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Research Article

GREEN SYNTHESIS OF SILVER NANOPARTICLES USING MURRAYA KOENIGII LEAF EXTRACT AND ITS CATALYTIC DEGRADATION OF METHYL ORANGE

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ABSTRACT

Here we report on the synthesis of silver nanoparticles using the leaf extract of Murraya koenigii as both the reducing and stabilizing agent. The prepared nanoparticles are characterized by UV-vis, FTIR, XRD and HR-TEM studies. The FTIR spectra are used to detect organic compounds capping AgNPs. The XRD and TEM analyzed clearly show the crystalline nature of the nanoparticles. The silver nanoparticles are found to be almost spherical with an average diameter of 26.8 nm confirmed by TEM images. The efficiency of silver nanoparticles as a promising candidate for the catalysis of organic dyes by methyl orange.

INTRODUCTION

A vital area of investigate in nanotechnology is the fabrication of nano silver particles. The metallic silver is used in the field of medicine, optoelectronics, optics, catalysis; sensors are well known (Nam *et al*., 2009, Narayanan *et al*., 2011, Jinwei *et al*., 2011, Fayza *et al*., 2011). Particularly, the nanoscaled metallic silver is of large research attention due to its high performance and relatively low cost in catalysis for a variety of chemical reactions (Zhang *et al*., 2011, Zhang *et al*., 2012). Further to decrease or remove substances dangerous to human health and the environment the development of green chemical processes and products is appropriate more and more important in the past decade (Poliakoff and Anastas 2001, Poliakoff *et al*., 2002). Also, Silver is a rather cheap metal catalyst as compared to Au. Therefore in this study, we expected to develop a green method for the preparation of Ag nanoparticles used as the catalytic reduction of methyl orange. A number of methods, i.e., physical, chemical and biological methods have been used for the synthesis of silver nanoparticles by several researchers. Presently microbes and plants are being exploited for large-scale synthesis of silver nanoparticles. This processes to avoid the use of toxic and harmful chemicals in their synthesis also known as "Green synthesis". Green synthesis of silver nanoparticles may advantages over the physical, chemical and microbial synthesis methods, as this is cheap, eco-friendly, suitable single-step method. Several researchers reported the plant-mediated green synthesis of silver nanoparticles using extracts of different plant parts such as root, stem, bark, leaf, fruit, bud, and latex as natural resources (Mariselvam *et al*., 2014, Danai-Tambhale *et al*., 2014, Chandran *et al*., 2006, Gnanajobitha *et al*., 2013). Various biomolecules obtained in these extracts contain polysaccharides, polyphenols, aldehydes, ketones, proteins/enzymes, amino acids and caffeine that can reduce metal ion and stabilize the nanoparticles to desired shapes and sizes (Husen and Siddiqi 2014, Khan *et al*., 2015).

In persistence of the efforts to synthesize of silver nanoparticles by green route, here, a rapid green synthesis of silver nanoparticles by reducing the resultant aqueous salt solutions of metal using the leaf extract of curry leaf (Murraya Koenigii) is reported.

Murraya koenigii belongs to family Rutaceae, is an aromatic, pubescent, deciduous shrub or small tree. It is widely distributed in south-east Asia, Australia, and the Pacific islands.

Curry leaf is an important leafy vegetable. The leaves are generally used in Indian cooking for flavoring food product. The curry leaf tree is native to India, Sri Lanka, Bangladesh and the Andaman Islands. The bioactive compound of Murraya koenigii has many functional properties. The Murraya Koenijii contains alkaloids like muconicine, mahanimbine, koenimbine, mahanimbidine, isomahanimbine, Koenen, koenigine, and koenidine which have bioactive functions like anticancer, antidiabetic, antioxidative and antiulcer (Kesari *et al*., 2007, Ningappa *et al*., 2008, Arulselvan and Subramanian 2007, Borse *et al*., 2007).Optical properties of the prepared silver nanoparticles were evaluated by UV-visible (UV-VIS)

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spectroscopy. Crystalline nature and capping of biomolecules of the prepared silver nanoparticles had been done by X-ray diffraction (XRD) and Fourier transformed infrared spectroscopy (FTIR). Morphology of prepared nanoparticles was analyzed by transmission electron microscopy (TEM) was used to investigate the particle size. Furthermore, the catalytic degradation of methyl orange using silver nanoparticles synthesized from Murraya koenigii aqueous leaf extract as bio-reducing and bio-stabilizing agent.

MATERIALS AND METHODS

Silver nitrate and methyl orange are obtained from Sigma-Aldrich Chemicals.

Preparation of leaf extract

Fresh leaves of Murraya koenigii are collected from local market, chidambaram, Cuddalur Dist, Tamil Nadu, India. The fresh leaves are washed several times with running tape water, followed by distilled water. 10 g of leaves added in 100 mL sterile distilled water, 80° C boiled in water bath for 15 min, and after that cooling at room temperature. Finally, this extract was filtered through Whatman No.1 filter paper and stored at 4° C for further synthesis.

Synthesis of Silver nanoparticles

1X 10-3 M aqueous solution of silver nitrate solution was taken in Erlenmeyer flask and different quantity of 5, 10 and 15 mL Murraya koenigii leaf extract was added to 100 mL of 1X10-3 M silver nitrate solution for bio-reduction process at room temperature. After 30 min the solution were turned from yellow to dark brown indicate the formation of Ag NPs.

Synthesized AgNPs on the reduction of methyl orange

The catalytic activity of synthesized AgNPs, two reactions are carried out in a 3 ml standard quartz cuvette (path length 1 cm) and absorbance values are monitored using UV-visible spectrophotometer. In the first reaction, 1 ml of methyl orange (2×10^{-3} M) is mixed with 2 ml of water, this reaction was monitored after 30 min (I). In second reaction, 2 ml of methyl orange (2×10^{-3} M) is mixed with 1 ml of synthesized AgNPs and this reaction is monitored at three different time intervals viz., 30 min, 45 min and 60 min (II). In all the reactions total volume of the mixture is made up to 3 ml. The values of absorption maxima (λmax) are compared, with that of methyl orange.

Characterization of silver nanoparticles

The UV-vis spectroscopy measurements were recorded on a JASCO dual beam spectrophotometer (Model SHIMADZU UV-1650) operated at a resolution of 2 nm. Photoluminescence (PL) spectra were recorded using Perkin Elmer LS 55 fluorescence spectrometer. Fourier Transform Infrared Spectrometer spectra were recorded under identical conditions in the 4000-400 cm^{-1} region using Fourier Transform Infrared Spectrometer (spectrum RX-I, FT-IR system, Perkineliner Model). The phytoreduced silver colloidal solution was drop-coated onto a glass substrate; and the XRD measurements were carried out using a powder diffractometer (PANalytical X'per PRO model X-Ray diffractometer), on the instrument operating at a voltage of 50 kV and a current of 30 mA. Morphological characterization of the samples was carried out using FE-SEM (JEOL JSM 6701-F). A pinch of dried sample was coated on a carbon tape. It was again coated with gold in an auto fine coater and then the material was subjected to analysis. The films on the grids were allowed to dry prior to TEM measurement in a TECHNAI10-Philphs instrument.

RESULTS AND DISCUSSION

X-ray diffraction analysis

The X-ray diffraction study was conducted to collect information about the crystalline nature of the synthesized nanoparticles. Fig. 1 shows the XRD patterns of the silver nanoparticles obtained from different concentrations.

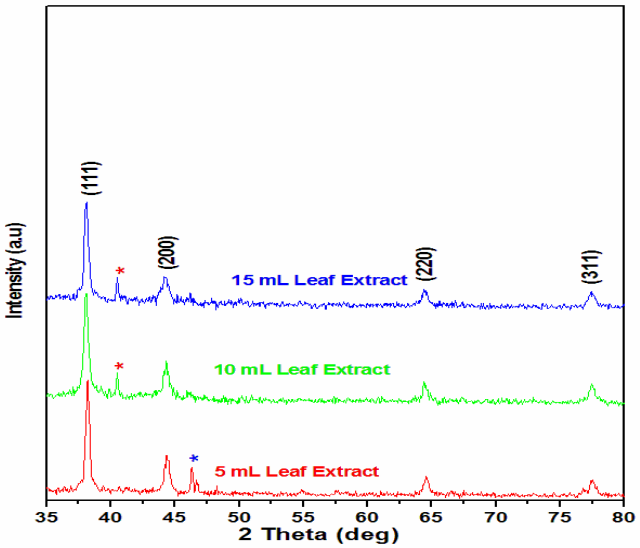

Fig 1 XRD pattern of silver nanoparticles using Murraya koenigii leaf extract.(* show unassigned Peaks).

The four diffraction peaks appeared at positions 38.21°, 44.30°, 64.52°, and 77.47° represent to the 111, 200, 220 and 311 lattice planes of face-centered cubic crystal structure confirms the crystalline nature [JCPDS No. 04-0783]. A similar pattern of XRD for silver nanoparticles has been reported (Bar *et al*., 2009, Mukundan *et al*., 2015). Also, three unassigned peaks in XRD pattern of silver nanoparticles appeared all the concentrations, which may be related to the phytochemical compounds in the leaf extract as a capping and stabilizing agent (Suvith and Philip 2014, Kharat and Mendhulkar 2016). The average crystal size of silver nanoparticles was calculated by Debye-Scherrer's formula,

$$D = K\lambda/\beta cos\theta$$

Where, D - the crystal size, λ - the wavelength of the X-ray radiation (λ=0.15406 nm) for CuKα, K - usually taken as 0.89, β- the line width at half-maximum height. The Scherrer formula was used to calculate the particle sizes in the range of 23–29 nm.

UV-Vis analysis

Fig. 2 shows the synthesis of silver nanoparticles using Murraya koenigii leaf extract was monitored by using UV-Vis spectroscopy. The Murraya koenigii leaf extract was added to the $AgNO_3$ solution, resulting in reduction and formation of silver nanoparticles and visual color change from light yellow to reddish brown. The brown color confirms the reduction of Ag+ to Ag0 and the formation of AgNPs. The change in color was described for by Surface Plasmon Resonance (SPR) excitation in the collective oscillation of free conduction electrons forced by an interacting electromagnetic field.

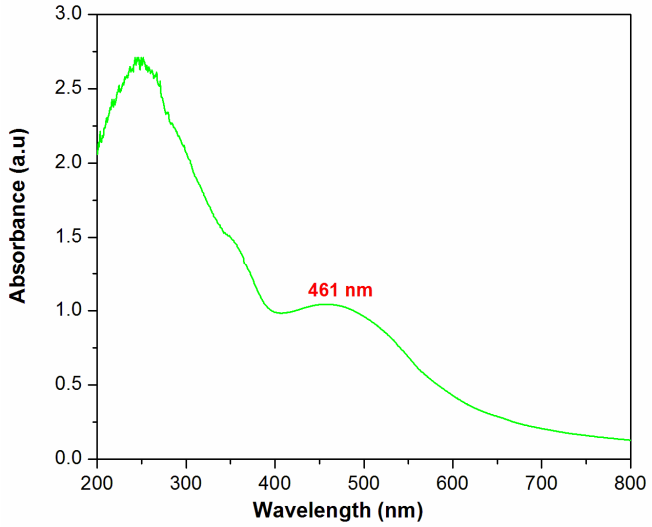

Fig 2 UV-Vis spectrum of silver nanoparticles at 15 ml Murraya koenigii leaf extract

The characteristics SPR peak for AgNPs was monitored by observing the color change and maximum absorbance in the range of 400–480 nm that is confirmation of the incidence of the AgNPs SPR (Bhakya *et al*., 2015, Ramalingam *et al*., 2014). The position and shape of the SPR peak depend on the size, shape and dielectric constant of solutions (Mata *et al*., 2015). The absorbance maxima peak of the AgNPs was observed at 461 nm that confirmed the formation of AgNPs using 15 ml Murraya koenigii leaf extract.

FT-IR analysis

The FT-IR spectra suggest the information about chemical changes in the biomolecules involved in the bioreduction. The spectrum of AgNP (Fig. 3) shows a broad peak at 3362 cm^{-1} which is due to the stretching vibrations of hydroxyl (-OH) group.

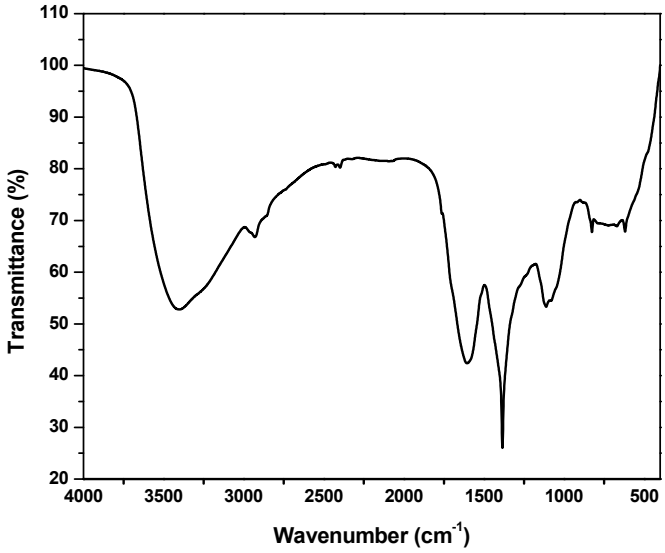

Fig 3 FT-IR spectrum of silver nanoparticles at 15 ml Murraya koenigii leaf extract

A weak absorption at 2924 cm^{-1} could be assigned to aliphatic C-H stretching vibrations. The moderately strong band at 1606 cm^{-1} could be assigned to C=C stretching vibrations of an aromatic ring. Moreover, the peak at 827 cm^{-1} is characteristic of an aromatic ring. The other peak at 1385 cm^{-1} could be attributed C-N stretching of amine respectively (Stuart 2004). It is clear from the above vibration bands appear from various alkaloids, flavonoids, and further phytochemicals which are richly present in the leaf extract of Murraya koenigii and they are responsible for the reduction and stabilization of silver nanoparticles.

SEM-EDX analysis

The SEM micrograph (Fig. 4a) of synthesized nanoparticles was rough, spherical structures with size range of 23-29 nm. The rough morphology of the NPs provides excellent catalytic activity for the synthesized nanoparticles.

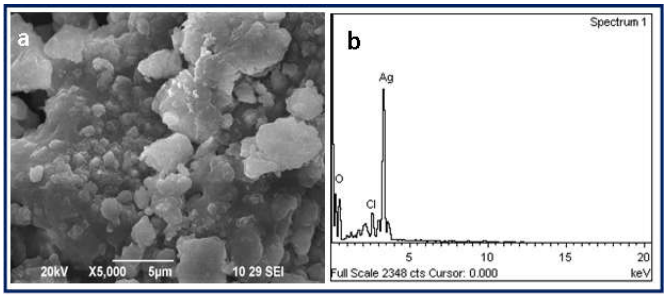

Fig4 (a) SEM and (b) EDX spectrum of silver nanoparticles at 15 ml Murraya koenigii leaf extract

The results of EDX analysis are shown in Fig. 4b. Elemental silver can be seen in the graph presented by the EDX analysis in support of XRD results, which indicated the reduction of silver ions to elemental silver.

TEM analysis

The size and morphology of the synthesized silver nanoparticles were performed by Transmission electron microscopy (TEM) analysis. Fig. (5a-c). shows the TEM images of the different magnification of silver nanoparticles synthesized using Murraya koenigii leaf extract. The images suggest that the nanoparticles are almost spherical in shape. The average particle size measured from the TEM images is to be 23-29 nm which is in good agreement with the particle size calculated from XRD analysis.

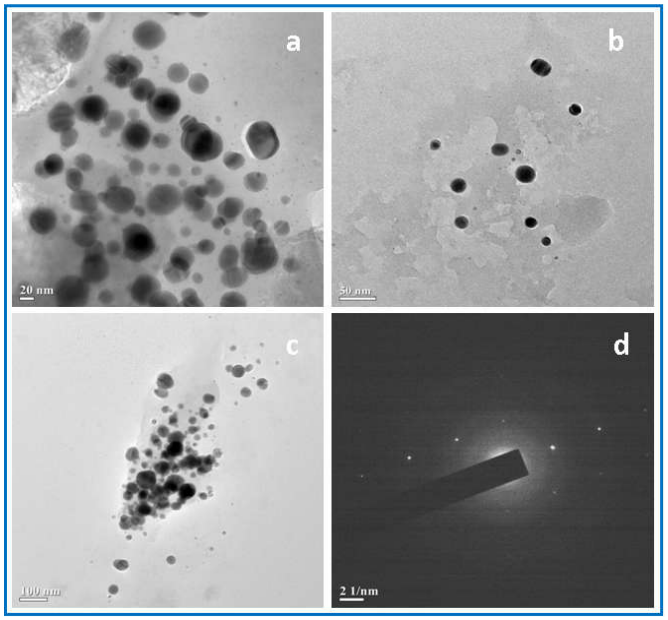

Fig. 5 TEM Images of silver nanoparticles at 15 ml Murraya koenigii leaf extract (a-c) under different magnifications and (d) SEAD Pattern

The selected-area electron diffraction (SAED) patterns showed in Fig. 5(d) demonstrate concentric rings with irregular bright dots, indicating that these nanoparticles are highly crystalline

in nature. This distance is in good agreement with the lattice spacing of (111) planes of face centered cubic (fcc) silver.

Photocatalytic analysis

Photocatalytic activity of silver nanoparticles was investigated by selecting the photocatalytic degradation of methyl orange. Evaluation of absorption peak at 464 nm of methyl orange was used for checking the catalytic degradation process. The absorption spectra of an aqueous solution of methyl orange tested in the presence of silver nanoparticles at different time intervals (Figure 6). The absorption peak decreased gradually with the addition of the exposure time, which indicates that photocatalytic degradation of methyl orange. In the absence of nanoparticles (control), the reaction did not have any improvement. The photocatalytic activity is dependent on the crystallographic structure, morphology, and size of the particles (Rajeswari *et al*., 2012, Kamat 1993, Balazs *et al*., 2008).

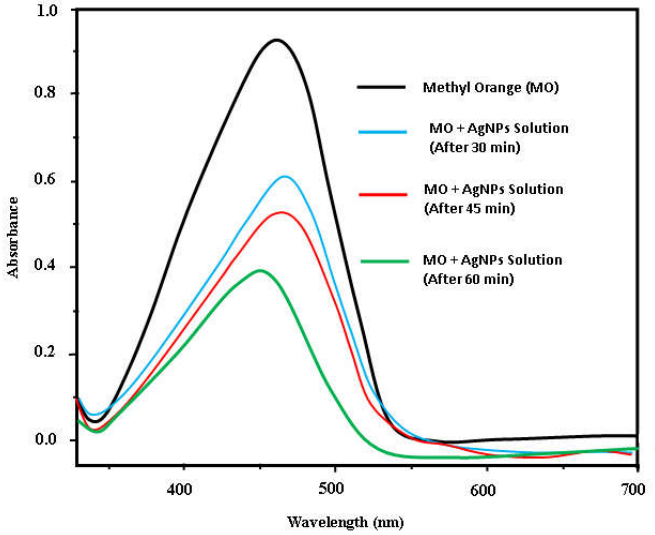

Fig 6 UV–visible spectra of methyl orange reduction by Murraya koenigii in the presence of AgNPs

CONCLUSION

In this study, we have synthesized silver nanoparticles in an aqueous medium by a novel green method using the leaf extract of Murraya koenigii as both the reducing and capping agent. This is a very easy, fast, economical and eco-friendly method for the large-scale fabrication of silver nanoparticles. The nanoparticles are stable in an aqueous medium for a long time without any aggregation. The synthesized silver nanoparticles are characterized by UV-VIS, TEM, XRD and FTIR techniques. Finally, the nanoparticles are proved to be remarkable size-dependent catalytic activity of organic dyes, methyl orange.

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