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Research Article

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Research on preparation of modified polyacrylamide-gel and its performance of removing lead from aqueous solution

Yang-zhi Zhu[1], Zhen Li[2] and Jian Liu[2]

[1]State Key Laboratory of Fine Chemicals, The R&D Center of Membrane Science and Technology, School of Chemical Engineering, Dalian University of Technology, Dalian
[2]School of Environmental Science and Engineering, Chang'an University, Xi'an, China

ABSTRACT

According to the Mannich-Reaction, a kind of chelating hydrogel grafted with dithizone was prepared from polyacrylam (PAM), formaldehyde and dithizone. And the optimum adsorption condition of disposing Pb (II) from aqueous solution was studied. The result indicated that at 30 ℃, pH=6-7,0.5g of the chelating hydrogel grafted with dithizone processed 10mg/L 100mL Pb (II) solution for 2h, and then 99.5% of Pb (II) was removed.

Key words: waste water; dithizone, chelating hydrogel, capturing Pb2+

INTRODUCTION

Lead is a typical heavy metals contamination because of its impact on public problems when it is discharged into water resources. Lead accumulates mainly in liver, bones, kidney and nervous system and may cause serious developmental disorders, anaemia, mental retardation and even death [1]. So far Lead can be reduced by Chemical Precipitation process, Electrolysis, Biological process, Ion-exchange technique and Electrodialysis [2]. However, such methods still have some weak points, so seeking for a nontoxic, no secondary pollution and high adsorption capacity material of removing Pb (II) can still be regarded as the key technology when studying removing lead ions from water.

Hydrogel is a kind of polymer materials with three-dimensional network structure, which can maintain its original structure after swelling in water, and respond to foreign stimulates, which is widely used in many areas especially in environmental pollution control[3-5]. The use of hydrogels as adsorbents for the removal of heavy metals, recovery of dyes and removals of toxic components from various effluents has been studied, absorbents with carboxl, phosphonic, sulphonic and nitrogen groups on their surface favor metal ions adsorption [6].

In this study, the behavior of lead ions removal from aqueous solution through grafted and modified polyacrylamide-gel was evaluated.

EXPERIMENTAL SECTION

Materials
Dithizone; N,N'-methylene, diacrylamide; Oxammonium, hydrochloride; Lead-nitrate and Ammonium citrate. All the reagents are Analytical Reagent (AR).

SYNTHESIS OF GRAFTED AND MODIFIED POLYACRYLAMIDE-GEL
Preparation of Polyacrylamide Gel (Crosslinking Degree=10%)
14g Acrylamide and 1.6g N,N'-methylene diacrylamide were dissolved into 50mL distilled water, and little

Ammonium persulphate (APS) and TMEDA were added. Then raised the pH of the solution to 10, keeping still at room temperature. The resulting Polyacrylamide Gel(crosslinking degree=10%) was washed in distilled water to remove any residual reagents and dried in a vacuum oven. After drying, the gel was cut into small cubes and stored in a desiccator until use.

Preparation of grafted and modified polyacrylamide-gel

1.58g Polyacrylamide Gel(crosslinking degree=10%) were added into 100mL distilled water at a three-necked bottle, then the pH of the solution was changed to 10, and 3.6 mL formaldehyde were added in to the three-necked bottle, then 50 mL 2g/L dithizone solution were dropped into three-necked bottle at a low speed. After dropping, reacted for 2h at 30℃. The resulting grafted and modified polyacrylamide-gel were rinsed for 4-5 times to remove residual monomers, and then dried in a vacuum oven. After drying, the gel was cooled before use. The reaction mechanism of preparation of modified Gel and its removal of Pb (II) are discussed in Fig.1.

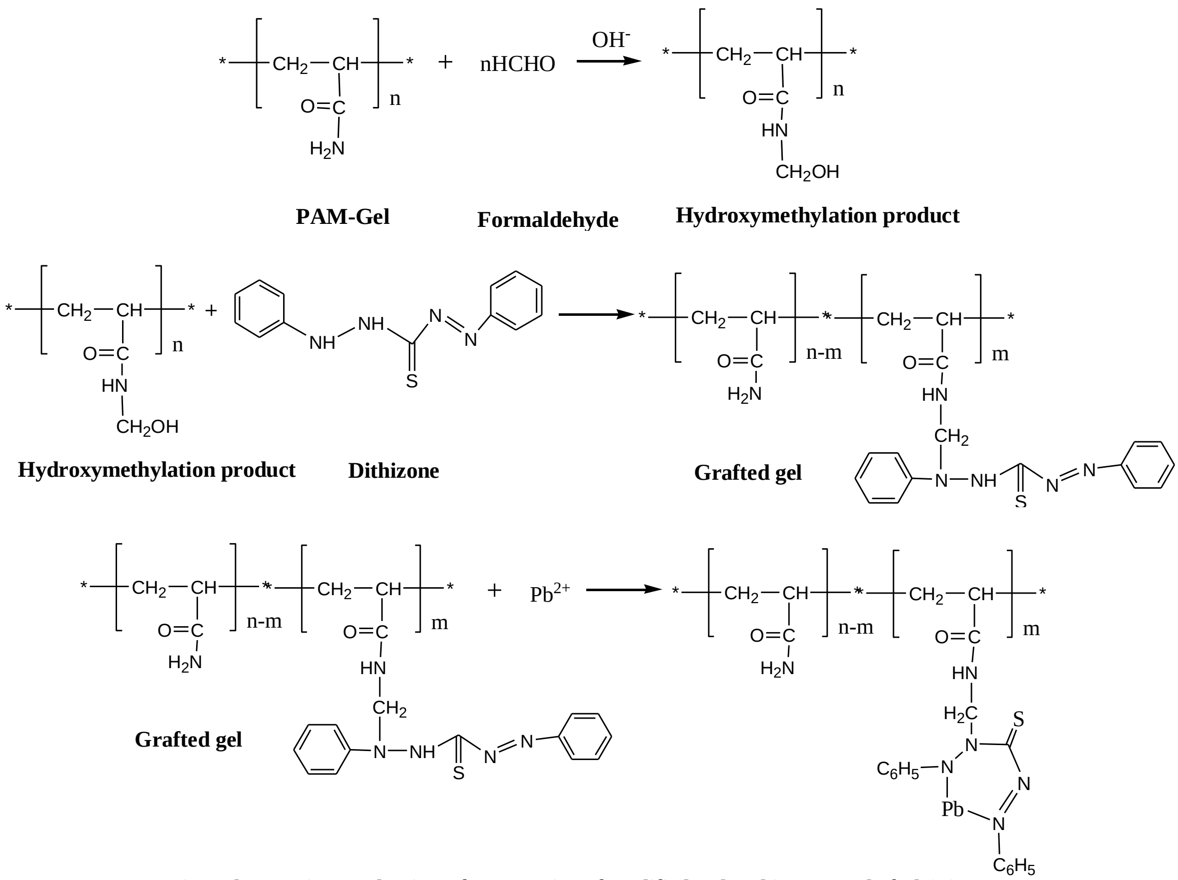

Fig.1 The reaction mechanism of preparation of modified Gel and its removal of Pb(II)

ADSORPTION OF Pb(II) BY GRAFTED AND MODIFIED GEL

In order to evaluate the Pb(II) removal capacity of the grafted and modified gel, batch equilibrium tests were conducted. The gel samples were put in contact with the aqueous Pb(II) solutions under various conditions, all solutions were preapared with analytical grade reagents, using lead nitrate and distilled water. The mixtures were stirred in a oscillator and then the Pb(II) in solution were evaluated after reaction. The contacting time varied from 0.5h to 3h. and the effect of the pH on lead absorption was determined by using different pH values from 2 to 10. Also the dosage of the gel (varied from 0.5g to 2g) and reaction temperature(ranged from 20℃ to 60℃) were evaluated. The initial lead concentration in all cases was 10mg/L, Duplicate experiments permitted averaging of the results.

The lead removal was calculated as follows:

$$\text{removal} = \frac{C_1 - C_2}{C_1} \times 100\% \qquad (1)$$

Where C_1 , initial lead concentration;C_2, final lead concentration.

Quantification of Pb(II) in solution

So far, there are many methods to test the lead concentration like Flame atomic absorption method, graphite furnace atomic absorption spectrometry (GFAAS), Inductively coupled plasma atomic emission spectrometer (ICP AES) and Stripping voltammetry[7-10]. In this paper, The concentration of Pb(II) ions in solution, before and after the absorption process, was determined by dithizone spectrophotometric method and carbon tetrachloride were used as extractant to detect lead content.

RESULTS AND DISCUSSION

pH effect on lead adsorption

The pH effect of solution on the adsorption of lead ions is shown in Fig.2, the grafted and modified gel display a sharp trend increasing Pb(II) removal with increasing pH of solution until peaking at pH=6 and decreasing sharply at higher pH values. The pH effect in gel was drastic with Pb(II) removal close to 80% at pH=6 in contrast to 40% at pH=4. This is likely due to increased competition of hydroxyl ions with metal ions in more alkaline conditions and increased consumption of protons with grafted gel in more acidic conditions. So pH=6-7 was the optimum operating condition.

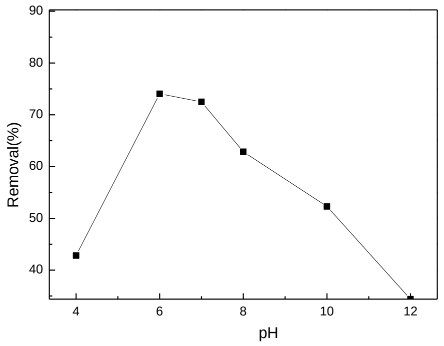

Fig.2 Effect of the pH removing Pb(II) removal at 2h of solution as a function of contact time. With Pb(II) initial concentration of 10mg/L.

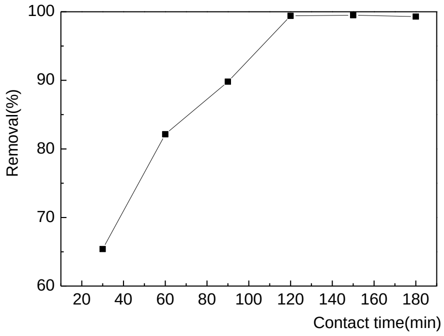

Fig.3 Experimental Pb(II) removal from aqueous contact time with a Pb(II) initial concentration of 10mg/L

Contact time effect on lead adsorption

Fig.3 shows the expermental plot obtained for Pb(II) removal from aqueous solution as a function of contact time with the modified gel. In 2h, it achieved a Pb(II) reduction around 99.5%, this confirms that dithizone group is one of the most effective groups for adsorption of heavy metal ions from aqueous solutions. It is also observed that the adsorption of lead ions increased rapidly in the first 2h, and then augumented slowly. This can be explained by the fact that adsoption rate is depended on the number of available adsorption groups (dithizone groups)on the surface of the adsobent. Hence 2h can be the optimum contact time.

Reaction temperature effect on lead adsorption

The efficiency of the removal of Pb(II) from aqueous solutions at different temperature was studied in Fig.4, Note that at 30℃, it achieved a Pb(II) reduction around 85% and as the temperature increases, the efficiency decreased sharply. This confirms that it was an exothermic reaction and thereby increasing the temperature could not favor the elevation of the efficiency of the removal of Pb(II) .

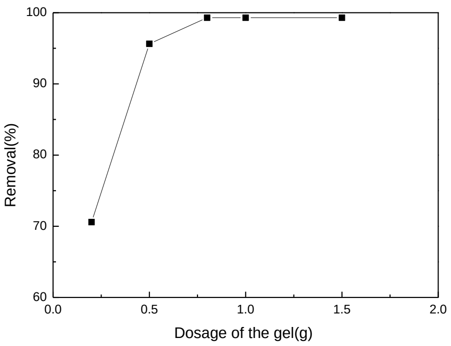

Fig.5 Effect on removing Pb(II) of modified Gel dosage with Pb(II) initial concentration of 10mg/L

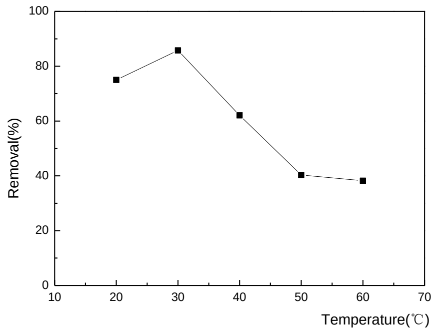

Fig.4 Effect on grafted and modified Gel removing Pb(II) of temperature with Pb(II) initial concentration of 10mg/L

Dosage effect on lead adsorption

Fig.5 shows that the efficiency of removal of Pb(II) increased as the dosage of the gel raised. It was observed that 0.5g gel could make a Pb(II) reduction around 99.5% and the efficiency increased slightly at higher dosage of gel. This can be explained by the fact that the concentration of Pb(II) was fixed despite the increasing of adsorbent. Therefore the efficiency of removal of Pb(II) could be obtained to 99.5% when the dosage could be grater than 0.5g.

CONCLUSION

(1) According to the Mannich-Reaction, a kind of chelating hydrogel grafted with dithizone was prepared from polyacrylam(PAM), and this is a noval grafted chelating gel.

(2) The optimum adsorption condition while disposing Pb(II) from aqueous solution were below: At 30℃, pH=6-7,0.5g of the grafted chelating hydrogel processed 10mg/L 100mL Pb(II) from aqueous water for 2h, and 99.5% of Pb(II) was removed. This synthesis method was simple with a higher removal efficiency of Pb(II).

Acknowledgments

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