



Removal of Lead from Drinking Water by Bioadsorption Technique: An Eco-friendly Approach

Prasenjit Mondal†, B. P. Yadav and N. A. Siddiqui

University of Petroleum and Energy Studies, Dehradun, U.K., India

†Corresponding author: Prasenjit Mondal; pmondal@ddn.upes.ac.in

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ABSTRACT

India is the world's largest user of groundwater, accounting for 25 per cent of the world's extracted groundwater. Contamination of water bodies is the main problem for degrading the potable water quality in India. The contaminants, mainly cadmium, chromium, nickel, lead, zinc, copper, calcium, fluoride, nitrates, etc. have a significant impact on waters. There have been several advancements in technology for removal or reduction of these contaminants in water such as reverse osmosis, UV filtration, distillation and ion exchange. But these methods are not found to be eco-friendly and cost-effective. This paper elaborates various techniques of organic nature that can be used to reduce/remove the heavy metals from water bodies using orange peels as a bioadsorbent and detailed experimentation for the removal of lead ion concentration from water. In the present study, synthetic water with various heavy metals of known concentration was treated by a bioadsorbent (orange peels) by changing various parameters such as adsorbent particle size, dose, initial pH, etc. At pH 7, dose 0.5 g, and adsorbent particle size of 250 µm, a drastic reduction in the concentration of lead from 10 ppm to 0.213 ppm in 50 mL synthetic sample was observed.

INTRODUCTION

Water stands first among the important fundamental requirements and also a basic nutrient having critical importance to human life (Howard et al. 2003) and is involved in almost every bodily function from digestion to excretion (Feldman et al. 1996). Water is transparent, colourless, odourless, and tasteless and covers almost 71% per cent of the earth's surface (Lindstrom 2012, Khyade & Swaminathan 2016). Coming to drinking water, it plays an important role in the life of every living organism (Vitousek et al. 1997). It boosts the metabolism and helps in breaking down the food but not all people get the quality drinking water (Congress 1995).

The problem we are facing currently is the pollution of water through various sources. Urbanization and industrialization are the main reasons for the pollution of water (Shiklomanov 1991, Czech et al. 2000, Le et al. 2010). With the increase in industrialization, there has been an increase in the high usage of different metals, chemicals and materials, etc. for the production of various products which resulted in producing waste which is being released into nearby water resources (Shukla et al. 2002). The chemicals, when mixed with water which is the source for drinking water in many areas, makes it toxic. Most of the people in many households are drinking the water which contains pathogens that may cause various diseases like typhoid, jaundice, diarrhoea, etc.

(Niemczynowicz 1999). A filtering mechanism uses techniques like RO, activated carbon filters to filter the water to make it fit for drinking (Matilainen et al. 2010).

The present work mainly focused on the removal of lead ions from water, a persisting problem in India, by using organic methods such as orange peels.

Heavy Metal Pollution in Water

Heavy metals have higher atomic weights and are approximately five times heavier than water. Many ecological and global public health concerns with the contamination by these metals have been emerged in recent years (Demirbas 2005). Heavy metals extensive use in several industrial, agricultural, domestic and technological applications has increased their exposure to the environment (Mulligan et al. 2001).

Pollution resulting from land runoff, precipitation, atmospheric deposition or land drainage is known as non-point source pollution (World Water Development Report 2017). Non-point sources are not easy to identify like the use of excess fertilizers or pesticides from agricultural lands or toxic chemicals from urban run-off and thus heavy metals find their way into water by industrial, agricultural, pharmaceutical, domestic effluents and atmospheric sources (Tarver 2008).

Heavy metals are naturally occurring in the earth's crust but the pollution caused by them is mainly due to the an-

thropogenic sources like mining, industrial production and using compounds containing metals (Annadurai et al. 2003). Some of the industrial sources include metal processing in refineries, coal burning in power plants, textiles, microelectronics, nuclear power stations and high tension lines, and combustion of petroleum (Hegazi 2013).

The metals like Co, Cu, Cr, Fe, Mg, Mn, Mo, Ni, Se and Zn are essential nutrients to the human body in adequate amounts. Insufficiency of these micronutrients in the human body results in a variety of ailments (Dobaradaran et al. 2017). Heavy metals are classified as human carcinogens (known or probable) according to the U.S. Environmental Protection Agency, and the International Agency for Research on Cancer (Howard et al. 2003). They are systemic toxicants which can induce multiple organ damage, even at lower levels of exposure (Pehlivan et al. 2008).

Heavy metals pollute the groundwater bodies and the surface water bodies like rivers, lakes, ponds. The primary metals like lead, arsenic, copper, cadmium, mercury and nickel are also known as trace elements, which are normally found in low concentrations. Surface water contamination is due to mining, dumping of solid wastes containing metal salts and discharge of effluents from battery and paint manufacturing, electroplating, viscous-rayon manufacturing, copper

picking industries (Tokalioğlu et al. 2000).

Previous Works on Heavy Metal Removal Techniques

The rapid growth in industries over the start of the 20th century has increased the demand for freshwater and thereby discharging the wastewater into the nearby water sources (Barlow & Clarke 2017). The discharged effluents from the industries contain harmful heavy metals like lead, chromium, cadmium, etc. in the form of dyes, coatings, pigments and so on and many of them are considered as hazardous to health because of their toxicity to human health. There are many methods available for the removal of these heavy metals from water such as reverse osmosis, chemical oxidation, adsorption, membrane separation, ion exchange, etc., out of which adsorption is considered as the simple and efficient method because of its ease of operation and versatility. The summative assessment on the removal of heavy metals from water by adsorption technique using organic wastes as adsorbents is listed in Table 1.

MATERIALS AND METHODS

Preparation of Adsorbent

Orange peels were used as adsorbents for the removal of

Table 1: Summative assessment on the removal of heavy metals from water using organic waste as adsorbent.

S. No.	Objective	Brief Summary	Outcome	References
1	Water purification using different waste fruit cortexes for the removal of heavy metals.	Water can be purified by using different fruit cortices to remove heavy metals. They used the peels of fruits like kiwi, tangerine and banana and crushed them into powder to an optimum size of 1 mm and 2 mm. A water sample was made by taking 50 mL wastewater and 0.5 g of the crushed powder of particle size both 1 mm and 2 mm is added into the flask as adsorbent. It is then kept in a shaker at 158 rpm for 60 minutes. It was later filtered and then the concentration of the metals was analysed by Inductively Coupled Plasma (ICP)	It has shown that the particles of size 1 mm had better adsorption when compared to the particles of size 2 mm. Of the fruits used, kiwi fruit gave better adsorption results than banana. The order of adsorption capacity of the metal ions for banana was $Cr < Cd < Zn$ and for Kiwi and tangerine, it was $Cd < Cr < Zn$. The capacity of adsorption will depend upon the pH, adsorbent dosage and the contact time. After conducting the experiments, the optimum value of pH was found to be 6.0	(Al-Qahtani 2016)
2	Adsorption of heavy metals from water using banana and orange peels.	banana peels and orange peels for the removal of heavy metals like Cu^{2+} , Zn^{2+} , Co^{2+} , Ni^{2+} and Pb^{2+} . Banana and orange peels, both were cleaned with double distilled water, dried and crushed into powder with particle size of 1-5 mm. The peels are treated with 0.4 mol/L Sodium Hydroxide, 0.4 mol/L Nitric Acid & distilled water. 15 g of banana peels and orange peels were soaked in 0.4 mol/L HNO_3 for 24 hours. "Synthetic solutions of $CuSO_4$, $CoSO_4$, $NiSO_4$, $ZnSO_4$ and $Pb(NO_3)_2$ are prepared". Experiments are done by adding 0.1gms of adsorbent in the 100 mL metal solution at a speed of 180rpm for 24 hours and centrifugation at 10,000rpm for 20 minutes, then later the concentration of metals were analysed by an atomic absorbance spectrophotometer (Varian Model 202FS)	The results from the experiments have shown that the maximum adsorption was at around pH 6-8 for banana and orange peels and the adsorption decreased with the increase in pH. The adsorption capacity was found to be 7.97 mg/g for lead, 6.88 mg/g for nickel, 5.80 mg/g for zinc, 4.75 mg/g for copper, and 2.55 mg/g for cobalt using banana peels. And for orange peels it was 7.75 mg/g for lead, 6.01 mg/g for nickel, 5.25 mg/g for zinc, 3.65 mg/g for copper and 1.82 mg/g for cobalt. The experiment also shown that maximum adsorption was attained at high pH with max level of lead by using banana peels was 7.97 mg/g and by using orange peels it was 7.75 mg/g.	(Annadurai, Juang et al. 2003)

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S. No.	Objective	Brief Summary	Outcome	References
3	Removal of heavy metals from wastewater using agricultural and industrial wastes as adsorbents.	Synthetic wastewater with known concentrations of copper sulphate, nickel nitrate and iron sulphate metal solutions are made separately in double distilled water using Cu, Ni, Fe. The stock solution consisting of 20 mg/L as the adsorbent dose and concentration of metal (Cu, Ni, Fe) is around 10 mg/L. The agitation speed is 200rpm. The adsorbent doses of 20,30,40,50 and 60 mg/L are taken. The adsorbents and the metal ions are added and stabilized by agitating to attain pH of range 2-10	water is purified by removing heavy metals using agricultural wastes like rice husk and industrial wastes like fly ash. First, The experiments are done and the results shown were that the agricultural waste rice husk found to be efficient in removing the metals like Fe, Pb and Ni whereas the industrial waste, fly ash has shown to be effective in removing i.e. adsorbing Cu and Cd. The experiments were also done by increasing the adsorbent concentration of rice husk and it was found that the Fe removal by using rice husk increased from 68.59% to 99.25%. Same when done for Pb, it was increased from 22.22 % to 87.17 %. When the concentration of fly ash was increased, the Cd removal increased from 25.21% to 73.5%. Same when done for Cu, the removal increased from 37.38% to 98.54%. The adsorption capacity was dependent on the level of pH, dosage of the adsorbent and the time of contact with the respective adsorbent. The contact time for attaining the desired level of metal ions was found to be 2 h and pH range was 6-7 for the effective adsorption.	(Hegazi 2013)
4	A laboratory study using maple leaves as a bio-sorbent for lead removal from aqueous solutions.	Lead was removed from aqueous by using maple leaves as a bio-sorbent. The materials used were one kg of maple leaves, salt, lead nitrate solution, Pb (II) and water.	10 mg/L removed 98.2% Pb (II) ions than other concentrations and 0.5 g Maple leaves posed higher removal for Pb (II) ions. A sharp increase in bio-sorption occurred in the pH range 2.5-4.5. The maximum bio-sorption was 98.5% for Pb(II) ions at pH 6.3. Smaller particles (<75 µm) have greater Pb (II) removal capacity.	(Hossain et al. 2014)
5	Self-purification of the marine environment for heavy metals: A study on removal of lead and copper by cuttlebone.	Self-purification marine environment done by the cuttlefish to remove lead and copper. In this experiment, cuttlebone from cuttle fish was collected and washed with tap and deionized water and dried at 105°C for 24hrs and grinded to particles with sizes between 0.3-0.7mm. The solutions of lead and copper are prepared by dissolving the required amount of lead nitrate and copper nitrate Pent hydrate in de-ionized water. The pH values were modified and calibrated by using 0.1-1.0 M HCl and 0.1-1.0 M NaOH. Experiment was conducted in batch reactor on shaker at 120rpm at room temperature. The pH is maintained at 5 initially. Experiments were done with metal concentrations of 10, 20, and 50 mg/L and the adsorbent dose of 0.1 to 1.0 g/L with constant ion concentration of 50 mg/L initially. The adsorption was studied by keeping the range of pH between 2-7. Later, the concentration of metals was analysed by using AAS.	The results were that the capacity of cuttlebone to adsorb Pb and Cu were found to be 45.9 mg/g and 39.9 mg/g. The adsorption of Pb and Cu increased with an increase in pH and the max adsorption occurred when the pH was 7.0 and the adsorption was found to be 18.7 mg/g and 19.8 mg/g for Pb and Cu. And the maximum uptakes were found when at the pH values 5.0 and 5.5.	(Dobaradaran, Nabipour et al. 2017)

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S. No.	Objective	Brief Summary	Outcome	References
6	Adsorptive removal of Arsenic from aqueous solution by waste litchi pericarps.	Litchi pericarps are washed with tap water and ultrapure water. They are dried at 70°C and then sized through a 60 mesh and stored in a polyethylene bottle. A stock solution of sodium arsenate NaAsO ₂ (1,000 mg/L) was prepared with ultrapure water. Polyethylene tubes of 100 mL are taken with 50 mL of aqueous arsenic solution. Different adsorption capacities are observed with LPs (1–20 g/L) with varying initial arsenate concentration (0.01–100 mg/L) at 293.15 K. The pH is maintained between 2–11 by using 0.1 M HCl and 0.1 M NaOH solutions. The polyethylene centrifuge tubes were shaken in a vertical temperature oscillation incubator at 220 rpm for 5 to 180 min. These suspensions are filtered through 0.45 µm “cellulose acetate membrane filters”. Arsenic filtrates are observed through AAS.	The removal rate increases from 24.0% to 97.74% as the adsorbent dose increases from 1.0 to 10.0 g/L. No further increase in the removal rate of As (III) is observed for LP additions from 10.0 to 20.0 g/L. Arsenic removal efficiency increases from 88% to 93% as the contact time increases from 10 to 60 min. The percentage removal rate increases by an increase in pH from 2 to 5, and then it decreases slightly with a further pH increase from 6–7.	(Li, Qi et al. 2016)
7	“Arsenic removal from aqueous solutions by adsorption on red mud”.	Arsenic is removed from water by using red mud as an adsorbent. In this paper it is mentioned that mud was wet sieved through a 200 mesh screen and was washed for five times with distilled water. The suspension was filtered and was dried at 105°C, ground and sieved through a 200 mesh screen. Stock solutions of 1 As (III) were prepared by adding 1.320 g of As ₂ O ₃ in 10 mL of 5M NaOH and made it up to 1 L with distilled water. Another stock solution of 1 g/L As(V) was prepared by adding Na ₂ HPO ₄ ·7H ₂ O. Solutions with 125 to 1500 mg of arsenic and 5 mL of 0.1 M NaCl were made to 50 mL using distilled water. The red mud was added as an adsorbent into the solution and shaken at the rate of 800 cycles/min with a mechanical shaker. Later, the mixture was centrifuged at 10000 rpm for 10 mins	As (III) and As (V) adsorptions are obtained within 45 and 90 min respectively at 25°C, 133.5 µmol/L concentration and 20 g/L red mud dosage. As(III) and As(V) adsorptions take places at pH 9.5 and 3.2. The adsorption densities at these conditions are 4.31 and 5.07 µmol/g for As(III) and As(V).	(Altundo an et al. 2000)
8	Removal of Pb(II) and Cd(II) from water by adsorption on peels of banana.	Banana peels were dried in sunlight for 5 days and in an oven at 70°C. The dried peels were sieved through 60 mesh screen. Standard solutions with desired concentrations (10–100 µgm/L) of lead nitrate and cadmium nitrate were prepared. Six solutions with concentrations of 30, 40, 50, 60,70 and 80 µgm/L of lead and cadmium were made with pH adjusted to 5 for lead and 3 for cadmium. Then adsorbent of 2.0 g for lead and 1.5 g for cadmium were added 50 mL of each solution which was agitated for half an hour. Later the concentration of the metals was analysed by AAS.	From the results, it was observed that banana peels were effective in removing cadmium compared to lead. 1 g of banana peels adsorbed 5.71 g of cadmium and 2.18 g of lead. Max adsorption of cadmium was observed at pH 3 and for the lead at pH 5.	(Anwar et al. 2010)

lead from water. Orange peels were collected from the juice shops in Dehradun, India. They were washed with water for removal of any dirt and dried in a hot air oven for 4 h at 80°C and 6 h at 105°C (total of 10 hours). They were cut into pieces using a grinding machine and sieved to various sizes like 250 µm, 500 µm and 850 µm for the experimentation.

Preparation of Stock Solution

1000 ppm stock solution was prepared and diluted with distilled water to make standard solutions of 5 ppm, 10 ppm,

15 ppm, 20 ppm and 25 ppm. The experiment was carried out using 10 ppm and 15 ppm stock solutions and rest were used as standards in atomic absorption spectroscopy.

Equipment and Chemicals Used

pH was checked by using a digital pH meter. The metal solution was taken and the exact selected size and dose of adsorbent was added and stirred using a magnetic stirrer at 600 rpm for 30 min. The metal solution was given 2-h time for settling and filtered using a Waterman Filter paper of size

400 micron. The pH was adjusted to the required value (4, 6, 8) using 0.1 N HCl or 0.1 N NaOH solutions.

Size of adsorbent: 250 μm , 500 μm , 850 μm

Dose: 0.5 g, 1 g and 2 g

Study of Process Parameters

Effects of various parameters like adsorbent particle size, pH and dose were studied by keeping agitation speed at 600 rpm for 30 min constant. For studying the effect of one parameter, the other three parameters were kept constant. Standard values of various heavy metals are mentioned in Table 2.

RESULTS AND DISCUSSION

It is observed that the removal efficiency increases with increase in pH and is maximum at pH 7 and started decreasing from pH 8. The removal efficiency was tested at three different pH values starting from pH 4, pH 7 and pH 8. Anwar et al. (2010) mentioned that the pH of the stock solution is one of the most important factors that decides the extent of adsorption of heavy metal ions. As shown in Fig. 1, adsorp-

tion efficiency increases from pH 4 to pH 7 which is acidic and declined after pH 7 which is optimum pH. At lower pH, the hydrogen ions (H^+) compete with heavy metal cations for active adsorption sites. But beyond the optimum pH, the adsorption sites will not be activated and metal cations will react with (OH^-) ions and form metal hydroxide precipitate which makes adsorption uncertain (Anwar et al. 2010).

It was observed that at 0.5 g as the adsorbent dose 250 μ size and pH 7, the maximum metal removal efficiency was observed, i.e. (97.87%) (Fig. 2). As the adsorbent size increases, the available surface area for adsorption decreases and its available active sites decrease, thereby decreasing the metal removal efficiency (Fig. 3).

Same experiments were carried out by preparing 15 ppm lead concentration stock solution in 50 mL distilled water.

It was observed that for 10ppm metal concentration stock solution at 0.5 g as adsorbent dose and 250 μ size and pH 7, the maximum metal removal efficiency was 97.87%, and for 15ppm metal concentration at 0.5 g as adsorbent dose and 250 μ size and pH 7, the maximum metal removal efficiency was 96.57% (Fig. 4). As the adsorbent size increases,

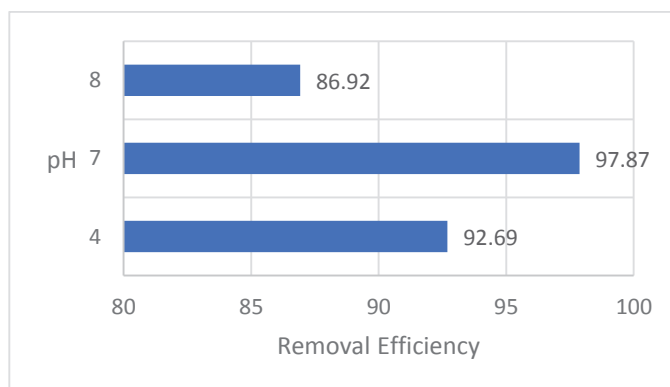


Fig. 1: Removal Efficiency with respect to varying pH at 10ppm lead concentration, adsorbent size-250 μm and adsorbent dose-0.5g.

Table 2: Drinking water standards: IS 10500:2012.

S. No.	Element	Acceptable Limit $\mu\text{g/L}$ (BIS 10500:2012)	PEL $\mu\text{g/L}$ (BIS 10500:2012)
1	Arsenic	10	50
2	Cadmium	3	No relaxation
3	Chromium	50	No relaxation
4	Copper	50	1500
5	Iron	300	No relaxation
6	Lead	10	No relaxation
7	Mercury	1	No relaxation
8	Nickel	20	No relaxation

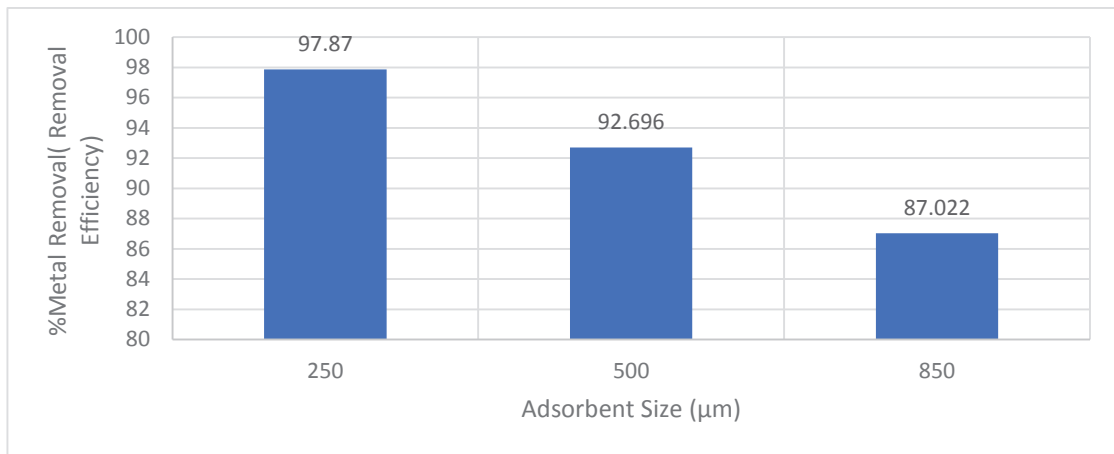


Fig. 2: Effect of adsorbent size for 10 ppm lead concentration at adsorbent dose-0.5g and pH 7.

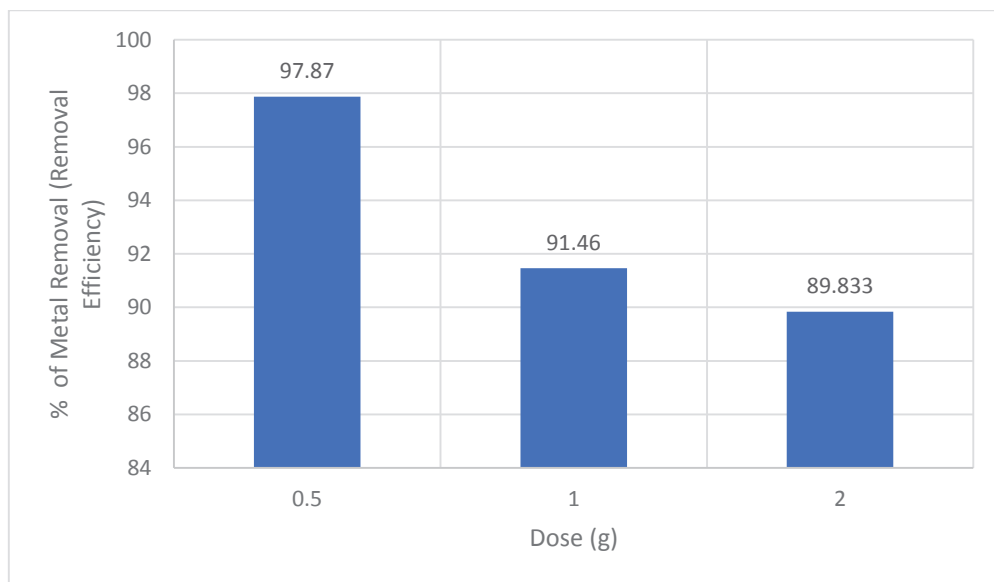


Fig. 3: Effect of adsorbent dose at adsorbent size-250µ and pH-7 on metal removal efficiency.

the available surface area for adsorption decreases and its available active sites decrease thereby decreasing the metal removal efficiency (Fig. 5).

CONCLUSIONS

A total of 20 samples were collected and tested by atomic absorption spectroscopy. The following observations were made.

1. The removal efficiency was found to mainly depend on adsorbent particle size, adsorbent dose and pH.
2. The RPM of the magnetic stirrer was kept constant throughout (600 RPM for 20 mins). Particles having

sizes in the range of 250-500 µm are expected to have greater adsorption tendencies as compared to larger particles because of the large surface areas of smaller particles allowing adsorption on the surface.

3. The optimum pH range can be between 5 and 7 for maximum adsorption of lead using orange peels. The minimum adsorption can be expected to occur at a pH of 3 because of its acidic nature. As the pH value increases, the adsorption capacity is also increased till a pH of 7 and it may remain stable in case of a further increase in pH.

From the above study it can be concluded that instead of using harmful chemicals for the removal of heavy metals

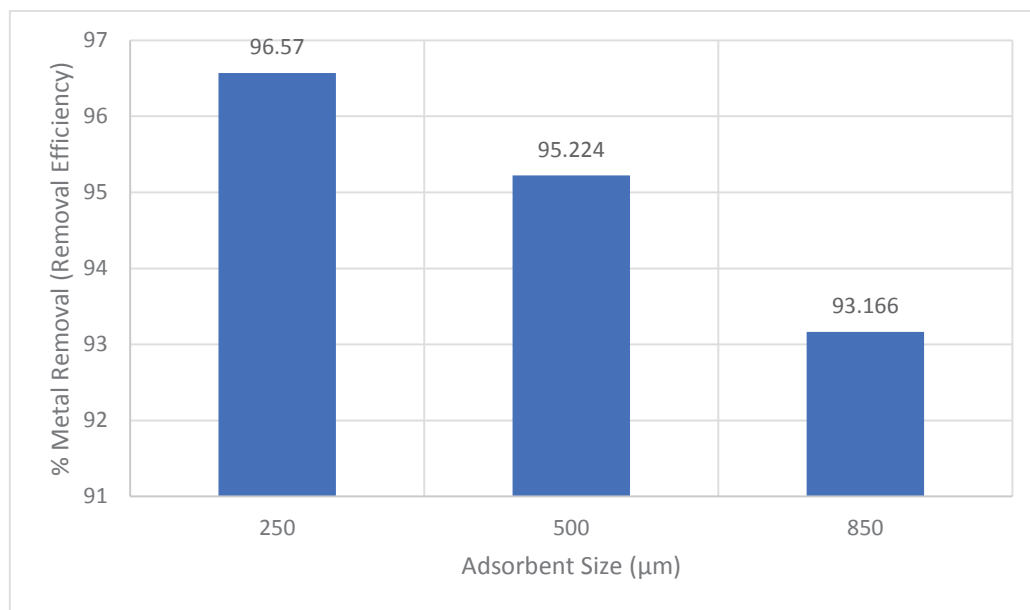


Fig. 4: Effect of adsorbent size at stock solution concentration of 15ppm at pH 7 and adsorbent dose of 0.5g.

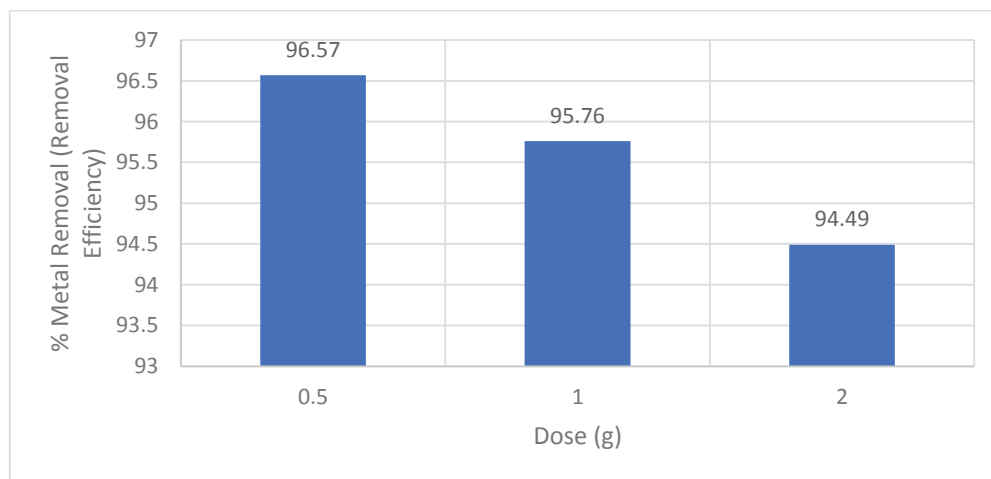


Fig. 5: Effect of adsorbent dose at adsorbent size-250 μ and pH-7 on metal removal efficiency at 15 ppm concentration.

from water, organic wastes such as orange peels, banana peels and rice husk can be used for the same. Detailed analysis of experimental data has been carried out for maximum adsorption capacity. It was found that maximum adsorption efficiency is at pH 7.

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