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Pro-ecological possibilities of using metallurgical waste in the production of aggregates

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Abstract
Waste management is a very important issue for the sustainable development of the modern world. The metallurgical industry is an industry that has been generating and still generates large amounts of waste that may have a negative impact on the natural environment and human health. Metallurgical waste comes from current production and is collected in landfills/heaps. Any research enabling the management of waste, including metallurgical waste, is justified. This study presents the results of research on waste that can be used in the production of aggregates – research related to natural radioactivity and the introduction of hazardous substances into water or soil. The study highlights the diversified chemical composition of metallurgical waste, which requires detailed research of the waste before it is directed to the production of aggregates. Aggregate, as a building material, is subject to specific legal (normative) regulations. Metallurgical waste that meets the requirements for the protection of the natural environment and human health should be used for the production of building materials - it is an environmentally friendly activity that implements the principles of sustainable development.

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1. Introduction

The main goal of waste management should be to recover valuable raw materials from them or manage waste in other areas, and thus protect the natural environment by reducing the use of deposits and energy.

The substances referred to as "waste" are strictly regulated in order to protect human health and the environment. Decisive influence on the choice of waste management technology are not only economic aspects, but also aspects of environmental protection. Due to the possibility of using waste as raw materials in specific production processes, its storage is avoided, and it also creates the possibility of liquidating the existing landfills, heaps.

The raw materials policy of the European Union in the document "Raw Materials Initiative" as one of the pillars predicts the maximum use of recyclable materials in ecological technologies to produce products of a quality that is not inferior to those made of natural resources. This approach is also dominant in the Europe 2020 strategy, which replaced the Lisbon Strategy. One of the three priorities proposed in this strategy is sustainable growth, which consists in supporting the resource-efficient economy, the so-called "green growth". A number of European Union countries have developed their own documents based on the Europe 2020 document, taking into account the use of deposits, international cooperation to obtain raw materials and the management of recyclable materials obtained from waste. According to the new Directive 2018/851/EU amending Directive 2008/98/EC on waste, waste management in the Union must be improved and transformed into a sustainable material management. It should include, inter alia, preparation of materials for reuse, recycling, reprocessing of waste into secondary raw materials for engineering purposes in the construction of roads or other infrastructure.

The metallurgical industry is an industry in which, in addition to mining and energy, large amounts of waste are generated. More and more attention is paid to the need to manage metallurgical waste due to the protection of the natural environment and economic aspects (Jursowa, 2010; Yang, 2012; Xu, 2006; Ilutiu-Varvara, 2016; Matinde, 2018; Soultana, 2019), incl. by conducting research on the possibility of using waste in production, thus protecting natural deposits and reducing energy consumption.

This study presents proposals for the management of the waste collected in a heap and from current production – the waste with a maximum iron content of 5%.

2. Secondary raw materials for the production of aggregate

The package of standards for aggregates (EN 12620:2002+A1:2008; EN 13043:2013; EN 13055:2016; EN 13139:2013; EN 13242:2013; EN 13383-1:2013; EN 13450:2013) provides, inter alia, breakdown of aggregates. In 2010-2011, the CEN Committee TC 154 introduced a more precise division of aggregates. The breakdown is in line with CEN/TC154/TG10/ N736 Recyclable materials. Artificial aggregates. Final report for aggregates from secondary deposits. The particular types of aggregates are defined as follows:

- natural aggregates, these are aggregates of mineral origin that, apart from mechanical processing, have not undergone any other treatment;
- artificial aggregates, these are aggregates of mineral origin obtained as a result of an industrial process involving thermal or other modification;
- recycled aggregates, these are aggregates that have been created as a result of the processing of nonorganic material previously used in construction;
- secondary aggregates, these are aggregates from secondary raw materials (products) produced in industrial processes.

According to the above-mentioned document, for the iron and steel industry and the non-ferrous metals industry as secondary materials – the following artificial aggregates were listed, (Góralczyk, 2011):

- steel slag (subtype D1),
- crystalline blast furnace slag – air cooled (subtype D2),
- glassy blast furnace slag (subtype D3),
- electric arc furnace slag (subtype D4),
- stainless steel waste (subtype D5),
- copper, crystalline and granular slag (subtype E1).

Alternative aggregates (artificial, recycled) supplement the natural aggregates market. In some European Union countries, the production of alternative aggregates is at the level of 15-25% of the total aggregate production (Great Britain, the Netherlands, Belgium, Germany) (Kozioł, 2016).

In places where there were steel mills in the past, piles of waste materials in the form of heaps remained after the production of these enterprises. Mostly these are slags from old metallurgical processes, mainly blast furnace and steel slags, e.g. open hearth. In such landfills, in addition to slags, there are ceramic materials, which include refractory bricks, construction debris from renovations. The proportions in which these materials were mixed are not constant and vary depending on what work was carried out at the steelworks at that time. Nevertheless, many metallurgical heaps should qualify for crushing and use as aggregate for road construction. It is worth mentioning that the mineral material from old landfills is seasoned and does not require any stabilisation treatments. Aggregates produced from the slag from the current production, as opposed to the slag from the heap, must be aged in a stabilisation landfill for a period of at least 6 months.

3. Research

This study presents the results of tests of material collected from the heap of a non-functioning ironworks (A) and a working metal foundry (B), tests for the content of hazardous substances in the field of natural radioactivity and harmful substances introduced into water or soil.

3.1. Natural radioactivity

The determination of the concentration of natural radioactive elements in the tested materials allows for the calculation of the following activity indexes:

- f1 – determines the content of natural radioactive isotopes (total gamma radiation): potassium K-40, radium Ra-226 and thorium T-228;
- f2 – indirectly determines the amount of Rn-222 gaseous radon emission and compares the results with the standard requirements.

The values of the activity indicators f1 and f2 for building materials, including aggregates, used in facilities intended for the permanent stay of people and livestock may not exceed by more than 20% f1 = 1 and f2 = 200 Bq/kg (Dohojda, 2009; Regulation (EU) No 305/2011).

The conducted radioactivity tests showed that the tested secondary materials, both from the heap and the operating foundry, meet the criteria for qualification coefficients: $f1 \leq 1$ and $f2 \geq 185$ Bq/kg.

3.2. Hazardous substances discharged into water or soil

In the European Union, directives are the basic legal acts to which the member states must comply. Pursuant to Directive 2003/33/EC, the tested waste was assessed in terms of the conditions that it must meet in the case of generating sewage into water or soil and the content of substances particularly harmful to the aquatic environment (Directive 91/271/EEC). The tests were carried out in laboratories accredited by the Polish Centre for Accreditation AB213 and AB146 in the field of collecting and testing waste samples.

The compliance test (water extract) was prepared in accordance with the requirements of the Polish standard PN-EN 1744-3:2004. Part 3 introducing the European standard EN 1733-3:2002. This European standard specifies a method for the preparation of eluates, by leaching of aggregates, for subsequent investigation of physical and chemical properties by existing standard methods for the purpose of compliance testing. It applies to unbound aggregates that have a particle size below 32 mm with or without size reduction.

Water extracts of these wastes were tested for determination of arsenic, barium, cadmium, chromium, copper, molybdenum, nickel, lead, antimony, zinc, mercury, chloride, fluoride, sulfate, alkalinity (ISO 11969:1999; ISO 11885:2007; EN 1483:2007; ISO 10304-1:2007; ISO 10523:2008). The obtained results were compared with the maximum permissible values of pollutants that can be introduced into the aquatic environment (Table 1).

Table 1. The values of pollution indicators of substances particularly harmful to the aquatic environment in water extracts of the tested waste (values for industrial wastewater, mg/l) – according with the directive 91/271/EEC with the changes.

Pollution	Results		The highest limit	Designation according to the standard
	A	B		
Arsenic	< 0.0010	0.005	0.1	15
Barium	0.0531	0.090	2	16
Cadmium	< 0.0005	0.015	0.4	16
Chromium total	0.0038	**6.932**	0.5	16
Copper	< 0.0040	0.037	0.5	16
Mercury	0.00095	0.003	0.06	17
Molybdenum	0.0665	0.019	1	16
Nickel	0.0046	0.088	0.5	16
Lead	0.010	0.045	0.5	16
Antimony	< 0.050	<0.050	0.3	16
Zinc	0.0398	0.098	2	16
Chloride	8.7	1.9	1000	18
Fluoride	0.94	0.88	25	18
Sulfate	50	14.1	500	18
pH	7.3	**4.2**	6.5-9.0	19

Values that do not meet the permissible pollution indicators have been marked in bold

4. Discussion of the results

The tested secondary materials in terms of natural radioactivity meet the criteria for qualification factors, which means that they can be used for the production of aggregates used in both residential and road construction.

In accordance with the requirements of the European Union (Directive 91/271/EEC), substances particularly harmful to the aquatic environment, causing water pollution, have been divided into two categories – substances that should be eliminated and substances that should be limited. In water extracts of tested waste, substances that should be eliminated include mercury and its compounds as well as cadmium and its compounds.

For the waste collected from the heap (A), the mercury content in the water extract is 0.00095 mg/l, and cadmium < 0.0005 mg/l and these values are significantly lower than the highest limit set for these elements, therefore they should not represent an obstacle in reusing these materials.

For the waste produced on an ongoing basis in the metal foundry (B), the mercury content in the water extract is 0.003 mg/l, and cadmium 0.015 mg/l. These values are lower than the highest limit for these elements, but higher than the values in the heap waste. In addition, in the waste from the current production of metal foundries, the content of total chromium is exceeded, which is 6.932 mg/l with the maximum content allowed of 0.5 mg/l, and the water extract has an acidic pH = 4.2 (recommended pH 6.5-9.0).

Both tested waste materials contain fluorides, which are classified as substances particularly harmful to the aquatic environment, causing water pollution, which should be limited. he fluoride content in both analysed wastes is similar: A-0.94 mg/l and B-0.88 mg/l with the highest limit being 25 mg/l. The content of fluorides on the above-mentioned level should not be an obstacle to the re-use of this waste.

Before making a decision to use waste materials for the production of aggregates, tests of the given waste should be carried out in terms of its physical and chemical properties, including the impact of this waste on the natural environment and human health.

The research results presented in the study show that the tested waste collected in the heap of a non-operating ironworks meets the requirements related to the protection of the natural environment and human health, because in water extracts the highest limit was not exceeded for any element or chemical compound, and the radioactive activity indicators meet criteria for qualifying coefficients.

The waste material from the current production of the metal foundry does not meet the requirements with regard to the protection of the natural environment and human health, despite the fact that the radioactive activity indicators meet the criteria of qualification indicators. However, in water extracts, the highest limit for chromium total was exceeded and the extract was acidic, pH = 4.2.

5. Possibilities of using waste

Metallurgical waste can be used for the production of building materials (Millosan, 2013; Suvorova 2020), including as a substitute for natural aggregates (Pizoń, 2020). This is doubly beneficial – the waste is recycled, which protects the deposits of natural resources from over-exploitation. Waste is collected in landfills occupying large areas, waste recycling causes the liquidation of existing landfills. In this way, the principles of sustainable development and pro-ecological activities are implemented, as well as the commissioning of landfill sites for development.

Metallurgical waste (slags) can also be used for the production of polyester-polymer concretes that can replace conventional concretes (Seco, 2020) and as an input filling concrete instead of natural gravel (Baricova, 2011). Research was also conducted on the use of metallurgical waste as a sand substitute for cement production, a positive effect on the initial setting time and the release of heat of hydration was noted (Alwaeli, 2020). Metallurgical waste (steel dust) can be used in the non-metallurgical industry – the production of glass and ceramic products (Lis, 2012).

The chemical composition of metallurgical waste, both those deposited and from current production, depends on the metallurgical processes in which they were generated and may affect the natural environment (Wowkonowicz, 2018). The results of the research on the content of elements in various slags (Pitak, 2015) indicate that selected substances are leached from some slags, and their concentrations in the leachate are correlated with their content in the slags. The diversified chemical composition of waste indicates the need for individual examination of dumps/heaps (Jonczy, 2014). Such an approach will allow for the elimination of possible environmental hazards related to the use of this waste.

Waste materials are used more and more frequently in road construction. Thanks to their properties, they are an ideal foundation for roads and motorways. They add stability to the soil, thus improving the performance and life of the asphalt surfaces themselves. Metallurgical slags can also be used as a bonding element for bituminous mass, thus improving road adhesion (https://poradnik.pkt.pl).

Slag aggregates from old heaps are characterised by a lower specific weight than natural aggregates. In addition, they are characterised by high fire resistance and high insulating properties. Many aggregates of this type are porous, which makes them absorb more water than natural materials. Due to its porosity, this type of aggregates have a very good frost resistance, because the pores are not connected to each other (https://poradnik.pkt.pl).

The production of alternative aggregates from waste and accompanying raw materials is the proper implementation of the basic objectives and principles of waste management, which are: limiting the consumption of natural resources, reducing landfill sites, reducing or eliminating the costs associated with waste storage and/or the purchase of raw materials for production. n terms of strength, artificial (alternative) aggregates are comparable to natural aggregates. Well composed, they meet the quality and ecological requirements, and do not pollute the soil and groundwater. Due to these properties, such aggregates are used in road construction (construction of roads, motorways, hard shoulders, car parks, etc.) and in railway construction.

The advantages of steel slag aggregates include: high strength, high frost resistance, low abrasion, low water absorption, high quality, competitive price (https://harscometals.pl).

6. Conclusion

From July 1, 2013, in the European Union, the Regulation No. 305/2011 of the European Parliament and of the Council of March 9, 2011 applies, abbreviated as CPR from the Construction Products Regulation establishing harmonised conditions for the marketing of construction products [39].

The regulations of the member states require that the facilities built are constructed in a way that ensures the safety of people and domestic animals and that they do not have a harmful effect on the environment. Each member state had different requirements for construction products, which generated obstacles to trade within the Union. Therefore, member states have been obliged to introduce national regulations relating to essential construction characteristics in accordance with harmonised technical specifications.

Due to the above, the producers of building materials were obliged to perform internal production control ensuring the constant quality of production (product). The chemical composition of waste collected in landfills and generated from current production is not constant, it depends both on the process generating the waste and other works carried out at that time. The varied chemical composition of waste materials can be a serious obstacle in maintaining a constant quality of the aggregate produced from such waste. This means that the waste, before it is used for the production of aggregate, should be thoroughly tested and identified. This mainly applies to type testing, taking into account the presence of hazardous substances, including the determination of radioactivity and harmful substances released into water or soil.

Based on own research and literature, it should be concluded that the use of metallurgical waste collected in dumps/heaps for the production of building materials, including aggregates, is safe, after appropriate research, for the natural environment and human health. The use of metallurgical waste for the production of various building materials implements the principles of sustainable development and is pro-ecological.

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在骨料生产中使用冶金废料的生态可能性

關鍵詞
冶金废料
环境的
有害物质
化学元素
骨料

摘要
废物管理是现代世界可持续发展的一个非常重要的问题。冶金行业是一个一直在产生并且仍在产生大量废物的行业，这些废物可能对自然环境和人类健康产生负面影响。冶金废料来自当前的生产，并在垃圾填埋场/堆中收集。任何能够管理废物（包括冶金废物）的研究都是合理的。本研究介绍了可用于生产骨料的废物的研究结果——与天然放射性和将有害物质引入水或土壤有关的研究。该研究强调了冶金废料的多样化化学成分，需要在将废料用于骨料生产之前对其进行详细研究。骨料作为一种建筑材料，受特定的法律（规范）法规的约束。符合保护自然环境和人类健康要求的冶金废料应用于生产建筑材料——这是一种贯彻可持续发展原则的环保活动