

Development of Chemical Process for Recovering High-quality Rare-earth Oxides from HV Motor Magnets

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ABSTRACT

In the automobile industry, interest in the prevention of global warming has always been high. The development of eco cars (HV, EV etc.), aimed at reducing CO₂ emissions during operation, has been progressing. In the announcement of its "Toyota Environmental Challenge 2050", Toyota declared its commitment to creating a future in which people, cars, and nature coexist in harmony. In this declaration, Toyota committed to reducing CO₂ emissions not only during operation but also over the entire life cycle of vehicles, and to using resources effectively based on a 4 R's approach (refuse, reduce, reuse, and recycle). Although eco cars decrease CO₂ emissions during operation, most of them increase CO₂ emissions during manufacturing. For example, the rare-earths (Nd, Dy etc.) used in the magnets of driving motors are extracted through processes that produce a significant amount of CO₂ emissions. The common process for recycling the rare-earths used in magnets can recover high-purity rare-earths by electrolysis. However, this process is costly and also produces a significant amount of CO₂. Toyota has developed a chemical recycling process for producing high-quality rare-earth oxides that is economical and reduces CO₂ emission during material production by 80% without the use of electrolytic refining. This paper describes this chemical recycling process for rare earth components and, in addition, introduces an energy-saving, vehicle-to-vehicle resource recycling flow.

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INTRODUCTION

In its "Toyota Environmental Challenge 2050", Toyota set itself the six challenges described in Fig. 1. One of the targets is to increase the production and adoption of eco cars, as shown in Fig. 2. However, it is imperative to perform a life cycle assessment (LCA) for the production of the eco vehicles and to target the reduction of CO₂ emissions across the entire life cycle of the vehicles, including raw material, parts sourcing and production.

All of the next-generation eco cars listed in Fig. 2 will use an electric motor as the main source of power. Fig. 3 summarizes the amount of CO₂ discharged during the manufacture of motor materials: manufacturing 1 kg of rare-earths contained in motor magnets results in the discharge of as much as 18.8 kg of CO₂. Clearly, from the viewpoint of LCA, technologies for recycling of rare-earth materials are critical to the viability of eco cars.

This section explains why rare-earths are needed for a magnet of high-performance electric motor. A neodymium magnet (also known as Nd-Fe-B), is typical magnet which includes rare-earths (Nd, Dy) [1][2]. A neodymium magnet has 10 times higher performance than a ferrite magnet. Dy is added to enhance magnetic coercive force. Recent studies have been trying to development less or free rare-earth magnet [3][4]. However, it's difficult that those magnets show the

equal performance to a neodymium magnet and use of rare -earths is presently indispensable to miniature and/or superior performance of an electric motor.



Figure 1. Toyota Environmental Challenge 2050

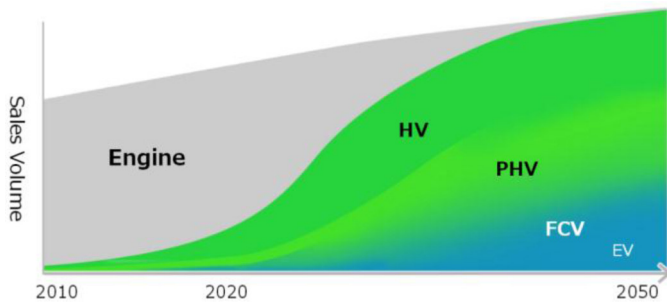


Figure 2. Next generation vehicles to accelerate technological development to follow market expansion of HV

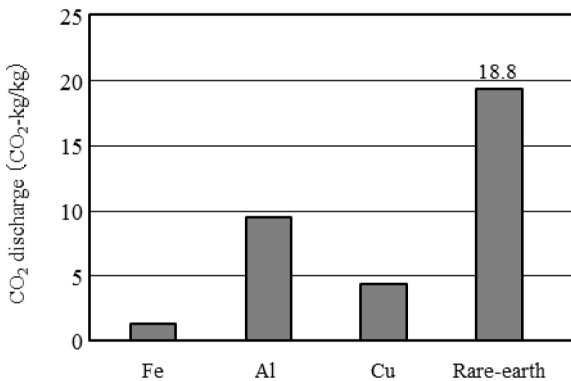


Figure 3. CO₂ discharge per 1 kg of the motor materials when manufacturing

Fig. 4 shows some vehicle applications for rare-earth resources. In addition to magnets, rare -earths are used in catalysts and lamps. These products must be manufactured using high-quality materials and components in order to obtain high performance. Therefore, end-of-life components, like magnets, cannot be reused; instead the high-quality rare-earths they contain must be recovered and reprocessed into new components.



Figure 4. The adaptation example of the rare-earths in the car

This paper will focus on a method of extracting magnets from motors in order to recycle the rare-earths contained in the magnets. In the latter part, the paper will briefly describe vehicle-to-vehicle implications of rare-earths recycling.

Fig. 5 shows the structure of a motor. The drive motor, used in models such as the Prius, has magnets incorporated into the magnetic steel sheet of the rotor. Various methods for extracting these magnets have been investigated [5]. For example, the magnets can be separated by heating and vibrating the magnetic steel sheet after the rotor shaft is removed. However, because this separation method is costly, the author believes that a structural design for improving recyclability will also be essential in the future.

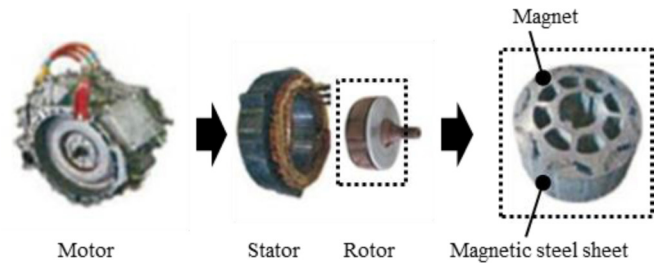


Figure 5. Structure of the driving motor

Fig. 6 shows the conventional flow for recovering rare-earths from magnets. As shown, high-quality, pure rare-earths can be obtained using an Fe removal process utilizing heat and electrolytic refining that requires a large amount of energy [6][7].

Metallic element can't be taken apart without being melted like this case. So electrolysis refinement and hydrometallurgy, like chemical processing using acids and solvent extraction, are indispensable technology in a process of the dissociation, concentration and collection of the rare metal [8][9].

However, this process is insufficient to meet the total life cycle CO₂ emissions targeted in the Toyota Environmental Challenge 2050. A new process is required that produces less CO₂ while at least maintaining the efficiency of rare earth recovery.

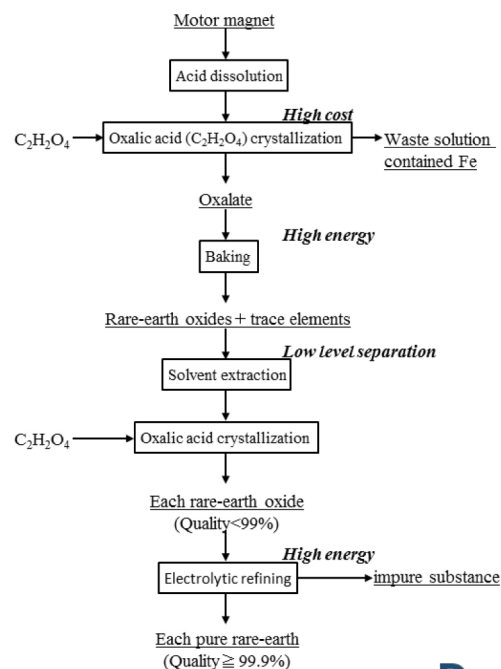


Figure 6. The general recycle process of the rare-earths

Fig. 7 shows the Toyota developed process for recycling of rare earth magnets. The process consists of four steps: (1) a magnet-dissolution process that utilizes waste acid; (2) an Fe removal process with reduced heat energy requirement; (3) a rare-earth separation process; (4) a crystallization process that removes the remaining trace elements. The use of this process, which does not involve electrolytic refining, yields a 99.9% separation of the rare earths while reducing CO₂ emissions during material recovery by 80%. Further, the produced rare earth oxides contain less than 100 ppm of impurity elements. Rare earth oxides of this purity level are appropriate for exhaust gas filters and so recycling of rare earth magnets towards this application has begun. The following section will describe the chemical recycling process yielding the high-quality rare-earth oxides in more detail.

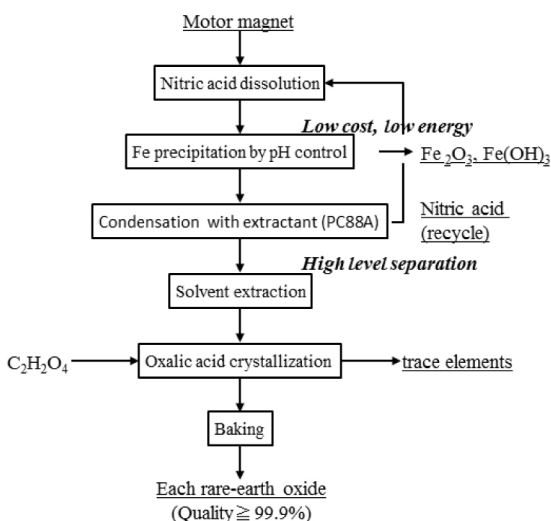


Figure 7. The developed recycle processes of the rare-earths

CHEMICAL PROCESS FOR RECYCLING HIGH-QUALITY RARE-EARTH OXIDES

Magnet-Dissolution Process Utilizing Waste Acid

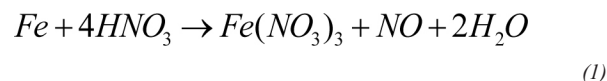
First, an acid suitable for dissolving magnets was identified. One organic acid and five inorganic acids, shown in Table 1, were tested. All these acids are waste acids generated in other industries and therefore, can be obtained on a recycled-for-free basis or free of charge. For example, if nitric acid were to be used, relatively high-quality waste acid used for cleaning electronic printed circuit boards could be obtained.

The suitability of the acid for the dissolution of magnets was judged based on: (a) ability to dissolve the magnet; (b) the quantity and type of gas generated; (c) the stability of the acid. The dissolvability evaluation determined whether dissolution was impeded by the compound generated on the surface of the magnet. The gas generation evaluation determined whether any toxic gas was generated. The stability evaluation determined whether the rare-earths precipitated as salts due to pH fluctuations of the acid, worsening the processing efficiency and yield.

Table 1. The select of the optimum acid to dissolve a magnet by three points evaluations (solubility, gas generation and stability)

		Solubility	Gas generation	Stability
Organic	CH ₃ COOH	×	○	—
Inorganic	HCl	○	×	○
	HF	○	×	○
	H ₃ PO ₄	Δ	○	×
	H ₂ SO ₄	Δ	○	×
	HNO ₃	○	Δ	○

Based on the evaluation results, nitric acid was selected as the ideal acid. Nitric acid reacts with the magnet as shown in Equation (1), generating NO_x (nitrogen oxide). This NO_x can be easily recovered using an alkali scrubber and therefore, the decision was made to use nitric acid.

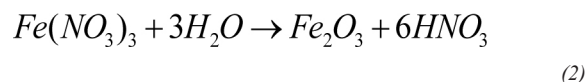


Next, using the selected acid, dissolution conditions were studied, using magnets described in Table 2, to identify conditions leading to high efficiency dissolution.

Table 2. Composition of the sample magnet in Percentage

	Nd	Dy	Fe	Co	B	Cu	Mn	Al	Ca
Sample magnet	22.3	5.2	69.5	1.3	1.0	0.2	<0.1	0.5	<0.1

It is known that nitric acid reacts with iron as shown in Equation (2) [10].



When the temperature of nitric acid is maintained at 50°C or higher, the reaction described in Equation (2), in which the Fe(NO₃)₃ generated by dissolving iron returns to nitric acid (HNO₃), occurs repeatedly. However, because the dissolution of the magnet by nitric acid as described in Equation (1) is a strongly exothermic reaction, it is necessary to control the temperature by controlling the reaction rate. To this end, the processing method shown in Fig. 8 was used. A 17.6kg mass of magnet is soaked in 24 L of water, to which is added, at a controlled rate, a solution of 67.5% nitric acid. This methodology maintained the temperature of the reaction bath between 60 and 70°C.