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

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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

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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

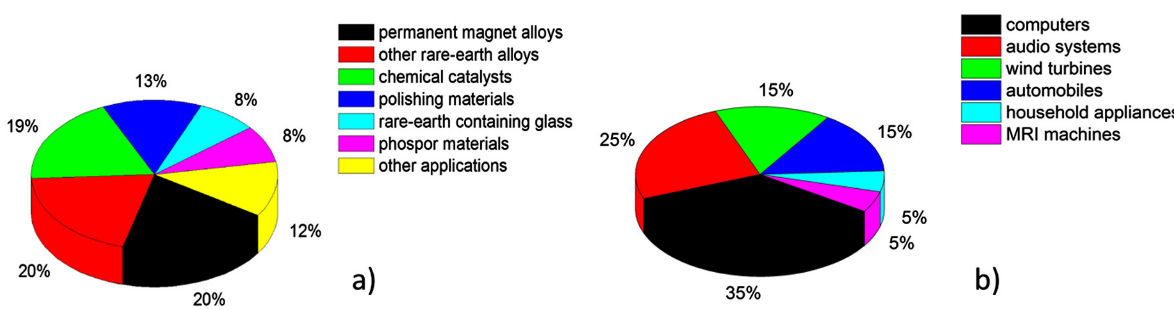

Fig. 1. Industrial use of mined REEs (a) and applications of Nd-Fe-B permanent magnets (b) (Wübbeke, 2013; Du and Graedel, 2011).

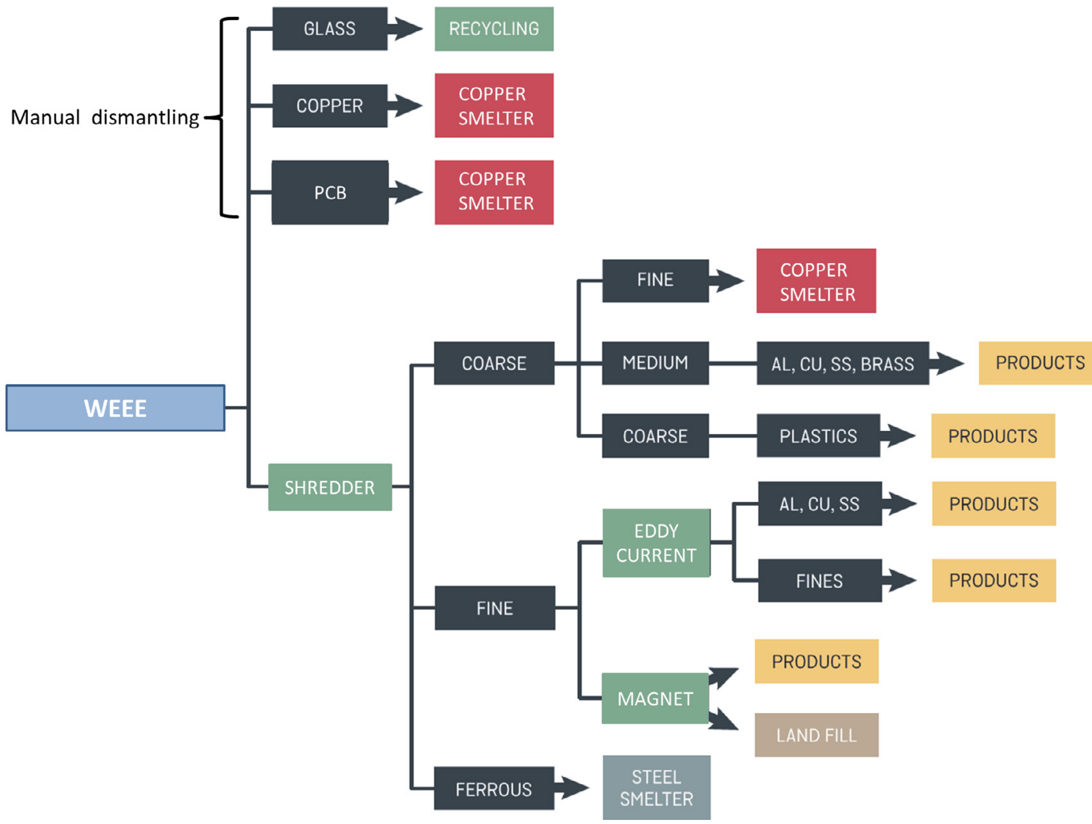

Fig. 2. Diagram of the sorting and recycling process developed at Stena Technoworld's WEEE recycling plant.

REEs such as neodymium, dysprosium and praseodymium. For exact elemental composition quantification, ICP-OES measurements were performed using a PerkinElmer - Optima 8300 system after a complete sample dissolution in HCl-HNO_3 acid mixture. Prior to the ICP-OES measurements the magnet coating was removed.

The following devices were chosen for disassembly and analysis from different waste categories:

1. Large household category: microwaves, electric fans and air conditioner appliances
2. Small household appliances: vacuum cleaners (including hand-held and smaller vacuum cleaners)
3. IT and telecommunication equipment: printers and copying equipment, pocket calculators, mobile telephones, desktop computers, minicomputers, laptops, notebook computers, notepad computers
4. Consumer equipment and photovoltaic panels: radio sets, television sets and video cameras
5. Electrical and electronic tools: power tools including drills, saws, and screwdrivers
6. Toys, leisure and sports equipment: small electric toys with electric motors, electric scooters
7. Monitoring and control instruments: alarms (Directive 2012/19/EU).

An important stream that contains Nd-Fe-B magnets is composed of hard disk drives (HDDs), which have been studied thoroughly and identified as an important secondary resource for the RE permanent magnets (Zakotnik et al., 2008; Gutfleisch et al., 2013; Walton et al., 2015; Sprecher et al., 2014; München et al., 2017). HDDs contain between 1 g and 30 g of Nd-Fe-B magnet depending on its size (Ueberschaar and Rotter 2015). Habib et al. (2015) tracked the REEs during shredding and further separation of hard disks in a WEEE recycling plant in Denmark and found that 90% of the Nd-Fe-B magnets remained stuck to various ferrous parts of shredder and other equipment. Walton et al. (2015) demonstrated that hydrogen can be used as a processing gas to demagnetize and break down the sintered Nd-Fe-B magnets contained in HDD's into a friable alloy powder which can then be separated from the other components prior to shredding (Zakotnik et al. 2008; International Patent Application WO2012/072989; Zakotnik M. et al., 2015). At Stena's recycling plant, an amount of circa 2500 kg of HDDs is separated and collected every month. An estimation based on an average HDD weight of 544 g and an average 2.6 ± 1.5 wt% Nd-Fe-B magnet per HDD (Ueberschaar and Rotter 2015), leads to an amount of 27.5–102.5 kg of Nd-Fe-B magnets that could be recycled per month.

Laptops are manually disassembled at Stena Technoworld. Lithium ion batteries are collected separately for recycling and safety reasons, PCB and the screens are also taken out and sorted. Apart from the HDDs, Nd-Fe-B magnets were found in loudspeakers in all laptops that were disassembled in this study. The laptop loudspeakers can be easily dismantled/collected as they are present mostly beneath the laptop screens (which are removed from the laptop body) or just under the keyboard, and can easily be plucked out.

3. Results and discussions

3.1. WEEE fraction containing Nd-Fe-B magnets

From the IT and telecommunication equipment category, appliances such as computers, printers, copying equipment, and mobile phones were analyzed. Laptops, notebooks, minicomputers and notepads are some of the most valuable secondary resources as

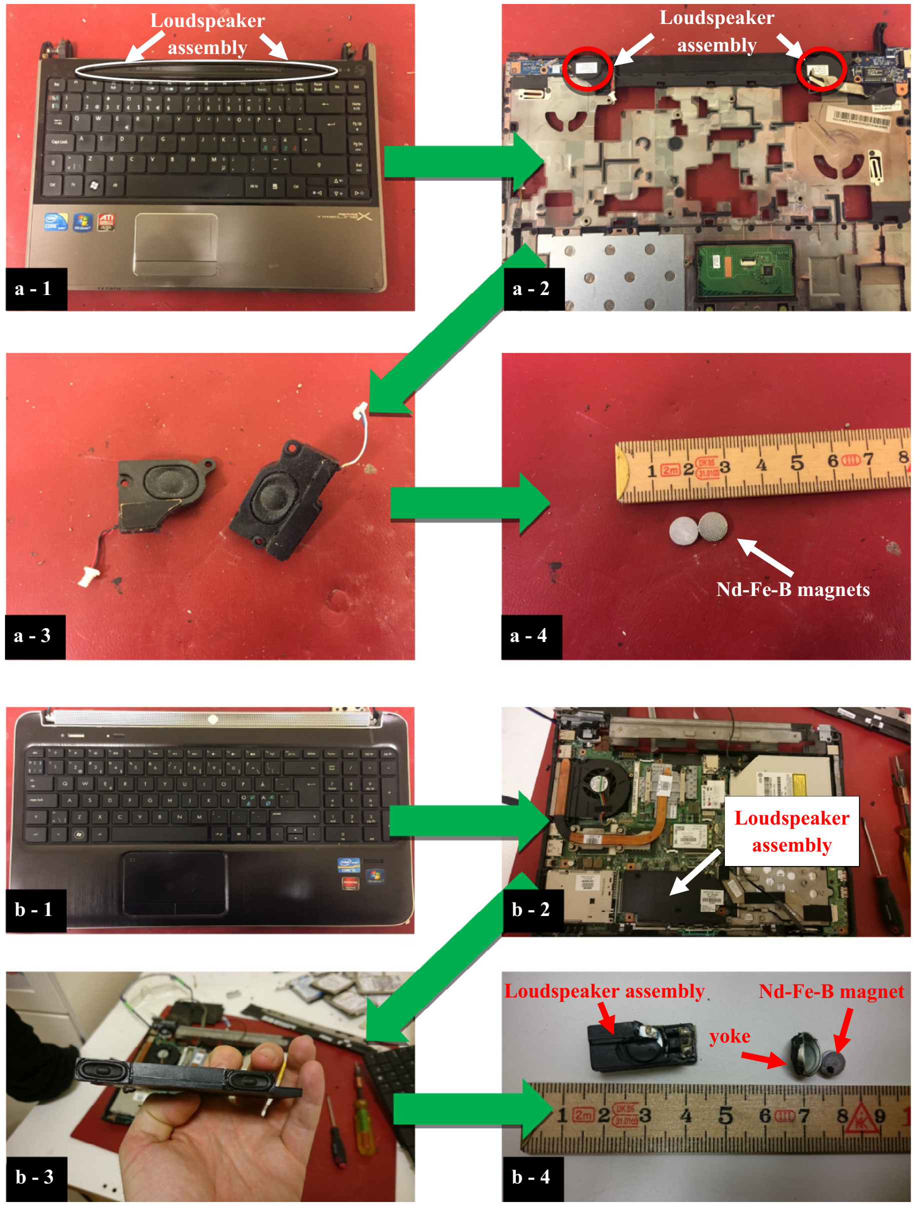

Fig. 3. Example locations of loudspeakers in laptops: **(a: 1–4)** beneath the laptop screens or **(b: 1–4)** under the keyboard.

they contain many critical metals such as gallium, indium, platinum-group metals (PGM's), cobalt and REEs.

Fig. 3 exemplifies the locations of the loudspeaker in laptops. Normally only a few screws needed to be removed before the speakers or the speaker assembly could be plucked out. The speakers were usually found inside a plastic casing that had to be broken to reach the speaker. The speaker itself most of the time contained one magnet and sometimes two magnets together with one or two discs made of iron and zinc, inside a yoke. The Nd-Fe-B magnets are coated with a Zn layer.

The laptop product market is very broad, from larger high-performance gaming laptops to small ultra-compact systems. Table 1 provides a summary of the evolution of the laptop loudspeaker magnets. The mass of the magnets represents a mean of the studied laptops that were produced in the same year.

For a more precise content of the elements which are present in the collected magnets from laptops loudspeakers, 4 magnets were selected randomly and ICP-OES measurements were performed. The chemical composition of the analyzed loudspeaker magnets is presented in the Table 2.

Table 1
Average mass of laptop loudspeakers magnets by year of production.

Year	No of studied laptops	Loudspeaker magnets mass [g]
1997	1	1.0 ± 0.05
1998	4	2.4 ± 0.05
2002	5	1.5 ± 0.05
2003	5	1.4 ± 0.05
2004	1	1.9 ± 0.05
2005	1	0.9 ± 0.05
2007	5	2.4 ± 0.05
2008	5	2.3 ± 0.05
2009	4	2.0 ± 0.05
2010	5	1.3 ± 0.05
2011	5	1.2 ± 0.05
2012	5	1.4 ± 0.05
2013	3	1.1 ± 0.05

The evolution of the Dy, Pr, and Nd over the years is presented in the Fig. 4. Our findings reflect two main trends: the Dy content of the magnets systematically decreased over the years, that is, from 1997 onwards. Dy was completely removed starting with

Table 2
Elemental compositions of the randomly selected magnets from laptops loudspeakers.

	Fe wt%	Nd wt%	Pr wt%	Dy wt%	Gd wt%	Co wt%	Nb wt%	Cu wt%	Al wt%	Ga wt%	Zn wt%	B wt%
Sample 1	64.91	22.92	7.47	0.19	0.06	0.37	0.09	0.19	0.54	0.20	0.58	0.84
Sample 2	66.43	24.18	4.42	2.75	–	0.03	0.03	0.05	0.54	–	0.86	0.94
Sample 3	62.96	24.75	8.02	0.26	0.16	0.03	0.05	0.05	0.96	–	0.55	1.03
Sample 4	65.06	21.69	6.75	3.44	–	1.34	0.02	0.19	0.59	–	0.02	1.01

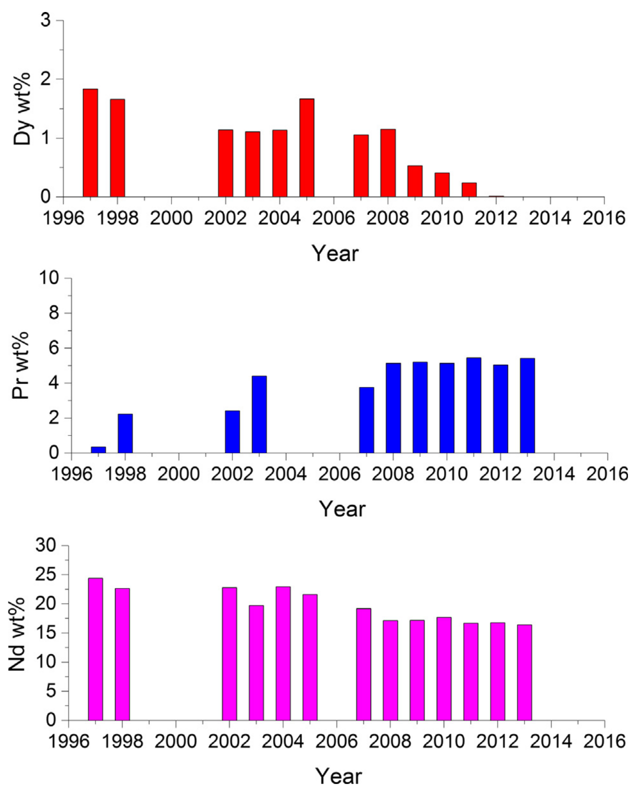

Fig. 4. Content evolution of the Dy, Pr and Nd in loudspeaker magnets collected from laptops.

2012, that is, in the very aftermath of and in reaction to the RE crisis. In fact, during the 2010/2011 crisis, particularly the price of neodymium and dysprosium increased drastically. In this time, the price of neodymium and dysprosium were approximatively two times and ten times, respectively, higher than that of praseodymium. Over this time period, also the neodymium content decreased. It can be observed also that the Pr contend is increasing over the years. This can be explained by the fact that small amount of Nd is substituted with Pr, in order to allow the use of didymium (a mixture of Nd and Pr) in the magnet manufacturing process, reducing slightly the price. The year represents the laptop release year, corresponding to the magnets from which were extracted. The content of REs in wt% represents an average of the studied magnets from laptop loudspeakers that were produced in the same year (sample size: 5 laptops for each year, except the years 1997, 2004 and 2005 with 1 laptop; 1998, 2009 with 4, and 2013 with 3 laptops – no laptops were found in the waste for the unrepresented years).

Devices of the consumer equipment category, such as audio systems, television sets, PC screens and video cameras, were also explored. In older generation TV sets based on cathode ray tubes (CRT), all the identified magnets used in their loudspeakers were ferrites.

In the case of recent TV set generations based on flat screens and flat panels, the loudspeakers were fabricated using Nd-Fe-B magnets. The technological choice for Nd-Fe-B makes sense considering the fact that those higher energy densities are required when it comes to integrating high quality speakers in a much reduced device volume of a flat screen compared to a CRT TV set. A range of different configurations was found. Some flat TVs contained only ferrites or Nd-Fe-B magnet speakers, while a majority of the units studied contained a combination of both. A typical configuration and the location of the loudspeakers in the flat TVs/PC screens are illustrated in Fig. 5.

In Fig. 6 a picture of one of the loudspeaker assemblies of the previous shown TV is presented. In this case, the TV has two loudspeaker assemblies and each assembly has two loudspeakers. Both loudspeakers contain Nd-Fe-B magnets.

The mass of the flat TV/PC screens loudspeakers magnets is shown in the following Table 3. This mass represents an average of all the magnets found in loudspeakers from flat TV/PC screens. Similar to the laptops, the magnets used in the TV/PC screen loudspeakers have compositions with Nd, Pr, and Dy as the key RE metal constituents. The Dy/Nd/Pr content is given here as an average calculated from the composition of the magnets produced in the specified year.

At Stena Technoworld, about 4.5 % of the WEEE feed (total 1000 t per month) are flat screen TVs, of which two thirds are liquid crystal display (LCD) TVs and one third are plasma or light emission diode (LED) TVs (the amount of the latter type is constantly increasing). Between 3 and 30 g of Nd-Fe-B was found per unit (average 12 g), and the size of the loudspeakers and that of the magnets was found to vary with the size of the screen.

Around 220 t of PC monitors are recycled every month, 70 t of which are flat screens and 150 t represent CRT monitors. Some flat PC screens contain loudspeakers and are found to be the ones containing exclusively Nd-Fe-B magnets (being located at the bottom of the PC screen, once the plastic casing is unscrewed, the speakers beneath the casing could be easily removed).

External loudspeakers were mainly found sorted into a stream of wood containing electronics. They required a large amount of force to break the structures and extract the speakers from the wooden boxes. More than 50 loudspeakers of various sizes were studied, and although our focus was to find speakers with Nd-Fe-B magnet, only 3 were found with the RE magnets, the rest being based on ferrites. Only two docking stations/(portable) loudspeakers were found, which is quite a new type of product where compactness is important, and the loudspeakers in both of them were based on Nd-Fe-B magnets. This was also the case for the two high-end over-ear headphones that were studied.

3.2. WEEE fraction containing non-rare earth permanent magnets

In this section, all the analyzed equipment that did not contain Nd-Fe-B magnets is presented and discussed. All studied motors that engage the fans used in microwave ovens (sample size: 8 devices), electric fans (sample size: 5 devices) and air conditioners (sample size: 5 devices) contained ferrite magnets.

A total number of 25 vacuum cleaners (appliance included in the small household category) produced by different manufactures and of different dimensions were analyzed. All these were driven by permanent magnet motors based on ferrites. In the study, also

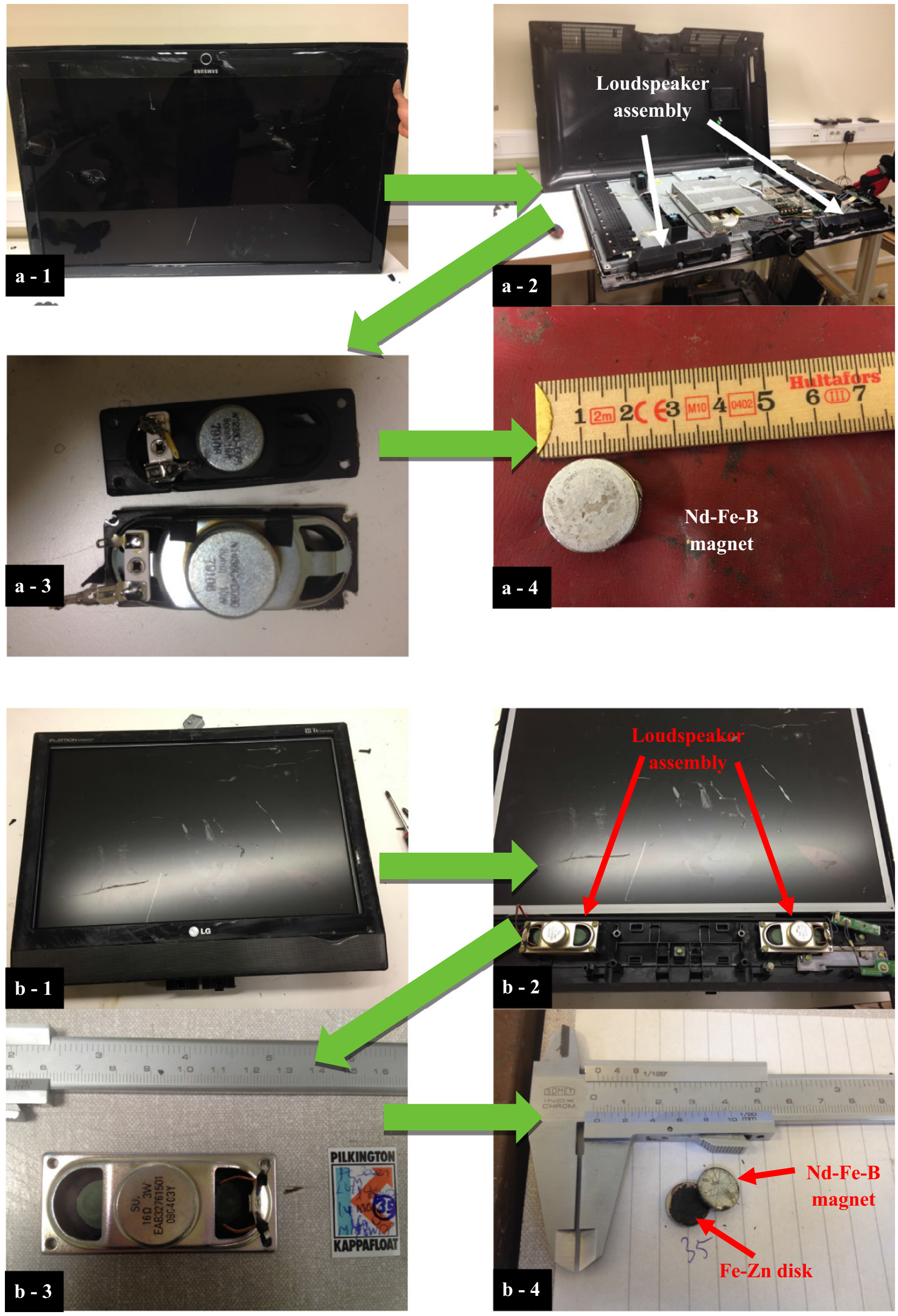

Fig. 5. Exemplification of the disassembling process and the location of the loudspeakers/Nd-Fe-B magnets for flat TV (a: 1–4) and PC screen (b: 1–4).

one vacuum cleaner from the 80′s was found and this was equipped with an induction motor. It must be noted that we provide here a snapshot of real scrap and one should consider that RE permanent magnets are generally used in the above-mentioned applications but they were not to be found in the waste streams that we studied so far.

Another part of the studied appliances that did not contain RE permanent magnets came from the IT and telecommunication category, that is, printers and copying equipment, which contains small motors with ferrite magnets. Here a total number of 15 printers were dismantled and it was found that all the small motors had ferrite magnets.

From the electrical and electronic tools category, the following appliances were studied: 12 hand drills, 5 electric saws, 16 electric screwdrivers, 3 hand sanders, 2 grass trimmers and 2 electric chainsaws. All of the appliances here were equipped with electric motors based on ferrite magnets.

From the monitoring and control instruments category and from the toys, leisure and sports equipment all the small electric motors found where equipped with ferrite magnets and also all the loudspeakers found in these categories used ferrite magnets. In these two categories it is less likely to find RE permanent magnets mainly because of the fact that these products have generally low prices.

Other EEE categories such as lighting equipment, medical devices, and automatic dispensers were not covered in the study, as they were not part of the waste streams recycled at Stena. It should also be noted that although NdFeB magnets have not been

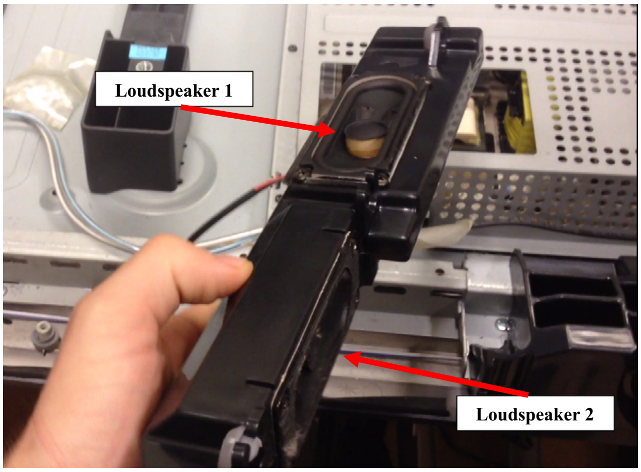

Fig. 6. Closer look of the TV loudspeaker assembly, and the location of the two loudspeakers.

found within these products at this specific site, they may be found elsewhere within the same products if a broader search is conducted.

3.3. Estimation of NdFeB magnet waste

Table 4 summarizes the possible amount of Nd-Fe-B magnets that could be obtained from loudspeaker and HDD resources at the Halmstad plant owned by Stena Technoworld in Sweden. The assumptions made were based on the data collected for the different streams and in each case, a higher and lower estimate of possible Nd-Fe-B magnet recovery is given. In the case of laptops, approximately 5 t are disassembled per month. A rough estimation assuming a laptop weight of 2.5–3.5 kg and of 1.5–2.0 g (average 1.8 g) of Nd-Fe-B magnet from loudspeakers per laptop, results that between 1.3 and 4 kg Nd-Fe-B magnet per month could be recycled. The estimation of magnets from TV is complicated as the devices were found to contain either ferrites, mixture of ferrites and Nd-Fe-B or purely Nd-Fe-B magnets in their loudspeakers. About two-thirds of flat screen TVs are considered to contain Nd-Fe-B magnets. Furthermore, with an average TV weight of 10–15 kg and an average of 10–15 g Nd-Fe-B per TV assumed, between 20 and 45 kg of Nd-Fe-B magnet can be recycled per month. Though all the PC screen magnets were found to be made of Nd-Fe-B, not all PC screens had loudspeakers. Hence, a modest assumption of 20% of PCs screens with loudspeakers is taken. The total weight of the magnets in them varies between 2 and 15 g. With an average screen weight of 4–6 kg and 4–7 g Nd-Fe-B per screen (average 5.4 g), around 7.0 and 21.0 kg of Nd-Fe-B magnet can be recovered in a month. Overall, it can be noticed that the total Nd-Fe-B magnet recovered from these various loudspeakers can be substantial and is in fact comparable to the possible magnet recovery from HDDs.

4. Conclusions

Various WEEE streams and products containing permanent magnets were studied in order to identify those that contain Nd-Fe-B magnets. Nd-Fe-B magnets were found in loudspeakers integrated into flat screen devices, where the components have to be small, and therefore the magnets have to be magnetically strong. It was observed that all the loudspeakers in the laptops studied and most of the loudspeakers in the flat TV sets and PC screens contained Nd-Fe-B magnets, whereas normal external loudspeakers contain mainly ferrites. Since LCDs and laptops are being dismantled already in the recycling process, it would be an easy task to extract the speakers from them. LEDs and plasma TVs, which are not dismantled, would require extra labor if the magnets were to be extracted.

Hard disk drives have been suggested by several projects or researchers to be the perfect candidate for Nd-Fe-B recycling. It

Table 3
Magnet mass and REs content of flat TV/PC screens loudspeakers.

Year	PC screens						TV					
	No of studied units	Loudspeaker magnets mass [g]	Dy wt%	Nd wt%	Pr wt%	REEs ∑ wt%	No of studied units	Loudspeaker magnets mass [g]	Dy wt%	Nd wt%	Pr wt%	REE ∑ wt%
2002	1	2.4	0.43	19.10	1.85	21.38	–	–	–	–	–	
2003	5	3.0	1.34	20.70	4.44	26.48	–	–	–	–	–	
2004	5	5.2	0.82	21.75	4.81	27.38	–	–	–	–	–	
2005	3	2.9	0.67	20.36	4.04	25.07	2	2.83	0.72	23.13	1.28	25.13
2006	2	5.5	0.40	19.58	4.56	24.54	1	3.60	0.54	20.57	4.72	25.83
2007	–	–	–	–	–		1	29.88	0.95	17.31	4.48	22.74
2008	1	3.7	0.70	16.48	4.69	21.87	–	–	–	–	–	
2009	–	–	–	–	–		–	–	–	–	–	
2012	–	–	–	–	–		1	12.34	0	17.95	5.36	23.31

Table 4
Possible quantities of Nd-Fe-B magnet that can be collected from different electronic waste streams at the Halmstad plant owned by Stena (for the HDDs mass it was used Ueberschaar and Rotter, 2015).

Product type	Product flows [kg/month]	Nd-Fe-B per product			Nd-Fe-B flows estimation	
		Grams			kg/month	
		Min	Average	Max	Low	High
HDDs (3.5″)	2500	6.0	14.1	22.0	27.5	102.5
Laptops	5000					
Speakers		0.6	1.8	5.3	1.3	4.0
HDDs		2.0	2.5	5.0	3.1	5.0
PC Screens	70,000	1.7	5.4	15.0	7.0	21.0
TVs	45,000	3.0	12.0	33.0	20.0	45.0

has also been suggested, that in order to recycle the Nd-Fe-B magnets, the magnet has to be removed before the shredding step, where a lot of it would be lost otherwise. If such a solution is implemented, where all the magnets are collected together, a rough estimation based on the flows at this recycling plant suggests that the amount of Nd-Fe-B could be almost doubled if also including loudspeakers from laptops and flat screens. It should also be noted that although the overall amount of the Nd-Fe-B magnet is still relatively small for this one recycling plant, there are multiple collection sites for WEEE across Europe. For an effective recycling process to be set up in Europe for Nd-Fe-B magnets then multiple streams of feedstock would have to be used for a range of devices and from a range of different companies. A detailed feasibility study is needed to determine a minimum economic scale, based on the findings of the plant data from this study.

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