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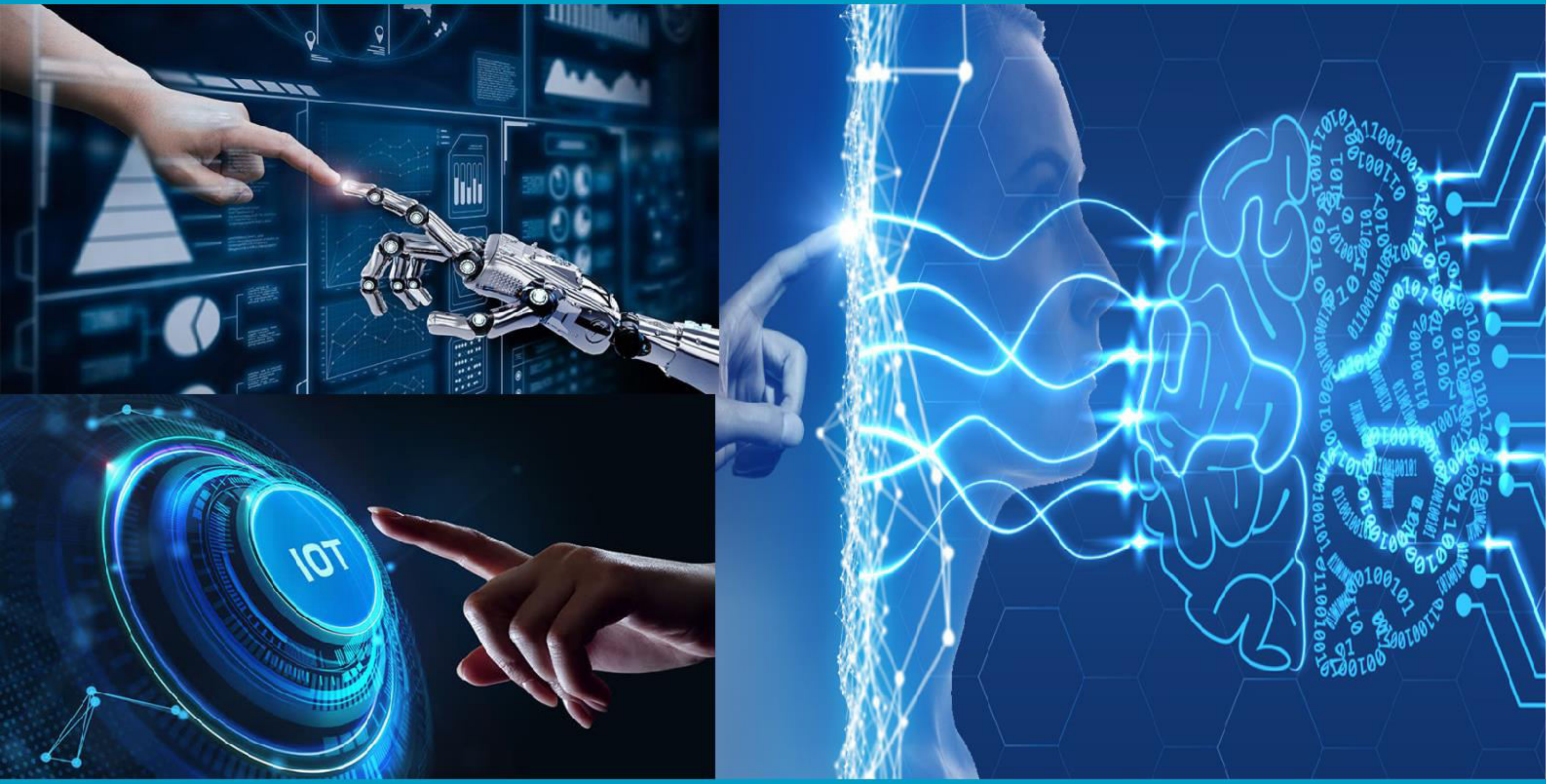

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Biosorption of Heavy Metals Using Rice Milling By-Products, Characterization and Application for Removal of Metals from Laboratory Prepared Waste Water

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ABSTRACT**-** The importance of removal of heavy metals from drinking water has received significant attention of researchers and decision makers across the globe. Bio-adsorbents have emerged as potential remediation materials for the removal of heavy metals and metalloids from both groundwater and surface water. Plants, algae, fungi are some of the biomass derived adsorbents which are capable of removing heavy metals and metalloids from aqueous solution by adsorption. For increasing the adsorption efficiency of bio-adsorbents, treatment of adsorbents is done by using various kinds of treating agents like tartaric acid, NaOH. The bio-adsorbents have affinity for heavy metal ions to form metal complexes or chelates due to having functional groups including carboxyl, hydroxyl, imidazole, sulphydryl, amino, phosphate, sulfate, thioether, phenol, carbonyl and amide etc. Rice husk (RH) is a low cost (agricultural by-product) bio-adsorbent which has been studied intensively for the removal of various heavy metals and metalloids (such as Pb, Cd, Zn, Ni and As) from both groundwater and surface water. The present study is focused on critical review of previous and current available information on potential of treated and untreated rice husk for the removal of heavy metals and metalloids (arsenic). Various studies on adsorption efficiency of rice husk considering the parameters contact time, adsorbent dose (rice husk), initial concentration of heavy metals, pH, and temperature have been evaluated by many researchers. The present study analyzed those studies and compiled the adsorption efficiency of rice husk and concluded that treated rice husk gave comparatively better adsorption efficiency of heavy metals with compared to that of untreated rice husk. The treated rice husk can be implemented on large scale industrial applications after field studies.

KEYWORDS- Adsorption Efficiency, Rice Husk, Bio-adsorbents, Drinking Water, Heavy Metals

I. INTRODUCTION

Biosorption can be defined as an ability of certain biomass to bind and concentrate heavy metals from even dilute aqueous solutions. In particular, the cell wall structure exhibits this property (Ahluwalia and Goyal 2007). Biosorption involves a solid sorbent, liquid phase, and the dissolved species to be sorbed. Biosorption can be defined as a fast process independent of energy on which biological materials or biopolymers acting as sorbents have the ability to remove pollutants, such as heavy metals from wastewater through metabolically mediated or physico-chemical pathways of uptake. With rapid development in agriculture, industry, commerce, hospital and healthcare facilities, many activities are consuming significant quantities of toxic chemicals and generating a large amount of hazardous waste. Currently, there are about 110 000 types of toxic chemicals commercially available. Each year, another 1 000 new chemicals are added into the market for industrial and other uses. One of the most hazardous pollutants in environment is heavy metals (Sud et al., 2008).

Heavy metals are elements having atomic weights between 63.5 and 200.6, and a specific gravity greater than 5.0 (Fu and Wang, 2011). Heavy metals generally refers to the elements such as Cd (cadmium), Cr (chromium), Cu (copper), Hg (mercury), Ni (nickel), Pb (lead), Fe (ferum) and Zn (zinc) which are commonly associated with pollution and toxicity problems. Heavy metals occur naturally in rock formation and ore minerals and so a range of normal background concentration is associated with each of these elements in soils, sediments, waters and living organisms. These heavy metals are of specific concern due to their toxicity, bio-accumulation tendency and persistency in nature. Several past disasters due to the contamination of heavy metals in aquatic streams are Minamata tragedy in Japan due to methyl mercury contamination and „„Itai-Itai" due to contamination of cadmium in Jintsu river of Japan (Sud et al., 2008).

Industrial uses of metals and other domestic processes have introduced substantial amounts of potentially toxic heavy metals into the atmosphere and into the aquatic and terrestrial environments. In small quantities, certain heavy metals are nutritionally essential for healthy life. Some of these are referred to as the trace elements (e.g., iron, copper, manganese, and zinc). These elements, or some form of them, are commonly found naturally in foodstuffs, in fruits and vegetables, and in commercially available multivitamin products. Heavy metals are also common in industrial applications such as in the manufacture of pesticides, batteries, alloys, electroplated metal parts, textile dyes, steel, mining, refining ores, fertilizers industries, paper industries and so forth (Sud et al., 2008). Many of these products are in our homes and actually add to our quality of life when properly used.

II. LITERATURE REVIEW

[1] Application of chemically modified rice husk for the removal of heavy metals from aqueous solution (2010)- The removal efficiency of lead, cadmium and zinc from aqueous solution on adsorption by using rice husk, a non-conventional material in its natural and chemically modified form has been presented in this paper. It has been observed that rate of adsorption is dependent on the nature of the adsorbent, adsorbent dose, particle size of the adsorbent, concentration, pH, contact time, temperature, etc. Under identical experimental condition chemically modified rice husk was found to possess greater adsorption capacity for all metals than untreated rice husk and chemically modified rice husk ash. Chemically modified rice husk could remove 99.8% Pb, 95% Cd and 97% Zn from aqueous solution at room temperature.

[2] Adsorption of lead and mercury by rice husk ash (2004)- An attempt at the use of rice husk ash, an agricultural waste, as an adsorbent for the adsorption of lead and mercury from aqueous water is studied. Studies are carried out as a function of contact times, ionic strength, particle size, and pH. Rice husk ash is found to be a suitable adsorbent for the adsorption of lead and mercury ions. The Bangham equation can be used to express the mechanism for adsorption of lead and mercury ions by rice husk ash. Its adsorption capability and adsorption rate are considerably higher and faster for lead ions than for mercury ions. The finer the rice husk ash particles used, the higher the pH of the solution and the lower the concentration of the supporting electrolyte, potassium nitrate solution, the more lead and mercury ions absorbed on rice husk ash. Equilibrium data obtained have been found to fit both the Langmuir and Freundlich adsorption isotherms.

[3] Use of rice straw as biosorbent for removal of Cu(II), Zn(II), Cd(II) and Hg(II) ions in industrial effluents (2009)- Adsorption experiments were carried out using waste rice straw of several kinds as a biosorbent to adsorb Cu(II), Zn(II), Cd(II) and Hg(II) ions from aqueous solutions at room temperature. To achieve the best adsorption conditions the influence of pH and contact time were investigated. The isotherms of adsorption were fitted to the Freundlich equation.
Based on the experimental data and Freundlich model, the adsorption order was Cd(II)>Cu(II)>Zn(II)>Hg(II) on the rice straw. This quick adsorption process reached the equilibrium before 1.5h, with maximum adsorptions at pH 5.0. Thermodynamic aspects of the adsorption process were investigated. The biosorbent material was used in columns for the removal of ions Cu, Zn, Cd and Hg of real samples of industrial effluent and its efficiency was studied.

[4] Adsorptive removal of Cd(II) from aqueous solution using natural and modified rice husk (2010)- In this study, the natural and modified rice husk were tested to remove Cd (II) ions from water. The modified rice husk was prepared by being treated with alkali. The results showed the Cd (II) adsorption capacity was 73.96, 125.94 mg/g, respectively, for the natural and modified rice husk. The modified rice husk had faster kinetics and higher adsorption capacities than the natural rice husk, which can be attributed to the surface structural changes of the material. Equilibrium adsorption data are more consistent with the Langmuir isotherm equation than with the Freundlich equation. The Cd(II) adsorption on the two adsorbents tends to increase with the increase of pH. The optimum pH for Cd(II) adsorption is 6.5. Both pseudo-first-order and pseudo-second-order equations were able to describe properly the kinetics of Cd(II) adsorption. The desorb ability of Cd(II) is about 95.8-99.1% by 0.1M HCl solution.

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[5] Recent advances on pollutants removal by rice husk as a bio-based adsorbent: A critical review (2019)- Rice husk is an attractive bio-based adsorbent material for pollutant removal since it is one of the low-cost and renewable resources. The objective of this review is to give a summary of the key scientific features related to pollutants removal using rice husk, with a specific emphasis on the effect of factors on adsorption capacity of rice husk. According to the results, rice husk has the removal potential of various pollutants and it can be more used in the wastewater treatment. On the other hand, untreated bio-based adsorbent in large-scale application can usually cause some difficulties and selection of appropriate pretreatment method for rice husk is also one of the major challenges. Therefore, this review studies different pretreatment methods as well as regeneration of adsorbent and the fate of adsorbed contaminants.
According to the literature, pretreatment methods increase the rice husk capability and adsorption capacity and the chemical treatments have been more used than thermal treatments. Also, regeneration of rice husk adsorbent and adsorbed contaminants is applicable. Finally, examples of some applications and possibility of biocatalyst immobilization on the rice husk as a promising approach are presented. Results confirmed that rice husk has an excellent prospective potential for biocatalysts immobilization.

III. PROPOSED METHODOLOGY

3.1 MATERIALS REQUIRED

1. Rice Husk

Rice Husk is collects from Hariom Rice Mill, Nagpur, Maharashtra, India. Rice hulls are the hard protecting coverings of grains of rice. In addition to protecting rice during the growing season, rice hulls can be put to use as building material, fertilizer, insulation material, or fuel. Rice hulls are part of the chaff of the rice. Rice hulls provide a dense source of carbon which, when composted, can increase the water and nutrient holding capacity of soil, improve soil aggregation, porosity, infiltration, and many other key beneficial soil physical characteristics. Rice husk ash (RHA) fillers are derived from rice husks, which are usually regarded as agricultural waste and an environmental hazard. Rice husk, when burnt in open air outside the rice mill, yields two types of ash that can serve as fillers in plastics materials.

3.2 REAGENTS AND SAMPLES

4.2.1 Sample-1: Grounded rice husk

Rice husk shown in is obtained from rice mills. To prepare the powdered husks, they are initially ground and homogenised using a food blender with steel blades for 10 min. Particle sizes <355µm were obtained by passing the milled material through a steel sieve shown in fig.4.1. Afterwards, the ground husks are stored in polyethylene bottles (high density) and used without any other physical or chemical treatment. The experiments are to be carried out in conical flask. (C.R. Teixeira Tarley,M.A. Zezzi Arruda, Biosorption of heavy metals using rice milling by-products, Chemosphere, 54(2004): pp.988) The wastewater used is prepared synthetically in laboratory by taking 100ml solution of Cu, Pb, Zn with varying concentration from 1ppm to 20ppm and, finally, solutions of metals used for calibration procedures in atomic absorption spectrometry.

[Fig.4.1: 355 µm sieve, used for sieving sample (powdered rice husk)]

4.2.2 Sample-2: Carbonized rice husk

Rice husk was washed 3-4 times with de-ionized water to remove all dirt in its original particle size followed by filtration and were oven dried at 100°C. The cleaned and dried rice husk was then kept inside the muffle furnace (shown in fig.2) at 500°C for 3 hours. The burned rice husks were soaked in 0.6M of citric acid for two hours at 20 degree temperature. The acid slurry is then oven dried at 50 degree and then the product was cleaned and dried and used without any other further treatment (I Nhapi, N Banadda, R Murenzi et.al., Removal of Heavy Metals from Industrial Wastewater Using Rice Husks, The Open Environmental Engineering Journal, 2011, 4: pp 172). It is called Carbonized Rice Husks (CRH) shown in fig.4.2.

[Fig.4.2: Rice husk after treatment]

3.3 EXPERIMENTAL SETUP

For these experiments 100 ml of a solution containing Cu(II), Zn(II), and Pb(II) at 1ppm to 20ppm concentrations is to be added with the adsorbent and stirred continuously at 250 rpm speed in a electromagnetic stirrer for 24 hours at 40 degree constant temperature. Then the sample is allowed for settlement till clear water is seen on the surface, the sample the filtered and final concentration of metals is measured from the analysis using a Perkin-Elmer Model Analyst 200 atomic adsorption spectrometer. The experimental parameters affecting the bioaccumulation of Cu (II), Zn and Pb (II) species are examined. The effect of pH on the ability of rice husks to adsorb metal ions was investigated. For this purpose, the pH values of the Cu (II), Zn and Pb (II) solution are varied from 2 to 6. In order to evaluate the treatment efficiency for other metals, after establishing the optimal conditions for Cu, Zn and Pb(II) laboratory effluent treatment. Thus, the initial and final concentrations of these metals are also determined, and the results are recorded.

IV. RESULTS

4.1 ADSORPTION ON GROUNDED RICE HUSK

A series of adsorption experiment were conducted to establish the isotherms for Pb, Cu, Zn adsorption on grounded rice husk. This section presents the results result of the adsorption isotherm of different metal with grounded rice husk.

1. Zn

Table 5.1: Isotherms for Zn adsorption on grounded rice husk

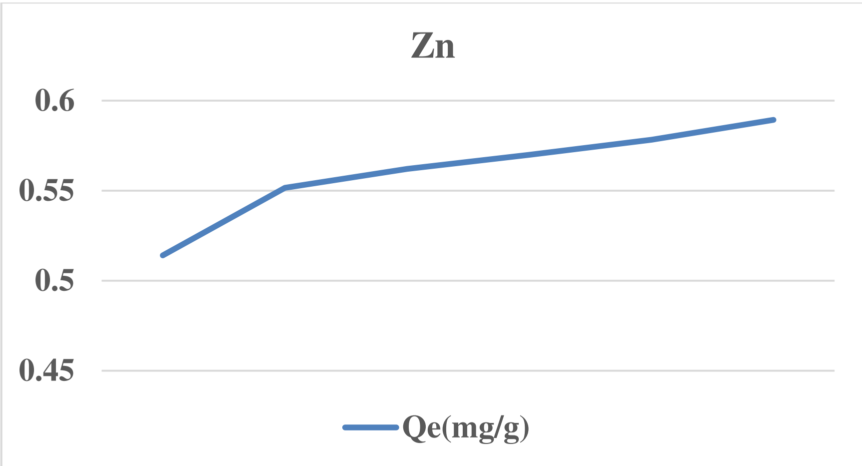
Time(hr)	Initial Concentration (mg/l)	Final Concentration (mg/l)	Qe(mg/g)	% Removal
0.5	6.248	1.101	0.514	82.37
1	6.246	0.729	0.5517	88.32
2	6.210	0.589	0.5621	90.51
3	6.162	0.463	0.5699	92.48
5	6.153	0.370	0.5783	93.98
18	6.063	0.170	0.5893	97.17

The amount of metal absorbed per unit mass is calculated as:

Qe=(Ci-Cf)V/m

Where Ci and Cf are the initial and final concentration (mg/l),m is mass of adsorbent, V is the volume of the solution (m3).

With increase in time or duration of treatment of effluent the adsorption capacity of the material used shows an increase. After long duration around 48 hours the there will be a negligible amount of heavy metals left in the solution.



[Fig.5.1: Zn vs time for grounded rice husk]

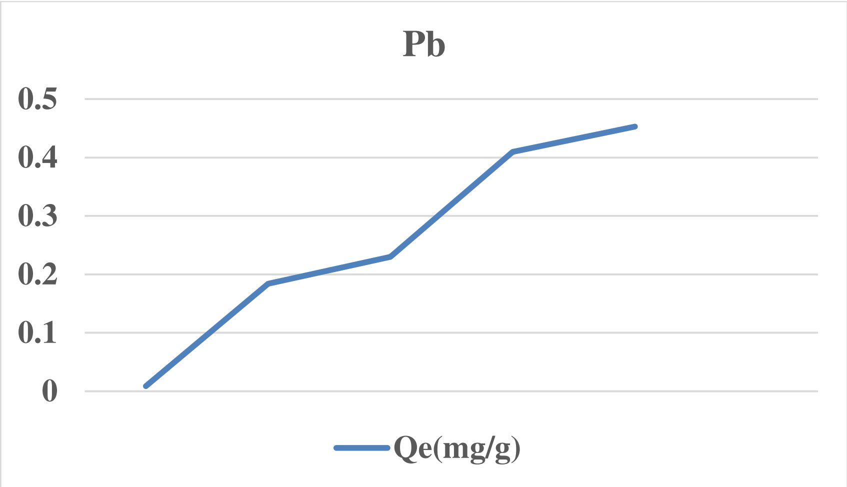
Table 5.2: Zn on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)

Initial Concentration (mg/l)	Final Concentration (mg/l)	Qe(mg/g)	% Removal
1.033	0.135	0.089	86.93
2.589	0.144	0.2445	94.43
3.079	0.028	0.3051	99.09
3.733	0.14	0.359	96.24
4.778	0.773	0.4005	83.82

2. Pb

Table 5.3: Pb on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)

Initial Concentration (mg/l)	Final Concentration (mg/l)	Qe(mg/g)	% Removal
0.179	0.093	0.0086	48.044
2.348	0.508	0.184	78.36
2.567	0.267	0.230	89.59
4.212	0.198	0.4095	95.29
4.729	0.117	0.4531	97.52

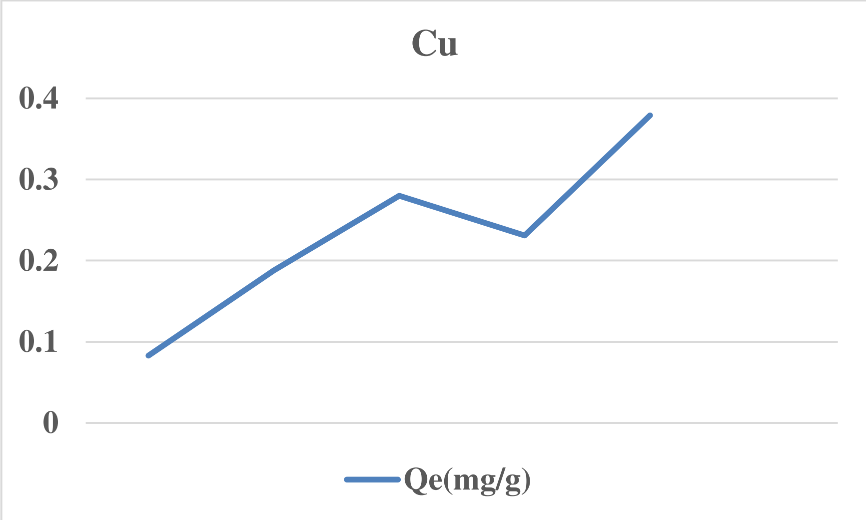


[Fig.5.2: Effect of initial concentration of Pb on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)]

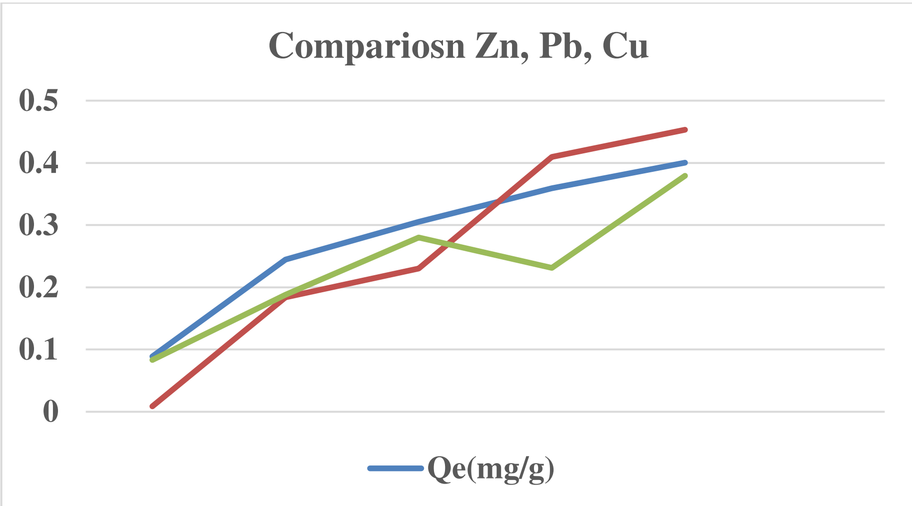
3. Cu

Table 5.4: Cu on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)

Initial Concentration (mg/l)	Final Concentration (mg/l)	Qe(mg/g)	% Removal
0.889	0.059	0.083	93.36
2.307	0.420	0.188	81.79
3.271	0.468	0.280	85.69
3.624	1.312	0.231	63.79
4.762	0.969	0.3793	79.65



[Fig.5.3: Effect of initial concentration of Cu on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)]



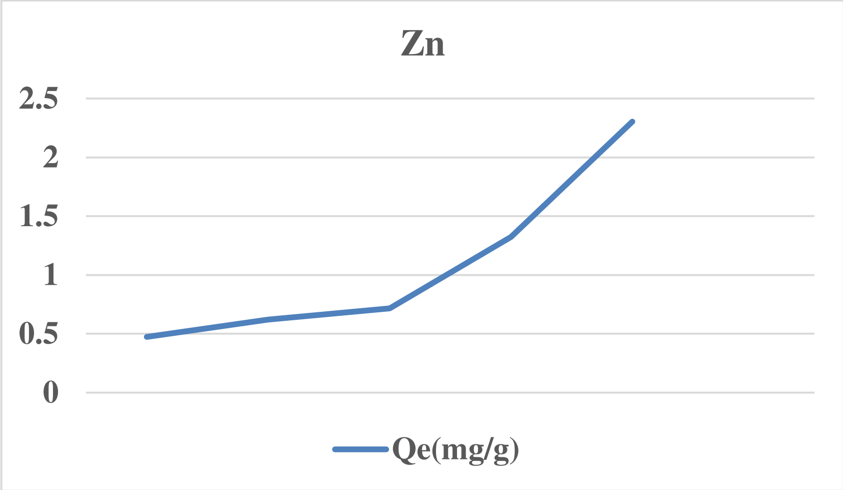
[Fig.5.4: Effect of initial concentration of metals Zn, Cu, Pb on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)]

5.2 ADSORPTION ON CARBONIZED RICE HUSK

1. Zn

Table 5.5: Zn on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)

Initial Concentration (mg/l)	Final Concentration (mg/l)	Qe(mg/g)	% Removal
5.38	0.65	0.473	87.91
6.93	0.72	0.621	89.61
7.89	0.74	0.715	90.62
14.02	0.78	1.324	94.43
23.85	0.80	2.305	96.64

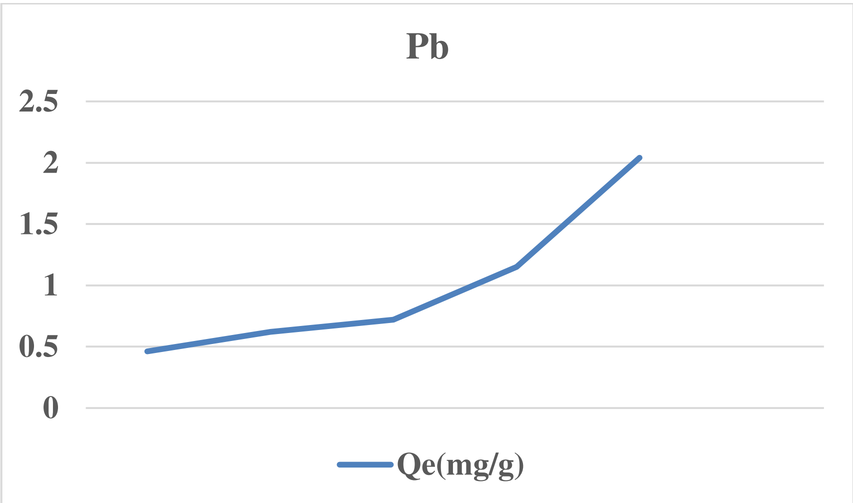


[Fig.5.5: Effect of initial concentration of Zn on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)]

2. Pb

Table 5.6: Pb on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)

Initial Concentration (mg/l)	Final Concentration (mg/l)	Qe(mg/g)	% Removal
5.3	0.68	0.462	87.16
6.9	0.7	0.62	89.85
7.92	0.72	0.72	90.78
12.23	0.73	1.15	94.03
21.15	0.75	2.04	96.45

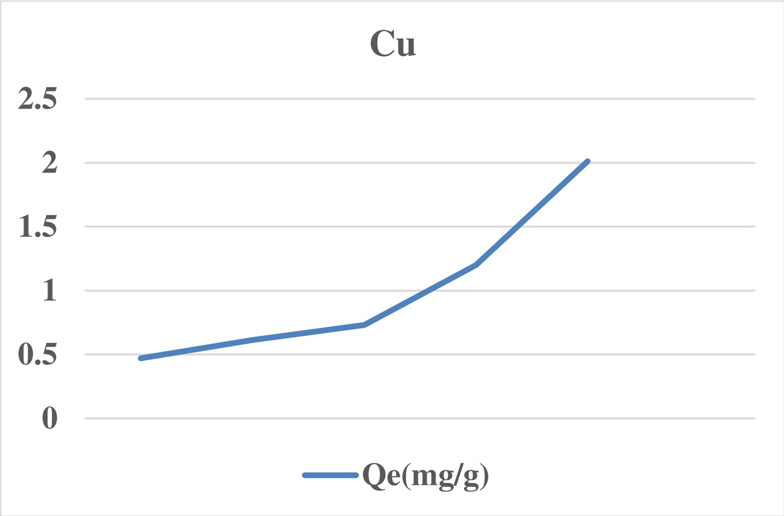


[Fig.5.6: Effect of initial concentration of Pb on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)]

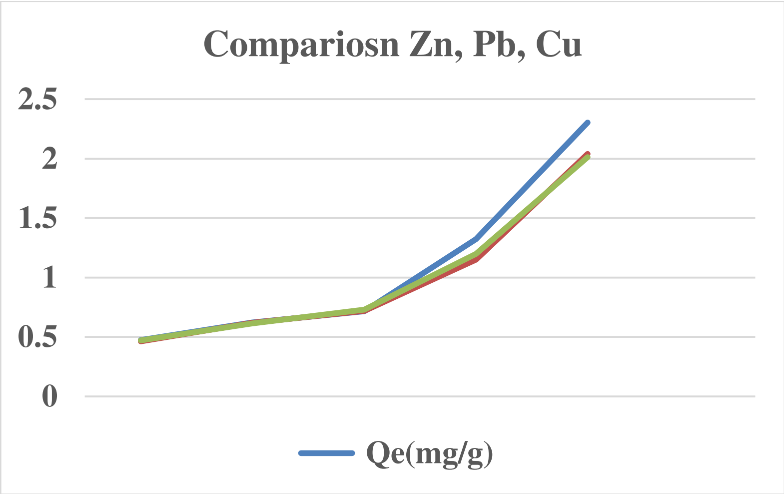
3. Cu

Table 5.7: Cu on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)

Initial Concentration (mg/l)	Final Concentration (mg/l)	Qe(mg/g)	% Removal
5.43	0.73	0.47	86.55
6.87	0.74	0.613	89.22
8.05	0.75	0.73	90.68
12.76	0.76	1.20	94.04
20.89	0.77	2.012	96.31



[Fig.5.7: Effect of initial concentration of Cu on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)]



[Fig.5.8: Effect of initial concentration of metals Zn, Cu, Pb on the adsorption capacity of grounded rice husks (mass of rice husk: 1.0g; particle size<355 micron)]

V. CONCLUSION

The chemical characterization obtained from the analysis of scanning electron microscope of rice husk shows carbon and silica content in the grounded rice husk as 47.28% and 3.73%, respectively. The results show carbon, oxygen and silica content in carbonized rice husk as 8.52%, 36.27% and 43.41%, respectively. After the chemical characterization, the material was used for removing Zn, Cu and Pb from laboratory prepared effluents. Under certain optimized conditions such as constant temperature of 40 degree centigrade, speed of stirring action as 250rpm for 24 hours, 1 g of adsorbent per each 100ml solution of heavy metals, the results obtained are summarized as:

1. The adsorption capacity of grounded rice husk decreases in order Pb > Zn > Cu.
2. Longer duration of time gives high values of adsorption capacity.
3. High initial concentration of metals mostly gives high values of adsorption capacity.
4. The adsorption capacity of carbonized rice husk decreases in order of Zn > P b > Cu.
5. Carbonated rice husk and grounded rice husk demonstrated higher potential to remove relatively all selected heavy metals.
6. After 48 hours it will leave a negligible amount of metals in the treated water.
7. The efficiency of both the adsorbents in the removal of desired heavy metals was 95-100%, or nearly 100%.
8. The results show the removal efficiency of grounded rice husk for Zn, Pb and Cu as 99.09%, 97.52% and 93.36 %, respectively.
9. The removal efficiency of carbonized rice husk for Pb, Cu and Zn is found to be 96.45%, 96.31% and 96.64%, respectively. Increase in the duration of stirring action the removal efficiency of the adsorbent material increases. As per the observation, removal of Zn from 5ppm solution of Zn for 0.5,1,2,3,5,18 hours are 82.37%, 88.32%,90.51%,92.48%,93.98% and 97.19%, respectively.

VI. FUTURE SCOPE

The importance of removal of heavy metals from drinking water has received significant attention of researchers and decision makers across the globe. Bio-adsorbents have emerged as potential remediation materials for the removal of heavy metals and metalloids from both groundwater and surface water. Plants, algae, fungi are some of the biomass derived adsorbents which are capable of removing heavy metals and metalloids from aqueous solution by adsorption. For increasing the adsorption efficiency of bio-adsorbents, treatment of adsorbents is done by using various kinds of treating agents like tartaric acid, NaOH. The bio-adsorbents have affinity for heavy metal ions to form metal complexes or chelates due to having functional groups including carboxyl, hydroxyl, imidazole, sulphydryl, amino, phosphate, sulfate, thioether, phenol, carbonyl and amide etc. Rice husk (RH) is a low cost (agricultural by-product) bio-adsorbent which has been studied intensively for the removal of various heavy metals and metalloids (such as Pb, Cd, Zn, Ni and As) from both groundwater and surface water.

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