

# REMOVAL OF ARSENIC (V) FROM WATER BY ADSORPTION ONTO LOW-COST AND WASTE MATERIALS

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## Abstract

*In the present study, waste and low-cost materials like Cast-Iron Filings (wastes from mechanical workshops, lathes) and Steel Wool (commercially available, used for cleaning of wood surfaces prior to polishing) were investigated to assess their potential for removal of Arsenic from water by the process of adsorption by conducting different types of batch adsorption experiments, continuous flow column studies and candle filter studies. Batch sorption experiments brought forth the effectiveness of sorbent materials for removal of Arsenic from water achieving a removal of 90-95% at favorable pH conditions which lie in the normal range. Isothermal equilibrium data fitted well into the Langmuir model suggesting formation of unimolecular monolayer of sorbate over a homogenous surface of uniform energy. Both desorption studies and the shape of the saturation curves denote high affinity of the sorbent for the sorbate. Regeneration studies indicate repeated regeneration of the sorbents thus extending the usage of the sorbent following regenerations. While sulfates, Fluoride and chlorides have little influence on sorptive removal, phosphates and silicates exercise considerable influence by reducing removal.*

*Fixed bed continuous down-flow column studies conducted using ground water spiked with 300ug/l Arsenic (V) at flow rates of 5.0 and 10.0 ml/min indicated the usefulness of the sorbents for engineering applications for removal of Arsenic from water. Candle filter studies conducted by filling the hollow portion of the candles used in the domestic filters filled with sorbent material further demonstrated the utility of the existing household filters for removal of Arsenic (V) to less than the permissible limits of BIS/WHO by simply filling the hollow portion with the sorbent material. No leaching of Iron was found in the filtered water.*

**Key words:** Arsenic, water, removal, adsorption, cast iron filings, steel wool, household filter.

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## 1. INTRODUCTION

As a universal solvent, water effectively dissolves many minerals and organic compounds. These compounds find their way into waters as a result of complex interactions between precipitants, soil, rock and underlying geological strata. In addition, anthropogenic activities through industrialization and agricultural activities have a tremendous influence on its purity. Thus, our water resources have become unfit for direct consumption. Arsenic is one such toxic metalloid commonly occurring in the natural ecosystem. It is the 20<sup>th</sup> most abundant element in the earth's crust, moderately soluble, common in sedimentary mineral deposits and is harmful for human health. There are 48 countries in the world where Arsenic in drinking water has been detected at concentration greater than the guideline value of 0.01 mg/L or the prevailing national standards. However, the world's six largest cases of groundwater contamination and the worst sufferings of the people have been in Asia (Bangladesh, Nepal, West Bengal (India), Mongolia, China and Taiwan) where 60 million people are at the risk of Arsenicosis.

Arsenic enters into the human body through Arsenic-bearing food stuffs, water, or air. Intake of Arsenic associated with food

is a common phenomenon, but this Arsenic is the low-toxicity organic arsenic. The major passage of entrance is through, consumption of Arsenic laden water (groundwater). Arsenic is naturally found in atmospheric air in concentration levels of about 0.4 to 30ng/m<sup>3</sup>, in food at concentration levels of about 0.4 to 120 µg/kg and in water at concentration levels from undetectable to few mg/L.

Arsenic is found in organic and inorganic forms. The organic form of Arsenic is insignificant in most drinking water sources. Inorganic Arsenic is encountered in surface waters contaminated with industrial Arsenic wastes and groundwaters from Arsenic bearing aquifers. Environmentally important oxidation states of inorganic arsenic in aqueous chemistry are the pentavalent Arsenate species (H<sub>3</sub>AsO<sub>4</sub>, H<sub>2</sub>AsO<sub>4</sub><sup>-1</sup>, HAsO<sub>4</sub><sup>-2</sup> and AsO<sub>4</sub><sup>-3</sup>) and the trivalent Arsenite species (H<sub>3</sub>AsO<sub>3</sub>, H<sub>2</sub>AsO<sub>3</sub><sup>-1</sup>, HAsO<sub>3</sub><sup>-2</sup> and AsO<sub>3</sub><sup>-3</sup>). The trivalent form of Arsenic is more toxic and has also been found more difficult to be removed.

Arsenic is known as the king of all poison. Prolonged use of water having Arsenic even at low concentrations for drinking purpose is detrimental to human beings and animals, causing

several neurological, dermatological, gastro-intestinal and vascular disorders such as blackfoot disease, keratosis, hyper pigmentation and depigmentation, besides being carcinogenic. Because of health implications, several countries are initiating measures for reduction in its Maximum Contaminant Level (MCL). The U.S Environmental Protection Agency (EPA) has revised the maximum contaminant limit of Arsenic from 0.05 to 0.01 mg/L. However, since 1975, India and Bangladesh are still adopting 0.05 mg/L as the maximum contaminant limit of Arsenic in drinking water. Because of its detrimental effects the ideal level of Arsenic in drinking water should be nil.

The proposed paper envisages use of locally available, low-cost and waste materials as Cast Iron Filings (scrap Cast Iron turnings from drilling and lathe shops) and Steel Wool (commercially available from hardware shops) are proposed to be investigated for removal of Arsenic from water employing batch sorption studies and continuous down flow column studies. Further, it is proposed to develop point-of-use filters for removal of Arsenic at household level using the above materials without incurring additional investment cost. Use of pre-rusted Cast Iron Filings and Steel Wool in domestic candle-type filters is an innovative idea wherein the existing domestic filters can be converted into point-of-use filters.

## 2. SORPTION MEDIA

Cast Iron Filing used in this work was obtained from local workshops/lathe shops as a waste scrap, is a by-product of mechanical process such as lathing and drilling. The material was sieved to a geometric mean size of 212  $\mu\text{m}$ .

Steel wool used in this work was purchased from local hardware shops and is used for scraping of wood surfaces before painting/polishing. The material was in the form of fibrous coils. Steel wool was cut into fibres of 3-5 mm size. All the sorbents were washed with distilled water to remove fines and other adhered impurities and then air dried for 24 hours to allow air oxidation and rusting.

### 2.1 EXPERIMENTATION

Stock solution of 1.0 ml = 100  $\mu\text{g}$  As(V) was prepared from Sodium hydrogen arsenate ( $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ ) and working solution of desired Arsenate concentration were prepared fresh by appropriate dilution of stock solution. Agitated, non-flow batch sorption studies were conducted to study the effect of controlling parameters like contact time (15-540 min), sorbent dosage (2-20 g/L), solution pH (3-12), influence of interfering anions (Sulphates, Fluorides, Chlorides, Silicates and Phosphates) on Arsenate uptake by the sorbents.

To an aliquot of 100 mL of test Arsenate solution taken in a reagent bottle, premeasured quantity of sorbent material was added, and the resultant mixture was agitated in a rotary shaker at a rate of 35-40 rpm. To find the equilibrium contact time, the bottles were agitated for time intervals of 5, 15, 30, 60, 120,

180, 240, 300, 360, 420 and 480 minutes, withdrawn from the shaker at designated time intervals and the sorbent was separated from the solution by filtering through a Whatman ashless filter paper No.42. The filtrate was analyzed for residual Arsenate concentration using Varian Spectra AA240FS Fast Sequential Atomic Absorption Spectrometer having a detection limit of 1.0  $\mu\text{g/L}$  (1.0 ppb) and equipped with an autosampler. Equilibrium isothermal studies were conducted by varying the doses of the sorbent and agitating the reaction mixture for equilibrium contact time at the end of which residual concentrations of Arsenate were determined. Effect of pH and influence of interfering anions were studied by varying pH of the test solution and adding predetermined quantities of anions, agitating to equilibrium time and analyzing residual Arsenic concentration. All the experiments were conducted at room temperature ( $29 \pm 2^\circ\text{C}$ ).

To investigate the engineering aspects of sorptive removal of Arsenic, fixed bed down-flow column experiments were conducted using polymethyl methacrylate (Perspex/acrylic) column of internal diameter of 40 mm and 1.0 mm length fitted with appropriate inlet and outlet arrangements. Tap water was used to prepare the Arsenate test solution of 300  $\mu\text{g/L}$  and two flow rates of 5.0 mL/min and 10.0 mL/min were used. Constant flow rates were maintained through out the experiment and residual Arsenate was analyzed at regular time intervals. To explore the feasibility of using the sorbents for removal of Arsenate at household level, candle is filled with sorbent material and the opening is plugged with cotton.

Tap water spiked with Arsenate (concentration: 300  $\mu\text{g/L}$ ) is used as feed water. Water coming out from the candle-type filter is collected and analyzed for Arsenate and iron at regular time intervals.

## 3. RESULTS AND DISCUSSION

Important physicochemical aspects for parametric evaluation of sorptive removal of Arsenate from water by Cast Iron Filings and Steel Wool were investigated and the results of all the experiments are presented either in graphical form or in tabular form and is followed by a discussion of the experimental findings.

### 3.1 EFFECT OF CONTACT TIME

Sorptive uptake of Arsenate by cast iron filings and steel wool from water at a solution pH of 5.0 is shown as a graphic representation of percent of Arsenate removal with contact time in Figure 1. It follows from the Figure that the rate of uptake by CI Filings is the highest at 84% and is followed by steel wool at 82%. Almost 50% removal occurred within 5 minutes and 68-70% in 60 minutes. Kinetics of sorption i.e. % sorption of sorbate as a function of contact time indicated a triphasic pattern with an initial steep portion denoting high rate of uptake; a transitional phase in which rate of up take decreases gradually and approaches a more or less constant value denoted by

curvilinear portion and a final equilibrium reached after a period of 240 minutes (4 h) and 360 minutes (6 h) in the case of Cast Iron Filings and Steel Wool respectively, in which rate of uptake is almost constant which is denoted by a straight line plateau. Initial high uptake is due to the diffusion of Arsenate from bulk of the solution onto the surface of the sorbent and is termed instantaneous sorption.

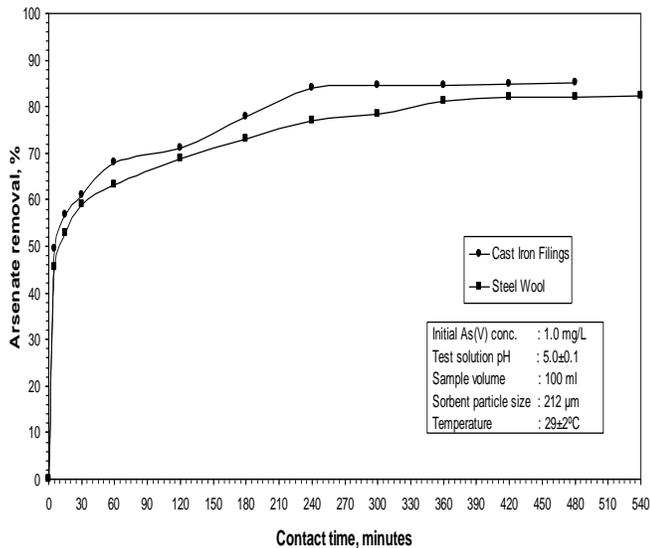


Figure1: Kinetics of sorptive uptake of Arsenate

3.2 EQUILIBRIUM ADSORPTION ISOTHERMS

Adsorption isotherms describe the relationship between the quantity of sorbate held by the sorbent and that remaining in the solution at equilibrium at a constant temperature. Adsorption isothermal studies were conducted by contacting a given amount of sorbate in a fixed volume of solution with varying amount of sorbents for the equilibrium contact time and the residual concentrations were found. The experimental results were processed in accordance with the Langmuir or Freundlich or BET equations to find the best applicable isotherm equation and thence to determine the sorption capacity and intensity and deduce the nature of the sorbent-sorbate reactions. The sorption data were fitted into the linearized Langmuir, Freundlich and BET equations and the values of sorption capacities and coefficient of correlation are presented in Table 1. It follows from the values of coefficient of correlation (r<sup>2</sup>) that Langmuir equation gives a best fit compared to Freundlich and BET equations.

Table1: Sorption capacities and coefficient of correlation (r<sup>2</sup>) for Arsenate

S. No	Sorbent	Values of coefficient of correlation (r <sup>2</sup> )			Slope	Intercept	Q <sup>0</sup>	b
		Langmuir	Freundlich	BET				
1	Cast Iron Filings	0.921	0.883	0.308	2.996	0.271	3.69	0.09
2	Steel wool	0.926	0.907	0.310	3.776	1.711	0.59	0.45

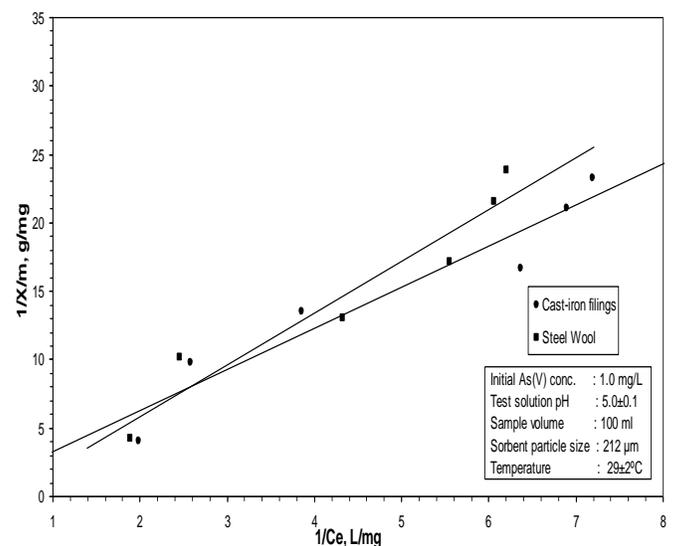


Figure2: Linearized Langmuir adsorption isotherm plot for Arsenate

Fitting of the Langmuir isotherm model suggests complete coverage and formation of a monolayer of sorbate over a homogeneous sorbent surface of uniform energy and that the adsorbed layer is unimolecular. However, a complete monolayer is not usually formed, adsorption sites may not have equal affinities for sorbate molecules and the presence of adsorbed molecules at one site may affect the adsorption of molecules at an adjacent site. Linearized Langmuir adsorption isotherm plot for Arsenate is shown in Figure 2. It may also be observed that the isotherm fit that Steel Wool has a negative intercept which indicates that the removal is good at lower concentrations, but not as good, at higher concentrations.

Experimental data of equilibrium isothermal adsorption may also be represented as saturation curves (sorption isotherms) in which the amount of sorbate sorbed per unit weight of the sorbent (X/m) is expressed as a function of concentration of sorbate remaining in solution (Ce) on an arithmetic scale. The shape of the saturation curve is often an indication of the

underlying sorbent-sorbate interaction. Saturation curves of CI Filings and Steel Wool closely resemble L- type curves of Giles classification (Giles, 1960) that denote a high affinity between the sorbent and the sorbate in the initial stages of the sorption interaction/process. As more sites in the sorbent matrix are filled, it becomes increasingly difficult for the solute molecules to find a vacant site for sorption.

### 3.3 EFFECT OF SORBENT DOSAGE

Sorption being a surface phenomenon, the amount of sorptive removal depends on the amount of sorbent surface available for sorption. Results of equilibrium isothermal experiments conducted by varying the dose of sorbent in the range of 2-20 g/L is shown graphically in Figure 3.

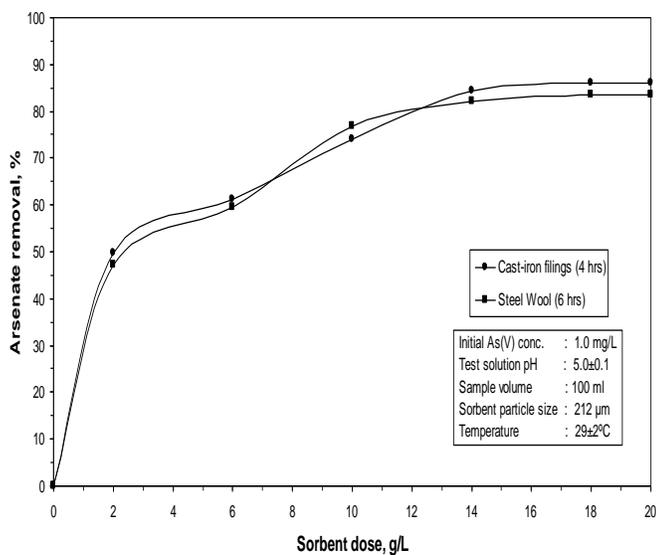


Figure3: Effect of sorbent dose on removal of Arsenate

The sorption curve is a typical sigmoidal curve denoting an increase in sorption as the sorbent dose is increased. The curve leveled off indicating exhaustion of sorption reaction though good amount of sorbent surface is available perhaps due to sorbate-solvent, sorbent-solvent and sorbent-sorbate interactions.

### 3.4 EFFECT OF PH

Equilibrium sorption studies were conducted at varying initial pH of the test As(V) solution in the range of 3.0 to 12.0. It follows from the results that the amount of sorption reduced considerably as pH was increased progressively from 3.0 to 9.0 and sorption decreased dramatically at high pH values i.e. pH>9.0. Highest removal of 93% and 89% was observed at pH 3.0 for an initial Arsenate concentration of 1.0 mg/L for Cast Iron Filings and Steel Wool respectively and sorptive removal decreased to around 20% at a pH of 12.0. The effect of pH on the removal of Arsenate is shown in Figure 4. Lackovic et al

(2000) and Manning and Goldberg (1996) also observed that Arsenate removal by zero valent iron is effective at low pH. Depending upon the pH, four species of Arsenate ions ( $H_3AsO_4$ ,  $H_2AsO_4^-$ ,  $HAsO_4^{2-}$  and  $AsO_4^{3-}$ ) may exist in aqueous solution (Snoeyink and Jenkins, 1980). At a pH of 3.0, Arsenate ions predominantly exist as monovalent negative species like  $H_2AsO_4^-$  (> 80%) and as a few natural species like  $H_3AsO_4$  (<20%). Zero valent Iron oxidized to ferrous iron undergo corrosion and the resulting corrosion products like ferrous oxide ( $Fe(OH)_2$ ) play a major role in effective removal of Arsenate species. At pH 9.0 and above, Arsenate ions exist as divalent and trivalent species having negative charges like  $HAsO_4^{2-}$  and  $AsO_4^{3-}$ . Corrosion products would also be negatively charged like  $FeO^-$ . This would result in poor removal due to charge-charge repulsion and hence less removal beyond pH 9.0. As the decrease in removal is more pronounced at pH values above 9.0, removal should be optimized at a pH of 6.0 or less.

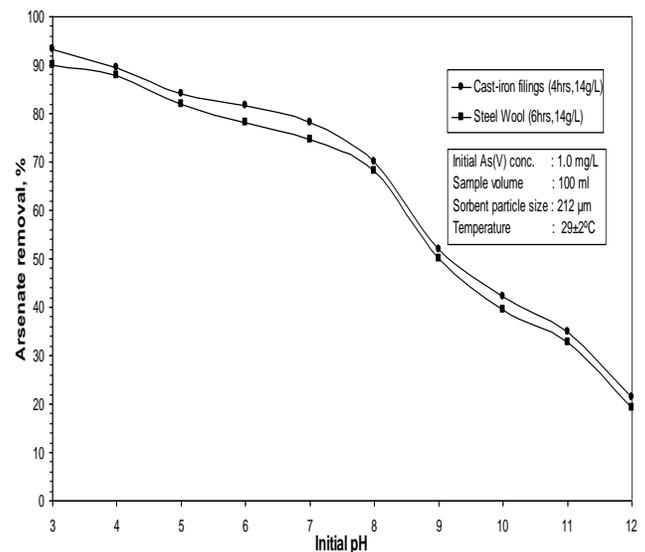


Figure4: Effect of pH on removal of Arsenate

## 4. DESORPTION AND REGENERATION STUDIES

Desorption and regeneration studies were conducted to understand the nature of sorption reaction and to investigate the possibility of reuse of spent adsorbent. Desorption experiments conducted employing distilled water as a desorbing agent yielded a desorption of about 24% and 28% in the case of CI Filings and Steel Wool respectively. It may therefore be inferred that the sorption of Arsenate is mostly irreversible in nature with a chemisorption type of interaction occurring between the Arsenate and sorbent surface. Regeneration studies were conducted with 0.1N HCl, 0.1N NaOH and 1% Alum solution. Insignificant regeneration (<15%) indicate that 0.1N HCl and 1% Alum are not useful as regenerants whereas regeneration with 0.1N NaOH was significant with about 65-73%. Therefore sorbent material may be regenerated using 0.1 N NaOH

solution. Perhaps, higher concentration of NaOH may result in higher regeneration which needs to be investigated.

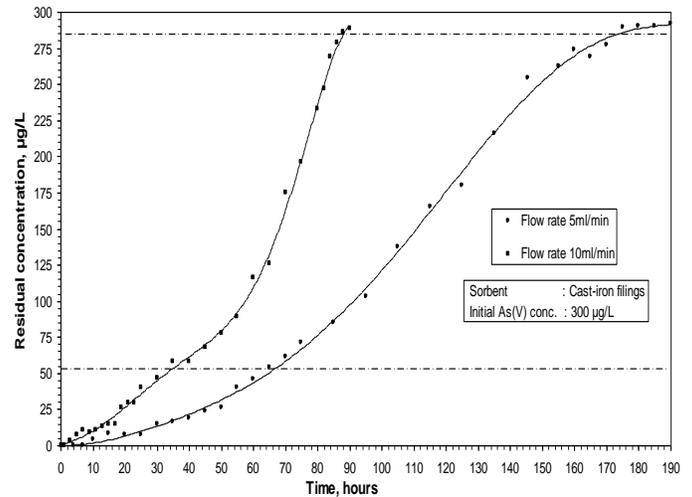
**5. FIXED BED DOWN-FLOW COLUMN STUDIES**

In real field conditions, adsorbents will be operated in continuous flow pattern. The capacity of sorbents for adsorption of a given sorbate is usually determined by equilibrium adsorption isotherm studies which are used with moderate success for scaling-up batch type process. However, isotherm data are inadequate for design of continuous flow systems since adsorption in a column is not in equilibrium condition and uneven flow pattern occurs in beds of adsorbent column. Therefore, in order to assess the applicability of the sorbents for continuous flow systems, extensive column studies are to be conducted and from the data of column studies, breakthrough time and breakthrough volume which are essential for operation and regeneration/replacement of the sorbent may be predicted.

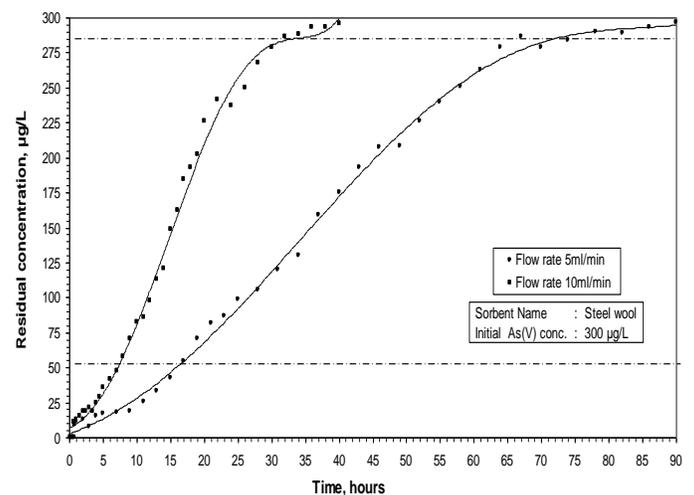
Fixed bed down-flow continuous column studies were conducted using tap water spiked with 300 µg/L concentration of As(V) and employing flow rates of 5.0 ml/min and 10.0 ml/min. Breakthrough curves for both the sorbent yield typical ‘S’ in shaped curves. As the flow rate is increased from 5 ml/min to 10 ml/min, the breakthrough curve becomes steeper. Results of column studies conducted at different flow rates of CI Filings and Steel wool are portrayed in Figure 5 and Figure 6 respectively. Performance data of column studies are presented in Table 2.

**Table2:** Performance data of fixed bed-down flow column studies

Sorbent	Qty. of sorbent (g)	Breakthrough time (hours) at flow rate		Breakthrough volume (liters) at flow rate	
		5ml/min	10ml/min	5ml/min	10ml/min
Cast-iron filing	732	66	32.5	19.80	19.50
Steel wool	191	16	7.5	4.80	4.50



**Figure5:** Breakthrough curves for removal Arsenate by Cast Iron Filings at different flow rates



**Figure6:** Breakthrough curves for removal Arsenate by Steel wool at different flow rates

It may be observed from the figure that the zone of adsorption moves gradually from one end to the other end which denotes the exhaustion of adsorbed bed and the breakthrough curves are typical ‘S’ in shape and the trends of the curves are similar. It may be deduced from the data presented in Table 2 that the capacity of CI Filings is 27 mL/g and that of Steel wool is 24 mL/g.

**6. STUDIES WITH DOMESTIC POINT-OF-USE CANDLE-TYPE FILTER**

Candle-type domestic filters are two-tier two compartment stainless steel/ceramic/porcelain/plastic units, cylindrical in shape and are of 16-24 L capacity. The upper compartment

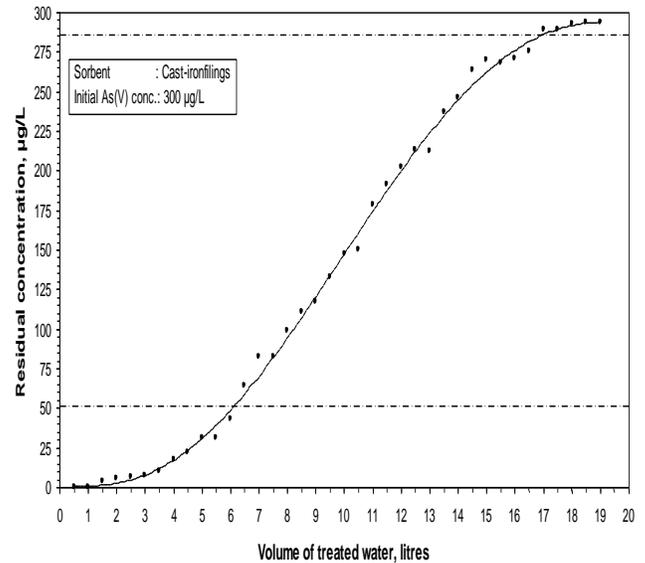
contains 1 to 3 candles fitted at the bottom into which the water to be filtered is poured manually. The water after filtration through the candle(s) is collected in the bottom vessel to which a tap is fixed to facilitate use of the filtered water. Candles are hollow, cylindrical in shape and are made of high porosity ceramic materials with appropriate arrangement for fixing to the bottom of the vessel. These filters are strong and porous allowing a maximum water flow of 1L/h and are effective in filtering out solids of sizes as minute as 0.01 mm. Candles are generally cleaned by autoclaving in domestic pressure cookers (121±10C @ 15 psi). These filters are easy to operate, cheap and sustainable.

The hollow portion of the candle is filled with the sorbent material and plugged with cotton. Tap water spiked with Arsenic is used as feed water. Filtered water was collected at regular intervals and analyzed for residual Arsenic concentration by Atomic Absorption Spectrophotometer and the data is used for plotting of breakthrough curve from which breakthrough volume and breakthrough time were computed.

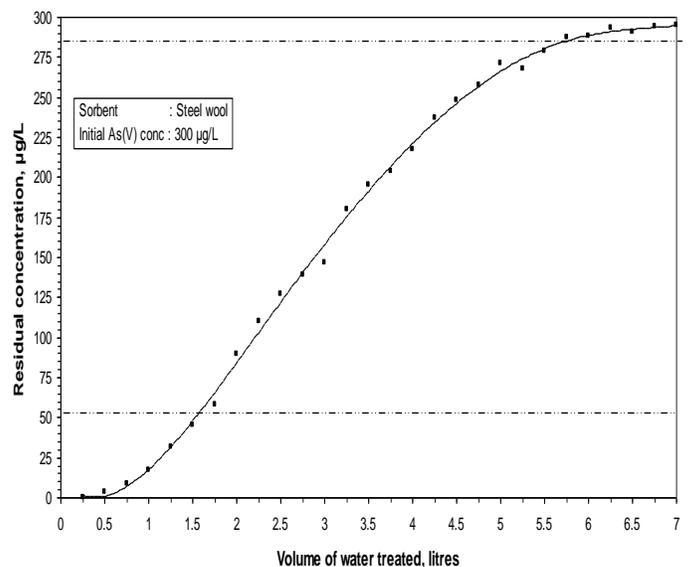
A single candle filter filled with 176 g of Cast Iron Filings or 47 g of Steel Wool could effectively remove Arsenic from 300µg/L to less than permissible limit of BIS guidelines and was capable of producing 6.0 L and 1.5 L per run. A filter fitted with 3 candles thus would produce 18.0 and 4.5 L of Arsenic free water per run. The output of Arsenic free water depends on the concentration of Arsenic in feed water. Lower feed concentration result in higher output of Arsenic free water and vice-versa. Iron content in the effluent of candle-type filters was less than the permissible limit of BIS/WHO guidelines. Results of candle-type filter studies are presented graphically as break through curve in Figures 7 and 8 and performance data in Table 3.

**Table3:** Performance data of domestic point-of-use candle-type filter studies

Sorbent	Amount of sorbent (g)	Breakthrough volume (liters)
Cast Iron Filing	176	6.00
Steel Wool	47	1.50



**Figure7:** Breakthrough curve for removal Arsenate by Candle filters using Cast Iron Filings



**Figure8:** Breakthrough curves for removal Arsenate by Candle filters using Steel wool

Arsenic-rich waste can be disposed of by converting it into volatile organic forms through the activities of the microbes in soil or sediments. One such disposal method is disposal in soil with cow-dung added.

**SUMMARY AND CONCLUSIONS**

Agitated, non-flow batch sorption studies indicate that sorption process proceeded quite rapid initially and attained equilibrium gradually and the contact time required to reach equilibrium was observed as 4 hrs and 6 hrs for Cast Iron Filings and Steel

Wool respectively. Isothermal equilibrium adsorption data fitted well to the Langmuir model indicating formation of a unimolecular monolayer of Arsenate over a homogeneous sorbent surface of uniform energy. Sorptive removal of Arsenate increased with increased sorbent dosage from 2 to 20 g/L but beyond a dosage of 14 g/L, increment in removal was insignificant. Sorption reduced as pH was increased progressively from 3.0 to 9.0 and decreased dramatically beyond pH 9.0. As the decrease in removal is more pronounced at pH values above 9.0, removal should be optimized at a pH of 6.0 or less.

Fixed bed down-flow column studies conducted at two flow rates yielded typical 'S' shaped breakthrough curves which became steeper as the flow rate is increased from 5 to 10 ml/min. Cast Iron Filings and Steel Wool column could produce 19.80 liters and 4.80 liters of potable water from an initial Arsenate concentration of 300 µg/L. A single domestic candle filter filled with 176 g of Cast Iron Filings or 47 g of Steel Wool could effectively remove Arsenic from 300µg/L to less than permissible limit and was capable of producing 6.0 L and 1.5 L per run. A filter fitted with 3 candles thus would produce 18.0 and 4.5 L of Arsenic free water per run.

The output of Arsenic free water depends on the concentration of Arsenic in feed water. Lower feed concentration result in higher output of Arsenic free water and vice-versa. Iron content in the effluent of candle-type filters was less than the permissible limit of BIS/WHO guidelines and hence is of no concern. The procedure can be replicated in any problematic areas of Arsenic as they are based on sound scientific principles.

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