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The Green Synthesis of Nanoparticle Zinc Oxide (ZnO) Using Aloe Vera Leaf Extract: Structural and Optical Characterization Reviews

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ABSTRACT

This work is to review the previous, current and latest findings on the structural and optical properties of the ZnO nanoparticles formed by the various green synthesis methods. Each method involves a different chemical reaction, and it affects the quality of the ZnO formed on the substrates. Zinc oxide (ZnO) is a wide and direct semiconductor with a wurtzite crystal structure. Nano-scaled ZnO as today has been synthesized through green synthesis using natural plant extract as an effective 'reducing agent' of the metal precursor, which has been reported to be an effective cleaner and environment-friendly alternative towards the physical and chemical methods. This Final Year Report is based on the green synthesis and the main physical optical properties of pure ZnO nanoparticles synthesized by a completely green chemistry process using the natural extract of Aloe Barbadensis Miller (Aloe Vera) Leaf Extract to bio-reduce the zinc precursor. The different zinc precursor concentration is recognized to affect the particle size as well as on the optical properties such as surface morphology, porosity, particle size and the polymeric properties of ZnO nanoparticles. The green synthesis of NPs ZnO use is much better and affordable because the process provides more eco-friendly, economical, free toxic, and easy to compose rather than the chemical and physical methods. The main characteristics and the applications of these synthesized zinc oxide are also reported by using different analysis equipment to be the main focus on such as X-Ray Diffraction (XRD) Analysis, Field Emission Scanning Electron Microscopy (FE-SEM), Fourier Transform Raman (FT-Raman), Photoluminescence Spectra (PL)

Key words : Zinc Oxide, Aloe Vera, Green Synthesis, ZnO.

1. INTRODUCTION

Zinc Oxide (ZnO) is a distinctive compound due to its unmatched optical and electrical properties where it is acknowledged for its non-toxic inorganic metal [1], unparalleled optical and electrical properties [2], plus with the multifunctional with many application in the different field [3]. ZnO has a variety of applications such as in piezoelectric devices, chemical sensors, environmental protection, optoelectronics, pharmaceutical and cosmetic industries [4] [5]. Moreover, ZnO also has photocatalytic properties [6] with its application in the food and agriculture industry [7]. Each of the properties and its application can be modified by changing the size and morphology [8]. Other than that, ZnO Nanoparticles (NPs) also have broad applications in biological and medical such as healing nanomedicine [9], and cosmetic industries due to its antioxidant, antimicrobial, and antibacterial activity [10]–[12].

There are different methods in synthesizing nanoparticles for examples the physical, chemical and green methods [13]. Nowadays, green synthesis methods which are highly safe, environmentally friendly, cost-effective and rapid are highly recommended because of its advantages in comparison towards the chemical and physical methods [14], [15]. In this green synthesis method, plants, alga, microorganisms and even enzymes are used in synthesizing. In addition, by using the different NPs properties, the shape and size can be controlled [16].

Plant parts such as root, stem, leaf, fruits and even seed can be used for ZnO Nanoparticles synthesis [17], [18]. The plant extract used to green synthesis the ZnO Nps is a more widely

used process. The plant extract collected contains secondary metabolites [19]. Other than that, it also contains some phytochemical compound as stabilizing, phytochemical compound and capping agents for the synthesis of ZnO Nps [20], [21].

Aloe Barbadensis Miller (Aloe Vera) leaves have countless beneficial properties which many researchers discovered it can be a benefit in many fields especially in medicinal, agricultural, cosmetics etc. [22], [23]. On top of that, a high amount of phenolic compound is also found in Aloe Vera, which are important in cancer treatment nowadays [24]. All part of the Aloe Vera plant such as leaves, roots and stem have been used in agriculture cosmetics and medicinal because of the plant's properties. Properties such as the increasing of effectiveness in antibiotics, the antifungal properties in agriculture and also inhibiting cancer cells [25], [26]. Hence, the extraction of the properties in a safe way by green synthesis the ZnO NPs is crucial.

Zinc acetate dihydrate ($Zn(CH_3COO)_2 \cdot 2H_2O$) affects the optical properties of ZnO Nps. The fast nucleation during the synthesis process may cause the inverse relationship between the optical properties of ZnO nanoparticles and zinc acetate dihydrate concentration [27]. To analyse the optical characterization, a few methods were completed to confirm the properties of the ZnO Nps. The methods used were XRD analysis where the structure of hexagonal wurtzite of ZnO NPs can be observed [28], Field Emission Scanning Electron Microscopy (FE-SEM) analysis to determine the surface morphology, porosity, particle size of the ZnO NPs [29] and Fourier Transform Infrared (FTIR) Spectroscopy Analysis is to determine the polymeric properties of ZnO NPs by relying on infrared light to scan samples and observe bond properties [30]. Besides that, the Ultraviolet-visible spectroscopy (UV-vis) is used to determine the concentration of an analyte in a solution [31].

Due to the high demands of environmental-friendly nanoparticles, high cost and harmful consequences of the commercial method of nanoparticles technology, green synthesis of nanoparticles were recently and widely research. However, through the green synthesis of the ZnO NPs, the concentration of Zinc acetate dihydrate affected the optical characterization of the NPs. Previous research demonstrates that the nanostructure's size is corresponding to the precursor [32], ($Zn(CH_3COO)_2 \cdot 2H_2O$) concentration. Controlled concentration could be obtained by the different characteristics of ZnO and with the different characteristics of the ZnO NPs which applies in the different fields on the usage of ZnO NPs.

2. REVIEW

2.1 Green Synthesis

Green synthesis or biosynthesis is one of the three methods of synthesizing metal nanoparticles which are physical synthesis, chemical synthesis and biosynthesis. In comparing with the physical and chemical synthesis, aside from the toxic nature of the method, the method also produced various disadvantages such as distorted structure, limited growth and slow production rate of the synthesized nanoparticles [33]. Biosynthesis is safer, cost-saving and environmentally friendly.

In obtaining the desired nanoparticles, the two separate basic principles of synthesis are the top-down and bottom-up approaches [13]. Usually, in using these commercial methods, physical and chemical techniques are used in producing the nanoparticles. However, physical techniques are considered highly expensive, while the chemical techniques are considered harmful to the environment and living organisms. Figure 1 shows the different synthesis approaches available for the preparation of metal nanoparticles. Figure 2 shows the example synthesis route of ZnO nanostructures from the leaf extracts.

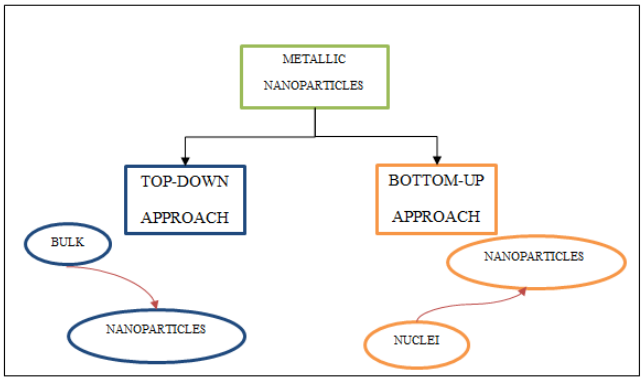

Figure 1: Possible synthesis methods for metal nanoparticles preparation [13]

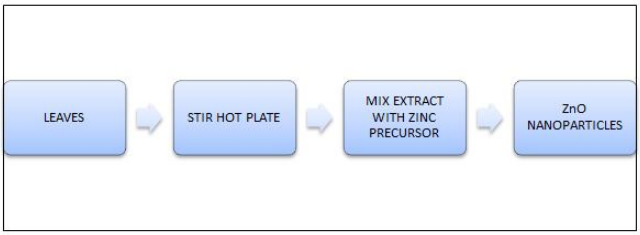

Figure 2: Example workflow of the ZnO nanostructures synthesis method from leaf extracts [34].

2.2 Zinc Oxide

Crude zinc oxide is a yellow-grey granular solid with no odour. It is insoluble in water. Table 1 shows the general properties of ZnO.

Table 1: Properties of ZnO [36]

Properties of ZnO	
Chemical Structure	ZnO
Molecular Weight	81.389 g/mol
Appearance	White solid
Odour	Odourless
Crystal Structure	Wurtzite
Coordination Geometry	Tetrahedral
Band Gap	3.37 eV
Density	5.606 g/cm3

Zinc Oxide is considered a primary hazard and danger to the environment. To restrict its spread to the community, urgent measures should be taken. Metal fume fever with symptoms of

chills , fever, muscle pain , nausea and vomiting can result in prolonged dust inhalation [35]. Hence proper personal protection equipment is required to prevent any unwanted incident from occurring.

2.3 Zinc Oxide Nanostructure

Zinc Oxide is known for its crystalline structure. Wurtzite, Zinc blende and rocksalt are the three crystalline structures of the ZnO [37]. The structure of rocksalt exists during high-pressure condition while the structure of zinc blende would be formed when the ZnO growth on the substrate and the structure in a proper cubic lattice. Wurtzite structure is in hexagonal close-packed, which consist of zinc (Zn) and oxygen (O) atom with attached at tetrahedral sites in alternate arrangement [38].

2.4 Application of Zinc Oxide

In the semiconducting and piezoelectric properties, ZnO has a variety of applications such as in the piezoelectric devices, chemical sensors, environmental protection, optoelectronics, pharmaceutical, cosmetic industries and even textile with its coating technology [5],[4]. Moreover, ZnO also has photocatalytic properties with its application in food as in food packaging and agriculture industry for its antifungal properties [6],[7]. All the properties and its application can be modified by changing the size and morphology [8]. In addition, ZnO Nanoparticles (NPs) also have wide applications in biological and medical such as healing nanomedicine [9], and cosmetic industries due to its antioxidant, antimicrobial, and antibacterial activity [10]–[12].

2.5 Aloe Vera

Aloe Vera is a juicy plant species in the family of Aloe. The plant, which is an evergreen perennial is originating from the Arabian Peninsula which also can be found in hot, semi-tropical and arid climates throughout the world. It is cultivated for agricultural and medicinal uses [39]. All parts of the plant can be used in various ways of application which makes the plant a multifunctional plant [22], [40]. Particularly in medicinal nowadays, Aloe Vera gives the benefits in treating health problem from aiding small wounds to treating cancer cell [23], [39].

3. STRUCTURAL AND OPTICAL CHARACTERIZATION REVIEWS

3.1 Structural Review of Zinc Oxide Synthesis with Aloe Vera

This topic is about the morphology of the surface, the structural and optical characterization of zinc oxide nanoparticles using an extract from the banana leaves. For this study, the characterizations are important in achieving high-quality zinc oxide nanoparticles through the usage of plant extraction. A few devices are needed to identify and investigate nanoparticles with zinc oxide, such as scanning electron microscopy (SEM), XRD, FT-Raman Spectroscopy, UV-Vis, FTIR, and Photoluminescence (PL) Spectroscopy as per in the previous study. Table 2 shows the biosynthesis analysis of zinc oxide nanoparticles using an extract from aloe vera leaves.

The optical characteristics of ZnO were determined by using SEM, XRD, Raman Spectroscopy and Photoluminescence. Parameters like the pH, temperature of the mixture and the concentration of the zinc precursors, were compared through literature review. In addition, the equipment used to determine a process that scans a sample with an electron beam to produce a magnified image was by SEM [40]. Meanwhile, XRD was primarily used for crystalline materials' phase identification as well as obtaining information on unit cell dimensions [40]. As for the technique which provides detailed information about chemical structure, phase and polymorphy, crystallinity and molecular interactions Raman spectroscopy was used, and it is also used to stimulate the emission of a photon from any matter by Photoluminescence [11].

Table 2: Reviewed Aloe Vera Literature

No.	Authors / Years	Optical And Structural Findings
1	E. Varghese and M. George, 2015 [40]	The key advantage of this synthesis is, the synthetic path, cost-effective and straightforward. Thus, we can extract the aloe vera leaf for large-scale ZnONP growth. The synthesized nanoparticles are very active in the degradation in the zinc precursor solution, and in the presence of visible light, illumination catalyzes the decolonisation.
2	R. Majumder et al, 2019 [39]	The structure of the surface morphology is obtained in irregular shapes and confirms the existing ZnONPs by the strongest single bond detected, which is oxygen and hydrogen bond.
3	G. Parthasarathy et al, 2017 [38]	Aloe vera leaves contain alkaloids, tannins, flavonoids, carbohydrates, and terpenoids while the neem plant contains alkaloids, steroids, flavonoids, carbohydrates, glycosides, and terpenoids. Such compounds may

		account for their medicinal uses
4	K. Ali et al, 2016 [43]	Green synthesis is a safer option that avoids the use of high temperature, strain, energy and harmful chemicals without the need for extrinsic surfactants or capping agents in the synthesis of metal NPs.
5	T. K. Mendy et al, 2019 [27]	Green synthesis made using Aloe vera extract with zinc precursor and zinc oxide can be produced with a specific ratio between plant extract and metal ion and the observation on the colour change has corroborated formation of nanoparticles.
6	C. E. Farrugia et al, 2019 [22]	In ALE-ZnONP formation, the study on ALE-ZnONPs revealed the compounds preponderance such as pentadecanoic acid and tetra-conate in ALE. In addition to proteins and other auxiliary phytochemicals, the elements or compounds may be accountable for bestowing stability to associated ZnONPs. The specificity of phytochemicals as presumptive stabilisers and capping agents is speculative.
7	B. H. Abbasi et al, 2019 [42]	This study indicates that, given their source, these substrates serve as reducers and stabilizers or as chelating substances. It is important to note that parameters such as temperature conditions, reaction time, pH and concentrations, in addition to the disparity between the compositions found in biological extracts, greatly alter the final properties of the synthesized nanoparticles.
8	G. Canche-Escamilla et al, 2019 [24]	The concentrations of both the biological extract and the source of zinc, as well as the pH of the solution, play an significant role in the final properties of ZnONPs obtained by a green path.
9	C. Peng et al, 2019 [23]	Due to its bio-compatibility nature and efficient synthesis protocols, ZnO NPs were selected for the current analysis. Throughout the current analysis, all the NPs synthesized demonstrated potent biological activities. S-ZNPs have proved to be the most potent antibacterial agent against the bacterial species being studied. It was also found that S-ZNPs possess the most potent antioxidant capacity.

3.1.1 The Effect of pH on Zinc Oxide Structure

A systematic study of ZnO nanostructure morphological variation by fluctuating the pH of the precursor solution through the green solution process was presented in this review. ZnO nanorod morphology differs markedly from sheetlike to rod-like zinc oxide structure. Due to the diffraction patterns at all pH levels had been well correlated with regular ZnO [27]. High-resolution transmission electron microscopy (HR-TEM) and selected area electron diffraction pattern have confirmed crystallinity and nanostructures, indicating that the structure has evolved along with an ideal lattice fringe [24]. It is deduced that the size or morphology structure can be configured by the differences in pH. In this context, lower pH is best suited for 2D structure, whereby the rod-like structure is developed from the higher pH values. In this context, the consistency of the substance is not affected by the pH solution as observed from the XRD pattern and FTIR spectroscopy significantly. Besides that, the optical properties (UV-vis spectroscopy) of the grown samples were also shown to have good optic properties compared to the bulk ZnO under different conditions.

3.1.2 The Effect of Temperature on Zinc Oxide Structure

At higher temperatures, due to agglomeration, the size of the zinc oxide nanoparticles increases. Raman spectra bands at extend explicitly indicate that the solution contains zinc oxide upon making the high peaks annealed while the FTIR converted radiation to zinc oxide. Also, the amount of zinc precursors being converted to zinc oxide depends on the temperature of the annealing process [39]. Optical transmittance data were used to determine the optical properties of the ZnO nanoparticles. As for the commodity optical band, temperature PL spectrum displays a strong UV band. Optical transmittance data were used to determine the optical properties of the ZnO nanoparticles. The crystalline consistency also can be improved by heat treatment of the

crystals in an O2 atmosphere due to the effects of oxygen entering the crystal lattice, whereby there was an increasing in the stoichiometric proportion of the sample and reducing the vacancies of oxygen in ZnO. For commodity optical band, temperature PL spectrum displays a strong UV band [23].

3.1.3 The Effect of Precursor Concentration on Zinc Oxide Structure

The concentration of the precursor effect on the structural, morphological and optoelectrical characteristics was investigated. XRD characterizations show that the samples are poly-crystalline with a preferential orientation. SEM analyzes however reveal that film morphology depends significantly on the concentration of the precursor including the outlining mechanisms governed by nucleation and/or growth. Based on the next layer built to be deposited in a stacked structure, it has been shown that roughness increases with an increasing precursor concentration while wet capacity can be adjusted by adjusting the precursor concentrations [38]. The present optical transmittance spectrum curves of ZnO films with different concentrations of precursors suggest that the films are highly transparent in the visible field. The opto-electrical analyzes in the studies also show that films that demonstrate higher electrical conductivity also have better photosensitive properties compared to the concentration variations [43]. In the future work, the application of ZnO as a photodetector in fiber optic [44] and X-ray shielding [45] applications will be reviewed.

4. CONCLUSION

As a conclusion, numerous studies note that the possibility of using a green synthesis to obtain ZnONPs by using aloe vera is achievable. Besides that, the studies cited here suggest that with the given source, these substrates act as the reducing and stabilizing substances, or as chelating substances. It is interesting to note that parameters such as temperature conditions, reaction time, pH, and concentrations, in addition to the difference between the discovered compositions in biological extracts, significantly alter the final properties of the synthesized nanoparticles. Between these parameters, the concentrations of both biological extract and zinc source and also the pH, temperature of mixture and concentration of zinc precursor play a major role in the final properties of ZnONPs obtained using the green path, according to the cited literature. Although the complexity of biological substrates still poses a challenge to evaluate the green synthesis of nanoparticles, further investigations on the mechanism of formation of the biological synthesis of ZnONPs are necessary to achieve a better understanding of the chemical processes and reactions that occur during the synthesis. It seems that the green synthesis process, which is important for the large-scale production of ZnONPs, will be regulated and optimized with the designation of the described mechanism. Therefore, the rapidly advancing understanding of green synthesis outlined herein indicates that ZnONPs' enormous potential towards the industrial production in the near future using biological extracts.

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