Recovery of Neodymium from Waste Permanent Magnets by Hydrometallurgy Using Hollow Fibre Supported Liquid Membranes

Achmad Chusnun NI’AM1,2, Yi-Hsien LIU1, Ya-Fen WANG1,3, Shyh-Wei CHEN1,4, Gen-Mu CHANG1 and Sheng-Jie YOU1,3*

1 Department of Environmental Engineering, Chung Yuan Christian University, 200 Chung Pei Road, Chung-Li 320, Taiwan; 2 Department of Civil Engineering, Chung Yuan Christian University, 200 Chung Pei Road, Chung-Li 320, Taiwan; 3 Research and Development Center for Membrane Technology, Chung Yuan Christian University, 493 Xinzhong N. Road, Chung-Li 320 Taiwan; 4 Environmental Protection Administration, Executive Yuan, Taipei 100, Taiwan

(Received September 28, 2019; Accepted October 21, 2019)

The recycling of the rare earth neodymium (Nd) is vital to providing greener technology for many industries. In this paper its recovery from waste permanent magnets (WPMs) is explored using a hydrometallurgical process involving separation using hollow fiber supported liquid membranes (HFSLMs). After demagnetization, the options for WPMs leaching were investigated by varying mineral acids, acid concentrations and solid/liquid (S/L) ratios. The separation Nd from iron (Fe) was carried out by precipitation using oxalic acid. Nd in WPMs was recovered by solvent extraction using D2EHPA in Isopar-L. The influence of various parameters such as pH and organic/aqueous (O/A) phase ratio were investigated. Optimum leaching was achieved using 3 M H2SO4 with an S/L ratio of 1 g/50 mL to obtain 485 g/kg of Nd. Nd-separation from Fe was successfully performed using 0.6 M D2EHPA with an O/A phase ratio of 3 at a pH of 1.26 – 2.0 resulting in Nd-recovery of 97.48%. The optimum stripping was achieved using 6 M HNO3. In the HFSLM experiments, the concentration of Nd in the aqueous strip solution was 4120 mg/L whilst that in the aqueous feed solutions was 658 mg/L. The HFSLM module used recovered 90.82% of the Nd within 35 min.

1. Introduction

The rare earth elements comprise 17 metals. These are divided into two groups: the heavy rare earth elements (HREEs), viz. Y, Tb, Dy, Ho, Er, Tm, Yb, and Lu and the light rare earth elements (LREEs), viz. Pr, Nd, Pm, Sm, Eu, Gd, Sc, La, and Ce [1-3]. Rare earth metals and their compounds are essential constituents in industrial and in technological products such as electrical vehicles, batteries, energy efficient lighting, magnets, catalysts, wind turbines, etc. [4,5]. Consequently, the demand is increasing rapidly and is predicted to exceed worldwide supply by 40,000 tons annually [6].

At present, permanent magnets in electrical machines are used mostly in sectors involving technology and transportation. The global demand for permanent rare earth magnets is expected to grow by more than 10% by 2022 [7]. Permanent magnets incorporate substantial quantities of Nd and dysprosium (Dy) [4]. Nd has particularly high commercial value and is also in high demand because of its extensive use in magnets for wind
turbines, motors, and generators [6,8]. Nd is a key element used in NdFeB permanent magnets, containing 15-30% of Nd [9].

Techniques using pyrometallurgical and hydrometallurgical processes to recycle materials in magnets and enhance the recovery of rare earth elements, especially Nd, have been reported [6,10,11]. The chemical and physical characteristics of REEs make their purification and separation complicated [12,13]. Liquid-liquid extraction can be easily applied on a lab or industrial scale [4,14,15]. One of the obstacles using hydrometallurgical processes to recover REEs from magnets is the large amount of Fe present. [13,16] Usually the affinity for Fe(III) shown by acidic solvent extractants or for ion exchange materials is higher than for REEs [17].

Separation using membranes usually is energy-efficient and thus has attracted the attention of several researchers [12,18,19]. The supported liquid membrane (SLM) is a simple and promising technology for the removal, extraction, separation of valuable metal ions from various resources [4,19]. SLMs have advantages such as high selectivity and combine extraction and stripping into a single stage [20]. In addition, a small amount of organic extractant is needed, a significant factor in relation to operating costs and to using environmentally friendly processes [21,22]. In SLM processes the aqueous feed and the strip phase are separated by a thin membranes [20]. Flat sheet supported liquid membrane (FS-SLM) and hollow fiber supported liquid membrane (HFSLM) are the major classes of membranes used [20,21]. Previous studies were successful in separating REEs using SLMs containing organophosphorus extractants such as di(2-ethylhexyl)phosphoric acid (D2EHPA) [23] and dinonylphenylphosphoric acid (DNPPA) [18,24,25]. The most widely used extractant is D2EHPA due to its high selectivity for REEs [1,14,26,27] and its miscibility with all common diluents, even at 90% (v/v) [28].

The main objective of this study is the recovery of Nd from WPMs. The hydrometallurgy recovery of REEs, especially Nd, was investigated varying several parameters. Nd separation from Fe also is a key feature of the study. In addition to conventional solvent extraction methods, Nd-recovery was also investigated using HFSLMs.

2. Experimental

2.1 Material

Waste permanent magnet samples used in this study were provided by one of Taiwan magnet manufactures.

2.2 Chemical reagents

Nd nitrate hexahydrate Nd(NO\textsubscript{3})\textsubscript{3}.6H\textsubscript{2}O, 99.9% was purchased from Merck and bis(2-ethylhexyl)hydrogenphosphoric acid (D2EHPA) from Acros Organic. D2EHPA, 95%, molecular weight of 322.426 g/mol was used as an acidic extractant in Isopar-L, obtained from ExxonMobil Chemical. Sulfuric acid (95-97%) was purchased from Scharlau, nitric acid (69%) from PanReac, hydrochloric acid (37%) from Aencore and oxalic acid from Merck. The ultra-pure water system of PURELAB flex 3 was used as the source for deionized water.

2.3 Experimental process

2.3.1 Pretreatment

Demagnetization of WPMs was investigated at various elevated temperatures i.e. 300, 350, 400 and 450°C for 15 and 30 min. Subsequently, the WPMs were crushed and ground in a planetary ball mill PM 100 (Retsch, Germany) at 450 rpm for 120 min to produce a with a 0.250 mm particle size. The determination of the Ni and other metal-content of the WPMs was performed by the NIEA S321.63B method. [29]
2.3.2 Leaching studies

Leaching of Nd from demagnetized WPMs into acids involved an agitation speed of 300 rpm at room temperature for 24 h. The influence of varying the mineral acids (HCl, H2SO4, and HNO3), acid concentration (0.1 to 5 M) and S/L ratio (0.02 to 0.3) was investigated to optimize conditions for dissolution of Nd. The pregnant leach solution and the solid residues were separated by centrifugation and filtration using a 0.45 µm PVDF filter. The aqueous phase was analyzed by an inductively coupled plasma-atomic emission spectrometry (ICP-AES, Shimadzu ICPE-9820).

2.3.3 Solvent extraction studies

A stock solution of Nd(NO3)3.6H2O in 0.5 M nitric acid was contacted with D2EHPA in Isopar-L. Process parameters were: the concentration of extractant (0.2-1 M), the organic/aqueous volume ratio (1 : 1 to 1 : 5), the pH of aqueous phase (0.2 - 2) and the nature of stripping agents (HCl, H2SO4, and HNO3) and their concentrations (3-7 M) were investigated. Experiments were carried out at room temperature with an agitation speed of 300 rpm for 24 h. The concentration of metals in the aqueous phase was determined by ICP-AES, and the extraction efficiency was calculated using:

$$E(%) = \frac{[\text{REE}]_i - [\text{REE}]_f}{[\text{REE}]_i} \times 100$$

where [REE]i and [REE]f refer to the initial and final concentration of Nd in the aqueous phase.

2.3.4 Precipitation studies

The REEs in leach solutions were precipitated by the addition of oxalic acid 0.5 M and heated to 70±5°C for 40 min. After 6 h the mixture was centrifuged, washed with 3% H2O2, 3% HCl, and deionized water and then dried to produce the solid rare earth oxide mixture, the composition of which was determined by ICP-AES.

2.3.5 Stripping studies

The stripping of Nd from the loaded organic phase was investigated using various mineral acids at different concentrations. The organic and aqueous phases were put in Erlenmeyer flask and stirred at 300 rpm at room temperature. A funnel was used to separate the two phases.

2.3.6 Hollow fiber supported liquid membrane (HFSLM) experiments

An HFSLM module was arranged with two gear pumps, two variable flow rate controllers, two rotameters, and four pressure gauges. A membrane contactor (Liquid Cell 2.5 x 8) was purchased from Polypore Company, USA with hydrophobic microporous polypropylene hollow fibers enclosed in a 2.5” x 8” polypropylene shell [30]. The polypropylene membrane is hydrophobic in nature thus the organic phase wets and even passes through the membrane fibers [30]. The characteristics of the membrane module used are shown in Table 1.

| Characteristic       | Unit                  |
|----------------------|-----------------------|
| Hollow fiber type    | Liquid-Cell Extra-Flow 2.5 x 8 |
| Material             | Hydrophobic polypropylene |
The module was used with the liquid flow rate in tube side of 0.25 - 0.3 L/min, resulting in an inflow pressure at the end tube of the tube side of about 5 psi. The flow rate of the strip solution was 0.3 - 0.35 L/min, resulting in an outflow pressure at the end tube of the shell side of about 0 to 1 psi. The feed and strip solution reservoirs were stirred at 100 rpm. Samples (5 mL) of feed and strip solutions were collected every 10 min for 120 min. The experimental set-up of the HFSLM module is shown in Figure 1.

2.3.7 Equipment and analytical methods

The characterization of the demagnetized WPMs was conducted using scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDS) (Hitachi S-3000N). The metal composition was determined after total dissolution in aqua regia at 80±1°C. Aqua regia was prepared by mixing HNO₃ and HCl in the volume ratio of 1:3 [14,31]. The concentration of metal ions in the samples was measured by ICP-AES.
3. Results and Discussion

3.1 Pretreatment

The anti-corrosion layer and epoxy resin in the magnet surface were removed by the demagnetization process. Complete demagnetization was achieved by heating at 450°C for 15 min. Samples obtained after heating at 300, 350 and 400°C had weak magnetic properties. Accordingly, demagnetization at 450°C for 15 min was chosen for this study. A magnetic performance can be influenced by demagnetization with high temperature [10,32]. Thermal demagnetization especially under high temperature steadily eliminated the magnetic force [5,33].

| Element | Fe      | Nd      | Pr      | Dy      | B       |
|---------|---------|---------|---------|---------|---------|
| g/kg    | 509.81  | 417.04  | 52.24   | 11.7    | 9.18    |

3.2 Characterization of samples

The morphology of the demagnetized waste permanent magnet samples was examined by SEM-EDS and Figure 2 indicates that the demagnetized samples consist mainly of Fe (see also Table 2) and Nd, and much smaller quantities of praseodymium, dysprosium, and boron. SEM-EDS analysis indicates and that the sample consists of irregularly shape particles. A previous study also records that the demagnetized material is not homogeneous and contains irregularly shaped particles with Fe as the major component, followed by Nd, Dy, Pr, and B [31,34].

Figure 2. SEM-EDS analysis of the sample.

3.3 Factor affecting Nd leaching

3.3.1 Influence of acid concentration

The influence of acid concentration on the leaching of Nd from WPMs was studied with concentrations in the range 0.1-5 M and an S/L ratio of 2.5g/50 mL, stirring for 24 h. Figure 3 indicates that on increasing the HNO₃ concentration from 0.1 to 5 M, the yield of Nd increased from 33.68 g/kg to 395 g/kg. Increasing the concentration of HCl from 0.1 to 1 M increased the yield from 49.6 g/kg to 362 g/kg. The yield of Nd increased from 38.08 g/kg to 456 g/kg when the H₂SO₄ concentration was increased from 0.1 to 3 M. In contrast, the yield
of Nd was drastically reduced to 116.8 g/kg when using 5 M H₂SO₄. This is ascribed to the predominant species being HSO₄⁻ rather than SO₄²⁻ at high acid concentrations [35]. In the work reported below 3 M H₂SO₄ was chosen for leaching.

Figure 3. The influence of acid concentration on the leaching yield of REEs; a) HNO₃, b) HCl, c) H₂SO₄.

3.3.2 Influence of S/L ratio

The influence of the S/L ratio in leaching is shown in Figure 4. The S/L ratio was increased, using 1, 2.5, 5, 10 and 15 g solid samples in 50 mL of acid. The other parameters, an agitation speed of 300 rpm, duration 24 h using 3 M H₂SO₄, were kept constant. The leaching yield of Nd decreased significantly when the S/L ratio exceeded 1:5. This indicated that the excessive addition of solid sample causes the metal was insoluble and more produced residue in solution. The reduced dissolution is related to the reduced surface area available to the leachant [36]. An S/L ratio of 1 g/50 mL (485 g/kg) was considered appropriate for further work.
3.4 Precipitation by oxalic acid

Since the leach solution has a high Fe concentration, a study of precipitation methods was carried out to separate Nd(III) by converting into an oxalate precipitate [13]. The precipitation was carried out using 0.5 M oxalic acid. Table 3 shows that REEs, especially Nd, were effectively precipitated using 0.5 M oxalic acid, recovering 99% of Nd, 97% of Pr, 94% of Dy, and also 82% of Fe. The reaction between Nd and oxalic acid is represented by Eq. (2)

$$\text{Nd}_2(\text{SO}_4)_3(\text{aq}) + 3\text{H}_2\text{C}_2\text{O}_4^{2-} (s) \rightarrow \text{Nd}_2(\text{C}_2\text{O}_4)_3 (s) + 3\text{H}_2\text{SO}_4$$  (2)

In order to remove Fe, the precipitate was rinsed with aqueous H$_2$O$_2$ and with HCl. In this process, Fe$_2$(C$_2$O$_4$)$_3$ is oxidized to Fe$_2$O$_3$ by H$_2$O$_2$ and then converted to an aqueous solution of FeCl$_3$ by HCl. The result shown in Table 3 suggest that the purification was successfully using oxalic acid.

| Elements | Fe   | Nd   | Pr   | Dy   |
|----------|------|------|------|------|
| Precipitation efficiency (%) | 82.21 | 99.99 | 97.20 | 94.62 |
| Purification efficiency (%)   | 4.00  | 84.00 | 10.00 | 2.00  |

3.5 Solvent extraction

3.5.1 Influence of pH

D2EHPA is a cation exchanger that produces H$^+$ on metal- cation uptake. This lowers the pH which in turn displaces the equilibrium, lowering metals-uptake [6,37,38]. In this work the pH of the aqueous feed fell in the range 0.2 to 2. The extraction of Nd was carried out using 1 M D2EHPA in Isopar-L, with an O/A ratio of 1, a contact time 24 h using 300 rpm agitation at room temperature.

As recorded in previous reports, the pH of aqueous phase crucially influences extraction efficiency [7,27,39,40]. Figure 5 shows that as the pH is raised extraction increases from 75 to 99.6%. The optimum range of pH for Nd extraction is 1.26-2. In an earlier report the optimum extraction efficiency of Nd is recorded at a
pH of 1.5 [27]. The mechanism of extraction of the REEs with organophosphorus reagents has been considered [14,41] to occur as shown in Eq. 3:

\[
M^{3+} + m(\text{HR})_2 \leftrightarrow \text{MR}_3(\text{HR})_{2m-3} + 3\text{H}^+ \quad (3),
\]

where \(M^{3+}\) is the lanthanide ion in the aqueous phase, HR is the organophosphorus extractant in the organic phase, existing as a dimer \((\text{HR})_2\), and \(\text{MR}_3(\text{HR})_{2m-3}\) is the complex formed in the organic phase.

\[\text{Figure 5. Influence of pH.}\]

3.5.2 Influence of O/A phase ratio

Nd-loading by D2EHPA in Isopar-L at various O/A phase ratios (Figure 6) is decreases slightly as the ratio is decreased from 1 : 1 to 1 : 5 Any further increase in the O/A phase ratio caused very little increase in Nd extraction. An O/A phase ratio of 1 : 3 which gives 99.72% uptake was used in subsequent experiments.

\[\text{Figure 6. Influence of O/A phase ratio.}\]
3.5.3 Influence of extractant concentration

The extraction of Nd by D2EHPA (Figure 7) increases from 56 to 99% as the extractant concentration is increased from 0.2 to 1 M in accordance with previous studies [4,11,42]. The extractant concentration is the operational parameter that has the major influence on selectivity and conceivable to control selectivity in a certain range [21]. Several extractant systems used for Nd recovery are summarized in Table 5. A concentration of 0.6 M D2EHPA was selected as the optimum value for further experiments.

![Figure 7. Influence of extractant concentration.](image)

| Extractant   | Diluent | Medium | pH   | Extraction [%] | References |
|--------------|---------|--------|------|----------------|------------|
| 0.6 M D2EHPA | Isopar-L| Nitrate| 1.26-2| 97.48          | This study |
| 0.1 M TODGA  | Nitrate | -      | -    | 98.6           | [43]       |
| 0.5 M Cyanex 921 | Kerosene | Nitrate | - | 98              | [44]       |
| 0.1 M Cyanex 272 | Escaid 110 | Chloride | 4.94 | 93.2           | [45]       |
| 0.3 M NaCyanex 302 | Kerosene | Chloride | 4.62 | 99.61          | [7]        |
| [336At][NO3]) | Kerosene | Chloride | 3    | 82             | [46]       |
| Cyanex 272   | Kerosene | Chloride | 2.7  | 95.04          | [47]       |

3.5.4 Stripping of Nd

3.5.4.1 The influences of stripping agents and concentrations

The loaded organic phases from the experiments described above were used to investigate the influence of stripping agents (HCl, H2SO4, and HNO3) and their concentrations (3 to 7 M), see Figure 8. Greater than 85% stripping of Nd was consistently achieved with the exception of 7 M H2SO4 when less than 70 % was obtained. It is caused by metallic complexes; a high concentration of hydrogen ions would combine with the metal ions to form complexes. The most efficient stripping agent (6 M HNO3) found was used in the rest of this study.
3.6 Nd recovery by HFSLMs

The optimized parameters in the solvent extraction batch experiments were applied in Nd recovery by the HFSLM module. 1 g of precipitated powder of Nd recovered from leaching solution was dissolved in 2 l of 0.5 M HCl. The aqueous feed used in the HFSLM experiments contained Fe: 30 mg/L, Nd: 658 mg/L, Pr: 77 mg/L, and Dy: 20 mg/L. D2EHPA (0.6 M in Isopar-L) was used as the organic phase in the membrane and 6 M HNO₃ as the aqueous strip solution. The volume ratio of feed : organic : aqueous strip was 6 : 3 : 1. Figure 9 indicates that Nd-recovery does not increase from a value of 90.82% after 35 min. Whilst, the recovery of Pr and Dy was achieved 11.89% and 1.50% after 35 min, respectively. The concentration of Nd in the aqueous feed was 658 mg/L and that in the aqueous strip was 4120 mg/L (ca. 6 times higher), in accordance with a previous study [48].
4. Conclusion

The recovery of Nd from WPMs using leaching followed by solvent extraction or membrane separation has been investigated. The optimum leaching of Nd (485 g/kg) from roasted WPMs used 3 M H₂SO₄ with an S/L ratio 1:50. Leaching efficiencies follow the order: HNO₃ ≥ HCl > H₂SO₄ and a stripping efficiency of Nd from the loaded organic of 99.95% was obtained using 6 M HNO₃. Precipitation using 0.5 M oxalic acid successfully decreased iron concentration to 4%. The optimum concentration of D2EHPA in Isopar-L for solvent extraction was 0.6 M with an O/A phase ratio 1 : 3 with aqueous feed having a pH of 1.26 - 2. The optimum recovery of Nd by an HFSLM was 90.82% of after 35 min of operation. The Nd concentration in the aqueous strip was increased by a factor of 6 over that in the feed solution, suggesting that HFSLM technology should be considered for industrial scale development for REE recovery.

Acknowledgement

The study was financially supported by EPA-106-XB08, Recycling Fund Management Board, Environmental Protection Administration, Taiwan.

References

1) T. Wannachod, N. Leepipatpiboon, U. Pancharoen, K. Nootong, Chem. Eng. J., 248, 158-167 (2014).
2) P. Enghang, Encyclopedia of the Elements, WILEY-VCH Verlag GmbH & Co. KGaA, 2004.
3) T. Akahori, Y. Miyamoto, T. Saeki, M. Okamoto, T. H. Okabe, J. Alloys Compd., 703, 337-343 (2017).
4) H.-S. Yoon, C.-J. Kim, K.-W. Chung, S.-D. Kim, J.-Y. Lee, J. R. Kumar, Hydrometallurgy, 165, 27-43 (2016).
5) D. D. Munchen, H. M. Veit, Waste Manage., 61, 372-376 (2017).
6) M. K. Jha, A. Kumari, R. Panda, J. Rajesh Kumar, K. Yoo, J. Y. Lee, Hydrometallurgy, 161, 77-101 (2016).
7) E. Padhan, A. K. Nayak, K. Sarangi, Hydrometallurgy, 174, 210-215 (2017).
8) L. Grandell, A. Lehtilä, M. Kivinen, T. Koljonen, S. Kihlman, L. S. Lauri, Renewable Energy, 95, 53-62 (2016).
9) M. A. R. Önal, E. Aktan, C. R. Borra, B. Blanpain, T. Van Gerven, M. Guo, Hydrometallurgy, 167, 115-123 (2017).
10) M. A. R. Onal, "Recycling of NdFeB Magnets for Rare Earth Elements (REE) Recovery," Ph.D, Materials Engineering, KU Leuven, Science, Engineering & Technology, 2017.
11) H.-S. Yoon, C.-J. Kim, K. W. Chung, S.-D. Kim, J. R. Kumar, J. Braz. Chem. Soc., 26, 1143-1151 (2015).
12) T. Wannachod, V. Mohdee, S. Suren, P. Ramakul, U. Pancharoen, K. Nootong, J. Ind. Eng. Chem., 26, 214-217 (2015).
13) E. Yamada, H. Murakami, S. Nishihama, K. Yoshizuka, Sep. Purif. Technol., 192, 62-68 (2018).
14) M. Gergoric, C. Ekberg, B.-M. Steenari, T. Retegan, J. Sustainable Metall., 3, 601-610 (2017).
15) S. Riaño, K. Binnemans, Green Chem., 17, 2931-2942, (2015).
16) Y. Yang, A. Walton, R. Sheridan, K. Güth, R. Gauß, O. Gutfleisch, M. Buchert, B.-M. Steenari, T. V. Gerven, P. T. Jones, K. Binnemans, J. Sustain. Metall., 3, 122–149 (2017).
17) T. A. Z. F. Xie, D. Dreisinger, F. Doyle, Miner. Eng., 56, 10-28 (2014).
18) M. Anitha, D. N. Ambare, M. K. Kotekar, D. K. Singh, H. Singh, Sep. Sci. Technol., 48, 2196-2203 (2013).
19) N. M. Kocherginsky, Q. Yang, L. Seelam, *Sep. Purif. Technol.*, **53**, 171-177 (2007).
20) P. K. Parhi, *J. Chem.*, **2013**, #618236 (2013).
21) J. Martínez, R. Rodríguez Varela, K. Forsberg, Å. Rasmuson, *Chem. Eng. Sci.*, **191**, 134-155 (2018).
22) I. Van de Voorde, L. Pinoy, R. F. De Ketelaere, *J. Membr. Sci.*, **234**, 11-21 (2004).
23) L. Pei, L. Wang, G. Yu, *J. Rare Earths*, **30**, 63-68 (2012).
24) M. Anitha, D. N. Ambare, D. K. Singh, H. Singh, P. K. Mohapatra, *Chem. Eng. Res. Des.*, **98**, 89-95 (2015).
25) D.N. Ambare, S.A. Ansari, M. Anitha, P. Kandwal, D.K. Singh, H. Singh, P.K. Mohapatra, *J. Membr. Sci.*, **446**, 106-112 (2015).
26) T. Wannachod, N. Leepipatpiboon, U. Pancharoen, K. Nootong, *J. Ind. Eng. Chem.*, **20**, 4152-4162 (2014).
27) M. Mohammadi, K. Forsberg, L. Kloo, J. Martinez De La Cruz, Å. Rasmuson, *Hydrometallurgy*, **156**, 215-224 (2015).
28) N. K. Batchu, K. Binnemans, *Hydrometallurgy*, **177**, 146-151 (2018).
29) E. I. Institute. "Detection method of heavy metals in soil-Aqua regia digestion method (NIEA S321.65B)," Environmental Protection Administration Executive Yuan.
30) M. A. Kartikey K. Yadav, D.K. Singh, V. Kain, *Sep. Purif. Technol.*, **194**, 268-271 (2018).
31) A. C. Ni'am, Y.-F. Wang, S.-W. Chen, S.-J. You, *J. Taiwan Inst. Chem. Eng.*, **97**, 137-145 (2019).
32) S. Högborg, F. B. Bendixen, N. Mijatovic, B. B. Jensen, J. Holboll, "Influence of demagnetization-temperature on magnetic performance of recycled Nd-Fe-B magnets.pdf," presented at the IEEE International Electric Machines & Drives Conference (IEMDC), 2016.
33) C.-H. Lee, Y.-J. Chen, C.-H. Liao, S. R. Popuri, S.-L. Tsai, C.-E. Hung, *Metall. Mater. Trans. A*, **44**, 5825-5833 (2013).
34) T. Itakura, R. Sasai, H. Itoh, *J. Alloys Compd.*, **408-412**, 1382-1385 (2006).
35) G. Senanayake, S. Jayasekera, A. M. T. S. Bandara, E. Koenigsberger, L. Koenigsberger, J. Kyle, *Miner. Eng.*, **98**, 169-176 (2016).
36) S. S. Behera, P. K. Parhi, *Sep. Purif. Technol.*, **160**, 59-66 (2016).
37) S. Acharya, S. Mishra, P. K. Misra, *Hydrometallurgy*, **156**, 12-16 (2015).
38) A. Battsgengel, A. Batnasan, A. Narankhuu, K. Haga, Y. Watanabe, A. Shibayama, *Hydrometallurgy*, **179**, 100-109 (2018).
39) S. Pavon, A. Fortuny, M. T. Coll, A. M. Sastre, *J. Environ. Manage.*, **222**, 359-367 (2018).
40) X. Li, Z. Deng, C. Li, C. Wei, M. Li, G. Fan, H. Rong, *Hydrometallurgy*, **156**, 1-5 (2015).
41) F. V. C. Basualto, L. Molina, J. P. Munoz. E. Fuentes, J.Sapag, *J. Chil. Chem. Soc.*, **58**, 1785-1789 (2013).
42) A. Geist, W. Nitsch, J. Kim, II, *Chem. Eng. Sci.*, **54**, 1903-1907 (1999).
43) D. Xu, Z. Shah, Y. Cui, L. Jin, X. Peng, H. Zhang, G. Sun, *Hydrometallurgy*, **180**, 132-138 (2018).
44) N. Panda, N. Devi, S. Mishra, *J. Rare Earths*, **30**, 794-797 (2012).
45) R. Banda, H. S. Jeon, M. S. Lee, *Hydrometallurgy*, **121-124**, 74-80 (2012).
46) A. Kumari, K. K. Sahu, S. K. Sahu, *Metals*, **9**, 269 (2019).
47) R. Panda, M. K. Jha, J. Hait, G. Kumar, R. J. Singh, K. Yoo, *Hydrometallurgy*, **165**, 106-110 (2016).
48) F.-C. Yen, T.-C. Chang, S. Laohaprapanon, Y.-L. Chen, S.-J. You, *Solvent Extr. Res. Dev., Jpn.*, **23**, 63-73 (2016).