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Green Synthesis and Antibacterial Activities of 11th Group Transition Elements of Copper and Silver Nanoparticles by Waste Peel Extracts of Citrullus Lanatus Fruits

C. Rajalakshmi[1*]
PG & Research Department of Chemistry, Periyar Arts College, Cuddalore, Tamil Nadu, India-607001

Abstract
The present study demonstrates the bioreductive green synthesis of nanosized 11th transition metals of copper(Cu) and silver(Ag) nanoparticles, using waste peel extracts of watermelon (Citrullus lanatus). Transition metals are highly reactive and higher positive charge metal cations, these ions are easily accepted to the electrons from phyto-constituents. The peel extracts of Citrullus lanatus seem to be environmentally friendly, so this protocol could be used for rapid production of nanoparticles of CuNPs and AgNPs. The formation of the plant constituent coated Cu-O, and Ag-O nanoparticles were first monitored using UV–Vis absorption spectroscopy, FTIR Spectroscopy. The average particle sizes are measured by XRD SEM and also screened antibacterial activity
Keywords: *CuNPs, AgNPs, Citrullus lanatus, UV, FTIR, XRD, SEM and Antibacterial activity*

1. INTRODUCTION
Nanotechnology, and alongside nanostructured materials, play an ever increasing role in science, research and development as well as also in every days life, as more and more products based on nanostructured materials are introduced to the market. The advance and very applicable technology is nanotechnology and it was derived from the term of nano it is the billionth of meter or 10^{-9} m. The Nano come ultimately from the Greek word for dwarf, and is also related to the Spanish word Nino [1]. The synthesis of silver nanomaterials or nanoparticles extensively studied by using chemical and physical methods, but the development of reliable technology to produce nanoparticles is an important aspect of nanotechnology. Biological synthesis process provides a wide range of environmentally acceptable methodology, low cost production and minimum time required. At the same time the biologically synthesized silver nanoparticles has many applications includes catalysts in chemical reactions [2]. Microbial source to produce the silver nanoparticles shows the great interest towards the precipitation of nanoparticles due to its metabolic activity. Of course the precipitation of nanoparticles in external environment of a cell, it shows the extracellular activity of organism. Extracellular synthesis of nanoparticles using cell filtrate could be beneficial over intracellular synthesis, the fungi being extremely good candidates for extracellular process and also environmental friendly. There are few reports published in literature on the biosynthesis of silver nanoparticles using fungal as source [3]. The use of bacterial strain in the bio-manufacturing process has the advantage that ease of handling than the fungal sources [4-7]. Plant biotechnology has opened up unexpected new ways for finding new way for trapping their potential resources. Biosynthesis of nanoparticles is an approach of synthesizing nanoparticles using microorganisms and plants having biomedical applications. This approach is an environment-friendly, cost- effective, biocompatible, safe, green approach [8]. Green synthesis includes synthesis through plants, bacteria, fungi, algae etc. They allow large scale production of CuO and AgO NPs free of additional impurities [9]. NPs synthesized from metallic approach show more catalytic activity and limit the use of expensive and toxic chemicals. These natural strains and plant extract secrete some phyto- chemicals that act as both reducing agent and capping or stabilization agent; for example, synthesis of CuO and AgO NPs nanoparticles from cell soluble proteins of waste peel extracts of watermelon (*Citrullus lanatus*).

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2. MATERIAL AND METHODS

2.1 Sample collection

The peel of *citrullus lanatus* (watermelon) were collected from the watermelon road shop in, Chidambaram main road, Cuddalore, Cuddalore district, Tamil Nadu, India

2.2 Reagents

All analytical grade chemical and solvents used in the sample preparation were purchased from local suppliers of Singma-Aldrich chemical company Pondicherry scientific company, Trichy.

2.3. Preparation of *citrullus lanatus* (watermelon) peel extract for synthesis of nanoparticles

The sliced *citrullus lanatus* (watermelon) peels were washed several times with de-ionized water and tried (Fig:1) five days in room conditions, then grained and powdered. A 20 g of powdered peel taken in 250 mL round bottomed flask and stirred with 100 mL de-ionized water, heated at 80oC for three min and filtered by using whatmann 1≠ filter paper to got the extract. The filtrate is used in the further synthesis process.

Figure: 1 Images of watermelon waste peel and dried peel

2.4 Synthesis of nanoparticles

2.4.1 Synthesis of Copper nanoparticles

10 ml of waste peel extract of *citrullus lanatus* (watermelon) was added to 100 ml of 0.1M aqueous solution copper sulphate pentahydrate (M.F:$CuSO_4 \cdot 5H_2O$, M.W:249.68) for the reduction of Cu ions. The mixture was boiled for approximately 10-20 minute at 50°C-70°C. Centrifuged the extract for 15 minutes at 4000 rpm.. The pale blue colour copper sulphate solution is turns to brownish green in colour. This colour change is indicated as the CuNPs generation. The collected supernatant was then double filtered by Whatman filtered paper [10]. The solution was diluted two times of its volume. The obtained product was dried, collects and stored the airtight clicklock tube. Finally, CuNPs were thoroughly characterized.

2.4.1 Synthesis of silver nanoparticles

The synthesis of silver nanoparticles carried out by, 100mL of 0.1M of silver nitrate (M.F: $AgNO_3$, M.W: 169.87g/mol) solution is taken in a 250 ml conical flask. Then, 10 ml of waste peel extract of *citrullus lanatus* (watermelon) was added drop by drop into silver nitrate solution under continuous stirring for 30 min at the room temperature[11], the milky white solution of lead nitrate is turn to pale whitish block in colour. The resultant residue was filtered by using whattman 1≠ filter paper, the precipitate is washed several in double distilled water, finally obtained the glittering crystalline product.. The obtained final product was dried, collects and stored the airtight clicklock tube. These, AgNPs were thoroughly characterized.

3. RESULT AND DISCUSSION

3.1 UV spectral analysis

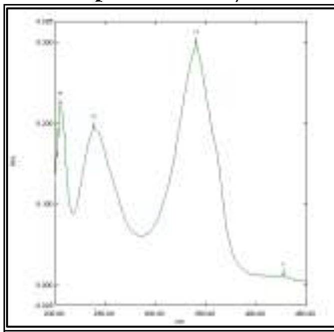

Fig:2a UV Spectrum of CuNPs

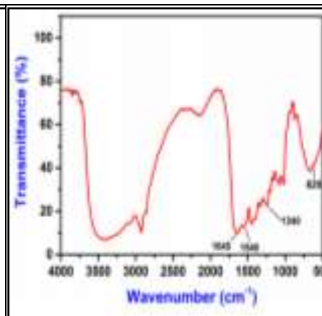

Fig:2b IR Spectrum of CuNPs

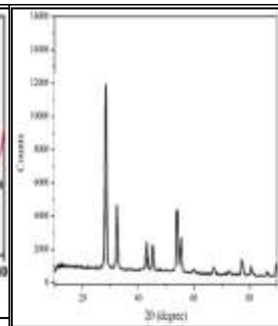

Fig:2c XRD pattern of CuNPs

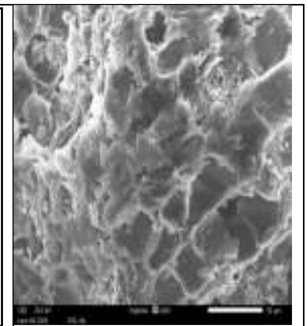

Fig:2d SEM image of CuNPs

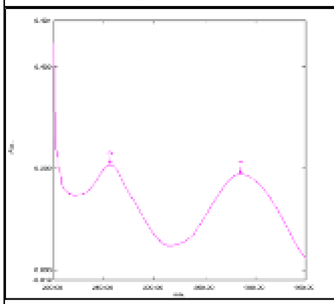

Fig:3a UV Spectrum of AgNPs

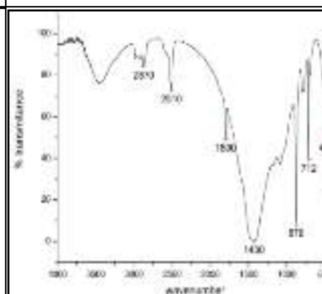

Fig:3b IR Spectrum of AgNPs

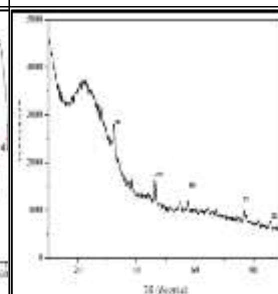

Fig:3c XRD pattern of AgNPs

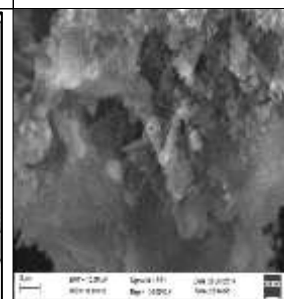

Fig:3 SEM image of AgNPs

UV–vis absorption spectra were recorded for AgNPs and CuNPs in the range of 200–800 nm using an aqueous AgNPs and CuNPs suspension. The samples displayed an optical absorption peaks of absorption maximum value have been shown 383 nm(Fig:3a) for AgNPs. 340nm for CuNPs(Fig:2a). FTIR spectrum used as identification of phytochemical constituents, these are capping to form the nanoparticles of such metal cations. IR spectra peak shows the copper oxide(Fig:2b) nano particle showing a high intensity broad band at 620 cm-1 is give CuO stretching frequency is due to the stretching mode of the copper and oxygen bond. Broad band at 2110 and 1933 cm-1 is due to the stretching mode of the silver and oxygen(Fig:3b) bond. XRD Analysis showed(Fig:2c) four distinct diffraction peaks at 26.24°, 32.26°, 38.53° and 49.04° and can be indexed 2θ values of (111), (211), (222) and (311) crystalline planes of cubic Cu. and size of copper oxide nanoparticles was obtained by Debye–Scherrer's formula to gives 15.4nm.Fig:3c of the AgNPs XRD pattern have shown 38.28°, 43.18°, 65.64° and 78.04° and can be indexed 2θ values of (111), (200), (220), (311) crystalline planes of cubic Ag. The average size of the silver nanoparticles is calculated to give 18.2nm. The SEM image of synthesized Copper nanoparticles is shown in Fig. 2d. From the SEM images were seen in different magnification ranges like 1μm–10 nm which clearly demonstrated the presence of spherical and mountain rock shaped nanoparticles. SEM images were seen in different magnification(Fig:3d) ranges like 1μm–50μm which clearly demonstrated the presence of spherical and mountain rockshaped nanoparticle of AgNPs.

4. ANTIBACTERIAL ACTIVITY

The synthesized CuNPs and AgNPs by waste peel extract of *citrullus lanatus* (watermelon) and standard Ciproflexine against three Gram Positive (*S. aureus, B.subtills and S.Pyogens*) were tested[12] among these test solution. AgNPs have shown good activity compared CuNPs for all the gram positive bacterial starins, **Table:1**. Zone of inhibition values, Figure:4 The plate of antibacterial activity of Gram positive bacterial strains, **Figure:5** and the cluster column chart of antibacterial activity

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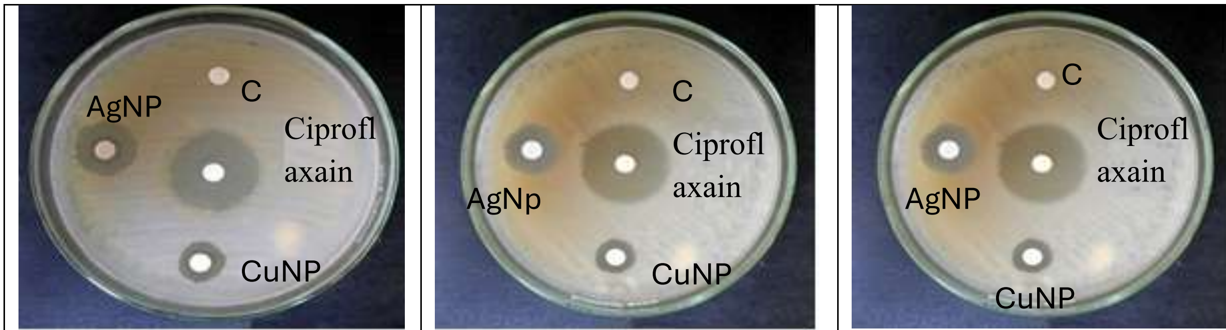

S. aureus	***B.subtils***	***S. pyogenes***

Figure:4 Plate of Antibacterial activities of CuNPs and AgNPs against Gram positive Bacterial strains

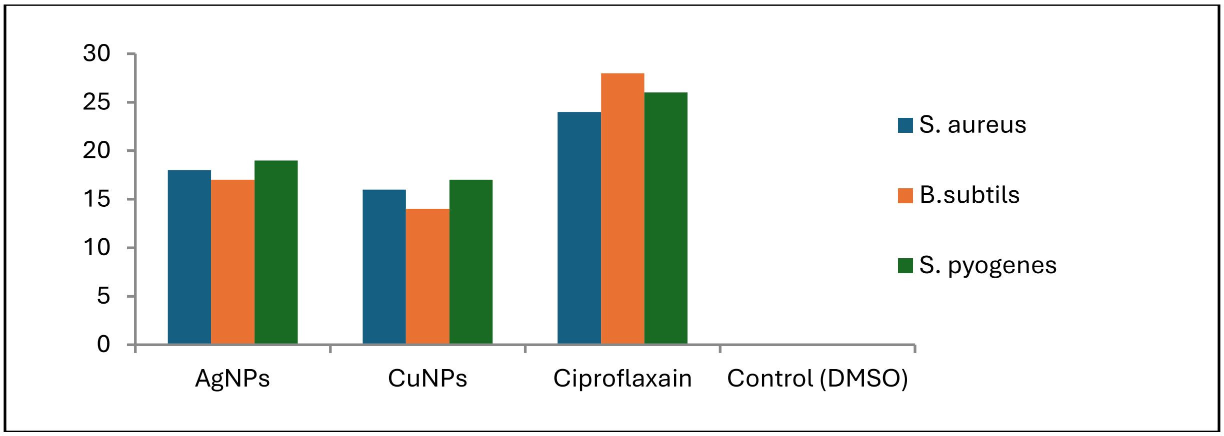

Figure:5 Clustercoloumn chart of antibacterial activities of AgNPs and CuNPs

Table:1 Zone of inhibition values of AgNPs and CuNPs

Compound	Zone of Inhibition (mm)		
	Gram positive Bacteria		
	S. aureus	*B.subtils*	*S. pyogenes*
AgNPs	18	17	19
CuNPs	16	14	17
Ciproflaxain	24	28	26

5. CONCLUSION

The green synthesis of transition metal of copper and silver nanoparticles from nanoparticles from *Citrullus lanatus* (Watermelon) peel extract was studied. Million tons of waste watermelon peels not utilized and thrown in the lands. The antibacterial activates of have shown silver nanoparticles have shown good activity compared to copper nanoparticle especially *S. pyogenes* bacterial strains have shown more active. The average size of the copper nanoparticles has shown1μm–10 nm and silver nanoparticles have shown1μm–50μm.

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