Effect of galena contained in dust from Zn–Pb metallurgical processes on environment

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Received: 24 February 2020 / Accepted: 16 March 2021 / Published online: 2 April 2021
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Abstract

This paper assesses the effect of galena emitted with dust from an electrostatic precipitator at Miasteczko Śląskie Zinc Smelting Plant (Poland) onto a soil and water environment. The Eh–pH diagrams plotted in the temperature range of – 3 to 25 °C serve to determine the conditions of PbS stability and conditions under which its decomposition may occur, resulting in the release of ionic forms of lead into the soil and water environment. Determination of the mobility of galena emitted with metallurgical dust is a current and important issue in view of the geochemistry of mineral components in the environment.

Keywords Galena · Dust · Pyrometallurgy · Soil · Water · Environment

Introduction

During the pyrometallurgical extraction of zinc and lead large amounts of dust are discharged into the environment.

The negative impact of particulates on the environment depends primarily on their chemical and mineral composition, which are the factors that determine the mobility and bioavailability of pollutants, and on the size of their particles, which determines the extent of dispersion of the dust in the atmosphere, as well as in water and soil.

The chemical and mineral composition of dusts from Zn–Pb smelting depends on: the type of charge material, which has the form of charge mixture consisting of zinc–lead concentrates and Zn and Pb containing waste materials (among others dust, dross, steelmaking dust, zinc and lead scrap) and on the point of their generation in the technological process (emission source) (Bernasowski et al 2017; Carpenter et al 2015; Gregurek et al., 2015; Ray and Ghosh 1993; Zhao 2013; Wu et al 2019).

Metallurgical dusts are characterised by coarse grain size, varying chemical composition and complex mineral composition (Adamczyk et al 2010; Adamczyk and Nowińska 2016; Czaplicka and Buzek 2011).

The main mineral components of Zn–Pb metallurgical dust are sulphides (ZnS, PbS, FeS₂), oxides (ZnO, PbO, FeO, Fe₃O₄) and sulphates (Pb, Zn and Fe). These components usually occur as complex conglomerates containing numerous elements, including heavy metals, i.e. Cd, As, Sb, Cu, Tl, Mn, Sn (Adamczyk et al 2010; Ettler et al 2005a, b; Ettler et al 2001; Simonyan et al 2019; Sobanska et al 1999).

Minerals emitted with the dust into the hypergenic environment are broken down in the environment, causing the release of the elements they contain.

The mobility of elements in the environment is determined by a number of external factors, which include ambient temperature, amount of precipitation, insolation, oxidation–reduction potential and contact time of the material with precipitation water. One of the key parameters affecting the stability of elements is also the pH of the soil–water environment where the dust is deposited (Czaplicka and Buzek 2011; Piatak et al. 2015).

Zinc, lead and iron are the elements released from dusts in the highest amounts and characterised by significant solubility > 2.0 mg/L, with Pb showing low mobility, due to the formation of new stable phases in an alkaline environment, e.g. cerussite PbCO₃, anglesite PbSO₄. In acid soils and waters, lead mobility is increased and the main forms in which lead occurs there are organic complexes and Pb²⁺ and PbHCO₃⁺ ions. The increased mobility of lead is significantly enhanced by the salinity of the near-surface waters, which favours the formation of soluble PbCl₂ and PbCl⁺ complexes (Adamczyk and Nowińska 2013; Adamczyk et al

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2010; Bril et al. 2008; Cabała 2009; Ettler et al 2005a, b; Hoffmann et al 2012; Piatak et al 2015).

The aim of this study is to present the behaviour of PbS contained in the dust from one of the emission sources (electrostatic precipitator) at the Miasteczko Śląskie zinc smelter (Poland) in a hypergenic environment. On the basis of geochemical modelling (Pourbaix diagrams), the PbS stability and mobility regions were mapped in detail and thus the possibility of negative effects of the dust on the environment of the smelter area was determined.

Materials and methods

10 samples of dust from the electrostatic precipitator of the Sinter Unit (sintering machine) of Miasteczko Śląskie Smelting Plant were taken in monthly intervals.

The content of trace elements in dust samples was determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES) using a JY 2000 spectrometer, while their chemical composition was determined by X-ray fluorescence spectroscopy (XRF) on a RIGAKU ZSX PRIMUS spectrometer, equipped with a rhodium X-ray tube, with stepless voltage adjustment between 20 and 60 kV, analysing crystals LiF, Ge and a number of synthetics.

Phase identification by X-ray diffraction (XRD) was performed on a Seifert-FPM XRD 7 diffractometer using a cobalt lamp, Fe filter, 35 kV voltage, 25 mA current. The results of this analysis enabled the identification of the main phases present in the dust.

Chemical composition was determined in micro-areas using a JEOL JCXA 733 X-ray micro-analyser equipped with a wavelength-dispersive spectrometer, yielding information on the qualitative and quantitative chemical composition of the micro-area of the phases present in the slags in trace amounts. Due to the high resolution, sensitivity and low detection limit, the WDS method commonly used to identify the phase composition of grains of many types of samples, both environmental and industrial.

Measurements were performed under the following conditions: focused beam (diameter 1...2 µm, accelerating voltage 20 kV, current 3×10^{-9} A). For each preparation, a series of microanalyses were performed, comprising several to several dozen measurements of the chemical composition of characteristic grains to determine the predominant chemical forms of the occurrence of individual elements, primarily the major elements, taking into account the morphology of individual phase precipitates in the grains. About 10 chemical composition measurements were performed on any given grain, and the average of these measurements was taken as the final result.

To determine the mobility of lead sulphide, which is one of the major identified mineral constituents of the dust from

the electrostatic precipitator, geochemical modelling based on Eh–pH diagrams was applied. These diagrams were plotted using HSC Chemistry software for conditions characteristic of the soil and water environment of the Smelting Plant area. For this reason, the following parameters were implemented: mean precipitation: 700 mm/year, mean temperature in winter season: $-3\text{ }^{\circ}\text{C}$, maximum temperature in summer season: $+25\text{ }^{\circ}\text{C}$. Studies were performed within the water stability field (Eh range: $-0.6...+1.2\text{ V}$) and pH range of 0...14, wherein the diagrams were plotted for pH values in the range of 2 to 10.

Results and discussion

The dust from the electrostatic precipitator of the Sinter Unit of Miasteczko Śląskie Smelting Plant was the subject of previous studies, conducted by the authors, involving chemical and mineral composition.

Chemical composition of the electrostatic precipitator dust

Based on the results of spectrometric tests, the basic constituents of electrostatic precipitator dust are Pb and Zn, which jointly constitute 88 wt% of the dust (Table 1). The other two prevailing elements in the composition of the dust are S and Cd, at concentrations of 4.50 wt% and 4.08 wt%, respectively (Adamczyk et al. 2010; Nowińska et al 2015; Adamczyk and Nowińska 2016).

Based on the identification of the mineral composition of the electrostatic precipitator dust, it was found that the main phase constituents of that dust were sulphides, PbS (galena)

Table 1 Chemical composition of dust from electrostatic precipitator of the ISP plant (wt%) (Adamczyk et al 2010)

Component	Pr-4
SiO ₂	0.8343
TiO ₂	0.5949
FeO	0.0441
MnO	0.1519
CaO	0.0641
K ₂ O	0.0655
PbO	73.2720
ZnO	15.2169
S	4.5020
As ₂ O ₃	0.5641
CdO	4.0795
SeO ₂	0.0504
Sb ₂ O ₃	0.0508
SnO ₂	0.0343
In ₂ O ₃	0.4752
Total	100.0000

and ZnS (sphalerite), with lead oxide (PbO, minium) and zinc oxide (ZnO, zincite) present in lower amounts. Apart from these four constituents the dust from the electrostatic precipitator also contains an amorphous substance (glaze) (Adamczyk et al. 2010; Hongxu et al 2016; Cappuyns et al 2014; Kucha et al 1996; Kushnir 2014).

Interpretation of the results of phase composition examination of electrostatic precipitator dust in the context of the environmental impact thereof should mainly apply to one of the basic phases; that phase is lead sulphide. This constituent has admixtures of such elements as Fe, Mn, Zn, As, Se, Sb, Sn and In (Table 2).

If lead sulphide is released into the soil and water environment, the decomposition of this phase may mobilize these elements into the environment, depending on the conditions of that environment, primarily Eh and pH (Ettler and Johan 2014, 2015; Mizerna 2016; Mizerna and Król 2015; Piatak et al 2015; Songa et al 1999, Taylor and Lapa 1983).

Galena mobility in the soil and water environment

The basic phase component of the dust, lead sulphide, will decompose in hypergenic processes occurring mainly under the influence of factors such as: seasonal temperature variability, atmospheric precipitation, insolation and Eh and pH conditions of soil–water environment. The Eh and pH conditions, in turn, depend on many other factors, principally on chemical interactions with precipitation water and on soil quality. The Eh–pH diagrams that show the stability regions of the

main chemical constituents of lead sulphide (sulphur and lead) under adopted temperatures (− 3 and + 25 °C) and additionally in temperatures of + 5 and + 11 °C are presented in Fig. 1a, b. The Eh–pH diagrams plotted for the additional temperatures (+ 5 and + 11 °C), enable observation of the changes in the behaviour of the forms of occurrence, mainly of lead, with rising temperature. The final task required for plotting the diagrams was establishing the concentrations of Pb and SO_4^{2-} in an aqueous environment, which were assumed to be equal to permissible limits specified in the Ordinance of the Minister of Environment of 24 July 2006 on the conditions of discharging wastewater to waters and soil and on substances particularly harmful to the aqueous environment (Dz.U. 2006, No. 137, item 984): 0.5 mg Pb/L, 500 mg SO_4 /L.

The water and soil environment under investigation (Miasieczko Śląskie) has a pH range of 4.5 to 7.5.

In the surface region lead sulphide is decomposed by acidic solution in the following reactions:

Table 2 Chemical composition of lead sulphide in electrostatic precipitator dust (sampling point PR-4) and average chemical composition of lead sulphide from other processing points in the Miasieczko Śląskie Smelting Plant (MS-1—Raw Materials Storage 1, PR-2—dust from bagfilter FT12 at the sinter unit, PR-3—dust from bagfilter FT24 at the sinter unit, PR-4—dust from bagfilter FT12R at the sinter unit, PR-5—dust from bagfilter FT12R at the grinding unit, PR-7—dust from bagfilter FT10 at the lead refinery (Adamczyk et al 2010)

Element	Sampling point			Average chemical composition of lead sulphide from other points of the process line					
	PR-4-6	PR-4-7	PR-4-8	PR-4	MS1	PR2	PR3	PR5	PR7
S	13.077	12.558	12.364	12.666	14.567	14.633	14.309	13.771	13.837
Ca	0.000	0.000	0.000	0.000	0.080	0.013	0.023	0.000	0.000
Mn	0.189	0.577	0.238	0.335	0.169	0.042	0.050	0.013	0.010
Fe	0.000	0.000	0.000	0.000	0.183	0.220	0.275	0.167	0.064
Cu	0.000	0.000	0.000	0.000	0.119	0.079	0.271	0.100	0.538
Zn	0.366	0.000	1.413	0.593	3.849	4.234	2.892	3.135	2.619
As	0.189	0.590	1.063	0.614	0.171	0.137	0.033	0.170	0.201
Se	0.000	0.000	0.000	0.000	0.252	0.092	0.228	0.033	0.000
Ag	0.000	0.000	0.000	0.000	0.172	0.229	0.000	0.033	0.000
Cd	2.913	12.019	3.975	6.302	0.348	0.451	0.000	0.000	0.471
Sn	0.000	0.000	0.050	0.017	0.243	0.098	0.000	0.000	0.742
Sb	0.000	0.000	0.075	0.025	0.336	0.097	0.000	0.063	0.155
Pb	83.266	74.257	79.935	79.153	79.510	79.674	81.919	82.514	81.363
In	0.000	0.000	0.888	0.296	0.000	0.000	0.000	0.000	0.000
Total	100.000	100.000	100.000						
<i>PbS and CdS fraction</i>									
PbS	95.6143	83.9486	91.4987						
CdS	3.6416	14.8847	4.7758

Fig. 1 Eh–pH diagrams of lead sulphide stability at: – 3 °C (**a**), 5 °C (**b**), 11 °C (**c**) and 25 °C (**d**)

Reaction (1) represents the oxidative decomposition of PbS which produces Pb^{2+} and sulphur. Pb^{2+} may also be formed as a result of oxidation (Scheetz and Rimstidt 2009; Czaplicka and Buzek 2011):

Reaction (2) suggests that PbSO_4 forms when the redox potential of the solution is too high, and reaction (3) indicates that H_2S forms in the course of PbS decomposition in acidic solution when the redox potential is too low. Pb^{2+} formed in reaction (1) or (5) may undergo hydrolysis: $\text{Pb}^{2+} \rightarrow \text{PbOH}^+$.

Changes of Eh and pH that determine the stability region of lead sulphide in aqueous environment, may cause its decomposition. The products of this decomposition in the specified environment which include Pb, PbO_2 , PbSO_4 , Pb^{2+} , PbOH^+ , whereas sulphur, depending on the conditions, adopts the following forms: HS_2O_8^- , HS_2O_7^- , $\text{S}_5\text{O}_3^{2-}$, $\text{S}_4\text{O}_3^{2-}$, SO_4^{2-} . Among the forms listed, Pb, PbO_2 and PbSO_4 are solid, the remaining products will occur in the aqueous environment in ionic form (Pb^{2+} , PbOH^+ , HS_2O_8^- , HS_2O_7^- , $\text{S}_5\text{O}_3^{2-}$, $\text{S}_4\text{O}_3^{2-}$, SO_4^{2-}) (Fig. 1).

At -3°C , under oxidative conditions, Pb^{2+} prevails at a lower pH (below 6.20), whereas Pb(OH)^+ starts to prevail at higher pH (above 6.20). At $+5^\circ\text{C}$ the boundary pH value between Pb^{2+} and Pb(OH)^+ prevalence is 6.18, at $+11^\circ\text{C}$ it is 6.19, and at $+25^\circ\text{C}$ it is 6.21. The range of pH value variability is between 6.18 and 6.21; this corresponds to seasonal differentiation of temperature from -3 to $+25^\circ\text{C}$ and may in fact be neglected when determining the boundary pH value between Pb^{2+} and Pb(OH)^+ prevalence.

At -3°C , under alkaline conditions ($\text{pH} > 8.05$), for slightly reductive environments, (Eh below -0.32) lead sulphide will decompose to Pb and $\text{S}_4\text{O}_3^{2-}$; this is in contrast with the increase of temperature to 25°C , where the pH value shifts towards 7.26 and Eh remains virtually unchanged.

The environmental Eh and pH values, derived from the Eh–pH diagrams (Fig. 1), are listed in the table below. This list clearly shows that with increasing temperature, the stability region of lead sulphide is gradually reduced (decreasing absolute values of $|\Delta\text{Eh}|$ and $|\Delta\text{pH}|$) (Table 3).

The other phase that is important from the viewpoint of the soil and water environment is lead sulphate, which may form as a result of lead sulphide decomposition. The stability of this component under the conditions of the soil and water environment, as determined by the diagrams, is presented in Table 4. The values of these parameters indicate that with increasing temperature the stability region of lead sulphate is gradually reduced (decreasing absolute values of $|\Delta\text{Eh}|$ and $|\Delta\text{pH}|$), and at ca. 12°C this component is completely decomposed in the environment.

Table 3 Boundary Eh and pH values of lead sulphide stability and absolute differences between the values ($|\Delta\text{Eh}|$ and $|\Delta\text{pH}|$) corresponding to seasonal temperatures

Temperature ($^\circ\text{C}$)	Eh		pH	
	Value	$ \Delta\text{Eh} $	Value	$ \Delta\text{pH} $
-3	$-0.67\dots-0.13$	0.54	$2.47\dots12.44$	9.97
$+5$	$-0.66\dots-0.14$	0.52	$2.62\dots11.94$	9.32
$+11$	$-0.65\dots-0.15$	0.50	$2.72\dots11.58$	8.86
$+25$	$-0.64\dots-0.17$	0.47	$2.96\dots10.86$	7.90

Table 4 Boundary Eh and pH values of lead sulphate stability and absolute differences between the values ($|\Delta\text{Eh}|$ and $|\Delta\text{pH}|$) corresponding to seasonal temperatures

Temperature ($^\circ\text{C}$)	Eh		pH	
	Value	$ \Delta\text{Eh} $	Value	$ \Delta\text{pH} $
-3	$0.10\dots0.36$	0.26	$4.97\dots6.41$	1.44
$+5$	$0.09\dots0.30$	0.21	$5.37\dots6.27$	0.90
$+11$	$0.08\dots0.25$	0.17	$5.69\dots6.20$	0.51
$+25$	No phase			

The findings presented above show that:

- the stability region of lead sulphide is defined by a wide span of pH values (from ca. 2.50 to 12.50) and a narrow span of Eh values (from -0.67 to -0.13),
- the stability region of lead sulphate is defined by a narrow span of pH values (from 4.97 to 6.41) and a narrow span of Eh values (from 0.10 to 0.36).

The narrow range of Eh and pH values within which lead sulphide and lead sulphate are stable is unfavourable for the soil and water environment. This is mainly due to the possibility of changing this environmental parameter within a narrow range beyond the boundary values. The effect will be the decomposition of this phase into metallic Pb, Pb^{2+} , PbOH^+ and sulphate ions (Fig. 1), posing a hazard for surface and underground waters. In addition to lead, other elements present as admixtures in lead sulphide (Fe, Mn, Zn, As, Se, Sb, Sn and In) will also be released into the environment (His-Hsiung 2016; Morrison et al. 2016; Nang-Htay 2016; Lia 2010; Sobanska et al. 1999).

Similar analyses have been performed for fayalite, the main constituent of slag generated and landfilled by the Miasteczko Śląskie Smelting Plant (Atlas of Eh–pH diagrams 2005; Adamczyk and Nowińska 2013; Puziewicz 2007; Ettler et al. 2001; Ray 1993). The Eh and pH parameters of the stability region of this constituent, established under similar environmental assumptions using diagrams, indicate that maintaining the pH value of soils at above 7

is favourable for the stability of fayalite. In the case of lead sulphide, the Eh potential seems to be the parameter of the soil that should be controlled to ensure stability.

Conclusions

The investigations carried out allow to draw the following conclusions.

The main mineral constituents of roaster dust are the oxides: ZnO (zincite) and PbO (minimum). As a result of oxidizing and high-temperature conditions of the roast sintering process, ZnO and PbO form as a result of the exogenic conversion of minerals, ZnS (sphalerite) and PbS (galena), contained in the charge mixture. The roasting dust was found to also contain sulphides, ZnS and PbS, that weren't the products of oxidizing roasting. These sulphides are relict minerals contained in the charge mixture that have not undergone conversion.

Interpretation of pH-Eh diagrams has shown that in the soil and water environment of the surroundings of the Miasteczko Śląskie Zinc Smelting Plant, at temperatures of between $-3\text{ }^{\circ}\text{C}$ and $+25\text{ }^{\circ}\text{C}$, lead sulphide will decompose into Pb, PbO_2 and PbSO_4 , as well as Pb^{2+} , PbOH^+ , HS_2O_8^- , HS_2O_7^- , $\text{S}_5\text{O}_3^{2-}$, $\text{S}_4\text{O}_3^{2-}$, SO_4^{2-} .

The main phases that may be present under the soil and water conditions are lead sulphide and lead sulphate, the stability region of which is defined by the following parameters:

- lead sulphide—wide span of pH values (from ca. 2.50 to 12.50) and a narrow span of Eh values (from -0.67 to -0.13),
- lead sulphate—narrow spans of pH values (from 4.97 to 6.41) and Eh values (from 0.10 to 0.36).

With increasing temperature, the stability region of lead sulphide is gradually reduced (decreasing absolute values of $|\Delta\text{Eh}|$ and $|\Delta\text{pH}|$), while that of lead sulphate is reduced rapidly, and at above $12\text{ }^{\circ}\text{C}$ the latter constituent is completely decomposed in the environment.

The ionic forms, which are readily assimilated by plants, constitute the main hazard for the soil and water environment. Therefore, narrow ranges of Eh and pH values that determine the stability of solid forms of lead (lead sulphide or sulphate) pose a hazard for this environment, as slight changes of these values may result in PbS decomposition into ions.

Identification of the forms of element occurrence in the dust from the oxidizing roasting process constitutes the basis for geochemical modelling. Determining the mobility in a hypergenic environment of heavy metals contained in the roasting dust enables the assessment of the impact of the smelting plant operation on that environment.

On the other hand, knowledge of the forms of occurrence of metals in the metallurgical waste (including process dust) helps identify the optimum technology for obtaining elements from this waste, thereby reducing dust emissions to the environment.

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