*Materials Transactions*, Vol. 44, No. 4 (2003) pp. 798 to 801

RAPID PUBLICATION

# Direct Extraction and Recovery of Neodymium Metal from Magnet Scrap*1

Toru H. Okabe[1], Osamu Takeda[2], Kazuhiro Fukuda[3,*2] and Yoshiaki Umetsu[3]

[1]*Institute of Industrial Science, The University of Tokyo, Tokyo 153-8505, Japan*
[2]*Santoku Co. Ltd., Kobe 658-0013, Japan*
[3]*Institute of Multidisciplinary Research for Advanced Materials (IMRAM), Tohoku University, Sendai 980-8577, Japan*

An environmentally sound process for the extraction of neodymium (Nd) from magnet scrap was devised and its feasibility was demonstrated. The developed Nd extraction apparatus circulates the magnesium (Mg) as an extraction medium by maintaining a temperature difference within the reaction vessel, thus achieving continuous extraction of metal Nd from scrap, re-extraction of Mg from a Mg–Nd alloy, and finally, pure Nd metal with 97.7% purity was directly recovered from magnet scrap without oxidation. The concept of "scrap combination" for recycling variable materials, that is demonstrated in this study, is an important technology that can be instrumental in creating a highly developed self-sustainable society.

(Received February 3, 2003; Accepted February 19, 2003)

***Keywords:*** *neodymium, magnet scrap, magnesium, extraction, recycling, recovery, rare earth*

## 1. Introduction

Since the development of the iron–neodymium–boron (Nd–Fe–B) permanent magnet (the magnet with the strongest magnetic power) in 1980, the production volume of neodymium (Nd) metal has increased dramatically. Currently, about 10,000 tons of Nd–Fe–B alloy magnets are produced annually.[1,2] With improvements in mass production processes, the price of alloy material has decreased to less than one tenth its cost twenty years ago. The development of the ultra-strong magnet has significantly improved the performance of small motors, and has contributed to reducing the weight of portable electronic appliances. For example, most mobile phones have small motors that use small Nd–Fe–B magnets for their vibrating functions. This alloy magnet is primarily used in the voice coil motor (VCM), which is currently an essential device for computer hard disks. The use of the alloy magnet in hard disks has dramatically improved the performance of this storage device, and has also contributed to the reduction in the size of computers. These hard disks are now beginning to be used as storage mediums for animated digital images of movies instead of video tape recorder (VTR). With this background, the demand for metal Nd is expected to increase further.[1]

Neodymium is one of the rare earth elements. These elements always co-exist in natural ores. Multiple energy-consuming procedures are necessary for the separation of Nd from other rare earth elements by mainly solvent extraction methods. A large amount of energy is necessary to obtain the metal or alloy by reduction of the feed material, since the feed material is thermodynamically very stable. Large quantities of solid and liquid wastes are generated during the neodymium refining process, and therefore it is important to develop an effective recycling process for neodymium to protect the environment. Furthermore, a large amount of scrap is generated while manufacturing alloy magnets for VCMs, which is the primary application of Nd. This scrap contains not only sludge powder that is produced when machining the magnet, but also "off-spec" products in which the impurity of oxygen content exceeds the required specification. Even though approximately 50% of Nd charged as feed material is disposed of as scrap during the manufacturing process, Nd is currently not being recycled. This is because Nd forms an extremely stable compound with many elements including oxygen, which makes it very difficult to reuse or recycle the scrap. Most developed countries import almost 100% of their Nd requirement, and a high percentage of the valuable material is simply disposed of without being utilized as a product. Considering the fact that a large proportion of the total Nd produced worldwide comes from a few particular countries,[2] the establishment of an efficient process for the recovery of Nd from alloy scrap is a major concern from the point of view of better industrial policy as well as secure resource procurement.

## 2. Experimental

Although several methods for recovering valuable metals from magnet alloys have been investigated in the past, most of these recycling processes are based on the re-melting or oxidation of the alloy to recover Nd in the form of compounds such as $Nd_2O_3$. At this stage, an efficient recycling process has not been developed. Recently, a new recycling process has been developed in which only valuable Nd is extracted directly from scrap using liquid metal, without oxidation of Nd.[3,4] To develop a recycling process for Nd–Fe–B alloy magnet scrap, a new and effective Nd recovery method, which uses continuous magnesium (Mg) circulation in the extraction system was investigated in this study. The concept of this process of direct extraction and recovery of metallic Nd from magnet scrap is shown in Fig. 1.

The reasons for employing Mg as the extraction agent are: (1) Mg shows a very strong chemical affinity with Nd, and forms a liquid alloy with low viscosity; (2) it hardly reacts

*1A Part of Study was Presented at the Spring Meeting of the Japan Institute of Metals, held in Chiba, on March 29, 2001, and the Yazawa International Symposium (TMS/JIM), held in San Diego on March 4, 2003.
*2Graduate Student, Tohoku University.

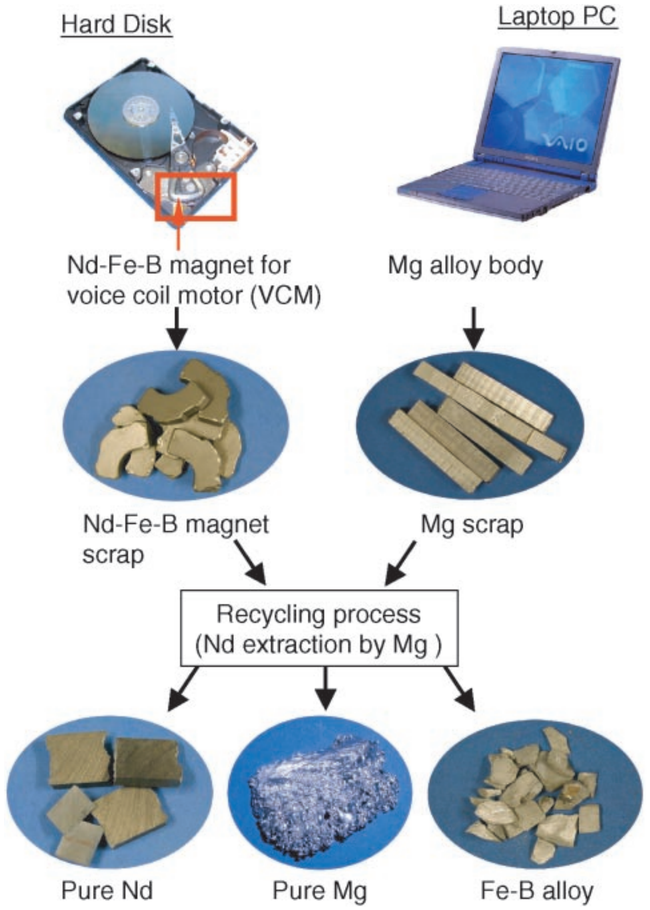


Fig. 1 Neodymium recovery process from Nd–Fe–B magnet scrap with magnesium metal as the extraction agent.

with iron; (3) it shows high vapor pressure over 1073 K (0.045 atm @ 1073 K), and the elimination and transportation of Mg through the gas phase is easy; and (4) its melting point is 922 K (649°C), and it can be recovered and re-used by condensing as a pure solid. Furthermore, Mg is one of the newer materials whose usage is expected to increase with the growth of lightweight structural material such as the casings of laptops, and as a result, the availability of Mg scrap is also expected to increase.

The extraction experiment was conducted using several self-devised apparatuses shown in Fig. 2. Nd–Fe–B alloy scrap (Fe–31 mass%Nd–1 mass%B, Mass of scrap: $w_{\mathrm{scrap}} =$ 70–250 g), which was pulverized into chips less than a few mm in size, was placed in an iron crucible with slits of 1–2 mm width. The crucible was fixed to and suspended from the top of a stainless steel reaction vessel. A tantalum crucible for recovering the Mg–Nd alloy was placed in the bottom part of the reaction vessel. Lumps of Mg (99.95 mass%, Mass of Mg: $w_{\mathrm{Mg}} = 30$–70 g) were placed in the tantalum crucible, and the reaction vessel was sealed by TIG welding. The extraction was carried out by placing the reaction vessel in an electric furnace. The bottom part of the reaction vessel was heated to a temperature of 1073–1273 K (High temperature zone: $T_{\mathrm{bottom}}$) by controlling the output of the furnace, while the top part was adjusted to a temperature of 1002–1207 K (Low temperature zone: $T_{\mathrm{top}}$) by supplying coolant gas from the top of the furnace through a

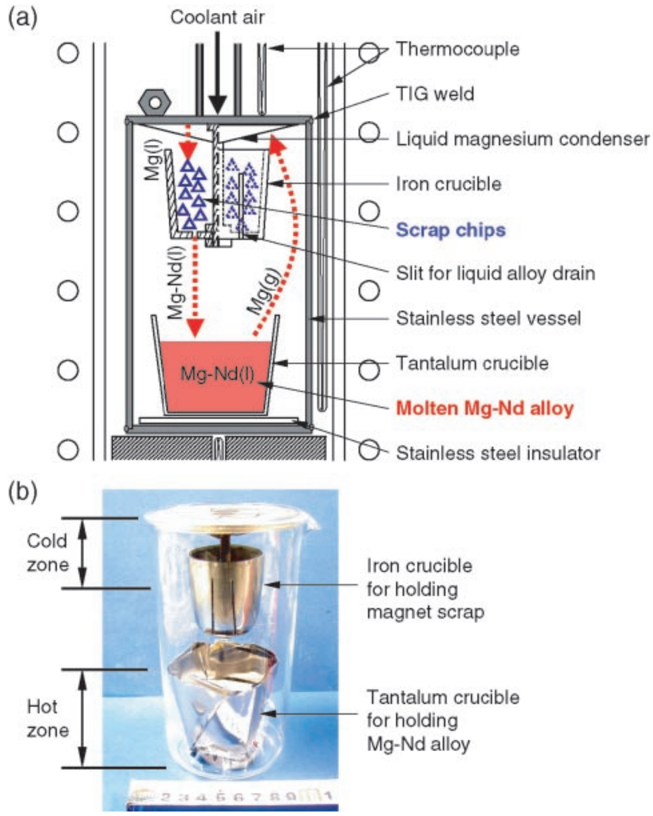


Fig. 2 Apparatus for extracting neodymium from scrap alloys using magnesium circulation. a: Schematic illustration of reaction vessel. b: Geometry of iron and tantalum crucible tentatively installed in transparent beaker.

gas tube. The experiment was conducted for approximately 86–259 ks (24 to 72 h).

By maintaining a temperature difference between the top and bottom parts of the reaction vessel, the Mg placed in the bottom part of reaction vessel evaporates, and then condenses in the top part of the reaction vessel. The condensed liquid Mg drips into the scrap in the iron crucible, and Nd in the scrap alloy combines with liquid Mg. The Mg–Nd liquid alloy thus formed is drained through the slit of the iron crucible into the tantalum crucible placed at the bottom of the reaction vessel. As the vapor pressure of Nd is extremely low, *c.a.* $10^{-6}$ atm even at 1300 K[5] (*cf.* $p_{\mathrm{Mg}} = 0.73$ atm @ 1300 K), only Mg evaporates out of the Mg–Nd liquid alloy, and Nd accumulates in the tantalum crucible. The evaporated Mg once again, condenses at the top of the vessel, and acts as an extraction medium. Thus Mg circulates in the reaction vessel, and Nd is continuously extracted from the scrap. If the scrap was simply dipped in molten Mg, the extraction of Nd is hindered when the concentration of Nd in Mg–Nd alloy increases. However, by using the circulating Mg method employed in this study, fresh Mg is constantly supplied to the scrap and extraction efficiency is expected to be high even when a small amount of Mg is used.

The experiment was terminated by cooling the reaction vessel in the furnace. When the vessel is cooled from the top, pure solid Mg condenses in the top part of the reaction vessel. The scrap alloy that is left over after Nd extraction, remains in the iron crucible and the solid Mg–Nd alloy with a high Nd concentration forms in the tantalum crucible. Mg is removed from the Mg–Nd alloy by reheating the obtained Mg–Nd alloy at higher temperatures in the range of 1123–1313 K. To enhance Mg removal efficiency, the obtained Mg–Nd alloy sample (about 8 g each) was heated at 1123 K for two to six hours under vacuum, then at 1313 K, and was then slowly cooled in the furnace. This two stage heat treatment prolongs the life time of the reaction vessel, and it is effective for minimizing processing time.

## 3. Results and Discussion

The extraction experiment was terminated by cooling the reaction vessel. As shown in Fig. 3, pure Mg condensed in the top part of the reaction vessel after the experiment. The scrap chips retained their original shape, and were left in the iron crucible (Fig. 3(b)). Results of the extraction experiments are summarized in Table 1. Depending on the mass ratio of Mg to scrap ($= w_{Mg}/w_{scrap}$) and the experimental temperatures ($T_{bottom}$ and $T_{top}$), the Nd concentration of the obtained Mg–Nd alloy varied, and was found to be in the range of 22–63 mass%. It was shown that more than 95% of Nd in the scrap could be extracted within 24 h of reaction time (Exp. # E3). Experiments that maintained a larger temperature difference and an increased amount of Mg produced an even more effective extraction ratio. In some experiments, the initial Nd concentration in the scrap, $C^0_{\text{Nd in Sc}}$ (= 31.2 mass%), decreased to 1 mass% or less, and it was shown that under some conditions, high extraction rates of 99% or more could be achieved (Exp. # E1). The boron (B) concentration in Mg–Nd alloy after extraction was 0.01–0.05 mass%, and it was found that boron in the scrap was not extracted by Mg, but rather, remained in the scrap alloy. On the other hand, a relatively high iron content, in the range of 0.37–2.26 mass% was found in the Mg–Nd alloy. The binary phase diagram for the Mg–Fe system indicates that the solubility of iron in pure Mg is 0.5 mass% or less at

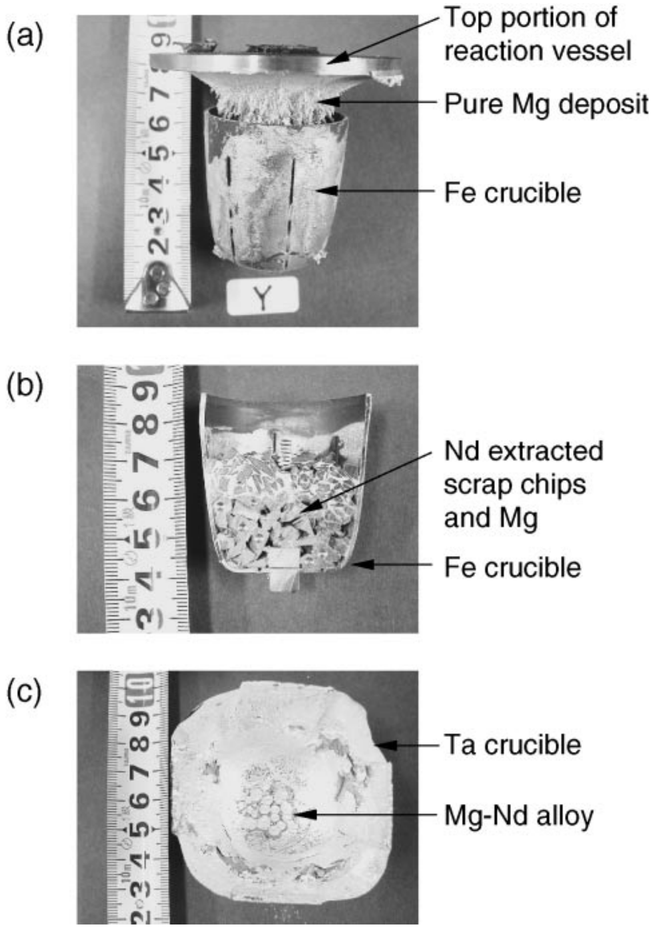


Fig. 3 Recovered samples from Nd–Fe–B scrap after Nd extraction experiment. a: Side view of iron crucible and top plate of the reaction vessel. b: Cross section of iron crucible containing scrap alloys. c: Top view of tantalum crucible containing Mg–Nd alloy. (Experimental condition: Mass of scrap, $w_{scrap} = 100.09$ g; Mass of Mg, $w_{Mg} = 69.55$ g; Bottom temp., $T_{bottom} = 1273$ K; Holding time, $t'' = 24$ h.)

Table 1 Experimental results of neodymium extraction from Fe–Nd–B magnet scrap using liquid magnesium.

| Exp. # | Mass Ratio, $w_{Mg}/w_{scrap}$ | Vessel Height, $h''$/mm | Temp., Bottom[a] $T_{bottom}$/K | Temp., Top[b] $T_{top}$/K | Holding Time, $t''$/h | Chemical analysis of samples after extraction[c] (mass%): Obtained Mg–Nd alloy, Nd $C_{\text{Nd in Mg}}$ | Obtained Mg–Nd alloy, Fe $C_{\text{Fe in Mg}}$ | Obtained Mg–Nd alloy, B $C_{\text{B in Mg}}$ | Scrap[d] after extraction, Nd $C_{\text{Nd in Sc}}$ | Scrap[d] after extraction, B $C_{\text{B in Sc}}$ | Extraction rate[e], ***R*** (%) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| E1 | 0.71 | 93 | 1098 | 884 | 40 | 49.3 | 1.60 | 0.02 | 0.07 | 0.82 | 99.8 |
| E2 | 0.70 | 100 | 1272 | 1207 | 72 | 21.6 | 2.26 | 0.01 | 2.15 | 1.21 | 93.1 |
| E3 | 0.69 | 100 | 1273 | 1200 | 24 | 33.8 | 0.70 | 0.03 | 1.57 | 1.20 | 95.0 |
| E4 | 0.65 | 92 | 1240 | 1118 | 42 | 27.8 | 0.78 | 0.05 | 0.43 | 0.91 | 98.6 |
| E5 | 0.50 | 100 | 1267 | 1196 | 72 | 27.0 | 0.37 | 0.01 | 2.46 | 1.18 | 92.1 |
| E6 | 0.50 | 100 | 1173 | 1095 | 24 | 49.9 | 0.45 | <0.01 | 2.84 | 1.24 | 90.9 |
| E7 | 0.49 | 100 | 1273 | 1193 | 24 | 63.1 | 0.90 | 0.01 | 1.86 | 1.25 | 94.0 |
| E8 | 0.49 | 100 | 1174 | 1103 | 72 | 54.2 | 0.41 | 0.01 | 4.83 | 1.15 | 84.5 |

a: Measured at exterior of the bottom part of reaction vessel.
b: Measured at exterior of the top part of reaction vessel.
c: Determined by ICP analysis.
d: Initially Fe–31 mass%Nd–1 mass%B alloy.
e: $R = 100 \times (C^0_{\text{Nd in Sc}} - C_{\text{Nd in Sc}})/C^0_{\text{Nd in Sc}}$, where $C^0_{\text{Nd in Sc}}$ is initial Nd concentration in scrap ($C^0_{\text{Nd in Sc}} = 31.2$ mass percent).

Table 2 Experimental results of neodymium metal recovery from Mg–Nd alloy by vacuum distillation.

| Exp. # | Composition of alloy before and after experiment, $C_i$ (mass%)[a] | | | |
|---|---|---|---|---|
| | Nd | Mg | Fe | B |
| Initial[b] | 54.2 | 45.5 | 0.41 | 0.01 |
| After exp. (Exp. # V1)[c] | 97.7 | <0.001 | 0.72 | <0.002 |
| After exp. (Exp. # V2)[d] | 96.2 | 0.049 | 0.76 | 0.011 |

a: Determined by ICP analysis.
b: Sample obtained in Exp. E8 in Table 1.
c: 8 g sample, 1123–1313 K for 6 h in vacuum.
d: 8 g sample, 1123–1313 K for 2 h in vacuum.

temperatures investigated in this study.[6)] It is suspected that the solubility of iron in Mg increases with an increase in Nd content, but the details of this mechanism are unknown at this point.

Results of Mg removal from a Mg–54.2 mass%Nd alloy obtained from Exp. # E8 in Table 1 is shown in Table 2. After the removal of Mg under vacuum, metallic Nd with 97.7% purity was successfully obtained. Pure Mg metal with 99% purity or more was obtained from the low temperature-condensing zone. Removal of Mg in an argon gas atmosphere was also performed in some experiments, but was found to be less effective than in the vacuum condition.

The results of this study clearly demonstrate the feasibility of a new, environmentally sound recovery process for recycling valuable metals, by combining scrap metals. It was found that liquid Mg is a suitable extraction agent, and can extract Nd from iron alloys with high efficiency at around 1100 K. The newly developed extraction apparatus for Nd is a mechanically simple and static device without moving parts, where the Mg extraction agent circulates due to differences in temperature inside the reaction vessel. This developed device can simultaneously accomplish the continuous extraction of metal Nd from scrap, the re-extraction of Mg from a Mg–Nd alloy, and the eventual recovery of pure Mg.

As major patents related to the Nd–Fe–B magnet will expire worldwide by 2007,[7,8)] the production of Nd–Fe–B magnets is expected to increase over the next few years, resulting in scrap being generated in large quantities. The authors believe that the development of new recycling processes, such as the one presented in this study, is important for the creation of a society sustained in its use of materials.

## 4. Conclusion

An environmentally sound process for the extraction of neodymium (Nd) was devised and its feasibility was demonstrated. The process involves combining scrap that contains valuable metals. The developed Nd extraction apparatus circulates the magnesium (Mg) extraction agent by maintaining a temperature difference within the reaction vessel, thus achieving continuous extraction of metal Nd from scrap, re-extraction of Mg from a Mg–Nd alloy, and finally, recovery of pure Mg. As a result, pure Nd metal with 97.7% purity was directly recovered from magnet scrap. It was also shown that the extraction agent, Mg, could be reused. The results confirmed the possibility of the direct extraction of pure Nd metal without the oxidization process.

## Acknowledgements

T.H.O. thanks Professor K. T. Jacob and Dr. Uda for their involvement throughout the project. The authors thank Mr. M. Horizawa and Mr. K. Hirota for supplying the scrap samples. This work was financially supported by a Grant-in-Aid for Scientific Research (B) from the Ministry of Education, Culture, Sports, Science and Technology, Japan (Project ID. # 12555206). All correspondence and request for material should be addressed to T.H.O. (E-mail: okabe@iis.u-tokyo.ac.jp).

## REFERENCES

1) S. Sugimoto, M. Okada and K. Inomata: *Proceedings of the Seventeenth International Workshop of Rare Earth Magnets and Their Applications*, G. C. Hadjipanayis, M. J. Bonder, Ed., (Rinton Press, 2002) 13–24.
2) S. R. Trout: *Proceedings of the Seventeenth International Workshop of Rare Earth Magnets and Their Applications*, G. C. Hadjipanayis, M. J. Bonder, Ed., (Rinton Press, 2002) 1003–1015.
3) Y. Xu, L. S. Chumbley and F. C. Laabs: J. Mater. Res. **15** (2000) 2296–2304.
4) K. Fukuda, O. Takeda, T. H. Okabe and Y. Umetsu: Proc. of Mining and Materials Institute of Japan (MMIJ), **2** (2001) 61.
5) I. Barin: *Thermochemical Data of Pure Substances*, (VHC. Weinheim, Germany, 1989).
6) T. B. Massalski: *Binary Alloy Phase Diagrams Second Edition*, (ASM International, U.S.A., 1990).
7) M. Sagawa, S. Fujimura and H. Matsu-ura: Japan Patent, S61-34242, Aug. 6 (1986).
8) J. J. Croat: U. S. Patent, No. 4802931, June. 25 (1989).