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Rare Earth Extraction from NdFeB Magnets

Jiro Kitagawa and Masami Tsubota

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Abstract

There is a considerable interest in the extraction of rare earths (RE) from NdFeB magnets in order to recycle rare earth elements. Although the wet process using acid is in practical use in the in-plant recycle of sludge, higher selectivity between rare earths and Fe at room temperature is desired. We have recently proposed a pretreatment of corrosion before the hydrochloric acid (HCl) leaching and the oxalic acid precipitation. Almost full recovery of rare earths can be achieved even at room temperature process. In practical extraction methods, employing wet processes, the discharge of waste acid solution is a problem that needs to be solved to reduce the environmental impact. We further present an encouraging demonstration of rare earth extraction from NdFeB magnet using a closed-loop HCl-based process. Triple extraction has been conducted, and the recovery ratio of rare earths is approximately 50% in each extraction, which is reduced from almost 100% recovery in a one-shot extraction. Despite the reduced extraction efficiency, our method with a rather small number of procedures at almost room temperature is still highly advantageous in terms of cost and environmental friendliness. This study represents the initial step toward the realization of a closed-loop acid process in the recycling of rare earth elements.

Keywords: recycle, corrosion, closed-loop acid process

1. Introduction

Rare earths (RE) are widely consumed in polish, catalysts, rare earth magnets, and so on [1]. Due to the skewed distribution of production countries for RE, many countries depend on the imports from other countries. For example, in Japan, the amount of import of RE metals reached 6479 tons in 2014. The import price, severely depending on international markets, fluctuates widely. Japan is promoting the provisions such as development of alternative materials and the recycle of rare earths. Because the demand of NdFeB magnets has been growing rapidly in recent years because of their use in motors of electric vehicles, wind

turbines, etc., the recycling of RE elements extracted from used magnets has become an important research area [2–5].

There are two main classes of recycling of RE: dry and wet processes. As for the dry process, the recovery of Nd metal has been demonstrated [6] by employing Mg acting as an extraction medium, which forms a low-viscosity liquid alloy with Nd. It has been reported that RE can be separated by a selective reduction and a distillation [7]. The large difference of vapor pressure between RECl_2 and RECl_3 is skillfully utilized, and the selection efficiency is highly improved. Recently, the difference of oxygen affinity between RE and transition metals has also received attention in that this difference is used as a RE separation. For example, the mixture with flux $\text{FeO} \cdot \text{B}_2\text{O}_3$ is a promising method for high purity and high extraction ratio of RE oxide [8]. Thermal isolation of RE oxides from NdFeB magnets using carbon as a reducing agent has been reported [9]. However, many attempts based on the dry process proposed so far are still at the initial laboratory stage.

Development of ore dressing technologies has promoted wet process methods [10, 11], which have already been applied to recycling the sludge of in-plant scrap. After the acid leaching of scrap using HCl , HNO_3 , and H_2SO_4 , and the filtration of insoluble material mainly containing Fe, acid solution is reacted with oxalic or carbonic acid to form a precipitate containing RE elements. The calcined precipitate becomes RE oxides, which can be returned to the initial manufacturing process of NdFeB magnet. The roasting of NdFeB magnet [12–14] as a pretreatment improves the selectivity between rare earths and Fe, but the recovery ratio of RE is usually rather low with acid (especially HCl) leaching at room temperature. Nearly 100% recovery is achieved [12, 13] when HCl solution is heated to 80–180°C. In the acid leaching method for sludge [15], it is also necessary to heat the acid solution up to 80°C. We have recently proposed a pretreatment of corrosion [16] before the HCl leaching and the oxalic acid precipitation. In this method, the recovery ratio of Nd reaches 97% even when a room temperature process is used.

From the standpoint of sustainability and ecology, the main issue of the wet process is the discharge of waste acid solution. The recyclability of waste acid crucially depends on the efficient extraction of the constituent elements of the magnet from the used acid. One of the promising methods for Fe extraction involves a reaction with an ionic liquid [12, 17–20], which often possesses a high selectivity between rare earths and Fe. Trihexyl(tetradecyl)phosphonium chloride (Cyphos® IL101) is a well-characterized ionic liquid that can extract Fe^{3+} ions in HCl solution with no extraction of trivalent rare earth ions [12, 18]. A possible closed-loop acid process for roasted NdFeB magnet has also been proposed, and the elemental technologies are well investigated [12]. However, an actual demonstration with reuse of waste acid solution has not been performed.

In this study, we introduce the full recovery of rare earth from NdFeB magnet using a wet process with the pretreatment of corrosion but without a closed-loop acid process. After that we describe the detailed experimental results of multiple rare earth extraction in a closed-loop acid process [21].

2. Materials and methods

We used two kinds of commercial NdFeB magnets (Niroku seisakusyo). The elemental component of one magnet (denoted as magnet (1)) according to the manufacturer is Nd:Fe:B:the other elements (Dy et al.) = 28:66:1:5 in wt%. The composition of the other magnet (denoted as magnet (2)) was checked by an energy-dispersive X-ray spectrometer equipped in a field emission scanning electron microscope (JEOL, JSM-7100F) and determined to be $\text{Nd}_{1.6}\text{Pr}_{0.6}\text{Fe}_{14}\text{B}$. **Figure 1(a)** shows the process flow without a closed-loop acid process. A demagnetized and pulverized NdFeB magnet (1), weighing approximately 0.5 g, was immersed in 3% NaCl solution (300 mL) for 1 week. An air pump provided constant air flow to the solution to accelerate the corrosion.

Figure 1. (a) Procedures for rare earth recovery from NdFeB magnet (1) without closed-loop acid process. (b) Procedures for rare earth recovery from NdFeB magnet (2) with closed-loop acid process. S, L, and IL denote the solid, liquid, and ionic liquid, respectively. In procedure (b), elements expected to be present in the solid or solution are denoted.

The corroded sample was leached into HCl solution (100 mL) ranging from 0.1 to 0.3 mol/L, at room temperature. The insoluble material was calcined at 800°C for 5 h to obtain α -Fe₂O₃. The solution after removal of the insoluble was reacted with 0.26 g oxalic acid. The precipitate after the reaction was also calcined at 800°C for 5 h to obtain cubic Mn₂O₃-type Nd₂O₃ (c-Nd₂O₃).

To examine the feasibility of closed-loop acid process, the process flow was modified as shown in **Figure 1(b)**. In this case, the corroded sample was leached in HCl solution (100 mL) with 0.2 mol/L or 0.5 mol/L for 1–2 h. After removal of the insoluble material, the solution was reacted with ionic liquid Cyphos® IL101 (HCl:ionic liquid = 4:1 in volume ratio), purchased from Sigma-Aldrich. The salting-out agent 10 mol/L NH₄Cl was added to the solution. The mixture underwent magnetic stirring at 750 rpm and 60°C for 10 min. Then, it was centrifuged at 2500 rpm for 10 min and split into each component. The HCl solution was reacted with oxalic acid.

Several samples, calcined at 800°C for 5 h in the air, were evaluated using a powder X-ray diffractometer (Shimadzu, XRD-7000 L) with Cu-K α radiation. We employed an inductively coupled plasma atomic emission spectrometer (Shimadzu, ICPE-9000) to analyze the concentrations of Nd, Pr, Fe, and B dissolved in HCl or NaCl solution. The concentrations were determined by the working curves of standard Nd, Pr, Fe, and B liquids.

3. Results and discussion

The XRD pattern of corroded magnet (1) is shown in **Figure 2**, in which the XRD pattern of magnet (1) itself is also displayed. The main phase of magnet (1) is Nd₂Fe₁₄B with additional minor phase of NdFe₄B₄. The XRD pattern of Nd₂Fe₁₄B completely disappears in the corroded magnet, partially containing the XRD pattern of γ -FeOOH denoted by the filled triangles. In order to investigate the origin of the rest of the diffraction peaks (open circles) in the corroded sample, we have corroded NdFeB magnet (1) by hydrogenating it at 600°C for 12 h under a high pressure of hydrogen. The hydrogenated sample [22], as shown in the bottom pattern of **Figure 2**, shows the decomposition into Nd hydride (NdH_{2+x}) and α -Fe. Therefore, in each compound, the corrosion process would independently occur. The XRD pattern of corroded sample after the hydrogenation almost coincides with that of directly corroded magnet (1). Considering that α -Fe corroded into the Fe hydroxide (γ -FeOOH), a Nd hydroxide is probably responsible for the rest of the diffraction peaks (open circles) in the corroded sample. We note here that the NaCl concentration is not optimized. We simply suppose the sea water which is an abundant resource. The preliminary result using more concentrated NaCl solution (10%) also leads to the same results.

We checked the XRD pattern of insoluble material after HCl leaching of the corroded sample as shown in **Figure 3(a)**. The diffraction peaks match well with those of the XRD pattern of γ -FeOOH, which transforms into α -Fe₂O₃ through the calcination (see **Figure 3(b)**). **Figure 3(a)** supports that the Nd hydroxide would be selectively dissolved into HCl solution, in which Nd ions are generated. The oxalic acid precipitation was performed to recover Nd. The precipitate has been calcined and evaluated by XRD pattern, which is displayed in **Figure 3(b)** with the simulated pattern of c-Nd₂O₃. The XRD patterns are well matched between the calcined precipitate and c-Nd₂O₃, suggesting the successful recovery of Nd in the form of Nd oxide.

Figure 2. XRD patterns of corroded magnet, corroded sample after hydrogenation, and hydrogenated magnet. The employed magnet is magnet (1). The origin of each pattern is shifted by an integer value for clarity. Based partly on Ref. [16].

Figure 4 shows the leaching time dependences of effective recovery ratio R of Nd through 0.1, 0.2, and 0.3 mol/L HCl leaching. R was obtained by the equation.

$$R = (\text{Mass of Nd in Nd}_2\text{O}_3) / (\text{Mass of Nd in magnet}) \times 100 \quad (1)$$

For each HCl concentration, R increases with increasing leaching time, and approximately saturates, however exceeds 100% at some conditions. The excess above 100% would be due to the existence of impurity phases in c-Nd₂O₃ and/or incomplete filtration technique.

Next, we show the experimental results of rare earth extraction using the closed-loop acid process. The starting magnet is magnet (2) with the mass of approximately 0.5 g. The distribution of constituent elements in our method mentioned above is partially unknown. Thus, a one-shot extraction with 0.2 mol/L HCl and 0.26 g oxalic acid but with no use of ionic liquid was performed. **Table 1** shows the distribution of each ion in the NaCl solution after removal of the corroded sample and in the HCl solution after removal of precipitates produced by the reaction with oxalic acid. In the NaCl solution, only B is detected. A sufficient amount of oxalic acid can efficiently separate rare earths. A large amount of Fe stays in the HCl solution. The expected ion concentrations of the completely dissolved 0.5 g magnet are 1041 mg/L for Nd, 381 mg/L for Pr, 3527 mg/L for Fe, and 49 mg/L for B, respectively. The summation of the B concentrations in the two states listed in **Table 1** is not far from 49 mg/L. Then, approximately 30% of B can be separated by the NaCl solution, and the remaining B stays in the HCl solution. Approximately 60% of the Fe ions are contained in the insoluble material obtained after HCl leaching. The recovery ratio of Nd (Pr) is 99% (97%).

Figure 3. (a) XRD patterns of corroded magnet (1) and insoluble material after HCl (0.1 mol/L, 30 min) leaching and γ -FeOOH. (b) XRD patterns of insoluble material and oxalic acid precipitate in HCl solution. They were calcined in the air. The simulation patterns of α -Fe₂O₃ and c-Nd₂O₃ are also shown. The HCl concentration is 0.2 mol/L, and the leaching time is 2 h. The origin of each pattern in (a) and (b) is shifted by an integer value for clarity. Based partly on Ref. [16].

Following the results of Ref. [12] which reported that salting-out agent NH₄Cl plays an essential role in full extraction of Fe by an ionic liquid, the dependence of extraction efficiency on NH₄Cl concentration was determined. Without NH₄Cl, the extraction efficiency of Fe is only 40%, increasing to 75% with 5 mol/L NH₄Cl and 95% for 10 mol/L NH₄Cl. In this experiment, we also found that B is fully extracted by the ionic liquid. We speculate that Cyphos® IL101 represented by C₃₈H₆₈ClP transforms into C₃₈H₆₈FeCl₄ after the incorporation of Fe³⁺ ions. Thus, the extraction efficiency of Fe severely depends on the Cl concentration, and the salting-out agent NH₄Cl is necessary to provide enough Cl ions.

Figure 4. Leaching time dependences of effective recovery ratio of Nd. The examined HCl solutions are 0.1, 0.2, and 0.3 mol/L. Based partly on Ref. [16].

Solution	Nd (mg/L)	Pr (mg/L)	Fe (mg/L)	B (mg/L)
NaCl after removal of corroded magnet	ND	ND	ND	12.3
HCl after removal of oxalic acid precipitates	10.6	13.0	1480	28.6

ND means not detected. Based on Ref. [21].

Table 1. Distribution of Nd, Pr, Fe, and B in the one-shot recovery process.

The preliminary experiment for a closed-loop process of HCl solution was performed using 0.5 mol/L HCl to reduce the experimental time. The oxalic acid mass was maintained at 0.26 g. Hereafter, the HCl solutions after removal of insoluble material in the acid leaching, after Fe extraction by the ionic liquid, and after removal of rare earths by oxalic acid precipitation are denoted state [I], [II], and [III], respectively (see also **Figure 1(b)**). **Table 2** shows the Nd, Pr, and Fe concentrations of these states during each cycle. In the first cycle, approximately 24% of rare earths are extracted together with Fe by the ionic liquid. Only Nd and Pr are separated by the reaction with the oxalic acid. In state [I] during the second cycle, the concentrations of Nd and Pr are approximately one-quarter of those during the first cycle, which means an immediate precipitation due to excess oxalic acid in the previous cycle.

The preliminary experiment suggested that the weight of oxalic acid needs to be adjusted. The weight of oxalic acid for a 0.5 g magnet, reproducing the initial concentrations of Nd and Pr in state [I] in the second cycle, has been searched as shown in **Figure 5**. The vertical axis shows the difference in Nd (Pr) ion concentration in state [I] between the first and second cycles, which is denoted as Δc . A positive value for Δc indicates a poor precipitation efficiency, and a negative value indicates an excess oxalic acid. For each element, Δc linearly decreases with increasing weight of oxalic acid. A weight of 0.1675 g oxalic acid can reproduce the initial Nd (Pr) ion concentration of state [I] in the second cycle. The chemical equation for oxalic acid precipitation is.

If the starting magnet weight is 0.5 g, the ideal amount of oxalic acid is 0.1335 g. However, as shown in **Figure 5**, Nd and Pr would not fully precipitate for 0.1335 g of oxalic acid. To achieve sufficient precipitation, the amount of oxalic acid that is consumed must be 1.3 times larger.

Cycle	State of solution	Nd (mg/L)	Pr (mg/L)	Fe (mg/L)
First	[I]	929	346	2320
	[II]	707	264	105
	[III]	16.9	17.4	109
Second	[I]	218	94	2770
	[II]	231	99	132
	[III]	ND	ND	134

The notations of solutions are defined in **Figure 1(b)**. ND means not detected. Based partly on Ref. [21].

Table 2. Distribution of Nd, Pr, and Fe in the preliminary experiment of a closed loop for HCl solution.

Figure 5. A plot of Δc vs. the weight of oxalic acid. Δc represents the difference in Nd (Pr) ion concentration after HCl leaching of corroded magnet between the first and second cycles. Based partly on Ref. [21].

From the preliminary experiments for the closed-loop process of HCl solution, we have obtained the conditions of 10 mol/L NH_4Cl and 0.1675 g oxalic acid for the recycling of 0.5 g magnet. **Table 3** shows the results of a demonstration of triple rare earth extraction from 0.5 g magnet with the closed loop of HCl solution. The HCl concentration was 0.5 mol/L to expedite the experiment. The concentrations of Nd and Pr ions are measured in states [I], [II], and [III]. In each cycle, 10–15% of Nd and Pr ions are extracted together with Fe ions by the ionic liquid, as in the preliminary experiment (see **Table 2**). Contrary to our expectation, 30–35% of Nd and Pr ions remain in solution after the oxalic acid precipitation. However, these ions apparently do not contribute to the rare earth concentrations of state [I] in the next cycle; the

Cycle	State of solution	Nd (mg/L)	Pr (mg/L)	Recovery ratio of Nd (%)	Recovery ratio of Pr (%)
First	[I]	971	370	50	47
	[II]	818	311		
	[III]	328	136		
Second	[I]	980	406	49	50
	[II]	836	357		
	[III]	352	156		
Third	[I]	919	410	61	56
	[II]	842	360		
	[III]	280	130		

The notations of the solutions are defined in **Figure 1(b)**. Based partly on Ref. [21].

Table 3. The distribution of Nd and Pr and the recovery ratio of each element in a triple rare earth extraction.

concentrations of Nd and Pr ions in state [I] are the same as those during the previous cycle. The recovery ratio of each element, calculated by eliminating the amount of element extracted by the ionic liquid, is approximately 50% in all cycles. **Figure 6** shows the XRD pattern of calcined insoluble material after HCl leaching in the second cycle. The simulated patterns of $\text{Nd}_{0.73}\text{Pr}_{0.27}\text{FeO}_3$ and $\alpha\text{-Fe}_2\text{O}_3$ are also exhibited. Elemental Nd (Pr) is partially recovered together with Fe. The XRD pattern in **Figure 6** supports the idea that the Nd and Pr elements, which are present in the same concentration as the ions in state [III], would enter insoluble material in state [I] in the next cycle.

In our method, Cyphos® IL101 is rather expensive, and the regeneration of the ionic liquid by stripping Fe^{3+} is required to reduce the cost. We examined two stripping methods. The first one is the stripping using NaOH solution. After the reaction of the ionic liquid containing Fe^{3+} ions with NaOH solution, $\text{Fe}(\text{OH})_3$ is expected to be precipitated, and Fe_2O_3 would be obtained after the calcination. The ionic liquid was reacted with NaOH solution (1 mol/L) with a volume ratio of Cyphos® IL101:NaOH solution = 1:2. The calcined precipitate was checked by the XRD pattern, and it is shown in **Figure 7(a)**. The obtained pattern is in good agreement with the XRD pattern of NaFeO_2 . The recovery ratio of Fe is estimated to be 17%. The second method is the employment of ammonia solution, for which 100% stripping of Fe^{3+} has been already reported [23]. We roughly followed the reported recipe [23]. The yellow-colored ionic liquid containing Fe^{3+} , which was obtained in the actual process flow, was reacted with the ammonia solution (approximately 3 wt.%) with a volume ratio of Cyphos® IL101:ammonia solution = 1:10.

Figure 6. XRD pattern of calcined insoluble material after HCl leaching in the second extraction. The simulated $\text{Nd}_{0.73}\text{Pr}_{0.27}\text{FeO}_3$ and $\alpha\text{-Fe}_2\text{O}_3$ patterns are also shown. The origin of each pattern is shifted by an integer value for clarity. Based partly on Ref. [21].

Figure 7. (a) XRD pattern of the calcined sample recovered from used Cyphos® IL101 reacted with NaOH solution. The simulated NaFeO₂ pattern is also presented. The origin of each pattern is shifted by an integer value for clarity. (b) XRD pattern of calcined sample recovered from used Cyphos® IL101 reacted with ammonia solution. The simulated α-Fe₂O₃, Fe₃PO₇, and C patterns are also presented. The origin of each pattern is shifted by an integer value for clarity. (Based partly on Ref. [21]).

The precipitate at the interface between the liquids was collected using a cellulose filter and calcined together with the filter in the air. **Figure 7(b)** shows the XRD pattern of the calcined sample recovered from used Cyphos® IL101. The figure also displays the simulated patterns of α-Fe₂O₃, Fe₃PO₇, and C. The experimental XRD pattern closely matches the superposition of simulated patterns. The sources of P and C atom contamination would be Cyphos® IL101 containing P and the cellulose filter, respectively. Despite the contamination due to our insufficient filtration technique, Fe can be stripped. Furthermore, the yellow-colored ionic liquid once again became transparent after Fe³⁺ stripping, which means the ionic liquid is regenerated.

Focusing on the one-shot recovery processes, a comparison between our method and other methods [12–15] based on the acid leaching mentioned in Introduction is shown in **Table 4**. The number of steps in the recovery processes and the rare earth recovery ratio of each method are similar. The roasting used in Refs. [12–14] would have a cost disadvantage. Although Ref. [15] does not employ roasting, HNO₃ has a disadvantage because waste discharge containing nitrate salt is severely controlled by law for environmental reasons. The acid leaching process in our method and in Ref. [14] is performed at room temperature, which means that these are

Items	Our method [16]	[12]	[13]	[14]	[15]
Pretreatment	Corrosion at RT	Roast at 950°C	Roast at 500–1000°C	Roast at 750°C	Sludge
Acid	HCl	HCl	HCl	H ₂ SO ₄	HNO ₃
Acid leaching temperature	RT	80°C	180°C	RT	80°C
B separation	30%	Not separated	Not separated	Not separated	Not separated
Recovery ratio of rare earths (%)	97	100	100	100	94

RT means room temperature. Based partly on Ref. [21].

Table 4. Comparison between our method and other methods based on acid leaching in a one-shot recovery process.

safe processes. Since B is harmful, its separation is highly desired. Our method has achieved 30% B separation, whereas the other methods do not report a clear B separation. If a closed-loop acid process with a high recovery ratio of rare earths is realized, our method is promising because each step except Fe extraction by ionic liquid is performed at room temperature. This condition and the rather simple procedures lead to a safe and low-cost recovery process. In addition, the peculiar feature of B separation in our method is environmentally friendly.

4. Summary

Rare earth extraction methods based on acid leaching are entering the stage of practical use. To address the issue of rather low selectivity between rare earths and Fe at the room temperature acid-process, we have proposed the pretreatment of corrosion. Our method has improved the selectivity, and rare earth recovery ratio, in one-shot extraction, reaches to almost 100% even at room temperature. For sustainability and environmental considerations, the recyclability of waste acid solution is one of the central issues in rare earth recycling, and this has not been well investigated. In this work, we have experimentally determined the recovery ratio of rare earth elements in our method with the closed-loop acid process. This ratio is approximately 50%, reduced from almost full recovery in a one-shot extraction. Although the recovery ratio is rather low at the present stage, our encouraging result should lead to rapid advancement of the study of recycling using a closed-loop acid process.

5. Future directions

The demonstration of closed-loop process for HCl solution indicates that the precipitation by oxalic acid is not sufficient, although the amount of oxalic acid is larger than the ideal amount calculated using the chemical formula of precipitation. To increase the recovery ratios of rare earth elements, if the amount of oxalic acid is increased, it will result in a reduced recovery ratio in the second cycle, as deduced from **Table 2**. Thus, a trade-off between the number of rare earth extractions and the recovery ratio of rare earths might exist for the present precipitation condition. The main cause of the reduced recovery ratio is the insufficient ionization of oxalic acid. The degree of ionization of oxalic acid strongly depends on the pH of the solution. The ionization concentration generally increases with increasing pH, and the full ionization of oxalic acid with an ideal weight of 0.1335 g would be realized. Another issue to be considered is the partial rare earth extraction by the ionic liquid. If the oxalic acid precipitation process is performed before the process of Fe^{3+} extraction by the ionic liquid, only rare earth elements would be separated due to the high selectivity between rare earths and Fe under oxalic acid precipitation. Thus, the issue would be resolved by reversing the sequence of the two processes. As shown in **Figure 6**, unassigned peaks of material other than $\alpha\text{-Fe}_2\text{O}_3$ and $\text{Nd}_{0.73}\text{Pr}_{0.27}\text{FeO}_3$ are present in the XRD spectrum. In our study, complete separation of the ionic liquid from the HCl solution is difficult, which results in contamination of the calcined sample. Further improvement of the separation technique is needed to obtain a pure calcined sample.

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Author details

Jiro Kitagawa^{1*} and Masami Tsubota²

*Address all correspondence to: j-kitagawa@fit.ac.jp

1 Department of Electrical Engineering, Faculty of Engineering, Fukuoka Institute of Technology, Fukuoka, Japan

2 Physonit Inc., Hiroshima, Japan

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