Journal of Environmental Management 222 (2018) 325–337

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Weathering of historical copper slags in dynamic experimental system with rhizosphere-like organic acids

Anna Potysz[a,*], Jakub Kierczak[a], Malgorzata Grybos[b], Artur Pędziwiatr[c,d], Eric D. van Hullebusch[e]

[a] University of Wroclaw, Institute of Geological Sciences, Cybulskiego 30, 50-205, Wrocław, Poland
[b] Université de Limoges, PEIRENE – EA 4330 - Faculté des Sciences et Techniques, 123 Avenue A. Thomas, 87060, Limoges, Cedex, France
[c] Wrocław University of Environmental and Life Sciences, Institute of Soil Sciences and Environmental Protection, Grunwaldzka 53, 50-357, Wrocław, Poland
[d] Warsaw University of Life Sciences (SGGW), Faculty of Agriculture and Biology, Department of Soil Environment Sciences, Nowoursynowska 159, 02-776, Warsaw, Poland
[e] IHE Delft Institute for Water Education, Department of Environmental Engineering and Water Technology, P.O. Box 3015, 2601, DA, Delft, The Netherlands

ARTICLE INFO

Keywords:
Artificial root exudates
Metal
Fayalite
Sulfides

ABSTRACT

This study was undertaken to simulate experimentally the weathering of slags disposed nearby soil rhizosphere. The aim of the research was to differentiate the effect of pH and organics on slags dissolution as well as to indicate weathering sequence of phase components. The studied slags are mainly composed of Fe (34.5 wt%) and Si (17.9 wt%) and contain up to 3761 mg kg^{-1} of Cu and 3628 mg kg^{-1} of Zn. The main identified phases are fayalite and glass, whereas sulfides and metallic Cu are volumetrically minor. A 30 days long slag weathering experiment was carried out with artificial root exudates (43.7 mM) and demineralized water at initial pH = 3.5 and pH = 6.7. The highest metal release (up to 10.9% of Zn and 4.6% of Cu) was observed in ARE solution at initial pH 3.5. Dissolution of sulfides and fayalite was mainly driven by pH. Artificial root exudates enhance glass dissolution as compared to demineralized water regardless of initially fixed pH. Based on this study following weathering sequences are delineated: i) under ARE 3.5 conditions: silicates > glass > sulfides, ii) under DW 3.5 conditions: sulfides > silicates > glass, iii) under near-neutral conditions: sulfides > glass > silicates.

1. Introduction

Numerous historical industrial districts are characterized by the presence of waste dumps where various by-products including mine tailings and slags have long ago been disposed of (Ettler et al., 2003; Ettler, 2016; Kierczak and Pietranik, 2011; Piatak and Seal, 2010; Potysz et al., 2018; Tyszka et al., 2018). Slags are important waste streams of pyrometallurgical operations due to their large production volume and high residual metal content. Former disposal options were not considered carefully enough and the landfills lack efficient barriers isolating the wastes from the environment (Kominková et al., 2018). Thus, percolating water and/or soil solution cause that the slags are exposed to the environmental factors such as pH, redox variations, organic acids, microorganisms which affect the geochemical stability of disposed wastes (Potysz et al., 2015 and references therein). Ongoing weathering processes often result in the release of toxic elements from slags and their irretrievable input into the environment (Ettler, 2016; Gee et al., 1997; Piatak and Seal, 2010; Tyszka et al., 2014; Parsons et al., 2001; Potysz et al., 2017).

The rate and strength of the weathering vary depending upon slag disposal conditions. The wastes located inside the dump are exposed to specific conditions (*e.g.* pH, temperature, humidity) differing from those encountered at the top of the dump (Potysz et al., 2017; Tyszka et al., 2014). Furthermore, a straightforward contact with top soil layer or dump vegetation cover enables the slags to interact with the soil solution (Bunzl et al., 2001; Houben et al., 2013; Potysz et al., 2017). In addition, the vegetation cover hosts various plant assemblages and creates organic-rich and microorganism-rich conditions highly resembling these encountered in the rhizosphere (Baetz and Martinoia, 2014; Hinsinger et al., 2003; Jones, 1998: Ash et al., 2014). Organic acids (*e.g.* citric, succinic, malic, and lactic acid) in the rhizosphere occur at concentration varying from micromolar up to millimolar levels (Ash et al., 2014; Jones, 1998; Sposito, 2008). For example, Montiel-Rozas et al. (2016) investigated root exudation from various plant species and demonstrated exudation of citric acid reaching 2.92 mM per hour. Steinauer et al. (2016) used root exudates cocktails at concentration 20 mM. The research studies considering various interactions of root exudates in the environment implemented ARE at concentration

* Corresponding author.
E-mail address: anna.potysz@uwr.edu.pl (A. Potysz).

https://doi.org/10.1016/j.jenvman.2018.05.071
Received 26 January 2018; Received in revised form 20 May 2018; Accepted 22 May 2018
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reaching even up to 1000 mM (Gao et al., 2010; Jia et al., 2018). Furthermore, there is a close dependency between the root exudates/organic acids and microbial activity in rhizosphere system (Steinauer et al., 2016). Rhizosphere is enriched in microbial communities mainly because organic compounds serve as a source of carbon for heterotrophic organisms. In turns, neighboring microbial consortia may also degrade organic acids (Ahkami et al., 2017). Furthermore, organic acids have a deteriorative impact on slag surfaces that may result in the release of toxic elements (Drever and Vance, 1994; Drever et al., 1994; Potysz et al., 2017). Geochemical stability of industrial wastes in the presence of organics has already been evaluated (Ash et al., 2014; Banks et al., 1994; Ettler et al., 2004, 2009; Potysz et al., 2017). Nevertheless, more detailed studies considering the release of major and minor elements in the column-like experimental system as well as geochemical prediction of secondary precipitates are still required for elucidating dissolution of individual slag phases (Potysz et al., 2017).

The main objective of this study was to explore root exudates driven weathering of metallurgical slags formerly disposed of nearby the industrial district located in south-western Poland in order to assess the environmental risk associated to this type of waste. This study examined: i) slags behavior under dynamic conditions simulating several root exudation events, ii) the role of organic compounds in metal (Cu, Zn, Pb, Co) mobility and iii) formation of secondary precipitates using geochemical modeling. To reach this goal, weathering experiments with artificial root exudates and ultrapure water solutions, both at initial pH (t_0) = 3.5 and pH (t_0) = 6.7 were carried out. The experiments were executed in a dynamic system with solution renewal every 5 days during 30 days. The reason for choosing low pH values (*i.e.* 3.5) was that rhizosphere is known for its acidic conditions (Nye, 1981). Although weathering simulation under near neutral pH conditions (pH (t_0) = 6.7) is not realistic in a real-time rhizosphere soil fraction, this experimental approach was required for clarifying whether slag dissolution process is exclusively driven by pH as well as for elucidating the relevance of organics assistance in an acidic and near-neutral pH conditions (*i.e.* 3.5 and 6.7). As the result of this study the weathering sequence of slag phase components was delineated.

2. Materials and methods

2.1. Slag sampling and characterization

Metallurgical slags used in this study were collected from historical industrial district located in the Rudawy Janowickie Mountains (south-western Poland). The slags are remains of pyrometallurgical activity held in 14–16th century (Kierczak and Pietranik, 2011). The slags have been randomly disposed and currently they may be found in soil profiles, stream beds and stream waters. Typical slag-abundant disposal spot located near soil rhizosphere is presented in Fig. 1. This disposal situation justifies the reason for simulating rhizosphere-like weathering undertaken in the frame of the present study. Detailed information about soils, stream beds and stream waters as well as characterization of historical slag have been described elsewhere (Kierczak et al., 2013; Potysz et al., 2016a).

The slag samples were characterized by the ACME Analytical Laboratory (Vancouver, Canada) for its chemical composition. The sample fusion by $LiBO_2/Li_2B_4O_7$ was carried out following digestion of the residue with HNO_3. The analytical reproducibility (2σ), as estimated from replicate analysis of sample JK113 (*i.e.* slag used in the present study) and eight analyses of standards DS7 and SO-18, ranges from 3% (Zn) to 16% (Cu) at 95% confidence limits. Analytical accuracy (2σ), as estimated from 8 measurements of standards (SO-18 and DS7) is from 3.5% (Zn) to 6.3% (Pb) at 95% confidence limits.

The mineral composition of the slags was analyzed using electron microprobe analyzer (Cameca SX100) with employment of two individual analytical programs for silicates and sulfides. The standards used were following: fayalite (Si, Fe), forsterite (Mg), Al_2O_3 (Al), orthoclase (K), wollastonite (Ca), TiO_2 (Ti), rhodonite (Mn), GaAs (As), $CuFeS_2$ (Cu) and willemite (Zn). Analytical conditions for both programs were 15 kV accelerating voltage, 10 nA beam current, and counting time 10 s for all the elements. The total number of spot analyses was 40 (Tables S1 and S2; Supplementary materials).

2.2. Experimental procedure

The slag grains used for the experiments were prepared by crushing using mechanical crusher and sieving using a vibratory sieve shaker (FRITSCH Analysette 3 Pro) to obtain fraction size at 1–2 mm. The sieving conditions corresponded to 1.0 (± 0.1) amplitude and 15 min of agitation with no interval time. Slag grains were rinsed 3 times with ultrapure water in order to remove possibly high number of fine slag particles. The experiments were designed such that 4 g of the slags were placed into polypropylene tubes (15 mL capacity) with uniformly drilled holes (0.5 mm size). Tubes were introduced into glass bottles containing 100 mL of the weathering solutions (*i.e.* artificial root exudates or demineralized water) and shaken at 100 rpm in order to allow the solution to penetrate the column tubes. The reactors were gently closed to avoid entrance of microorganisms and to limit evaporation, but to keep the oxygenated conditions (van Hullebusch et al., 2015; Potysz et al., 2016b). Organic solution was modified after Baudoin et al. (2003) and was composed of sugars: glucose (9.2 mM), fructose (9.2 mM), and sucrose (4.6 mM) as well as organic acids: citric (4.6 mM), lactic (9.2 mM), and succinic (6.9 mM). The experiments were carried out with organic acid rich solutions fixed at initial pH (t_0) = 3.5 (ARE 3.5) and pH (t_0) = 6.7 (ARE 6.7). Demineralized water adjusted to the initial pH (t_0) = 3.5 (DW 3.5) and pH (t_0) = 6.7 (DW 6.7) were used as inorganic experimental controls. The pH adjustment of ARE 6.7 solution was done using 1 M KOH, whereas DW 3.5 was adjusted using 1 M HNO_3. The experiments were performed during 30 days with a regular sampling interval set at each 5 days when each leach solution was replaced by fresh weathering solution. All experiments were carried out under sterile conditions in order to rule out the effect of microbial organisms on slags weathering and to avoid biodegradation of organic compounds. The glassware, slags, and DW solution were autoclaved for 20 min at 120 °C. During sterilization, the slags were kept in polypropylene tubes closed with cotton plug and aluminum foil in order to avoid moisture penetration and to maintain dry sterilization conditions. Organic solutions were sterilized using sterile polyethersulfone 0.45 μm pore size filters (Chemland). The bottles were wrapped up with aluminum foil in order to maintain dark conditions and to avoid organic matter photodegradation. All experiments were performed in duplicates and accompanied by procedural blanks (*i.e.* each weathering solution without the slags). The pH and Eh values were measured immediately after opening reactors using pH/Eh meter Elmetron CP-411. The leachate for elements assaying was filtered using nitrocellulose filters (0.45 μm pore size), acidified using 1% vol. of 65% HNO_3 and stored at 4 °C until analysis. The non-acidified leachate was also kept at 4 °C and analyzed for anions content.

2.3. Leach solution analysis

Elemental composition of the leachates was determined using inductively coupled plasma atomic mass spectrometry (ICP-MS Nexion 300D Perkin Elmer). Detection limits were typically 0.1 μg/L and 1 μg/L for minor and major elements, respectively. The elements measured were following: Si, Fe, Al, Ca, Mg, K, Na, Cu, Zn, Pb, Co, Cd, As. The anion content was determined using ion chromatography (Compact IC Flex chromatograph, Metrohm). Quantification limits were typically 5 ppm for F^-, 10 ppm for Cl^- and 3 ppm for NO_3^-, PO_4^{3-} and SO_4^{2-}. Content of bicarbonates (HCO_3^-) was determined by titration with HCl. All the anion concentrations are presented in Table S3 (Supplementary materials).

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