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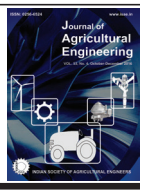

Evaluating Performance of a Nano-Filter in Removal of Arsenic Trioxide from Water

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ABSTRACT

Arsenic is a toxic element present in groundwater causing severe health issues, which require proper treatment before using groundwater for drinking purpose. A study was conducted to remove arsenic trioxide [As (III)] from groundwater using iron oxide-coated sand (IOCS). Batch study was conducted as a function of contact time (30 and 120 min), flow rate (1, 2, 4 and 7 $l.h^{-1}$), thickness of filter media (50, 75 and 100 mm) for initial arsenic concentration (0.10 and 0.25 $mg.l^{-1}$). Surface characteristics study of IOCS was done using scanning electron microscopy, Fourier transformation infra-red, energy dispersive X-Ray and X-Ray diffraction. Scanning electron microscopy showed the presence of iron oxide on the surface of coated sand, Fourier transformation infra-red analysis showed the chemical bonding of iron at 525 cm^{-1} and energy dispersive X-Ray analysis showed 20.76 % of iron on coated sand. During study, maximum As(III) removal was 100 % for initial concentration of 0.25 $mg.l^{-1}$ and 0.10 $mg.l^{-1}$, respectively, for contact time of 120 min at flow rate of 1 $l.h^{-1}$ for 100 mm thickness of filter material. Results of the batch study suggested that IOCS can be effectively used to achieve low level of arsenic in drinking water.

Wastewater released into water bodies without treatment potentially leads to contamination of groundwater, which is a serious ecological problem as some of these like arsenic and lead are toxic even at low concentrations (Singh *et al*., 2008). Arsenic, highly toxic in its inorganic form, is classified as one of the most carcinogenic chemical elements (Anon., 1999). According to drinking water guidelines suggested by the World Health Organization (WHO), Geneva, the maximum permissible limit of arsenic is 0.01 $mg.l^{-1}$ (WHO, 2011), and its pollution has been reported in Canada, China, India, Japan, New Zealand, Taiwan and United States of America (Mohan and Pittman, 2007). Higher concentrations of arsenic (As) greater than 10 $mg.l^{-1}$ in many states of India have become a major concern in recent years. About 0.2 million groundwater samples have been recently analysed for determination of arsenic contamination from different parts of the country (Mishra *et al.,* 2016). However, 90 % of those samples covered only the eastern part, while several states and union territories (UTs) of the country are still unexplored. It is reported that groundwaters of 18 Indian states and 3 UTs are contaminated with arsenic to different extents due to natural and/or anthropogenic sources (Mishra *et al.,* 2016). Among these, As greater than 300 $\mu g.l^{-1}$ in groundwater has been reported from at least one locality from 14 states. However, the problem is more serious in the state of West Bengal, followed by Bihar and Uttar Pradesh. Five out of 8 north-eastern states are also affected by arsenic contamination. Manipur is ranked as the first As-contaminated State and Assam as the second, followed by Arunachal Pradesh, Tripura and Nagaland. Groundwaters in the above-mentioned states are naturally arsenic-enriched, and therefore, wide spatial distribution of arsenic has been found (Mishra *et al.,* 2016). In north India, Punjab and Haryana, Andhra Pradesh and Karnataka in south India are also suffering with arsenic contamination of groundwater. Low levels of As (up to 17 $\mu g.l^{-1}$) have also been reported from Tamil Nadu in south India

(Mishra *et al.,* 2016). In the northern and central parts of Punjab, the concentration of arsenic in groundwater varied from 0.003 mg.l^{-1} to 0.042 mg.l^{-1}, and 0.009 mg.l^{-1} to 0.042 mg.l^{-1}, respectively (Hundal *et al*., 2007).

Various adsorption materials as activated alumina, activated carbon, fly ash, ferric hydroxide and zero valent iron have been used to remove the contamination of arsenic (Ladeira *et al*., 2001; Twindell *et al*., 2005; Jeon *et al*., 2009). In third-world countries, the present focus is on arsenic removal using iron-containing compounds due to their economy and effectiveness (Ramaswami *et al*., 2001). Iron oxide-coated sand-filter is an emerging technology for arsenic removal from groundwater (Gupta *et al*., 2005). It has several advantages including formation of highly sorptive properties against the contaminants, formation of highly porous media with minimum clogging risks and easy possible regeneration and reuse to amend the reactive properties (Cantrell and Kaplan, 1997). Iron oxides have a relatively high surface area and surface charge, and often regulate free metal and organic concentrations in water through adsorption reactions (Davis and Bhatnagar, 1995; Bajpai and Chaudhuri, 1999; Lin *et al*., 2006). Iron hydroxide-coated sand has high efficiency in removing micro-organisms, turbidity and heavy metals (Ahammed and Meera, 2010).

Iron oxide-coated sand along with adsorption materials has thus been used for arsenic removal from groundwater. However, information on use of nano-filter with varying thickness of filter media, contact time and flow rate is lacking. Therefore, the present study was undertaken to evaluate nano-filter by using iron oxide-coated sand in combination with other filtering media for removal of arsenic trioxide with varying thickness of filter media, contact time and flow rate.

MATERIALS AND METHODS

A laboratory study was conducted in the Department of Soil and Water Engineering, Punjab Agricultural University (PAU), Ludhiana for preparation and study of surface characteristics of iron oxide-coated sand, fabrication of a filter and arsenic removal from water through nano-filter during the years 2014-15. Fine sand, coarse sand, gravel and charcoal were selected for coating of iron oxide on the basis of geometric size. Double-distilled water was used for the preparation of solutions. Arsenic detection kit manufactured by M/SE. Merck India Private Limited, Merck, Germany, was used for determination of arsenic trioxide [As(III)].

Fabrication of Nano-Filter

An acrylic sheet box (700 mm height, 200 mm length, 120 mm width) was used for fabricating the filter. The filter was divided into three compartments: (i) reservoir tank (at top), (ii) material container (at middle), and (iii) filtered storage tank (at bottom) as shown in Fig. 1. The dimension of reservoir tank and filtered storage tank was 200 mm×200 mm×120 mm. It could store water up to 4.8 litres. The container dimension of filter-material was 300 mm×200 mm×120 mm. The container was divided into three parts for filling different types of material (sand, charcoal and iron oxide-coated sand).

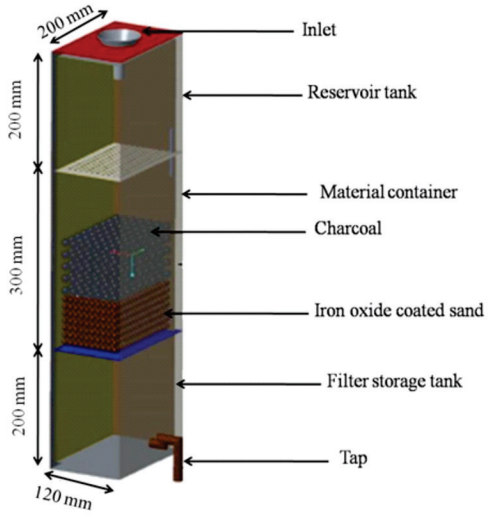

Fig. 1: Schematic view of developed filter

Preparation of Iron Oxide-Coated Sand

Iron oxide-coated sand (IOCS) was prepared by following the method described in Bailey *et al.* (1992). River sand of 0.30 mm geometric mean size and chemicals of analytical grade were used for coating process in this study.

Two hundred gram of river sand was soaked in 100 ml (50 %) sulphuric acid solution for 24 h in a beaker. After 24 h, the acid-soaked sand was washed with de-ionized water until the drained water was clear and pH of water was 7 (neutral). Sand was then put in a hot air oven at 105 ^{o}C for 24 h for drying. Washed and dried river sand was mixed with 80 ml of 2.47 M ferric chloride, and the pH was adjusted to 11 by adding 2 ml of NaOH solution and mixed for 2 min. The mixture was then placed for oven-drying at 110 ^{o}C for 24 h. The coated sand was washed with distilled water until the drained water was clear, and then dried at 105 ^{o}C for 3 h. After

drying, coated sand was put in a furnace at 550 ^{o}C for 3 h to increase coating strength.

Surface Characteristics of Iron Oxide-Coated Sand

A combination of scanning electron microscope (SEM), energy dispersive X-Ray (EDX), X-Ray diffraction (XRD) and Fourier transformation infrared (FT-IR) was used to study the physical and chemical characteristics of iron oxide-coated sand (IOCS).

SEM is a microscope that produces image of a sample by scanning it with a focus beam of electrons. SEM analysis of the adsorbents was done using a Philips SEM (Model SEM-515) at resolution of 420× at Electron Microscopy and Nano Laboratory, PAU, Ludhiana. Samples for SEM analysis were coated with their carbon film in order to avoid the influence of charge effect during the operation of SEM. The chemical composition of IOCS sample was studied with an EDX analyser, and was characterized by XRD having range of 0° to 160° for 2θ (Model Bruker S8 Tiger). FT-IR gave the interfacial bonding mechanism between iron and silica. FT-IR spectra were obtained over the range of 400-4000 cm^{-1} (Model Perkin Elmer Spectrum RX1).

Experimental Variables

Arsenic adsorptions were evaluated for simple sand, charcoal and iron oxide-coated sand through batch tests. Batch experiments were performed to investigate the efficiency of As (III) removal with uncoated sand and iron oxide-coated sand. The study was carried out for material thicknesses of 50, 75 and 100 mm and at flow rates of 1, 2, 4 and 7 l.h^{-1} with contact time of 30, 120, and 240 min, respectively. The initial concentration of spiked As (III) water was taken as 0.10 mg.l^{-1} and 0.25 mg.l^{-1}.

Thickness of filter material, flow rate and contact time were the dependent variables, whereas arsenic concentration was the independent variable. The experiment was run for ten cycles for all the treatments, and arsenic concentrations of water were tested before and after filtration using Merck Arsenic detection kit. The detection range was between 0 and 0.5 mg.l^{-1}. The upper detection limit could be extended to 2.5 mg.l^{-1} with a simple 1 to 5 dilution.

Statistical Analysis

Data were tested for laboratory experiment by using factorial completely randomized design at 5 % of level of significance. The effect of concentration of arsenic in water, flow rate through the filter, thickness of filter media, contact time on performance of filter was evaluated by computing arsenic removal efficiency.

RESULTS AND DISCUSSION

Surface Characteristics

The SEM image of uncoated sand showed that it had a relatively uniform and smooth surface, with small cracks, micro-pores or light roughness found on the surface (Fig. 2a). The surface of coated sand was rougher as compared to uncoated sand (Fig. 2b). No cracks were visible except for the crystalline iron oxide found on the surface. Similar results were reported by Chang *et al*. (2008) in Korea for SEM study on coated sand. The presence of carbon was due to carbon film used for coating during the operation of SEM. The spectra showed the absence of iron in the uncoated

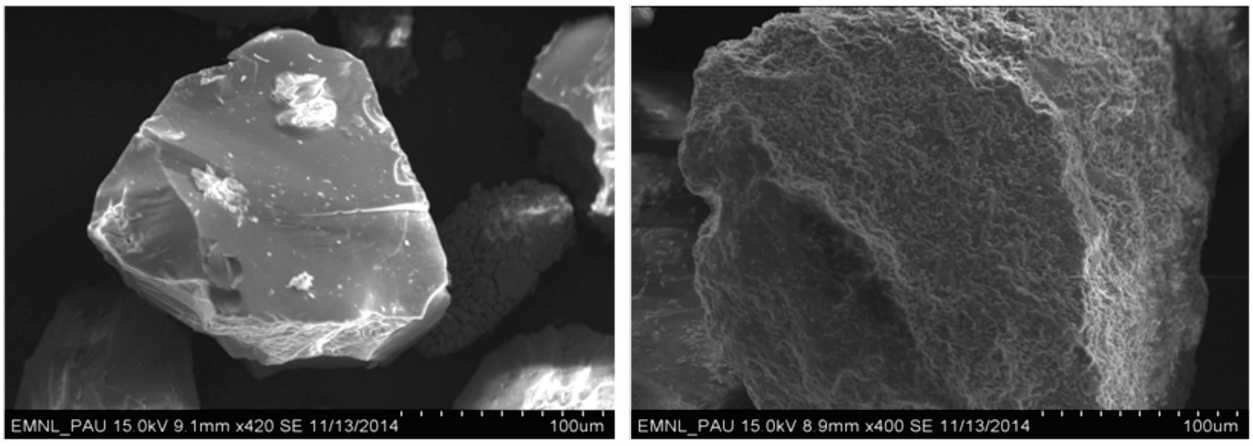

Fig. 2: SEM image of (a) uncoated sand, and (b) iron oxide-coated sand

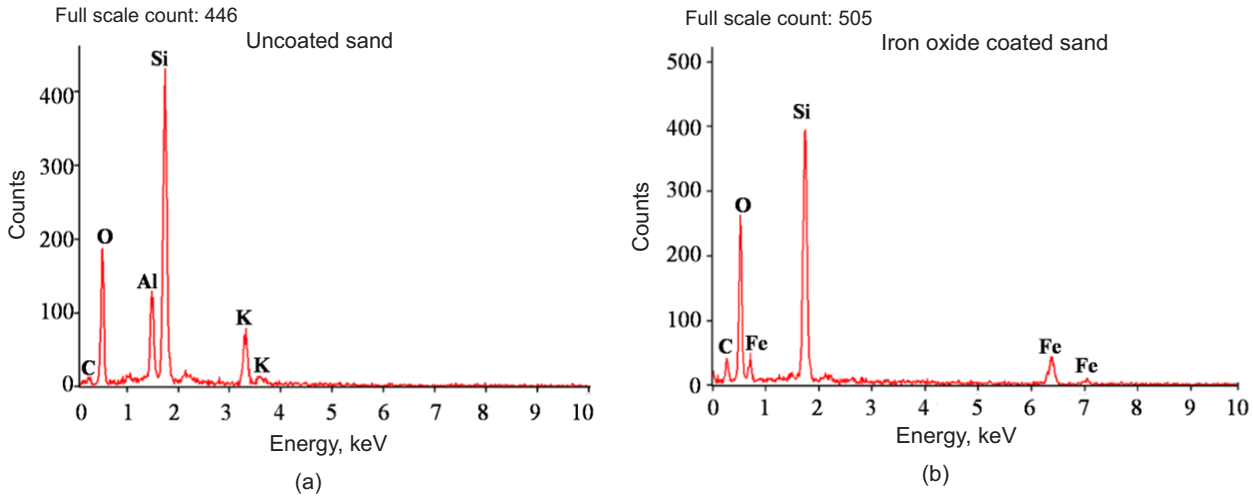

Fig. 3: EDX spectra of (a) uncoated sand, and (b) iron oxide-coated sand

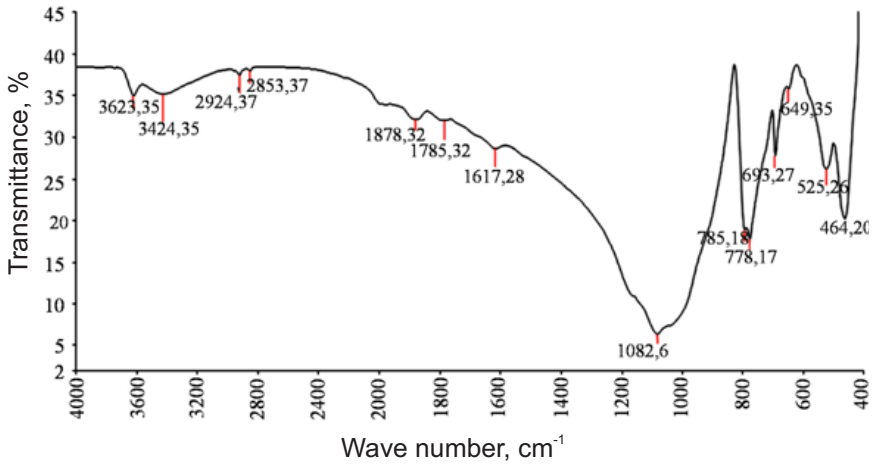

Fig. 4: FT-IR spectra of iron oxide-coated sand

sand. The EDX result showed 40.59 % oxygen, 7.03 % aluminium, 30.82 % silica and 11.84 % potassium. The EDX spectra of uncoated sand showed the presence of silica, oxygen, aluminium, potassium and carbon as shown in Fig. 3(a). The EDX spectra of IOCS showed the presence of iron, along with silica, oxygen and carbon as shown in Fig. 3(b). The iron content of coated sand was 20.76 % on weight basis. The presence of iron oxide at a high percentage indicated the effectiveness of the iron-oxide coating.

The surface chemical-structure of Fe_2O_3 nano-particles was characterized by FT-IR spectroscopy as shown in Fig. 4. The IR spectra of the iron oxide-coated adsorbents were recorded in the range of 4000-400 cm^{-1}. The result of FT-IR spectroscopy showed that the bands at 3623.35 cm^{-1} and 3424.35 cm^{-1} were due to O-H stretch. The peak at 1617.28 cm^{-1} was due to the existence of Fe-O stretch. Si-O band was shown at 1082.6 cm^{-1}, and 795.18 cm^{-1} was due to Si-O stretch. Si-O strong band was shown at 778.17 cm^{-1}, and at 693.27 cm^{-1} was due to Si-O-Si band.

The FT-IR spectrum of iron oxide exhibited strong bands in the low-frequency region at 1000–500 cm^{-1} due to iron oxide skeleton, which was consistent with the magnetite (Fe_2O_3) spectrum. The band at 525.26 cm^{-1} was due to weak adsorption of Fe-O, and a medium band at 465.20 cm^{-1} might be due to Fe-O. The FT-IR result showed presence of ferric oxide coating on sand (Palanivel and Velraj, 2007).

XRD was used to identify the crystalline structure of synthesized Fe_2O_3 nano-particles, which was obtained for IOCS using CuK radiation (λ = 1.5406A°) with a scan speed of 2.4θ min^{-1} and scan range of 10–80$^{\theta}$.

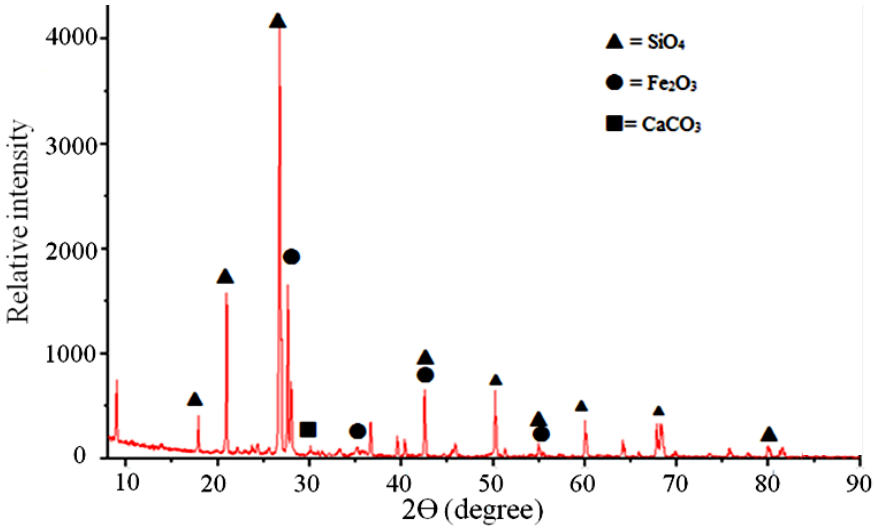

Fig. 5: Peak identification of iron oxide-coated sand by XRD

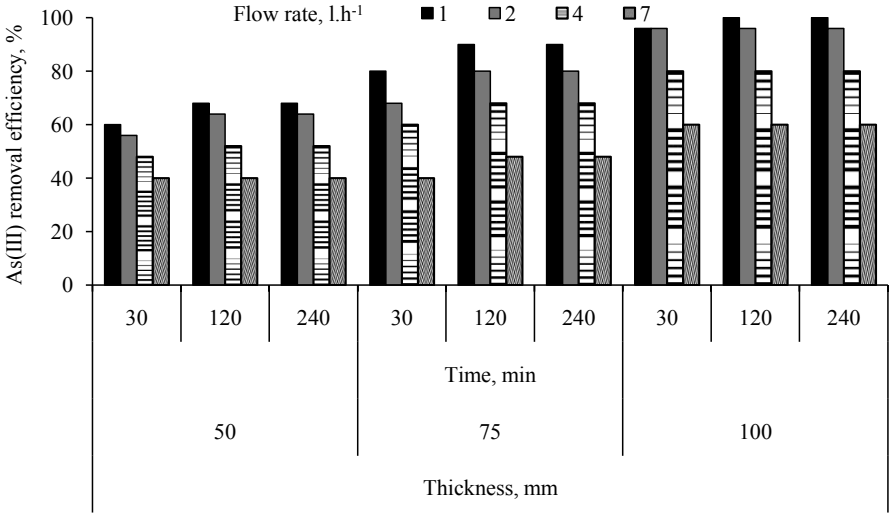

Fig. 6: Effect of contact time, thickness and flow rate on As (III) removal efficiency for initial concentration of 0.25 mg.l^{-1}

Peaks corresponding to Fe_2O_3 (ASTM data card) were present in the XRD spectrum along with the peaks of SiO_4, which confirmed the coating of the sand with Fe_2O_3 nano-particles as shown in Fig. 5.

Batch study for adsorption of As (III)

After passing spiked As (III) water through uncoated sand and charcoal, the initial and final concentration of As (III) remained unchanged, reflecting that As (III) was not removed by uncoated sand and charcoal for all considered thicknesses of media, specific flow rates and for specific contact time. The As (III) containing water was passed through IOCS. During the first study, the thickness of filter media (IOCS) was kept at 50 mm with contact time of 30, 120 and 240 min at flow rates of 1, 2, 4 and 7 l.h^{-1}. Results revealed that 40 – 70 % As (III) removal efficiency was achieved for initial concentration of 0.25 mg.l^{-1} and 0.10 mg.l^{-1}, and for 30 min contact time within all flow rates (Figs. 6, 7). For same filter thickness, As (III) removal efficiency increased to 40 – 75 % when contact time was increased from 30 to 120 min as well as 240 min. Further, As(III) removal efficiency was found to be same for adsorption time of 120 min and 240 min so adsorption time of 120 min was considered. Highest As (III) removal efficiency (75 %) with 50 mm thick filter was observed

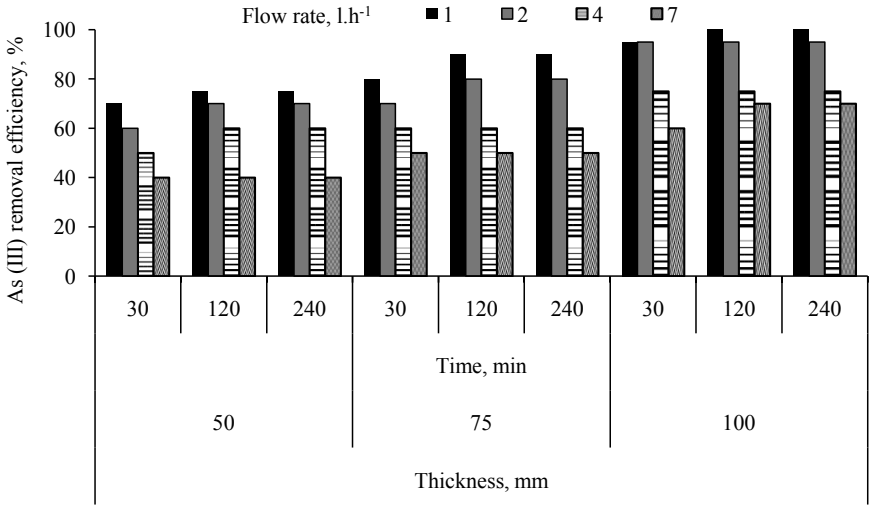

Fig. 7: Effect of contact time, thickness and flow rate on As (III) removal efficiency for initial concentration of 0.10 mg.l^{-1}

Table 1. Statistical analysis of adopted combinations

Treatment	Arsenic concentration (A)		Flow rate (B)				Thickness of media (C)			Contact time (D)		
	0.10	0.25	1	2	4	7	50	75	100	30	120	240
Mean removal efficiency, %	70.41	68.77	84.83	78.61	64.61	50.33	56.74	68.33	83.70	66.20	71.29	71.29
Critical Difference (5 %)	A=0.46		B=0.66				C=0.57			D=0.57		
	AB=0.93		BC=1.15				AC=0.81			AD=NS		
	BD=1.15		CD=0.99				ABC=1.62			ABD=NS		
	ACD=1.40		BCD=1.99				ABCD=2.81					

for initial concentration of 0.10 mg.l^{-1} with contact time of 120 min and flow rate of 1 l.h^{-1} (Fig. 7).

Thickness of filter media was then increased from 50 mm to 75 mm to examine the effect of increasing thickness on As (III) removal efficiency. For initial concentration of 0.10 mg.l^{-1} and 0.25 mg.l^{-1}, highest percentage of As (III) removal was found to be 80 and 90 % for contact time of 30 min and 120 min, respectively, at flow rate of 1 l.h^{-1} (Figs. 6, 7). The final concentration of As (III) was still higher than the maximum permissible limit of 0.01 mg.l^{-1} (WHO, 2011). Therefore, thickness of the filter media was further increased from 75 mm to 100 mm. With filter thickness of 100 mm, As (III) spiked water sample with 0.10 mg.l^{-1} and 0.25 mg.l^{-1} initial concentration could have 95 % removal efficiency at contact time of 30 min; and 100 % As (III) was removed from As (III) spiked water when contact time was increased to 120 min, which was found to be within maximum permissible limit of 0.01 mg.l^{-1}.

The effects of various discharge rates on As (III) removal were compared for contact time of 30 min and 120 min, and for different filter media thicknesses. On increasing the discharge rate, As (III) removal efficiency was found to decrease. The minimum removal efficiency was at 7 l.h^{-1} discharge rate, whereas the maximum efficiency was at 1 l.h^{-1} discharge rate. Different combination of concentration of arsenic, flow rate, media thickness and contact time showed the significant effect (critical difference=2.81) on removal efficiency for As (III) (Table 1). Increasing efficiency of arsenic removal might be due to the availability of reaction sites which were able to capture more ions around or inside the cells at a lower flow rate of 1 l .h^{-1}, and also due to the effect of contact time, thickness and flow rate of the filter (Ghorai and Pant, 2005; Devi *et al.,* 2014). IOCS was also observed as a good filter media for removal of As (III) from the arsenic contaminated groundwater (Joshi and Chaudhuri, 1996; Thirunavukkarasu *et al.,* 2003).

CONCLUSIONS

Surface characteristics of the iron oxide-coated sand showed the presence of iron oxide coating on sand. EDX analyser of uncoated sand showed absence of iron, and presence of iron (20.76 % on weight basis) in iron oxide-coated sand, indicating the effectiveness of iron oxide coating. FT-IR result also showed presence of iron oxide coating on sand. Removal of arsenic through uncoated sand and charcoal was negligible. Contact time of 120 min at flow rate of 1.0 $l.h^{-1}$ through 50 mm thick filter material gave maximum As (III) removal efficiency of 68 % and 75 % for initial concentration of 0.25 $mg.l^{-1}$ and 0.10 $mg.l^{-1}$, respectively. With increase in filter thickness from 50 mm to 75 mm, and from 75 mm to 100 mm, the removal efficiency increased to 80 % and 100 %, respectively. The results of the study suggested that a filtration system containing iron oxide-coated sand of 100 mm thickness can be used in small-scale or household purposes for effective removal of As (III) from contaminated water.

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