Green Synthesis of Copper Nanoparticles Using *Ocimum sanctum* L. (Tulsi) and *Piper nigrum* L. (Pepper Seed) for Pollution Free Environment

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**ABSTRACT**

Synthesis of nanoparticles using plant leaf extract is an eco-friendly, non-toxic and cost efficient approach in the field of nanotechnology. In this study, biosynthesis of stable copper nanoparticles were prepared by using *Ocimum sanctum* and *Piper nigrum* extract. These biosynthesized Cu nanoparticles were characterized with the help of X-Ray diffraction (XRD), Scanning Electron Microscopy (SEM) and Fourier transforms infrared spectroscopy (FTIR). The crystalline size of the Copper Nanoparticles prepared from Tulsi extract is little smaller than Copper nanoparticles prepared from Pepper extract. SEM images of Copper nanoparticles showed that the cluster formation, presence of smaller and larger grains. FTIR analysis revealed that the functional groups present in the Copper nanoparticles prepared from both Tulsi and Pepper extracts. Thus, this method can be used for rapid and ecofriendly biosynthesis of stable copper nanoparticles and can be used for catalytic agent, anti-microbial, anti-fungal, anti-biotic, nanosensor, super conducting material, solar cell applications.

**Keywords:** *Ocimum sanctum*, Tulsi, *Piper nigrum*, Pepper, Diffraction, Green synthesis, Copper, Cu, Copper nanoparticles
1. INTRODUCTION

The skillful fabrication of nanoparticles has aspiring nanotechnology into one of today’s most promising and prevalent fields of scientific research [1]. Potential future advancement requires the ability to prepare nanomaterials in a reproducible and controlled manner [2]. Currently, there is an aggregate attention to synthesize metallic copper nanocrystals not only for the expansion of synthetic advancement, but also for the assessment of their electrical, catalytic, sensing and surface properties [3-5]. Copper nanoparticles has been employed as heterogeneous catalysts for numerous environmental progressions, e.g., selective reduction in nitric oxide, oxidation of carbon monoxide and decomposition of nitrogen dioxide [6]. Metallic Cu nanoparticles are striking materials primarily because of their exceptional properties and low cost compared to other metallic nanomaterials such as gold and silver [7].

The ability to prepare nanomaterial with well-defined morphologies and sharp faces should enable the appraisal of their properties [8]. Synthesis of Cu nanoparticles is quiet challenging due to its high tendency for oxidation. It is extremely sensitive to air, and the oxide phases are thermodynamically more stable [9]. The high oxidation rate of Cu nanoparticles may limit their applications [10]. Oxidation of copper nanoparticles can be eliminated if the synthesis is conducted in the presence of CO or H₂. On the other hand, handling these gases is rather cumbersome, and use of such gases is avoided when possible [11].

Copper nanoparticles have been used in various fields, including agricultural, industrial engineering, wound dressings, gas sensors, catalytic process, high temperature superconductors, solar cells and technological fields [12-15]. Although the use of Nano science in agriculture has been predominantly theoretical up to now, effective antibacterial activities exhibited by Copper nanoparticles in agricultural research have increased development in the field of nanotechnology, leading to the establishment of intensively clean, cost-effective and efficient biosynthesis techniques of Copper Nano particles. Generally the Cu nanoparticles are synthesized from vapor deposition, electrochemical reduction, radiolysis reduction, thermal decomposition, chemical reduction of copper metal salt.

These methods involve high temperature, high pressure and hazardous chemicals, and some toxic chemicals absorbed on the surface of nanoparticles may cause adverse medical effects. Green synthesis of nanoparticles has several advantages over chemical and physical synthesis, a method using leaves extract. In addition, the plant-mediated synthesis is a rapid, flexible, and suitable process for large-scale production of nanoparticles. Among nanoparticles copper oxide nanoparticles have been used enormously due to their potent antibacterial activity.

In recent, green synthesis of Cu nanoparticles was achieved by using microorganisms, plant extract, Ocimum sanctum (local name Tulasi) is a traditional medicinal plant of India has a source of bio-reduct on and stabilizers. Recently Ocimum sanctum extracts have been used in the synthesis of silver nanoparticles and gold nanoparticles [16-18]. In the group of medicinal plants, the Piper nigrum possess excellent medicinal properties due to the presence of enormous phytochemicals.

The piperine is an alkaloid, majorly found in Piper nigrum, which belongs to the Piperaceae family that is massively cultivated at India and Sri Lanka [19, 20]. Owing to the presence of large amount of phytochemicals, the leaf and stem of Piper nigrum are taken into account for the synthesis of Copper nanoparticles. In the present study attempts were made to synthesis Copper nanoparticles by using Ocimum sanctum and Piper nigrum extracts.
2. EXPERIMENTAL MATERIALS AND METHODS

2.1. Synthesis of Copper Nanoparticles Using Tulsi Extract

For the preparation of copper nanoparticles copper sulphate (CuSO₄) and tulsi leaf extract were used as precursors. The roughly washed leaves (100 gm) were cut and boiled with 100 ml of ionized water for 15 minutes using a heating mantle at 80 °C. The extract was filtered and stored in refrigerator. 10 ml of those tulsi leaf extract was added to 100 ml of the aqueous CuSO₄·5H₂O solution. Finally flask was then kept overnight at room temperature. The copper nanoparticles solution thus obtained was purified by repeated centrifugation at 12,000 rotations per minute for 15 minutes. Then the obtained Cu nanoparticles are dried using hot air oven at 80 °C for 3 hours.

2.2. Synthesis of Copper Nanoparticles Using Pepper Seed Extract

For the synthesis of copper nanoparticles Copper Sulphate (CuSO₄) and Pepper Seed Extract were used as precursors. For the preparation of copper nanoparticles 1mm CuSO₄·5H₂O solution mixed with 20 ml of seed extract. Another set of same experiment conducted with a supply of microwave irradiation as an alternate energy source. The mixture was incubated for 3 hours at room temperature. The change in color indicating the formation of copper nanoparticles. The settled Copper nanoparticles were filtered and dried using a hot air oven. Figure 1 shows the Copper nanoparticles synthesized through chemical (green) and Microwave irradiation methods (Black).

![Figure 1. Photograph of Copper Nanoparticles.](image)

The XRD pattern of the Copper nanoparticles was analyzed by using a powder X-Ray diffractometer (Schimadzu model: XRD 6000 using CuKα (λ = 0.154 nm) radiation, with a diffraction angle between 0° to 80°. The surface morphology of the nanoparticles was recorded from Scanning Electron Microscopy (SEM). Functional group of synthesized Copper nanoparticles was observed from Fourier Transform Infra-Red spectroscopy (FTIR).
3. RESULTS AND DISCUSSION

3.1. Structural analysis (XRD)

XRD pattern of synthesized Cu nanoparticles using a leaf extract of thulsi is shown in Figure 2. The XRD pattern reveals that a high crystallinity of Cu nanoparticles observed at different diffraction angles of 25.9°, 28.3° and 44.8°, which correspond to the characteristic face centered cubic (FCC) of copper lines indexed at (101), (200) and (220) respectively. The diffraction angle observed at 21.1° is related to the tulsi leaf extract medium. The estimated size of the nanoparticles is about 26.47 nm from Debye-Scherer Equation, which may indicate a high surface area, and surface area to volume ratio of the nanoparticles.

![XRD Pattern of Cu nanoparticles from Tulsi extract](image)

**Figure 2.** XRD Pattern of Cu nanoparticles from Tulsi extract

The Crystallite size of the nanoparticles is calculated from Debye Scherer formula which is written below:

\[ D = \frac{0.9 \lambda}{(\beta \cos \theta)} \]

where: K, known as Scherer’s constant (shape factor), ranges from 0.9 to 1.0, is 1.5418 Å, which is the wavelength of the X-Ray radiation source, \( \beta_{1/2} \) is the width of the XRD peak at half height and \( \theta \) is the Bragg angle. XRD patterns obtained for the Cu NPs synthesized using pepper juice has been shown in Figure 3.
The presence of intense peak corresponds to (101), (200) and (220) indexed a crystalline copper FCC phase which gives an average size of 28.54 nm [21]. A small peak observed at around 25° indicates that a small amount of copper is oxidized and converted into copper oxide.

![XRD Pattern of Cu nanoparticles from Pepper seed extract](image)

**Figure 3.** XRD Pattern of Cu nanoparticles from Pepper seed extract

### 3. 2. Scanning Electron Microscopy (SEM) analysis

The surface morphology of the nanoparticles was taken by Scanning Electron Microscopy (SEM) analysis. Scanning Electron Microscopy gives further insight into the morphology and size details of the copper nanoparticles. The SEM photograph shows that the synthesized Copper nanoparticles are asymmetrical dispersed and aggregated infrequently to form free crystal structures.

The Surface Morphology of synthesized copper nanoparticles from Tulsi extract is shown in Figure 4. From the image it is clear that the existence of unsymmetrical spherical tiny copper nanoparticles settled on extract residue as the sample was prepared by the method of evaporation. However, it was not possible for us to investigate the exact surface texture of the observed nanoparticles. SEM images of copper nanoparticles prepared from Pepper Seed extract are shown in Figure 5.

Copper nanoparticles prepared by this method show nearly monodispersed distribution of particle sizes, with some traces of lumped particles observed forming bulky micron sized aggregates.
Figure 4(a,b,c,d). SEM images of Cu nanoparticles prepared from Tulsi extract.
Figure 5(a,b,c,d). SEM images of Cu nanoparticles prepared from Pepper seed extract.
3.3. FTIR analysis

FTIR spectrum of Copper nanoparticles synthesized from thulsi extract is shown in Figure 6. The broadband around 3200 cm\(^{-1}\) which are characteristics of the hydrogen bonded phenols and alcohols.

![FTIR Spectrum of of Cu nanoparticles prepared from Tulsi extract](image)

Figure 6. FTIR Spectrum of of Cu nanoparticles prepared from Tulsi extract

The band observed around 1610.45 cm\(^{-1}\) are assigned to N-H bending as well as C=O stretching vibration of amino acids. The bands in the region 1100.21- 1600.65 cm\(^{-1}\) were due to the stretching vibrations of various C–O and C–N present in the thulsi leaf extract. Thus, it is clear that the presence of flavones and polysaccharides in the leaf extract and the shifting of the respective bands confirm the binding of copper with the functional groups or its capping to provide further stability [22].

This also confirms that water soluble compounds such as terpenoids, which are present in leaf extract have the ability to perform dual functions of reduction and stabilization of copper nanoparticles.

Figure 7 shows the FTIR spectrum of Cu nanoparticles prepared from pepper extract. The IR spectrum of Cu nanoparticles shows band at 3373 cm\(^{-1}\), 1635 cm\(^{-1}\), 1516 cm\(^{-1}\), 1376 cm\(^{-1}\), 1198 cm\(^{-1}\) corresponds to O-H Stretching H-bonded alcohols and phenols, carbonyl stretching, N-H bend primary amines, corresponds to C-N stretching of the aromatic amino group and C-O is stretching alcohols, ethers respectively. The band at 624 cm\(^{-1}\) is due to acetylenic C=H bending vibrations.

FTIR spectrum of Cu nanoparticles suggested that Cu nanoparticles were surrounded by different organic molecules such as terpenoids, alcohols, ketones, aldehydes and carboxylic acid.
4. CONCLUSIONS

Copper nanoparticles have been synthesized by Green Synthesis method (from Tulasi extract and Pepper seed extract). The XRD analysis confirmed the crystalline size of Copper Nanoparticles.

The calculated crystalline size of Copper nanoparticles prepared from Tulsi extract is found to be 26 nm and for the Copper nanoparticles prepared from Pepper extract is about 28 nm. The crystalline size of the Copper Nanoparticles prepared from Tulsi extract is little smaller than Copper nanoparticles prepared from Pepper extract.

SEM images of Copper nanoparticles showed that the cluster formation presence of smaller and larger grains. FTIR analysis revealed that the functional groups present in the Copper nanoparticles prepared from both Tulsi and Pepper extract.

From the above results showed that the synthesized Copper nanoparticles could have a large number of applications in the field of electrode materials in different rechargeable batteries, catalytic agent, anti-microbial, anti-fungal, anti-biotic, nanosensor, super conducting material, solar cell application.

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