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Biogenic Green Synthesis of Zinc Oxide Nanoparticles from Defatted Coconut Presscake for Wastewater Treatment

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ABSTRACT

Defatted coconut meat presscake is a by-product of coconut milk production, often discarded or used as low value products, but has the potential to treat wastewater. The aim of this study was to characterize zinc oxide nanoparticles (ZnO NPs) from defatted coconut meat presscake and determine its effect on the degradation of contaminants in wastewater and antibacterial properties. The nanoparticles were created via a co-precipitation method using zinc nitrate as a precursor and defatted coconut meat presscake extract as a reducing agent. The characteristics of the nanoparticles were then characterized using FTIR (Fourier Transform Infrared Spectroscopy), FE-SEM (Field-Emission Scanning Electron Microscopy), XRD (X-ray diffraction), and TGA (Thermogravimetric Analysis). Characterization revealed ZnO NPs with a hexagonal wurtzite structure and an average size of 54 nm. The photocatalytic degradation of methylene blue under UV light irradiation showed a degradation efficiency of 79.5 % within 180 min. Treated wastewater with Zn ONPs at concentrations of 0.5, 1, and 5 mg/L showed a significantly reduced microbial load, with an average of 10.2×10⁶, 8.3×10⁶ and 3.8×10⁶ CFU/mL, respectively compared to untreated wastewater (16.5×106 CFU/mL), indicating the antimicrobial properties of ZnO NPs. Therefore, ZnO NPs, synthesized from defatted coconut meat presscake extract, offer an eco-friendly solution for efficiently treating wastewater from food industry.

INTRODUCTION

Wastewater treatment is crucial for the food industry in Malaysia, serving to safeguard the environment, ensure regulatory adherence, conserve water resources, and uphold public health. The sector's growth has resulted in a surge of wastewater discharge containing a spectrum of pollutants, from organic matter to potentially hazardous substances. While conventional treatment methods are effective, they often present challenges in terms of cost and environmental impact [1, 2]. Utilizing biogenic nano-materials from food by-products like defatted coconut meat presscake from coconut milk extraction, presents a sustainable solution. These nanomaterials, synthesized from waste extracts, have demonstrated significant potential in previous studies for efficiently treating the diverse

pollutants present in food industry wastewater [3-5]. Nanoparticles offer promising solutions for treating wastewater through various mechanisms, including antimicrobial action and photocatalysis.

This study focuses on developing such materials from defatted coconut meat presscake for food industry wastewater treatment. By using the Taguchi method, the aim is to optimize the synthesis of ZnO nanoparticles from defatted coconut meat presscake extract. The goal is to enhance their photocatalytic activity for better pollutant degradation, particularly under UV irradiation. UV light serves as the primary energy source for initiating and driving photocatalytic reactions involving nanoparticles, thereby enabling efficient and sustainable wastewater treatment processes. Additionally, the study

explores the potential antibacterial activity of these nanoparticles, expanding their application in food industry wastewater treatment. By investigating defatted coconut meat presscake as a sustainable source for biogenic nanomaterials, this research aims to provide a sustainable solution to the wastewater treatment challenges faced by Malaysia's food industry.

MATERIALS AND METHODS

Materials

Defatted coconut meat presscake was obtained from a Public Market in Serdang, Selangor. Zinc acetate dihydrate was obtained from R&M Chemicals, UK and methylene blue was purchased from Nacalai Tesque, Kyoto, Japan. Distilled water and all chemicals were analytical grade. Glassware used was thoroughly washed and dried. Defatted coconut meat presscake was washed with tap water followed by removal of coconut shell specks, then cleaned by mixing with boiled water and pressed using a coconut milk screw presser to remove any excess water or oil. The residue was frozen at -40°C, freeze-dried to 7% moisture in a Scanvac Coolsafe Freeze Dryer, ground into fine particles with a kitchen blender, vacuum packed, and stored at 4°C.

Sample preparation

In order to obtain defatted coconut meat residue extract (CMRE), 2.5 g of defatted coconut meat presscake powder was stirred with 100 mL distilled water for 45 min at 200 rpm, then centrifuged at 5000 rpm for 30 min to obtain a cloudy extract. The filtered extract was refrigerated for use. ZnO NPs were synthesized via a co-precipitation method with CMRE and zinc acetate solution. Zinc acetate (0.1 M) in 100 mL water was stirred, and 50 mL CMRE was added dropwise until the pH reached 12. After stirring for 2 h, the solution was centrifuged, pellets washed, dried, ground, and stored.

The Taguchi method was used with a fractional factorial design (L9) for nine experimental runs, varying precursor concentration, defatted coconut meat presscake extract volume, pH, and temperature. The experimental parameters were within the following ranges: precursor concentration ranged from 0.05 to 0.20, extract volume from 20 to 100 mL, pH from 8 to 12, and temperature from 0°C to 90°C. From the Taguchi analysis and UV-Vis results, it was determined that a combination of zinc acetate (0.1 M) in 100 mL water and 50 mL CMRE at pH 12 yielded optimal results in terms of particle size. Subsequently, this optimized sample was selected for further characterization.

UV-Vis, X-ray diffraction (XRD), FT-IR spectrometer, and Field Emission Scanning Electron Microscope (FESEM)

Confirmation of ZnO NPs was done with UV–Vis spectrophotometry (200–800 nm), XRD for crystal structure, FT-IR spectrometer for functional groups, and FESEM for surface morphology. Thermal stability was analyzed with TGA-DSC. Methylene blue (MB) was prepared, with 15 mg catalyst added to MB. After equilibration, MB concentration was recorded, followed by UV-light exposure and subsequent concentration measurements. Photocatalytic degradation was calculated. Antibacterial activity against pathogenic bacteria in fish processing industry wastewater was assessed by the colony counting method. Varying concentrations (0, 0.1, 1, 5 mg/L) of ZnO nanoparticles were mixed with wastewater and stirred for four h. Serial dilutions were plated on agar, incubated at 37°C for 24 h, and colonies were counted using a colony counter device.

RESULT AND DISCUSSION

CMRE successfully facilitated the synthesis of zinc oxide nanoparticles (ZnO NPs), evidenced by the formation of a white precipitate when added to a zinc acetate solution. The UV-VIS spectrum in **Fig. 1** displayed a prominent absorbance peak at 367 nm, characteristic of ZnO NPs. This aligns with established ranges for ZnO NPs (350-375 nm) as seen in previous studies [6,7,8]. The optical band gap of the ZnO NPs was calculated at 2.74 eV as shown in **Fig. 2**, indicating their semiconductor nature suitable for applications in photocatalysis and optoelectronics.



Fig. 1. UV-Vis spectrum of ZnO NPs synthesized from defatted coconut meat presscake extract.



Fig. 2. Optical band gap of the synthesized ZnO NPs from defatted coconut meat presscake extract.

XRD patterns in **Fig. 3** revealed distinct peaks corresponding to the hexagonal wurtzite ZnO structure, with peaks at (100), (002), (101), (102), (110), (103), (200), (112), and (201) planes. The sharp, intense peaks indicated high crystallinity and the average crystallite size was determined as 20.68 nm using the Debye-Scherrer formula, consistent with literature findings [9-11].



Fig. 3. XRD pattern of synthesized ZnO NPs from defatted coconut meat presscake extract

FTIR spectra in **Fig.** 4 exhibited characteristic peaks indicative of functional groups in the ZnO NPs. Peaks at 3372 cm⁻¹ indicated O–H stretching, 2852 cm⁻¹ suggested C–H bonds, and 1558/1411 cm⁻¹ indicated C=O stretching vibrations. The peak at 876 cm⁻¹ confirmed tetrahedral coordination of Zn, with 607 cm⁻¹ supporting ZnO stretching vibrations, further confirming ZnO NPs formation.



Fig. 4. FTIR spectra of synthesized ZnO NPs from defatted coconut meat presscake extract.

FESEM images in Fig. 5 illustrated the morphology of ZnO NPs from defatted coconut meat residue, showing irregular shapes and sizes ranging from 34 to 75 nm. The observed agglomeration was likely due to the uniform distribution of Zn cations, indicating successful ZnO NPs synthesis with the extract. Fig. 6 demonstrated the thermal stability of the ZnO NPs. Minimal weight loss, especially beyond 400 °C, indicated strong stability, essential for various applications of ZnO NPs.



Fig. 5. FESEM image of ZnO NPs from defatted coconut meat presscake extract under 50000× and 100000× magnifications.



Fig. 6. TGA analysis of ZnO NPs from defatted coconut meat presscake extract.

Fig. 7(a) shows the evaluation of the photocatalytic activity using methylene blue degradation where a 79.5% reduction after 180 min of exposure to ZnO NPs was obtained. This effective degradation was attributed to the generation of reactive species like hydroxyl radicals and superoxide radical anions under UV light [12]. The degradation of methylene blue solution without photocatalyst was shown in **Fig. 7(b)** and it can be seen that there was no considerable reduction.



Fig. 7. (a) Photocatalytic degradation of methylene blue dye by ZnO NPs and (b) without ZnO NPs from defatted coconut meat presscake.

For the assessment of antibacterial activity, **Table 1** revealed a significant reduction in bacterial count, with a 77% reduction at the highest ZnO NPs concentration of 5 mg/L. This highlights the potential of ZnO NPs as an antimicrobial agent in wastewater treatment.

 Table 1. Bacteria count (CFU/mL) of wastewater with different concentration of zinc oxide nanoparticles and percentage of removal.

Concentration zinc oxide nanopartic (mg/L)	of Bacteria count cles (CFU/mL)	Reduction (%)
0 (control)	16.5 x 10 ⁶	0
0.5	10.2 x 10 ⁶	38
1	8.3 x 10 ⁶	49
5	3.8 x 10 ⁶	77

ZnO NPs exhibit enhanced antibacterial effects with increased concentration, generating H_2O_2 on their surface. This leads to damaged bacterial cell membranes, where the nanoparticles attach firmly to inactivate bacteria, preventing further action [13].

CONCLUSION

The findings of this study concluded that ZnO NPs have been successfully synthesized in an environmentally friendly and sustainable manner using an aqueous extract of defatted coconut meat presscake as a reducing agent. The FTIR analysis showed the formation of Zn–O bond. The XRD image analysis proved high crystallinity hexagonal ZnO crystals of the wurtzite type were formed. The findings of the photocatalytic dye degradation demonstrated that ZnO NPs effectively separate photogenerated electron–hole pairs, which is a prerequisite for enhanced photocatalytic performance. Under UV exposure, the breakdown of MB reached 79.5% in 180 min. The ZnO NPs

also showed a significant microbial reduction in wastewater. The results of this study highlight the potential of defatted coconut meat presscake to synthesize multifunctional ZnO NPs used for wastewater treatment and microorganisms' inactivation.

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