



Rare earth elements and critical metal content of extracted landfilled material and potential recovery opportunities



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ABSTRACT

Rare earth elements (REEs), Platinum group metals (PGMs) and other critical metals currently attract significant interest due to the high risks of supply shortage and substantial impact on the economy. Their uses in many applications have made them present in municipal solid waste (MSW) and in commercial and industrial waste (C&I), since several industrial processes produce by-products with high content of these metals. With over 4000 landfills in the UK alone, the aim of this study was to assess the existence of these critical metals within landfills. Samples collected from four closed landfills in UK were subjected to a two-step acid digestion to extract 27 metals of interest. Concentrations across the four landfill sites were $58 \pm 6 \text{ mg kg}^{-1}$ for REEs comprising $44 \pm 8 \text{ mg kg}^{-1}$ for light REEs, $11 \pm 2 \text{ mg kg}^{-1}$ for heavy REEs and $3 \pm 1 \text{ mg kg}^{-1}$ for Scandium (Sc) and $3 \pm 1.0 \text{ mg kg}^{-1}$ of PGMs. Compared to the typical concentration in ores, these concentrations are too low to achieve a commercially viable extraction. However, content of other highly valuable metals (Al and Cu) was found in concentrations equating to a combined value across the four landfills of around \$400 million, which increases the economic viability of landfill mining. Presence of critical metals will mainly depend on the type of waste that was buried but the recovery of these metals through landfill mining is possible and is economically feasible only if additional materials (plastics, paper, metallic items and other) are also recovered for reprocessing.

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1. Introduction

The global demand for metals continues to increase and the costs of extraction are reflected in their prices since sources of high quality ores have reduced and so lower quality mines must be now exploited (DEFRA, 2012; House of Commons Science and Technology Committee, 2011). Availability and sufficiency of certain metals are causing great concerns to governments and industries (Kaartinen et al., 2013a). Several reports (British Geological Survey, 2012; DEFRA, 2010; House of Commons Science and Technology Committee, 2011; U.S. Department of Energy, 2011; European Commission, 2014) investigated the demand and supply risk of several metals including their scarcity, the political stability of the producing countries, the price volatility and the potential of substitution, trying to identify the most critical. Rare earth elements (REEs), Platinum group metals (PGMs), Lithium (Li), Indium (In), Cobalt (Co) and Antimony (Sb) were identified as high risk of supply shortage and increased impact on the economy (Hislop and Hill, 2011).

The most common use of Li, In, Co and Sb is in ceramics and glass materials, but they can also be found in permanent magnets (Co), fluorescent lamps (In and Sb), rechargeable batteries (Li and Co) and in electrical and electronic equipment (Co, In and Sb). Also, In is used in fibre optic telecommunications and alkaline batteries whereas Sb is commonly used as a flame retardant in plastics and textiles (Jorgenson and George, 2005; USGS, 2011; Sadyrbaeva, 2014; Jaskula, 2013; Butterman and Carlin, 2004). PGMs are mainly used in fuel cells, catalysts, glass and glass fibre manufacture, electrical and electronic uses and jewellery. They are essential constituents of automotive catalysts as they significantly decrease the emission of harmful compounds such as carbon monoxide, hydrocarbons and nitrogen oxides (British Geological Survey, 2009; Hagelüken, 2012; Chaston, 1982; Ayres and Peiró, 2013). A typical auto-catalyst has a concentration between 300 and $1000 \mu\text{g g}^{-1}$ of Pt, 200 and $800 \mu\text{g g}^{-1}$ of Pd and from 50 to $100 \mu\text{g g}^{-1}$ of Rh (Jimenez de Aberasturi et al., 2011).

REEs comprise the periodic group of lanthanides plus Scandium (Sc) and Yttrium (Y) and can be divided in 3 groups; Light (LREEs), Heavy (HREEs) and Sc. LREEs comprise Lanthanum (La), Cerium (Ce), Praseodymium (Pr), Neodymium (Nd) and Samarium (Sm) and the remaining are included in the HREEs. This classification is based on the chemical properties, geological availability, supply

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sources, supply demands, market values and end-markets of REEs (European Commission, 2014).

REEs are used in magnets, metallurgy, phosphors, glass and polishing, catalysts, ceramics and for nuclear and defence technologies (Hayes-Labruto et al., 2013; Meyer and Bras, 2011). World-wide, in 2010, 1300 tonnes of REEs were placed in magnets for wind turbines and 8250 tonnes in phosphors applications. Particularly for magnets applications, Nd Dy were used for electrical vehicles (EVs) (4160 tonnes of Nd and 265 tonnes of Dy), in MRI units (450 tonnes of Nd and 35 tonnes of Dy), in electrical and electronic devices (12,450 tonnes of Nd and 830 tonnes of Dy) and in wind turbines (910 tonnes of Nd and 70 tonnes of Dy). While Eu, 50 tonnes were used for phosphors applications; 24.5 tonnes in lighting, 19.5 tonnes in LCDs (Liquid-crystal display) and 6 tonnes in plasma panels (Ayres and Peiró, 2013).

All these products become a great source of critical metals when reaching end-of-life, however, reported recycling rates show poor reuse of these metals; typically between 1% and 25% Sb, <1% for REEs, In, Li and Os, and $\geq 50\%$ for Pt, Pd, Rh and Co (Graedel et al., 2011). With the Waste and Electronic Equipment (WEEE) Directive being established in 2002 (EC, 2003), it is presumed that many of the waste products containing these valuable metals were disposed in landfills prior to this date. For decades landfills have been the solution for disposing all types of waste; without any segregation they are a heterogeneous source of materials (Kaartinen et al., 2013a). For example, mobile phones contain several of these critical metals; Co, Nd, and Sb are present in quantities of $52 \mu\text{g g}^{-1}$, $1.454 \mu\text{g g}^{-1}$, $1 \mu\text{g g}^{-1}$, respectively, and between 0.05 and $0.2 \mu\text{g g}^{-1}$ of Pd and from 0.005 to $0.1 \mu\text{g g}^{-1}$ of Pt (Valero Navazo et al., 2014). The presence of REEs and other critical metals have been reported in municipal solid waste (MSW) (Morf et al., 2013), ashes from the incineration of different types of waste (Zhang et al., 2001, 2008) and in sewage sludge and food industry sludge from wastewater treatment plants (Kawasaki et al., 1998). Furthermore, landfilled industrial wastes such as metallurgical slags, bauxite residue (red mud), phosphogypsum and mine tailings are rich in REEs (Binnemans et al., 2013), although these industrial wastes are more commonly contained in mono-landfills.

Once metal-containing wastes are placed in landfill the mobility of the metals will be limited by a number of attenuating mechanisms (Jain et al., 2005). Metal recovery and extraction from landfill using leachate is hard to achieve as high pH values (>7) of the leachate decreases the mobility of metals and their tendency to leach out (Esakku et al., 2003; Ross et al., 2000; Bozkurt et al., 1999). Also, leachate anions such as sulphides and hydroxides significantly reduce the solubility of metals and induce their precipitation. Where leachate recirculation is applied the concentration of metals in the treated leachate is considerably low due to the reduction of sulphate to sulphide during waste decomposition. Approximately 0.02% of the total heavy metals in a landfill are leached out in 30 years (Reinhart and Al-Yousfi, 1996; Kjeldsen et al., 2002; Esakku et al., 2003; Gould et al., 1990; Ross et al., 2000).

Organic compounds present in landfilled waste greatly influence metals mobility. High organic matter content, as humus material, tends to increase the sorption of metals. After methane production ceases, the remaining organic substrate is harder to degrade and the microbial activity significantly decreases, resulting in the start of the humic phase. Humic substances (humic and fulvic acids) bind metals to hydroxyl and carboxyl groups, and either mobilise metals or delay their release; in aerobic conditions the humic substances lose their binding properties (Bozkurt et al., 1999; Klavins et al., 2006; Kochany and Smith, 2001; Leung and Kimaro, 1997). Almost 99.9% of metals are still found at the beginning of the humic phase (Bozkurt et al., 1999). The presence of clay in landfills (liner or daily cover) affects the adsorption of metals. Clay is usually negatively charged and in high

content increases the ion exchange with metals and induces their retention within the clay particles (Bradl et al., 2005; Moldoveanu and Papangelakis, 2012).

There are approximately >4000 open and closed landfills in the UK (Kohler and Perry, 2005) and as these critical metals are currently sourced from overseas (British Geological Survey, 2012), it is appealing to consider excavating closed landfills as they have since evolved into local “mines” of metals (Masi et al., 2014). Landfill mining (LFM) is gaining more attention due to the opportunity to source materials for energy production, recover metals, allow site remediation and land reclamation for development (Hogland et al., 2004; Jain et al., 2014; Van Der Zee et al., 2004). Furthermore, it can yield new business opportunities and decrease dependency on metal exports from the producing countries (e.g. China for REEs and Sb) (Kaartinen et al., 2013a; Wübbeke, 2013; British Geological Survey, 2012).

Previous studies (Masi et al., 2014; Kaartinen et al., 2013a; Hogland et al., 2004; Quaghebeur et al., 2013; Kaartinen et al., 2013b; Spooren et al., 2012; Prechthai et al., 2008) have assessed the type of wastes that can be recovered through LFM for reuse. However they mainly focused on raw materials like paper, glass, plastics, textiles, wood, metallic items and the presence of heavy metals. Conventional LFM is the process of recovering materials contained within a landfill mainly with the purpose of remediating the site (Krook et al., 2012), whereas the concept of enhanced landfill mining [ELFM] targets resource recovery by combining integrated and innovative valorisation techniques for material and energy recovery (Jones et al., 2013). Furthermore in-situ, as part of an ELFM approach, is an interesting option for recovering critical metals from specific landfilled waste streams, such as industrial process residues (Binnemans et al., 2013). Therefore, the purpose of this study was to analyse the presence of REEs and critical metals in the landfill mass (involving mixture of cover material, degraded organics and other inorganic materials) within the closed landfills of the UK to assess the availability of these metals and feasibility of recovery regarding the potential economic impact. A possible relationship between the concentration of metals with the organic content of landfilled material and with the depth at which waste was buried in the landfill site was also investigated.

2. Materials and methods

Fifty-five samples were collected from four closed landfill sites (LFS) in the UK. Different cores were drilled vertically across the landfill area and samples were taken at different depths by a specialised private company hired by the LFS operator. An overview of the waste samples collected at the four LFS is provided in Table 1.

The samples were subjected to manual sieving and sorting for removal of plastic, metal, paper, textile, glass and other materials larger than 19 mm. The remaining fraction, the organic-like material, was dried at 105 °C overnight and ground to a size of ≤ 1.5 mm (Retsch SM 2000 grinder).

To achieve the highest quantity of the extracted metals, samples were submitted to nitric acid digestion followed by a digestion with aqua regia ($\text{HNO}_3 + \text{HCl}$ in a ratio of 1:3) (James, 2011), as it was not technically possible to use HF to digest the samples. Briefly, for the nitric acid digestion, 0.5 g of sample was mixed with 8 ml of HNO_3 (70%, 1.42 density, Fisher Scientific) in Teflon tubes and left overnight. The samples were placed in a microwave digester (Multiwave 3000, Anton Paar) for 45 min at 800 W. The mixtures were then filtered (150 mm, hardened low ash paper, Fisher Scientific) and the remaining liquid was diluted to 100 ml with distilled water and retained for analysis. The filter paper and residue was then dried for 2 h at 105 °C and placed into a digestion tube with 6 ml of HCl (37%, 1.18 density, Fisher

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