Adsorption Studies on Polymer Matrix-Mixed Metal (Fe-Mn-Zn) Oxide Nanocomposite for Removing Zn (II) ions from Contaminated Water.

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ABSTRACT

A huge number of contaminants are discharged in the environment by various industries. The water bodies are the major receivers of the contaminants. The major water contaminants are of biological, chemicals and physical types. Some of these contaminants include nitrogen, bleach, salts, pesticides, metals, toxins produced by bacteria, and human or animal drugs. Heavy metals contaminants in water are of major concern due to their serious effect on the health of living beings. Various methods like flocculation, ion exchange, precipitation, membrane filtration, reverse osmosis, adsorption, electrolyte removal, and reduction are used to purify contaminated water. In treating the contaminated water, adsorption processes have attained significant maturity due to lower cost, easy operation and environmental friendliness. In the present study we have employed polymer magnetic nanocomposites of mixed oxides of metals (Fe-Mn-Zn) to treat contaminated water for removal Zn (II) ions using the principle of adsorption. We report here the percentage removal of Zn ions with adsorbent dose, with contact time, with pH and initial concentration of adsorbate ions. The adsorption data are validated using Langmuir and Freundlich isotherms models. The surface area and porosity of sample was analyzed by BET analysis.

Keywords: Polymeric magnetic nanocomposites, heavy metals, adsorption, Langmuir and Freundlich isotherms, metal oxides.

INTRODUCTION

Fresh water is of top importance to human life. However, as reported about 70% of Earth's surface is water of which 97.5% is salty water and 2.5% is fresh water. Less than 1%of th is 2.5% amount of freshwater is accessible on the Earth, which makes the clean water scarcity a worldwide problem.[1] Various hazardous chemicals produced by various industries like Fertilizer, pesticides, chemical, textiles, petrochemical etc., are directly or indirectly introduced in water resources without following any proper safety measures.[2] Apart from the outspread of industrialization, the considerable release of pollutants into the surroundings is also caused by the people who are neglecting or unaware of the appropriate protocol for health and safety practices.[3] Contamination of fresh water by heavy metal is of major concern. The intake of heavy metals present in the contaminated water can cause serious health problems like failure of central nervous system, kidney, liver, pancreas, etc.[4] So, it is necessary to get rid of the heavy metals from waste water before discharge into water bodies.[5] Various techniques like flocculation, ion exchange, precipitation, membrane filtration, reverse osmosis, adsorption, electrolyte removal, and reduction are used to purify contaminated water.[6-9] Among these adsorption techniques is preferred due to its easiness and versatility.[8] The various polymer and carbon based magnetic nanocomposites are used for water treatment.[3,10] In the present report we report the adsorption studies of Zn ions on mixed metal oxides of (Fe-Mn-Zn) in polymer matrix of aniline-formaldehyde.

METHODOLOGY

To synthesize polymeric nanocomposites of mixed oxides, 10.0 mL (100 mmol) of aniline (Sigma Aldrich), 10mL (0.10 mol) formaldehyde (Merck), 12.0 mL (120 mmol) of concentrated HCl (Thermo Fischer),

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$FeCl_3.6H_2O$ (Otto Kemi), $MnCl_2.4H_2O$ (Otto Kemi), $ZnCl_2$ (Merk) and NaOH (Otto Kemi) were used. For the adsorption studies we have synthesized the polymeric mixed oxides nanocomposites with aniline (0.10 mol), concentrated hydrochloric acid (0.12 mol), 10 mL formaldehyde (0.10 mol), 50 mL of doubled distilled water along with metal ions concentrations as (0.04 mol) Fe, (0.0151 mol) Mn and (0.0039 mol) of Zn of respective salts. 1000 ppm stock solution of Zn (II) ion was prepared by using standard method as reported[11]. The details of synthesis process are reported elsewhere[12]. The synthesized material is used for adsorption studies and referred as sample in the whole study. The sample was shaken with the Zn (II) ion solution in a stoppered conical flask and the concentration of residual ions was determined using AAS (ECIL, AAS4141). The concentration of Zn (II) ion adsorbed, q_e (mg/L), was determined using following equation[13]

$$q_e = \frac{(C_0 - C_e)V}{W}$$

Where C_o and C_e are initial and equilibrium concentrations of adsorbate (mg/L), V is volume of solution (L) and W is mass of adsorbent (g). The percentage removal of Zn (II) ions was determined using following equation:

$$\%\ \text{removal} = \frac{(C_0 - C_e)}{C_0} \times 100$$

RESULTS AND DISCUSSIONS

Effect of adsorbent dose

Adsorbent dose study is an important parameter in adsorption studies that determines the capacity of the adsorbent for a given initial concentration of metal ion solution. To analyze the effect of dose on the adsorption 0.01 to 0.07 g of sample was taken. Increasing the amount of sample increased the percent removal of Zn (II) from 34.34% to 97.36% and 0.05 g resulted in the highest removal efficiency. The increase in removal efficiency was due to increase in number of free adsorption sites, however further increase in adsorbent dose resulted in aggregation of the adsorbent[14]. The variation of percentage removal of Zn (II) ions with adsorbent dose is given in figure 1.

Effect of contact time

The percentage removal efficiency of the Zn (II) ion increased as the contact time increased. The maximum percentage removal, 99.27% was achieved at 35 min with 0.05 g of sample and no significant improvement was observed after this contact time. This implies that further increase in contact time did not show significant rise in percentage removal either due to repulsive forces between the Zn (II) ions and sample or saturation of the binding sites making unoccupied binding sites unavailable[15]. The variation of percentage removal with contact time is shown in figure 2.

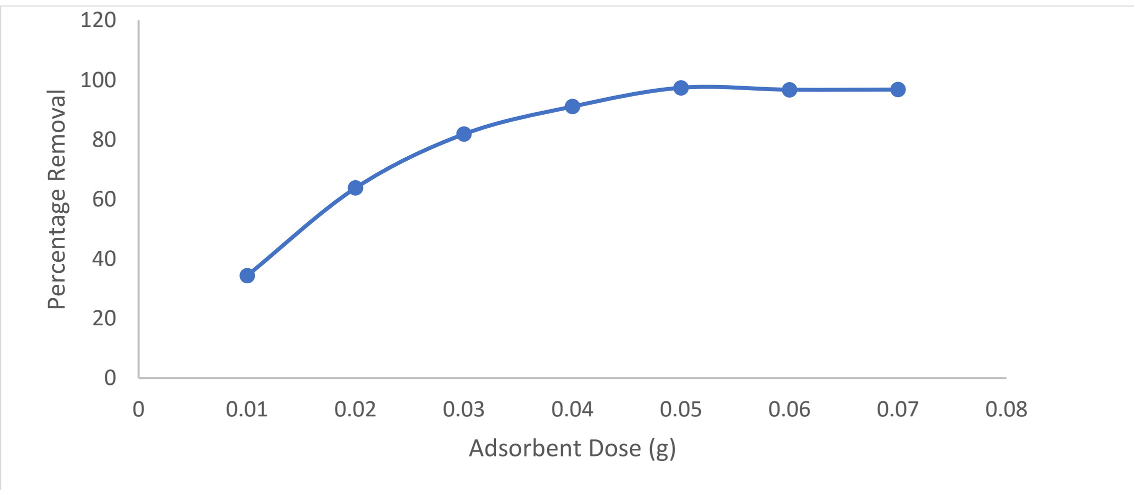

Figure 1: Variation of Adsorbent dose vs Percentage Removal

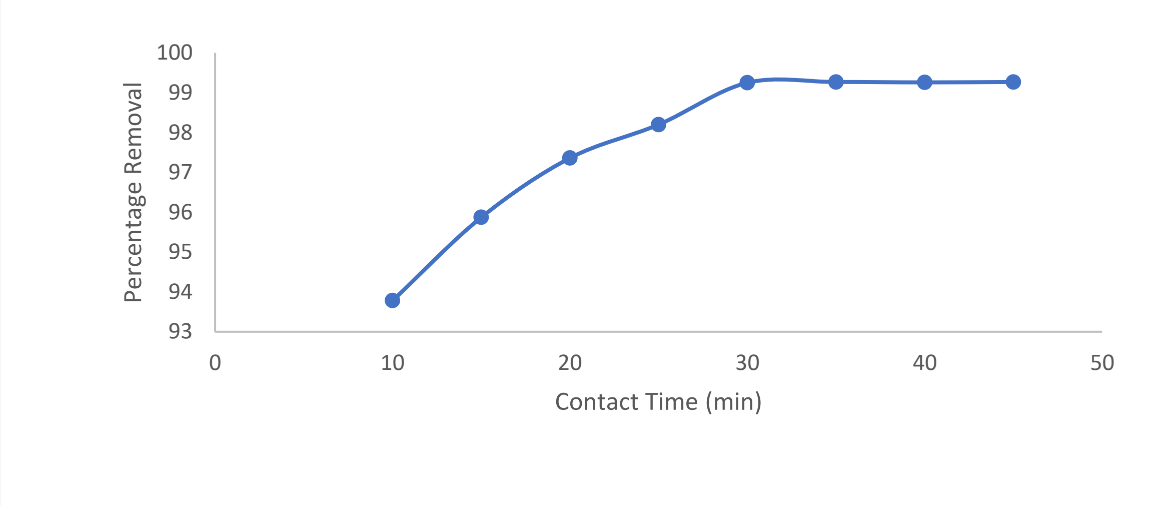

Figure 2: The variation of Percentage Removal vs Contact Time

Effect of pH

As pH is one of the most important parameters affecting adsorption yield, so the effect of pH on the recovery of Zinc(II) ions was investigated. The adsorption studies were executed with 0.05 g of sample and contact time of 35 min by varying the pH in the range of 1.0–7.0. The maximum percentage removal, 96.43% was observed at a pH 6.0. The surface of the nanocomposite might be deprotonated at pH 3.0-5.0 due to electrostatic attraction between the cations and the partially negatively charged adsorbent surface[16]. The variation of percentage removal with pH is shown in figure 3.

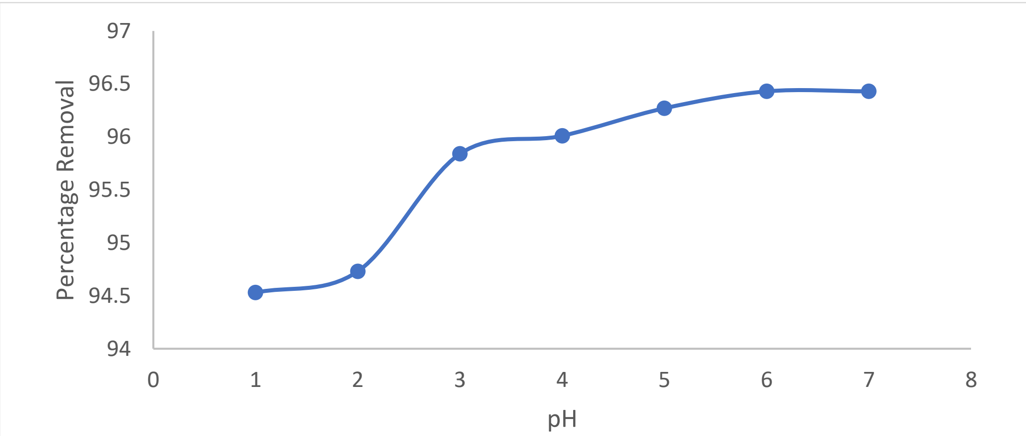

Figure 3: The Variation of Percentage Removal vs pH

Effect of Initial Concentration of Zn (II) ions

The effect of initial concentration of Zn (II) ions at pH 6.0, contact period of 35 min and 0.05 g of sample was studied. It shows that a lower percentage removal at higher Zn (II) concentrations was observed due to an increase in adsorbate ions to a constant number of available active sites on the adsorbent. As a result, 3.0 ppm was regarded as the optimum concentration for the removal of Zn (II) ions with removal efficiency of 79.50%. This variation of percentage removal of Zn (II) ions with initial concentration is shown in figure 4.

Equilibrium Adsorption Isotherms Models

The adsorption isotherms provide indication of the mass of the solute adsorbed per unit mass of adsorbent from the liquid phase at equilibrium. The data obtained for Zn (II) ion adsorption were also confirmed using Langmuir and Freundlich isotherms.

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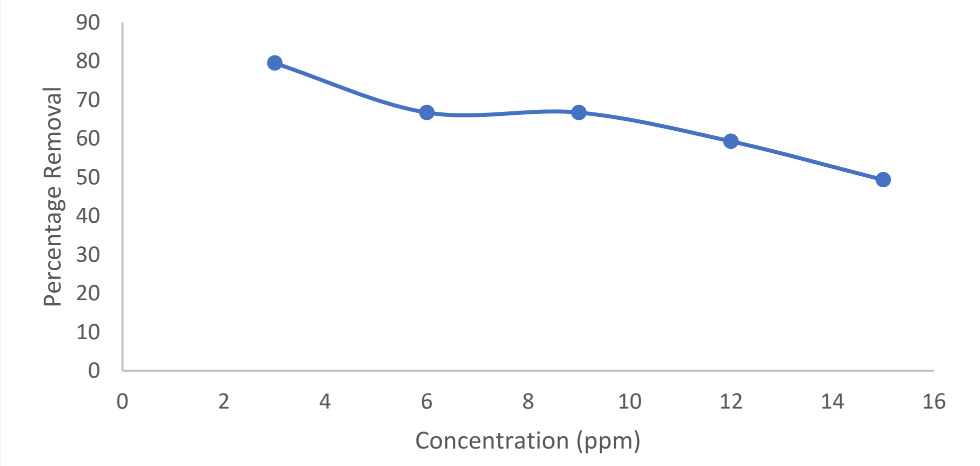

Figure 4: Variation of Percentage Removal vs Initial Concentration

Langmuir Isotherm Model

Langmuir isotherm is based on the assumptions that adsorption occurs on a homogeneous surface and each adsorption site can hold one metal ion at a time[17-18]. The linearized equation of this isotherm is as

$$\frac{C_e}{q_e} = \frac{1}{K_L q_{\max}} + \frac{C_e}{q_{\max}}$$

Where C_e (mg/L) is equilibrium concentration of the metal ion, q_e is the mass of metal ions adsorbed per mass of adsorbent (mg/g), q_{max} is the maximum monolayer capacity of adsorption (mg/g) and K_L is Langmuir constant. The Langmuir isotherm parameters are given in Table 1. The experimental data were fitted into the above equation by plotting C_e/q_e against C_e.

Table 1: Data for Langmuir adsorption isotherm for adsorption of Zn (II) ions by sample

S. No.	q_e (mg/g)	C_e (ppm)	C_e/q_e (g/L)	r
1.	11.925	0.615	0.0516	79.500
2.	20.020	1.996	0.099	66.733
3.	30.025	2.995	0.099	66.722
4.	35.575	4.885	0.137	59.291
5.	37.005	7.599	0.205	49.340

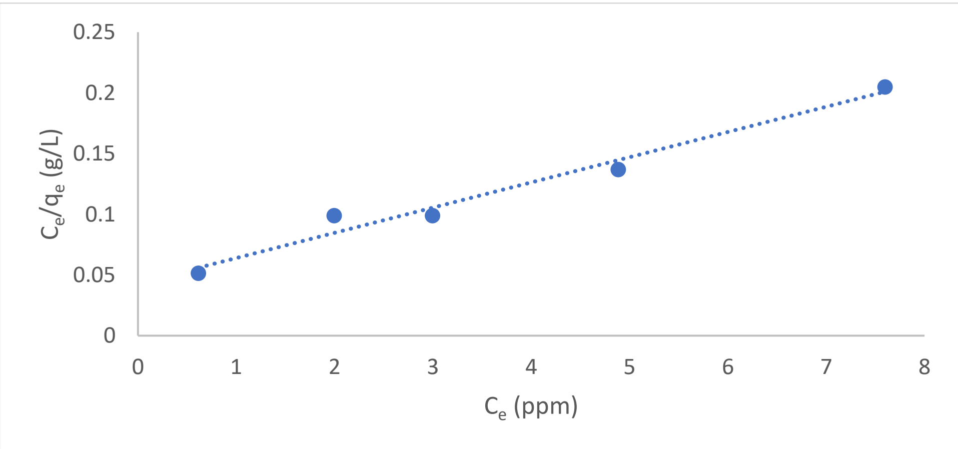

Figure 5. Langmuir Adsorption Isotherm

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The values of q_{max} and K_L were determined from the slope and intercept respectively. To determine the viability of the adsorption process, a separation factor, R_L was used as an indicator of the nature of the adsorption process

$$R_L = \frac{1}{1 + K_L C_1}$$

Where C_1 is the initial metal concentration (mg/L). The values of q_m and R_L being equal to 48.077 mg/g and 0.4113 respectively. R_L value less than 1 indicated favourable adsorption of Zn (II) ions over sample surface.

Freundlich Isotherm Model

Freundlich isotherm is based on the assumption that adsorption occurs on a heterogeneous surface with various adsorption sites which can hold more than one metal ion at a time[19-20]. The linearized Freundlich isotherm equation is as

$$\log q_e = \log K_F + \frac{1}{n} \log C_e$$

Where q_e (mg/g) is the amount of metal ions adsorbed onto the adsorbent at equilibrium, C_e (mg/L) is the heavy metal ions concentration in the solution at equilibrium, K_F and n are Freundlich constants determined from the slope and intercept respectively of the straight line graph between $\ln C_e$ and $\ln q_e$.

Table 2: Data for Freundlich adsorption isotherm for adsorption of Zn (II) ions by sample

S. No.	q_e (mg/g)	C_e (ppm)	$\ln C_e$	$\ln q_e$	$\ln C_e + 0.5$
1.	11.925	0.615	-0.4861	2.4786	0.0138
2.	20.020	1.996	0.6911	2.9967	1.1911
3.	30.025	2.995	1.0969	3.4020	1.5969
4.	35.575	4.885	1.5862	3.5716	2.0861
5.	37.005	7.599	2.0280	3.6110	2.5280

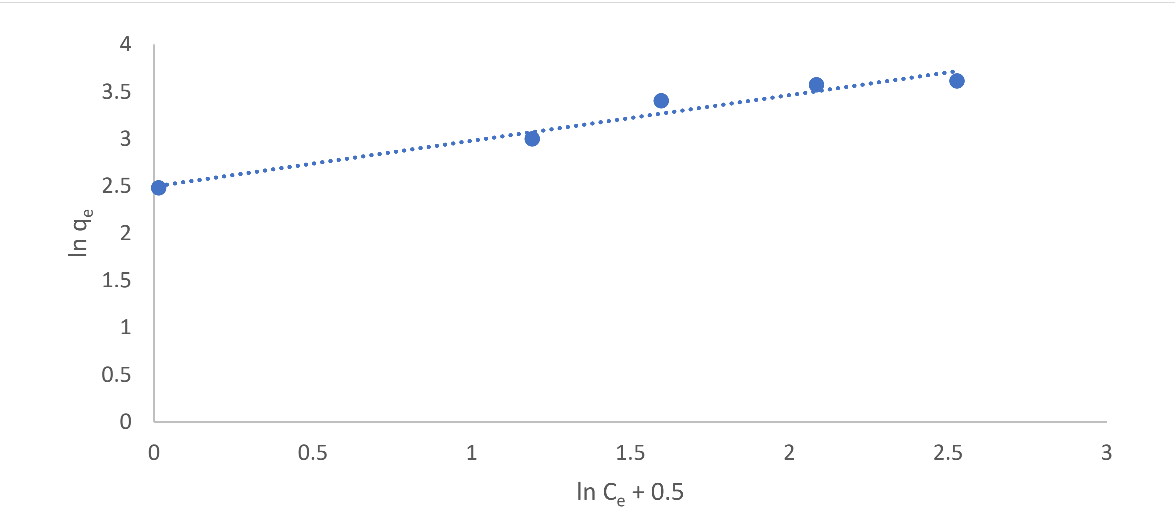

Figure 6: Freundlich Adsorption Isotherm

The favourability of Freundlich model was determined by values of n in the range of 1–10 represent favourable adsorption, while n<1 represent unfavourable adsorption. From the slope and intercept of this linear graph n = 2.0674, and K_f = 312.25 mg/g, values were obtained suggesting the favourable adsorption of Zn (II) ions on sample.

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BET Adsorption Isotherm Study of Sample

To investigate the surface area and porosity of sample BET analysis was performed by adsorption-desorption of N_2 (Figure 7). The surface area and the average pore diameter of the sample were found to be 12.06 m2/g and 43.98 Å, respectively.

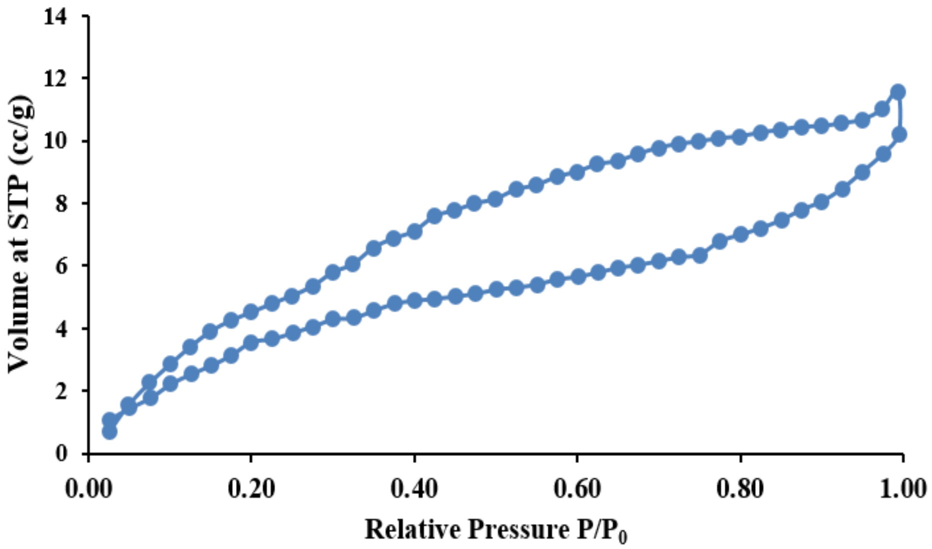

Figure 7. Nitrogen adsorption−desorption isotherm of sample

CONCLUSION

From the adsorption studies carried on sample for removal of Zn ions we report here the optimized parameters. The optimum value of parameters is pH as 6.0, contact time as 35 minutes, adsorbent dose as 0.05 g, initial metal ions concentration as 3.0 ppm and percentage removal as 79.5. From the Langmuir isotherm study the calculated value of separation constant we could establish the favourable adsorption of Zn (II) ions. From the Freundlich isotherm study the favourable adsorption of Zn (II) ions is also established. The BET adsorption study provided useful information about the surface area and average diameter of pores on the surface of sample.

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Conflict of Interest

Authors declare no conflict of interest for this work.

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