



Leaching behaviour of co-disposed steel making wastes: Effects of aeration on leachate chemistry and vanadium mobilisation



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ABSTRACT

Steelmaking wastes stored in landfill, such as slag and spent refractory liners, are often enriched in toxic trace metals (including V). These may become mobile in highly alkaline leachate generated during weathering. Fresh steelmaking waste was characterised using XRD, XRF, and SEM-EDX. Batch leaching tests were performed under aerated, air-excluded and acidified conditions to determine the impact of atmospheric CO₂ and acid addition on leachate chemistry. Phases commonly associated with slag including dicalcium silicate, dicalcium aluminoferrite, a wüstite-like solid solution and free lime were identified, as well as a second group of phases including periclase, corundum and graphite which are representative of refractory liners. During air-excluded leaching, dissolution of free lime and dicalcium silicate results in a high pH, high Ca leachate in which the V concentration is low due to the constraint imposed by Ca₃(VO₄)₂ solubility limits. Under aerated conditions, carbonation lowers the leachate pH and provides a sink for aqueous Ca, allowing higher concentrations of V to accumulate. Below pH 10, leachate is dominated by periclase dissolution and secondary phases including monohydrocalcite and dolomite are precipitated. Storage of waste under saturated conditions that exclude atmospheric CO₂ would therefore provide the optimal environment to minimise V leaching during weathering.

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1. Introduction

Steel slag is a ubiquitous byproduct of the steelmaking industry that is produced in large quantities worldwide. It is estimated that 160–240 million tonnes of steel slag were produced in 2016 (Ober, 2017) which corresponds to approximately 10–15% of crude steel output (Piatak et al., 2014). Primary steelmaking (i.e. conversion of iron to steel) produces steel slag via two principal production methods; (1) basic oxygen furnace (BOF) steelmaking in which molten iron from a blast furnace is used and (2) electric arc furnace (EAF) steelmaking which uses a combination of scrap steel, directly reduced iron and pig iron. In both processes lime (or limestone) is added to the furnace as a fluxing agent to remove impurities from the molten metal (Eloneva et al., 2010; Piatak et al., 2014). The composition of BOF and EAF slag is broadly similar and consistent across location and process (Tossavainen et al., 2007; Yildirim and Prezzi, 2011; Proctor et al., 2000). These predominantly consist of

Ca, Mg, Fe and Al oxides and silicates (Proctor et al., 2000); the relative proportions of which will vary according to the raw materials used during manufacture. Secondary steelmaking slags are formed during secondary steelmaking where both BOF and EAF derived steels are further processed in ladle furnaces, producing BOF(L) or EAF(L) slags respectively. These are much more variable in composition and are also relatively enriched in Mg and Al due to additives used in the process (Shi, 2002). In addition to slag production, a wide variety of refractories (MgO-C; Al-silicate; MgO-Al₂O₃-C) are used as furnace liners during steelmaking to protect the furnace (Quaranta et al., 2014). Refractories that are in contact with molten slag wear over time and the build-up of solidified slag above the melt (due to sputtering) and in conduits also require regular removal. Therefore, periodic renewal of the entire liner is required. The result is a mixed waste containing both slag and refractories which is difficult to separate and are often co-disposed (Hanagiri et al., 2008).

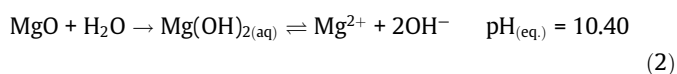
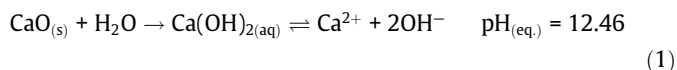
Primary steelmaking slags are recycled where possible, usually as aggregate in civil engineering applications, such as road construction and as a general fill material due to its stability (Geiseler, 1996;

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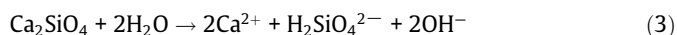
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Yi et al., 2012). However, in some cases elevated concentrations of free lime (CaO) and periclase (MgO), which expand on hydration, preclude reuse in engineering applications. Ladle slags and refractories can also be recycled during primary steelmaking (as an alternative source of CaO or MgO flux) but virgin materials are often preferred due to their more uniform composition and the increased effort that would be required in slag sorting and processing (Kwong and Bennett, 2002; de Sa et al., 2007). For this reason, and also because supply frequently exceeds the demand for secondary aggregates, steelmaking byproducts are often stored either in landfill or in open 'heaps'. However, as recycling rates increase, materials with problematic properties (e.g. high metal content, high % of CaO or MgO, or simply uncertain or variable composition) will make up an ever greater proportion of materials stored in landfill. It is therefore increasingly important to understand the leaching behaviour of such non-standard by-products as they become a significant part of the disposed inventory.

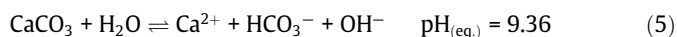
Steel slags contain free lime (CaO) and periclase (MgO; also in present in refractories) that hydrate to form portlandite (Ca(OH)₂) or brucite (Mg(OH)₂). These phases dissolve in water to generate high pH (10.5–12.5) leachate (Mayes et al., 2008):



Alkalinity may also be produced by the dissolution of Ca-silicates, (e.g. β-dicalcium silicate, larnite; Roadcap et al. (2005)):



In addition, under aerated conditions, reactions involving the in-gassing of CO₂ results in conversion of Ca/Mg hydroxide based alkalinity to carbonate alkalinity.



Whilst steel slag has historically been considered to be a non-hazardous waste, concerns have been raised in recent years regarding the high concentrations of potential toxic trace metal(loid)s (including Al, Cr, Pb, Mo and V) present in the slag (Tossavainen et al., 2007; Proctor et al., 2000; Matern et al., 2013) which may be mobilised in the alkaline leachate (Cornelis et al., 2008). V leaching in particular has received a lot of recent attention (Chaurand et al., 2006; De Windt et al., 2011; Huijgen and Comans, 2006; Navarro et al., 2010) due to its relative enrichment in steel slags and regulatory concern over high V concentrations in leachates (Environment Agency, 2014). Steelmaking wastes at disposal sites are often deposited in both saturated and unsaturated settings, however, few data currently exist concerning slag leachate generation and chemistry under aerated conditions (Bayless and Schulz, 2003; Roadcap et al., 2005; Mayes and Younger, 2006). Under aerated conditions, CaCO₃ precipitation results in a drop in solution pH as OH⁻ ions are consumed (Eq. (4)). Changes in both pH and redox have significant implications for the mobility of many potentially toxic metals; for example Al, Cr, and V mobility and toxicity are all highly dependent on their speciation (with higher oxidation states generally regarded as more toxic and mobile) (Pourbaix, 1966).

This study investigates leaching behaviour of co-disposed steelmaking waste under aerated and air-excluded conditions. These represent contrasting conditions present either near the surface (good contact with atmosphere) and below the water table deeper within waste heaps. The waste has been characterised using X-ray

diffraction (XRD) and fluorescence (XRF) techniques, as well as scanning electron microscopy (SEM) with energy-dispersive X-ray spectroscopy (EDX) to determine the mineral phases present. Fully aerated and air-excluded water leaching tests have been performed to determine how phase dissolution behaviour, secondary mineral formation and trace metal release are affected by changes in pH and availability of air. Results will assist prediction of metal release from waste when stored in landfill above and below the water table, enabling effective environmental risk assessment and cost-effective long-term management of the waste.

2. Methods and materials

2.1. Sample collection and characterisation

Samples were collected within one week of deposition from the Yarborough Landfill (British Steel, Scunthorpe, UK) in May 2013 (LAT 53°35'22.24" LONG 0°35'41.52"). The sample consisted of 50–500 g blocks (~100 kg total). A sub-sample of the collected material (approx. 500 g consisting of 50 g pieces) were brushed to remove any fines and crushed to provide a homogenised powder consisting of 20–100 μm particles. The crushed waste was stored in a polythene bag within an airtight glass jar containing soda lime to prevent weathering due to contact with atmospheric CO₂ and moisture.

Elemental analysis of the powdered waste was undertaken using a PANalytical Axios Advanced X-ray Fluorescence (XRF) spectrometer (data corrected for loss on ignition at 1050 °C). Samples were prepared for major element analysis as fused beads with lithium metaborate/tetraborate flux (Johnson Matthey Spectroflux JM100B) (0.6 g sample; 3 g flux). For minor/trace element analysis pressed pellets were prepared containing ~10 g of dried waste using ~10–20 drops of 6.6% w/v polyvinyl alcohol in a 1:6 mix of methanol and distilled deionized water as a binder (Moviol 88 solution). The elemental limit of detection (LoD) was generally <0.02 wt% and the analytical uncertainty (versus certified reference standards) was <±10% of the data value. Mineralogical analysis (LoD was approximately 3 wt% for crystalline phases) was undertaken by powder X-ray diffraction (XRD) using a Bruker D8 diffractometer, where powder samples were mounted on silicon slides and scanned between 2° and 70° 2θ using Cu K_α radiation. Diffraction peaks from unknown samples were then matched to known standard reference patterns using Diffrac.Suite Eva v3.0 software using the International Centre for Diffraction Data (ICDD) PDF2 database (powder diffraction file (PDF) reference numbers are reported for identified phases).

Two polished blocks were prepared by first cutting waste pieces to size under water and setting the resultant ~2 cm³ blocks into epoxy resin with the cut surface exposed. This surface was then polished using a water-free diamond paste to remove the top 1–2 mm of material potentially exposed to water during cutting. Electron micrographs were subsequently collected on a FEI QUANTA 650 FEG ESEM, which was equipped for Oxford Instruments INCA 350 energy-dispersive X-ray spectroscopy (EDS) system/80 mm X-Max silicon drift detector. EDS spectra and elemental maps were collected and analysed using Oxford Instruments AZtec software. The element and sample specific LoD for EDS analysis was between 0.1 and 0.5 wt%.

2.2. Acid neutralisation capacity (ANC)

Homogenised powdered waste (0.4 g) was mixed with 40 mL HCl with concentrations ranging from 1 M to 0.001 M in 50 mL polypropylene Oak Ridge tubes (Nalgene, USA). Experiments were performed in triplicate. Headspaces of the 50 mL tubes were

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