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Concentration of precious metals during their recovery from electronic waste

R. Cayumil^a, R. Khanna^{a,*}, R. Rajarao^a, P.S. Mukherjee^b, V. Sahajwalla^a

^a Centre for Sustainable Materials Research and Technology (SMaRT), School of Materials Science and Engineering, The University of New South Wales, Sydney, NSW 2052, Australia

^b Advanced Materials Technology Department, CSIR-Institute of Minerals and Materials Technology, Bhubaneswar 751013, India

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ABSTRACT

The rapid growth of electronic devices, their subsequent obsolescence and disposal has resulted in electronic waste (e-waste) being one of the fastest increasing waste streams worldwide. The main component of e-waste is printed circuit boards (PCBs), which contain substantial quantities of precious metals in concentrations significantly higher than those typically found in corresponding ores. The high value and limited reserves of minerals containing these metals makes urban mining of precious metals very attractive. This article is focused on the concentration and recovery of precious metals during pyro-metallurgical recycling of waste PCBs. High temperature pyrolysis was carried out for ten minutes in a horizontal tube furnace in the temperature range 800–1350 °C under Argon gas flowing at 1 L/min. These temperatures were chosen to lie below and above the melting point (1084.87 °C) of copper, the main metal in PCBs, to study the influence of its physical state on the recovery of precious metals. The heat treatment of waste PCBs resulted in two different types of solid products, namely a carbonaceous non-metallic fraction (NMFs) and metallic products, composed of copper rich foils and/or droplets and tin–lead rich droplets and some wires. Significant proportions of Ag, Au, Pd and Pt were found concentrated within two types of metallic phases, with very limited quantities retained by the NMFs. This process was successful in concentrating several precious metals such as Ag, Au, Pd and Pt in a small volume fraction, and reduced volumes for further processing/refinement by up to 75%. The amounts of secondary wastes produced were also minimised to a great extent. The generation of precious metals rich metallic phases demonstrates high temperature pyrolysis as a viable approach towards the recovery of precious metals from e-waste.

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1. Introduction

The manufacturing of electronic and electrical equipment (EEE) and the associated generation of electronic waste (e-waste) is experiencing a rapid growth due to the high consumption of electronic devices and their early obsolescence throughout the world. The lifespan of most of EEE is considerably shorter now than a decade ago. The emergence of electronics industry has impacted the environment in two different ways: large volumes of e-waste being disposed/or landfilled (this waste stream is increasing by 4–5% every year (Baldé et al., 2015)), and the increasing demand for rare and precious metals from primary resources (Veit and Bernardes, 2015). The disposal of e-waste has several health issues, such as the release of toxic and hazardous compounds, contamination of water, soil and air, and impacts on regions surrounding recycling

facilities. As a number of countries lack regulations regarding e-waste management, especially developing regions of the third world and Africa, operations in informal sectors extract materials from e-waste in an inappropriate manner using methods such as open burning, dumping and digestion in acids. The use of these inappropriate techniques generates significant amounts of secondary wastes and releases pollutants, dioxins and furans which have deleterious impact on the health of people near the recycling areas. There is an urgent need to develop environmentally sustainable approaches towards recycling e-waste and to reduce the amounts of wastes being trashed or landfilled. It is also desirable to prolong the life spans of precious materials in a range of applications and to minimise their loss and to reduce the amount of natural resources being used to manufacture EEE (Veit and Bernardes, 2015).

E-waste is a heterogeneous mixture of materials; its composition is evolving continuously due to rapid upgrades in functions and design of electronic products (Chancerel et al., 2009). Printed

* Corresponding author.

E-mail address: ritakhanna@unsw.edu.au (R. Khanna).

circuit boards (PCBs), the key central components of electronic equipment, are generally very complex in nature. Several precious metals are present in PCBs in much higher concentrations than typically found in corresponding ores. With appropriate recycling, e-waste could prove to be an important secondary resource of precious metals (Hagelüken, 2006; Teller, 2006). The electronics industry uses significant amounts of precious metals mainly due to their high chemical stability, corrosion resistance and electrical conductivity. These metals are frequently used as contact materials, for electroplating and/or as connectors. Specifically, silver is used in contacts, switches and solders; gold as a bonding wire, contacts and in integrated circuits; palladium in multilayer capacitors and connectors; and platinum in hard disks, for proton exchange membrane fuel cells and in thermocouples. These are also found as plated gold, palladium on copper laminates and plated silver/gold on nickel or iron (Cui and Roven, 2011; Kogan, 2006; Schluep et al., 2009).

Precious and other metal resources for the manufacture of EEE are mainly obtained from their respective primary production routes. The extraction of these metals through mining is associated with several negative features such as extensive land, energy and water usage, gaseous emissions such as SO₂ and CO₂ and the generation of large amounts of secondary solid and liquid wastes. The environmental footprint of the primary production of these metals is high, especially when the concentration of valuable components is quite low and requires the removal of large volumes of material involving several steps of treatment. Primary production is also associated with high levels of greenhouse gas emissions. For example, 1 tonne of gold produced through mining emits ~17,000 tonnes of CO₂, while palladium and platinum respectively produce ~10,000 and ~14,000 tonnes of CO₂ (Chancerel et al., 2009; Schluep et al., 2009). The demand for precious metals for use in electrical and electronic equipment has increased rapidly due to a significant increase in the number of electronic products being manufactured (Schluep et al., 2009). The annual consumption of silver in the electronics industry is currently estimated to be ~7554 tonne (34% of total primary production); the corresponding estimates for Au, Pd and Pt are 327, 44 and 7 tonnes/year are ~13%, 19% and 4% of the total primary production. The cost of these precious metals was estimated to be ~18.9 billion dollars (Reuter et al., 2013; USGS, 2010).

Environmentally sustainable recovery of precious metals from e-waste is also expected to have a significant economic impact. In waste TV boards and DVD players, the typical value of copper and precious metals amounts to ~40% of the total value of the entire device. In spite of the small amounts used, the estimated value of these metals in waste mobile phones and printed circuit boards can be up to 80% of the total worth (Hagelüken and Corti, 2010; Park and Fray, 2009). While the usage of precious metals in EEE has reduced to some extent in the manufacture of new devices, recent hikes in the price of these metals makes their recycling still very appealing. A rough estimate of precious metals present in mobile phones was determined as follows: One tonne of mobile phones without batteries contains 130 kg Cu, 3.5 kg Ag, 340 g Au and 140 g Pd (Schluep et al., 2009). If all mobile phones discarded in 2008 were recycled, a material flow of 1250 tonne Cu, 13 tonne Ag, 3 tonne Au and 2 tonne Pd would be recovered, representing a market value of USD 105 million (Yu et al., 2010).

In addition to economic incentives, several other benefits are associated with the recovery of precious metals from electronic waste. These include a reduction in the wastes being landfilled, resource utilisation, minimisation of gaseous emissions and toxic compounds generated. There are also potentially high savings in energy consumption. Recycling of gold can reduce energy usage by up to 65% as compared with primary production; corresponding energy savings for Pd were estimated to be 14% (Wang and

Gaustad, 2012). The relative concentration of precious metals in printed circuit boards (PCBs) is also much higher than the corresponding concentration in their ores, e.g., the concentration of silver, gold and palladium in minerals is less than 10 g/tonne, whereas in computer printed circuit boards, the typical average is 1000, 250 and 110 g/tonne PCB respectively (Hagelüken, 2006).

In this article, we report a novel approach to significantly concentrate precious metals present in PCB waste, an approach that is cost-effective and will produce minimal secondary waste. An in-depth investigation was carried out on the recovery of Ag, Au, Pd and Pt during the recycling of waste PCBs through high temperature pyrolysis under Ar atmosphere. The influence of copper, the main metal in PCBs, on the concentration of precious metals was investigated in the temperature range of 800–1350 °C. The physical state of copper, whether solid state or molten state, was one of the key experimental parameters. Precious metals recovered from this process can be refined further or be used as a feedstock in other metallurgical recovery approaches. The main focus of this research was on maximising the recovery of precious and other metals from e-waste while causing minimal environmental damage. The expected outcomes will result in to a significant reduction in the amount of wastes being landfilled or processed inappropriately and will also help develop safe practices for managing electronic waste.

2. Previous studies

Several metallurgical and chemical approaches have been used to recover precious metals from e-waste. Some well-known industrial pyro-metallurgical processes that recover these metals are Noranda, Rönnskar and Umicore. In the Noranda process, copper concentrates and e-waste (up to 14 wt.%) are melted at 1250 °C under an oxidising atmosphere. The resulting metallic fraction, composed of Cu and precious metals, is transferred to the conversion process, producing Cu blister with 99.1% purity. The remaining constituents include several precious metals, which can be recovered in subsequent electro-refining (Veldhuizen and Sippel, 1994). The input for Umicore process includes several types of wastes including up to 10 wt.% e-waste. The process is quite similar to Noranda in other aspects with leaching and electrowinning being carried out after conversion. From the resulting sludge, Ag, Au, Pt, Pd, Rh, Ir and Ru can be recovered with up to 99.9% purity. In the Rönnskar process, metal rich e-waste is recycled along with Cu concentrates, whereas the poor quality e-waste is recycled along with Pb concentrates. Black Cu (slag produced in the Pb process line) is later transferred to copper conversion. The product is then electrolytically refined; the generated slime contained Ag, Au and Pt-group metals in a granulated form (Bigum et al., 2012; Cui and Zhang, 2008; Schluep et al., 2009).

A few studies have been reported on the extraction of precious metals from PCBs through pyro-metallurgy. Zhou et al. (2007) heated a mixture of PCBs and 12 wt.% NaOH at 1200 °C. Complexes containing up to 99% Cu, Au and Ag were recovered in the metallic fraction; some slag was also generated as a secondary by-product. Flandinet et al. (2012) mixed molten KOH and NaOH with crushed PCBs and heated at 300 °C under 2 L/h of Ar. The generated metallic fraction (25 wt.%) contained up to 287, 0.959, 0.725 and 0.238 g/kg of Cu, Ni, Au and Ag, respectively. A brown powder containing fluorides, chlorides and bromides was produced (75 wt.%) as a bi-product; some volatile compounds were also released in ppm range.

A number of investigations have been reported on the use of hydrometallurgical techniques to recover precious metals from waste PCBs. Li et al. (2010) dissolved mixed waste PCBs in HNO₃; the leachate was then mixed with a lime-sulphur-synthetic solution (CaS_x/CaS₂O₃, CuSO₄, aqueous ammonia and sodium

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