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ZnO-CuO Nanoporous Composite Synthesized using Gum Arabic (*Acacia senegalensis*) as an Emerging Nanolarvicide for *Culex quinquefasciatus* Larvae Management

Zaccheus Shehu[1], Ezra Abba[2*], Blessing Emmanuel[3]
[1]*Chemistry Department, Faculty of Science, Gombe State University, Gombe, Nigeria*
[2]*Department of Zoology, Faculty of Science, Gombe State University, Gombe, Nigeria*
[3]*Department of Biology, Faculty of Science, Gombe State University, Gombe, Nigeria*

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Abstract

Culex quinquefasciatus is an intermediate host of several human pathogens and serve as a vector of filariasis worldwide. Recently, there are increasing efforts to develop new and effective nanoparticles to control mosquito vectors. The green synthesis of ZnO-CuO nanoporous composite **using Gum Arabic (*Acacia senegalensis*)** was successfully carried out with Gum Arabic and the composite was characterized by UV-Visible, FTIR, and SEM/EDX techniques. Three different larval instars (1st, 2nd, and 3rd) of *Culex quinquefasciatus* were exposed to various concentrations (10, 20, 25 mg/l) of the ZnO-CuO nanocomposite for 24 h. The LC_{50} values for the 1st, 2nd, and 3rd instars were found to be 4.702, 5.459, and 5.459, respectively, while the LC_{90} values were 22.923, 33.922, and 33.922, respectively. The correlation coefficient values for the three instars were found to be 0.982, 0.941, and 0.941, respectively, and these indicate concentration-dependent larvicidal activity. The novel ZnO-CuO nanoporous composite could serve as a new nanolarvicide against the filariasis-transmitting vector.

Keywords: Nanolarvicide, Nanotoxicity, ZnO-CuO Composite, Larvae Management, *Culex quinquefasciatus*

Introduction

ZnO, CuO, and their nanocomposites have been synthesized with confirmed nanolarvicidal activity against different species of mosquito larvae according to Ramdayevi *et al*. [1], Sharon *et al*. [2], Hassanain *et al*. [3], Kirthi *et al*. [4], Al-Dhabi and Arasu [5], Roopan *et al*. [6], Murugan *et al*. [7], Vijayakumar *et al*. [8] and Mostafa *et al*. [9]. New development of nanotechnology aimed to produce nanoparticles or nanocomposites through greener pathway with less or no effects on human health and environment [10]. Thus, it has been reported that semiconductors such as ZnO and CuO nanoparticles provide a ground of a novel green nanotechnology to control insect pests, including mosquitoes [11]. Ragavendran *et al*. [12] stated that infectious diseases caused by mosquitoes as vectors have recently been increased due to urbanization, travel, and trade. So far, there are no effective preventive measures such as vaccines available. As such, the only effective solution is to prevent the mosquito (vectors) from breeding and biting humans. The most common and synthetic chemicals used for mosquito control include diflubenzuron, malathion, pyrethroid, and methoprene [13]. Nonetheless, these synthetic chemicals have adverse effects on non-target organisms or humans and, moreover, the mosquitoes have developed resistance to the chemicals. Thus,

*Email: ezra.abba@gmail.com

nanotechnological approaches, especially ZnO and CuO based nanolarvicides, have proven better in controlling disease-carrying vectors and could solve the problem of resistance development. They also cause no or less effects to the non-target organisms [1-9]. Therefore, in this study, ZnO-CuO nanoporous composite was prepared **using Gum Arabic (*Acacia senegalensis*)** and used as an emerging nanolarvicide for *Culex quinquefasciatus* larvae management.

Materials and Methods

Experimental Site

The experiment was carried out in the Chemistry Laboratory of the Chemistry Department and the Insectary of the Department of Biological Sciences, Gombe State University, Nigeria. *Cx. quinquefasciatus* larvae were collected from different breeding sites within Gombe metropolis.

Collection and Extraction of Gum Arabic

Gum Arabic was collected from Billiri, the Local Government Area of Gombe State. They were collected on the basis of cost effectiveness, ease of collection, availability, and medicinal properties. The gum extract was neatly collected and allowed to dry properly under the sun. Gum Arabic was crushed to powder using pistil and mortar.

Synthesis of ZnO-CuO Nanoporous Composite using Gum Arabic (*Acacia senegalensis*)

The ZnO-CuO nanoporous composite **using Gum Arabic (*Acacia senegalensis*)** was synthesized based on the method reported by Fardood *et al*. [14]. To synthesize ZnO-CuO nanoporous composite **using Gum Arabic (*Acacia senegalensis*)**, 1 g of Gum Arabic (GA) was placed in a beaker and 40 ml of distilled water was added. It was then placed on a magnetic stirrer hotplate for dissolution for 10 minutes at 90 °C. Following this, 2 g of $Zn(NO_3)_2.6H_2O$ (DBH product) and 2 g of $Cu(NO_3)_2.6H_2O$ (DBH product) were added and stirred continuously for 2 hours. A blue-green resin was formed. The obtained resin was then transferred into a crucible, covered with a paper foil, and then placed in the laboratory furnace at 450 ^oC for 2 hours.

Characterization of ZnO-CuO Nanoporous Composite using Gum Arabic (*Acacia senegalensis*)

The formation of ZnO-CuO nanoporous composite using Gum Arabic as a reducing agent was monitored by spectral analysis. UV-VIS spectrophotometer (Perkin Elmer Lamda Spectrophotometer) was used to monitor the formation of the nanoparticles with absorbance in the range of 200–800 nm. Fourier transform infrared (FTIR) analysis of the sample was performed using Perkin Elmer Spectrum version 10.03. The FTIR spectra of the synthesized ZnO-CuO nanoporous composite were analyzed and discussed for the possible functional groups for the formation of the composite. The morphology of the composite was studied using scanning electron microscopy (SEM; JEOL, Model JFC-1600). The chemical composition of the nanocomposite was studied using energy dispersed x-ray (EDX) technique.

Mosquito Collection

Culex quinquefasciatus larvae were collected from different identified breeding sites (sewage and sullage water collections, including cesspools, cesspits, drains and septic tanks, etc.) in Gombe metropolis, using ladle and a collection bottle. The ladle was lowered into the water (breeding site) at an angle of about 45^0 until one side is just below the surface of the water. While dipping, care was taken not to disturb the larvae which may cause them to swim downward. The larvae were maintained and reared in the laboratory for larvicidal bioassay. The collection and identification were performed based on the method described by Kamaraj *et al*. [15], with minor modifications.

Larvicidal Bioassay

One gram of aqueous extract was first dissolved in 100 mL of distilled water (stock solution).

A solution of 100mg/L of the NPs was prepared. From the stock suspension, 5, 10, 20, 25 and 50mg/L concentrations were prepared subsequently by serial dilution. A 100 mL of these concentrations were placed together with twenty larvae of each of the first, second, and third instars in plastic cups. Test of each concentration against each instar was repeated two times. After 24 h exposure, larval mortality was recorded. In each case, the control comprised 25 larvae in 200 ml of distilled water, according to Wilson *et al*. [16]. Percentage mortality was calculated as follows:

$$\text{Percentage mortality} = \frac{\text{Number of dead larvae}}{\text{Number of larvae introduced}} \times 100$$

Data Analysis

Mean percent larval mortality data were subjected to analysis of variance. LC_{50}, LC_{90}, and their associated confidence intervals were estimated from 24 h concentration with mortality data using probit analysis (SPSS version 2016).

Results and Discussion

UV-Visible Analysis

Absorption spectrum of synthesized ZnO-CuO nanocomposite using Gum Arabic (*Acacia senegalensis*) (Figure 1) at different wave lengths, ranging from 280 to 800 nm, was measured using UV-Vis spectrophotometer (Perkin Elmer Lamda Spectrophotometer). The maximum absorption wavelength at 280 nm reveals the formation of nanocomposite. Findings also indicate that ZnO-CuO nanocomposite generally consists of a characteristic peak, ranging from 270 to 400 nm [17]. It was reported that low transmittance, i.e. below 380 nm, indicates strong absorbance for ZnO-CuO nanocomposite [18].

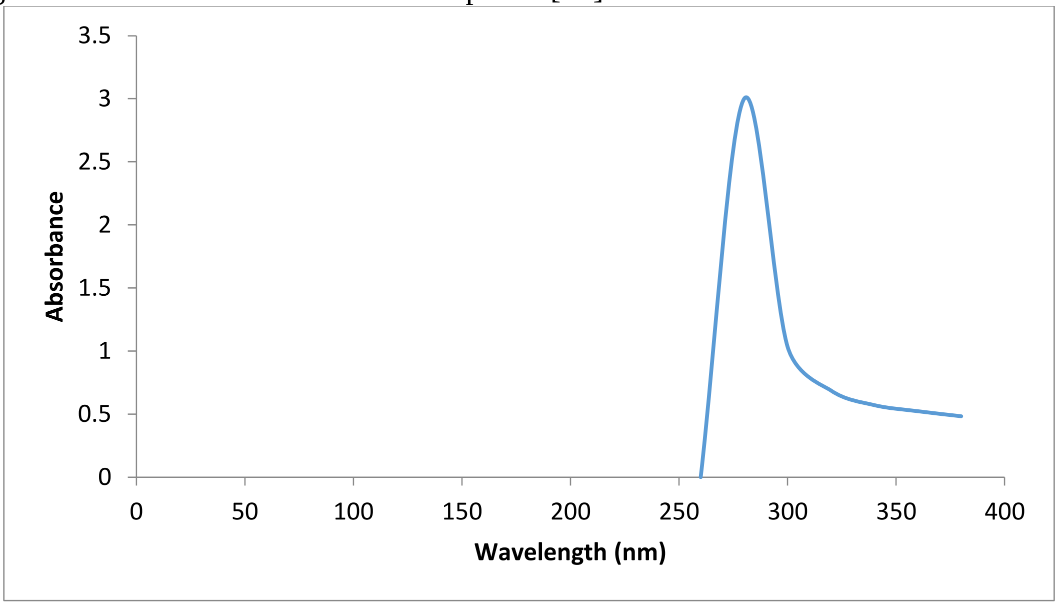


Figure 1-UV-Visible spectrum of ZnO-CuO nanocomposite synthesized using Gum Arabic (*Acacia senegalensis*)

Fourier Transform Infrared (FTIR) Analysis

FTIR spectrum of ZnO-CuO nanocomposite **using Gum Arabic (*Acacia senegalensis*)** (Figure 2) exhibited prominent peaks at 3428.88, 2923.33, 1621.11, 1384.49, 1093.29, and 482.92 cm^{-1}. These distinct peaks represent the multifunctional groups indicating –OH stretching, C-H (due to aldehyde), –OH bending of water molecules, and C-C for 3428.88, 2923.33, 1621.11, and 1384.49 92 cm^{-1} present in the nanocomposite. It was reported that due to inter-atomic vibrations, the absorption of metal oxides falls below 1000 cm^{-1}. Thus, absorption peaks at 1093.29 and 482.92 cm^{-1} are responsible for Zn-O/ Cu-O interactions.

This is in agreement with FTIR analysis results of ZnO-CuO nanocomposite reported by Sakib *et al*. [19], Widiarti *et al*. [20], Saravanakkumar *et al*. [21], Da Silva *et al*. [22] and Allaf and Hope-Weeks [23].

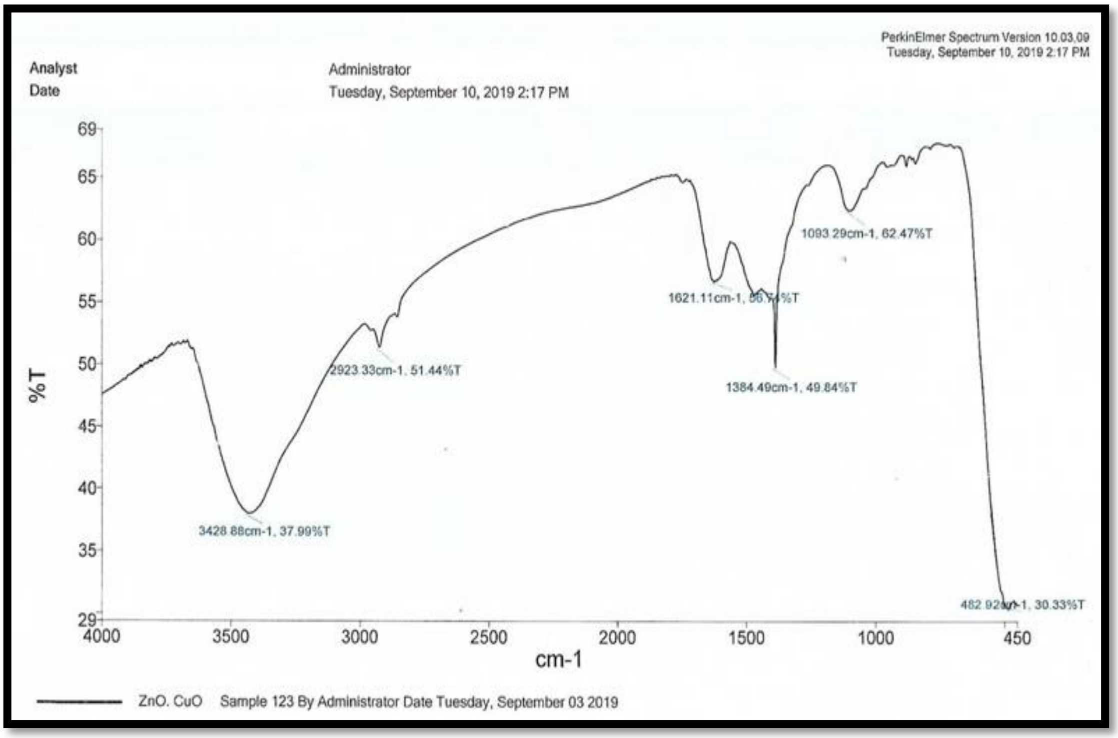
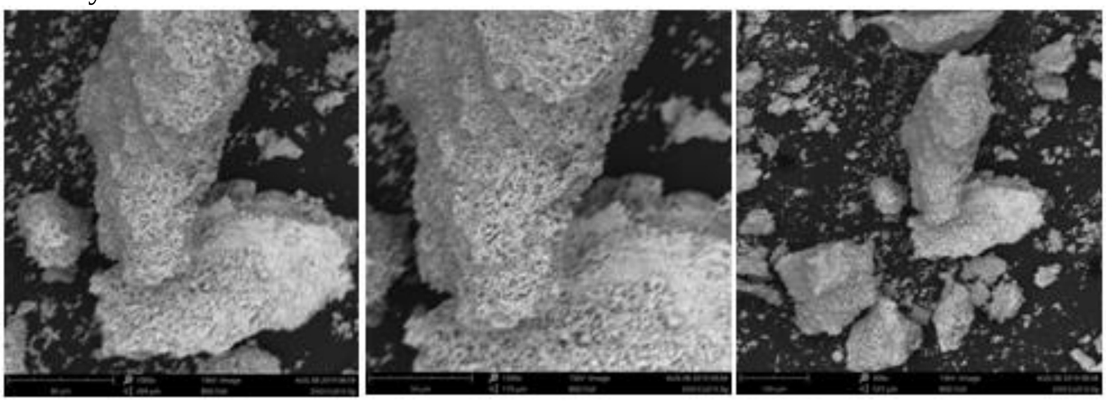


Figure 2- FTIR spectrum of ZnO-CuO nanoporous composite synthesized using Gum Arabic (*Acacia senegalensis*)

Scanning Electron Microscopy (SEM) Analysis

SEM determinations of the sample showed the formation of nanoparticles. SEM analysis showed that the synthesized ZnO-CuO nanoporous composite **using Gum Arabic (*Acacia senegalensis*)** was clearly distinguishable. The analysis at various resolutions showed that is the composite is nanoporous and has an irregular shape (Figure 3). However, the morphological structure of ZnO-CuO observed involve nanowire, nanorod, flower-like, spherinoid, nanoflakes, and networked flakes by Zhu *et al*. [24], Jung *et al*. [25], Sabry *et al*. [18], Sakib *et al*. [19], Saravanakkumar *et al*. [21] and Allaf and Hope-Weeks [23], respectively.



a b c

Figure 3-SEM image of synthesized ZnO-CuO nanocomposite synthesized using Gum Arabic (*Acacia senegalensis*): **a**(100µm), **b**(50µm, 1500×) and **c**(80µm, 1000×).

Energy Dispersed X-ray (EDX) Analysis

The elemental composition analysis of the ZnO-CuO nanoporous composite **using Gum Arabic (*Acacia senegalensis*)** demonstrated elements with different atomic concentrations (Figure 4). The results showed that copper, zinc, and oxygen have the percentage weight concentrations of 39.40%, 37.46% and 2.39%, respectively, as listed in Table 1. The EDX results showed that the compound composition with CuO and ZnO has the highest atomic concentrations of 46.0335, and 38.6654 respectively as described in Table 2. This also confirmed the formation of ZnO-CuO nanocomposite in a good proportion. However, it was confirmed that the presence of other elements is due to the Gum Arabic used in the synthesis, as reported by Mahendran *et al*. [26].

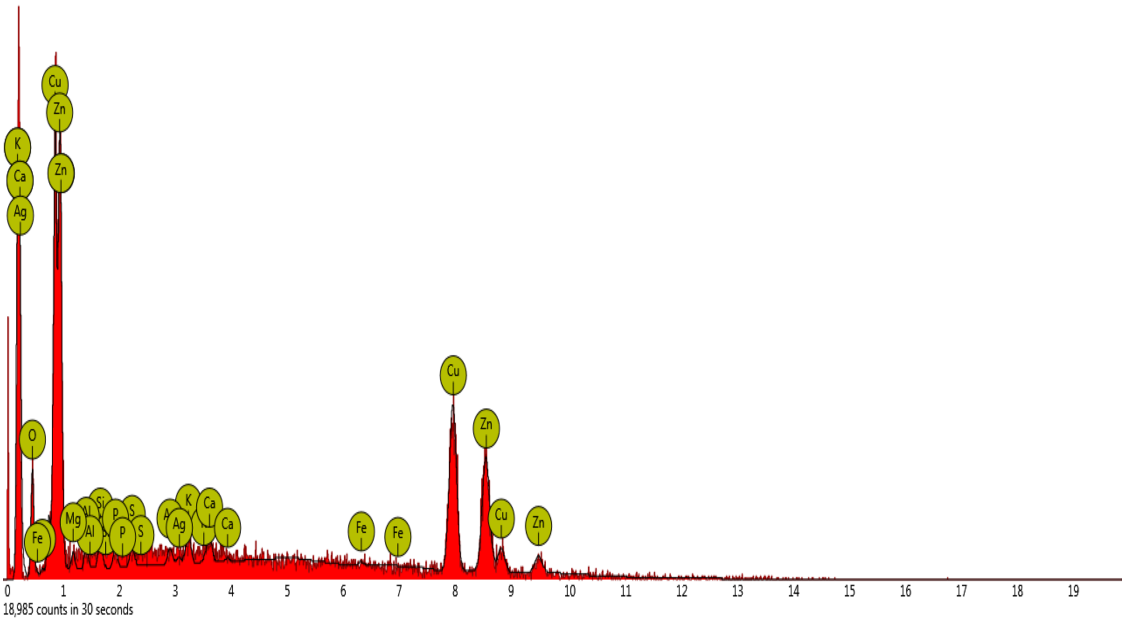


Figure 4-EDX spectrum of ZnO-CuO nanoporous composite synthesized using Gum Arabic (*Acacia senegalensis*)

Table 1: Elemental composition of ZnO-CuO nanoporous composite synthesized using Gum Arabic (*Acacia senegalensis*)

Element Number	Element Symbol	Element Name	Atomic Conc.	% Weight Conc.
29	Cu	Copper	28.76	39.40
30	Zn	Zinc	26.57	37.46
11	Na	Sodium	31.23	15.48
8	O	Oxygen	6.92	2.39
47	Ag	Silver	0.48	1.11
19	K	Potassium	0.84	0.71
14	Si	Silicon	1.10	0.67
20	Ca	Calcium	0.76	0.66
13	Al	Aluminium	0.92	0.53
12	Mg	Magnesium	0.94	0.49
16	S	Sulfur	0.65	0.45
15	P	Phosphorus	0.61	0.41
26	Fe	Iron	0.20	0.24

Table 2-Compound composition of ZnO-CuO nanoporous composite synthesized using Gum Arabic (*Acacia senegalensis*)

Element	Content	Detection limit	Error
SiO_2(%)	4.6831	0.0000	0.0443
K_2O(%)	0.1961	0.0000	0.0225
CaO(%)	0.7294	0.0000	0.0455
MnO(%)	0.0156	0.0000	0.0009
Fe_2O_3(%)	0.2567	0.0000	0.0040
NiO(%)	0.1826	0.0000	0.0043
CuO(%)	46.0335	0.0000	0.0504
ZnO(%)	38.6654	0.0000	0.0393
HfO_2(%)	0.0355	0.0000	0.0005
WO_3(%)	0.0610	0.0000	0.0019

Nanotoxicity Assay

In this study, *Culex quinquefasciatus* larvae were exposed to various concentrations of synthesized ZnO-CuO nanocomposite for 24 hours. The values of percentage mortality, lethal concentrations, chi square, and correlations are presented in Table 3. Larval average percentage mortality for the 1st, 2nd and 3rd instars were found to increase with the increase in concentrations of ZnO-CuO nanocomposite synthesized using Gum Arabic (*Acacia senegalensis*), as indicated by the correlation coefficient. The Lethal concentration (LC) increases with the increase in larval age; hence, the higher the instar stage, the higher the lethal concentration. The control (distilled water) showed nil mortality in the current assay. The lethal effect of *Culex quinquefasciatus* on the three instar larvae showed the values of LC_{50}=4.702, 5.459, and 5.459 mg/ml, respectively, and LC_{90}=22.923, 33.922, and 33.922 mg/ml, respectively. Similarly, concentration-dependent mortality was reported by earlier studies [10, 16, 27-30], . Previous findings of larvicidal activity of ZnO nanoparticles revealed that the LC_{50} and LC_{90} were 0.72 and 27.29 mg/L, respectively, on the 3rd instar of *Culex quinquefasciatus* [9]. These valuess are higher than those found in the current study. However, larvicidal activities of Ag [27], Al_2O_3-ZnO [9], Ag [30], Ag-Co [10], and Cu/Ni [16] nanoparticles on the third larvae of *Culex quinquefasciatus* showed LC_{50} values of 48.98 mg/L, 7.93 mg/L, 21.84 mg/ml, 13.63mg/L, and 18.50 mg/L, respectively. Thus, these values of larvicidal activities from the literature are lower than that of the current investigation, indicating the applicability of ZnO-CuO nanoporous composite as a potential nanolarvicide.

Table 3-Effects of synthesized ZnO-CuO nanocomposite synthesized using Gum Arabic (*Acacia senegalensis*) on larvae *Culex quinquefasciatus*

Instar	Conc. (Mg/l)	Mortality (%)	SD (±)	LC_{50} mg/l	LC_{90} mg/l	χ^2	R
	10	74	5.163978				
1st	20	84	7.302967	4.702	22.923	0.625	0.982
	25	94	2.309401				
	10	68	6.531973				
2nd	20	76	3.265986	5.459	33.922	0.986	0.941
	25	90	5.163978				
	10	68	6.531973				
3rd	20	76	3.265986	5.459	33.922	0.986	0.941
	25	90	5.163978				

LC_{50}, lethal concentration (mg/l) that kills 50% of larvae; LC_{90}, lethal concentration that kills 90% of larvae; SD, standard deviation, Mean value of five replicates; R, correlation coefficient, χ^2, chi square

Conclusions

Green synthesis of nanocomposite using plant derivatives was extremely studied in the last two decades. The plant metabolites induce the production of metallic nanoparticles in an ecofriendly manner. In the present study, focus was on green synthesis of ZnO-CuO nanoporous composite using Gum Arabic. The physical properties of the synthesized nanocomposite were characterized using relevant techniques. Furthermore, demonstration of the possible application of ZnO-CuO nanoporous composite as a substitute to insecticides to show possible mosquito larvicidal activity was performed. Our results showed that ZnO-CuO nanoporous composite has high efficiency against *Culex quinquefasciatus* larvae. One obvious advantage of using this composite as larvicide is the low risk of developing resistance by the insects in long term use.

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Conflict of Interest

Authors declare that there is no conflict of interest.

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