



# Opportunities for plasma separation techniques in rare earth elements recycling



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## ABSTRACT

Rare earth elements recycling has been proposed to alleviate supply risks and market volatility. In this context, the potential of a new recycling pathway, namely plasma mass separation, is uncovered through the example of neodymium - iron - boron magnets recycling. Plasma mass separation is shown to address some of the shortcomings of existing rare earth elements recycling pathways, in particular detrimental environmental effects. A simplified mass separation model suggests that plasma separation performances could compare favourably with existing recycling options. In addition, simple energetic considerations of plasma processing suggest that the cost of these techniques may not be prohibitive, particularly considering that energy costs from solar may become significantly cheaper. Further investigation and experimental demonstration of plasma separation techniques should permit asserting the potential of these techniques against other recycling techniques currently under development.

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## 1. Introduction

Owing to their unique ferromagnetism, superconductivity and luminescence properties, rare earth elements (REEs) are key components in a large number of technologies. Although REEs have long been used in mature markets, such as catalysts, glassmaking and lighting, REEs are now also sought after by emerging, high-growth markets, such as permanent magnets and battery alloys (Goonan, 2011; Zepf, 2013; Brumme, 2014). Quantitatively, the global REEs mining expanded on average by 7% annually between 1990 and 2006 (Goonan, 2011), while REEs demand for permanent magnet manufacturing alone grew by 280% between 2000 and 2007 (equivalent to 16% annually) (Yang et al., 2016). Looking ahead, the globalization of low-carbon energy systems, and in particular of wind turbines and electric cars, is projected to yield demand growth of 700% and 2600% over the next 25 years for respectively neodymium (Nd) and dysprosium (Dy) (Alonso et al., 2012) by virtue of their unmatched performances for high strength, high temperature magnets (Jacoby, 2013).

On the supply side, REEs production is extremely unequally distributed worldwide. A single country - China - is responsible for

over 80 percent of the current global mining production (and even for as much as 97% up until 2010 (Brown et al., 2013; Chakhmouradian and Wall, 2012)), and controls over 50% of worldwide mineral reserves (Hatch, 2012; U.S. Department of the Interior, 2013; Humphries, 2013; U.S. Department of the Interior, 2015, 2017). A direct consequence of this near-monopoly situation is a very vulnerable market and a high volatility of REEs price. This market volatility led to the price spike of 2011, during which the price of some REEs experienced a 10-fold increase over a few months (U.S. Department of Energy, 2011; Hatch, 2012). The uncertainty with respect to price, availability, and quality of raw materials is a serious concern for many industries in countries that are almost 100% import-reliant, such as the USA and multiple E. U. countries. This situation led panels and governmental agencies to place REEs, in particular neodymium (Nd), yttrium (Y), dysprosium (Dy), europium (Eu) and terbium (Tb), on their *critical* raw elements list (Moss et al., 2011; U.S. Department of Energy, 2011). Furthermore, the main consumer countries began implementing mineral strategies to minimise their vulnerability to the supply of REEs (United States Environmental Protection Agency, 2012; European Commission, 2013).

One strategy is supply diversification (Golev et al., 2014; Barteková and Kemp, 2016). This option has led to the start-up or reviving of exploration projects and mining production around the

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world to alleviate supply risks (Walters and Lusty, 2011; Mariano and Mariano, 2012; Sarapää et al., 2013; Goodenough et al., 2016; Machacek and Kalvig, 2016). The trigger effect of the 2011 price spike in the geographical diversification of REEs mining is hardly debatable. As a matter of fact, China had 97% of the REEs market share up until 2010 (Brown et al., 2013), whereas about a half dozen countries produced REEs in 2015, with Australia, India, the USA, and Russia combining for about 17% of the global production (Brown et al., 2016; U.S. Department of the Interior, 2017). Although mining expansion has obvious benefits from a market stability perspective, REEs mining comes at a significant environmental cost (Humsa and Srivastava, 2015; Charalampides et al., 2016; Lee and Wen, 2016; Browning et al., 2016; Huang et al., 2016). In addition, health hazards associated with REEs mining are only beginning to receive attention (Li et al., 2013; Rim et al., 2013; Pagano et al., 2015; Rim, 2016). Although it has been suggested that the environmental impact of mining could be reduced if subjected to stricter environmental legislation (Schreiber et al., 2016), it remains unclear whether REEs mining will be societally accepted in European countries, as exemplified by the ongoing debate in Denmark on REEs mining in Greenland (Rosen, 2016). Finally, mining expansion does not address the so-called *balance* problem (Falconnet, 1985; Binnemans and Jones, 2015), which stems from the fact that natural REEs abundance in ores does not match the market demand of each individual REE.

Another option to address the supply-risks while limiting environmental damage and mitigating the balance problem is recycling (Bloodworth, 2013; Rademaker et al., 2013; Tsamis and Coyne, 2014; Verrax, 2015; Barteková and Kemp, 2016). Correlation between recycling rate and market price stabilization has notably been observed previously for cobalt and platinum (Darcy et al., 2013; Verrax, 2015; Bandara et al., 2015). Despite these strong incentives, less than 1% of REEs were recycled in 2011 (Graedel et al., 2011; Reck and Graedel, 2012; Binnemans et al., 2013). Various reasons have been given to explain REEs low recycling level, such as the lack of effective collection systems, the difficulty to extract REEs from scrap, or the relatively low prices prior to 2011. However, one of the main impediment to the development of REEs recycling appears to be the low REEs content of most end-products (Golev et al., 2014). Mass content of REEs for most applications is lower than 5 g/kg (0.5%), and as low as 0.5 g/kg (0.05%) in LEDs (Chancerel et al., 2013). One exception is neodymium - iron - boron ( $\text{Nd}_2\text{Fe}_{14}\text{B}$ , or NdFeB for short) permanent magnets, for which the REEs mass content can be as large as 30%.

Interestingly, NdFeB magnets currently happen to dominate the permanent magnets market thanks to their superior energy product (Jacoby, 2013). Small NdFeB magnets are used extensively in consumer products, such as hard disk drives and loudspeakers, while large NdFeB magnets are increasingly used in electric vehicles and windmill turbines (Binnemans et al., 2013). Although large magnets can be efficiently disassembled to be recycled, disassembling of electronic goods is key in making REEs recovery from these products attractive (Tsamis and Coyne, 2014; Sprecher et al., 2014). Indeed, without efficient pre-processing, rare earth magnets in electronic goods are shredded along with waste electronics and electrical equipment, decreasing in turn significantly the mass content of REEs.

Recent reviews on NdFeB magnets recycling (Takeda and Okabe, 2014; Yang et al., 2016) show that the two main techniques considered to date are hydro-metallurgy and pyro-metallurgy. Hydro-metallurgical recycling typically involves processes very similar to those used for REEs extraction from primary ores (Krishnamurthy and Gupta, 2015). These processes rely heavily on the use of chemicals, including strong mineral acids. This generally leads to the production of large volumes of liquid wastes and a

significant environmental footprint, but new separation schemes based on green chemistry (Bandara et al., 2016), closed loops (Kitagawa and Uemura, 2017) and coordination chemistry (Bogart et al., 2015, 2016) are currently under development to limit the footprint of hydro-metallurgical NdFeB magnets recycling. On the other hand, pyro-metallurgical or high-temperature processing is believed to be more environmentally friendly thanks to reduced water consumption and hazardous waste production (Firdaus et al., 2016), but at the expense of a greater energy input. Yet, some high-temperature processes still require chemicals and generate wastes. For example, among pyrometallurgical techniques, some liquid-phase processes such as electroslag refining generate large amount of solid-waste (Yang et al., 2016) while certain gas-phase processes require large amount of chlorine gas (Firdaus et al., 2016). High-temperature processes are also believed to produce larger amounts of emissions (Akhori et al., 2014). A promising recent development in high-temperature NdFeB magnets recycling is the vacuum induction melting - magnetic separation (VIM-HMS) process (Bian et al., 2016), which requires no toxic chemicals and produces minimal solid waste. However, this process might be challenged by complex compositions, in particular other transition metals such as copper and nickel (Yang et al., 2016). Besides hydro- and pyro-metallurgical techniques, new recycling process have recently been proposed, for example REEs absorption on bio-materials such as salmon milt (Takahashi et al., 2014) and bacteria (Bonificio and Clarke, 2016; Park et al., 2016).

In this paper, we illustrate the potential of plasma mass separation techniques for rare earth elements recycling through the example of NdFeB magnets. First, the general features and intrinsic advantages of plasma separation are introduced. The potential of plasma mass separation for NdFeB magnets recycling is then discussed, and a preliminary cost estimate is derived. Finally, concept improvements are suggested.

## 2. Plasma separation

By operating on dissociated molecules, plasma separation can be used where chemical techniques are challenged. Applications for which these capabilities could prove extremely valuable include nuclear legacy waste disposal (Siciliano et al., 1993; Freeman et al., 2003; Gueroult et al., 2015) and spent fuel reprocessing (Zhil'tsov et al., 2006; Gueroult and Fisch, 2014; Timofeev, 2014). By accommodating straightforwardly complex chemical compositions, plasma separation could in principle handle coatings, additives and contaminants which are typically found in permanent magnets (Firdaus et al., 2016). In addition, plasma separation could handle equally sintered magnets or resin bonded magnets. Furthermore, plasma separation does not require chemicals, nor creates additional waste streams. By virtue of these properties, plasma separation could in principle offer an environmentally friendly pathway for REEs recycling. However, as it will be shown later, the energy input required to separate products scale linearly with the number of atoms in the feed for plasma separation. Plasma separation is therefore expected to be most attractive for concentrated feeds such as large magnets, and less for diluted feeds such as waste electronics.

Although plasma separation can be envisioned in many ways, it essentially boils down to three core technologies. First, the input stream has to be turned into a plasma. By heating material to very high temperatures, the bonds that commonly form chemical substances are broken. Upon further heating, atoms are ionized, leaving individual ions and electrons, *i.e.* a plasma. A variety of well established techniques can be used for plasma formation, including for example laser ablation (Boulmer-Leborgne et al., 1993), arc-discharges (Brown and Oks, 2005), or dust injection (Tanaka

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