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ZnO particles assisted removal of Arsenic [As(V)]

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Abstract

The problem of arsenic content in drinking water is one of the oldest issues and yet scientists and researchers are struggling to find affordable and fast methods to remove arsenic from drinking water. The ill effect of arsenic to human health is very serious and in this paper effort is being made to remove arsenic from water by following the method of adsorption assisted by ZnO nanoparticles. Hydrothermal method is used to synthesize the nano particles and different characterization techniques used to confirm the formation. The optimal values for parameter like pH value, temperature and contact time were found and the same were used to get the maximum adsorption at minimum time to remove the arsenic from drinking water.

Keywords: Arsenic, ZnO, nanoparticles, Adsorption, water purification

Resumen

El problema del contenido de arsénico en el agua potable es uno de los más antiguos y, sin embargo, los científicos e investigadores luchan por encontrar métodos asequibles y rápidos para eliminar el arsénico del agua potable. El efecto nocivo del arsénico en la salud humana es muy grave y en este trabajo se pretende eliminar el arsénico del agua siguiendo el método de adsorción asistido por nanopartículas de ZnO. Se utiliza el método hidrotermal para sintetizar las nanopartículas y diferentes técnicas de caracterización para confirmar la formación. Se encontraron los valores óptimos para parámetros como el valor de pH, la temperatura y el tiempo de contacto y los mismos se usaron para obtener la máxima adsorción en el tiempo mínimo para eliminar el arsénico del agua potable.

Palabras clave: Arsénico, ZnO, nanopartículas, Adsorción, purificación de agua

INTRODUCTION

Entire world is facing the problem of safe drinking water, especially the weaker section of the society who can afford expensive filtration processes, (Bailey et al. 1999). Due to industrialization and urbanization the quality of ground water is getting degraded day by day. Heavy metal contamination of water has become a concern to the society to researchers to develop affordable method to deal with the problem. Arsenic (As) is one of the most harmful metal contaminants found in ground water. As can be formed from the weathering of rocks, resulting in high concentrations of As in groundwater, posing health risks to people consuming water that contains As. As is considered to be a carcinogenic chemical. Controlling its concentration in the drinking water is of utmost importance for all human beings, (Helmer and Hespanhol 1997). Many methods and processes are developed to filter the water to make it suitable for drinking. Some of the conventional methods include oxidation, coagulation-flocculation, and membrane techniques. Adsorption is also another method which is discussed and used by many researchers using different adsorbents, (Sugunan and Dutta 2008). Nanomaterial as adsorbents has attracted the scientists because of the physical properties which exhibits large surface by volume ratio which makes them very suitable candidates as adsorpbents, (Pitoniak et al. 2005). ZnO nano particles are one of the most discussed form of nano material which have a vast horizon of applications including water remediation, (Baruah et al. 2021).

Experimental

Material used: The following chemicals were used as purchased: Zinc acetate di-hydrate [$(CH_3COO)_2Zn.2H_2O$], zinc nitrate hexahydrate ($Zn(NO_3)_2.6H_2O$), sodium sulphide (Na_2S), Sodium hydroxide (NaOH), hexamethylenetetramine ($C_6H_{12}N_4$), ethanol (C_2H_5OH), sodium Arsenite ($NaAsO_2$) and nitric acid (HNO_3).

Synthesis: Synthesis of ZnO nanoparticle: A 4 mM solution of $(CH_3COO)_2 Zn.2H_2O$ was prepared in 40ml 98% ethanol under rigorous stirring at 50°C. The prepared solution was cooled to room temperature and then to the cooled solution 20 ml of 2 mM sodium hydroxide was added under mild stirring. The solution was then kept for two hours in a hot water bath at 70°C. It was then allowed to cool naturally to room temperature, (Agarwal et al. 2016). The nanoparticle dispersion was then dried to powder using a lyophilizer. For the experiments, the dried powder was dispersed in sterile double distilled Milli Q water, (Bhattacharyya et al. 2018).

Preparation of arsenic As(V) standard solution: To make arsenic standard, 1.7334 mg of As_2O_3 was dissolved in 1000 mL of distilled water in order to achieve a concentration of 1 ppm Arsenic. Similarly for 10 ppm solution, 1.7334 mg of As_2O_3 was dissolved in 100 mL of distilled water. Arsenic standards of various concentrations were prepared in this manner using the dilution method.

CHARACTERIZATION

A JEOL JEM-3100F transmission electron microscope operating at 200 kV was used for high resolution transmission electron microscopy (HRTEM). A drop of the ZnO suspension in ethanol was placed onto a standard carbon coated copper grid to prepare the sample for TEM. Before taking the micrographs, the grid was dried. The sample's UV-visible absorption data was collected using an ELICO-SL-159 spectrophotometer in the wavelength range 200-800 nm. The elemental concentrations of various samples were determined using an ICE 3000 Series Thermo Scientific Atomic Absorption Spectrometer (AAS).

METHODOLOGY

As shown in figure 1, the ZnO nanoparticles and nanorods were prepared using the method as discussed above. The source of Arsenic used was Sodium Arsenate (Na_3AsO_4) which is a chemical source of As(V), a positive pentavalent ionic form. 4mM of ZnO nanoparticles solution was prepared and mixed with 300 ppb of spiked As(V). Multiple samples of the mentioned mixture were prepared and kept for different durations (15 mins, 30 mins, 90 mins, 300 mins and 600 mins) before passing them though a 100µm pore size Whatman filter paper.
The filtered and unfiltered samples were analyzed with Atomic Absorption Spectrophotometer (AAS) to determine the concentration of Arsenic.

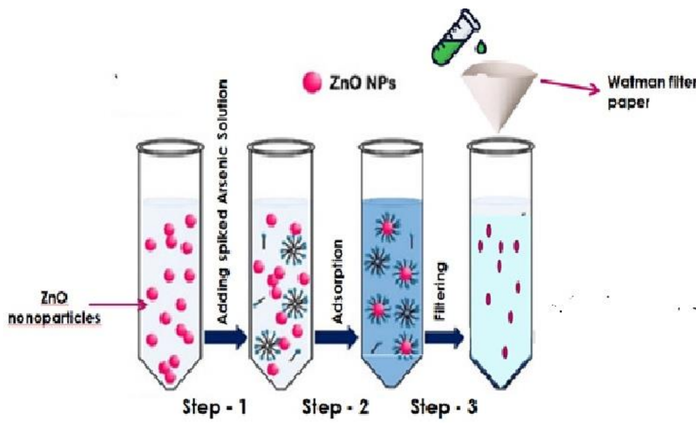

Figure 1: Schematic diagram of As(V) removal process

RESULTS AND DISCUSSION

The optical absorption spectrum for the ZnO nanoparticles in ethanol, shown in fig. 2(a) indicate that the ZnO nanoparticles have a very high absorption in the UV region of the electromagnetic spectrum with a shoulder in the wavelength range of 250-280 nm. Fig 2(b) shows the Tauc's Plot for ZnO nanoparticles from which the optical band gap was estimated as 5.54 eV indicating the presence of very small nanoparticles. The absorption edge at wavelength of 300nm indicates a blue shift, and an increment in the band gap energy, which should be due to the quantum confinement effect in the nano regime. In the HRTEM image shown in fig 2(c), the individual nanoparticles are clearly seen and the inset in the figure gives the particle size distribution where most of the particles are of the size of 4 nm.

Removal of arsenic ions through ZnO NPs: Arsenic ions were removed from various water samples using ZnO NPs. The removal of arsenic from water samples was investigated by varying parameter like pH of the solution, contact time, and temperature.

Effect of pH: The effect of pH on arsenic ion elimination at different pH levels (1-7) was studied at room temperature for 60 minutes. To adjust the pH, HNO_3 was used. The sample solution in this experiment must be neutral, with a pH of 7. Figure 3 shows that the maximum arsenic ions were evacuated at pH 5. Arsenic oxide

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generates oxy anions in aqueous media at high pH levels. Furthermore, because hydroxyl ions are abundant on the surface of produced NPs, the effectiveness of arsenic removal in acidic media is greater than in basic media. The arsenic removal efficiency was observed to have followed an exponentially decreasing trend defined by Eq. (1) limited to ±5% error.

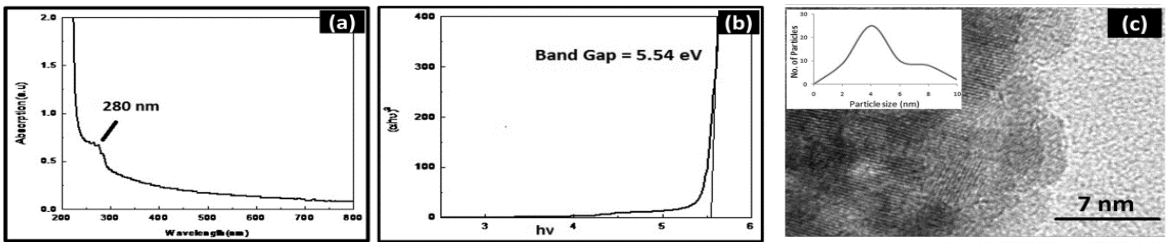

Figure: 2 (a) Optical absorption spectrum for the ZnO nanoparticles (b) Tauc's Plot for ZnO nanoparticles (c) HRTEM image for ZnO nanoparticles: Inset : Particle size distribution.

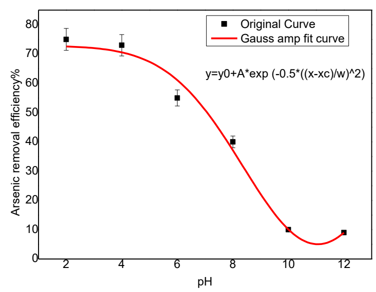

Figure 3: pH values vs arsenic removal efficiency.

$$y = y_o + A.e^{(-0.5((x-xc/w)^2)}\text{...}(1)$$

where,

y_0 = 72.81

x_c = 11.06

w= 2.704

A= -67.77

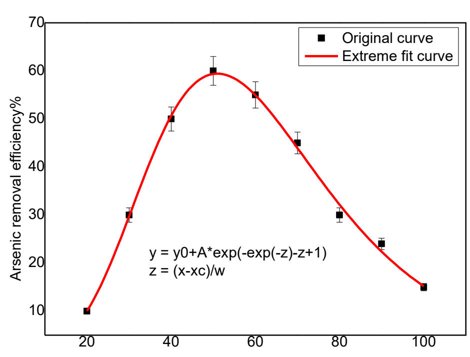

Figure 4: Temperature vs arsenic removal efficiency.

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Effect of temperature: Temperature effects on arsenic removal were investigated at temperatures ranging from 20ºC to 90ºC degrees Celsius while keeping the adsorbent content constant in acidic media. Figure 5 shows how the percentage uptake of arsenic by adsorbing material increases with increase in temperature from 20°C to 50°C and then falls out as shown in the figure 4. Desorption of arsenic from adsorbing material may occur at higher temperatures which caused the decrease in arsenic removal effectiveness. The arsenic removal efficiency was observed to have followed an increasing and then a decreasing trend defined by Eq. (2) limited to ±5% error.

$$y = y_o + (A/(w*\sqrt{(\pi/2)}))*e^{(-2*((x-Xc)/w)^2)} \qquad (2)$$

where,

y_0 = -11.251

X_c = 60.1074 ºC

w= 55.3569

A= 4515.40

Effect of contact time: At room temperature for 10 to 120 minutes, the effect of contact duration on arsenic removal was investigated while the adsorbent concentration remained constant. In an acidic atmosphere, the entire procedure was studied. There was a consistent increase in arsenic removal from 10 to 60 minutes, but no substantial arsenic removal after 60 minutes, as seen in figure 4. The early rate of arsenic removal was higher, which could be due to the adsorbent's availability of unoccupied sites, and the subsequent reduction could be due to the saturation of accessible sites on synthesized NPs. The arsenic removal efficiency was observed to have followed an increasing and then getting saturated trend defined by Eq. (3) limited to ±5% error.

$$y = A_1*e^{(-x/t1)} + A_2*e^{(-x/t2)} + y_o \qquad (3)$$

where,

y_0 = 77.19

A_1 = -101.7

t_1 = 13.53

A_2 = -101.7

t_2 = 13.53

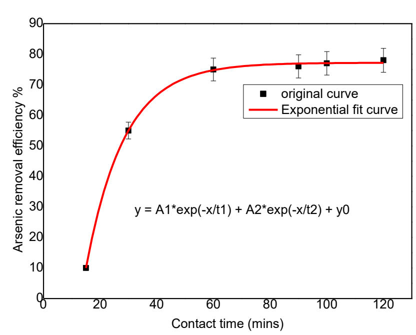

Figure 5: Contact time vs arsenic removal efficiency.

CONCLUSION

The process of adsorption is enhanced by using ZnO nanoparticles to remove Arsenic from water. The optimum working values for parameters like pH value, temperature and contact time were found following the above mentioned process and keeping the parameters in their optimum values, maximum adsorption was observed. The Arsenic phenomenon of ZnO nanoparticles demonstrated in this study shows potential in developing a practical one-step process for field applications. This can solve the requirement of expensive processes to remove Arsenic from water and making it suitable for drinking within the safe limits of contaminants.

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