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Design and optimization of Zn (Ii) adsorption conditions from aqueous solutions by Fe/Mn-Diatomite material

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ABSTRACT

Environmental issues such as the wastewater have influenced each aspect of our lives. For human and environmental health protection, it is necessary to remove excess zinc in industrial wastewaters before discharging them to environment. Modified diatomite displayed larger surface area and pore volumes in comparison with untreated natural diatomite, which favored heavy metals sorption behavior. In this study, the removal of Zn(II) ions from aqueous solution was studied using Fe/Mn modified diatomite sample at different adsorption parameters such as contact initial metal ions concentration, dosage of Fe/Mn-Diatomite and ionic strength Na_2CO_3 on ionic Zn^{2+} adsorption capacity of diatomite modified. The residual zinc concentration in the solution was determined using flame atomic absorption spectroscopy. The results showed that: the gravitational increase increases with increasing time and then becomes almost stable, with 120 minutes timeliness; absorption increases when Fe/Mn-Ditomite is increased, absorption reaches 89.48% at a dose of 1.5 g/l; additional different concentrations Na_2CO_3 ranged from 0 ppm to 80 ppm the results showed that performance treatment Zn^{2+} of correspond 94,85%.

This study could lay an essential foundation to develop modified diatomite for heavy metal removal from wastewater.

***Keywords:** Diatomite, adsorbed Zn^{2+}, Zn(II) removal, adsorption efficiency, Adsorption capacity*

1. Introduction

With the increase in technological development and industrialization in our society, this consequently leads to an increase in the contamination of the environment through various means. Industrial waste, exhausts from factory engines and turbines, products of nuclear weapons among others lead to the release of some toxic heavy metals to our waters and air which also invariably have some complicated effects on the environment and human beings. Heavy metal contamination of water, air and soil regimes continued unabated despite several decontamination processes that have been developed (Burri & Weatherl, 2019).

Adsorption is one of the most effective and economic techniques for removing heavy metal ions from aqueous solutions. The efficiency of adsorption relies on the capability of the adsorbent to concentrate or adsorb metal ions from dilute solution onto its surfaces and the rate of removing such ions from the solutions. Different adsorbents such as activated carbon, zeolites, resins, bio sorbents, hydrogel and magnetic hydrogel has been used for the removal of heavy metal ions by adsorption. Despite the availability of a number of adsorbents for the removal of low concentrations of heavy metal ions from the aqueous solution, there is still a need for the development of new adsorbent with superior adsorption capacity, facile desorption–desorption kinetics, high stability and easiness of operation (Bandura & Woszuk, 2017).

Zinc is one of the essential ions for life due to its micronutrients properties when present in trace quantities. However, excess of the maximum permissible amounts can lead to serious health problems. The World Health Organization recommended the maximum acceptable concentration of zinc in drinking water as 5.0 mg/L. Most of Zn(II) ions are generated by industries such as mineral extraction, metal plating and battery producing.

Diatomite is a siliceous natural material also used in water treatment. It is a sedimentary rock of biogenic origin, with a biomorphic structure. The main component of the rock is amorphous opal, the least stable form of silica; however, the rock can also contain quartz, calcite, clay minerals, iron compounds, or glauconitic (Haldar, 2014). As good sorbent, diatomite has many applications for removing various impurities from water (Flores-Cano, Leyva-Ramos et al, 2013). The specific surface area of diatomite varies depending on the origin, though is larger comparing to the silica rocks, the porous structure makes it an excellent filtration medium with a wide industrial application (Ediz, N.; Bentli, I.; Tatar,I 2010) and the porosity can be increased by modification.

This informs the choice of our contribution to this area of adsorption process which focus on modified diatomite by Fe/Mn for Zn(II) removal from aqueous solutions.

2. Arrangement of experiments

2.1. Materials

- ✓ Research subjects: Zinc solution (Zn^{2+}) (from $Zn(NO_3)_2$ 98% China)
- ✓ Research material : Fe/Mn-diatomite material was prepared by Diatomite in Phu Yen provine with $FeSO_4$ and $KMnO_4$ appropriate rate (According to Fangfang Chang et all (2009), Bui Hai Dang Son (2017)).
- ✓ Research chemicals: NaOH (China, 96%), HCl (China , 36%), $FeSO_4.7H_2O$ (China, 98%), $KMnO_4$ (China, 99%).

2.2. Arrangement of experiments

- ✓ **Experiment 1:** According to Fangfang Chang & Jiuhui Qu et all (2009), Zhongzheng Yanga, Yuejun Lu (2013), Dosage survey: 0.2, 0.5, 1, 1.8, 2g/l; Surveying time: 0, 30, 60, 60, 120, and 150 minutes.
- ✓ **Experiment 2**: According to Fangfang Chang & Jiuhui Qu et all (2009), Zhongzheng Yanga, Yuejun Lu (2013): Concentration survey: 25, 50, 75, 100, 150 ppm, Surveying time: 0, 30, 60, 60, 120, 150 minutes.
- ✓ **Experiment 3:** According to Fangfang Chang & Jiuhui Qu et all (2009), Bui Hai Dang Son (2017): Ionic strength Na_2CO_3: 0, 20, 40, 60, 80 ppm; Surveying time: 0, 30, 60, 60, 120, 150 minutes.

2.3. Evaluation methodology

- The residual zinc concentration in the solution was determined using flame atomic absorption spectroscopy. The samples were finally filtered and analyzed by atomic absorption spectroscopy Shimadzu 7000 (AAS) for determining of residual ion concentration, measurement wavelength of 213,9 nm.
- Take 50 ml of Zn^{2+} containing solution of known concentration in a 250 ml Erlenmeyer flask, add a certain amount of Fe/Mn-diatomite, according to different experimental requirements, under the conditions of changing the experimental conditions, shake adsorption for 1 hour, adsorption is completed .After that, the supernatant was taken, and its residual ion concentration was measured by visible spectrophotometry. Calculate the removal rate and the amount of adsorption using the formula below:

$$Removal\ efficiency = \frac{C_0 - C_t}{C_0} \times 100\%$$

3. Results and Discussion

3.1 Effect of adsorbent dosage

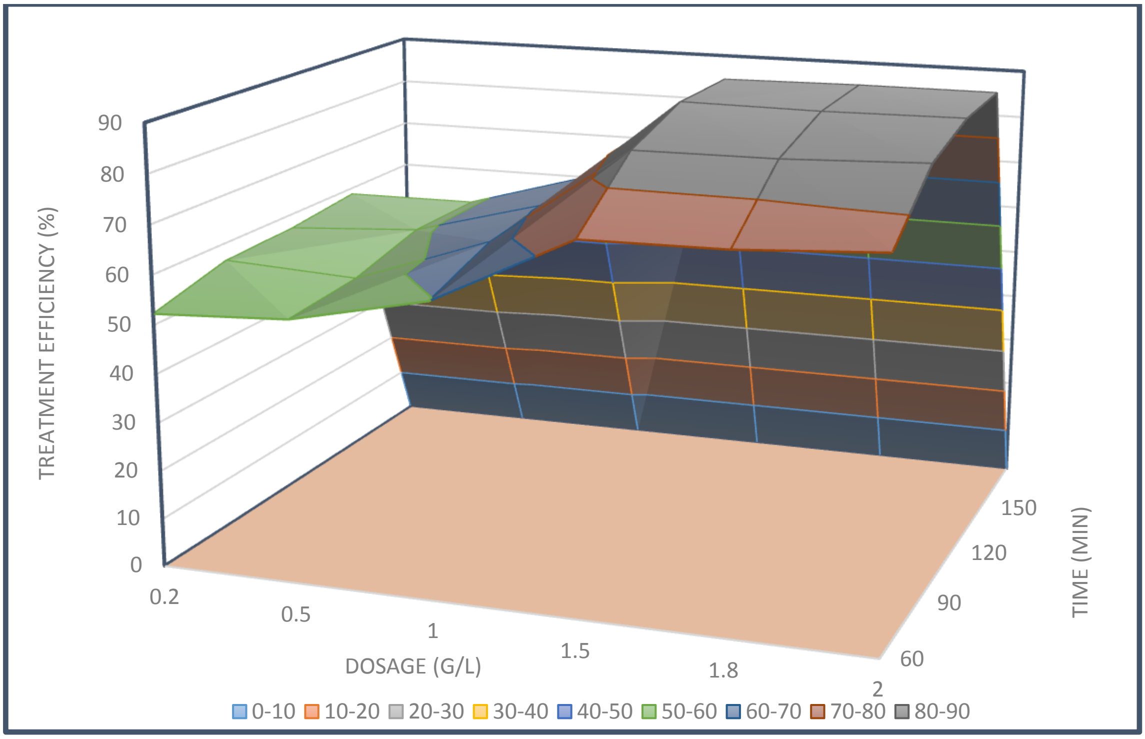

Figure 1. *Response surface, showing the effect of adsorbent dosage on the Zn(II) percentage removal.*

The mass of the adsorbent is one of the other factors that studies in adsorption experiments. The outcomes from this study (Fig.1) demonstrate that the adsorption efficiency of zinc removal goes up when the adsorbent mass increases. The research results capable of processing zinc metal at the Fe/Mn-diatomite material at time: 30, 60, 90, 120, and 150 min is expressed on Fig 1 shows, in the range of doses of 0.2, 0.5, 1.0, 1.5, 1.8, 2 g/l has the increasing performance from 58,62% to 89,90 %. Optimal adsorption is achieved at a dose of 1.5 g/l, reduction efficiency is 89.48%.

As the matter of fact, the increase in the adsorbent mass causes an increase in the number of the active adsorption site in solid phase and therefore causes an increase in the efficiency of zinc removal. With increasing adsorbent mass amount, the number of the active sites increases, so the contact area of the adsorbent and the pollutant increases and causes an increase in the adsorption capacity. For the initial blank surface, the adhering possibility is large, and therefore adsorption proceeded with a high rate. The slower adsorption rate at the end is probably due to the saturation of active sites and achievement of equilibrium (Pehlivan & Altun, 2009). As mentioned earlier, by increasing adsorbent dosage, the percentage removal of Zn (II) was increased.

According to research results of Liu Ling, Wei Qiye about Study on adsorption property of modified diatomite against Cu^{2+} in wastewater, Elimination efficiency increases with increasing dose of modified diatomite, When the amount of modified diatomite material was 1.0 g/l, the removal rate was 90.00%. This shows that, modified diatomite material adsorb metal best at doses between 1.0-1.5 g/l.

3.2. Effect of initial concentration

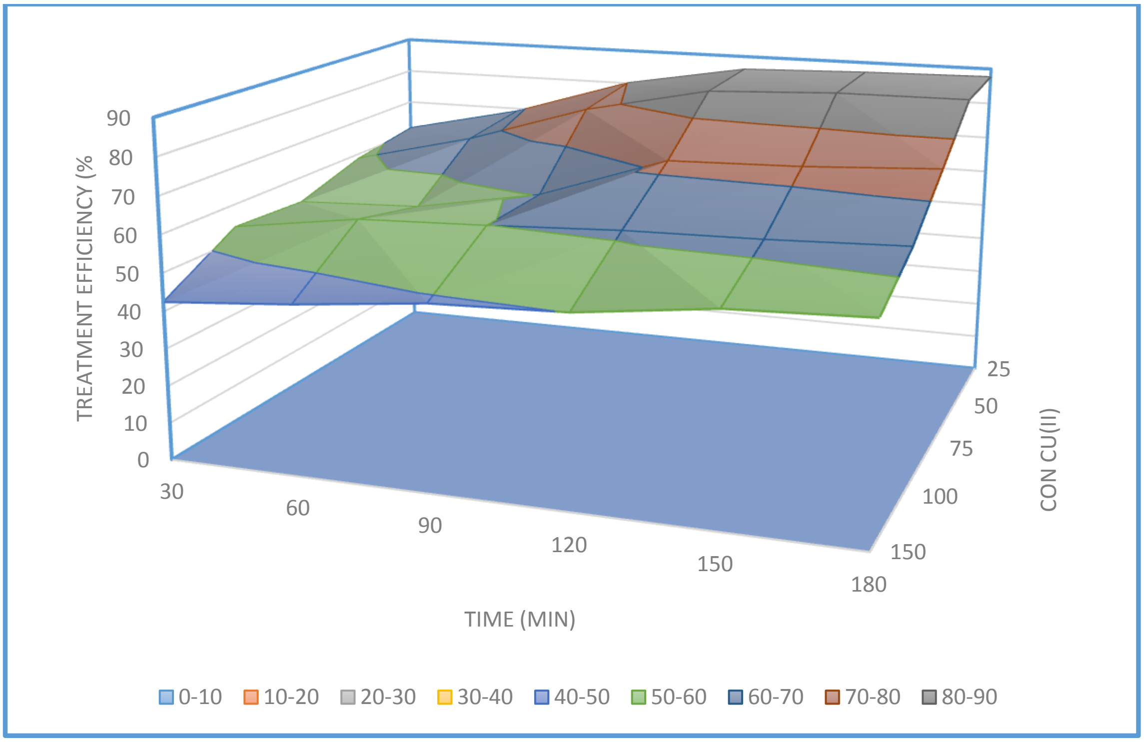

Figure 2. *Response surface, showing the effect of initial ion concentration, removal time and adsorbent dosage on the Zn (II) percentage removal*

The extent of adsorption can be better understood with the initial concentration studies. The possible interaction between heavy metals and the accessible sites on the surface of the adsorbent is influenced by the initial concentration of the heavy metal. Adsorption of Fe/Mn-diatomite with initial ions concentration from 25 ppm to 150 ppm and time adsorption from 30 minutes to 180 minutes at pH=5 was shown in Fig.2. It was shown that adsorption at initial ions concentration of 25, 50, 75, 100 and 150 ppm were 87.65%, 86.91%, 75.05%, 63.43% and 55.81% respectively. In other words, adsorption decreased with increase of initial ions concentration and then tended to be stable at lower than 50 ppm, 120 minutes. In order to gain best adsorption, appropriate Zn(II) ions concentration was 25 ppm ion at fixing adsorbent dosage, the adsorption reached up to 87%. We know that surface areas and defects are definite when a certain amount of Fe/Mn-diatomite, so its adsorption capacity for Fe/Mn-diatomite is also limited. When amount of Zn(II) ions are more than the maximum bearing capacity of Fe/Mn-diatomite, the Fe/Mn- diatomite could not adsorb any other ions. So the adsorption decreased fast when Zn(II) ions were increased. If we wanted to increase adsorption of Zn(II) ions with higher concentration, we should increase the amount of Fe/Mn-diatomite.

According to research results of the adsorption of Zinc(II) from aqueous solution on Mn-diatomite from Zhongzheng Yanga, Yuejun Lub, Yangyang Li (2013) with an efficiency of 95.7%. Compared with some previous studies such as research results Sari Tuomikoski, Riikka Kupila (2019) show that Adsorption of Zinc(II) from 10 ppm to 500 ppm using biomass-based activated carbon, heavy metal processing performance Zn^{2+}have equivalent processing efficiency of 90% at 75ppm.

Metal treatment Zn (II) of Fe/Mn-diatomite is effective at 50 ppm and the dosage is 1.5g/l with a removal efficiency of 87%. For best results we need to handle additional survey effect of ionic strength Na_2CO_3 to have a good performance press the best performance.

3.3. Effect of ionic strength Na_2CO_3

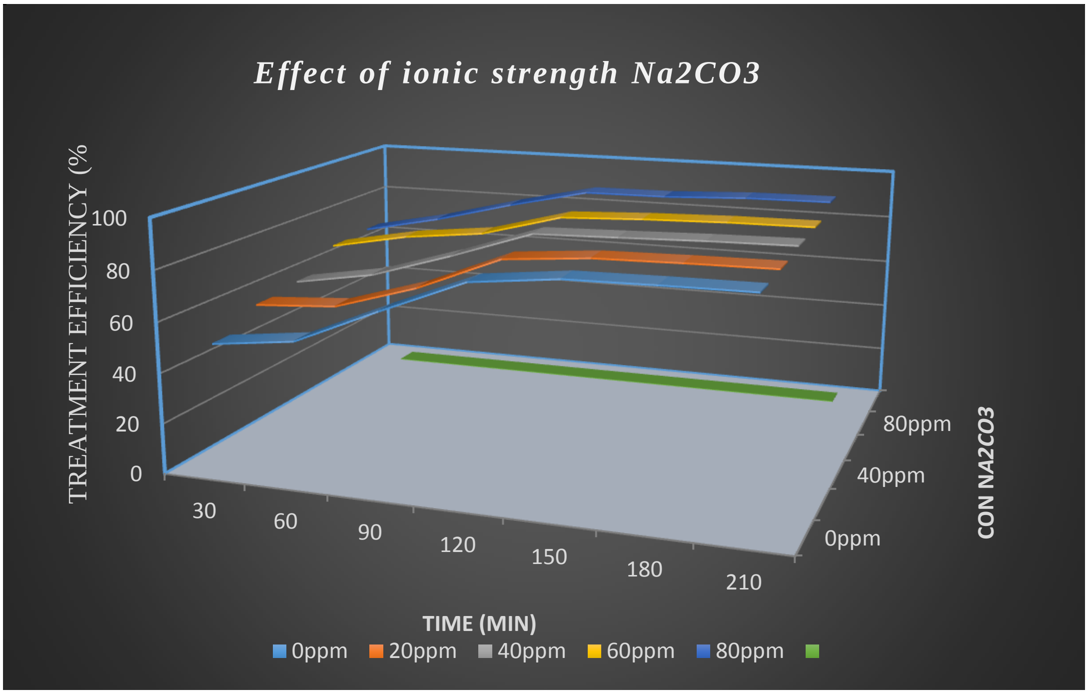

Figure 3. *Response surface, showing the effect of ionic strength Na_2CO_3 removal time and adsorbent dosage on the Zn (II) percentage removal*

For studying the influence of ionic strength on the efficiency of zinc removal by Fe/Mn-diatomite, experiments were carried out in optimum condition and six distinct time durations (30, 60, 90, 120, 180 and 210 min) with five different ionic strengths (0, 20, 40, 60, and 80 ppm) solution. The results of each experiment were calculated and introduced in fig.3. Whereby, with increasing the ionic strength of Na_2CO_3 solution from 0, 20, 40, 60, and 80 ppm, zinc removal efficiency by Fe/Mn-diatomite has increased. The mean removal efficiency for ionic strength of 0, 20, 40, 60, and 80 ppm are respectively 87.07%, 87.94%, 89.73%, 90.61%, 94.85%.

Following fig.3, the highest removal efficiency is 94.85%, which occurred when the contact time was 210 minutes and the ionic strength was 80 ppm; in the same way, the lowest removal efficiency is 50.69%, which occurred when the ionic strength was 0 ppm and 30 min.

From Fig. 3, it can be seen that with the increase of ionic strengths from 0, 20, 40, 60, and 80 ppm, the pH of the solution increases, the removal rate and the amount of Zn^{2+} in the wastewater from the modified diatomite increase. It shows that the modified diatomite has better adsorption when the ionic strength of Na_2CO_3 solution increases.

According to research results of the absorption of As(III) from aqueous Solution on Mn-diatomite from Bui Hai Dang Son (2017), removal efficiency As(III) raise from 35% to 80% when ionic strength of Na_2CO_3 from 0, 20, 40, 60, and 80 ppm. Thus, we see that the ionic strength force greatly affects the adsorption capacity of the Fe/Mn-diatomite material.

4. Conclusion

The factors influencing the Zn^{2+} sorption in the wastewater were studied by using Fe/Mn-diatomite. The three factors, such as dosage of Fe/Mn-diatomite, the initial concentration of Zn^{2+}, ionic strength of Na_2CO_3. The experimental results show that: the best adsorption effect 89.48% at a dose of 1.5 g/l; the increase of adsorption time is beneficial to improve the adsorption of Zn^{2+} by Fe/Mn-diatomite and zinc concentration 50 ppm and adsorption time 120 minutes, the adsorption of Zn^{2+} on the Fe/Mn-diatomite reaches a saturation state 87%; ionic strength has an effect on zinc removal efficiency by Fe/Mn-diatomite, concentration Na_2CO_3 with 80 ppm the results showed that performance treatment Zn^{2+} of correspond 94,85%.

To get the best zinc adsorption results from Fe/Mn-diatomite, we need to handle additional survey effect of ionic strength Na_2CO_3 to have a good performance press the best performance.

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