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BIOSYNTHESIS OF ZnO NANOPARTICLES USING LIME LEAF EXTRACT (*CITRUS AURAANTIFOLIA*) FOR IDENTIFICATION OF LATENT FINGERPRINTS

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ABSTRACT

Fingerprints are an identification tool in forensic science because of their unique properties. Unfortunately, some of the chemicals used in fingerprint powders are toxic and pose a potential health hazard. This study was conducted to analyze the ability of ZnO nanoparticles to identify latent fingerprints. ZnO nanopowder was synthesized with lime leaf extract using the green synthesis method with double-distilled water solvent and characterized by FT-IR at a wavenumber of 4000-400 cm^{-1} and SEM-EDX analysis to provide information about the morphology and to detect the elemental composition nanoparticles. The average particle diameter through SEM was around 173.4 nm and formed a spherical with a rough surface with beige color. Identification of latent fingerprints using the powder dusting method on various porous surfaces (craft paper and greaseproof paper) and non-porous surfaces (glass preparations, aluminium foil, and compact disk) shows visualization with the characteristics of the ridges that look good and clear. The study showed the highest frequency of loops (47%), followed by double loops (20%), plain whorls (30%), and central pocket whorls pattern (3%) from 30 fingerprint samples consisting of 14 men and 16 women. Development identification fingerprints using TiO_2 show visualization more clearly because color contrast from bright white color and detail ridges is shown better with ZnO nanopowder.

Keywords: *Zinc oxide, Nanoparticles, Latent fingerprint*

INTRODUCTION

Fingerprints are one of the essential pieces of evidence at a crime scene for establishing a person's identity [1]. Unfortunately, latent fingerprints are not apparent to the naked eye. Instead, they get onto the surface via bodily fluids and pollutants on the fingertips. Researchers have worked to improve procedures or invent novel fingerprint detection mechanisms for more than a century to improve efficiency, sensitivity, and selectivity [2], [3]. One is developing nano fingerprints, the latest generation of fingerprint development techniques [2].

The theory and its transport properties indicate nanoparticles as small objects with nanoscale dimensions. Metal nanoparticles, metal oxides, and polymer nanoparticles are all nanoparticles. Metal oxide nanoparticles are the most widely used due to their consistent properties and functions [4,5]. In addition, because of their nano size, nanoparticle contact is best on fingerprint ridges [6].

Zinc oxide is a non-toxic, inorganic, polar, and crystalline substance that is inexpensive, safe, and simple to obtain. Because of its unique nature, zinc oxide is employed in various sectors, including pharmaceuticals, cosmetics, food, and contemporary sunscreens [7]. ZnO is used as a nanopowder because of its adhesive properties and interaction with lipids and proteins [8]. Nanopowder having a small size, adheres more easily to the latent print residues [9]. Combination nanopowder of ZnO-SiO_2 synthesized by conventional heat method found the ridges detail on the fingerprint was found to be very good with less staining disturbance on the background [9]. Nitrogen functionalized carbon dots coated on zinc oxide nanoparticles (N-CDs/ZnO NPs) have also developed latent fingerprint detection using powder brushing [10].

Some of the chemicals used in fingerprint powders are toxic and potentially hazardous to health. In addition, chemical synthesis methods using organic solvents and toxic reducing agents are highly reactive and unsafe for the environment [11,12]. In this research, ZnO nanoparticles are synthesized through the biological synthesis method using leaves extract of *citrus aurantiifolia* (lime), an eco-friendly and non-toxic method for providing a solution for developing a non-toxic fingerprint powder material.

Lime is an annual plant native to Southeast Asia found as a wild or garden plant in Indonesia hundreds of years ago [14]. Using water as a solvent, the phytochemicals in lime leaf extract contain flavonoid compounds, steroids, alkaloids, saponins, and tannins [15]. In addition, the phytochemical content of lime leaf extract acts as a reducing agent to convert metal salts (precursors) into metal or metal oxide nanoparticles [16].

Several studies have found that alcohols, phenols, alkanes, and amines in Eclipta (*Eclipta prostrate*) plants participate in the synthesis of nanoparticles. In addition, fig leaf extract *(Ficus carica Linn*), which contains functional groups –OH, C=O, C-OH, and –NH, acts as a reducing agent in the biosynthesis of nanoparticles. [14,15]. Therefore, a biological approach from plant extracts can be an alternative to chemicals in synthesized nanoparticles used to fingerprint powder.

METHODS

1. Materials

Citrus auraantifolia (fresh lime leaves) were collected from gardens in Pematangsiantar, Indonesia. All experiments used double-distilled water, Zinc nitrate hexahydrate ($Zn(NO_3)_2.6H_2O$) (Smart-Lab), TiO_2 powder (Sirchie), transparent adhesive tape (Joyko), porous surface samples (craft paper and greaseproof paper), and non-porous surface samples (glass preparations, aluminium foil, and compact disk).

2. Instrumentation

Glassware (Pyrex), Whatman filter paper (No.1), blender (Willman), analytical balance (Fujitsu), magnetic stirrer (Thermo Scientific), furnace (Gallenhamp Hot Spot), mortar and pestle (PZ), Fourier transform infrared (Thermo Scientific Nicolet), Scanning electron microscopy-energy dispersive X-ray (Zeiss EPOMH 10Zss) and camera phone (Samsung A51).

3. Preparation of Lime Leaf extract

Fresh lime leaves (*Citrus auraantifolia*) were washed in double-distilled water to eliminate impurities. It was then broken into small pieces and dried for five days at room temperature before being mixed for four minutes to make powder. Next, two grams of lime leaf powder were added to a beaker with 100 mL of double-distilled water and stirred with a magnetic stirrer for 10 minutes at 70°C, then raised to 80°C for 10 minutes before cooling to room temperature. The filtrate was used to synthesize ZnO nanoparticles [19,20].

4. Synthesis of ZnO nanoparticles using lime leaf extract

Four grams of zinc nitrate were combined with 85 mL of lime leaf extract, and the mixture was stirred for one hour using a magnetic stirrer. The solution was then placed in a 60°C water bath for one hour. The mixture was then heated at 150°C using a magnetic stirrer to produce a suspension. The solution changed color and formed a suspension, suggesting that the bioreduction of ZnO salts to nanoparticles was complete. The nanoparticles were then calcined at 400°C to yield pure ZnO nanoparticles.

5. Characterization of nanoparticles

The functional groups in lime leaf extract involved in the creation of nanoparticles were identified using FT-IR at a wavenumber of 4000-400 cm^{-1}, and the morphology and composition of elemental substances contained in nanoparticles ZnO nanoparticles were determined using SEM-EDX.

6. Latent fingerprint identification

The participants in this study were 30 students from the State University of Medan's Chemistry Department's 2018-2021 class, with 14 men and 16 women selected randomly. Students who had their fingerprints taken were instructed to wash their hands with soap and air dry them before touching the fingertips of the right hand's thumb on the forehead or face to obtain an oil print (sebum), which helps the fingerprint powder adhere to the media [16].

They were also requested to press their right thumb's fingerprint on several types of porous surfaces (craft paper and greaseproof paper) and non-porous surfaces (glass preparations, aluminium foil, and compact disk). Then, the powder is gently and carefully brushed onto the latent fingerprint impression using the powder dusting process. Finally, the remaining powder should be removed with a gentle tap, and the latent fingerprints should be photographed using a Samsung A51 camera before being removed with masking tape [21,22].

RESULTS AND DISCUSSION

1. Synthesis of ZnO nanoparticles

Two grams of dried lime leaf powder were stirred with double distilled water to speed up the extraction process by supporting the diffusion of the analyte from the bulk solution to the extractant phase.

Lime leaf extract solution, will then be used to synthesize ZnO. nanoparticles [23].

The color change generated by physio-chemical changes in the aqueous solution during the process of stirring ($Zn(NO_3)_2.6H_2O$) with lime leaf extract was regarded as an early indicator of nanoparticle production [20]. The mixture was cooked at 150°C for 10 minutes with a magnetic stirrer, resulting in a dark brown hue and a suspension, suggesting that the nanoparticle bioreduction was complete. The leftover precursor is continually removed and emitted as CO_2 after reaching a temperature of 400°C throughout the calcination process, resulting in pure zinc oxide nanoparticles with beige color.

2. Characteristics of ZnO nanoparticles

The stretching vibration peak at wavenumber 3302 cm^{-1} is a hydroxyl functional group (O-H). The interaction of CO_2 absorption with water molecules on the surface of nanoparticles causes this peak [24]. The stretching vibration band indicates the C-H functional group with alkane compounds with medium intensity at wave number 1384 cm^{-1}. The peak shows an aromatic amine (C-N) functional group at wavenumber 1123 cm^{-1}, which corresponds to the infrared range (1250-1050 cm^{-1}). The stretching vibration band indicates the presence of metal oxides with a wavenumber of 426.72 cm^{-1}. This signal is a Zn-O signal, indicating that the substance is zinc oxide. The results of the FT-IR spectrum of ZnO nanoparticles showed in Figure 1.

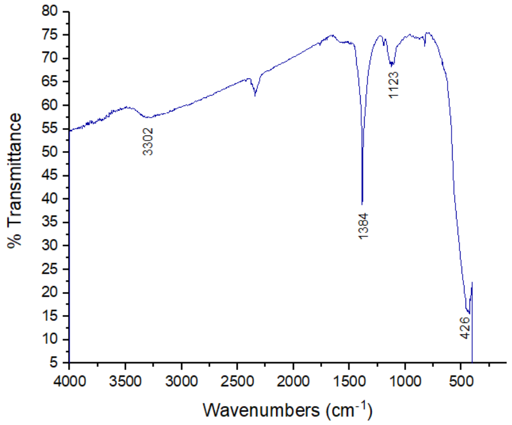


Figure 1. FT-IR spectrum of ZnO nanoparticles

Figure 2 shows the reaction mechanism of a zinc nitrate biosynthesis process with secondary metabolite compounds from lime leaf extract containing hydroxyl groups. Following research that shows the presence of flavonoid, alkaloid, and tannin compounds from the –OH functional group that can donate electrons and reduce ions divalent zinc (Zn^{2+}) and finally to zinc nanoparticles (Zn^0), the presence of the peaks verifies the presence of secondary metabolites in lime leaf extract [25].

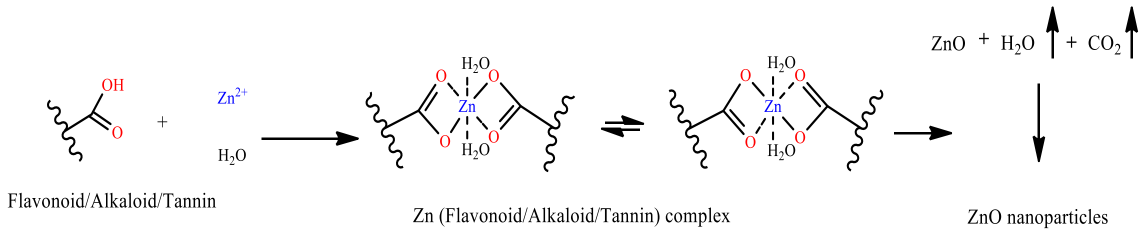


Figure 2. Proposed mechanism of synthesis of ZnO Nanoparticles through *Citrus auraantifolia*

Lime leaf extract's secondary metabolite compound's hydroxyl group lowers and oxidizes Zn^{2+} ions. Due to the release of hydrogen atoms, the hydroxyl group becomes a ketone group. A zinc flavonoid/alkaloid/tannin complex can be formed when a flavonoid/alkaloid/tannin molecule with a free –OH group reacts with $Zn(NO_3)_2$ [26]. The suspension was calcined once the process was finished, yielding pure ZnO nanoparticles [26]. The functional groups in lime leaf extract also have a role in the stability process of nanoparticle development, preventing agglomeration and stabilizing nanoparticles [27].

The surface morphology of zinc oxide nanoparticles synthesized with orange leaf extract at 4.000x and 5.000x magnification by SEM are shown in Figure 3. Morphology of ZnO showed it is close to spherical with a rough surface. The large surface area of the nanoparticles results in agglomeration, and the tremendous attraction between the particles causes the nanoparticles to agglomerate rapidly. [23]. The particle size distribution of ZnO nanoparticles synthesized with lime leaf extract showed that the average particle diameter is 173.4 nm.

The results of the measurement of the diameter of the nanoparticles from the ImageJ program are displayed in the histogram data as shown in Figure 4.

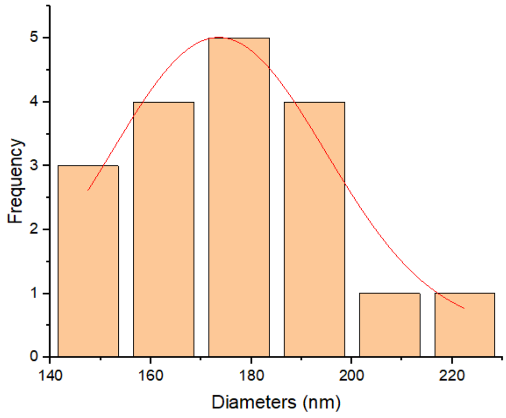
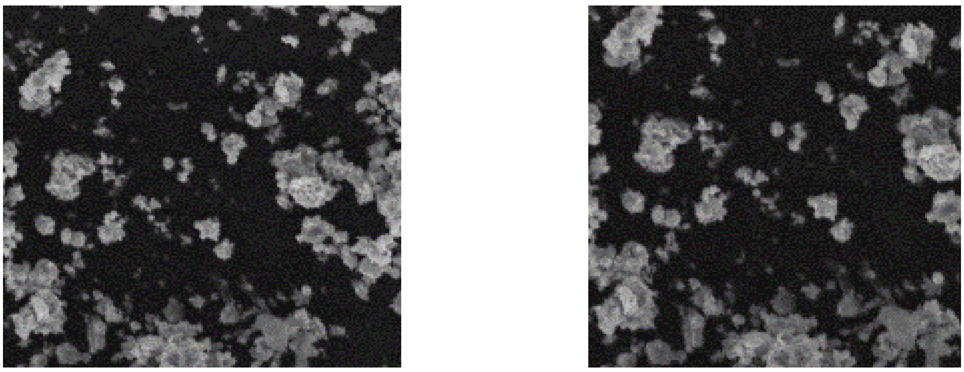


Figure 4. ZnO nanoparticle size distribution histogram

Figure 5 displays the results of the elemental composition for each of the samples as additional information from an EDX investigation. The sample contains components of Zn, O, Pt, In, K and Cl, with Zn accounting for the highest weight per cent, followed by O, Pt, In, K, and Cl accounting for 65.76, 20.69, 11.05, 1.22, 1.02, and 0.24 per cent, respectively.



(a) (b)

Figure 3. Morphology of ZnO nanoparticles based on SEM with a magnification: a) 4.000x b) 5.000x

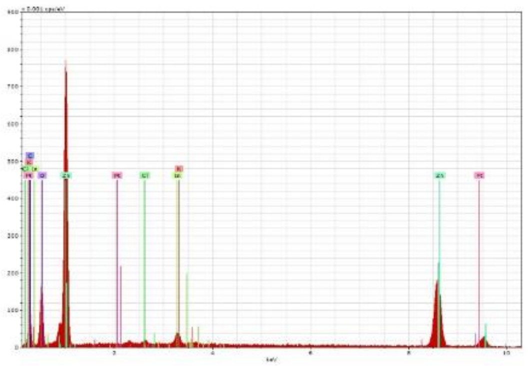


Figure 5. EDX spectrum of ZnO nanoparticles

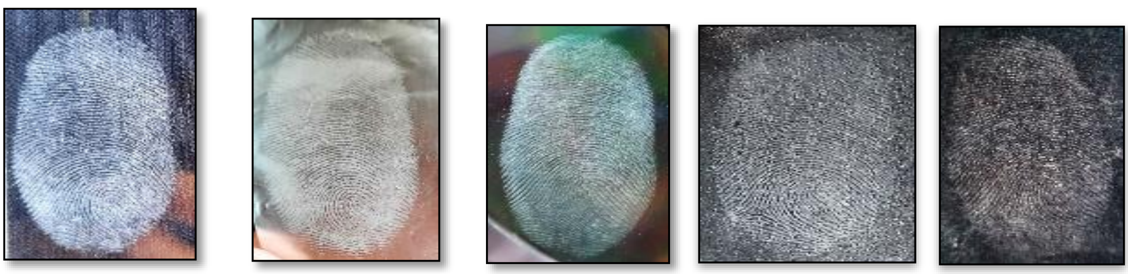
The presence of Zn and O elements confirmed the presence of zinc oxide in the sample and, at the same time, confirmed the purity of the nanoparticles formed, indicating that no element was lost in the process while confirming the presence of zinc as the majority label in the synthesis of zinc oxide nanoparticles [24]. Lime leaf extract residues are still present in ZnO nanoparticles, as evidenced by contaminants other than Zn and O components shown in the EDX spectrum.

3. Latent fingerprint identification using ZnO nanopowder

A total of 30 students had their fingerprint samples printed after following the procedure to get fingerprints. This visualization of latent fingerprint prints is caused by non-covalent/hydrophobic interactions between ZnO nanopowder and fingerprint residues rich in sebaceous secretions of organic compounds that are insoluble in water [9]. Furthermore, adhering the powder to the latent fingerprint can generate superior visualization of the smaller powder size [28].

Visualization of latent fingerprints identified on non-porous surfaces and porous surfaces with ZnO nanopowder in Figure 6 shows good fingerprint print ridges characteristics may be due to the size factor of nanoparticles in identifying latent fingerprints. The discovery backs up the theory that smaller, more delicate, and spherical particles adhere more efficiently than bigger, coarser particles [22]—the finer the powder's capacity to attach to latent fingerprints, the better the visualization.

Furthermore, the findings of this study revealed that one of the elements affecting the quality of latent fingerprints is the kind of media used. This discovery is consistent with prior research employing tea leaf extract to manufacture CuO nanopowder on non-porous surfaces such as glass, white paper, baking paper, and steel [29].



(a) (b) (c) (d) (e)

Figure 6. Visualization of latent fingerprints with ZnO nanopowder on porous and non-porous surfaces: (a) glass preparations, (b) aluminium foil, (c) compact disk, (d) craft paper, and (e) greaseproof paper

4. Distribution pattern of latent fingerprint

Fingerprint prints from 30 students were taken to examine what sort of fingerprints they had. Figure 7 illustrates the percentage of fingerprint patterns in 30 samples from the Department of Chemistry, State University of Medan.

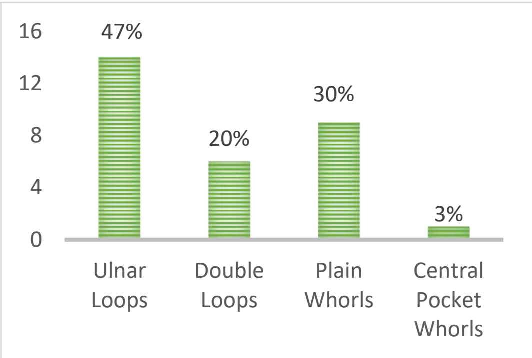


Figure 7. Diagram of the percentage of fingerprint patterns

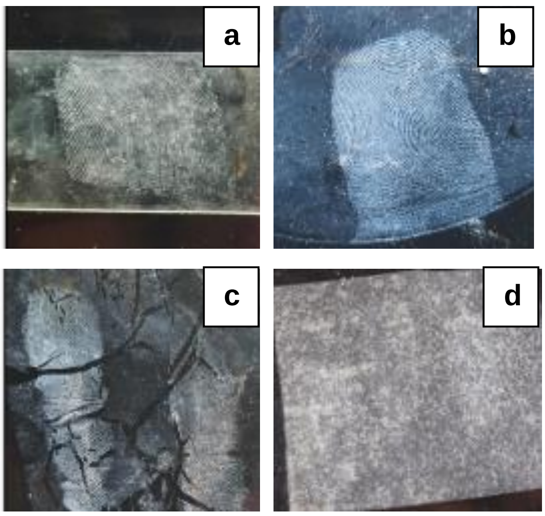


Figure 8. Visualization of latent fingerprints with TiO_2 powder on porous and non-porous surfaces: (a) glass preparations, (b) compact disk,(c) aluminum foil, and (d) craft paper.

The ulnar loops pattern has the most significant number of fingerprint patterns (47%) compared to other fingerprint patterns, indicating that it is quite dominant. The fingerprint pattern with double loops accounts for 20% of the four fingerprint patterns derived from 30 fingerprint samples. While the plain whorls fingerprint pattern accounted for up to 30% of the entire fingerprint pattern, the centre pocket whorls fingerprint pattern accounted for just 3%.

This conclusion was consistent with prior research, which found that 11.6 percent of 120 fingerprint samples had radial loops, 44.1 per cent had ulnar loops, 33.3 per cent had plain whorls, twisted loops, and 0.8 per cent had tented arches [30].

5. Latent fingerprint Development Identification

Latent fingerprint prints were identified with ZnO nanoparticle powder and titanium dioxide (TiO_2) powder with micro size using the powder dusting method. Visualization of latent fingerprints identified on non-porous surfaces and porous surfaces with TiO_2 powder is shown in Figure 8.

The color contrast in latent fingerprint prints was more clearly on the media identified with TiO_2 powder due to the difference that the powder is bright white. In contrast, the ZnO nanopowder was beige-coloured when applied to dark media. In addition, the fingerprint prints at the end of the groove line or branching points are more clearly visible on the ZnO nanopowder due to the nano size of the powder.

The smaller the powder size, the higher the surface area, thus providing better interaction with the ridges in the fingerprint [6] and the better the ability to attach to latent fingerprints and the better the visualization [31]. This may be due to the porous surface causing sebum to quickly absorb below the surface of the craft paper. Another factor that looks less good is caused by a slippery surface which makes it challenging to print ridges on fingerprints with the powder dusting method [30].

CONCLUSION

In summary, we found that ZnO nanoparticles were successfully synthesized with lime leaf extract with a diameter of around 173.4 nm and morphology spherical with a rough surface. The ability of ZnO nanoparticle powder to latent fingerprints on various porous and non-porous surfaces in this study showed visualization with ridge characteristics that looked good and clear. However, latent fingerprint identification using TiO_2 powder showed a more evident color contrast on powder media. Affected by the color difference with ZnO nanopowder, and visualization on porous media using ZnO nanopowder or TiO_2 powder can look less good if the surface is too smooth, which causes sebum to be removed when applying powder with the dusting method.

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