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Biologically synthesized PbS nanoparticles for the detection of arsenic in water

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ABSTRACT

Semiconductor nanoparticles have gained importance because of their interesting optical properties. Among these, lead sulfide (PbS) has been extensively studied due to its potential technological applications in field effect transistors, solar cells, photo-voltaics, light emitting diodes, photocatalysis, photoluminescence, infrared photodetectors, environmental and biological sensors. Hence there is a need to explore cost effective and eco-friendly biological routes for their synthesis. In this paper, biosynthesis of PbS nanoparticles were carried out using endophytic fungi, subsequently detailed characterization was also performed using UV–visible, fluorescence spectrometer, FTIR, SEM, TEM, EDX and XRD. TEM revealed the formation of PbS nanoparticles in typical size range of 35–100 nm. The application of these nanoparticles for detection of arsenic in aqueous solution through their absorbance properties was also dealt. Importantly, the results were demonstrated for detection of 50 ppb As (III) in water without any interference of other selected ions maintained upto 20 ppb under same conditions. Further, the correlation for the bio-sensitivity of PbS nanoparticles based on the quenching effect with arsenic concentrations ranging between 10 and 100 ppb in water samples was deduced.

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1. Introduction

The threat of arsenic pollution in drinking water is a serious environmental and health concern because of its toxicity on human beings and on other living organisms (Dominguez-Gonzalez et al., 2014). Compounds of arsenic have a variety of applications in agriculture as pesticides, alloying agents and manufacture of semiconductors, lasers, transistors, metal adhesives, explosives and pharmaceutical products (Shrivas et al., 2015). Arsenic enters human body either by drinking of contaminated water or by the consumption of plants commonly grown in the contaminated area (Song et al., 2016). It has been reported that in India, ground waters of West Bengal and Chhattisgarh states are most commonly contaminated with arsenic that exceeded the World Health Organization (WHO) guidelines for drinking water (10 ppb) (Muniyandi et al., 2011; Shahlaei and Pourhossein, 2014; Shrivas et al., 2015). In natural waters, arsenic exists primarily as inorganic forms in two predominant oxidation states: pentavalent arsenic (Arsenate or As (V)), predominating in oxygenated waters and trivalent arsenic (Arsenite or As (III)), prevailing in more reductive environments. Compared with As (V), As (III) is 60 times more toxic to human and has higher mobility in the environment. In groundwater, Arsenic is mostly present as As (V) but can be reduced to As (III) under anaerobic conditions (Boxi and Paria, 2015; Butwong et al., 2012; Divsar et al., 2015; Hagiwara et al., 2015; Luong et al., 2014; Morillo et al., 2015; Reddy et al., 2013; Yogarajah and Tsai, 2015). The resurgence of arsenic is a serious threat to environment. Inorganic arsenic compounds are classified by the International Agency for Research on Cancer (IARC) in Group 1 (carcinogenic to humans), on the basis of sufficient evidence for carcinogenicity in humans and limited evidence for carcinogenicity in animals (Dominguez-Gonzalez et al., 2014). In order to protect our environment and ensure our health, it is imperative to develop a fast, low-cost and sensitive As (III) detection method that is applicable to water environment. Traditional methods for quantitative detection of heavy metal include Atomic Fluorescence Spectrometry (AFS), Atomic Absorption Spectrometry (AAS), Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES), Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and High Performance Liquid Chromatography (HPLC),etc. (Forzani et al., 2007; Ghosh and Luwang, 2015; Hao et al., 2015; Huang and Chen, 2013). Although these methods can accurately measure arsenic in an environmental sample to microgram arsenic per liter concentrations, there is still a necessity for development of simple and rapid methods for field assays. This is due to the fact that, sophisticated analytical techniques need sample preparation procedure before

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the instrumental measurement, which is found to be tedious, time consuming and even more amount of reagents are required (Boxi and Paria, 2015; Shrivas et al., 2015).

Colloidal semiconducting nanomaterials have attracted extensive interest as active building blocks for low-cost solution-processed photovoltaics due to their size-tunable absorption from the visible to the near infrared. The remarkable optical perspectives of nanoparticles have accredited them as ideal nanomaterials for ultrasensitive optical sensing for the selective detection of pesticides, organic compounds, biomolecules and a multitude of metals (Frasco and Chaniotakis, 2009). Advances in colloidal synthesis over the last two decades have enabled the production of high quality, II-VI and IV-VI (such as CdSe, CdS, PbSe and PbS) nanoparticles of various sizes and morphologies. Of these nanomaterial systems, the lead chalcogenide of PbS which has small direct-band gap (0.41 eV) and large excitonic bohr radius of 18 nm, are particularly used as sensors in ammonia gas sensing (Hassan et al., 2013), detection of mercury (II) (LiDong et al., 2014), identification of cauliflower mosaic virus (Sun et al., 2008). However physical and chemical methods used for their synthesis suffer from limitations such as toxicity and bio-incompatibility, instability, uncontrolled crystal growth and aggregation of the nanoparticles (Bai et al., 2006; Hudlikar et al., 2012). Exploring cost effective biological routes for their synthesis holds importance in this scenario. According to literature PbS nanoparticles were synthesized by several biological routes namely *Torulopsis* sp. (Kowshik et al., 2002), *Desulfotomaculum* sp. (Gong et al., 2007), *Rhodosporidium diobovatum* (Seshadri et al., 2011), *Aspergillus* sp. (Kaur et al., 2014).

Lead and sulphur tolerance fungus (*Aspergillus flavus*) which have been isolated from endophytic fungi in our previous studies have been used for the biosynthesis of PbS nanoparticles under conditions akin to room temperature (Uddandarao and Balakrishnan, 2016). Nanoparticles synthesized from the fungal sources contains thiol group of its cysteine residues which have the capacity to bind to metal ions (Jacob et al., 2016). Since endophytic fungi which have been investigated to be a rich source of novel biological active secondary metabolites (Devi and Joshi, 2015; Fouda et al., 2015) such as novel antibiotics, antibacterial and anticancer compounds it has been used for the study. The principal factors that affect sensitivity and selectivity of these sensors are size and concentrations of PbS nanoparticles, the nature of the molecule for the recognition of the element, pH, ionic strength, concentration of nanoparticles and interferences (Shrivas et al., 2015). The novelty of the present work is *Aspergillus flavus* which have been isolated from medicinal plant *Nothapodytes foetida,* have been used for the biosynthesis of PbS nanoparticles for arsenic detection. To the best of our knowledge this is the first report on arsenic detection using biological PbS nanoparticles.

The aim of this paper is to develop a simple and inexpensive method for the arsenic determination in water samples at low levels using PbS nanoparticles and UV–visible spectroscopy as a detection technique.

2. Materials and methods

2.1. Tolerance studies, biosynthesis and characterization of PbS nanoparticles

Aspergillus flavus, an endophytic fungus isolated from a medicinal plant *Nothapodytes foetida*, was employed in this study (Uddandarao and Balakrishnan, 2016). The fungal cultures were maintained on potato dextrose agar plate at 4 °C and sub-cultured. Lead and sulphide tolerant *Aspergillus flavus* was subjected to biosynthesis for synthesis of PbS nanoparticles.

The concentration of lead acetate and sodium sulfide for the synthesis process is chosen based on the tolerance limit of the fungus. *Aspergillus flavus* was subjected to broth studies in potato dextrose broth medium containing lead acetate and sodium sulphide salts in a concentration range of 0–5 mM and 0–0.3% respectively (Mala and Rose, 2014). Control containing potato dextrose broth with fungal strain, but without lead acetate and sodium sulphide was used as a positive control. After 5 days of growth, the biomass is filtered using Whatman filter no. 42. The harvested fungal biomass was rinsed with double distilled water 3–4 times and dried in hot air oven at 80 °C. The dry weight of the biomass was measured and was compared with the control.

For fabrication of PbS nanoparticles, fungal strain was inoculated in potato dextrose broth and incubated at 30 °C for 72 h on a rotary shaker at 115 rpm. Thereafter, 0.5 mM lead acetate and 0.05% sodium sulfide were added to the culture, which was further incubated under the same conditions (Gong et al., 2007). After 5 days of incubation, color change was observed and filtrate was recovered from the cultures. Controls containing potato dextrose broth, with fungal strain but without lead acetate and sodium sulphide as positive control and with lead acetate and sodium sulphide but without cell biomass as negative control, were also run simultaneously along with the experimental flasks in three replicates. Further, the synthesized product was characterized using a UV–visible spectrophotometer, Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), X-ray Diffraction

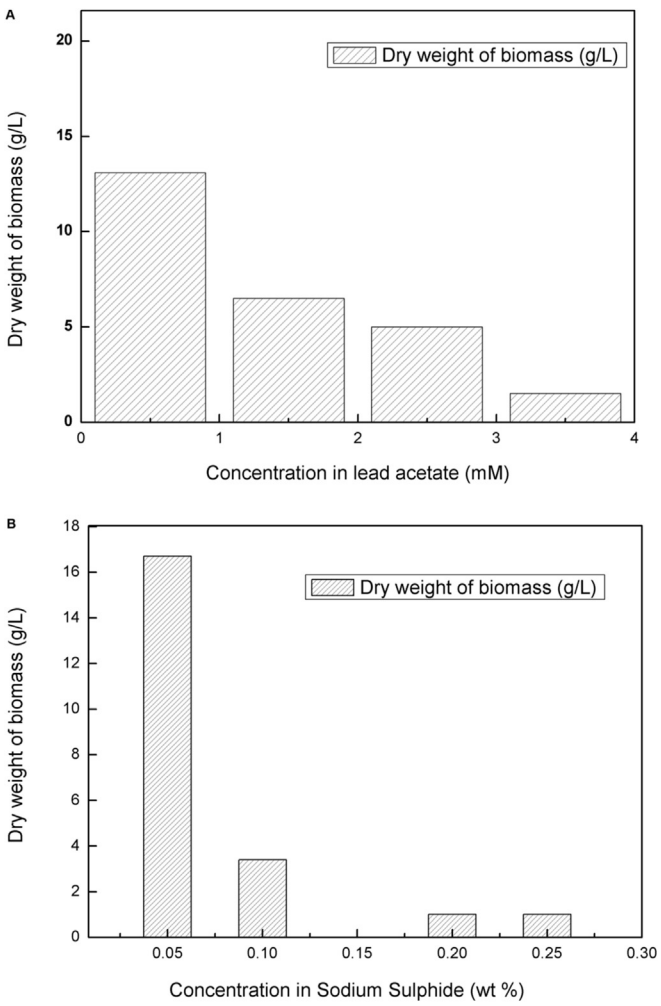

Fig. 1. (a) Lead acetate tolerance range of endophytic fungus *Aspergillus flavus*; (b) Sodium sulphide tolerance range of endophytic fungus *Aspergillus flavus.*

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