# GREEN SYNTHESIS OF ZINC OXIDE NANOPARTICLE USING *TAMARINDUS INDICA* BLENDED WITH ACTIVATED CARBON BASED OF ZINC OXIDE COMPOSITE FOR SUPERCAPACITOR APPLICATION

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**Abstract**
Tamarindus indica leaf extract was used as a bio-reductant in this study to synthesize zinc oxide nanoparticles (ZnO-NPs) blended with activated carbon for possible use in electrochemical energy conversion systems. The electrochemical behavior of the Tamarindus indica activated carbon (TIAC) and nanostructured ZnO composite mixture was also investigated using electrochemical impedance spectroscopy with 0.1 M $Na_2SO_4$ as the electrolyte. The wurtzite (hexagonal) form of the herb assisted synthesis of ZnO-NPs with size 35 nm. At a scan rate of 5 mV/s, the prepared AC-ZnO nanocomposite electrode has a specific capacitance of 275.3 $F/cm^2$. The electrodes specific capacitance decreased with increase of zinc oxide substance. The nanocomposites improved electrochemical behavior can be attributed to the activated carbons and electroactives high electrical conductivity.

**Keywords:** Tamarindus indica, ZnO, Activated carbon-ZnO nanocomposite, Supercapacitor, Specific Conductance.

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## 1. Introduction

The field of nanotechnology is one of the most important research fields of modern materials science [1,2]. Nanoparticles have new or enhanced properties that are accompanied by unique characteristics including scale, distribution, and morphology [3,4]. In contrast to bulk ZnO, the increasing degree of nanoscale ZnO has the potential to improve fabric activity potency. As a critical semiconductor with tremendous scientific and technological interest, ZnO has enormous excitons binding energy (60 meV) [5] and an on the spot large gap (3.37 eV), making it the most common multitasking metal and chemical compound with a long list of appealing properties. Because of its distinct optical and electrical properties [6], it is thought to be a potential material for use in optoelectronic applications that work in the visible and near ultraviolet spectral regions. UV light emitting devices [7,8], ethanol gas sensors [9,10], photocatalysis [11,12], all make extensive use of ZnO-NPs. Despite these accolades, ZnO is specific properties such as structure dependent, electrical, and thermal transport properties that differ depending on particle size, shape, morphology, orientation, and the ratio [13,14]. Super capacitors also known as electrochemical capacitors have a wide range of applications due to their high power density, natural attractiveness, long lifetime, and long shelf life [15,16]. Much research has been done to improve the overall performance of super capacitors. ZnO is a significant battery active material with a battery density of 650A/g, is environmentally friendly, and is a promising electrode for super capacitor applications [17]. *Tamarindus indica* was used as a bio reductant in this study to synthesize bio-inspired ZnO NPs. An activated carbon-ZnO nanocomposite electrode was prepared as a potential for super capacitor fabrication, and its electrochemical properties were assessed using electrochemical impedance spectroscopy.

## 2. Materials and Methods

### 2.1 Preparation of *Tamarindus indica* Leaf Extract

*Tamarindus indica* leaves have been collected and used for the preparation of aqueous leaf extract. The collected leaves were well washed deionized water, extract the grime particles, and then cut into small parts. The fresh chopped leaf pieces (40g) were boiled with 400 ml of deionized water and boiled at 80 ˚C for 1 hr. The raw extract obtained was cooled in a well ventilated region [18].

### 2.2. Eco-friendly green synthesis of ZnO nanoparticles

The extract was mixed with a solution of 10 mM $Zn(NO_3)_2$ in an RB flask at 60 °C for 6 h. The pale yellow solution thus obtained was dried in a hot air oven at 100 °C for 2 hrs. The resulting mass obtained was calcined at 400 °C to obtain a fine solid of ZnO NPs and to be stored in air-free containers. The Green Approach to the Composition of *Tamarindus indica* Mediated ZnO-NPs is shown in Figure 1.

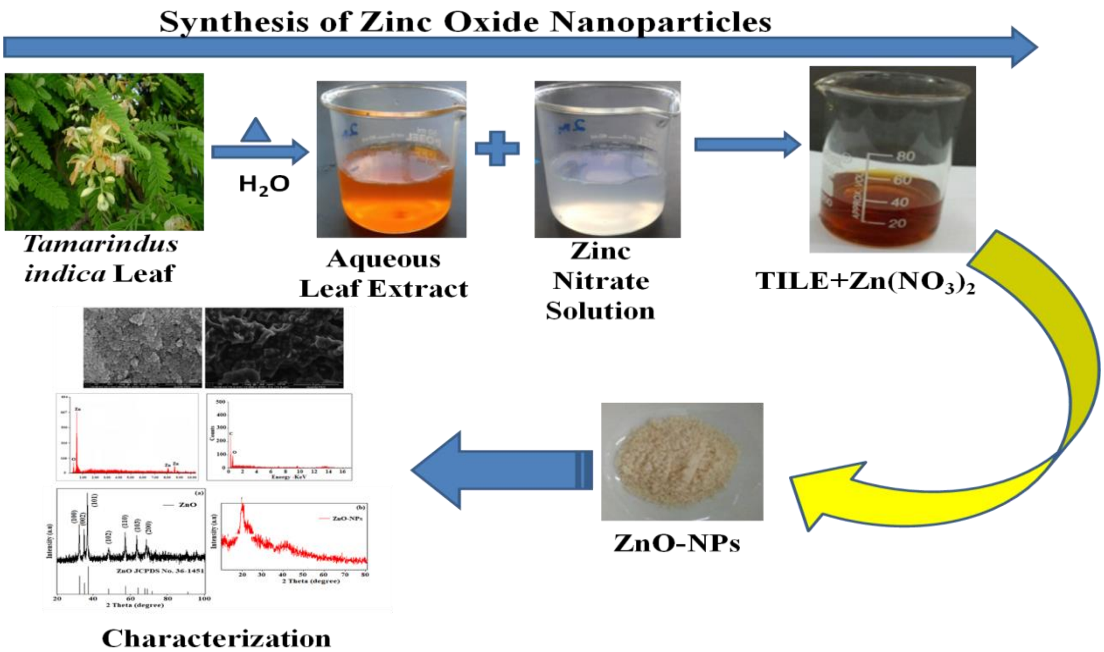


**Figure 1.** Schematic synthetic route of Zinc oxide nanoparticles

### 2.3. Preparation of *Tamarindus indica* Activated Carbon (TIAC)

The substance is grounded by the use of pestle and mortar to convert it into fine particles. Approximately 100g of powdered fine materials are mixed with 0.05 L of Conc. $H_2SO_4$ and held at room temperature for 24 hrs. Then wash with double distilled water to remove excess acid and dry at 110 °C for 14 hrs to remove moisture and store in a dry atmosphere.

### 2.4. Preparation of AC-ZnO Electrode

Activated carbon was blended with ZnO in three distinct proportions {2:0.5, 2:3, 2:2, 2:1} using n-methyl pyrrolidone together with a binding agent, polyvinylidene fluoride, and a paste. After that, the AC-ZnO composite was applied with a brush on a weighed stainless steel current collector and dried at 25°C.

### 2.5. Electrochemical Characterisation of Electrode

Electrochemical impedance measurements were performed in 0.01 L of 100 mM $Na_2SO_4$ electrolyte over a potential range of -0.9 V to +0.1 V, the solution being used as an experimental solution for ZnO nanoparticles. Electrochemical Impedance Spectroscopy (EIS) of nanoparticle modified electrodes was measured in 100 mM $Na_2SO_4$ at a disturbance amplitude of 0.01 V within a frequency range of 0.0001 mHz to 100 mHz.

### 2.6. Characterisation of *Tamarindus indica* Mediated ZnO-NPs

UV-1601 the Shimadzu spectrophotometer is used to record UV-Vis spectra that have confirmed the formation of ZnO-NPs. Functional groups of *Tamarindus indica* and biosynthesized ZnO-NPs were analyzed by FT-IR spectra with the help of the BRUKER-FTIR-TENSOR-27 spectrop-hotometer instrument. SEM, Hitachi S4700, equipped with Energy Dispersive Spectroscopy, investigated the size, shape, and elemental composition of bio-synthesized ZnO-NPs (EDS). Phase Identification of bio-synthesized ZnO-NPs analyzed with X-ray diffractometer (PAN-analytical X-Pert PRO).

## 3. Result and Discussion

### 3.1. Spectral analysis of Bio-Synthesised ZnO-NPs and ZnO nanocomposite

The absorption spectrum of ZnO-NPs, the bio-synthesized ZnO nanoparticles is shown in Figure 2.

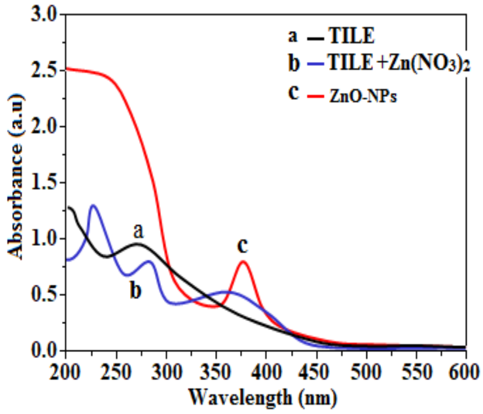


**Figure 2. (a)** The UV-visible spectrum of *Tamarindus indica* leaf extract; (b) ZnO-nanocomposite; (c) ZnO-NPs.

From the figure, it is confirmed that the Surface Plasmon Resonance (SPR) band for ZnO nanoparticles is deduced that the absorption occurred at 378 nm [19]. This is consistent with the study on the eco-friendly synthesis of ZnO-NPs (absorption peak at 376 nm) using various herb extracts [20,21], which have confirmed the existence of ZnO nanoparticles.

FT-IR spectrum of *Tamarindus indica* leaf extract spectra and ZnO-NPs in Figure 3a, b. The broad peak due to the –OH band is 3530 $cm^{-1}$, Weak absorption peaks at 2931 $cm^{-1}$ and 2863 $cm^{-1}$ are due to aliphatic asymmetrical C-H stretching vibrations and carboxylic acid stretching –C-H. The peak at 1754 $cm^{-1}$ corresponds to the C=O expansion of the carbonyl group. The poor absorption peaks at 1172 $cm^{-1}$ and 1332 $cm^{-1}$ are representative of C=N amide bond stretching. In addition, the C-O-C stretching vibrations align with the absorption peak of 1026 $cm^{-1}$. The crest at 753 $cm^{-1}$ is due to the presence of the R-CH group and the peak at 672 $cm^{-1}$ shows the vibration band of the ZnO-NPs according to the data reported distinctive peak at 510 $cm^{-1}$ can be attributed to the [Zn-O] bond of metal oxygen [22.23].

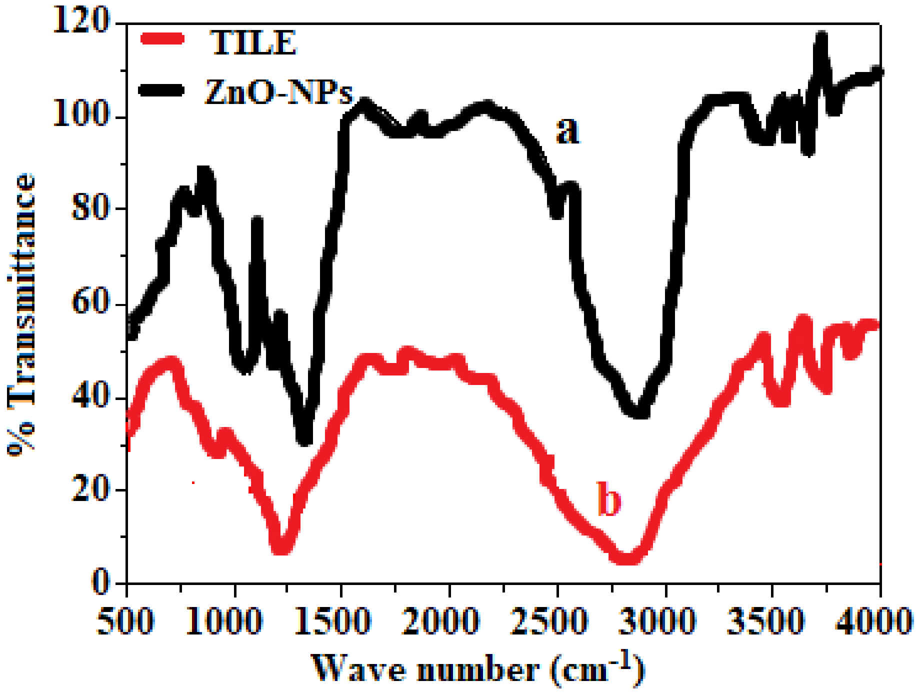


**Figure 3.** (a) The FT-IR spectrum of TILE, (b) *Tamarindus indica* mediated ZnO-NPs.

Figure 4(a) shows that the synthesized ZnO-NPs are ball-shaped and the particle size ranges between 35 nm. The EDX spectrum of eco-friendly synthesized ZnO NPs as shown in Figure 4(c) shows the chemical characterization of ZnO-NPs and the high purity of ZnO NPs except for impurities. Scanning electron microscopy was used to determine the form, size, and morphology of activated carbon produced from *Tamarindus indica* leaves (Figure 4(b)). Figure 4(b) confirms the size of the particle and further confirmation of the prepared activated carbon was made by the Energy Dispersive X-ray study (EDX). Figure 4(d) for activated carbon showed two characteristic C and O signals of 71.51 percent, 28.49 percent, respectively.

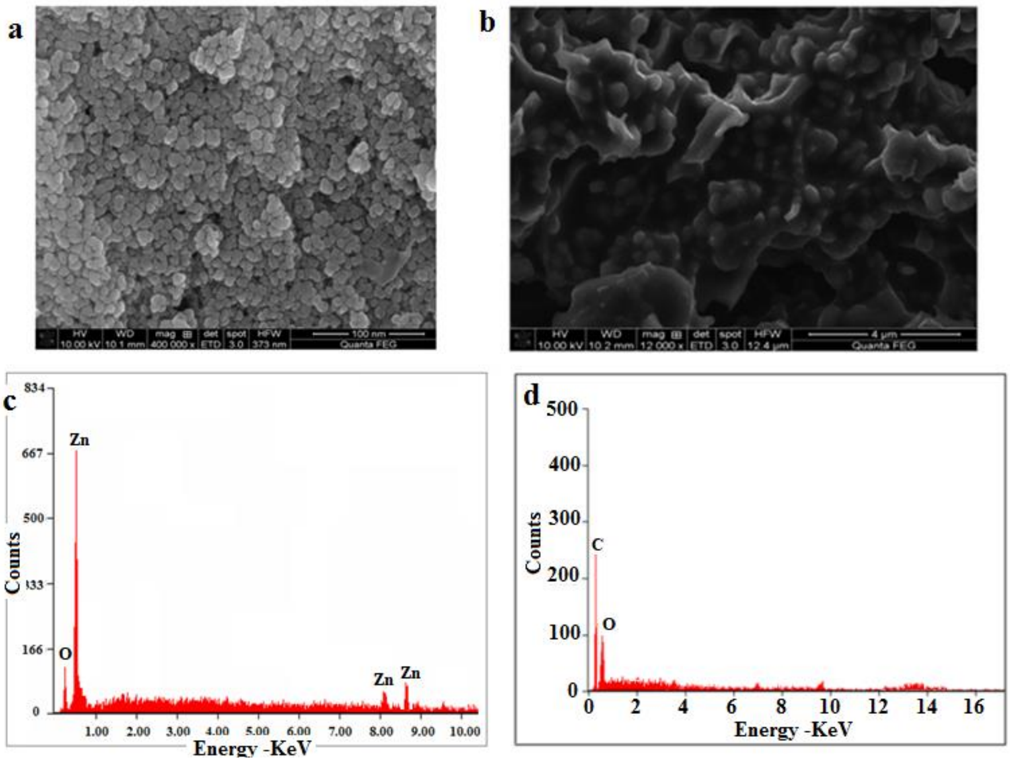


**Figure 4.** (a) SEM image of ZnO-NPs, (b) SEM image of Activated carbon prepared from *Tamarindus indica* Leaves, (c) EDX spectra of pure ZnO-NPs, (d) EDX spectra of activated carbon

The TEM image of biosynthesized ZnO-NPs is shown in Figure 5. The image shows hexagonal (Wurtzite) ZnO-NPs with particle sizes ranging from 35 to 90 nm, which is consistent with XRD results. The agglomeration of ZnO-NPs is caused by high surface energy due to their synthesis in an aqueous medium, as well as densification, which results in a narrow space between particles. Figure 5 (b) shows the selected region (electron) diffraction (SAED) pattern indicated that the particles are strongly crystalline in nature, as well as an arrangement of rings containing spots, suggesting that nanoparticles have a larger surface area. The selected area (electron) diffraction (SAED) pattern indicated that the particles are strongly crystalline in nature, as well as an arrangement of rings containing spots, suggesting that the nanoparticles have a larger grain size, uniform shape, and are polycrystalline in nature.

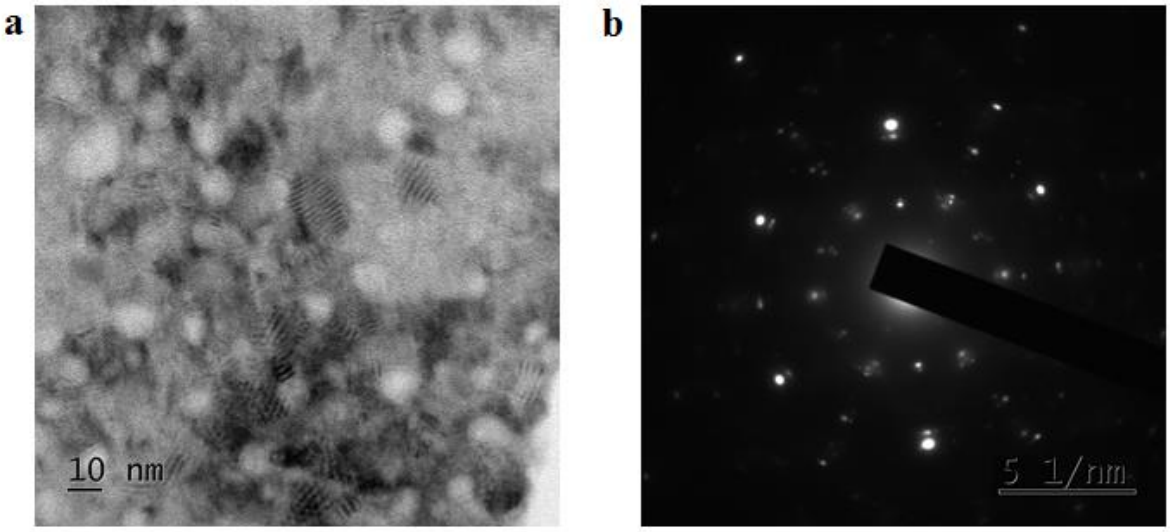


**Figure 5.** HR-TEM image of ZnO-NPs: (a) ZnO-NPs, (b) SAED patterns of ZnO-NPs.

The XRD analysis of the *Tamarindus indica* mediated zinc oxide nanoparticles shows the chemical composition, physical properties, and crystallographic structure of the ZnO NPs (Figure 6(a)). The diffraction peaks obtained at 31.9, 34.8, 36.8, 47.5, 56.3, 62.9, and 69.2 as shown in Figure 6(a) correspond to the miller indices of 100, 002, 101, 102, 110, 103, and 200 respectively. The results obtained were inappropriate agreement with the JCPDS file No. 036-1451 confirming the hexagonal wurtzite structure [24]. The clear, severe peak purchased indicates the excessive purity and crystalline nature of the synthesized ZnO-NPs [25]. No different diffraction peaks are recognized indicating the purity of the synthesized ZnO-NPs. Figure 6 (b) shows the XRD pattern of activated carbon. Characteristics 10-30° peaks of TIAC were discernible in carbon, the obtained diffraction spectrum did not show any apparent peak at a scan range of 10-90° indicating the amorphous phase of TIAC.

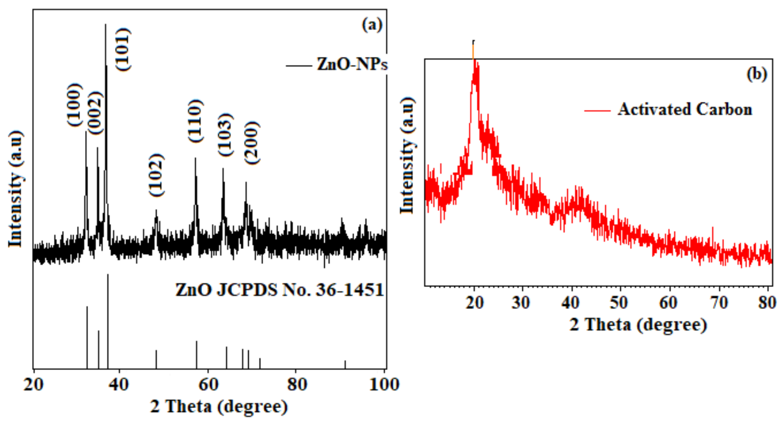


**Figure 6.** (a) XRD patterns of *Tamarindus indica* mediated ZnO-NPs, (b) XRD spectrum of activated carbon.

### 3.2. Electrochemical Properties of AC-ZnO Composite and Supercapacitors

On stainless steel plates, AC-ZnO nanocomposite electrodes with different composition ratios such as 2:0.5, 2:1, 2:2, and 2:3 were prepared. Figure 7 depicts the Nyquist plots for the AC-ZnO nanocomposite with various composition ratios of 2:1, 2:2, and 2:3. As shown in Table 1, the precise capacitance measurement indicates that composite composition in the ratio 2:0.5 has the highest specific conductance. Figure 8 depicts the Nyquist plot for a nanocomposite electrode with a composition of 2:0.5, which has the lowest resistance and the highest real capacitance. The improved electrochemical performance of the AC-ZnO nanocomposite can be due to the electroactive property of ZnO assisted on activated carbon, which provides a three-dimensional conducting system with active sites for the formation of an electrical double layer [26], and pseudocapacitance from the ZnO, which provides a higher specific capacitance [27,28].

**Table 1.** Electrochemical Impedance parameters for AC-ZnO electrode in 0.1 M $Na_2SO_4$

| Materials | Scan rate (mV/Sec) | Charge transfer Resistance ($cm^2$) | Capacitance ($F/cm^2$) |
|---|---|---|---|
| AC-ZnO (2:3) | 5 | 7.1 | 3.4 |
| AC-ZnO (2:2) | 5 | 42.5 | 33.5 |
| AC-ZnO (2:1) | 5 | 53.4 | 59.2 |
| AC-ZnO (2:0.5) | 5 | 1704.0 | 275.3 |

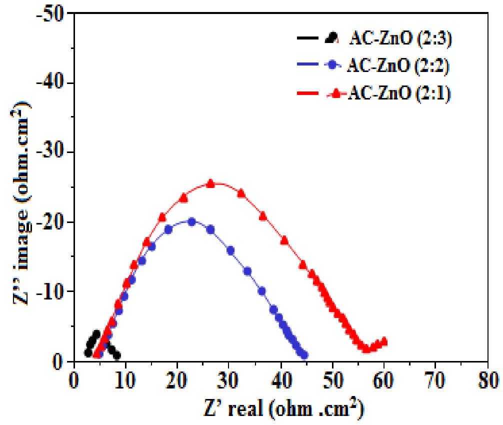


**Figure7.** Impedance plots for AC-ZnO composites at compositions of 2:3, 2:2 and 2:1.

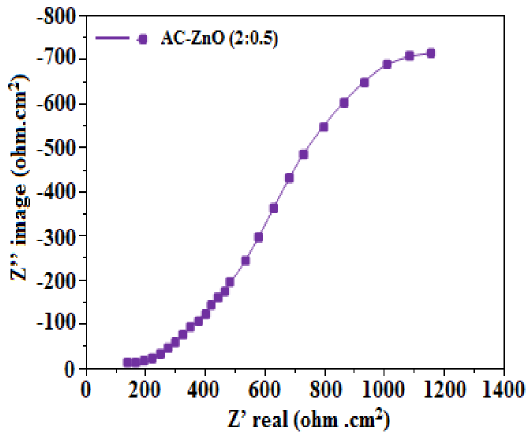


**Figure8.** Impedance plots for AC-ZnO composites at 2:0.5 mass ratio.

### 4. Conclusion

The physical property of nanostructured ZnO using *Tamarindus indica*, the particle size is 35-90 nm in SEM and TEM. They are also examined chemically by FT-IR, EDAX, and XRD, they show the presence of ZnO-NPs. The electrochemical behavior of the *Tamarindus indica* activated carbon (TIAC) and nanostructured ZnO composite mixture was also investigated using electrochemical impedance spectroscopy with 0.1 M $Na_2SO_4$ as the electrolyte and at a scan rate of 5 mV/s, the prepared AC-ZnO nanocomposite electrode has a specific capacitance of 275.3 $F/cm^2$. The electrodes' specific capacitance decreased as the zinc oxide substance increased. The nanocomposite's improved electrochemical behavior can be attributed to the activated carbon's and electroactive's high electrical conductivity. By composting with activated carbon, biosynthesized ZnO-NPs served as supercapacitors. According to the study, supercapacitors have a high capacitance and can be used as supercapacitor active materials.

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