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Simple Method for the Removal of Mercury(II) from Its Aqueous Solution Using Aluminum as a Reducing Agent

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Abstract

Removal of mercury(II) from its aqueous solution with the reduction technique by using aluminum metal as a reducing agent has been investigated. The experiments were performed by adding the amount weight of aluminum chips into testing solutions containing Hg(II) in various concentrations. Some factors studied in this investigation were pH and contact time. From the experimental data it is known that the temperature and contact time influenced on the reduction of Hg(II), where the optimum pH value and contact time of the reduction are 6 and 8 h, respectively. This simple method may be useful to remove the mercury(II) from any waste aqueous solution before their discharge into the environment.

Keywords: Aluminum, aqueous solution, contact time, mercury(II), pH, reducing agent, removal.

1. Introduction

Existence of mercury (Hg) in environment is one of the major concern nowadays, because of its toxicity to living beings (Rao et al, 2011). Mercury and its compounds are recognised as dangerous poisons and harmful to central nerveous system (Craig, 1986) and natural ecosystem (Clifton, 2007; Clarkson and Magos, 2006). Mercury can be adsorbed through the skin and lungs, and are stored in the liver, kidneys, brain, spleen and bone leading to development of carcinogenic, mutagenic, teratogenic, and also promote several health problems (Bhakta et al, 2009). Mercury encountered in environment is produced from various anthropogenic and industrial activities such as paint, pharmaceutical, paper and pulp, oil refining, rubber processing and fertilizer industries. In addition, the extensive use of HgCl₂ for routine activities in laboratories followed by the discharge of the analyte solution also contributes to the mercury pollution (Giri and Das, 2013). Due to potential accumulation of mercury onto human bodies and aquatic organisms as well as its toxixity, removal of mercury from its aqueous solution is a vital task. Therefore, efforts to develop the methods of mercury removal should be always carried out. Various techniques for removing of mercury have been reported including precipitation, coagulation, softening, reverse osmosis, ion-exchange and adsorption (Manohar, 2002), foam fractionation (Moussavi and Javidnejad, 2007) and others (Fagueirera et al, 2012; He et al, 2011). The recent researchers reported that reduction-oxidation reaction with aluminum metal (Giri and Das, 2013) and magnetite (Wiatrowski et al, 2009) are also effective for recovery of Hg(0) from waste analyte solutions. These latter reports has been inspired an extended search to investigate some factors affecting the reduction of Hg(II) using aluminum metal as reducing agent.

2. Experimental

The reducing agent used in this experiment was aluminum chips produced by Merck (GR) and all chemicals and reagents used were of AR/GR grade without purification process. The reaction vessels used in all redox experiments were 100-mL Erlenmeyer flasks with glass stoppers. A magnetic stirrer was used to mix the reductor and Hg(II) solution, and a glass beaker filled in with water was used as a thermostat to maintain a constant temperatureat experiment conditions. Experiment was performed by adding 0.21 g of aluminum chips into a 100-ml of HgCl₂ solution with a concentrations of 3.0 and 4,0 g/l. The pHs of solution (2-12) were adjusted by applying a suitable buffer solution. The flask was put in a thermostat at constant-temperature of $30\pm1^{\circ}$ C and stirred continuously with a speed of 300 rpm for a period of time 1-12 hours. After Hg(0) is completely settling down at the bottom of the flask, the residual Hg(II) concentration in solutions were analyzed by a double beam UV-vis Spectrophotometer (HITACHI U-2010 Model) at 576 nm after being used rhodamine 6G treated with tetraiodomercurate(II) according to procedure reported by Oubagaranadin et al (2007). The removal effectiveness ($E_{Removal}$) of Hg(II) from aqueous solution by the reduction process was calculated as:

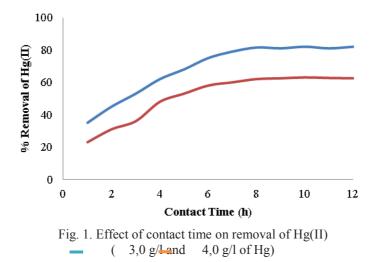
% Removal of Hg(II) =
$$\frac{(c_0 - c_t)}{c_0} \times 100\%$$
 (1)

where C_0 and $C_t(g/l)$ correspond to aqueous solution concentrations of Hg(II) at initial and sampling times, respectively. The same procedure but at different temperatures, pHs of solution and contact times were also performed to evaluate their effect on the reduction reaction.

3. Results and Discussions

3.1 Effect of contact time

The Fig. 1 shows the percentage removal of mercury using 3,0 and 4,0 g/l at various contact time by 0.21 g (100 ml) of aluminum.



It was observed that in the case of the experiments, the percentages removal of Hg(II) from test solutions with initial concentrations of 3,0 and 4 g/l were increased with increasing contact time until reaching 8 hours. And, the maximum percentages of Hg(II) removed were 81.5 and 62,5% from solutions having concentrations of 3,0 and 4,0 g/l, respectively. At higher contact times than 8 h, the percen reduced of Hg(II) by are relative was observed the same. It means that the optimum contact time of the reduction process is at 8 hours. It is seen that the rate of reduction is relatively slow and about 8 h is required to complete the reduction of Hg(II) to Hg(0) under the experimental condition used. The mercury recovered from the solution is analyzed. The purity of recovered Hg(0) is checked by measuring the density. The density of recovered Hg(0) from the aqueous solution and pure HgCl₂ solution is found 13.45 and 13.50 g/ml, respectively, which are close to that of standard value 13.53 g/ml indicating that recovered mercury is sufficiently pure and may be considered for its possible use in different purposes.

3.2 Effect of pH

The pH is one of the most important parameter controlling the reduction process ot mercury(II) from its aqueous solutions. Fig. 2 shows the influences of pH on mercury removal efficiencies by the reductor.

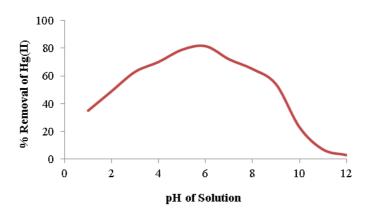


Fig.2. Effect of pH on the removal of Hg(II)

The studies were conducted at a constant initial mercury concentration and contact time of 8 hours. On treatment with aluminum chips, Hg(II) reduces to Hg(0), as follows:

$$3 Hg^{2+} + 2Al^0 \rightarrow 3 Hg^0 + 2Al^{3+}$$
 (2)

The percentage of Hg(II) reduced increases with an increase in the pH to attain a maximum at pH 6 and thereafter, it decreases with further increase in pH. The maximum removal of mercury at pH 6 was found to be nearly 80 percent. These results indicate that aluminum can be satisfactorily used at existing pH of the test solution. The lower pH values result in the protonation of Al, which leads to the extensive repulsion of Hg²⁺ ions. This results in a decrease in mercury(II) reduction. With increase of pHs from 2.0 to 6.0, the mercury exists as mercuric hydroxide Hg(OH) in the medium and surface protonation of adsorbent is minimum, leading to the enhancement of Hg²⁺ ions reduction. The reduction decreases at acidic pH due to the lower adsorption of HgCl, species (present at acidic pH) as compared to Hg(OH), species (present at pH 6). This hypothesis is supported by the results obtained in the present study, because the amount of mercury(II) reduced at pH 6 is much greater than that reduced at pH 2. The decreased reduction at pH 2 may be due to the lesser extent of the oxidation of HgCl to HgCl₂ at this pH as a result of the elevated HCI concentration in the medium. On increasing the pH from 2, the percentage removal of mercury increased and became maximum at pH 6. With increase of pHs greater than 6, the reduction process decreased. This can be explain by the formation of the yellow solid of Hg(OH)₂ as a result of reaction between Hg(II) and NaOH:

$$\text{Hg}^{2+}_{(aq)} + \text{NaOH}_{(aq)} \rightarrow 3\text{Hg}(\text{OH})_{2 (s)}$$
 (3)

3 Conclusions

The study indicates that aluminum metal could be used as an effective reducing agent for the treatment of mercury(II) aqueous solution. The reduction of mercury(II) by aluminum is influenced by pH and contact time. The optimum conditions of mercury(II) removal obtained from this study are pH 6.0 and contact time 8 h. And, sufficiently pure metallic mercury can be easily recovered from aqueous solution by reduction with Aluminum chips. This method is very simple to be performed without comsuming much energy, but in low rate of reduction. This method may be used for the removal and recovery of mercury(II) to reduce the ensuing environmental pollution.

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