



# Mineral phases and metals in baghouse dust from secondary aluminum production



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## HIGHLIGHTS

- Mineral phases, metals content & their leachability from baghouse dust were studied.
- High variability of metallic Al and total Al content in baghouse dust was observed.
- Leachability of heavy metals of baghouse dust was higher than that of salt cake.
- Not all BHD samples were below the TCLP limits in US.

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## ABSTRACT

Baghouse dust (BHD) is a solid waste generated by air pollution control systems during secondary aluminum processing (SAP). Management and disposal of BHD can be challenging in the U.S. and elsewhere. In this study, the mineral phases, metal content and metal leachability of 78 BHD samples collected from 13 different SAP facilities across the U.S. were investigated. The XRD semi-quantitative analysis of BHD samples suggests the presence of metallic aluminum, aluminum oxide, aluminum nitride and its oxides, spinel, elpasolite as well as diaspora. BHD also contains halite, sylvite and fluorite, which are used as fluxes in SAP activities. Total aluminum (Al) in the BHD samples averaged 18% by weight. Elevated concentrations of trace metals ( $>100 \mu\text{g L}^{-1}$  As;  $>1000 \mu\text{g L}^{-1}$  Cu, Mn, Se, Pb, Mn and Zn) were also detected in the leachate. The U.S. toxicity characteristic leaching procedure (TCLP) results showed that some samples leached above the toxicity limit for Cd, Pb and Se. Exceeding the TCLP limits in all sample is independent of facilities generating the BHD. From the metal content perspective only, it appears that BHD has a higher potential to exhibit toxicity characteristics than salt cake (the largest waste stream generated by SAP facilities).

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## 1. Introduction

Recycling through the process of secondary aluminum production (SAP) plays an important role in aluminum manufacturing (IAI, 2009; TAA, 2010). While salt cake (SC) is consisting of the primary residue from SAP activities (Huang et al., 2014), the processes also generate baghouse dust (BHD), which is a powdery waste of a very fine grain size captured in dry emissions control devices called baghouses. Baghouses are used in SAP to control particulate air emissions from furnace operation and other SAP processing activities (López et al., 2001; López-Delgado et al., 2007). In general, the

formation of BHD and the amount of BHD formed depends on several factors such as the type and quality of input material (e.g. aluminum scraps), the operating conditions, and the control technology applied (Peterson and Newton, 2002; Hwang et al., 2006; Schlesinger, 2007; Schmitz, 2007).

BHD is the second largest solid waste generated in SAP. Viland estimated in a 1990 study that for every one ton of scrap aluminum processed, 760 kg of secondary aluminum, 240 kg of dross residues and 3 kg of BHD are generated (Viland, 1990). López-Delgado et al. (2007) estimated that in Western Europe, approximately 13 kg of BHD are generated per ton of scrap aluminum recycled (López-Delgado et al., 2007). A 2013 document from TAA indicates that the BHD generation rate in Northern America is about 6.8 kg per ton of the aluminum scrap recycled (TAA, 2013). The total combined annual amount of these aluminum

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wastes (SC and BHD) from SAP is around one million tons in the U.S. (USDOE, 1999), which is 20% of all the SAP-related waste generated in the world (Azom, 2003). Depending on their constituencies, the end-of-life management of these wastes has been found to present challenges in the U.S. and elsewhere (Reuter et al., 2004; Gil, 2005; Lorber and Antrekowitsch, 2010; SINTEF, 2010; Huang et al., 2012).

BHD is usually co-disposed in municipal solid waste (MSW) landfills. It has been reported that many operational issues of landfill occurred after disposal aluminum solid wastes, including BHD. High concentration of hydrogen ( $H_2$ ) (30–50%) with gaseous ammonia ( $NH_3$ ) (up to 15000 ppmv) were detected (Gerbasi, 2006; Allen et al., 2009; Stark et al., 2012). An increase in landfill temperature, 60 to 93 °C over a period of several months to several years (Gerbasi, 2006; Allen et al., 2009) were also observed in a MSW landfill after disposal of these aluminum wastes. Above average concentration of  $F^-$ ,  $Cl^-$ ,  $NH_4^+$ ,  $CN^-$ , high pH and electric conductivity value in soil, leachate or groundwater, as well as the potential contaminations of heavy metals were also documented (USEPA, 1995, 2008; Gerbasi, 2006; Swackhamer, 2006; OhioEPA, 2007; Allen et al., 2009; Lorber and Antrekowitsch, 2010). It was known that MSW landfills are anaerobic systems that decompose the organic fraction of solid wastes with the temperature between 25 and 60 °C depending on the waste characteristics and location of landfill (Yeşiller et al., 2005; Hanson et al., 2010). On the other hand, BHD is recognized as a hazardous waste in European Union countries (European-Commission, 2000) because it is considered to be “highly flammable” (Category H3-A) and an “irritant” (Category H4) (European-Commission, 1991). When BHD comes in contact with water or damp air, highly flammable gases form, and these gases can be explosive, as well as act as irritants to skin and mucous membranes. Furthermore, BHD has been found to be harmful if inhaled or ingested (Category H5) (European-Commission, 1991). BHD is also in the category of substances that are capable, after disposal (landfill or other), of potentially yielding another substance (e.g. leachate), which can possess any of the characteristics associated with the solid BHD or gaseous products (Category H13) (European-Commission, 1991). It is believed that the reactivity of aluminum wastes, including BHD is related to the composition and mineral phases of wastes (López-Delgado et al., 2007; Lorber and Antrekowitsch, 2010; SINTEF, 2010; Huang et al., 2012; Stark et al., 2012; Tsakiridis, 2012).

When compared with SC, there is much less data available on the characteristics of BHD from SAP (López et al., 2001, 2004; Reuter et al., 2004; López-Delgado et al., 2007; Lorber and Antrekowitsch, 2010; Huang et al., 2011, 2012, 2014; Tsakiridis, 2012), although it has been reported that BHD contains 25–40% total Al, 15–25% metallic Al, 1–3% C, 0.2–1% S, 1–6% N, 6–11%  $SiO_2$ , 1–3% Ca, 2–5% Mg, 1–3% Na, 0.2–1% K, 0.5–2% Fe and 1–5% F (López-Delgado et al., 2007, 2009), and also contains aluminum nitrides, carbides and sulfides, as well as metal oxides derived from the particular alloys being processed (López et al., 2001, 2004; López-Delgado et al., 2007, 2009; Schlesinger, 2007; Schmitz, 2007). Thus, the objective of this study was to investigate BHD from SAP facilities in the U.S. by determining the mineral phases and the metal (Al, As, Ca, Cd, Cr, Cu, Fe, K, Mg, Mn, Na, Pb, Se and Zn) content of the samples. The study also examines the leachability of metals from BHD exposed to deionized water in an anaerobic and elevated temperature environment (Huang et al., 2014), which is designed to simulate key conditions in municipal solid waste (MSW) landfills. The U.S. Environmental Protection Agency (EPA) toxicity characteristic leaching procedure (TCLP) (USEPA, 1992) was also employed on these BHD samples. The information resulting from this study will help provide the scientific foundation to understand BHD waste material, its potential risk and strategies for its management.

## 2. Experimental

### 2.1. BHD sampling

A total of 78 BHD samples were collected over a period of four months from 13 different SAP facilities in the U.S. Prior to sample collection, the BHD was stored at the generation site. After ensuring sufficient cooling of the BHD, a subsample was collected following ASTM Method C702-98, “Standard practice for reducing samples of aggregate to testing size” (ASTM, 2003). Upon receipt at the EPA laboratory, each sample was mixed in an acid rinsed stainless steel pan. Samples did not require size reduction since they passed through a 2 mm sieve as received.

### 2.2. Mineral phases analysis

Because of analysis time constraints (16 h per sample), 44 BHD samples were randomly selected for XRD analysis from the original 78 collected. The samples' mineral phases were evaluated from 5° to 110°  $2\theta$  on a Philips X'Pert Pro Diffractometer using copper  $K\alpha$  radiation. The powder diffraction file (PDF) patterns database from the International Centre for Diffraction Data (ICDD) was employed for the search, match and identification. A subset of reference patterns was built for all studied BHD samples. The semi-quantitative phase analysis was used by the X'Pert HighScore Plus software, based on the CHUNG Normalized RIR Method (Chung, 1974). An example of XRD analysis processing and results were presented as supporting documents (Tables SI-1–3, and Fig. SI-1).

### 2.3. Total metal analysis

Extractable metal content was evaluated in all 78 BHD samples collected. After homogenization, 0.1 g subsample of BHD was acid digested following U.S. EPA SW846 Method 3051A with minor modifications due to the samples' high aluminum content (USEPA, 2007a). The details of digestion and metal recovery can be found in the supporting information. After acid digestion, metal compositions including Al, As, Ca, Cd, Cr, Cu, Fe, K, Mg, Mn, Na, Pb, S, Se and Zn were determined following EPA's SW846 Method 6010C using a Thermo ICP-AES (USEPA, 2007b).

### 2.4. Metal leachability

The same 44 samples that were randomly selected for the XRD analysis were targeted for the metal leachability evaluation [3]. Ten grams of the BHD sample (<2 mm) was placed into a 500 ml sealed-reaction vessel and then purged with argon for ten minutes. Then, 100 ml of argon-purged pre-heated deionized water (DIW) at 50 °C was added, and the reaction vessels were incubated for seven days at 50 °C in an insulated incubator. The temperature (50 °C) was used to simulate the high range of temperature that can be found in municipal solid waste landfill sites (Klein et al., 2001; Hanson, 2005; Hanson et al., 2010). In general, most BHD samples have low reactivity, even without temperature increase (an index of reactivity) at room temperature, whereas have high reactivity (e.g. temperature increase and gas generation) at 50 °C. It is important to investigate the metal's leachability after aluminum wastes reacted with water to understand these material's metal behavior in landfill environment (Huang et al., 2014). Leachate samples were then collected and preserved with concentrated  $HNO_3$  (final pH < 2.0) after filtration using 0.45  $\mu m$  filters. Leachable elements (Al, As, Ca, Cd, Cr, Cu, Fe, Mg, Mn, Na, K, Pb, S, Se and Zn) were analyzed using the Thermo ICP-AES with EPA Method 6010C (USEPA, 2007b). ANOVA analysis indicated that there were no statistical

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