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Biomass Mediated Synthesis of ZnO and ZnO/GO for the Decolorization of Methylene Blue under Visible Light Source

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Abstract: In this study, zinc oxide (ZnO) as well as ZnO/GO (zinc oxide/graphene oxide) were successfully synthesized. The Carica papaya leaf extract was used to synthesize ZnO and oil palm empty fruit bunch biomass to obtain graphene, which was further used to obtain graphene oxide. The samples were characterized through a variety of analytical methods such as scanning electron microscopy, transmission electron microscopy, X-ray diffraction analysis, Fourier transform infrared spectroscopy and UV–Visible spectroscopy in order to understand their morphology, size, structural phase purity, functional groups and optical properties. Various peaks such as O-H, Zn-OH and Zn-O were found in the case of ZnO. Some additional peaks, such as C-C and C=C, were also been detected while analyzing the sample by Fourier-transform infrared spectroscopy. The results of the XRD and SEM studies demonstrated that the synthesized material shows the crystalline nature of the substance in the case of ZnO, and the crystallinity decreases for ZnO/GO. The average crystallite size was found to 80.0 nm for ZnO and 74.0 nm for ZnO/GO. Further, a red shift was shown in the case of ZnO/GO, which was indicated by the UV–Vis absorption spectrum. In the TEM analysis, the particles were shown to be nanosized. For instance, the highest number of particles was found in the range of 100 to 120 nm in the case of ZnO, while 80–100 nm sized particles were found for ZnO/GO. Using synthesized ZnO and ZnO/GO, the decolorization of methylene blue was found to be 64% and 91%, respectively.

Keywords: ZnO; ZnO/GO; photocatalyst; photodecolorization; green synthesis; methylene blue

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1. Introduction

Water pollution has threatened the survival of human beings, as many industries have been expelling harmful substances into the environment in recent decades [1–3]. Even though chemical compound-based companies have played an important role in modern life (especially those that make dyes, insecticides, pharmaceuticals, etc.), a lot of waste is thrown into the environment by them, which potentially disturbs our ecosystem on a continuous basis [4]. For instance, compounds which are soluble in water, such as pesticides, dyes, and drugs, are predominately accountable for contaminating aquatic environments and have become a major cause of water pollution. These contaminants may also cause chronic diseases for human beings and are also harmful to our environment, even if they were present at very low levels. Therefore, the occurrence of such contaminants in water resources has emerged as a significant concern for environmental scientists [3,5]. In addition, several pesticides, drugs, and dyes have already been reported in various water resources across the globe in groundwater and surface water due to various types of activities [6–8]. Among them, dyes are producing a high level of water pollution, as yearly 70,000 tons is manufactured globally and 12% of these dyes are lost during the dying process in color industries, which eventually leads to the obliteration of our aquatic system [6,9]. Therefore, it is necessary to remove/degrade such pollutants from the polluted water before releasing them into the water sources.

The photocatalytic degradation method has emerged as a feasible process for the elimination of different pollutants from the aqueous solution. To address this issue, in the past several years various photocatalysts such as ZnO, TiO_2, etc, have been used [10–13]. Among them, ZnO is considered a prominent photocatalyst because of its higher stability, while irradiation is inexpensive, and has low toxicity [14]. Nevertheless, despite these benefits, the single semiconductor has certain drawbacks as it exhibits low photocatalytic activity in visible light because of its high bandgap energy [10,15]. The band gap energy was calculated for ZnO (3.3 eV), which demonstrates that it was only capable of absorbing close-ultraviolet light ($\lambda < 387$ nm) but is not suitable for working in visible light regions [13]. Therefore, there is a need for doing the necessary modifications to make it fit for working in the visible range.

Conversely, graphene derivatives are gaining considerable attention because their structure makes them ideal building blocks for a wide variety of novel applications such as photocatalysts [16], biosensors [17], batteries [18], and supercapacitors [19]. Moreover, because of graphene oxide's mechanical, electric, thermal, and optical properties, it has also been considered a material platform in modern technology. The properties of these materials can be modified through the application of a simple chemical functionalization process [20,21]. There have also been a number of publications in recent years that discuss the combination of ZnO with graphene oxide as a means of degrading a variety of pollutants [22–26]. Additionally, the synthesizing of ZnO in different sizes and shapes has been attempted using a variety of techniques, including hydrothermal, chemical microemulsion, wet chemical, sol-gel, direct precipitation, vapor phase, solvothermal, microwave, and sonochemical techniques [27,28]. However, a high amount of chemicals has been used while applying these techniques in the process of synthesis. Therefore, a simple and less chemically oriented method is still needed for the preparation of photocatalysts.

There has been much attention given to green synthesis methods for the preparation of metal oxides because they offer many advantages over chemical methods (non-green). The benefits of this synthesis environment include simplicity, energy efficiency, eco-friendliness, and low toxic levels [29]. Thus, there are many articles that report on synthesizing ZnO via the green synthesis method. For instance, the synthesis of ZnO using coffee powder as a photocatalyst [30,31]. In addition, plant extracts of Physalis alkekengi L. and Sedum Alfredii Hance were also used to synthesize crystalline ZnO [32,33]. As far as the extract of papaya leaf is concerned, which was used in this study, it contains many flavonoid and phenolic compounds with reducing and stabilizing properties that may play an important role in metal oxide synthesis [34–38].

Furthermore, the synthesis of graphene oxide using biomass such as agro-waste can also be considered a good idea to prepare a valuable compound as well as to reduce the waste from the environment, as few studies have been reported in the literature [39–42]. Hence, in the present work, we reported a green synthesis of zinc oxide (ZnO) using Carica papaya leaf extract. Moreover, the oil palm empty fruit bunch biomass was utilized to obtain graphene, which was further employed for the formation of graphene oxide. The synthesized ZnO/GO (zinc oxide/graphene oxide) was able to work as a photocatalyst active in visible light. There are some techniques that were used in the current work to