Waste Management xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Identification and recovery of rare-earth permanent magnets from waste electrical and electronic equipment

A. Lixandru [a,b,*], P. Venkatesan [c], C. Jönsson [d], I. Poenaru [a,b], B. Hall [e], Y. Yang [c], A. Walton [d], K. Güth [a], R. Gauß [a], O. Gutfleisch [a,b]

[a] Fraunhofer ISC – Project Group Materials Recycling and Resource Strategies IWKS, 63457 Hanau, Germany
[b] Technische Universität Darmstadt, Functional Materials, 64287 Darmstadt, Germany
[c] Delft University of Technology, Department of Materials Science and Engineering, 2628 CD Delft, The Netherlands
[d] University of Birmingham, School of Metallurgy and Materials, Birmingham B15 2TT, UK
[e] Stena Metall AB, 40040 Gothenburg, Sweden

ARTICLE INFO

Article history:
Received 16 February 2017
Revised 19 May 2017
Accepted 17 July 2017
Available online xxxx

Keywords:
Rare-earth scrap permanent magnets
Nd-Fe-B
Waste streams selection
WEEE
Recovery
Recycling

ABSTRACT

Nd-Fe-B permanent magnets are a strategic material for a number of emerging technologies. They are a key component in the most energy efficient electric motors and generators, thus, they are vital for energy technologies, industrial applications and automation, and future forms of mobility. Rare earth elements (REEs) such as neodymium, dysprosium and praseodymium are also found in waste electrical and electronic equipment (WEEE) in volumes that grow with the technological evolution, and are marked as critical elements by the European Commission due to their high economic importance combined with significant supply risks. Recycling could be a good approach to compensate for the lack of rare earths (REs) on the market. However, less than 1% of REs are currently being recycled, mainly because of non-existing collection logistics, lack of information about the quantity of RE materials available for recycling and recycling-unfriendly product designs. To improve these lack of information, different waste streams of electrical and electronic equipment from an industrial recycling plant were analyzed in order to localize, identify and collect RE permanent magnets of the Nd-Fe-B type. This particular type of magnets were mainly found in hard disk drives (HDDs) from laptops and desktop computers, as well as in loudspeakers from compact products such as flat screen TVs, PC screens, and laptops. Since HDDs have been investigated thoroughly by many authors, this study focusses on other potential Nd-Fe-B resources in electronic waste. The study includes a systematic survey of the chemical composition of the Nd-Fe-B magnets found in the selected waste streams, which illustrates the evolution of the Nd-Fe-B alloys over the years. The study also provides an overview over the types of magnets integrated in different waste electric and electronic equipment.

1. Introduction

The demand and consequently the production of consumer electrical and electronic equipment (EEE), -computers, TV-sets, sound systems, refrigerators, mobile phones, etc. - have increased continuously over the last decades (European Commission, 2016). The rapid rate of technological development combined with a decrease in the prices of EEE and a growing demand has led to significant increase of generated waste streams. It is estimated that a quantity of 30–50 million tons of waste electrical and electronic equipment (WEEE) is produced worldwide every year (Cucchiella et al., 2015). In Europe, WEEE are considered to have the highest growth rate (3–5% per year) of all waste consumer goods (European Commission, 2016). The amount of WEEE generated in Europe in 2012 amounted to 9.45 million tons and it is estimated to increase to 12 million tons by 2020 (European Commission, 2016).

WEEE come with a complex mixture of components and materials containing both valuable materials that can be recycled and hazardous substances that can have a negative environmental impact (Robinson, 2009). This kind of waste contains numerous economically interesting materials but industrial recycling processes are mainly focused on the recovery of bulk materials (polymers, glass and ferrous scrap) and on high value materials like precious metals (gold, silver, and platinum group metals), and base

* Corresponding author.
E-mail address: lixandru.alexandru@gmail.com (A. Lixandru).

http://dx.doi.org/10.1016/j.wasman.2017.07.028
0956-053X/

metals like copper and aluminum (Zepf et al., 2014). Rare earth elements (REEs) such as neodymium, dysprosium and praseodymium are also found in WEEE in volumes that grow with the technological evolution. There is a growing competition in the use of REEs, particularly in the automotive sector, wind power, electric bikes, and air conditioning, apart from their standard use in an endless number of electric motors and generators in industrial machinery and domestic appliances (Yang et al., 2016). Mined rare-earths are mainly used for the production of the permanent magnet and other types of alloys, chemical catalysts, polishing materials and phosphors (Wübbeke, 2013). Nd-Fe-B magnets are used in computers HDDs and audio systems loudspeakers, wind turbines, automobiles and to a lesser extent in household appliances and magnetic resonance imaging (MRI) machines (Du and Graedel, 2011) (see Fig. 1).

WEEE could therefore be a valuable source for rare-earth (RE) materials recovery and recycling as they contain Nd-Fe-B magnets (for instance in loudspeakers and hard disk drives), phosphors containing REs (in fluorescent lamps and LCDs) (Schüler et al., 2011) and NiMH batteries (Meshram et al., 2016). Since recycling of NiMH batteries already exist on a commercial scale, the recycling efforts should be focused on the magnets and phosphors. Nd-Fe-B magnets typically contain 27–31 wt% REEs (Croat et al., 1984; Sagawa et al., 1984), which is much higher compared to the phosphors. Apart from Nd, this share may also consist of Pr, Dy, Tb, Gd or Ho and, depending on the specific magnet alloy composition and the field of magnet application they can also contain additives like cobalt, zirconium, molybdenum, and niobium.

REs are marked as critical elements by the European Commission due to their high economic importance combined with significant supply risks (Binnemans et al., 2013; European Commission, 2014), however recycling of these metals from waste streams is not yet commercially conducted. In fact, less than 1 % of the REs are currently being recycled mainly due to low collection rates and lack of mature and economically feasible technologies (ERECON, 2014). However, during the last 5 years, research into the recycling of RE has been intensified significantly by many groups worldwide, and in Europe projects such as REMANENCE, REPROMAG, EREAN, DEMETER, MORE, PROSUM and ReCreew project were initiated. The EU also set up a Rare Earth Competency Network, ERECON, which in its final report (ERECON, 2014) suggested recycling of REs from WEEE as an urgent priority in order to develop a sustainable RE supply chain in Europe.

The main challenges of WEEE magnets recycling are that they are often present in small quantities in electronic equipment components. They are often embedded and glued in place within the products, have a protective coating layer of nickel, copper or zinc, and they are magnetized, making their extraction and recycling difficult. However, when extracted, Nd-Fe-B magnets can be directly re-used or recycled to regain the magnet alloys for the production of new magnets (e.g. using hydrogen (Zakotnik et al., 2008; Gutfleisch et al., 2013; Walton et al., 2015; Sheridan et al., 2012; Sheridan et al., 2016; Lixandru et al., 2017)) or alternatively, the REEs or oxides can be extracted using various pyrometallurgical or hydrometallurgical routes (Yang et al., 2016).

Although the RE contents of different end-of-life (EOL) electronic products such as HDDs have been estimated previously (Rademaker et al., 2013), a real life survey of RE identification in various streams of WEEE plants has not been carried out. In this work, we focus on the identification of Nd-Fe-B permanent magnets from different, well-defined streams of WEEE present at the recycling company Stena Technoworld in Halmstad, Sweden as a case study representing a large recycling company in Europe.

2. Materials and methods

This study was performed at Stena Technoworld, Halmstad, Sweden where the total inflow of WEEE is approximately 1000 tons per month, of which 750 tons comes from Sweden and 250 tons (only CRT and flat screen TVs) from Denmark. The first treatment of the WEEE at the recycling plant is the sorting and depolluting, where the objects containing hazardous waste are selected and removed manually. Batteries, mercury, Polychlorinated Biphenyls containing capacitors, lead and asbestos are examples of hazardous waste being removed. LCD screens are dismantled to remove their lead-containing lamps while CRT screens are also selected for of their lead-containing glass. During the dismantling, other constituents such as printed circuit boards (PCB), aluminum parts and copper coils are also plucked out and sorted. LEDs and Plasma screens are however not disassembled being sent directly to the shredder. Non-hazardous products are sorted depending on their main constituent, for instance, loudspeakers being sorted into a wood fraction, and vacuum cleaners and microwave ovens into a plastics fraction. Small electronic devices such as mobile phones and electric toothbrushes are sorted into a separate stream. Further recycling procedures of these different waste streams at Stena Technoworld is given in Fig. 2. Most of the materials are shredded and then separated using various techniques in the PMR (Precious Metal Recycling) process, and PRC (Plastics Recycling Center). The recycling process yields the following fractions: ferrous scrap, copper and aluminum scrap, PCB, precious metals and copper, recyclable plastics, brominated plastics, and other.

This study focused on sorting and identifying the streams that have the potential to become valuable secondary resources for recycling of Nd-Fe-B magnets. For this purpose various WEEE appliances were disassembled manually. The extracted magnets were demagnetized by heating to 350 °C in a muffle furnace and their surface was then ground in order to remove the protective coating layer. A portable XRF device (Thermo Scientific Niton XL3t) was used to determine the chemical composition of the collected scrap magnets. Americium-241 isotope was used as the X-ray generating source, since it permits distinguishing between

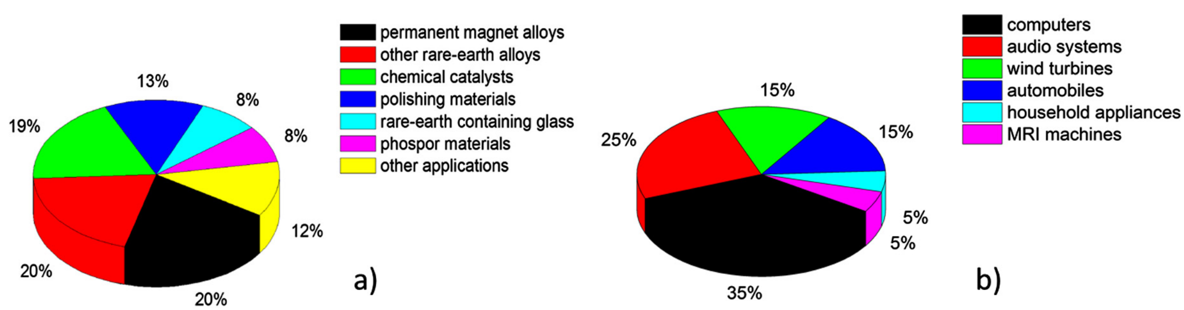

Fig. 1. Industrial use of mined REEs (a) and applications of Nd-Fe-B permanent magnets (b) (Wübbeke, 2013; Du and Graedel, 2011).

Please cite this article in press as: Lixandru, A., et al. Identification and recovery of rare-earth permanent magnets from waste electrical and electronic equipment. Waste Management (2017), http://dx.doi.org/10.1016/j.wasman.2017.07.028