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Author(s)	Machida, Motoi; Cao, The Ha; Tatsumoto, Hideki
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Removal of Heavy Metal from Aqueous Solution by Magnesium Oxide Loaded Granular Activated Carbon

Motoi Machida¹, Cao The Ha² and Hideki Tatsumoto¹

¹ Faculty of Engineering, Chiba University, Yayoi-cho 1-33, Inage-ku, Chiba 263-8522, Japan

² Research Centre for Environmental Technology and Sustainable Development, University of Science, Vietnam National University, Hanoi, Vietnam

ABSTRACT

Improvement of adsorption capacity of lead(II) in aqueous solution was examined by introducing magnesium oxide to a commercially available granular activated carbon (GAC). Comparing the magnesium loaded granular activated carbon with no magnesium loaded GAC, lead(II) adsorption was significantly enhanced by magnesium oxide loading. Influence of pH on lead(II) adsorption and desorption was also experimented to clarify the difference between GAC and magnesium loaded GAC. Lead(II) removal was always higher for magnesium loaded GAC than for GAC. The results indicated that increase in lead(II) adsorption for magnesium loaded GAC would be caused by increasing the number and strength of adsorption sites.

Key words: heavy metals, lead, activated carbon, adsorption, magnesium oxide

INTRODUCTION

Heavy metals are widely known as a threat towards not only humans but also ecosystems as there are toxic [1]. Most of the cases, they cannot be decomposed into harmless materials. Lead is one of the most toxic heavy metals as mercury, cadmium, chromium and arsenic. Lead has been detected on the road sides [2], exhausted gas from chimney of factories, disposal sites and their acidic leachate, and even in the food and the drinking water from lead tubes. Since lead mostly exists as ion species in environmental water, precipitations by pH control or ion exchanges were used to remove it. Adsorption can be an option to remove lead from, for example, acidic leachate and the leachate at waste disposal landfill [3]. Activated carbons have been widely and practically used for water purification in their forms of powder, granular and fibers [4]. While activated carbons exhibit excellent behavior for removal of organic compounds from aqueous solutions [5], they show poor performance to remove heavy metals [3]. A major difference in adsorption between organic compounds and heavy metals would be principally caused by difference in adsorption sites. In general, activated carbon consists of graphite layers on which oxygen were substituted for edge atoms forming such carbon-oxygen complexes as carboxyl, lactonic, phenolic hydroxyl and carbonyl groups [6]. Heavy metal ions as lead and cadmium would adsorb to a part

of carbon-oxygen functional groups [7], while organic compounds as phenols and chlorinated aromatic compounds can adsorb both on carbon's basal planes and to carbon-oxygen functional groups on the edge of graphite layers [8].

On the other hand, it has been reported that fly ash [9] and some natural inorganic materials [10] as zeolite and clay [11, 12] could be superior adsorbents for heavy metals. Oxygen atoms of metal oxide might be adsorption sites in the inorganic materials. For instance, magnesium oxide is known to be a solid base [13], and it would also uptake a large amount of heavy metals. The magnesium oxide was, therefore, introduced to an activated carbon to improve the adsorption capacity of heavy metals [14]. Influence of pH on lead adsorption and desorption on the adsorbents was also studied.

MATERIALS AND METHODS

Adsorbents

Commercially available granular activated carbon (GAC) was employed as adsorbents. Magnesium nitrate hexahydrate was used to introduce magnesium to the GAC. All chemicals were reagent grade in this study. The 5.0-g GAC was mixed with 500mL of 0.1 mg-Mg/L solution, and then agitated for 3days at 100 rpm and room temperature of 25°C. After the magnesium adsorption reached the equilibrium state, the GAC was separated from the magnesium solution, dried in oven at 110°C over night, and then calcined in air at 350°C for 3 hours. The procedure was repeated for 4 times to increase the amount of magnesium loading. The magnesium loaded GAC was referred to as MgO-GAC. As a reference the GAC was also calcined in air for 3 hours without the magnesium loading.

Pb solutions and pH adjustment

1-L Pb(II) stock solution of 1000 mg-Pb/L was obtained from lead chloride by dissolving it to de-ionized water. Pb(II) solutions of 120 mg/L were prepared from the stock solution for the adsorption experiments. Equilibrium pH of the solutions was adjusted by adding hydrochloric acid. The resultant Pb(II) concentration of the solution to be mixed with the adsorbents was 60 mg/L as the 120 mg/L solution was diluted with the same amount of de-ionized water or hydrochloric acid.

Adsorption experiments and analysis

The 50mL of 60 mg/L Pb(II) solution and 100 mg of GAC or MgO-GAC adsorbent were mixed in 100 mL of conical flask, and the vials were sealed and shaken at 25°C and 100 rpm for 5 days to reach equilibrium. The equilibrium pH was measured by a pH meter. After equilibrium solution was separated from adsorbent by decantation technique. 0.1N hydrochloric acid was added into the Pb(II) solutions to convert all Pb(II) species to Pb^{2+} to minimize errors in quantitative

analysis. The amount of Pb(II) was determined by an atomic absorption spectroscopy (AAS). The amounts of Pb(II) on adsorbents were calculated from difference between initial and equilibrium concentrations of Pb(II).

RESULTS AND DISCUSSION

Effect of magnesium loading

As mentioned in the experimental section, magnesium was repeatedly loading to GAC. **Fig.1** shows the change in percent Pb(II) removal as a function of the number of magnesium loading. A pronounced increase in Pb(II) uptake was observed at 1st and 2nd magnesium loading. **Fig. 2** also represents the change in equilibrium pH as a function of the number of magnesium loading. The increase in Pb(II) adsorption by the 1st magnesium loading would be directly attributed to the introduction of magnesium oxide, because equilibrium pH was not changed from non-magnesium loading. For the 2nd through 4th magnesium loadings, the enhancement of Pb(II) uptake would be due to not only magnesium loading but also pH increase as Pb(II) adsorption was generally improved with increasing pH value above 5-6 [15]. For using precipitation technique, solution pH must be increased up to more than 8.0 to make precipitated $\text{Pb}(\text{OH})_2$. **Fig.3** depicted speciation diagram of Pb(II) at 25°C calculated from formation constants between Pb^{2+} and OH^- . Since the maximum pH value was 7.5 in the study, most of Pb(II) species were limited to Pb^{2+} , $\text{Pb}(\text{OH})^+$, $\text{Pb}_3(\text{OH})_4^{2+}$, and little $\text{Pb}(\text{OH})_2$ can be generated. Surface areas were more than 1000 m^2/g , and mesopore and macro pore defined by IUPAC were dominant for the pore size distribution of all GAC and MgO-GAC's used in the experiments.

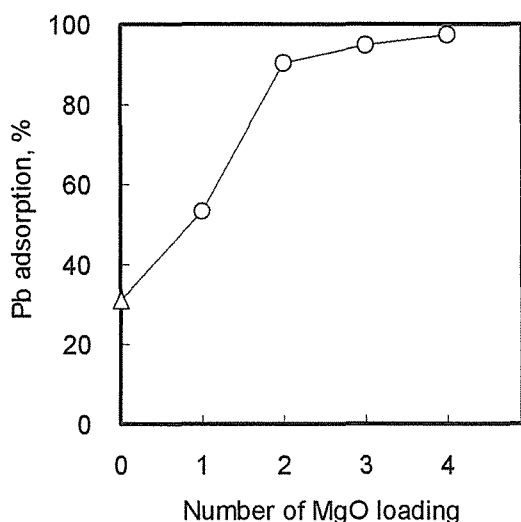


Fig.1 Influence of number of magnesium loading on Pb adsorption

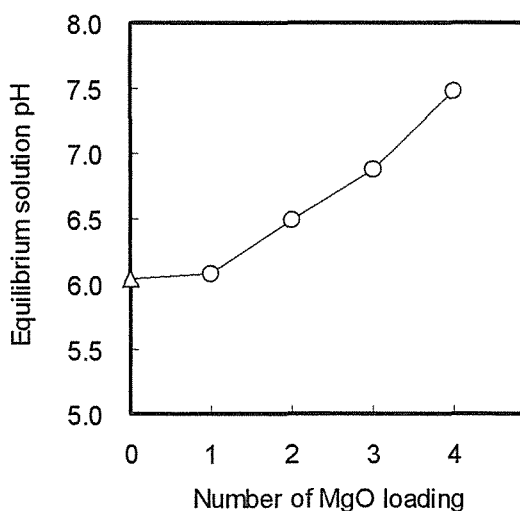


Fig.2 Influence of number of magnesium loading on equilibrium solution pH

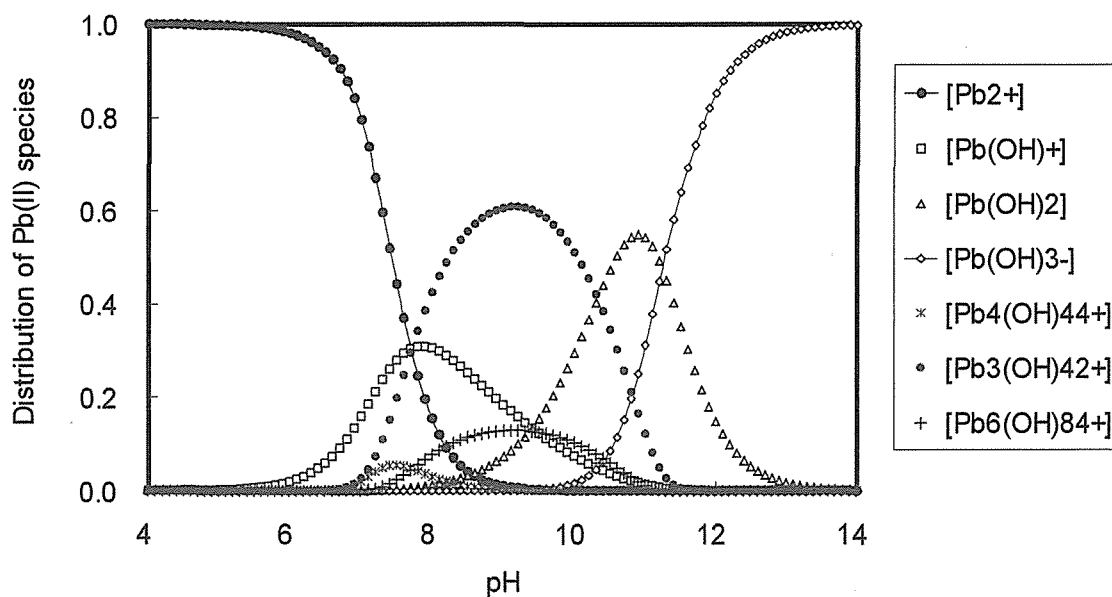


Fig.3 Calculated speciation diagram of Pb(II) as a function of pH
 C_0 ; 50 mg/L of Pb(II)

Influence of pH on Pb(II) adsorption and desorption

Figs.4 and 5 show the influence of pH on Pb(II) adsorption and desorption, respectively, on the MgO-GAC and the GAC. It is obvious that Pb(II) adsorption is more pronounced for MgO-GAC than for GAC at any pH value examined. Even in equilibrium pH less than 3.0, more than 30% of total Pb(II) was remained on the MgO-GAC. Almost all Pb(II) was removed from the adsorbents at pH below 3.0 for most of GAC's [16]. Similarly the amount of Pb(II) released from the adsorbents to the solution in desorption was smaller for MgO-GAC than for GAC, even though the amount of Pb(II) was more in the MgO-GAC than in the GAC. These results reveal that not only increasing the number of adsorption sites but also increasing the strong binding sites for Pb(II) adsorption would be generated by introducing MgO to GAC; most of the surface functional groups could be switched from carbon-oxygen complexes to magnesium-oxygen complex. The increase in Pb(II) adsorption for MgO-GAC was, again, attributed not only to rising in pH but also to the formation of magnesium-oxygen complex. Therefore, combinational effects of strong Pb(II) binding with magnesium-oxygen complex and rising in pH were expected for the MgO-GAC to remove Pb(II) for practical applications of the adsorbents.

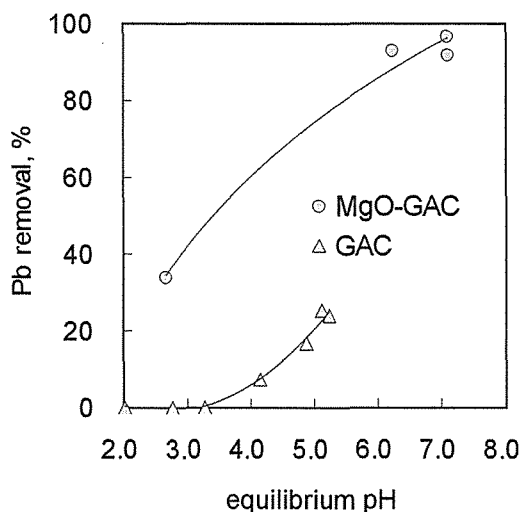


Fig.4 Adsorption of Pb(II) as a function of equilibrium pH

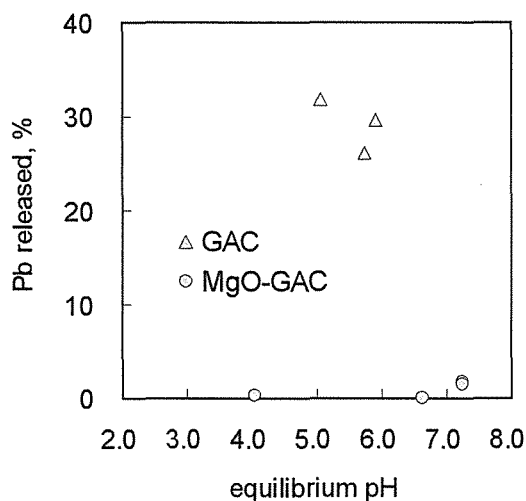


Fig.5 Desorption of Pb(II) as a function of equilibrium pH

CONCLUSIONS

For the enhancement of Pb(II) adsorption, magnesium was loaded onto commercially available granular activated carbon as an adsorbent, and Pb(II) adsorption was examined in aqueous solution by GAC and MgO-GAC. The following conclusions were deduced from the results.

- 1) The amount of Pb(II) adsorption was increased by introducing magnesium oxide, and further improvement of the adsorption was also observed by repeated loading of MgO on GAC.
- 2) By introducing MgO to GAC, not only increasing adsorption capacity for Pb(II) but also generating strong adsorption sites was found out.
- 3) For practical use of MgO-GAC, the Pb(II) adsorption was expected to be enhanced both by the effects of basic solid introduction in itself and by consecutive increase in pH.

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