Synthesis of zinc oxide nanoparticles using tea leaf extract and its application for solar cell

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Abstract. We report the synthesis of zinc oxide (ZnO) nanoparticles and its composite with natural graphite (NG) powder for application in solar cell. ZnO nanoparticles were synthesized using green tea leaf extract as non-toxic and eco-friendly reducing material under microwave irradiation. The formation of ZnO nanoparticles was monitored by the colour changes during the reaction. The synthesized ZnO nanoparticles were characterized by particle size analyzer (dynamic light scattering), scanning electron microscope, UV-visible spectroscopy, atomic force microscope and fluorescence spectroscopy. The average particle size of the ZnO nanoparticles was found to be 26 nm. The synthesized ZnO nanoparticles were further used to prepare ZnO/NG composite material with commercially available NG powder. The current–voltage (I – V) characteristics of thin film of ZnO/NG nanocomposite were investigated. J_{SC} (short-circuit photocurrent), V_{OC} (open-circuit photovoltage), FF (fill factor) and η (efficiency of the solar cell) were measured for ZnO/NG nanocomposite. Interestingly, the cell showed a good power conversion efficiency of 3.54% with high stability.

Keywords. Tea leaf; ZnO; NG; microwave; solar cell.

1. Introduction

Zinc oxide (ZnO) is being extensively used in the fabrication of solar cells, namely quantum dot-sensitized solar cells.^{1–3} It has many technological applications, because of its exceptional optical and electrical properties, such as thin film transistors, gas sensors, transparent conductor, biomedical and piezoelectric application.^{4–8} It is expected to be an alternating material for TiO_2 because of its properties such as being environment friendly, stability and that, it can be synthesized into different shapes and sizes very easily. It is well known that ZnO is one of the most important semiconductor materials with direct wide bandgap (3.2–3.37 eV) and good transparency⁹ at room temperature. Besides, due to large exciton binding energy of 60 meV,¹⁰ it has potential applications in optoelectronic devices such as solar cells.¹¹ Thin films of ZnO can be prepared by various techniques like sputtering,¹² chemical vapour deposition,¹³ laser ablation,¹⁴ sol–gel,¹⁵ spray pyrolysis,¹⁶ from clear emulsion of monodispersed ZnO nanocrystals^{17,18} etc.

Recently, there is a growing necessity to develop environmentally friendly methods that does not use toxic materials in the synthesis procedures. Till now, a large number of physical, chemical and biological methods are available to synthesize different type of nanoparticles.^{12–16}

Synthesis of nanoparticles using different parts of the plants is quite novel, leading to truly green chemistry which

is effective also at a very affordable cost,^{17–20} as there is no need of high pressure, energy, temperature and toxic chemicals. More recently, some of the researchers have described that the use of eco-friendly benign materials like leaf extract (aloe barbadensis),²¹ bacteria²² and fungus²³ for the synthesis of ZnO nanoparticles offers numerous advantages of eco-friendliness and compatibility for pharmaceutical and other biomedical applications.

In continuation of our earlier research work on green synthesis of metal nanoparticles,²⁴ the present study describes the rapid synthesis of ZnO nanoparticles in mass level, using green tea leaf extract under microwave irradiation. These ZnO nanoparticles were further used to prepare thin film of composite material with natural graphite (NG) powder to study its suitable application in solar cells.

2. Materials and methods

Tea leaf was collected from tea garden, Tripura. Zinc nitrate, NG powder and polystyrene (PS) were purchased from Sigma Aldrich. Double distilled water was used throughout the experiments.

2.1 Preparation of tea leaf extract

A quantity of 20 g of finely grounded tea leaf was dissolved in 200 ml of water and boiled for around 3 h. After cooling at room temperature, it was centrifuged for 15 min and filtered. The filtrate was stored at 5–10°C for further experiments.

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2.2 Preparation of ZnO nanoparticles using tea leaf extract

Zinc nitrate was used as precursor for the synthesis of ZnO nanoparticles. A ratio of 1 : 3 of zinc nitrate and tea leaf extract was mixed and the solution was subjected to microwave irradiation at 540 W, which produced brownish-black precipitate after 7 min. The precipitate was filtered and then dried at hot air oven for 4–5 h. The possible mechanism of the formation of ZnO nanoparticles from tea leaf extract is shown in scheme 1.

2.3 Characterization of ZnO nanoparticles

The ZnO nanoparticles obtained was characterized by dynamic light scattering (DLS; nanotracer wave W3222), UV-vis spectroscopy (Shimadzu 1800), scanning electron microscope (SEM; JSM-6360 JEOL), atomic force microscopy (AFM; multimode V8) and fluorescence spectroscopy (Perkin Elmer LS 55).

2.4 Preparation of thin film of ZnO/NG nanocomposite

Thin film of ZnO/NG nanocomposite was prepared by mixing 1 : 1 ratio of ZnO nanoparticles and NG powder with small amount of PS using xylene as solvent. The solution mixture was sonicated for 30 min followed by stirring at 60°C for 4 h. A drop of the resultant slurry was spread on a glass and a thin film was obtained using another glass slide. PS was chosen because of its chemical stability and excellent solubility in ordinary solvents, which helped in making the thin film.

2.5 Photovoltaic characteristics

The photovoltaic measurements of thin film of ZnO/NG nanocomposite were carried out using a PGSTAT 101 solar

simulator with an irradiance of 100 mW cm⁻². The current-voltage (*I*–*V*) characteristics of the cell were measured by applying external potential bias to the cell and measuring the generated photocurrent. The monochromator was incremented through the visible spectrum to generate the IPCE (incident photon to current conversion efficiency). Parameters such as *J*_{SC} (short-circuit photocurrent), *V*_{OC} (open-circuit photovoltage), FF (fill factor) and *η* (efficiency of the solar cell) were measured for ZnO/NG nanocomposite.

3. Results and discussion

The green tea leaf extract was used as reducing as well as stabilizing agent for ZnO nanoparticles. The microwave irradiation accelerated the processes and the reaction was completed in a very short period of time (7 min). The reaction does happen in the absence of microwave heating, but it takes almost 4–5 h at room temperature. This further gave strong evidence for the involvement of polyphenol contents of tea leaf in the rapid biosynthesis and for the stability of metal nanoparticles in the aqueous medium.²⁵ The reaction mechanism in scheme 1 showed that, ZnO nanoparticles can be obtained through the reduction of Zn²⁺ using tea leaf that contains epigallocatechin gallate (EGCG), which acts as both the reducing and capping agent. EGCG is a highly water-soluble compound with strong polarity. As such, the structure of EGCG gave enough reducibility to convert Zn²⁺ ions into Zn(0) nanoparticles. As illustrated in scheme 1, EGCG served as a stable (electron + proton) donor in interactions, and was converted into the radical ion called 'semihydro-EGCG' and then dehydro-EGCG. Dehydro-EGCG and EGCG together constituted the redox

Scheme 1. Reaction for the formation of ZnO nanoparticles.

system, which was sufficient to reduce Zn^{2+} to Zn. Besides, it should be noted that EGCG has also played the role of stabilizing agent during the reaction process. The lone pair electrons in the polar groups of EGCG can occupy two sp orbitals of the zinc ion to form a complex compound. The EGCG was capped with zinc ions, then synthesized Zn(0) nanoparticles through reduction of Zn^{2+} inside the nanoscopic templates. In the presence of nanoscopic templates, small zinc nanoparticles were easily formed. The other explanation may be attributed to the dispersion effect of the oxidation product of EGCG on the zinc nanoparticles after the completion of the reduction reaction. EGCG was converted into dehydro-EGCG through oxidation. The formation of ZnO nanoparticles from tea leaf extract was initially monitored by the colour change occurring during the reaction period, as shown in figure 1. The colour change arises due to the excitation of surface plasmon resonance in the metal nanoparticles, indicating the formation of ZnO nanoparticles. Figure 2 shows the DLS of the nanoparticles, which confirmed the average size of the ZnO nanoparticles to be around 26 nm. The SEM image clearly indicated the formation of spherical ZnO nanoparticles with

Figure 1. (a) Aqueous solution of zinc nitrate; (b) tea leaf extract and (c) colour of the solution after reaction.

Figure 2. Particle analyzer histogram of as-prepared ZnO nanoparticles.

the size ranging between 20 and 50 nm (figure 3). The UV-visible spectrum of ZnO showed the absorption peak at 345 nm (figure 4), which matched with the expected value of ZnO nanoparticles.²⁶

AFM showed the topo-graphic image of well-dispersed ZnO nanoparticles (figure 5). The luminescence of ZnO nanoparticles is of particular interest from viewpoints of both physical and applied aspects. Figure 6 showed the room temperature photoluminescence spectrum of the ZnO nanoparticles with excitation peak at 438 nm. The excitation peak corresponded to the band to band transition, which also confirmed the blue shift in the bandgap of ZnO nanoparticles. Figure 7 shows the UV-vis spectra of thin film of ZnO/NG nanocomposite, where two characteristics peaks were found at 231 and 272 cm^{-1} for NG powder and at 368 cm^{-1} for ZnO nanoparticles.

Figure 3. SEM image of ZnO nanoparticles.

Figure 4. UV-vis spectra of ZnO nanoparticles.

Figure 5. AFM of ZnO nanoparticles.

Figure 6. Fluorescence spectra of ZnO nanoparticles.

The photovoltaic measurements were carried out using two electrode systems. Xenon lamp was used as a light source and the incident light intensity was maintained at 100 mW cm^{-2} . Figure 8 shows the photocurrent density–voltage measurements (J – V curves), which can be obtained by applying a potential scan, from 0 V (short-circuit conditions) to the open-circuit potential, under constant illumination.

The FF was calculated by

$$FF = V_M J_M / V_{oc} J_{sc} \quad (1)$$

Figure 7. UV-vis spectra of ZnO nanocomposites.

Figure 8. Current density vs. voltage curve.

Where V_M and J_M were photovoltage and photocurrent density for maximum power output, respectively.

Efficiency was calculated by

$$H = V_{oc} J_{sc} FF / P_{in} \quad (2)$$

Where P_{in} is the power of incident white light from Xe lamp.

Under the irradiation of 100 mW cm^{-2} , the ZnO/NG composite gave a short-circuit photocurrent (J_{sc}) of 8.02 mA cm^{-2} , open-circuit photovoltage (V_{oc}) of 0.614 V and FF of 0.733. Efficiency in the fabricated cell was measured as 3.54%. The FF was about 73.3%. Obviously the weak value of this cell was due to the low value of the FF and the efficiency. This may be due to the higher series resistance.^{28,29}

4. Conclusion

In this paper, microwave-assisted green chemistry has been used for the synthesis of ZnO nanoparticles. A facile approach has been reported using green tea leaf extract, acting as reducing agent for the synthesis of ZnO nanoparticles of well-defined dimensions in bulk amount. This eliminated the need of toxic chemicals for the synthesis of nanoparticles. Besides, excellent reproducibility of these nanoparticles, without the use of any additional capping agent or stabilizer, will have a great advantage in comparison with microbial synthesis, avoiding all the tedious and hygienic complications. ZnO nanoparticles were successfully used to prepare thin film of ZnO/NG composite material for photovoltaic application. Interestingly, this ZnO/NG nanocomposite showed a significant efficiency of 3.54%.

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