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Mobility of heavy metals from metallurgical waste in the context of sustainable waste management

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Abstract: Metallurgy is one of the industry producing the most diverse range of wastes in terms of their composition and level of heavy metals contamination. With the development of economy, the amount of produced wastes increases. It involves with the searching for new methods of waste management. The one of priority in the activity of companies is the use of principles of sustainable development. Constantly a new technological solutions are searched in order to improve the environment protection and increasing the reuse of waste. This behaviour is the basis for sustainable waste management. Unfortunately, it is not possible to economic use of all waste. It is caused by the level of contamination of wastes, as well as the risk of environmental contamination by inappropriate disposal of waste. In order to increase the protection of the environment from the effects of uncontrolled impact of waste, the leaching tests of contaminants are carried out. This practice is indispensable operation of every enterprise that produces the wastes that may negative affect the environment, especially the water environment. In the paper, the results of heavy metals concentrations from metallurgical slags (zinc and copper metallurgy) were presented in order to assess the mobility of contaminants into the environment. The levels of heavy metals release from waste from variety of industrial sectors were also analysed.

Keywords: sustainable waste management, metallurgy waste, heavy metals mobility

JEL codes: Q01, Q53

1. Introduction

Sustainable development is a concept born in the late 20th century. The Commission on Environment and Development in the U.N. report "Our Common Future" published in 1987 defined the sustainable development as the "development that meets the needs of the present

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without compromising the ability of future generations to meet their own needs." The report was a breakthrough in the adoption of the sustainable development concept in the field of science, politics and economy. This future-oriented concept is very clearly related to the quality of human life. It involves balancing various areas: natural and environmental (nature conservation), technical (new technologies, saving of resources), economical (taxes, subsidies), social (tackling unemployment) and political areas (implementation and monitoring of the sustainable development strategy) (Pawłowski and Pawłowski, 2004: 277). One of the goals of the implementation of sustainable development principles in manufacturing processes is to minimise a negative environmental impact of the industry, which is a challenge to technical sciences. In the field of sustainable waste management, technological processes should be carried out in such a manner to reduce the flow of raw materials by decreasing the amount consumed. This can be achieved by the more efficient use of raw materials and the implementation of material-efficient technologies. However, the most important aspect of waste management is the recovery and reuse of materials and waste. Both strategies are a part of the concept of "cleaner production", which aims to reduce expenses and decrease harmful effects of industry on people and environment (Pawłowski and Pawłowski, 2004: 278).

Sustainable development in the metallurgy industry involves the application of new principles and techniques related to issues such as: the optimisation of production processes to improve the energy efficiency, decrease the unit consumption of energy and resources used for the production, reduction of harmful impact on the environment, in particular: reduction of emission of hazardous substances into the atmosphere and increase the reuse level of by-product and waste (Peng et al., 2015: 1931). Such actions necessarily involve large costs for companies. However, the implementation of practices for the improvement of the environmental quality is necessary to achieve sustainable development. Sustainable development contributes to the economic growth, while reducing threat to natural resource management (El-Haggar, 2007: 1). Metallurgy industry uses a variety of technologies depending on the production process applied. It may also affect the environment to a different extent, polluting the air, soil and water. The processing of raw materials is accompanied by the emission of dust and gas pollutants. The water consumed in technological processes may be contaminated with suspensions, mineral oils and various chemical compounds. Steelworks with the full production cycle (coking, sintering, iron blasting and converting processes) are most harmful to the environment. However, recently, more and more attention has been paid to the need to improve environmental protection. Currently, dust pollutants are captured by dust removal devices. Moreover, a significant gas emission reduction was observed as a result of the modernisation of sintering processes, the installation of seals in iron blast furnaces or the removal of open hearth processes. Thanks to the sewage treatment and recirculation, water management was improved. Dumps used for the waste disposal are also removed from plant sites. Such actions are consistent with the sustainable development principle (PPTS, 2006: 9).

Waste landfilling is the final and least desirable method of waste disposal. The interest in the problem of metallurgical waste stored at dumping grounds located near steelworks made it necessary to conduct multi-directional research on the leaching of heavy metals. Such research is also important in case of using waste as secondary raw materials for the production of materials placed in the environment. It provides important information on the content of heavy metals in waste, as well as their mobility and migration to the environment (Jonczy et al., 2014: 162). Waste deposited in the natural environment is exposed to various physical, biological and chemical factors that affect the mobility of heavy metals. Contaminants contained in waste, when exposed to water, may leach into soil and enter surface and groundwater (Sloot and Zomeren, 2012: 92). Therefore, before discharging waste to the environment, an entrepreneur has to perform leaching tests in order to assess the mobility of contaminants in the environment.

The paper raises the issue of sustainable waste management and heavy metals mobility from metallurgical waste. The aim of the paper is to evaluate the mobility of heavy metals from selected metallurgical wastes: copper slag and slag from zinc metallurgy. Additionally, the impact of tested waste on the environment was assessed. The paper also presents the results of heavy metals leaching from slags analysed by other authors.

2. Metallurgical waste management

The metallurgical industry is one of the largest branches of industry producing various mineral and metallurgical waste. Each waste-producing company should strive to prevent and reduce waste production. The waste that cannot be avoided should be reused. If it is necessary to dispose waste, its negative impact on the environment and human health has to be reduced. Such actions should comply with the waste management hierarchy set out in the Waste Framework Directive (European Commission, 2008).

The main types of waste generated in the metallurgical industry are as follows (Lis et al., 2015: 283):

- slag and dust from blast furnace process,
- dust and sludge from the sintering.
- dust and sludge from steel production in converters,
- slag and dust from steel production in electric arc furnaces,
- fine metal-bearing wastes,
- grinding dust,
- ceramic debris.

The reuse of metallurgical waste more and more often becomes a requirement, not only in economic terms, but also in environmental ones (PPTS, 2006: 60). Sitko (2014: 533) reports that slag is the waste the largest amounts of which are produced by the steel and iron industry. Currently, the average slag weight produced globally by the steel and iron industry is about 300 kg per each 1 Mg of crude iron, and for individual steelworks it varies from approx. 180 kg/Mg to over 400 kg/Mg. In Poland, this value was approx. 700 kg/Mg until 1980 whereas it currently varies between 300 and 400 kg/Mg of crude iron. For years metallurgical slag has been stored at dumping grounds. Nowadays, there is however an ongoing search for various possibilities of using such waste. When reusing waste, one should pay attention to the content of harmful elements (especially heavy metals). Therefore, contaminants in waste should be monitored and balanced in the metallurgical cycle. It is also important to develop safe technologies for the processing of waste containing hazardous substances (PPTS, 2006: 60; Jonczy, 2012: 63).

The main direction of the metallurgical slag management is to use it in the constructions, as a cement component and for the production of aggregates (Kuterasińska and Król, 2015: 61). Iron blasting slag is almost entirely recyclable. It is commonly used in the road construction for the construction of asphalt and concrete surfaces, and road foundations. In case of asphalt surfaces, iron blasting slag-based aggregate is used both in the binder and wearing course. As a cementing additive in a granular form, it is used in concrete surfaces (Jonczy, 2012: 63).

Another direction of the metallurgical slag management is to use it as a source of metals (Jonczy, 2012: 63; Sitko, 2014: 537). Such actions are becoming more and more popular in non-ferrous metallurgy (copper, zinc, tin, lead and aluminium). Metal recovery technologies differ depending on their form in the slag. However, a common advantage of metal recovery from slag, next to the metal yield maximisation, is a decrease in the mass of landfill waste. In the long-term perspective, such actions also allow for the reduction of costs of production processes.

For the metallurgical waste management, it is necessary to understand its phase and chemical composition in detail. Such studies enable us to identify forms of heavy metals in waste, the way they are bound to other components and possibility of releasing elements as a result of chemical weathering or contact with water (Jonczy, 2012: 64).

3. Mobility of heavy metals from waste

The potential environmental impact of waste will mainly depend on its chemical and physical properties and conditions of its reuse or neutralisation. The chemical and mineralogical composition of waste is the main aspect to which attention has to be paid when determining the environmental impact of waste. Such properties determine the chemical stability of waste when exposed to various factors. The potential environmental impact of waste is also determined by the content and mobility of heavy metals as well as the solubility of individual components (Wiertz, 1999: 404). The mobility of elements in the landfill zone is mainly determined by their form and environmental conditions. Changing environmental conditions, e.g. by changing pH, can lead to the concentration of heavy metals in various environmental components and pollution. In case of good bioavailability of toxic elements, the metabolism of living organisms may become disturbed. Elements may be present in a stable form (bound in crystalline structures of mineral components), or in a mobile form (present at ion-exchange positions or on the surface of waste particles). When an element is in a stable form, a mineral component may be subject to decomposition due to changes in environmental conditions. As a result, the element will be released to the environment (Nowińska and Adamczyk, 2013: 79; Król, 2012: 29-30). The main factors which influence the process of release of heavy metals are: fragmentation and shape of waste, ambient temperature, ratio of liquid to solid L/S, redox potential and the time of contact with rain water (Król, 2011: 72; Sloot and Zomeren, 2012: 92).

There are various methods used throughout the world for the assessment of the mobility of heavy metals in waste deposited in landfills. The TCLP method (*Toxicity characteristic leaching procedure*) developed by the US Environmental Protection Agency is the procedure that is most frequently used for the waste toxicity assessment (Environmental Protection Agency, 1992). This method allows for the determination of the mobility of organic and inorganic substances present in water, soil and waste, on the basis of which waste can be classified in terms of harmful effects on the environment. Laboratory conditions used in this method are to simulate the leaching of contaminants occurring in landfills. However, conditions (including weather conditions) of waste storage may change. Therefore, the assessment of contaminant leaching from waste should always be adjusted to the type of waste deposited and disposal conditions (Wiertz, 1999: 407).

In Europe, including Poland, tests related to the leaching of heavy metals from granular materials (with a grain size of <4 mm and <10 mm) are carried out according to the procedure described in the standard EN 12457:2002 (CEN, 2002). The mobility of heavy metals is most often presented using one-stage 24-hour extraction at the liquid/solid (L/S) ratio equal to 10 dm^3/kg. The leaching results obtained with the use of this method may also be used for the assessment of the potential release of hazardous components from waste, when stored in the natural environment and exposed to weather conditions, mainly precipitation. The analysis of the potential environmental impact of waste is one of the sustainable waste management principles. Entrepreneurs are obliged to conduct tests related to the contaminant leaching from waste in order to assess the potential effects of waste on the environment.

4. Materials and methods

The assessment of the mobility of heavy metals was carried out from zinc metallurgy slag and shaft copper slag. The former waste was produced in a rotary furnace at the department of lead refinery. The slag had various grain size distribution. It is disposed in the steelworks' hazardous waste landfill. "Fresh" slag (stored in the landfill for a short period of time) samples were collected for testing. Copper slag collected for testing was in the form of irregular chunks of various sizes. It is produced in the process of smelting a briquetted copper concentrate in a shaft furnace. Due to the low copper content, slag is the final waste of the melting process. It is transported in a liquid form to the dump, where it solidifies.

The test of the heavy metal leaching from metallurgical slag was performed according to the procedure based on the standard EN 12457:2002 (CEN, 2002). To that end, water extracts of L/S ratio = 10 dm^3/kg were prepared from ground waste. Material was subjected of agitation with water for 24 hours. Subsequently, the eluates were filtered through membrane filter and analysed.

Heavy metal concentrations in water extracts were determined by means of the atomic absorption spectrometry with flame atomization.

5. Results and discussion

The concentration of heavy metals (Cd, Cr, Ni, Cu, Zn, Pb) in water extracts are presented in Table 1. The concentrations of individual elements indicate their leachability to the aqueous phase and thus their mobility in the environment. Additionally, the results were compared with the permissible heavy metal concentrations set out in the Regulation of the Minister of Environment of 18 November 2014 on the conditions to be met when discharging sewage into water or soil and on substances particularly harmful to the aquatic environment (Dz. U. 2014).

Table 1. Heavy metals concentrations in water extracts from metallurgical slag compared with the limit values in (Ministerstwo Środowiska, 2014)

Type of waste	Concentrations of heavy metals (mg/dm^3)					
	Cd	Cr	Ni	Cu	Zn	Pb
Slag from zinc metallurgy	5.19	0.015	4.43	1.35	113	3.57
Copper slag	< 0.005[1]	0.002	< 0.010	0.026	0.100	< 0.050
Limit values of leaching	0.40	0.50	0.50	0.50	2.00	0.50

Source: Author's own research results compared with the limit values in (Ministerstwo Środowiska, 2014)

Copper slag demonstrated a low leachability of heavy metals. Due to the low content of heavy metals not exceeding the permissible values, such waste poses no significant environmental risk. Moreover, it is temporarily stored in the dumps. It can be widely used in the construction industry and road engineering. Unlike the copper slag, slag from zinc metallurgy is problematic and requires proper neutralisation methods due to the high level of heavy metal leaching. Currently, it is stored in a hazardous waste landfill adapted to this type of waste. The landfill must have appropriate facilities protecting the natural environment (soil, water) against contamination. The correct design of the landfill is an important aspect of waste management. Heavy metals are easily solubilised

[1] The result with the sign "<" means value of leaching below the detection limit.

under weather conditions and transferred to the aqueous phase. Therefore, it is required to carry out tests related to the mobility of heavy metals in waste in case of the waste disposal in the environment. Meeting the heavy metal leachability criteria is one of the conditions for the acceptance of waste for storage in the landfill of a given type. In Poland, there are the following types of landfills: landfills for neutral waste, hazardous waste and non-hazardous waste. Waste is classified based on the leachability criteria set out in the Regulation of the Ministry of Economy of 16 July 2015 on the acceptance of waste for depositing in a landfill (Ministerstwo Gospodarki, 2015). This regulation may also be used for the assessment of the level of heavy metal leaching from waste and thus an environmental risk posed by given waste.

The article also focuses on the assessment of the leaching of selected heavy metals from metallurgical slags from various metallurgical industry branches in Poland. Table 2 presents the concentrations of selected heavy metals (Cd, Cr, Ni, Cu, Zn, Pb) in water extracts from waste obtained by other authors (Jonczy et al., 2014: 171; Sanak-Rydlewska, 2011: 83; Gambal, 2013: 92; Rzechuła, 1994: 212). The leachability results have been given for information purposes only. The release rate of heavy metals will depend on the type of the technology used, which generated waste, the waste structure and the storage time.

Table 2. Heavy metals leachability from various types of metallurgical slags: a review

Type of waste	Concentrations of heavy metals (mg/dm^3)					
	Cd	Cr	Ni	Cu	Zn	Pb
Vitrified metallurgical slag (zinc and lead metallurgy) (Jonczy et al., 2014: 171)	< 0.001	< 0.002-0.005	< 0.007	0.006-0.055	0.009-0.017	< 0.005
Slag from closed steel plant landfill (Sanak-Rydlewska, 2011: 83)	n.d.[2]	0.003-0.015	0.008-0.019	0.008-0.014	n.d.	< 0.016
Copper slag (granulated) (Gambal, 2013: 92)	0.09	0.068	< 0.11	0.36	0.53	< 0.20
Spent copper slag (Rzechuła, 1994: 212)	n.d.	n.d.	n.d.	0.50	0.80	0.50

Source: Author's own compilation based on the results of the literature (Jonczy et al., 2014: 171; Sanak-Rydlewska, 2011: 83; Gambal, 2013: 92; Rzechuła, 1994: 212)

[2] no data

Jonczy et al. (2014: 171) studied the leachability of heavy metals from vitrified metallurgical slags after the production of zinc and lead, taken from an old, disused dump. The waste was produced as a result of rapid setting of liquid slag. The concentration range given (Table 2) is an average value obtained in 5 water extracts. The analysed slag had a trace leachability of heavy metals. However, due to the fact that the waste has been deposited in the dump at the turn of the 19th and 20th century, during this period, the leaching of the maximum amounts of contaminants might have occurred. Additionally, a dense, crystallised waste structure could have limited the release of heavy metals in the environment.

Slag collected from closed steelworks landfill also demonstrated a low leaching rate of Cr, Ni, Cu and Pb (Sanak-Rydlewska et al., 2011: 82-83). The authors (2011: 82-83) analysed the heavy metal leaching from three slag samples taken from different places of the dump. Copper metallurgy slags constitute another group of slags. Gambal (2013: 92) determined the zinc and copper content in water extracts from granulated copper slag (from blister copper production in electric furnace) at levels of 0.53 mg/dm^3 and 0.36 mg/dm^3, respectively. The tested waste was also characterised by the chrome release in the amount of 0.068 mg/dm^3. In spite of the results of the determination of above-mentioned heavy metals in water extracts, copper slag is not environmentally hazardous waste. It is commonly used as road aggregate or abrasive for cleaning metal parts (Gambal, 2013: 5). The release of heavy metals (Cu, Zn, Pb) from spent copper slag used as an abrasive for material cleaning was analysed by Rzechuła (1994: 212). Water extracts based on the test slag had the highest concentration of Cu, Zn and Pb from all the studied wastes. The increased heavy metal content in water extract was partly a result of the treatment process, during which slag was ground and enriched with paint coating and, to some extent, with the material treated.

On the basis of own research and literature data, can be observed that water extracts prepared from various copper slags contained low concentrations of heavy metals. This indicates that the tested slags characterised by low mobility of heavy metals into the environment. It is different in the case of slag from zinc metallurgy. Unprocessed slag during storage may release significant amounts of heavy metals into the soil and water.

5. Conclusion

A comprehensive approach to waste, including economic and ecologic aspects as well as social conditions, is the basis for the sustainable metallurgical waste management. One should bear in mind that waste has a measurable material value. Due to the depleting natural resources, the efficient waste management requires to treat waste as valuable secondary raw materials. Entrepreneurs should strive to reduce the amount of waste-to-landfill as the waste disposal is the final method of its utilisation. Metallurgical slag is the waste the largest amounts of which are produced by the metallurgical industry. The actions in the field of environmental protection have forced entrepreneurs to search for new methods of waste management, e.g. by reusing it the construction industry and road engineering. However, if it is impossible to reuse waste due to the high level of contamination, e.g. with heavy metals, it should be stored in a way that is safe for human health and the environment. Therefore, entrepreneurs are obliged to carry out contaminant leachability tests before the disposal of waste in the environment. The tests on the release of heavy metal from waste are an important aspect of waste management. It is possible to assess the potential effect of waste on the soil and water environment on the basis of tests on the mobility of contaminants. The research results presented in the paper show various concentration values of heavy metals depending on the source of the waste, structure and type of used technology. The highest leachability of heavy metals was determined for slag from zinc metallurgy. This waste can be hazardous for the environment. The copper slag characterised by low leachability of heavy metals. A low release rate of heavy metals to the aqueous phase makes it possible to reuse such waste usable for the production of aggregates or construction materials.

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Analiza mobilności metali ciężkich z odpadów hutniczych w aspekcie zrównoważonej gospodarki odpadami

Streszczenie

Hutnictwo jest jedną z gałęzi przemysłu wytwarzającą najbardziej zróżnicowaną gamę odpadów pod kątem ich składu, jak i stopnia zanieczyszczenia metalami ciężkimi. Wraz z rozwojem gospodarki wzrasta masa wytwarzanych odpadów przemysłowych, co wiąże się z poszukiwaniem nowych sposobów ich zagospodarowania. Jednym z priorytetów w działalności przedsiębiorstw jest stosowanie zasad zrównoważonego rozwoju. Nieustannie poszukuje się nowych rozwiązań technologicznych mających na celu zwiększenie ochrony środowiska przy jednoczesnym wzroście ponownego wykorzystania odpadów. Działanie takie jest podstawą zrównoważonej gospodarki odpadami. Niestety nie jest możliwe gospodarcze wykorzystanie wszystkich odpadów. Wpływ na to ma między innymi stopień ich zanieczyszczenia oraz ryzyko skażenia środowiska poprzez nieodpowiednie unieszkodliwianie odpadów. W celu zwiększenia ochrony środowiska naturalnego przed niekontrolowanym oddziaływaniem odpadów wykonuje się testy wymywalności zanieczyszczeń. Praktyka ta jest nieodzownym działaniem każdego przedsiębiorstwa wytwarzającego odpady mogące mieć negatywny wpływ na środowisko, zwłaszcza środowisko wodne. W pracy omówiono aspekty zrównoważonego rozwoju w gospodarce odpadami hutniczymi. Przedstawiono wyniki stężeń metali ciężkich z żużli pochodzących z hutnictwa cynku i miedzi w celu oceny mobilności zanieczyszczeń do środowiska. Przeanalizowano także poziomy wymywania metali ciężkich z odpadów pochodzących z różnych gałęzi hutnictwa.

***Słowa kluczowe**:* zrównoważona gospodarka odpadami, odpady hutnicze, mobilność metali ciężkich