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Recovery of neodymium from a mixture of magnet scrap and other scrap

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Abstract

A novel neodymium (Nd) extraction process was devised and its feasibility was demonstrated in order to establish an environment-friendly process that combined scraps containing valuable metals. The newly developed device can simultaneously accomplish continuous extraction of Nd metal from scraps, re-extraction of magnesium (Mg) from Mg–Nd alloy, and finally, recovery of pure Mg. As a result, Nd metal of 98% purity was directly recovered from magnet scraps under certain conditions. Major process parameters, such as Mg/scrap mass ratio and extraction temperature, were investigated in order to evaluate the optimum conditions. The results of the investigated process indicated the possibility of extracting Nd metal directly from magnet scraps.

Keywords: Neodymium; Magnesium; Recycling; Rare earth magnet scrap; Extraction

1. Introduction

The production volume of neodymium (Nd) has dramatically increased since the development of a neodymium–iron–boron (Nd–Fe–B) permanent magnet, which had the strongest magnet power, in the 1980s [1]. Presently, more than 10,000 tons of Nd–Fe–B alloy magnets are annually produced in Japan [2]. Development of this magnet has improved the performance of electronic devices and contributed to reducing their weight. At present, the Nd–Fe–B magnet is indispensable for high-performance electronic products, such as the hard disk drive (HDD) in a personal computer. Neodymium is one of the rare earth elements that always coexist in natural ores. Multiple processes are required for the separation of Nd from other rare earth elements. Since the feed material is thermodynamically extremely stable, a large amount of energy is necessary in order to obtain the metal or alloy by reduction of the feed material. Although Nd is a relatively abundant rare earth element, it is important to develop an effective recycling process for this element, in order to conserve the environment. This is because a large quantity of solid waste and waste solution is generated during the Nd refining process. Furthermore, a large quantity of scrap is generated during the manufacturing of the Nd–Fe–B magnets for voice coil motor (VCM) in HDD, which is the primary application of Nd [3,4]. These scraps consist of not only sludge powder that is produced during magnet machining, but also “off-spec” products in which the impurity, such as oxygen content, exceeds the required specification. Although approximately 50% of Nd charged as feed material is disposed as scrap during the manufacturing process, recycling of Nd is not presently sufficient. This is because Nd forms extremely stable compounds with several elements including oxygen, thereby making it very difficult to reuse or recycle the scrap. Most developed countries import almost the entire quantity of Nd for their requirement, and a large proportion of valuable material is disposed of without being utilized as a product. The establishment of an efficient recovery process for Nd from the magnet scrap is an important issue from the viewpoint of better industrial policy as well as secure resource conservation, considering the fact that Nd is produced almost exclusively in one particular country.

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Recently, a new recycling process has been investigated, in which only valuable Nd is directly extracted from scrap using molten metal without oxidation [5,6]. Additionally, the feasibility of a new Nd extraction process in which the magnesium (Mg) extraction medium circulates due to a controlled temperature difference inside the reaction vessel was demonstrated [7]. The extraction limit of Nd from Nd–Fe alloy by molten metal was also investigated, and the theoretical extraction limit of Nd from Nd–Fe–B magnet scrap by Mg or silver (Ag) was determined by equilibrium experiments in the system Nd–Fe–X (X = Mg, Ag) at 1076 or 1363 K [8,9]. This study discusses the major process parameters for extracting Nd from magnet scraps, which are essential for developing the new Nd extraction process.

2. Experimental

Characteristics of Mg as an extraction medium are: (1) it has an extremely strong chemical affinity with Nd and forms a liquid alloy with low viscosity; (2) it is insoluble with Fe; (3) it exhibits high vapor pressure over 1073 K (800 °C), and the removal and transportation of Mg through the gas phase is easy; (4) its melting point is 922 K (649 °C), and it can be recovered by condensation as a pure solid and reused. In addition, the availability of Mg scraps is expected to increase since the use of Mg as a light-weight structural material, such as the outer casing of portable devices, is presently increasing. In our previous investigation, we experimentally demonstrated that the Mg–Nd alloy containing 24–25 mol% (65–66 mass%) Nd equilibrates with the Fe/Nd_2Fe_{17} mixture in the Fe–Mg–Nd system at 1073 K, which indicates that the theoretical extraction limit of Nd by molten Mg–Nd alloy is 24–25 mol% at 1073 K [9].

Fig. 1 shows the representative extraction apparatus used in this study. Crushed Nd–Fe–B alloy magnet scrap chips (Fe-31 mass% Nd-1 mass% B, mass of scrap: $w_{scrap} = 70–250$ g) were charged in an Fe crucible with slits of 1–2 mm, and the Fe crucible was fixed to and suspended from the top of a stainless steel vessel. Lumps of Mg (99.95 mass%, mass of Mg: $w_{Mg} = 30–70$ g) were charged in a tantalum (Ta) crucible for

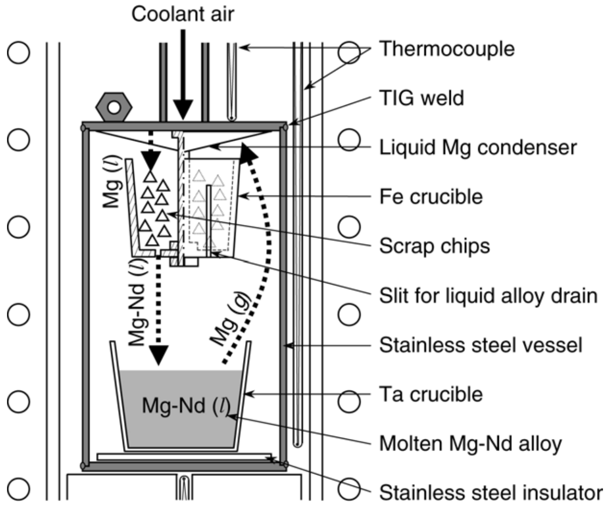

Fig. 1. Schematic illustration of the apparatus for neodymium extraction from scrap alloys using magnesium circulation.

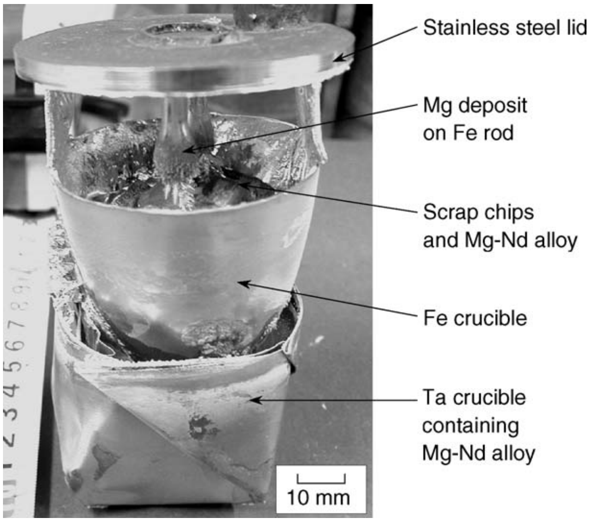

Fig. 2. Apparatus in the reaction container after neodymium extraction. Experimental conditions: mass of scrap, $w_{scrap} = 250.6$ g; mass of magnesium, $w_{Mg} = 61.4$ g; bottom temperature, $T_{bottom} = 1299$ K; holding time, $t' = 74$ h.

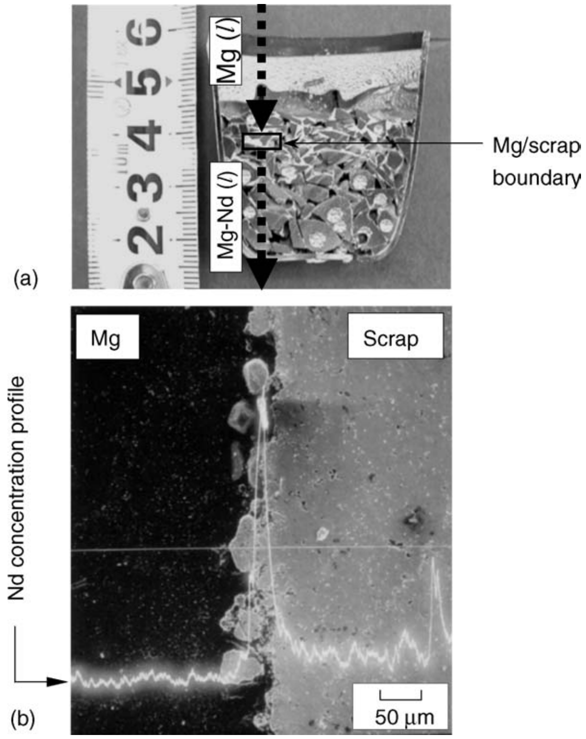

Fig. 3. (a) Sectioned iron crucible after neodymium extraction using magnesium circulation. (b) Scanning electron micrographs of the boundary between magnesium and extracted scrap. The neodymium concentration profile is also shown. Experimental conditions: mass of scrap, $w_{scrap} = 100.1$ g; mass of magnesium, $w_{Mg} = 50.4$ g; bottom temperature, $T_{bottom} = 1273$ K; holding time, $t' = 72$ h.

recovering the Mg–Nd alloy, and the Ta crucible was placed in the bottom part of the stainless steel vessel. The vessel was sealed by TIG welding and placed in an electric furnace. The bottom part of the vessel was heated to a temperature of 1073–1299 K (high temperature zone: T_{bottom}) by controlling the furnace output. Simultaneously, the top part was adjusted to a temperature of 955–1207 K (low temperature zone: T_{top}) by supplying coolant air from the top of the furnace through a gas tube. The extraction experiment was carried out for approximately 86–266 ks (24–74 h).

In the experiment, the Mg placed in the bottom part of the vessel evaporates and then condenses in the top part of the vessel. The condensed liquid Mg drips into the scrap in the Fe crucible, and the Mg reacts with Nd in the scrap alloy to form Mg–Nd liquid alloy. The Mg–Nd liquid alloy is drained through the slit of the Fe crucible into the Ta crucible. Since the vapor pressure of Nd is much lower than that of Mg ($p_{\text{Nd}}{}^{\circ} = 10^{-6}$ atm, $p_{\text{Mg}}{}^{\circ} = 0.73$ atm at 1300 K [10]), only Mg evaporates from the Mg–Nd liquid alloy, and Nd accumulates in the Ta crucible. The evaporated Mg again condenses in the top part of the vessel and acts as an extraction medium. Thus, Mg circulates in the vessel and Nd is continuously extracted from the scrap. If the scrap is merely dipped in molten Mg, the extraction of Nd is hindered as the Nd concentration in the Mg–Nd alloy increases. In this method, however, Mg without Nd is constantly supplied to the scrap and high extraction efficiency is expected even with a small amount of Mg. The experiment was terminated by cooling the vessel from the top.

3. Results and discussion

As shown in Fig. 2, the pure Mg condensed in the top part of the vessel after the experiment. The scrap chips maintained their original shape and were left in the Fe crucible. The Mg–Nd alloy was contained in the Ta crucible. The scanning electron microscopy (SEM) image of the boundary between the extracted scrap chip and Mg deposit after the experiment is shown in Fig. 3. The Nd concentration profile analyzed by energy-dispersive spectroscopy (EDS) is also shown in this figure. Neodymium in the Mg deposit was not detected by EDS, and the Nd concentration in extracted scrap was 1.3 mass%. This result indicates that Mg containing Nd was removed from the boundary, and pure Mg was continuously

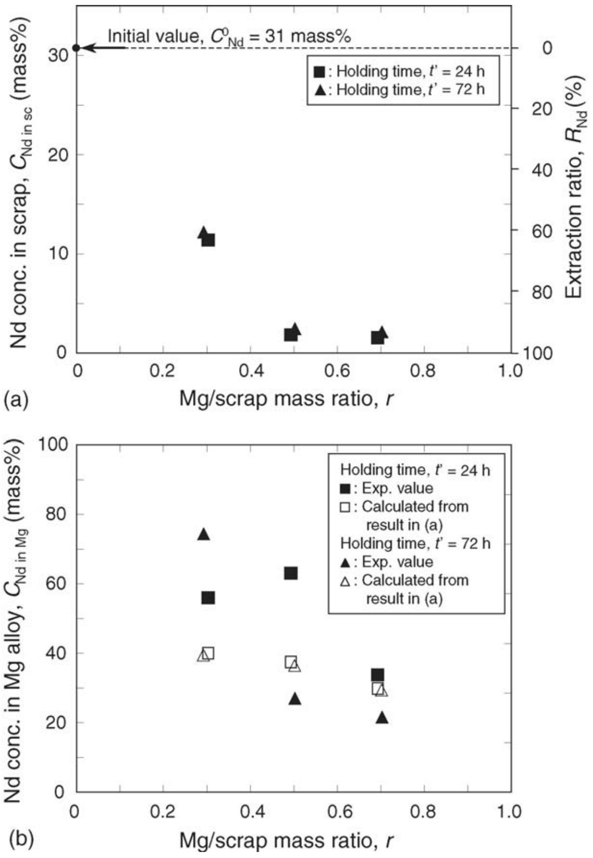

Fig. 4. Analytical results of neodymium extraction from Nd–Fe–B magnet scrap determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES). (a) Neodymium concentration in extracted scrap, (b) neodymium concentration in Mg–Nd alloy. Experimental conditions: bottom temperature, $T_{\text{bottom}} = 1273$ K; vessel height, $h'' = 124$ mm.

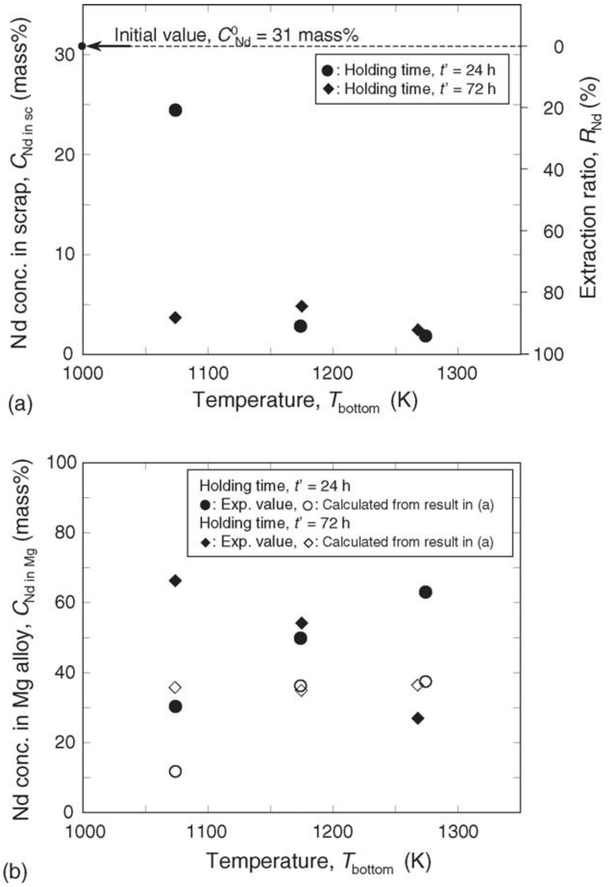

Fig. 5. Analytical results of neodymium extraction from Nd–Fe–B magnet scrap determined by ICP-AES. (a) Neodymium concentration in extracted scrap, (b) neodymium concentration in Mg–Nd alloy. Experimental conditions: Mg/scrap mass ratio, $r = 0.5$; vessel height, $h'' = 124$ mm.

supplied to the boundary by circulation of Mg in the vessel. A small region (~10 μm) with high Nd concentration exists at the boundary (Fig. 3). This is probably due to the presence of Nd oxide at the boundary. However, the exact reason is not clear at this stage.

Representative results of the extraction experiments are shown in Figs. 4 and 5. In Fig. 4, the extraction ratio increased with an increase in mass ratio of Mg to scrap (Mg/scrap: $r = w_{\mathrm{Mg}}/w_{\mathrm{scrap}}$). Fig. 4(a) shows that 95% of the Nd in the scrap could be extracted at 1273 K for 24 h. The Nd concentration in Mg–Nd alloy obtained after extraction was in the range of 22–74 mass% (Fig. 4(b)). The extraction ratio increased with an increase in T_{bottom} (Fig. 5). The relationship between the extraction temperature and Nd concentration in Mg–Nd was not highly reproducible under the conditions employed in this study. The Nd concentration in scrap decreased from 31.2 mass% to less than 1 mass%, and a high extraction rate of 99% was achieved under certain experimental conditions, i.e., with a higher mass ratio and a greater temperature difference between T_{bottom} and T_{top} [7]. Furthermore, Nd metal and Mg metal of 98% and 99% purity, respectively, were successfully obtained by evaporating Mg from Mg–Nd liquid alloy under vacuum [7]. Thus, an efficient recycling process for magnet scrap can be constructed by employing the optimized extraction condition investigated in this study ($r > 0.5$, $T_{\mathrm{bottom}} > 1173$ K).

4. Conclusions

In this study, a novel Nd extraction process was devised and its feasibility was demonstrated in order to establish an environment-friendly process that combined magnet scraps (Nd–Fe–B alloy) and Mg. Major process parameters for Nd extraction, such as mass ratio of Mg to scrap and extraction temperature, were investigated and the optimum extraction conditions of Nd from scrap were determined. The concept of "scrap combination" for recycling variable materials, demonstrated in this study, is an important technology for developing an advanced and sustainable society in the future.

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