Recycling of High-Purity Strontianite and Hematite from Strontium-Bearing Sludge

Rui Bian, Ting Su, Yu Chen, Zhan Qu, Suiyi Zhu,* Xi Tian, and Yang Huo*

ABSTRACT: Sr-bearing sludge is a hazardous waste that is commonly generated by nuclear power plants and mineral refining operations. In this work, Sr-bearing sludge was simulated and then cleanly recycled into high-purity strontianite with hematite nanoparticles as a byproduct via a novel hematite precipitation route. The sludge contained 26.1% Fe, 3.5% Sr, and Si impurities. After dissolution in 1.2 M nitric acid, the sludge was treated hydrothermally with the addition of glycol to precipitate Fe effectively. Without the addition of glycol, only 52% Fe was hydrothermally precipitated in the form of hematite aggregates. With the addition of glycol at the optimal molar ratio of 0.4, nearly 100% Fe was removed in the form of hematite nanoparticles with an average diameter of 50 nm, whereas over 98% of Sr was retained in the leachate. The generated hematite was highly purified with an Fe2O3 content of 95.23%. Sr was present at a high concentration of 3.9 g/L in the treated leachate and further precipitated in the form of strontianite with a purity of 96.8% through Na2CO3 addition. Tertiary butanol (TeB) exhibited a similar Fe removal rate as glycol even though its optimal molar ratio was 0.1, which was approximately one-fourth the optimal molar ratio. Fe removal involved spontaneous Fe3+ hydrolysis under hydrothermal conditions and was promoted by increasing the pH of the redox reaction between nitrate and glycol and/or TeB. The method reported here successfully enabled the resource recycling of Sr-bearing sludge to generate high-purity strontianite and hematite products without producing any secondary waste.

1. INTRODUCTION

Nuclear power is a clean and cost-effective energy source and is commonly applied in more than 30 countries worldwide.1 Approximately 13% of the power in the world is supplied by nuclear energy.7 In nuclear power plants, Sr is generated as an important fission product of U and Pu in light water reactors3 and is present at concentrations of 1–10 mg/L in discharged wastewater.2,4 Conventionally, Sr-bearing wastewater is chemically precipitated through the addition of a flocculant; this approach results in the mass production of waste Sr-bearing sludge.5,6 This type of sludge is considered as low-level waste and is produced at the rate of 460 tons per 1000 MWe electricity in nuclear plants.7 Sr is the major contaminant of this type of sludge and easily released in the form of free ions into water with rain6 and/or underflow9 and spreads with runoff for random exposure in the environment, where it poses harm to nearby microbes and human health.8,10 Storing sludge in an insoluble form and sanitary landfills are appropriate treatment processes. The transformation of sludge into glasslike solids, ceramic–metallic forms, or bituminized solids has been performed in North America,9 Eastern Europe,5,10 and Indian districts.11 However, this process is tedious and complicated and consumes high amounts of carrier materials and power energy.5,12

Sr is a costly ingredient that is in high market demand for the production of high-quality alloy, special ceramics, and advanced luminescent materials.13 The resource recycling of Sr from waste Sr-bearing sludge is a valuable and feasible approach for reducing sludge production and pollution. Sludge is first dissolved in sulfuric, hydrochloric, and/or nitric acids14,15 to produce a Sr-bearing leachate used to recycle high-purity Sr-bearing products. Fe impurities in the sludge are dissolved by strong acids and decrease to residual concentrations of 5.7–44.7 g/L in the leachate.16 Second, Sr is separated from Sr-bearing leachate through the following approaches: cationic exchange,17 chemical precipitation,18,19 solvent extraction,20−22 and membrane filtration.23,24 However, Fe is present in high concentrations in Sr-bearing leachate and shows high affinity with resins, extraction agents, and membranes.25−27 For example, although the use of di-t-butylcyclohexano-18-crown-6 results in efficient Sr extraction
from the acidic leaching solution of a radioactive tank sludge, this approach results in Fe extraction and retention at a concentration of 12 mg/L in the extracting solution. Such defects limit the applications of this method for high-purity Sr recovery. In addition, Fe is an active cation and easily reacts with precipitators, such as sulfide, hydroxide, and carbonate. This characteristic accounts for the high Fe impurity concentrations of recycled Sr-bearing products. Therefore, Fe should be removed prior to Sr removal in Sr separation from Sr-bearing leachate.

Fe is rapidly hydrolyzed into Fe oxhydroxide in the leachate and further converted into highly crystallized Fe oxides, e.g., akageneite and hematite. Lu et al. achieved approximately 30% Fe and 0.1% Cu removal from Fe/Cu-bearing leachate generated through chalcopryite leaching with hydrochloric acid and subsequent hydrothermal treatment at 155 °C for 60 min. Fe removal rates can be increased to above 85% when the temperature is increased to 200 °C and to nearly 90.7% in the presence of H2O2. Although a large portion of Fe is removed from the leaching acid, residual Fe is present in high concentrations (nearly 1.5 g/L) and thereby requiring further removal to obtain high-purity Sr-bearing products.

This study aimed to remove Fe impurities efficiently from the leaching acid of Sr-bearing sludge via a novel hematite precipitation route. In contrast to the leachate with high residual Fe contents obtained through the conventional method, the leachate treated through the present method had an Fe concentration of less than 8 mg/L. This characteristic enabled the recycling of high-purity strontianite and hematite byproducts from Sr-bearing sludge. Glycol and tertiary butanol (TeB) were used as auxiliaries to promote Fe precipitation, and their dosages were also optimized.

2. MATERIALS AND METHODS

2.1. Sr-Bearing Sludge. Sr-bearing sludge was generated from simulated Sr-bearing wastewater through the following steps (Figure 1). First, 34.1 mg of SrCl2·6H2O was dispersed in 2 L of deionized water to simulate Sr-bearing wastewater. Second, 0.36 g of FeCl3·6H2O was added to the wastewater. The wastewater was then stirred at 70 rpm for 15 min and then made to stand for 30 min to form a brown sludge. The generated sludge was collected and vacuum dried at 55 °C for 10 h. The major composition of the sludge was characterized through X-ray fluorescence (XRF, Rint2200, Rigaku, Japan). The sludge contained 26.1 wt % Fe and 3.5 wt % Sr (Table 1).

2.2. Fe Separation from Sludge. High-purity hematite was recycled from the sludge via the following steps (Figure 1). First, the sludge was dispersed in 1.2 M nitric acid under constant stirring at 90 rpm for 24 h to generate an Fe/Sr-bearing leachate, and the undissolved solid was discarded. Second, the leachate was adjusted to pH 0.2 using 20% NaOH. Third, 30 mL of the leachate was added to a 50 mL Teflon vessel. Then, glycol or TeB was added. Fourth, the vessel was heated at 160 °C for 10 h and then water-cooled to below 25 °C. Fifth, the reddish deposit generated at the bottom of the vessel was collected and dried at 105 °C for 3 h, and the supernatant was collected for recycling the Sr-bearing product. Glycol dosage was optimized by varying the Mglycol/Mnitrate molar ratio from 0.2 to 0.4 and 1 in accordance with the above-mentioned steps. In contrast to glycol, TeB has four structural carbon atoms and one hydroxyl group, and its effect on Fe precipitation was investigated by following the above steps.

2.3. Recycling of Sr-Bearing Products from the Treated Leachate. Sr that remained in the supernatant after Fe precipitation was filtered and collected. The treated leachate was collected and then mixed with 5 g/L Na2CO3 under constant stirring at 70 rpm for 5 h. A whitish precipitate was generated and then collected for freeze-drying at ~80 °C for 24 h.

2.4. Characterization. The Fe and Sr contents in leachate before and after treatment were measured using an inductively coupled plasma emission spectrometer (ICP-OES, Avio-200, PerkinElmer). The nitrate concentration and pH value of the leachate were determined by applying an ion chromatography (Metrohm AG, Switzerland) apparatus and a pH meter (S210-S, Mettler Toledo), respectively. The total organic carbon (TOC) concentration in leachate before and after the hydrothermal reaction was detected with a TOC analyser (TOC-500, Shimadzu, Japan). The morphologies of the sludge and deposits were characterized using an X-ray diffractometer (XRD, Rigaku D/max, Rigaku, Japan) with Cu Ka radiation.

3. RESULTS AND DISCUSSION

3.1. Effective Fe Removal. The effect of glycol dosage on Fe precipitation was investigated (Figure 2). Without glycol, an Fe removal rate of only 52% was achieved, whereas Sr loss was below 2%. With the addition of glycol, the Fe removal rate considerably increased to 93.7% and further rose to 99.97% but decreased to 71.5% as the Mglycol/Mnitrate molar ratio was increased from 0.2 to 0.4 and 1. In parallel, Sr loss was initially 1.3%, then slightly increased to 1.9% and further rose to approximately 10% when the Mglycol/Mnitrate molar ratio was elevated from 0.2 to 0.4 and then to 1. Thus, the optimal molar ratio of glycol to nitrate was 0.4. Nearly 100% Fe precipitation with the Sr loss of 1.9% was achieved using this molar ratio.

In the absence of glycol, Fe was precipitated in the form of an irregular hematite block (Figures 3A and 4A). Smooth
hematite aggregates were generated by adding glycol at the molar ratio of 0.2 (Figures 3B and 4B). As the molar ratio of glycol was increased from 0.2 to 0.4, smooth aggregates disappeared and small spherical particles with an average diameter of 50 nm appeared instead (Figure 3C). These particles were identified as hematite units (Figure 4C). However, when the molar ratio of glycol was 1, the precipitated product was obtained in the form of a mixture of large microrods and fine particles (Figure 3D,E). The microrods consisted of humboldtine, and the small particles were affiliated with hematite (Figure 4D). The mechanism of Fe precipitation is discussed in Section 3.4.

Nitrate concentrations and TOC in the treated leachate were determined. As the molar ratio was increased from 0.2 to 0.4 and 1, nitrate was consumed, and its concentration decreased from 52.4 g/L to 11.6 and 0.2 g/L (Figure 5A). Accordingly, the leachate pH increased steadily from 0.2 to 0.5 and 2.7 (Figure 5C). However, the residual TOC concentration was less than 0.2 g/L at the molar ratio of 0.2 and 0.4 but increased to 22 g/L at the molar ratio of 1 (Figure 5B). This outcome demonstrated that nitrate reduction was related to glycol addition and that high pH played a key role in Fe precipitation.

3.2. Time-Course of Fe Precipitation in Sr-Bearing Leachate. The time for the hydrothermal removal of Fe was also investigated (Figure 6). The Fe removal rate reached 81% at the initial 0.5 h, steadily increased to 97.3% after 3 h, and was close to 100% after 10 h. By contrast, Sr loss was less than 0.5% in the initial 3 h and slightly increased to 1.9% after 10 h. During Fe removal, nitrate concentration decreased rapidly from 29.9 g/L to 17.1 and 11.6 g/L as the hydrothermal treatment time was extended from 0.5 h to 1 and 10 h. Similar to nitrate, TOC decreased sharply from 0.21 g/L after 0.5 h of treatment and steadily rose to 0.12 g/L at 10 h of treatment. The consumption of added glycol in the hydrothermal process resulted in an increase in leachate pH from the initial value of 0.1 to 0.55 after 10 h.

No Fe-bearing precipitate was observed at the hydrothermal treatment time of <0.5 h. However, Fe was rapidly precipitated in the form of hematite aggregates at 0.5 h (Figures 7A and 8A), remained almost unchanged at 1 h (Figures 7B and 8B), and was converted into strawberry-shaped hematite at 3 h (Figures 7C and 8C) and spherical hematite nanoparticles at 10 h (Figures 7C and 8C). These results suggested that free Fe³⁺ was directly hydrolyzed into well-crystallized hematite particles in the hydrothermal process.

3.3. Effect of Tertiary Butanol on Fe Precipitation. The structural carbon content of TeB is 4, which is twice that of glycol. The dosage of TeB for Fe precipitation was optimized (Figure 9). As the $M_{TeB}/M_{nitrate}$ molar ratio was increased from 0.1 to 1, the Fe removal rate steadily decreased from 99.9 to 91%, and Sr loss steadily elevated from 1.6 to 15.1%. Accordingly, nitrate concentration decreased from 31.4 to 0.14 g/L, whereas the leachate pH increased from 0.27 to 2.43. Thus, although the Fe removal rate of TeB was similar to that of glycol, the optimal molar ratio of TeB for Fe precipitation was 0.1 or nearly one-fourth that of glycol. Hematite nanoparticles were also generated at the desirable $M_{TeB}/M_{nitrate}$ ratio of 0.1 (Figures 10A and 11A) and were similar to those generated with the addition of glycol (Figure 3C). The morphology of hematite nanoparticles remained almost unchanged at the molar ratio of 0.2 (Figures 10B and 11B) but coarsened as the molar ratio of TeB was increased from 0.2 to 0.4 (Figures 10C and 11C). Such an outcome was likely due to the formation of humboldtine at the hematite crystal surface. However, when the $M_{TeB}/M_{nitrate}$ molar ratio was 1, the Fe-bearing precipitates turned into a mixture of hematite and crystallized humboldtine microrods (Figures 10D and 11D,E). This change was suggestive of TeB overdosing, which promoted humboldtine formation.

3.4. Mechanism of Fe Precipitation. In the Sr-bearing leachate, Fe impurities were predominantly present in the form of Fe³⁺ and hydrothermally hydrolyzed into hematite aggregates. Glycol and TeB play an important role in this process (Figure 12). Fe³⁺ hydrolysis spontaneously occurred.
under hydrothermal conditions. H+ generation during Fe3+ hydrolysis accounted for the reduction in leachate pH from 0.13 to 0.02. The produced hematite also aggregated and provided numerous surface hydroxyl groups for coordinating metal cations and Sr2+. However, in strong acids, H+ was abundant and could replace the coordinated Sr on hematite surfaces to regenerate surface hydroxyl groups with the release of free Sr into the leachate. This outcome demonstrated that the coordination of Sr on the hematite surface was inhibited. As the leachate pH decreased from 0.13 to 0.02, Fe3+ hydrolysis in the leachate reached an equilibrium, and approximately 48% of Fe3+ was retained in the leachate.

Fe was continuously hydrolyzed into hematite with the introduction of glycol and/or TeB into the hydrothermal system. Glycol and/or TeB are organics that can be oxidized by nitrate to generate the intermediate oxalic acid (eqs 1 and 2) with the final products of CO2 and H2O (eqs 3 and 4). H+ consumption during this process raised the leachate pH from 0.13 to 2.7. Thus, Fe3+ hydrolysis continued and considerably decreased the concentration of residual Fe3+ in the leachate.

\[
8H^+ + 8NO_3^- + 5(CH_2OH)_2 = 4N_2 + 5(COOH)_2 + 14H_2O \tag{1}
\]
Glycol and TeB could react with nitrate to consume H⁺ (eqs 3 and 4). This process led to the continuous hydrolysis and precipitation of Fe in the leachate. In the reaction, the oxidation of 1 M glycol consumed 2 M H⁺ and 2 M NO₃⁻ (eq 3), which was nearly half of that consumed by the oxidation of TeB (eq 4). This result demonstrated that compared with glycol, TeB employed more electrons for nitrate oxidation. Other organics, e.g., glucose, ethylenediaminetetraacetic acid (EDTA), and acetic acid, also react with nitrate to consume H⁺. Thus, organics with low cost and high molecular weight can also be used as desirable reagents for the hydrothermal precipitation of Fe³⁺.

In the presence of excessive glycol and/or TeB, nitrate was consumed and was present in concentrations that were inadequate for oxidizing the two organics completely. Therefore, Fe³⁺ was reduced to Fe²⁺ by the organics. Subsequently, the intermediate oxalic acid was accumulated, and two hydroxyl groups were used to coordinate divalent metal ions, e.g., Sr²⁺ and Fe²⁺. Oxalic acid reacted with Fe²⁺ to generate ferrous oxalate and was further precipitated in the form of humboldtine when the leachate became saturated with ferrous oxalate. Thus, residual Fe appeared in the leachate even though the leachate pH had increased to 2.7. Similarly, Sr²⁺ was precipitated in the presence of oxalic acid. This effect...
resulted in the Sr loss of 15%. TeB contained four carbon atoms in its molecule and consumed more nitrate and H⁺ in its oxidation than glycol.

3.5. Generation of High-Purity Sr-Bearing Products. At the optimal \( \frac{M_{\text{glycol}}}{M_{\text{nitrate}}} \) molar ratio of 0.4, nearly 100% of Fe was removed in the form of hematite particles with an \( \text{Fe}_2\text{O}_3 \) content of 95.23% (Figure 13A). In the treated leachate, residual Fe was present at a concentration of 7.9 mg/L, and residual Sr was present at a concentration of 3.9 g/L and further precipitated with the addition of 5 g/L \( \text{Na}_2\text{CO}_3 \). Sr-bearing particles were precipitated in the form of whitish irregular strontianite particles (Figures 14 and 15) with a \( \text{SrCO}_3 \) content of 96.8% (Figure 13B). Thus, high-purity strontianite was successfully recycled from the Sr-bearing sludge.

3.6. Environmental Application. This method exhibited two merits in the recycling of Sr from wastewater. First, Sr was effectively enriched and recycled as a highly purified strontianite, which was a costly industrial active pharmaceutical ingredient. Second, the added flocculant, FeCl₃·6H₂O, introduced impurity Fe into the generated sludge. However, after the sludge dissolution, the impurity Fe was effectively precipitated as hematite nanoparticles. Such hematite nanoparticles contained 95.23 wt % \( \text{Fe}_2\text{O}_3 \) and were of great value in the economy. FeCl₃·6H₂O is commonly used as a flocculant to coprecipitate heavy metals (e.g., Cu and Cr) from wastewater, and its addition introduces Fe impurities into the generated sludge. However, Fe impurities can be effectively precipitated in the form of hematite nanoparticles. Thus, the method also has potential applications in the recycling of other heavy metals from wastewater.

4. CONCLUSIONS

Fe impurities were successfully precipitated from the leaching acid of Sr-bearing sludge. Fe impurities were present in the form of Fe³⁺ at a high concentration of 27.8 g/L in the leachate obtained by dissolving sludge in 1.2 M nitric acid. Approximately 52% of Fe³⁺ was hydrothermally precipitated in the form of hematite aggregates without the addition of glycol and/or TeB. With the addition of glycol at the \( \frac{M_{\text{glycol}}}{M_{\text{nitrate}}} \) molar ratio of 0.4, nearly 100% of the Fe was hydrothermally precipitated in the form of hematite particles, with the Sr loss of <2%. The precipitated hematite particles were approximately 50 nm in size and contained 95.23% \( \text{Fe}_2\text{O}_3 \) with an impurity of 0.6% Sr. The residual Sr in the acid was 3.9 g/L and further precipitated as strontianite with a purity of 96.8% with the addition of 5 g/L \( \text{Na}_2\text{CO}_3 \). The Fe removal rate of TeB was similar to that of glycol. However, the optimal
molar ratio of TeB was 0.1, which was one-fourth that of glycol.

**AUTHOR INFORMATION**

**Corresponding Authors**

Suiyi Zhu – School of Environment, Northeast Normal University, Changchun 130117, China; Changchun Institute of Technology, Changchun 130117, China; orcid.org/0000-0002-9784-2698; Email: papermanuscript@126.com

Yang Huo – Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China; orcid.org/0000-0002-4477-4636; Phone: 86-0431-89165610; Email: huo0814@outlook.com

**Authors**

Rui Bian – School of Environment, Northeast Normal University, Changchun 130117, China

Ting Su – School of Environment, Northeast Normal University, Changchun 130117, China

Yu Chen – Jilin Institute of Forestry Survey and Design, Changchun 130022, China

Zhan Qu – School of Environment, Northeast Normal University, Changchun 130117, China

Xi Tian – Changchun Institute of Technology, Changchun 130117, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.0c01539

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**Notes**

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