



Removal of arsenic from an aqueous solution by coprecipitation with manganese oxide



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ABSTRACT

The removal of arsenic (As) from wastes is important to ensure good environmental practice within the nonferrous metallurgical industry. In the present study, the coprecipitation of As and manganese was investigated as a possible means of achieving such removal. As(V) was found to be removed from a Mn–As(V) aqueous solution during stirring. Such removal of As(V) from an aqueous solution is likely caused by coprecipitation with Mn_3O_4 and $Mn(OH)_2$, both of which are assumed to have isoelectric pH of 10.9. Higher removal of As(V) from an aqueous solution was found at pH of 6–10, and can be attributed to the adsorption of $H_2AsO_4^-$ and $HAsO_4^{2-}$ onto the positively charged surface of Mn_3O_4 and $Mn(OH)_2$, which is caused by electrostatic interaction. Conversely, the lower removal of As(V) from an aqueous solution at a pH of 11 can be attributed to the electrostatic repulsion between As(V) species and the negatively charged surfaces of Mn_3O_4 and $Mn(OH)_2$.

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Introduction

Arsenic (As) is a typical unwanted constituent of many of ores and concentrates that are processed in the nonferrous metallurgical industry; accordingly, various types of As-bearing intermediate products and process wastes must be treated in an environmentally acceptable manner. Previously, coprecipitation techniques with Fe(III) species have been proposed to eliminate As from process liquors and effluents [1,2], although As(V) is known to be more amenable to treatment with coprecipitation techniques than As(III). Typically, As(III) is oxidized to As(V) with chlorine before the As(V) can be treated, and the subsequent coprecipitation is caused primarily by the adsorption of As(V) species (i.e., $H_2AsO_4^-$, $HAsO_4^{2-}$, and AsO_4^{3-}) onto $Fe(OH)_3$. The point of zero charge for $Fe(OH)_3$ is reported to occur only at pH of 8 [3]. Thus, adsorption is efficient below a pH of 8 because the surface of $Fe(OH)_3$ is positively charged and As(V) is negatively charged. Conversely, for a pH greater than 8, the surface of $Fe(OH)_3$ is negatively charged, and adsorption is impaired [4].

Wastewater containing As typically also contains various heavy metal ions. For example, hot spring water in Hokkaido, Japan, has been shown to contain Mn, Ni, Cu, and Zn [5,6], and a river in

Hyogo, Japan, yielded Mn, Zn, Fe, and Cu [7]. These wastewaters contain more Mn than other heavy metals, and Mn is known to be a particularly harmful element [8]. Therefore, we have developed a new treatment method for As involving coprecipitation with Mn contained in wastewater. The primary advantage of this method is that it avoids the need to add Fe(III) salt, while also facilitating effective utilization of Mn in wastewater to treat As in the same wastewater. In the present study, the removal of As(V) from an aqueous solution using Mn was investigated. In particular, the effects of Mn/As molar ratio, pH, and initial concentration on the removal of As(V) during the stirring of a Mn–As(V) mixed solution were examined. Then, to allow consideration of the removal of As(V), the isoelectric point of the precipitate from a Mn solution was estimated.

Experimental

Atomic mass of As and Mn is 74.92 and 54.94 g/mol, respectively. Manganese solutions with initial concentrations of 10, 30, 50, and 100 mM were prepared by dissolving various amounts of $MnSO_4 \cdot 5H_2O$ in deionized water. Then, $Na_2HAsO_4 \cdot 7H_2O$ was added to the Mn solutions at Mn/As molar ratios of 1, 3, 5, and 10 to prepare Mn–As(V) mixed solutions. In this case, initial As(V) concentrations were 10 mM; 500 mL of Mn–As(V) mixed solution was stirred at 30 °C, and the initial pH was adjusted to 6–11. The resultant suspension was kept standing at 30 °C for 120 min with stirring. Samples of the suspension were

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withdrawn at various time intervals and immediately filtered through a 0.45 μm membrane filter. The filtrates were analyzed for residual As and Mn using inductively coupled plasma-atomic emission spectrometry (ICP-AES). The precipitate was washed repeatedly with deionized water and dried under reduced pressure (133 Pa) at 40 $^{\circ}\text{C}$ for 40 h. The precipitate was analyzed by making X-ray diffraction (XRD) measurements using Cu $\text{K}\alpha$ radiation. The precipitate from Mn-only solution without As(V) was also analyzed using XRD.

To examine the effects of initial As(V) concentration, initial As(V) concentrations were also prepared at 10 and 100 mg/L. Furthermore, the isoelectric point of precipitate from Mn solution was estimated. To achieve this, 500 mL of 100 mM Mn solution was stirred at 30 $^{\circ}\text{C}$, and the initial pH was adjusted to 9–12. Then, 100 mL of the resultant suspension was added to a mess cylinder immediately, and the height of sedimentation in 10 min was measured.

Results and discussion

Fig. 1 illustrates the effect of Mn/As molar ratio on the residual As(V) concentration during the stirring of a Mn–As(V) mixed solution at pH of 6 and 8. At both pH values, the residual As(V) concentrations decreased rapidly with time at all Mn/As molar ratios. Thus, it is clear that As(V) was removed from aqueous

solution using Mn. In addition, the residual As(V) concentration decreased with increasing Mn/As molar ratio at any time, which we attribute to the increasing amount of Mn precipitate. Moreover, for the same Mn/As molar ratios, the residual As(V) concentrations at pH of 8 were lower than those at pH of 6 for any given time, suggesting that As(V) removal was affected by pH. To investigate this further, the effect of pH on the residual As(V) and Mn concentrations during the stirring of the Mn–As(V) mixed solution with Mn/As molar ratio of 10 for a period of 120 min was examined (Fig. 2). The residual As(V) concentration decreased as pH increased from 6 to 8; then, the concentration remained constant until pH reached 9, before increasing with increasing pH from 9 to 11. Overall, the residual As(V) concentration was found to be lowest (highest) for pH of 8–9 (11). In contrast, the residual Mn concentration remained almost constant over the pH range 6–8 and decreased with increasing pH from 8 to 11. At all pH values, the Mn concentration decreased from the initial value (100 mM), indicating the precipitation of a Mn compound. Fig. 3 illustrates the XRD patterns for precipitates from both the Mn solution and the mixed Mn–As(V) solution with Mn/As molar ratio of 10 at pH of 9 for 120 min. The precipitate from the Mn solution was ascribed to Mn_3O_4 and $\text{Mn}(\text{OH})_2$. Firstly, $\text{Mn}(\text{OH})_2$ was probably precipitated from Mn solution, and then some Mn(II) in $\text{Mn}(\text{OH})_2$ was oxidized to Mn(III), leading to the formation of

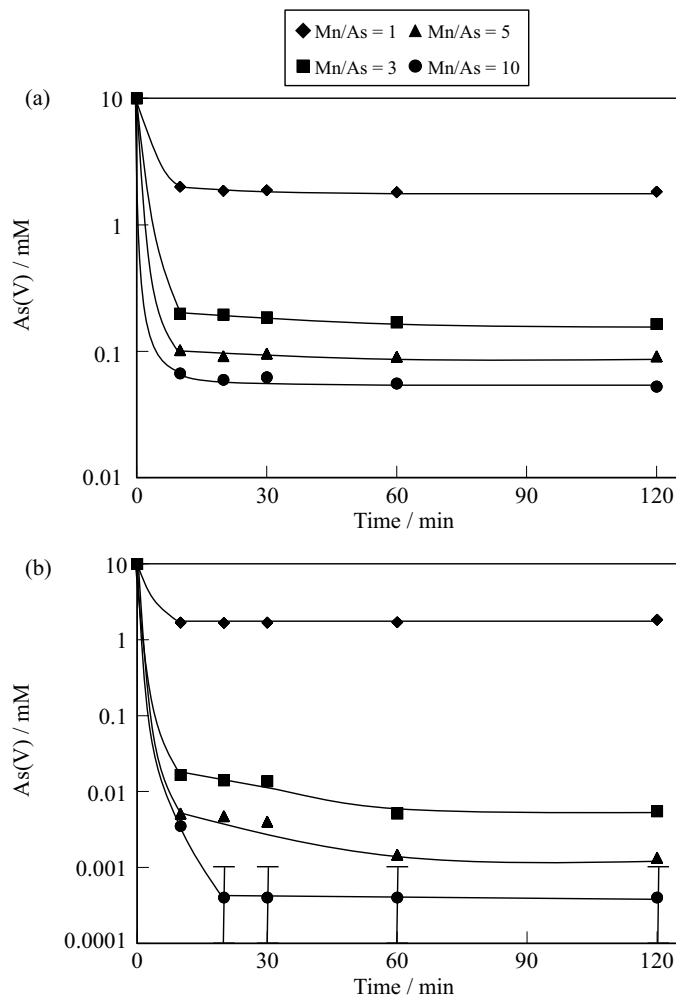


Fig. 1. Effect of Mn/As molar ratio on the residual As(V) concentration during the stirring of Mn–As(V) mixed solution at pH of (a) 6 and (b) 8 (initial As(V) concentration: 10 mM).

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