



Optoelectronic properties and characterization of green synthesized ZnO semiconductor nanoparticles obtained from *Polygonum aviculare* L. extract

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Abstract

This study aims to synthesize zinc oxide nanoparticles using *Polygonum aviculare* L. and compare their optical and electrochemical properties. The impact of varying extract concentrations during preparation was analyzed. The nanoparticles were formed into pellets to evaluate their potential in optoelectronic applications, and their electrochemical properties were investigated using Electrochemical Impedance Spectroscopy. The synthesized nanoparticles measured between 31.04 and 25.27 nm exhibited a Wurtzite tetragonal phase with band gaps ranging from 3.29 to 3.18 eV. The findings of this study demonstrate a simple, affordable, and eco-friendly method for synthesizing zinc oxide nanoparticles using a readily available plant extract.

Keywords Optoelectronic · Green synthesis · Electrochemical impedance spectroscopy · ZnO

Introduction

Nanotechnology has enabled the development of diverse materials to address societal needs, with applications spanning medicine, engineering, and electronics. Traditional nanoparticle synthesis methods often pose environmental challenges, prompting recent interest in green synthesis methods. This eco-friendly approach is cost-effective, utilizes natural materials, and avoids the production of toxic by-products, making it a sustainable alternative [1]. Knotweed (*Polygonum aviculare* L.) is a non-threatened herbaceous plant belonging to the *Polygonaceae* family, widely distributed throughout Europe, Asia, and the Americas. It

thrives in diverse environmental conditions ranging from arid habitats to moist, nutrient-rich soils, which reflects its high ecological adaptability [2, 3]. Traditionally, *P. aviculare* has been used in herbal medicine for the treatment of gingivitis, cardiovascular disorders, and various inflammatory conditions, and it exhibits documented antipyretic, antiparasitic, astringent, coagulant, diuretic, expectorant, and antioxidant activities [4]. These biological properties are attributed to its rich phytochemical profile, particularly its content of flavonols such as avicularin, myricitrin, astragalin, and juglanin [5]. The abundance of these reducing and stabilizing phytochemicals makes *P. aviculare* a strong candidate for environmentally friendly (green) nanoparticle synthesis. Zinc oxide nanoparticles (ZnO NPs) have garnered interest for their diverse properties, including optical, photocatalytic, biological, magnetic, and electrochemical characteristics [6]. Additionally, ZnO NPs are cost-effective, biocompatible, and non-toxic, and exhibit high thermal and chemical stability [7]. Moreover, ZnO showcases properties such as photocatalytic activity and temperature-dependent photoluminescence [8], which are beneficial for optoelectronic applications. Because of these attributes, ZnO NPs have been found to be helpful in solar cells, photocatalysis, batteries, sensors, and various electrochemical and optoelectronic devices [9, 10]. Electrochemical Impedance

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Spectroscopy (EIS) is a highly reliable technique due to its exceptional sensitivity, ability to detect capacitive changes, flexibility, and compatibility with various systems [11]. Furthermore, it is a non-destructive method [12]. This study synthesized ZnO NPs using varying percentages of *P. aviculare* L. extract and subsequently formed them into pellets to assess their electrochemical properties through EIS.

Materials and methods

Materials

For the green synthesis of NPs, *P. aviculare* L. leaves, 98% pure zinc nitrate [$\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$], obtained from Sigma-Aldrich, and deionized water were utilized.

Green synthesis

P. aviculare L. extracts were prepared in three concentrations (0.5%, 1%, and 2%) relative to deionized water. The leaves were immersed in water, stirred for 2 h, then placed in a thermal bath at 60 °C for 1 h, and filtered. Subsequently, zinc nitrate was added in a 1:1 ratio. The solution was stirred for an additional hour and dried in a thermal bath at 60 °C for 12 h. Following this, heat treatment was conducted at 400 °C for one hour, resulting in three distinct samples (PLA-1%, PLA-2%, and PLA-4%, respectively). The samples were subsequently ground and stored for future use.

Pellet formation

The stored NPs were compressed under 10 MPa pressure to produce tablets measuring 1 mm in thickness and 1 cm in diameter. Sputtering deposition was used to coat the four corners of these pellets with chromium to enhance contact with the measuring equipment.

Characterization

NP characterization was conducted using various instruments: XPS measurements were performed on a PHI 548 system using Al K α radiation (1486.6 eV) under ultra-high-vacuum condition; functional groups were analyzed employing an ATR-IR spectrometer from Perkin Elmer with a resolution of 0.5 cm⁻¹; crystallite size and defects of the nanoparticles were investigated by X-ray diffraction (XRD) using a Bruker D2-Phaser diffractometer operating at 30 kV with a step size of 0.02. The band gap analysis was performed using a Perkin Elmer spectrophotometer covering a wavelength range of 190–800 nm, with a resolution

speed of 600 nm/s. ZnO pellets were positioned between two polished copper electrodes for electrochemical measurements, using a parallel-plate capacitor model. The ZnO samples were compressed and a pellet with dimensions of $l = 1 \times 10^{-3}$ m, $S = 0.785 \times 10^{-6}$ m². Each pellet was placed between polished copper electrodes, with the aim of using a parallel-plate capacitor as a model. Then, each sample was connected to an E4980A Precision LCR Meter. As a result, a set of data ω , $|Z_T|$, D values were obtained. The measurements were obtained at room temperature, which can be considered as 25 °C.

Results and discussion

The analysis of ZnO nanostructures was conducted using X-ray photoelectron spectroscopy (XPS). Figure 1a illustrates the typical broad survey spectra obtained for different ZnO NP samples. Peaks corresponding to Zn, O, and C were observed in all three spectra. The presence of carbon is attributed to surface adsorption during exposure to ambient conditions and residual organic material from post-calcination. Binding energy corrections were made to account for charge shifts, using the C 1s peak of graphitic carbon as a reference [13].

The binding energies of the Zn 2p components, as shown in Fig. 1b, exhibit slight variations, likely due to differences in the surface morphologies of ZnO NP samples [14]. Two primary factors may account for these energy differences: (1) interactions within the chemical environment affecting surface atoms (comprising the top 5 nm thickness of the surface), and (2) changes in texture coefficients linked to morphology. Previous studies suggest that minor shifts in surface core-level binding energy, observed in clean and oxygen-covered surfaces, are related to variations in atomic packing densities across facets compared to rough surfaces [15]. The Zn 2p core levels of ZnO NPs feature fitting peaks at approximately 1024.0, 1024.4, and 1024.5 eV, corresponding to Zn 2p_{3/2} for PLA-1, 2, and 4%, respectively. These peaks demonstrate a shift to higher binding energies relative to the standard range of 1021–1023 eV for Zn 2p_{3/2}. This indicates that the Zn on the surface of the three ZnO morphologies is in the p2 oxidation state [16]. All Zn 2p_{3/2} peaks were fitted with a single Gaussian across all analyzed samples. Figure 1c) presents the XPS spectra of the O 1s region for the various ZnO NP samples. The results indicate that the O 1s core-level spectrum of ZnO NPs corresponds to a single oxygen species. The experimental data peak, positioned at a binding energy of approximately 532.6 eV, was fitted using a single Gaussian peak. This peak is attributed to O₂ ions in the Zn–O bonds of the wurtzite structure of ZnO.

Fig. 1 a XPS survey, b zinc high resolution, c oxygen high resolution

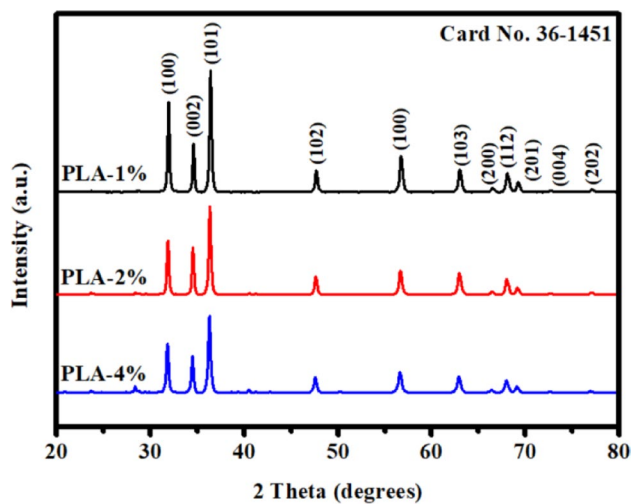
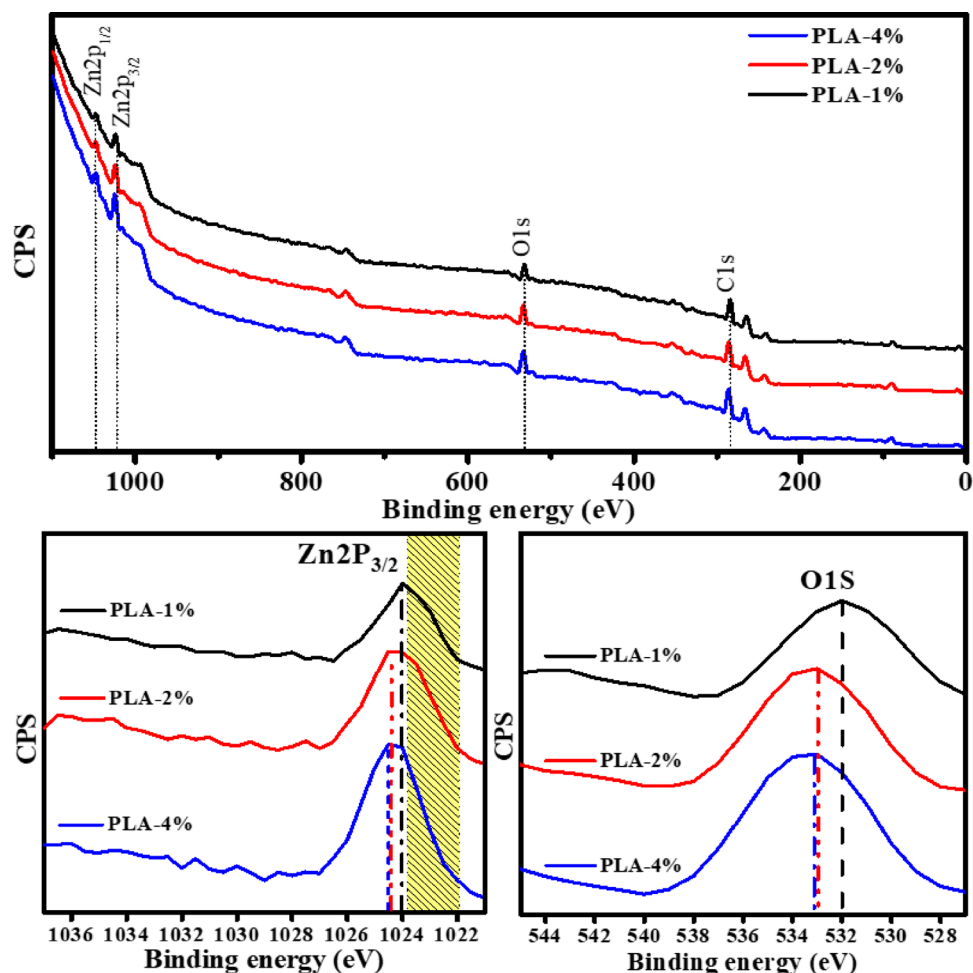


Fig. 2 XRD pattern of the synthesized ZnO semiconductor nanoparticles using *Polygonum aviculare* L. extract

The XRD results are depicted in Fig. 2. The nanoparticle diffractograms exhibit distinct ZnO diffraction peaks at 31.87° , 34.62° , 36.41° , 47.86° , 56.74° , 62.98° , 66.59° , 68.08° , 69.31° , 72.52° , and 77.16° which correspond to the

planes (100), (002), (101), (102), (110), (103), (200), (112), (201), (004), and (202), respectively [17, 18]. The obtained diffraction patterns match with a Wurtzite phase (JCPDS No. 36-1451) [19]. These results indicate that the synthesized ZnO NPs are highly pure and free from amorphous material due to the effect of organic molecules [20].

The crystal size calculation was performed using the Debye-Scherrer formula

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

Where D is the average crystallite size, λ is the wavelength of the incident X-ray, K is the Scherrer's constant, β is the full width at half maximum, and θ is the Bragg's angle [21]. The calculated sizes were 31.04, 29.05, and 25.27 nm for PLA-1%, PLA-2%, and PLA-4%, respectively. Due to the effect of the phytochemicals in the extract on the NPs, a decrease in crystal size is observed as the percentage of *P. aviculare* L. increases. The physical, chemical, and optical properties of the synthesized ZnO nanoparticles are strongly

influenced by the phytochemical constituents of *P. aviculare* L. Extracts from this plant contain abundant phenolic acids, flavonoids, and tannins, whose functions play a central role in the synthesis process. These biomolecules act as natural reducing, stabilizing, and capping agents. They promote the reduction of Zn^{2+} ions to ZnO and regulate the nucleation and controlled growth of the nanoparticles. In addition to directing particle formation, the extract's constituents strongly influence the surface chemistry of the resulting ZnO nanoparticles [22]. These changes have direct consequences for the optoelectronic behavior of the material, including its light absorption, charge-carrier dynamics, and electronic transitions [23–25].

Figure 3a–c presents the UV-Vis spectra and the band gap study, determined using the TAUC model [26]. The maximum absorbance point for each synthesis is located at 378 nm, 374 nm, and 371 nm for PLA-1%, PLA-2%, and PLA-4%, respectively. The difference in the absorption bands is directly related to the concentration of the extract used to perform the synthesis of the NP, and a greater availability of biomolecules that act as capping agents [27]. Samples PLA-1%, PLA-2%, and PLA-4% exhibited band gaps of 3.29, 3.23, and 3.18 eV, respectively, which fall within the reported ranges for ZnO NPs [28]. The reduction in band gap can be attributed to NP diameters; larger diameters result in smaller band gaps due to surface resonance effects [29]. Figure 3d shows the spectrum obtained from FTIR, where signals can be observed at $\sim 1381\text{ cm}^{-1}$, $\sim 1130\text{ cm}^{-1}$, and $\sim 1036\text{ cm}^{-1}$, which correspond to C-H rocking of alkenes;

C-O stretching vibrations; and C-O vibration modes of phenols and alcohols [30,31]. The signal at $\sim 871\text{ cm}^{-1}$ belongs to a weak vibration of the ZnO, and the prominent band at $\sim 400\text{ cm}^{-1}$ is attributed to a symmetric bending vibration [32]. The appearance of these signals confirms the formation of the Zn-O bond.

From Fig. 4a, it can be observed that impedance decreases after an initial increase in frequency for all cases. The phase of the samples shows that impedances PLA-2z and PLA-4z exhibit quite close values, while PLA-4z has the highest magnitude of all samples. This could be attributed to the variations in average crystallite size. In addition, the phase of all samples (PLA-1a, PLA-2a, PLA-4a) shows similar behavior in all cases, indicating similar conduction processes during the electrical excitation. However, the phase PLA-1a shows a shift to lower frequencies, which can be attributed to variations in sample morphology and/or composition. Regarding conductivity (Fig. 4b), the characteristic behavior of ionic conductors under Johnson's power law [33,34] is noted. After fitting the data to Johnson's equation, the corresponding results in Table 1 were obtained. Notably, the claim of an ionic conductor nature is supported by the power value n falling close to 0.6. Also, three regions can be distinguished in conductivity: region 1, where DC conduction processes are taking place; region 2, where the transition from DC to AC conductivity occurs; and finally, region three, which is related to AC conductivity.

The electrical model shown in Fig. 4d is proposed based on the Nyquist plot (Fig. 4c) and the material's understanding.

Fig. 3 Bandgap determination and UV-Vis spectra **a** PLA-1%, **b** PLA-2%, and **c** PLA-4%, and **d** FTIR spectra of the synthesized ZnO NPs

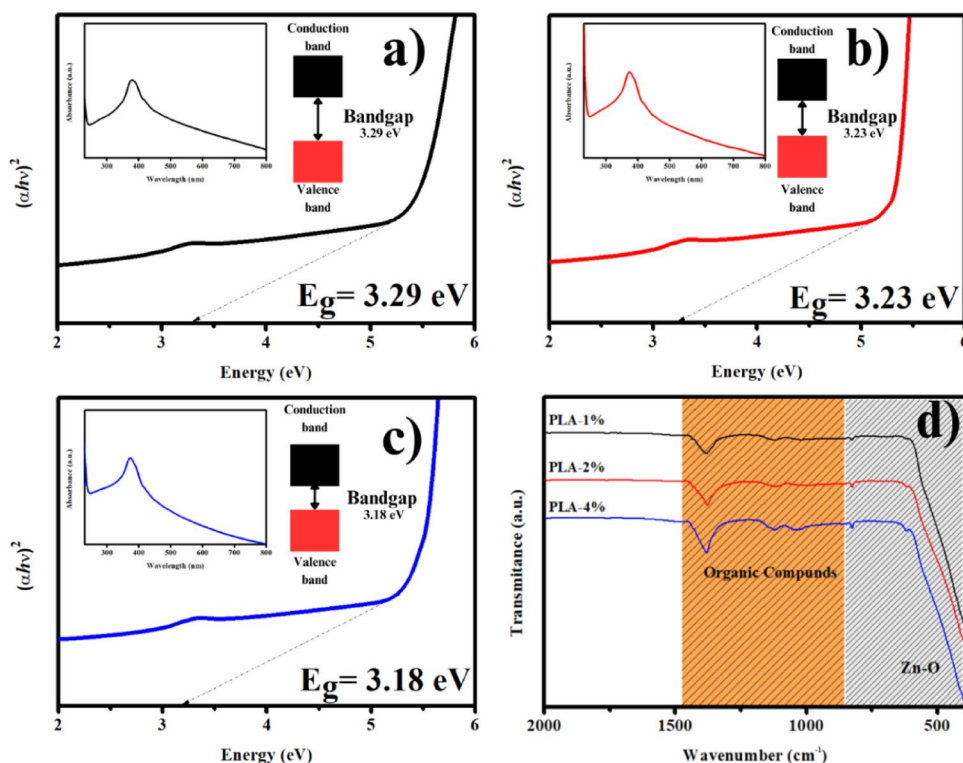


Fig. 4 Data obtained from analysis through electrochemical impedance spectroscopy: **a** the frequency-dependent magnitude of impedance (left axis) and frequency-dependent phase (right axis), **b** total conductivity, **c** Nyquist plot, and **d** equivalent proposed circuit

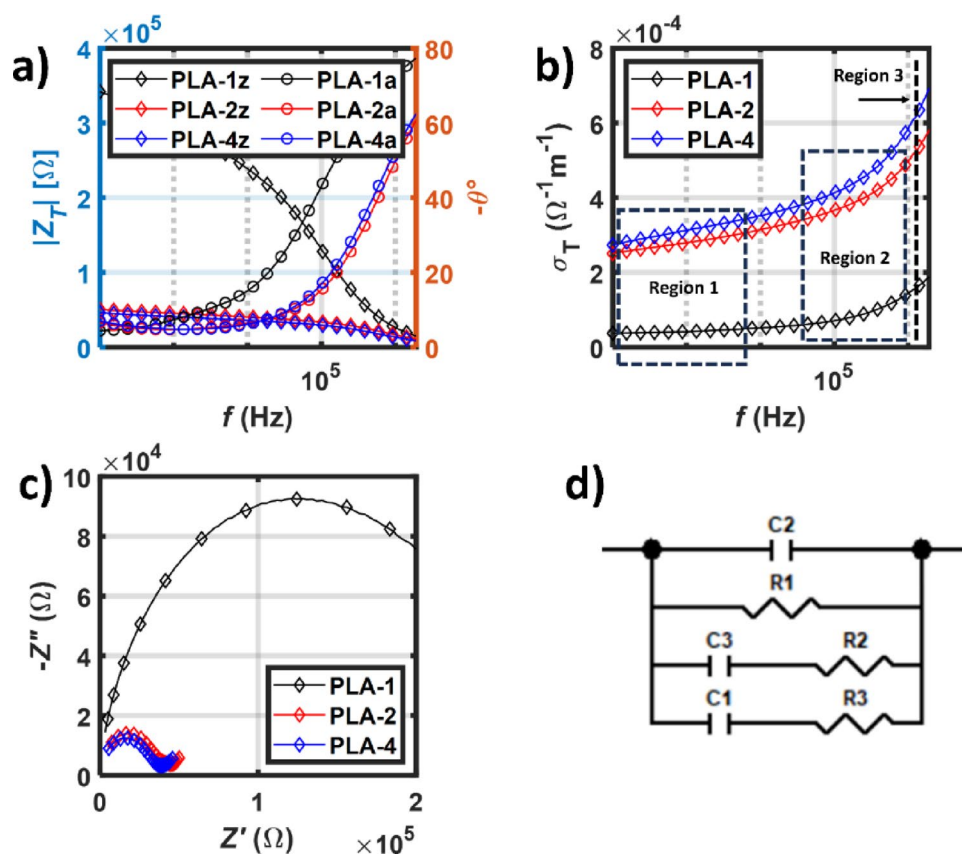


Table 1 Information from electrical characterization using EIS

	Circuit parameters						Johnsher's parameters		
	C_1 (10^{-10} F)	C_2 (10^{-12} F)	C_3 (10^{-11} F)	R_1 ($10^5 \Omega$)	R_2 ($10^5 \Omega$)	R_3 ($10^6 \Omega$)	σ_{dc} $10^{-5} \Omega^{-1} m^{-1}$	A (10^{-8})	n
PLA-1	1.03	7.07	0.99	3.36	5.61	1.27	4.5	3.29	0.57
PLA-2	13.7	8.28	3.17	5.15	1.39	0.246	31.8	6.34	0.58
PLA-4	16.7	10.5	3.08	4.68	1.18	0.196	34.6	10.4	0.56

The circuit parameters in Table 1 were derived from fitting the experimental data to this proposed circuit.

In the equivalent circuit, C_2 and R_1 are intended to model interface effects between the pellet and the electrodes. Since the samples have similar morphologies but differ in average crystallite size, these effects influence the observed electrical response. Specifically, interface effects due to particle surface density are comparable in samples PLA-2 and PLA-4, as indicated by the close values of R_1 and DC conductivity. Moreover, changes in sample microstructure affect hopping behavior, evidenced by variations in R_1 magnitude for sample PLA-1 and increased capacitance values for PLA-4 [35].

Conclusions

This study underscores the potential of green-synthesized ZnO NPs using *P. aviculare* L. extracts for optoelectronic applications. XPS analysis confirms that all samples exhibit the characteristic Zn^{2+} and O^{2-} species of wurtzite ZnO, with only minor morphology-related energy shifts linked to surface morphology. XRD results confirm high ZnO purity and crystallinity; Scherrer's formula revealed sizes of 31.04 nm, 29.05 nm, and 25.27 nm; the TAUC model presented band gap measurements of 3.29 eV, 3.23 eV, and 3.18 eV; the presence of the Zn-O bond, indicated by the 443 cm^{-1} signal in the FTIR spectrum, supporting the formation of ZnO NPs. The electrochemical characterization provided valuable insights, revealing the NPs potential as ionic conductors under Johnsher's power law. The proposed electrical model offers a framework for understanding interface effects and microstructural influences on conductivity. Future research

could focus on optimizing these NPs to enhance performance across diverse technological applications.

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Declarations

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

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