



A novel method for remediation of nickel containing wastewater at neutral conditions



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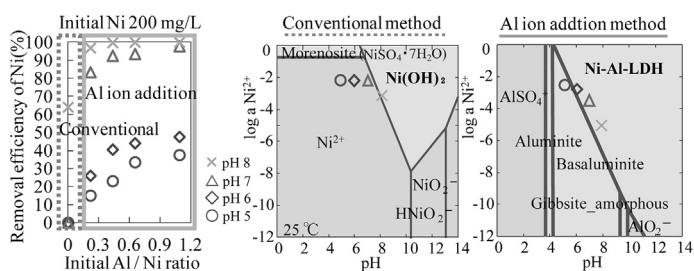
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HIGHLIGHTS

- High Ni removal efficiencies at neutral pH conditions were achieved.
- Layered double hydroxide was selectively precipitated by the addition of Al ions.
- Ni was mainly incorporated into the hydroxide structure at pH 7 and 8.
- A thermodynamic geochemical model reproduced the experimental results well.
- Ni can be removed by adding a small amount of Al ions in practical wastewater treatment.

GRAPHICAL ABSTRACT



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ABSTRACT

Heavy metals contained in wastewater are generally removed by adding antalkaline to increase the pH, and Ni is commonly precipitated as Ni-hydroxides at pH 10. However, a more sustainable remediation method of treatment at neutral conditions would be attractive due to the high cost of chemical reagents and inefficient treatment at present. Based on natural attenuation, the method of adding Al ions has been used in wastewater treatment to precipitate layered double hydroxides (LDH). Here, we investigated the use of Al ion addition in the Ni containing wastewater treatment, experimentally and thermodynamically. By co-precipitation experiments adding Al ions to Ni-containing water, Ni was selectively incorporated into the structure of LDH, and the removal efficiency of Ni was close to 100% even in pH 7 and 8 samples (lower pH than conventional methods) with initial Ni concentrations of 200–10,000 mg/L. Geochemical modeling results replicate the experimental results well when the Al/Ni ratio of LDH is assumed to be 0.33. This model makes it possible to estimate the amount of Al ions and additive agents necessary for use in treatment of wastewater containing different Ni concentrations.

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1. Introduction

Advances in technology such as the electronics and metal plating industries have increased the demand for Ni [1], and some industrial wastewaters contain large amounts of Ni, leading to environmental pollution as excessive intake of heavy metals is harmful to humans and as wastewaters lead to environmental pollution. Hence, there is a need to maintain the environmental standards for

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Ni and to dispose properly of stable precipitation products to protect human health and protect natural environments. At the same time, Ni-rich mines and grades of Ni ore have been decreasing with the increase in mined quantities of Ni [2]. Additionally, resources are unevenly distributed and found only in a few countries making the supply sources of Ni vulnerable. Because of this, there is an urgent need to recover Ni from wastewaters and other waste forms.

Current wastewater treatments can be divided into biological [3–8] and chemical treatment methods [9–15]. Chemical precipitation processes (especially hydroxide precipitation) are widely used among these treatments [16,17]. Generally, the removal of Ni from wastewater by adding antalkaline and flocculants to increase the pH to 10 (the lowest solubility of Ni) or above result in the generation of Ni-hydroxides, and after treatment it is necessary to decrease the pH below the effluent standards of Japan (pH 5.8–8.6) before discharge. However, the current method suffers from disadvantages including the high cost of antalkaline, flocculant reagents, and acid neutralizing agents. Therefore, it would be useful to develop a lower-cost and still effective wastewater treatment.

The concentration of heavy metals and toxic ions in soils and ground waters may be attenuated in natural processes such as neutralization and mineral precipitation [18,19]. The natural attenuation processes may be applied to wastewater treatment, something which would likely be safer, more environmentally friendly, and more cost-effective, than conventional methods. For example, a study [20] has clarified the natural attenuation mechanism of Cu and Zn ions at Dougamaru, an abandoned mine in Japan. The mine drainage here contains higher concentrations of Cu and Zn ions than effluent regulatory limits in Japan allow (3 mg/L for Cu and 2 mg/L for Zn). However, Cu and Zn in the mine drainage are incorporated in the structure of precipitates composed of Cu-bearing layered double hydroxides, hydro-woodwardite at around pH 6 because this is produced by the presence of Al ions in the drainage, resulting in natural attenuation. The general chemical composition formula of layered double hydroxide (LDH) is represented by $[M^{2+}_{1-x}M^{3+}_x(OH)_2(A^{n-})_{x/n} \cdot mH_2O]$ ($0.20 < x < 0.33$), and LDH is classified as anionic clay because of its anion exchange capacity [21,22]. It has a structure of octahedral hydroxide layers with alternating laminated interlayers of anions (A^{n-}) and interlayer water. In LDH, substitution of the divalent cation (M^{2+}) with a trivalent cation (M^{3+}) produces a positive charge on the hydroxide layers, requiring the presence of anions between the hydroxide layers to maintain the charge balance. Nickel could be incorporated in the LDH octahedral sheets because hydroxide layers of LDH are essentially composed of metal ions with coordination number 6. To further explore this, the objective of this study is to investigate the optimum conditions such as pH, Al, and Ni concentrations under which Ni-LDH is precipitated by the addition of Al ions and compare this with the conventional method to precipitate $Ni(OH)_2$, to understand the behavior of Ni in the precipitation process, and to develop a geochemical model to reproduce the experimental results. With these results, the final goal is to develop a remediation method for Ni-bearing wastewaters at neutral conditions and to investigate the applicability of the Al ion addition method for Ni-containing water using the model.

2. Experimental

Co-precipitation experiments were carried out to synthesize Ni-bearing LDHs with different Al/Ni ratios to confirm the applicability of LDH in Ni-bearing wastewater treatment. However, the uptake mechanism of Ni is considered to be complex because multiple slightly soluble salts are simultaneously precipitated in the co-precipitation processes. Therefore, adsorption experiments were

also carried out to elucidate the adsorption behavior of Ni onto the precipitated phases. In addition, extraction experiments were carried out to establish details of the chemical state of Ni in the precipitated phases.

2.1. Co-precipitation experiments

Co-precipitation experiments were carried out in Ni-containing water with SO_4^{2-} by Al ion addition under various conditions (pH, Al, and Ni concentrations) to establish whether Ni-LDH is precipitated and what the precipitating conditions of Ni-LDH are. Initially, simulated wastewaters were obtained by mixing $NiSO_4 \cdot 6H_2O$ and $Al_2(SO_4)_3 \cdot 14-18H_2O$ (Kanto Chemical Co., Inc., Guaranteed reagent) with 150 mL deionized water in 250 mL polyethylene bottles, then alkaline titrated by adding 1.0 M NaOH was below 1 mL/min at 25 °C, and adjusted to set pH values with vigorous stirring (300 rpm). The initial Ni concentrations were set to 200, 1000, and 10,000 mg/L because the actual wastewater contains various Ni concentrations, and the resulting initial Al/Ni molar ratios were 0, 0.218, 0.435, 0.653, and 1.087 to for determining the influence of the amount of Al ions. The resulting molar ratio is complicated by the fact that the unit of the initial concentration was set to “mg/L” as widely used in wastewater treatment (f.ex., the initial Al/Ni molar ratios of the solution with initial Ni and Al ion concentrations were respectively 1000 mg/L and 200 mg/L is 0.218). The pH values were 5.0, 6.0, 7.0, and 8.0 (± 0.1) allowing confirmation of the precipitates at neutral conditions. After titration, the suspensions were left standing for 24 h at 25 °C. The supernatant solutions were collected by centrifugation for 40 min at 3000 rpm, and the solid phases were recovered after solid-liquid separation.

The extraction experiment is a quantitative evaluation achieved by dissolving or extracting only specific components of the solid or liquid using different types of solvents. The first step was deionized water (pH 5.8) extraction for water soluble ions from the solid phases and the second was $CaCl_2$ extraction for the exchangeable ions around pH 6.5, using $CaCl_2$ solutions of 3.0, 15, and 150 mmol/L (in solutions respectively with 200, 1000, and 10,000 mg/L of initial Ni) [23,24]. The maximum concentration where Ni was adsorbed was considered the Ca concentration. The solid and suspension were shaking for 24 h at 200 rpm at 25 °C and the liquid-solid ratio was 500 mL/g, and then were separated.

Adsorption experiments were also carried out to determine the Ni adsorption behavior of each of main precipitates generated by the co-precipitation experiments. The adsorption solutions were 30 mL with Ni concentrations of 10, 50, 100, 300, and 1000 mg/L using $Ni(NO_3)_2 \cdot 6H_2O$ suspended in 0.02 M $NaNO_3$ as a supporting electrolyte solution at pH 5.45–5.60. Next, 100 mg of the solid phases were added to the solutions (giving a liquid-solid ratio of 300 mL/g). The solid and liquid phases were separated for the further analysis after shaking for 24 h at 25 °C.

2.2. Analytical methods

The concentrations of Ni and Al ions contained in the recovered solutions after the treatments were analyzed by inductively coupled plasma atomic emission spectrometry (ICPE-9000, Shimadzu). The SO_4^{2-} ions contained in the supernatant were quantitatively analyzed by using ion chromatography (Metrohm 861 Advanced Compact IC). Compact IC is comprised of 3 consecutive suppressor units that are used for suppression, regeneration with sulfuric acid (0.04 M), and rinsing with ultrapure water. The eluent was diluted mixed solution of Na_2CO_3 (2.70 mM) and $NaHCO_3$ (0.30 mM). The column was set to 35–38 °C.

The solid phases were freeze dried and were then analyzed to determine the mineral component of the precipitates. The minerals contained in the solid phases were analyzed by X-ray diffraction.

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