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ORIGINAL ARTICLE

Green synthesis of nanostructured silver particles and their catalytic application in dye degradation

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Abstract Today, discharge of hazardous dyes from textile industries in water bodies like lakes, rivers and groundwater has become a serious problem, which contributes to increase their pollution levels significantly. These pollutants are difficult to remove by traditional water treatment procedures. Thus, there is a need to develop more suitable methods of effluent treatment. Here, we describe use of green-synthesized nanostructured silver particles in degradation of hazardous dyes like Safranine O, Methyl red, Methyl orange and Methylene blue etc. The silver nanoparticles (AgNPs) used as nanocatalysts were synthesized using *Zanthoxylum armatum* leaves. The reduction of silver ions and the formation of AgNPs have been assessed by UV–Vis spectroscopy. DLS, SEM–EDX, TEM, SAED and XRD studies revealed that the AgNPs were crystalline in nature with size range from 15 to 50 nm. The report emphasizes that the AgNPs are observed to be an excellent catalyst on reduction of hazardous dyes, which is confirmed by a decrease in absorbance maximum values.

1. Introduction

Recent advances in the field of nanotechnology and greater applications of nanoparticles have led to learn about the unexploited resources that already exist in nature for development of new methods in synthesis of nanoparticles. Generally, nanoparticles can be readily produced using physical and chemical methods [1]. However, these methods cannot avoid the generation of toxic byproducts in the synthesis protocol. Instead of using physical and chemical methods for synthesis of metal nanoparticles, the use of biological resources available in nature including microorganisms [12] and plants [11] has received considerable attention for efficient and rapid synthesis of metal nanoparticles [22]. All the green methods are expected to minimize things that contribute to environmental problems and extremely offering a cost effective, eco-benign and energy efficient green alternative.

Foregoing facts revealed that the synthesis of metal nanoparticles by green chemistry approach is an exciting possibility that is relatively underexploited, which aroused our interest in the present investigation. The focus of the present work is to apply green chemistry approaches in the synthesis of silver nanoparticles (AgNPs) using aqueous leaf extracts of *Zanthoxylum armatum* as reducing and capping agents. *Z. armatum* is a perennial shrub belonging to the family Rutaceae

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and commonly known as toothache tree. The bark, fruits, seeds and leaves are extensively used in indigenous system of medicine as a carminative, stomachic, anthelmintic and in the treatment of fever, indigestion and abdominal pain [4].

Dyes are a major class of synthetic organic compounds used in variety of applications [10]. One of the applications of dyes is in textile industries which consumes about ~60% of total dye production for coloration of various fabrics. Moreover after the completion of their use nearly 15% of dyes are wasted. These dye compounds dissolve in water bodies with a concentration in between 10 and 200 milligram per liter results in significant water pollution worldwide [9,7,17]. Therefore, treatment of dye effluents from textile industries is a mandatory part of waste water treatment. The release of dye effluents in aquatic systems is major environmental concern because coloration not only decreases sunlight penetration and dissolved oxygen in water bodies, but also releases toxic compounds during chemical or biological reaction pathway that effects aquatic flora and fauna [17]. Reduction of these dye compounds using physical–chemical and biological processes is generally ineffective, time consuming and methodologically demanding in a high effluent concentration [24]. Among such process, reductive degradation of hazardous dyes with metal nanomaterials is a convenient degradation process because of their unique physiochemical and electronic properties which are not present in bulk materials [5,2]. Metal nanomaterials are versatile materials that can be used in applications such as environmental remediation, medical technology, energy, water treatment, and personal care products [2,14]. Gold and silver nanoparticles can act as good catalysts and hence catalyze many reduction reactions. The rate constant of the catalytic reaction is found to depend on the size of the nanoparticles. The silver nanoparticles synthesized here are used as such to investigate its size dependant catalytic potential in the reduction of hazardous dyes.

2. Material and methods

2.1. Plant and chemicals

Fresh *Z. armatum* leaves were collected from Govind Ballabh Pant Engineering College campus and the sample was authenticated with the help of plant taxonomist of Forest Research Institute, Dehradun, Uttarakhand, India. Silver nitrate (99.99%), Safranine O, Methyl red, Methyl orange and Methylene blue were purchased from Sigma–Aldrich, Delhi.

2.2. Aqueous Z. armatum leaves extract preparation

The fresh *Z. armatum* leaves extract was prepared by taking 20 g of thoroughly washed finely cut leaves along with 100 ml of Milli-Q water in a 250 ml Erlenmeyer flask. The cut leaves were then boiled at 40 °C for 15 min. The solution was filtered through Whatman filter paper No. 1 and stored at 4 °C for further experiment.

2.3. Phytosynthesis of silver nanoparticles

AgNPs synthesis was carried out by taking 5% of leaf extract and adding 95% of 10^{-3} M aqueous $AgNO_3$ solution at 40 °C. The change in color from colorless to brownish indicated the formation of silver nanoparticles.

2.4. Characterization of silver nanoparticles

Ultraviolet–visible spectral analysis was performed for all samples and the absorption maxima were recorded at a wavelength of 300–700 nm using spectrophotometer UV–Vis 3000+ LABINDIA. Fourier Transform Infrared Spectroscopy (FTIR) was used to detect the possible functional groups of biomolecules present in the plant extract. The surface morphology of the green synthesized AgNPs was examined using scanning electron microscopy (SEM) on NOVA-450 instrument, whereas shape, size, crystallinity and chemical composition were examined by transmission electron microscopy (TEM), selected area electron diffraction (SAED), X-ray diffraction (XRD) and energy dispersive spectroscopy (EDS) measurements on Tecnai G^2 20 S-TWIN instrument. The particle size distribution and surface charge of AgNPs were determined using particle size analyzer (Zetasizer nano ZS, Malvern Instruments Ltd., U.K.) at 25 °C with 90° detection angle.

2.5. Catalytic experiments

The catalytic activity of green synthesized AgNPs was demonstrated by degrading hazardous dyes, Safranine O, Methyl red, Methyl orange and Methylene blue. In general, 10 mg of each dye was added to 1 L of distilled water and used as stock solution. After that, 1 mg of green synthesized AgNPs was added to 10 ml of each dye solution and mixed ultrasonically for 15 min. Thereafter 3 ml of each mixed solution was used to evaluate the catalytic degradation of dyes.

The progress of reactions was monitored using UV–visible spectrophotometer by measuring absorbance maxima of dyes at different time intervals viz.,0.5 h, 1 h, 2 h, 3 h, 5 h, 8 h 12 h 18 h and 24 h. A control set was maintained without AgNPs for each dye and measured for absorbance.

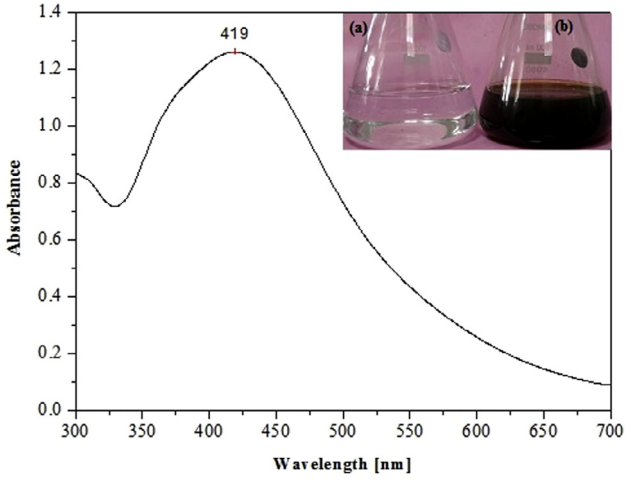

Figure 1 UV–visible spectra of synthesized silver nanoparticles. Insert: 1 mM $AgNO_3$ solution (a) without plant extract and (b) with plant extract.