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Green Synthesis and Characterization of $CuO@SiO_2$ Nanocomposite using Gum Arabic (*Acacia senegalensis)* (L) against Malaria Vectors

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Abstract

The nanocomposite $CuO@SiO_2$ was synthesized through green pathway using Gum Arabic. The green synthesized nanocomposite was characterized using Scanning Electron Microscopy (SEM), Energy Diffraction X-Ray (EDX), Ultra-Violet/Visible spectroscopy, and Fourier Transform Infra-Red (FTIR). These techniques confirmed the formation of $CuO@SiO_2$ nanocomposite. The toxicity studies were conducted for 24 h on 1st, 2nd, 3rd, and 4th instars of malaria vectors at various concentrations of 10, 20, and 25 Mg/L. The LC_{50} for the 1st, 2nd, 3rd, and 4th instars were found to be 7.525, 7.980, 8.026 and 9.741 Mg/L, respectively, whilst the LC_{90} for the 1st, 2nd, 3rd and 4th instars were found to be 14.839, 24.937, 57.77 and 48.5 Mg/L, respectively. The correlation coefficient values between concentrations and mortality for instars were obtained in the range of 0.944 - 0.984. These correlation coefficient values indicate that mortality rates increases with the increase in concentrations. The $CuO@SiO_2$ nanocomposite could be a new nanolarvicide for malaria vector control in tropical countries.

Keywords: Green synthesis, Larval instars, Nanocomposite, Toxicity, Vector control

Introduction

Mosquitoes are known to be vectors of several diseases, including malaria, yellow fever, Dengue, filariasis and chikungunya [1]. Anopheles gambiae, Anopheles coluzzi, and Anopheles arabiensis are the most common insect vectors in Northeastern Nigeria, transmitting mosquito-borne disease in the Sub-Saharan Africa [2,3]. According to the recent World Malaria Report, there were an estimated 228 million malaria cases globally, among which an estimated 405,000 deaths were recorded in the year 2018. The majority of the cases, about 213 million, were from the African Region. This region also accounted for 94 % of malaria deaths in the same year [3].

Vector control is one of the approaches that has been widely accepted as a means of curtailing the menace of vector borne diseases. In Nigeria, the current malaria vector control tool relies greatly on the use of chemicals in the form of insecticides. These insecticides have been classified into 4 classes: carbamates, organochlorines, organophosphates, and pyrethroids [2,4]. However, there have been reports of widespread resistance to these classes of insecticides in several countries, including Nigeria. This results in an enormous setback in the campaign against malaria in these countries [5]. Another disadvantage of using chemicals is that they pose the risk of environmental and human health damage. As a consequence of these challenges of insecticide resistance and environmental toxicity being reported, it has become necessary to develop alternative mosquito control measures that are natural and novel [6,7].

Recently, vector control biologists have been taking advantage of nanotechnology as an alternative measure to control insect vectors [8]. Nanotechnology is a rapidly developing interdisciplinary area of research and development, involving biology, physics, and chemistry [9].

There are different methods for the synthesis of nanoparticles, which include physical, chemical, and green routes. The green route provides an advancement over the other 2 methods. It is cost effective and environmentally friendly, because nontoxic reagents are normally used. Moreover, green synthesis of nanoparticles has the advantage of easy large-scale production [10,11].

Biological synthesis is done by biological organisms such as yeasts, algae, molds, bacteria, actinobacteria, plants, or plant products. The molecules found in these organisms perform nanoparticle synthesis by the reduction reaction [8,12-16].

The green synthesis of nanoparticles is initiated by the addition of extracts from parts of plants, such as roots, fruits, or leaves, into aqueous solutions of either metal ions or oxides [17]. There are wide reports of green synthesis of copper oxide nanoparticles using different plant extracts and their diverse applications as antibacterial agents [18-21] and as larvicides [22]. The literature also indicates the synthesis of silica oxide using the green route, applied for various purposes such as insecticide, antimicrobial agents, and molluscicides [23-25]

Gum Arabic, an exudate of Acacia senegalensis (L), comprises 3 distinctive fractions, referred to as: arabinogalactan (AG), consisting of 88.4 % of the total, with a low protein content (0.44 % w/w); arabinogalactan-protein (AGP), consisting of 10.4 % of the total, with a high protein of 9.18 w/w; and glycoprotein (GP), consisting of about 1 % of the total, with a protein content of 50 % w/w [26]. Gum Arabic is reportedly used as a reducing and stabilizing agent in the synthesis of metallic nanoparticles [26-27]. This plant species has naturally adapted to, and is very common in, Sub-Saharan countries, particularly Nigeria, Mauritania, Niger, Mali, Senegal, Sudan, and Chad [28]. This is the first report of the synthesis of CuO@SiO2 nanocomposite using gum Arabic as a biomaterial for the management of malaria vectors.

Materials and methods

Chemicals and apparatus/instrument

All the reagents used in this study were analytical grade and DBH products. The glass wares and apparatus used were Pyrex products, while the instruments included an Ultraviolet–Visible spectrophotometer (model 6705), an SEM/EDX (model PhenomWorld), and an FTIR (PerkinElmer Spectrum Version 10.0309).

Collection of gum arabic extrudes

A fresh *A. senegalensis* (L) extrude (Gum Arabic) was obtained from Billiri, a Local Government Area in Gombe state. The extrude was dried under shade and ground using mortar and pestle until it became a powder. It was kept under room temperature in the laboratory of the Department of Chemistry, Gombe State University.

Synthesis of CuO@SiO_2 nanocomposite

The synthesis of the nanoparticles followed the prescription of [29]. A beaker containing 40 mL of 1 g of Gum Arabic was magnetically stirred for 10 min at 90 °C on a hotplate for complete dissolution. Following this, 2 g each of copper nitrate and silica gel were added and stirred for 120 min and the same temperature was maintained. By addition of copper nitrate and silica, the aqueous solution was changed to blue-green. With time, the solution became viscous. A cloud formation at the bottom of the beaker indicated the formation of resin. The obtained resin was then transferred into a crucible and covered with a paper foil, then placed in the laboratory furnace at 450 °C for 2 h to obtain the CuO@SiO_2 nanocomposite powder.

Characterization of CuO@SiO_2 nanocomposite

The chemical bonding of CuO@SiO_2 nanocomposite was investigated using a PerkinElmer Spectrum Version 10.03.09 Fourier Transform Infrared Spectrophotometer (FTIR). The morphology and chemical composition of the synthesized CuO@SiO_2 nanocomposite was evaluated using an SEM/EDX (PhenomWorld model). A UV-Visible spectrophotometer model 6705 was employed for optical measurement between the ranges of 260 - 380 nm wavelength. A spectrum was plotted with absorbance against wavelength (nm).

Collection of mosquito larvae

Laboratory-reared larvae of *Anopheles gambiae complex* was obtained from the Gombe State Roll Back Malaria Insectary (Malaria Control Booster Insectary, Gombe, Nigeria). The methods of [30-31] were used for the identification of the larval instars of the mosquitoes. The larvae were kept and maintained under standard conditions of 25 ± 2 °C and were kept in de-chlorinated water. A mixture of dog biscuits and yeast powder in the ratio of 3:1 was used to feed the larvae, as per the methods of World Health Organization [32].

Larvicidal activity of CuO@SiO_2 against *Anopheles gambiae* complex

To obtain the stock solution, 0.1 g of CuO@SiO_2 nanocomposite was weighed and then diluted in distilled water using a 1,000 mL volumetric flask and was shaken to obtain 100 Mg/L concentration.

From the stock solution, 10 Mg/L, 20 Mg/L, and 25 Mg/L concentrations were prepared through serial dilution. The larvicidal activity of $CuO@SiO_2$ nanocomposite against *Anopheles gambiae complex* was assessed following [32] protocol, as per the method of [33]. 25 larvae (1st instar) were placed in 100 mL of each desired concentration of $CuO@SiO_2$ nanocomposite (10 Mg/L, 20 Mg/L, and 25 Mg/L). Tests of each concentration against each larval instar were replicated 4 times. In each case, the control comprised 25 larvae in 200 mL of distilled water. The same process was conducted for the 2nd to 4th instar larvae. Percentage mortality was calculated as follows:

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

Data analysis

The mean mortality was calculated. The LC_{50} and LC_{90} were also computed using probit analysis. SPSS (Statistical software package) version 25.0 was used.

Results and discussion

Larvicidal bioassay

This study reported the larvicidal activity of the synthesized $CuO@SiO_2$ nanocomposite against the larvae of *Anopheles gambiae complex* after 24 h exposure. The percentage mortality observed for the 1st instar larvae were 70 ± 6, 98 ± 3 and 98 ± 3 % for 10, 20 and 25 Mg/L concentrations, respectively. The mortality of the 2nd instar larvae revealed 60 ± 6, 85 ± 6 and 90 ± 0 % for 10, 20 and 25 Mg/L concentrations, respectively (**Table 1**). When exposed to the nanocomposite, the 3rd larval instars showed 55 ± 6, 75 ± 6 and 75 ± 6 % mortality to 10, 20 and 25 Mg/L concentrations, respectively, as shown in **Table 1**. Finally, for the 4th larval instar, the mortality revealed 50 ± 6, 75 ± 0 and 75 ± 0 % for 10, 20 and 25 Mg/L concentrations, respectively (**Table 1**). Overall, the highest larval mortality (98 ± 3 %) was observed in the 1st instar larvae when subjected to 25 Mg/L of the $CuO@SiO_2$ nanocomposite for 24 h. However, the lowest larval mortality (50 ± 6) was recorded in the 4th instar larvae after exposure to 10 Mg/L of $CuO@SiO_2$ nanocomposite (**Table 1**). Similarly, the highest mortality was reported in synthesized Ag NPs against *Culex quinquefasciatus* and *Culex gelidus* at the concentration of 25 Mg/L [34]. Contrarily, Danbature *et al.* [35] and Wilson *et al*. [36] reported a lower susceptibility of *Culex quinquefasciatus* subjected to varying concentrations of Ag-Co BMNPs and Cu/Ni BMNPs, respectively. According to these findings, it is obvious that the mortality recorded is dose dependent. Santhoshkumar *et al.* [33] and Velayutham *et al.* [37] reported similar findings indicating the progression of mortality based on the increasing concentration of the nanoparticles. More so, the mortality rate reported in this present study concurs with several reports which showed the efficacy of nanoparticles against the larvae of mosquitoes [34,37-38,]. The calculated LC_{50} for the 1st, 2nd, 3rd, and 4th larval instars were 7.525(5.69 - 8.842), 7.98(5.207 - 9.943), 8.026(2.762 - 11.130) and 9.741(0.001 - 14.47) Mg/L (LCL–UCL), respectively, and the calculated LC_{90} for the 1st - 4th larval instars showed 14.839(13.218-17.282), 24.937(20.771-34.725), 57.770(35.225-337.888) and 48.741(33.22 - 131.009) Mg/L (LCL–UCL), respectively (**Table 1**). [34] reported LC_{50} values that were slightly higher compared to the findings of this work. Their results showed LC_{50} (UCL-LCL) values of 12.00(9.3 - 13.01) Mg/L as a result of *C. quinquefasciatus* exposure to Ag NPs of *Ficus racemosa*. On the other hand, biosynthesized SiO_2 nanoparticle using marigold flower against the 1st to 4th instar larvae of *Aedes aegypti* showed LC_{50} values of 3.85, 4.24, 4.66 and 5.08 Mg/L [39], which are lower than the findings in this work. Hence, it can be deduced that the biosynthesized $CuO@SiO_2$ nanocomposite has good penetration capacity due to its small size and high surface to volume ratio, which results in likely disruption of organelles and enzymes in young juvenile instars, as they are easily susceptible to the action of $CuO@SiO_2$ nanocomposite.

Table 1 Larvicidal activity of $CuO@SiO_2$ nanocomposite against 1st to 4th instars larvae of *Anopheles gambiae complex*.

Larval instar	Conc. (Mg/L)	% Mortality (Mg/L) ± SD	LC_{50} Mg/L (LCL–UCL)	LC_{90} Mg/L (LCL–UCL)	χ^2	r
1st	10	70 ± 6	7.525(5.69 - 8.842)	14.839(13.218 - 17.282)	0.945	1
	20	98 ± 3				
	25	98 ± 3				
2nd	10	60 ± 6	7.98(5.207 - 9.943)	24.937(20.771 - 34.725)	0.001	0.984
	20	85 ± 6				
	25	90 ± 0				
3rd	10	55 ± 6	8.026(2.762 - 11.130)	57.770(35.225 - 337.888)	0.591	0.945
	20	75 ± 6				
	25	75 ± 6				
4th	10	50 ± 6	9.741(0.001 - 14.47)	48.741(33.22 - 131.009)	0.893	0.945
	20	75 ± 0				
	25	75 ± 0				

LC_{50}: Lethal concentration that kills 50 % of the exposed larvae, LC_{90}: Lethal concentration that kills 90 % of the exposed larvae, UCL: Upper confidence limit, LCL: Lower confidence limit, χ^2: Chi-square value, r: correlation coefficient, SD: Standard deviation

Characterization of $CuO@SiO_2$ Nanocomposite

Fourier Transform Infra-Red (FTIR) spectrum of the Gum Arabic and the synthesized $CuO@SiO_2$ nanocomposite are shown in **Figures 1(a)** and **1(b)**, respectively. For the Gum Arabic spectrum (**Figure 1(a)**), the characteristic absorption band at 3,435.15 cm^{-1} representing the amino group must have been masked by the broad OH group absorption band. The bands at 2,924.76 cm^{-1} indicated the presence of sugars, galactose, arabinose, and rhamnose, as well as the presence of alkane C-H stretch and aldehyde C-H stretch. The polymers also showed the characteristic band of C=C stretch, amide NH bend, NO_2 from both aliphatic and aromatic galactoproteins, and amino acids around 1,633.24 cm^{-1}. Alkane CH_3 bend, aromatic C=C stretch, ketone C-C stretch, carboxylic acid C-O stretch, anhydride C-O stretch, and amine C-N stretch from polysaccharides and galactoproteins were observed at the 1,384.37 cm^{-1} band. The band at 1,254.58 cm^{-1} represented alkane CH_3 bend, alcohol C-O stretch, ether C-O-C stretch, carboxylic acid CO stretch, amine C-N stretch, and alkyl due to sugar backbone showing alkane bend, alcohol stretch. Ether stretch is due to the attachment of 2 galactose sugars, and CO and CN stretches from galactoproteins. A distinct band at around 1,035.35 cm-^{1} represented alkene C-H bend from polysaccharides of the Gum Arabic, while the shoulder signals at around 450 cm^{-1} were assigned to metal residue in Gum Arabic. The $CuO@SiO_2$ nanocomposite spectrum (**Figure 1(b)**) exhibited prominent peaks at 3,445.19, 2,924.76, 1,633.08, 1,082.94 and 450.00 cm^{-1}. The band at 450.00 cm^{-1} identified vibration of the Cu–O and Si-O bond [40-42]. The band at 1,082.94 cm^{-1} corresponded to asymmetric stretching vibration of the Si-O-Si bond. The peaks at 3,445.19 and 1,633.08 cm^{-1} indicated H-O-H stretching and bending mode of silanol and adsorbed water, respectively [40-42]. The band at 1,384.37 cm^{-1} ascribed to polysaccharides and galactoproteins, and the band at 1,254.58 cm^{-1} that represented sugar backbone in the Gum Arabic spectrum were absent in the nanocomposite. This indicates that the polysaccharides and galactoproteins had decomposed during heat treatment at 450 °C. However, the presence of bands at around 2,900 cm^{-1} for both the Gum Arabic and the $CuO@SiO_2$ nanocomposite indicated the residual alkane C-H stretch and aldehyde C-H stretch in both samples.

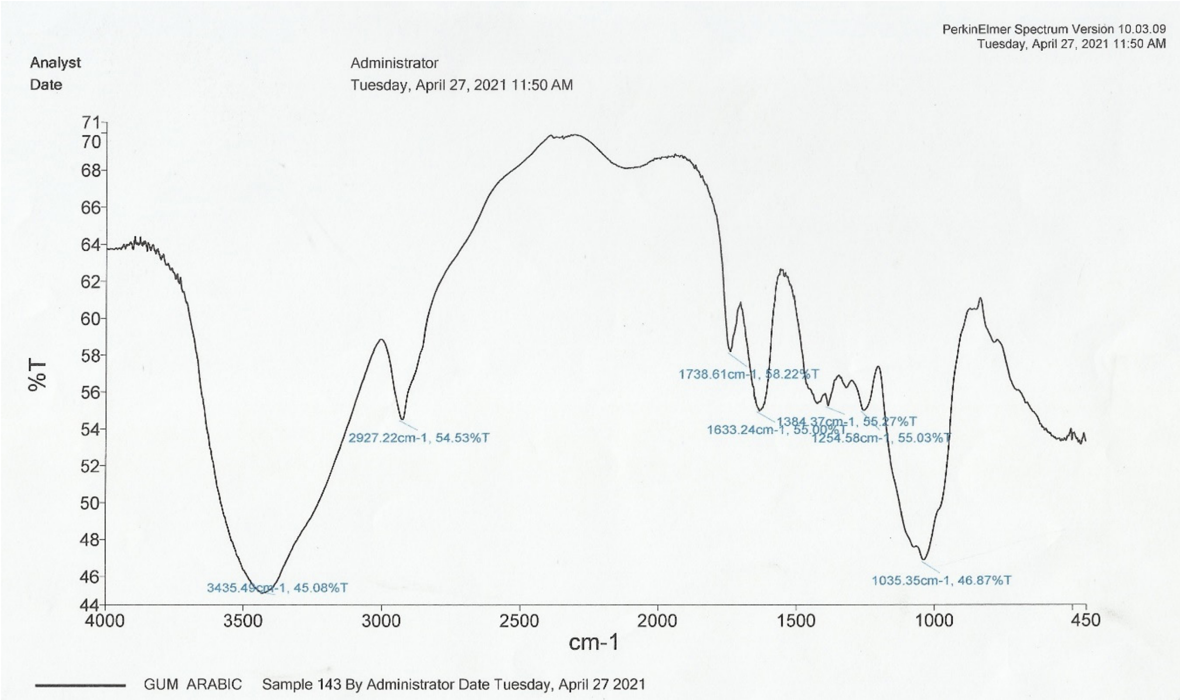

Figure 1(a) FTIR spectrum of Gum Arabic.

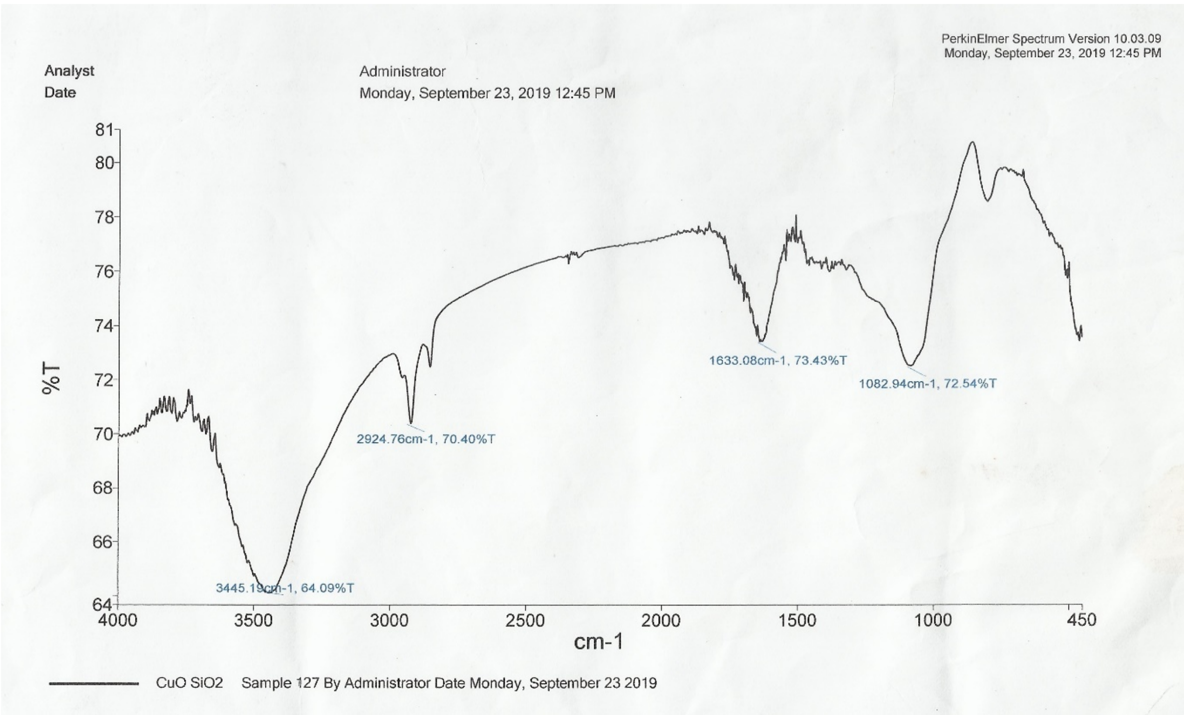

Figure 1(b) FTIR spectrum of CuO-SiO_2 nanocomposite.

Absorption spectrum of synthesized CuO@SiO_2 at different wavelengths, ranging from 280 to 380 nm, revealed a peak at 280 nm due to surface plasmon resonance (**Figure 2**). This absorption peak is in agreement with other findings as reported by [40,43-44].

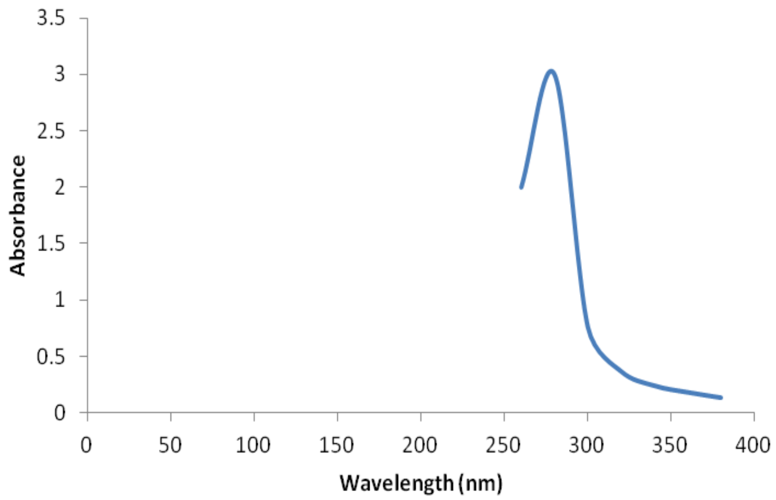

Figure 2 UV-spectrophotometric analysis of $CuO@SiO_2$ nanocomposite.

Scanning Electron Microscopy (SEM) image of $CuO@SiO_2$ nanocomposite, Energy Diffraction X-Ray (EDX) elemental composition of $CuO@SiO_2$ nanocomposite, and EDX micrograph of $CuO@SiO_2$ nanocomposite are shown in **Figures 3** and **4** and **Table 2**, respectively. The SEM image of $CuO@SiO_2$ nanocomposite revealed that the formed particles possessed larger size and were also irregular. Similarly, a larger size and irregular shape of $CuO@SiO_2$ nanocomposite has been reported by [40]. The composition of copper, silicon, and oxygen were found to be 43.63, 40.51 and 9.78 %, respectively. In total, $CuO@SiO_2$ nanocomposite made up 93.92 % of the sample. Other elements and compositions were found to be 6.09 % (**Table 2** and **Figure 4**). Within the $CuO@SiO_2$ nanocomposite, copper was found to be, by a small amount, the major component, followed by silicon and oxygen, respectively. Conversely, oxygen was the major component, which was followed by silicon and copper, respectively, in $CuO@SiO_2$ nanocomposite synthesized by [40].

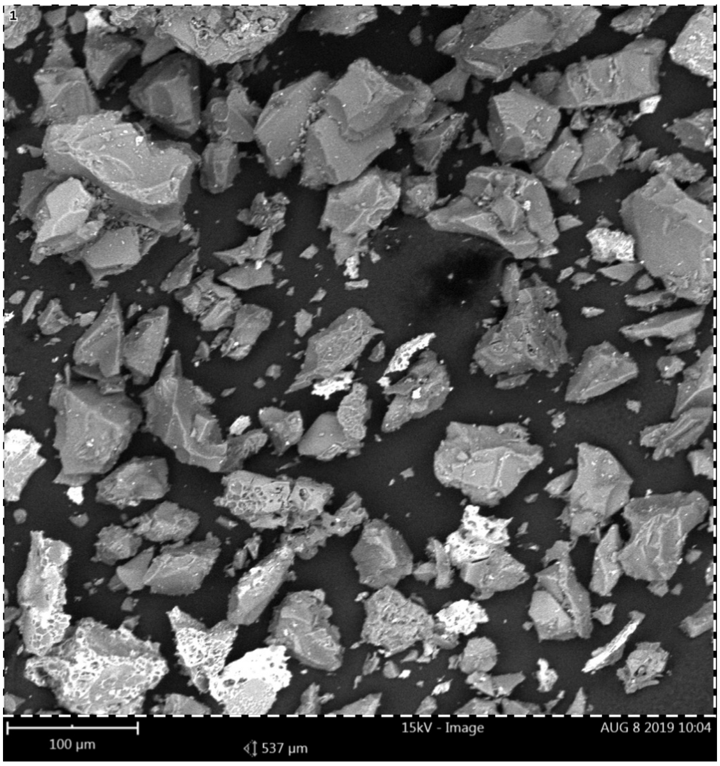

Figure 3 SEM image of $CuO@SiO_2$ nanocomposite.

Table 2 EDX elemental composition of $CuO@SiO_2$ nanocomposite.

Element number	Element symbol	Element name	Atomic Conc.	Weight Conc.
29	Cu	Copper	23.39	43.63
14	Si	Silicon	49.14	40.51
8	O	Oxygen	20.83	9.78
11	Na	Sodium	1.82	1.23
13	Al	Aluminum	1.53	1.21
47	Ag	Silver	0.29	0.92
12	Mg	Magnesium	1.06	0.76
15	P	Phosphorus	0.75	0.68
16	S	Sulfur	0.51	0.48
20	Ca	Calcium	0.37	0.43
19	K	Potassium	0.33	0.38

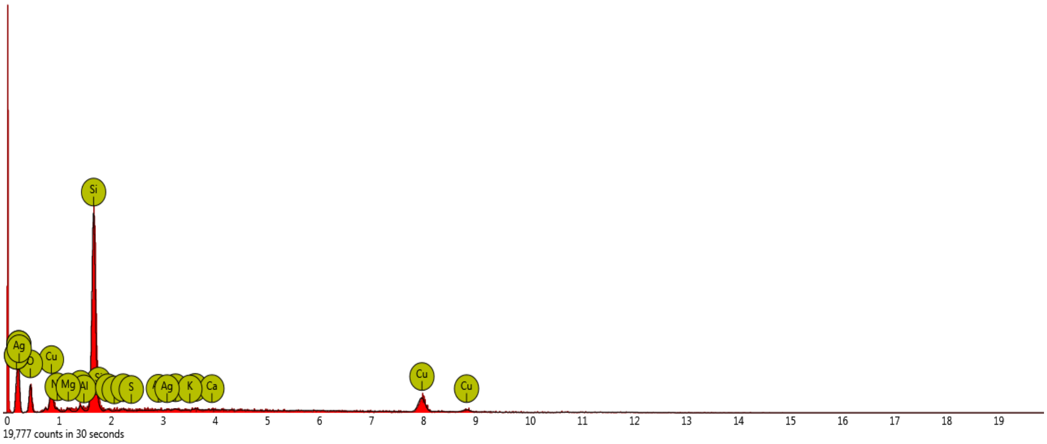

Figure 4 EDX micrograph of $CuO@SiO_2$ nanocomposite.

Mechanism of control of $CuO@SiO_2$ nanocomposite using Gum Arabic (GA)

The large chains of Gum Arabic are natural polysaccharides, which form complexes with Zn-$^{2+}$ ions which interact with functional groups such as –COOH, –OH, and –NH_2 [45]. The complexes formed lead to the prevention of excess growth, producing ZnO NPs of smaller size. A simple explanation of the mechanism of formation and growth of CuO in the presence of Gum Arabic (GA) and silica can be given as the following:

$$Cu(NO_3)_2 \xrightarrow{Aqueous\ solution} Cu^{2+} + NO_3^- \quad (1)$$

$$Cu^{2+} + AG \xrightarrow{Aqueous\ solution} [Cu(\mathrm{GA})]^{2+} \quad (2)$$

$$[Cu(GA)]^{2+} + Silica \xrightarrow{Heat} [Cu(GA)/Silica]^{2+} \xrightarrow{Calcination\ at\ 450\ o_C} CuO/SiO_2 \quad (3)$$

Conclusions

For the first time, the synthesis of CuO@SiO_2 nanocomposites using Gum Arabic is reported. The CuO@SiO_2 nanocomposite was ascertained using different characterization techniques, SEM, EDX, UV-spectrophotometric analysis, and FTIR. The larvicidal activity was also tested against the larval instars of malaria vectors, and the LC_{50} and LC_{90} were computed. The findings indicated that CuO@SiO_2 nanocomposites could be a potential nanolarvicide for larval source management of malaria vectors. Consequently, the toxicity of this nanocomposites against non-target organisms in the same habitat as the mosquito larvae needs to be ascertained, and a field trial of this nanocomposite is also recommended for validation. The mode of action of this nanocomposite needs to be investigated.

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