaterials show spherical nanocrystallites consistently distributed on the surface of the films (Figure 6a). Increasing the 3 wt % Al doping in ZnO materials changes the spherical grains to nanorod-like structures with a high packing density and granular background uniformly distributed throughout the surface, as shown in Figure 6b. Energy-dispersive spectroscopy (EDS) results,.
shown in Figure 6c, noticeably reveal the existence of Zn, Al, and O elements in the 3 wt % Al-doped ZnO nanoparticles.

### 3.4. Transmission Electron Microscopy Analysis

TEM micrographs of 3 wt % Al-doped ZnO thin-film nanomaterials are shown in Figure 7a. TEM images of 3 wt % Al-doped ZnO thin films show elongated/spherical grains uniformly distributed throughout the inside of the surface. In Figure 7b, the histogram of the TEM image exhibits 3 wt % Al-doped ZnO thin films with uniformly distributed grains with a mean size of ~150 nm. The grain size of the 3 wt % Al-doped ZnO thin-film observed in micrographs using Image J software shows good agreement with the grain size calculated from AFM and SEM analyses.

### 3.5. Humidity-Sensing Measurement

The humidity sensing of pure and 1, 2, and 3 wt % Al-doped ZnO thin films was measured in a specifically designed chamber. The synthesized samples were kept inside the humidity chamber, and variations of electrical resistance against the %RH were measured at room temperature. The resistances of the prepared samples were precisely measured by a multimeter with an accuracy of ±0.001 MΩ (VC-9808). The electrical resistances of samples were recorded by linking two copper sheets, with the resistance of the laid wire assumed to be negligible. The humidity of the chamber was increased using a drenched aqueous solution of potassium sulfate (95%) and decreased using a drenched aqueous solution of potassium hydroxide (20%). The variation of humidity in the chamber was tracked by a standard hygrometer (Huger, Germany, ±1% RH accuracy). Further, the aging effect of the sensing material placed in the humidity control chamber was studied after 3 months, and its stability was determined at fixed values of %RH.

### 3.6. Sensing Principle

The humidity-sensing approach of semiconducting metallic oxide sensors depends on physical adsorption, chemical absorption, and condensation of water vapor at the surface of the sample. The sensitivity of the humidity sensing measurement is generally based on an ionic or electronic mechanism. In the ionic-type sensing mechanism, absorption and condensation of water molecules on the surface of the sample may change the resistance. In the electronic-type sensing mechanism, chemical absorption takes place because H₂O molecules have electron-donating properties due to which the conductivity of the sensing material changes. The electronic-type sensing mechanism depends on the n-type or p-type characteristics of the material. In this study, the material was kept in a humidity variation chamber; the chemical absorption route occurred and water vapor was chemisorbed on the surface of the materials. AFM/SEM images show that grain growth of nanorods increases with increasing Al-doping concentration, which may be due to an increase in the surface-to-volume ratio.

### 4. RESULTS AND DISCUSSION

The results obtained after characterizations of different samples are critically analyzed in this section. Detailed literature surveys of various nanomaterials fabricated with different synthesis techniques were found to yield different surface morphologies, which modified the humidity-sensing properties with respect to the %RH; the results are tabulated below.

The humidity-sensing properties of pure and different metal-doped ZnO nanomaterials were investigated by several research groups, and they found an improved humidity sensitivity. Figure 8 shows the variation of resistance with changes in the %RH for sensing materials of pure and 1, 2, and 3 wt % Al-doped ZnO thin films annealed at 450 °C for the humidification process. It was found that the resistance...
constantly decreased with an increase in the %RH up to a value of <45, which may be due to the good conductivity of the sample. The humidity gas-sensing measurement results confirmed that the fabricated pure and 1, 2, and 3 wt % Al-doped ZnO thin-film sensors displayed quick-response characteristics as well as good reproducibility, revealing their promising potential for application in humidity gas sensors.

4.1. Variation of Sensitivity with the Al-Doping Concentration. The humidity sensitivity of pure and 1, 2, and 3 wt % Al-doped ZnO thin films were calculated from the equation, sensitivity = $\Delta R/\Delta \%$RH. The sensitivity values of Al-doped ZnO thin films from 0 to 3 wt % were calculated and found to be in the range of 9.8$-16.5$ M$\Omega$/%RH. The electrical properties of aluminum-doped and undoped zinc oxide nanoparticles were investigated by some research groups, and they reported that the conductivity decreases with increasing doping concentrations, which may be due to increasing disorder in the nanocrystalline materials. Furthermore, in the presence of artificial air, the conductivity increases by six orders of magnitude with increasing Al-doping content. It is also reported in the literature that the conductivity of the sample increases, possibly due to the increase of the surface-to-volume ratio, generating free charge carriers upon Al dopant incorporation.

4.2. Aging Behavior of Samples. Aging is a very serious problem and significantly affects sensing devices. After the study of humidity-sensing properties, the effect of aging was examined and recorded in the humidity control chamber after 4 months with variations of resistance versus %RH. Figure 9 shows the aging behavior of pure and 1, 2, and 3 wt % Al-doped ZnO thin film samples sintered at 450 °C, and it was found that the obtained values are repeatable within ±1.00% accuracy in the 20−100%RH range after 4 months. The pure and 1, 2, and 3 wt % Al-doped ZnO thin films show a significantly lower aging effect, thus giving good quality performance for up to 4 months; thus, Al-doped nanostructures also play a critical role in slowing down the aging effect.

5. CONCLUSIONS

X-ray diffraction analysis of pure and 1, 2, and 3 wt % Al-doped ZnO thin-film samples revealed a hexagonal wurtzite crystal structure with a $P6_3/mc$ space group. No additional characteristic peaks were observed in the XRD pattern, signifying the highly pure phase formation of undoped and 1, 2, and 3 wt % Al-doped ZnO thin films. The crystallite sizes of the pure and 1, 2, and 3 wt % Al-doped ZnO thin films sintered at 450 °C were calculated and found to be ~18.0, 22.5, 26.3, and 29.7 nm, respectively. AFM/SEM images of 3 wt % Al-doped ZnO thin films revealed the formation of uniform nanorods with grains of different sizes and shapes distributed throughout the surface of the films. The EDS spectra noticeably revealed the existence of Zn, Al, and O elements in the 3 wt % Al-doped ZnO nanoparticles. The growth of nanorods increases with increasing Al-doping concentration, which plays a vital role in humidity gas sensing. TEM images of 3 wt % Al-doped ZnO thin films showed elongated/spherical grains uniformly distributed throughout the inside surface. A histogram of the TEM image demonstrated that 3 wt % Al-doped ZnO thin
films exhibited uniformly distributed grains with a mean size of ~150 nm. The 3 wt % Al-doped ZnO thin films show lower hysteresis loss, less aging effect, and good sensitivity. Sensitivity values were calculated and found to be in the range of 9.8–16.5 MΩ/%RH for 0–3 wt % Al-doped ZnO thin films. The 3 wt % Al-doped ZnO thin film was used for the practical potential application in humidity gas sensors. The outcomes affirm that the Al-doped ZnO thin films are the most promising emerging nanomaterials that can be used for humidity sensing, and they require extensive research in the future.

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