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Synthesis of Copper Oxide Nanoparticles by a Novel Method and its Application in the Degradation of Methyl Orange

Henam Sylvia Devi[1] and Thiyam David Singh[2]

[1,2]Department of Humanities and Basic Sciences,
National Institute of Technology, Imphal -795001, Manipur.
E-mail: [1]henambi411@gmail.com,[2]davidthiyam@yahoo.co.in

Abstract

This work is to report an environmental benign route for the fabrication of copper oxide nanoparticles using Centella asiatica (L.) leaves extracts at room temperature. This method is completely a green method, free from toxic and harmful solvent. Copper oxide particles such prepared are in Nano scale and there morphology and size are characterized using SEM, UV –Visible spectroscopy, IR spectroscopy and EDX. Copper oxide NPs synthesized by this method can be used for the photo catalytic degradation of methyl orange. These NPs can reduce methyl orange to its leuco form in aqueous medium in the absent of reducing agents. It is more economy as compare to other methods. This catalytic effect of copper oxide nanoparticles can be contributed to its small size. Nanoparticles have many active sites as compared to the bulk materials because of its large surface to volume ratio. Copper oxide nanoparticles such prepared has good catalytic properties.

Keywords: copper nanoparticles, green method, catalyst, centella asiatica (L.), methyl orange.

1. Introduction

Nano size materials exhibit unique electronic, magnetic, optical, catalytic and medicinal properties as compared with the traditional and commercial bulk materials (Mingqing and Junhui et al, 2011). It is due to its quantum size effect, large surface to volume ratio. Copper nanoparticles are of great interest due to its easiness of preparation and significant promising physical and chemical properties. There are

many methods for the synthesis of copper nanoparticles (Cu NPs) like precipitation methods, decomposition methods, plasma methods, pulsed wire explosion methods, sol gel methods, vapor deposition, electrochemical, radiolysis methods and so on(Suleiman and Mousa et al,2013). Most of these methods are complicated and have drawbacks like use of hazardous organic solvents, expensive reagent, toxic by product, drastic reaction condition, difficult to isolate NPs, longer time etc. There arises a growing need to develop green synthesis routes for the particles. By following 12 principles of green chemistry, new routes for the synthesis of NPs using plants extracts and microorganism are given emphasis as compared to the commercial and traditional methods. Copper NPs such synthesized have similar physical as well as chemical properties as prepared by chemical methods. Copper NPs have antibacterial activity, catalytic properties, biocidal properties and use in wound dressings, gas sensors, super conductors, solar cells (Honary and Barabadi et al, 2012) and thermal conductivity and so on. Nano biotechnology synthesis of NPs using microorganism like fungi, bacteria, yeasts etc. intracellular or extracellular are also reported but these methods required lots of maintenance and it is complex. In this paper tailoring of CuO NPs is done by using *Centella asiatica* (*C.asiatica*) dried leaves extract. Nano size particles obtained from this method play very important role in the degradation of dyes which is one of the organic pollutants. Organic pollutants are of serious concern now. Heavy metals, Phenols and is derivatives, nitro groups containing fertilizers, insectides, pesticides , chemicals etc. are the main organic pollutants and are hazardous as the harmful to human health not only to human, it also affects the aquatic life. Main source of these pollutants is industrial waste water. These compounds should be eliminated from the water bodies to avoid the unwanted consequences. Techniques like advanced oxidation process with UV/H_2O_2(AOP),sonochemical degradation , microwave enhanced photo catalysis method are employed. Methyl orange dye (M.O) which is commonly used in the laboratory, coloring agents in leather, food, textile, pharmaceutical industries. Methyl orange is not easily degradable, it is mutagenic and carcinogenic (B.Choudhary, A. Goyal et al, 2009)]. In this work , M.O is the target organic pollutant CuO NPs synthesis by using *centella asiatica* leaves extracts at room temperature can be use for the degradation of methyl orange in the absent of reducing agents..

2. Materials and Experiment

2.1 Material

Copper acetate was obtained from NICE CHEMICALS pvt.ltd Cochin 682024, methyl orange was purchased from Merck. *Centella asiatica* plants were collected from local areas of Manipur, whole plant was washed several time with distilled water and dried in a dark room. The obtain chemicals were used without further purification as it is of analytical grade. Throughout the experiment double distilled water was used.

2.2 Preparations

2.2.1 Preparation of leaves extract: 0.1gm of dried leaves and stems of *Centella asiatica* was weight out and to it 10 ml of the double distilled was added, it was stirred

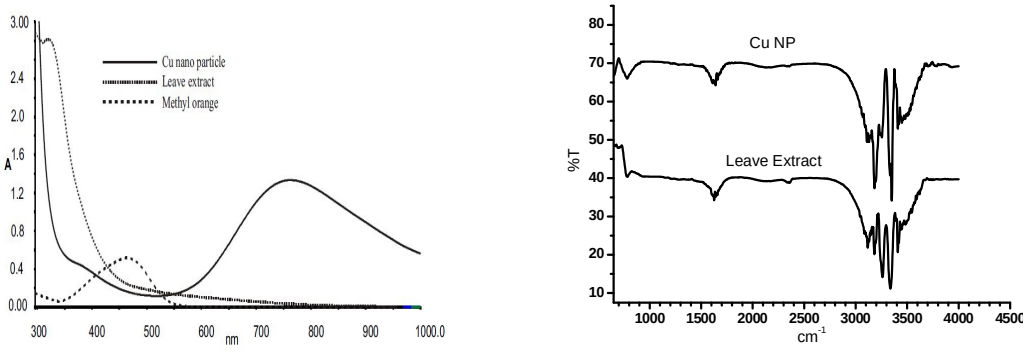
for about 20 mins in a magnetic stirrer at room temperature. After stirring, it was filtered using watman filter paper no.4 and followed by centrifuged to settle down the unwanted solids. Extract was taken up by using a syringe to avoid the solid residues. This very leave extract is used for the preparation of copper nanoparticles.

2.2.2 Environmental benign preparations of copper nanoparticles: 1% (w/v) of copper nanoparticles was prepared by dissolving 1 gm. of Cu $(CH_2COO)_2.H_2O$ was dissolved in 100 ml of double distilled water; it was stirred for 20 mins. To the above solution 10 ml of the *Centella asiatica* leaves extract was added and the mixture was stirred for about 3 hrs. The formation of the particles can be seen within 1 hr. The solution was aged for 12 to 13 hrs. Nanoparticles prepared were centrifuged and washed with double distilled water twice .Nanoparticles was collected, dried at 60^0C in an oven.

2.2.3 Kinetic study of photocatalytic degradation of methyl orange: The degradation of methyl orange in the absence and presence of CuO NPs were studied spectrotometrically by using Perkil Elmer L-35 UV-Vis spectrophotometer determining the decrease in the absorbance at 464nm. To a mixture containing 300µl of M.O (10^{-3}M) and 1ml of CuO NPs was added, distilled water was added to make up to 3ml.The reaction was study spectrophotometrically at room temperature (25^0C). The colour of the reaction mixtures faded, indicating that degradation had occurred. The same procedure was followed for uncatalyzed reactions, in absence of CuO NPs.

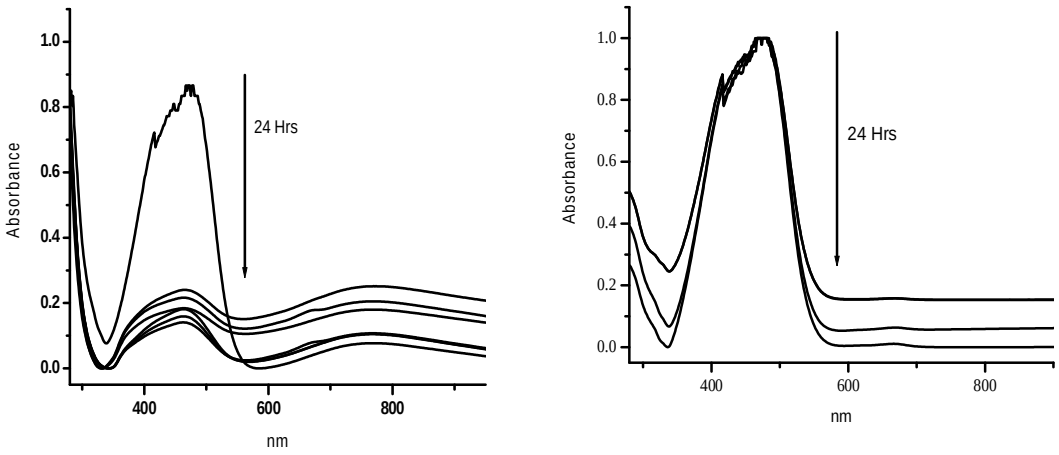
3. Result and Discussion

Fig.1 (a) present the UV-Visible spectra's of copper nanoparticles, methyl orange and *Centella asiatica* (L.) extract. Kinetic study for the degradation of M.O (10^{-3}M) in the present and absence of CuO NPs is shown in fig 2. Photocatalytic degradation of M.O was study using UV in aqueous medium and scanned in the range of 190-1100nm, in the absent of CuO NPs but in present of leaves extract there is no sign of degradation as shown in fig 2(b). In present of CuO NPs degradation of methyl orange can be observed as shown in fig 2(a).



(a) **(b)**

Fig. 1: (a) Comparative UV-Visible spectra of CuNP, leave extract and dye; (b) Comparative IR Spectra of CuNP and leave extract



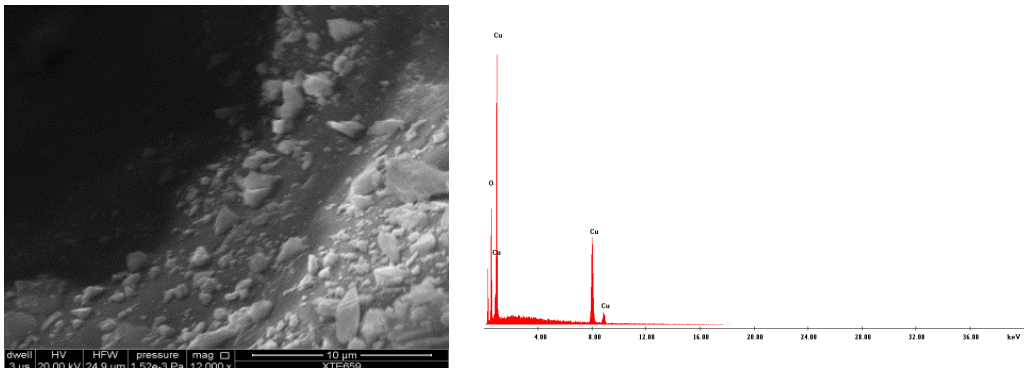
(a) **(b)**

Fig. 2: (a) Degradation of methyl orange in presence of NPs and (b) degradation of methyl orange in absence of methyl orange.

From this result it can be concluded that CuO NPs such prepared can act as catalyst and leaves extract has no role in the degradation of M.O. Mechanism for the degradation of M.O are summarized as below(S.S.Shinde, P.S.Shinde et al,2011).

$$CuO + hv \rightarrow h^+ + e^-$$
$$h^+ + H_2O \rightarrow H^+ + {}^{\cdot}OH$$
$$h^+ + OH^- \rightarrow {}^{\cdot}OH$$
$$e^- + O_2 \rightarrow O_2^{\cdot -}$$
$$O_2^{\cdot -} + \text{organic species} \rightarrow \text{organic species}^{\cdot}OO^-$$
$$OH + \text{organic species} \rightarrow \text{oxidized product}$$

The morphology of as prepared CuO particles was study by SEM as shown in Fig 2 (a). The average diameter of the NPs ranges from 2µm to 5µm.



(a) **(b)**

Fig. 2: (a) SEM image of Cu NPs and (b) EDX result.

Fig 2(b). Shows the EDX result of copper product, it can be concluded that the sample mainly consist of copper. IR spectrum of CuNPs and leaves extract is shown in fig1(b)., The IR bands at 3373 cm^{-1},1638 cm^{-1} and 725 cm^{-1} corresponds to O-H stretching H- bonded alcohols and phenols, carbonyl stretching, N-H bending. It suggest that Cu NPs are surrounded by different organic molecules such as terpenoids, alcohols, ketones, aldehydes etc. which match well with the chemical composition of *C.asiatica* ,it mainly consist of large amount of pentacyclic triterpenoids saponins [15, 16, 17]. Besides these it also contain numerous components like alkaloids, carotenoids , tannin etc[18,19], from the Fig. it can be concluded that copper nanoparticles were coated by leaves extract of *C.asiatica* .

4. Conclusion

Copper nanoparticles synthesized using *Centella asiatica* L. leaves extract is a simple and environmental benign method. This method has many advantages such as economic viability, ease to scale up and less time consuming. Copper nanoparticles such prepared can be used as catalyst for photocatalytic degradation of methyl orange which is an organic pollutant. The experiment revealed that the presence of copper nanoparticles is necessary for the degradation of methyl orange.

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