



Original Article

Effects of La Doping on Structural and Electrical Properties of ZnO Ceramics

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Abstract: ZnO materials have attracted significant attention due to their diverse functionalities and tunable properties via doping and microstructural engineering. However, the relationship between crystallographic texture and electrical transport in La-doped ZnO has not been clearly clarified. This study investigates the effect of La incorporation on the carrier concentration and mobility of La-doped ZnO (LZO) polycrystals. The results reveal that the improvement in electrical conductivity, characterized by reduced carrier concentration and enhanced mobility, is predominantly governed by crystallographic texture rather than doping-induced carrier generation alone. Specifically, an increase in the texture coefficient for the (002) plane [$TC_{(002)}$] and a corresponding decrease in the texture coefficient for the (101) plane [$TC_{(101)}$] indicate preferential grain alignment along the c-axis, which improves crystallinity and reduces defect-related scattering. This texture-driven microstructural refinement plays a key role in facilitating efficient carrier transport. As a result, the highest electrical conductivity of 544.6 Scm^{-1} was achieved in the LZO sample with a La doping ratio of 0.1 at%.

Keywords: Lanthanum doping, ZnO ceramics, electrical conductivity, texture coefficient.

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1. Introduction

Zinc oxide (ZnO) is a wide-bandgap semiconducting oxide that has attracted considerable attention due to its low cost, ease of fabrication, and unique optical, magnetic, and electronic properties. Owing to its broad range of practical applications, continued research is necessary to enhance its electrical performance. Doping ZnO with various metal ions (e.g., Al, Ga, In, Co, Mn) has been shown to improve carrier concentration, Hall mobility, and a range of functional properties, including optical, thermoelectric, and magnetic behaviour [1-5]. These doped ZnO materials are considered promising candidates for use in spintronics, solar cells, field-effect transistors, gas sensors, light-emitting devices, photocatalysts, and biomedical applications.

More recently, rare-earth elements such as La, Er, and Yb have been shown to dramatically alter the morphology and band gap of ZnO particles due to defect formation when La^{3+} , Er^{3+} , and Yb^{3+} ions incorporate into the ZnO host lattice [6, 7]. M. Khuili and his group also concluded that the electrical conductivity of ZnO improved after doping with rare-earth elements [8]. Among these, La doping is particularly notable for its ability to significantly increase the density of states in ZnO, attributed to the hybridization between the d-orbitals of La and Zn atoms and the p-orbitals of O atoms [8]. Substitution of Zn^{2+} by La^{3+} introduces additional free electrons, while the incorporation of larger La^{3+} ions disrupts the original periodicity and order of the ZnO lattice, leading to increased carrier scattering. It has been reported that the carrier concentration increases with La doping, ranging from approximately $0.08 \times 10^{19} \text{ cm}^{-3}$ to $0.45 \times 10^{19} \text{ cm}^{-3}$. However, this is accompanied by a decreasing trend in carrier mobility [8].

Previous studies have also reported the influence of La doping on the structural and electrical properties of ZnO, particularly in terms of carrier concentration and conductivity [9, 10]. In these works, the electrical behaviour is

mainly interpreted based on dopant-induced carrier generation and defect-related mechanisms.

While previous studies have examined how La doping influences the electrical properties of ZnO, the specific relationship between La-induced crystallographic orientation and the resulting electrical transport characteristics has not been thoroughly clarified. To address this gap, the present work focuses on elucidating the role of crystallographic texture in governing electrical transport properties in La-doped ZnO. In this study, we provide a systematic analysis linking Hall transport parameters with texture coefficients, showing that the variation in carrier concentration and mobility is strongly correlated with the evolution of $\text{TC}_{(002)}$ and $\text{TC}_{(101)}$. This result indicates that texture-induced microstructural effects, rather than doping-induced carrier generation alone, play a dominant role in determining the electrical performance of La-doped ZnO ceramics. To the best of our knowledge, such a direct correlation between crystallographic texture and carrier transport has not been clearly addressed in the above-mentioned studies.

2. Experimental

La-doped ZnO (LZO) bulk materials were fabricated using the solid-state reaction method with doping ratios of 0.1 at% (LZO 0.1) and 0.5 at% (LZO 0.5). Starting materials of ZnO (Merck, 99.9%) and La_2O_3 (Sigma-Aldrich, 99.99%) powders were accurately weighed and wet ball-milled for 5 hours. The resulting slurry was then dried in air at 120 °C for 24 hours. The dried powders were compacted using a hydraulic press and subsequently sintered at 1400 °C for 1 hour. A pure ZnO sample was also prepared using the same procedure for comparison.

To evaluate the impact of La doping on the electrical properties of ZnO bulk, the investigation of carrier concentration, mobility, and electrical conductivity for all the samples was performed using Hall effect-based measurement (Ecopia HMS-3000). XRD

analysis of bulk samples was performed using a Bruker D8-Advanced X-ray diffractometer with CuK_α radiation ($\lambda = 0.1541$ nm), operating at 40 kV and 30 mA. The average crystallite size (D) was then determined using the Debye–Scherrer formula (Equation 1) [11].

$$D = 0.9\lambda / \beta_{hkl} \cos\theta \quad (1)$$

where β_{hkl} is the full width at half maximum (FWHM) of the diffraction peak. To understand how the microstructure of a material changes with different La concentrations, the texture coefficient (TC) was estimated according to Equation 2 [11].

$$TC_{(hkl)} = \frac{I_m(hkl)}{I_o(hkl)} \left\{ \frac{1}{n} \sum_{i=1}^n \frac{I_m(hkl)}{I_o(hkl)} \right\}^{-1} \quad (2)$$

where $I_m(hkl)$ is the intensity of the (hkl) peak in the sample, $I_o(hkl)$ is the intensity of the (hkl) peak in the standard XRD pattern of ZnO (JCPDS #39-1451), and n is the total number of considered peaks ($n = 3$). Lattice parameters can be calculated by the relationship according to the 2θ value of the (002) and (100) planes [11]:

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

where $d_{(hkl)}^2 = l^2 / 2\sin\theta$ is the interplanar spacing according to Bragg's law. To confirm the crystallinity and lattice structure, high-resolution transmission electron microscopy (HRTEM, JEM 2100 JEOL) was analyzed. In addition, the morphology and elemental composition of these samples were characterized by using scanning electron microscopy (SEM, Jeol JSM-IT200) equipped with an energy-dispersive X-ray spectroscopy (EDS) detector.

3. Results and Discussion

The electrical properties, including the dependence of carrier concentration, mobility, and electrical conductivity on La concentration, were analyzed using Hall measurement at room

temperature, as shown in Figure 1. All the samples exhibit negative Hall coefficients, indicating n-type semiconducting behavior dominated by electron carriers. The carrier concentration value reduces in the presence of La dopant, as seen in Figure 1. The highest mobility and conductivity of $73.8 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and 544.6 Scm^{-1} , respectively, were observed for the LZO 0.1 sample.

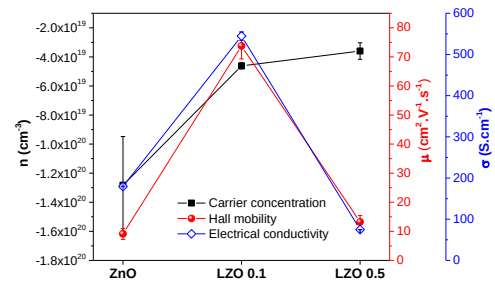


Figure 1. Hall-effect measurements of the carrier concentration (n), Hall mobility (μ), and electrical conductivity (σ) as a function of La concentration.

The effect of La doping on the ZnO host lattice was investigated by analyzing XRD patterns.

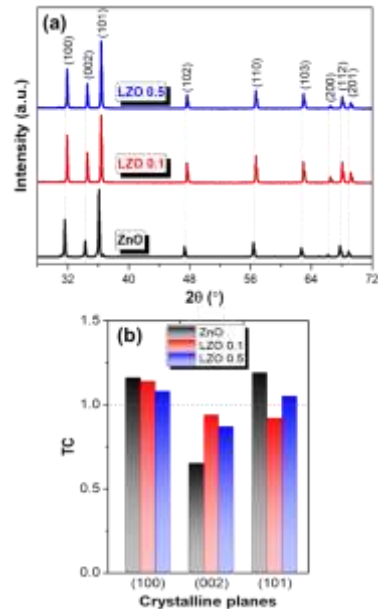


Figure 2. (a) XRD patterns and (b) texture coefficients corresponding to the (100), (002), and (101) crystallographic planes of ZnO, LZO 0.1, and LZO 0.5 samples.

Figure 2a presents the full XRD profiles of the pure ZnO, LZO 0.1, and LZO 0.5 samples in the 2θ range of 28° – 72° . All the samples exhibit characteristic diffraction peaks corresponding to the hexagonal wurtzite structure of ZnO, such as (100), (002), (101), (102), etc., consistent with the JCPDS card no. 36-1451 of the standard ZnO. Notably, no secondary phases were detected, indicating the good solubility and structural homogeneity of La^{3+} ions in the ZnO lattice up to a doping level of 0.5 at.%. A slight shift of the diffraction peaks toward higher 2θ angles was observed in the LZO 0.1 and LZO 0.5 samples, suggesting lattice distortion within the ZnO crystal structure due to the incorporation of La^{3+} ions [12].

To confirm the crystalline structure of the samples, HRTEM images were shown in Figure 3. Clear lattice fringes are observed for all the samples, indicating good crystallinity. The observed lattice fringes can be indexed to the characteristic (101) plane of hexagonal ZnO. A slight variation in the interplanar spacing is observed upon La doping, suggesting local lattice distortion induced by the incorporation of La ions. This result is consistent with the XRD analysis and further supports the successful incorporation of La into the ZnO lattice.

To further confirm the successful incorporation of La into the ZnO matrix, EDS analysis was performed, as shown in Figure 4. The spectra reveal the composition of Zn, O, and La elements in the samples. The measured La concentrations are approximately 0.28 at% and 0.63 at% for the LZO 0.1 and LZO 0.5, respectively, which are in reasonable agreement with the nominal compositions. Minor deviations may arise due to the limited accuracy of EDS for low-concentration elements.

The lattice constants a and c slightly decrease from $3.27 \text{ \AA}$ to $3.24 \text{ \AA}$ and from $5.24 \text{ \AA}$ to $5.19 \text{ \AA}$, respectively, upon La doping, as summarized in Table 1. Changes in the lattice parameters of metal-doped materials depend on the ionic radius of the dopant, which can substitute for Zn^{2+} ions or occupy interstitial

positions within the host lattice. In the case of La doping, the ionic radius of La^{3+} ($1.06 \text{ \AA}$) is significantly larger than that of Zn^{2+} ($0.74 \text{ \AA}$), which would typically result in lattice expansion [13].

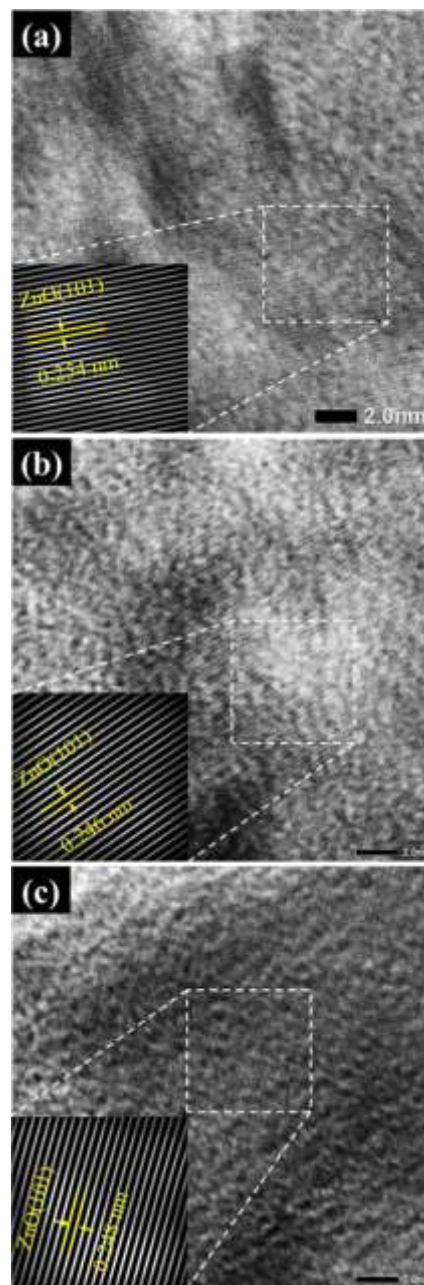


Figure 3. HRTEM images of (a) ZnO, (b) LZO 0.1, and (c) LZO 0.5 samples.

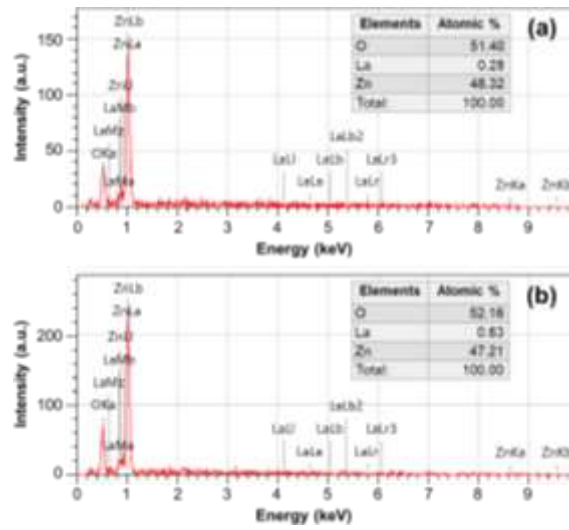
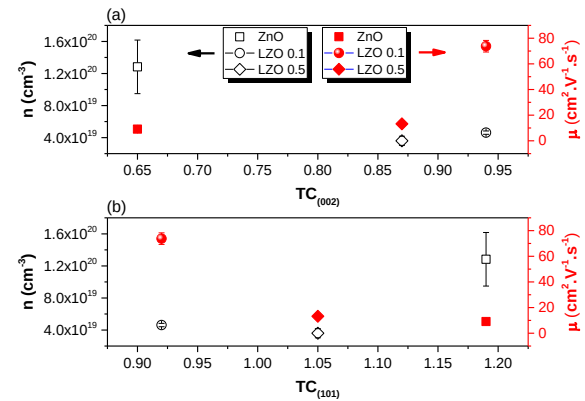


Figure 4. EDS analysis of (a) LZO 0.1 and (b) LZO 0.5 samples.

However, at low doping concentrations (0.1 and 0.5 at.%), the LZO 0.1 and LZO 0.5 samples exhibit smaller lattice parameters compared to undoped ZnO. It can be attributed to the formation of a high concentration of defects, such as oxygen vacancies, induced by the high-temperature sintering process [14]. Additionally, La^{3+} ions may not substitute directly at Zn^{2+} sites but instead occupy interstitial positions or promote defect formation, both of which can lead to lattice contraction [12, 14]. The average crystallite size, calculated from the XRD data, ranges from 50.5 nm to 63.4 nm, as summarized in Table 1.

In addition, three prominent diffraction peaks corresponding to the (100), (002), and (101) planes were recorded. To evaluate the preferential crystallographic orientation of the bulk samples, the texture coefficient (TC) for these planes was calculated, as shown in Figure 2b. The (100) and (101) orientations are identified as preferred growth directions, with TC values greater than 1, whereas the (002) orientation exhibits a lower degree of preferred growth. The $\text{TC}_{(100)}$ remains nearly unchanged upon La doping, while $\text{TC}_{(002)}$ and $\text{TC}_{(101)}$ show strong dependence on La concentration. Specifically,

$\text{TC}_{(002)}$ increases by 0.2 with La doping, while $\text{TC}_{(101)}$ decreases, with values of 1.19, 0.92, and 1.05 for the ZnO, LZO 0.1, and LZO 0.5 samples, respectively. This indicates that La incorporation can alter grain growth anisotropy, promoting c-axis alignment at the expense of the [001] direction, especially at low La content.

Figure 5. The variation of carrier concentration (n), Hall mobility (μ) as functions of (a) $\text{TC}_{(002)}$ and (b) $\text{TC}_{(101)}$.

To assess the relationship between electrical properties and crystallographic texture, the variations in carrier concentration (n) and Hall mobility (μ) as functions of $\text{TC}_{(002)}$ and $\text{TC}_{(101)}$

are plotted in Figure 5. In this study, the carrier concentration decreases while the Hall mobility increases with increasing $TC_{(002)}$. Conversely, the opposite trend is observed for $TC_{(101)}$, where higher $TC_{(101)}$ values correspond to increased carrier concentration and reduced mobility. These findings suggest that the electrical properties of La-doped ZnO bulk materials are strongly influenced by crystallographic texture and the associated microstructural quality. In the samples with high $TC_{(002)}$ values, such as LZO 0.1 and LZO 0.5, enhanced grain alignment and improved crystallinity along the c-axis contribute to a reduced density of native defects, such as oxygen vacancies (V_O) and zinc interstitials (Zn_i), which typically act as electron donors [15]. The reduction in these defects can minimize defect-carrier scattering, resulting in lower carrier concentrations and improved Hall mobility. In contrast, ZnO bulk samples with a dominant (101) orientation exhibit greater structural disorder and grain misalignment, which promote the formation of native donor defects and increase both grain boundary and defect-carrier scattering. Consequently, these samples display higher carrier concentrations but lower mobility. This behavior differs from previous reports [9, 10], where the electrical properties were mainly interpreted in terms of defect-induced carrier generation, without considering the role of crystallographic texture. These observations highlight the critical role of crystallographic orientation in governing both

the structural and electronic properties of La-doped ZnO ceramics.

SEM imaging was employed to evaluate the influence of La doping on the surface morphology and to assess the sintering quality of the bulk samples after sintering at 1400 °C, as shown in Figure 6. The images reveal that all the samples exhibit good densification and packing. At a magnification scale of 10 μm , voids are observed to be sparsely distributed throughout the bulk material. Among the samples, the LZO 0.1 sample exhibits the fewest voids, whereas the undoped ZnO sample shows the highest void density. The apparent grain (domain) size estimated from SEM images is in the range of a few micrometers ($\sim 1\text{--}5\ \mu m$), showing a decreasing trend with increasing La concentration. This reduction in grain size may be attributed to the influence of La on inhibiting grain growth during the sintering process. At the 50 μm scale, the microstructure exhibits grain clustering without well-defined grain boundaries. It should be noted that the grain sizes observed in SEM are significantly larger than the crystallite sizes ($\sim 50\text{--}60\ nm$) estimated from XRD, indicating that each grain consists of multiple crystallites. This observation suggests that grain boundary scattering, a known mechanism that impedes electron transport and reduces mobility, is minimal or negligible in these samples [16, 17].

Table 1. Crystallographic information of ZnO and LZO 0.1 and LZO 0.5 samples

Samples	Orientation	2θ (°)	FWHM (°)	D (nm)	TC	a (Å)	c (Å)
ZnO	(100)	31.59	0.1368	60.3	1.16	3.27	5.23
	(002)	34.28	0.1548	53.7	0.65		
	(101)	36.07	0.1497	55.8	1.19		
LZO 0.1	(100)	31.90	0.1303	63.4	1.14	3.24	5.19
	(002)	34.56	0.1565	53.1	0.94		

	(101)	36.38	0.1657	50.5	0.92		
LZO 0.5	(100)	31.90	0.1523	61.4	1.08	3.24	5.19
	(002)	34.56	0.1453	57.2	0.87		
	(101)	36.38	0.1523	54.9	1.05		

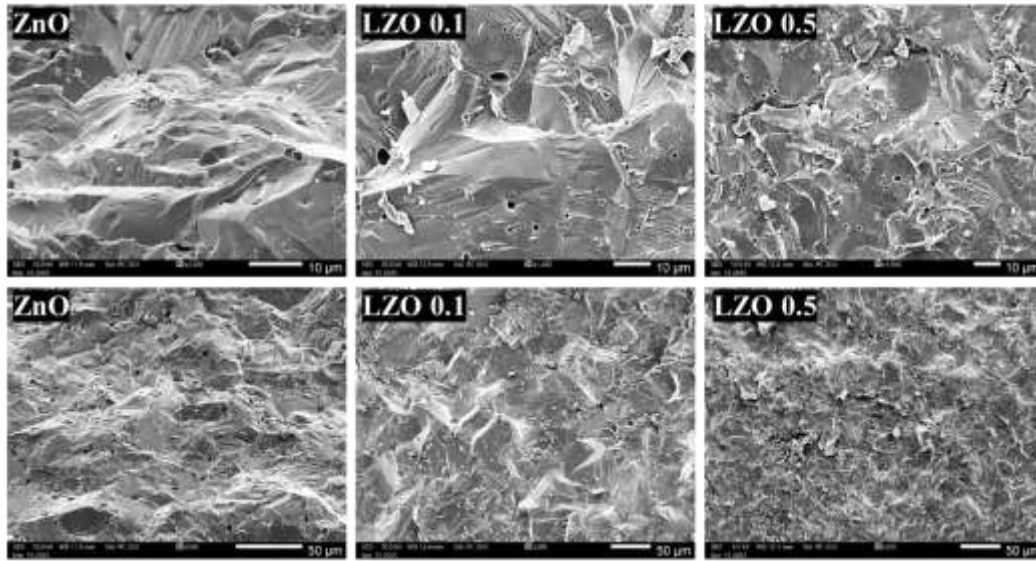


Figure 6. SEM images of ZnO, LZO 0.1, and LZO 0.5 samples at the scales of 10 μm and 50 μm .

4. Conclusion

The electrical properties of La-doped ZnO polycrystals were investigated. The observed reduction in lattice parameters is primarily attributed to chemical doping effects, including the incorporation of La^{3+} ions and the suppression of native donor defects. However, the simultaneous improvement in electrical properties, specifically the decrease in carrier concentration and the increase in Hall mobility, is more closely associated with the evolution of crystallographic texture. As the texture coefficient $\text{TC}_{(002)}$ increases and $\text{TC}_{(101)}$ decreases, grain alignment along the c-axis becomes more pronounced, leading to enhanced crystallinity, reduced defect density, and diminished defect-related scattering. These results demonstrate that crystallographic texture plays a decisive role in governing carrier

transport in La-doped ZnO ceramics. This texture-induced microstructural refinement, rather than lattice contraction alone, is identified as the dominant factor controlling the electrical transport properties in the material. As a result, the highest electrical conductivity of 544.6 Scm^{-1} was recorded in the LZO 0.1 sample. These findings further suggest that La-doped ZnO ceramics with controlled crystallographic texture are promising candidates for applications in thermoelectric devices, transparent conductive materials, and electronic components requiring optimized carrier transport.

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