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Effect of Ash from Biomass Combustion on Tailings pH †

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Abstract: This article deals with the use of ash from biomass burning for the remediation of thermally active dump in Heřmanice, Czech Republic. Nowadays, various chemical, physical, or biological methods of remediation are used for the remediation of dumps. The authors discuss the complex use of ash from biomass as a possibility for biological remediation of the Heřmanice dump. The main advantage of obtaining ash when burning biomass is primarily the fact that it is a renewable energy source, which produces electricity and large amounts of ash, which can be used, for example, for the remediation of the Heřmanice dump. Tailings are characterized by their acidity, while fly ash is characterized by high alkalinity. This study deals with which ratio (tailings:ash) would achieve the necessary neutral values in order to prevent the release of heavy metals into the surroundings of the Heřmanice dump. The value of the active soil reaction (pH/H_2O), the value of the exchange soil reaction (pH/$CaCl_2$), the value of hydrolytic acidity (H_a), together with the value of soluble salts in the tailings, i.e., electrical conductivity, were also studied. Based on the obtained results, it can be concluded that the addition of biomass combustion ash had a positive effect on the pH value of the tailings. Based on this fact, Al^{3+} is excreted more slowly into the environment. A higher content of aluminum is toxic to plants, while in a smaller amount, its content is necessary and at the same time an important factor in the process of plant growth. The mixing of alkaline ash with unburned surface tailings from the thermally active Heřmanice dump significantly influenced its acidity, which had a positive effect on increasing the active and exchange acidity.

Keywords: thermally active dumps; tailing; biomass combustion ash

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1. Introduction

During the deep mining of black coal and its subsequent processing, waste is generated, which used to be deposited in dumps. This waste consisted of carboniferous rocks originating from mining, preparatory works, shaft sinking, roof rocks (sandstones, shales, conglomerates) coated with coal substance, waste material produced during coal washing, and coal slurry [1–3]. Carboniferous rocks are characterized by the presence of finely dispersed coal substance, which is very difficult to separate using conventional beneficiation methods. It is precisely this characteristic of carboniferous rocks that is the reason why the material deposited in the Ostrava-Karviná region (OKR), Czech Republic, contains a large amount of coal substance. The spoil heaps in the OKR were not covered or protected, and therefore they were exposed to mechanical and chemical weathering. Thermal processes, triggered by the oxidation or combustion of coal substance or other combustible materials (wood residues, plastics, etc.) deposited on the spoil heaps for a relatively long period, were a common occurrence. Endogenous combustion is still taking place in some spoil heaps. Currently, in the Czech Republic, within the OKR territory, there are three such thermally active spoil heaps of black coal waste: Heřmanice, Hedvika, and Ema, the latter of which can already be classified as a burning landfill [4,5].

The thermal activity of the dump causes the release of harmful gases into the surroundings and also disrupts the stability of the spoil heap structure (posing a risk of sinkholes forming in areas of hot spots) [6]. Residues of pyrite, which is present in the spoil material, can be oxidized by bacteria, particularly the *Acidithiobacillus ferrooxidans* genus [7–9]. Sulfuric acid, formed during this process of pyrite oxidation, is the reason for the acidification of the spoil. Due to the very low content of basic rocks (particularly carbonates) in the spoil of the Ostrava-Karviná region (OKR), the neutralization of the resulting acids occurs very slowly. As a result of the acidity of the spoil, there can be leaching of hazardous metals from the spoil heap into the immediate surroundings [2,4]. However, the main problem affecting the immediate surroundings of the spoil heap is the ongoing thermal activity, which is closely associated with gas emissions, potential suspension of dust particles, and the presence of hazardous elements that may be present in the deposited material. Thermally active spoil heaps can thus represent a significant ecological burden [10,11].

The remediation of such a dump and its surroundings is necessary to minimize the negative impacts of the thermal process, not only on the environment but also on the health of people living in the immediate vicinity. Currently, physical, chemical, and biological methods are used for the remediation of dumps, each with its advantages and disadvantages (economic costs, inefficiency, or negative impacts on the surrounding environment). During the 1990s, mining activities gradually declined, resulting in a significant reduction in the amount of mining waste (tailing). Nowadays, tailing is considered a useful material for further utilization. It finds application in construction (road construction) or as a material for terrain modifications (remediation, reclamation). However, the use of mining tailing as a construction material in road infrastructure comes with many challenges. Tailing can only be used in road embankments if it does not contain trace amounts of coal. A higher coal content in spoil can cause endogenous combustion and subsequent self-ignition of the road embankment. Tailing is also utilized in the reclamation of mining landscapes. For these purposes, it can be used regardless of its petrographic composition, grain size, or water content [1,2,4].

Within legislation and societal thinking, there is increasing pressure for the reutilization and reduction of disposed waste, especially through landfilling. The focus of discussion lies primarily in finding ways to transform waste, whether newly generated or stored in dumps, into resources that can be utilized in further development such as construction and reclamation. As mentioned earlier, the main issue with spoil material is its acidity, which poses a challenge for biological reclamation. One potential remediation method that could address the acidity issue of the spoil is the utilization of the alkaline properties of biomass ash resulting from biomass combustion. Our objective was to contribute to a more efficient and sustainable remediation of thermally active dump heaps through the use of biomass ash as an alternative material, as demonstrated by this pilot study.

2. Material and Methods

2.1. Characterization of Sampeling Site

The Heřmanice dump was formed during deep coal mining in the 19th and 20th centuries. It received tailing from the Ida deep mine and industrial waste from former mining companies and coking plants. The Heřmanice dump covers an area of approximately 103 hectares and is located within the cadastral territory of Slezská Ostrava in the municipal districts of Hrušov and Heřmanice, Czech Republic. Currently, the area consists of a reclaimed section of the dump in the west and remediated areas in the south. In the central part of the Heřmanice dump, there are three sludge ponds, the thermally active portion, and a secured landfill for chemical waste owned by Ostrava-Karviná Coke Plants, a joint-stock company. The amount of tailing imported to the storage area for mining waste is approximately 20 million cubic meters. The unaffected part of the tailing, still affected by thermal activity, contains approximately 12 million tons of spoil [12].

Over the past few years, a series of remediation interventions have been carried out in the area. The first remediation works took place in the late 1990s when a more extensive thermal process was observed. A reduction of thermal activity was attempted by layering ash combined with clay soil on the surface of the thermally active area. However, this attempt proved to be ineffective. Subsequent efforts were made but also proved unsuccessful, resulting in the removal of all vegetation from the ridge of the thermally active dump. The most effective method to prevent thermal activity has been determined to be the removal of the mining tailing. However, thermal processes are unpredictable, and it cannot be guaranteed that complete cessation of activity will occur [13].

2.2. Methodology of Sample Collection and Preparation

Samples of the tailing were collected from the thermally active part of the Heřmanice dump. The sampling locations were chosen to be as representative as possible of the spoil in that area. A total of 20 sampling points were selected, encompassing the sloping terrain and flat surfaces (see Figure 1). Initially, the top layer of the dump, approximately 10–20 cm thick, which was not part of the collected sample, was removed. The sampling was conducted from a depth of 20–50 cm. After cooling down, the spoil samples from all sampling points were homogenized to create a composite sample, which was stored in a sealable plastic container.

Figure 1. Sampling locations at the Heřmanice dump.

The tailing samples were mechanically processed in the laboratory using a jaw crusher, Retsch type BB200 WC (Haan, Germany). After homogenization, the tailing was further size-reduced using a Retsch jaw crusher, type BB200 WC (Haan, Germany), to approximately 2 mm. Subsequently, the samples were dried to a constant weight in a vacuum dryer, model VO29 MEMMERT (Schwabach, Germany). The dried tailing samples were stored in a desiccator.

The sample of grate ash from biomass combustion of plant origin was obtained from an industrial boiler in a thermal power plant. At the time of sampling, wood chips were being burned. The ash was stored in a sealable plastic container with a lid to prevent air and moisture infiltration. The grate ash was size-reduced to a particle size of 2 mm using a Retsch jaw crusher, type BB200 WC (Haan, Germany). The dried ash samples were stored in a desiccator.

The samples of ash and tailing were blended in ratios of 1:6, 1:7, and 1:8, ensuring an excess of the tailing sample.

2.3. Determination of Monitored Parameters

The active acidity (pH/H_2O) and passive acidity (pH/$CaCl_2$) were determined following the ČSN EN ISO 10390 (836221) [14]. The measurements were performed using an inoLab® pH 7110 laboratory pH meter from Xylem Analytics Germany Sales GmbH (Weilheim, Germany). To assess the active and passive acidity, criteria specified in the Table 1 were utilized.

Table 1. Criteria for assessing active and passive acidity [15].

pH/H_2O	Interpretation	pH/$CaCl_2$	Interpretation
<5.4	Strongly acidic	<4.8	Strongly acidic
5.5–6.9	Moderately acidic	4.8–5.2	Moderately high acidic
6.5–6.9	Slightly acidic	5.2–5.5	Moderately acidic
7.0	Neutral	5.5–7.5	Moderately acidic to sightly alkaline
7.1–7.5	Slightly alkaline	>7.5	Moderately to strongly alkaline
7.6–8.3	Moderately alkaline	-	-
>8.4	Strongly alkaline	-	-

The determination of soluble salts based on electrical conductivity (EC) was carried out according to ČSN ISO 11265 (836210) [16] using the WTW Multi 3320 instrument (WTW Germany, Weilheim, Germany). Water-soluble electrolytes were extracted from the soil in a ratio of soil to water of 1:5, and their concentration was determined based on the increase in specific electrical conductivity of the filtered extract. The criteria for assessing electrical conductivity were applied as presented in Table 2.

Table 2. Criteria for assessing active and passive acidity [17].

Electrical Conductivity mS cm^{-1}	Soils
<0.70	Non-saline
0.71–1.20	Moderately saline
>1.20	Saline

The chemical composition of the samples was determined semi-quantitatively by X-ray fluorescence on the XEPOS (Spectro, Kleve, Germany) energy dispersion spectrometer. After trituration, the samples were placed in a plastic cuvette with a Mylar protective foil and then analyzed in a protective atmosphere (He). The phase composition and microstructural properties were determined using the X-ray powder diffraction (XRD) technique. XRD patterns were obtained using a Rigaku SmartLab diffractometer (Rigaku, Tokyo, Japan) with a D/teX Ultra 250 detector. The measured XRD patterns were evaluated using PDXL 2 software (version 2.4.2.0) and compared with the PDF-2 database, 2015 release (ICDD, Newton Square, Worcester, MA, USA).

3. Results and Discussion

From a legislative perspective, tailing is classified as mining waste and must be handled accordingly. In the Czech Republic, this matter is addressed by Act No. 157/2009 Coll., which establishes relevant regulations for the management of mining waste. The law also sets rules for mitigating the impacts on water, air, soil, plants, animals, and the landscape that may result from the handling of mining waste [18]. However, the law does not apply to mining waste generated during mineral deposit exploration, extraction, and processing. This exception also includes water containing substances resulting from the extraction of oil and minerals, as well as waste generated during mineral deposit exploration and mining activities not directly related to these operations. The mentioned law also does not apply to materials obtained during the extraction and processing of non-metallic minerals, which are used for reclamation and remediation, or for the stabilization and disposal of mining works. The aforementioned law is supplemented by Decree No. 428/2009 Coll. and No. 429/2009 Coll., which regulate the requirements for a mining waste management plan, as specified in Decree No. 281/2018 Coll. These decrees fall within the framework of environmental protection regulations and aim to minimize the risk of soil and water pollution and improve the quality of the environment [19–21]. Additionally, within the Waste Catalog, specifically in the Decree No. 8/2021 Coll. of the Ministry of the Environment of the Czech Republic, waste from mining activities is classified in Group 01, which includes all waste from geological exploration, mining, processing, and further treatment of minerals and stones [22]. Under Czech legislation, waste generated from mining activities and activities carried out by mining methods, which are deposited in spoil heaps and tailings ponds, are not part of waste management under Act No. 541/2020 Coll., but are subject to Act No. 44/1988 Coll. (Mining Act) [23,24].

In 2001, the question of whether mining waste (tailing) should be considered as waste was discussed. After discussions among state authorities and mining organizations, mining waste was eventually recognized as a filling material for reclamation projects, such as spoil heaps, embankments, or road construction, according to Act No. 22/1997 Coll. As a result, mining waste was removed from the category of waste and is now only utilized with the permission of a certificate and conformity confirmation [25]. These certificates are regularly renewed through tests conducted in accredited laboratories, and individual mines perform regular sampling and analysis of the mining waste to compare its parameters with those stated in the certificate. Currently, mining waste can be used for road construction, but without endangering the environment. From an environmental protection perspective, the material can be considered suitable and non-threatening if the concentrations of leaching substances do not exceed the values specified in Appendix C of the ČSN 73 6133 standard [26]. When using mining waste for reclamation and remediation, it is necessary to adhere to the rules outlined in Decree No. 428/2009 Coll., which also regulates the requirements that must be included in the project documentation for remediation [19].

The tailing from thermally active dumps can also contain various hazardous organic substances, such as polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and others. These substances can be a source of contamination in the immediate surroundings, and therefore their remediation is necessary [10]. The procedures and methods for removing these hazardous substances from soil and tailing are specified in Decree No. 429/2009 Coll., which is regulated in terms of the requirements for the waste management plan by Decree No. 281/2018 Coll.

Knowledge of the mineralogical–petrographic characteristics of tailing is crucial for assessing its potential impact on the environment. The petrographic composition of tailing in the Ostrava-Karviná region (Czech Republic) is practically identical among individual dumps. The most prevalent rock types are various types of aleuropelites, accompanied by fine-grained and medium to coarse-grained sandstones in significant and variable quantities, which are more resistant to weathering. Calcite is rare in the rocks of the Ostrava-Karviná region. Small amounts of clay minerals (such as mixed illite–smectite structures) may exhibit swelling when in contact with water. Based on the elemental analysis (see Table 3), it can be observed that elements such as Si (27%), Fe (25%), and Al (11%) were predominant in the tailing. Pertile et al. state that considering the percentage of silicon, it can be assumed that acidification will not occur due to its loss, but rather due to the deficiency of basic cations during the decomposition of aluminosilicates [2]. The sulfur content in the dry coal samples from the Ostrava-Karviná region is generally low. Pešek et al. report that the total sulfur values vary between 0.4% and 4.8% with an average of 2.1% across different layers and areas [3]. This was confirmed by the sample of tailing from the Heřmanice dump, which had a total sulfur content of around 3.5%. Generally, the content of heavy metals in the carbonaceous rocks of the Ostrava-Karviná region is low and does not exceed the background levels of other industrial emissions from an ecological perspective. The composition of ash from biomass combustion depends on various factors, with the main ones being the origin of the biomass and the method of combustion.

Table 3. Elemental analysis of tailings and cyclone ash samples using XRF method in % [2].

%	Tailing	Ash	%	Tailing	Ash	%	Tailing	Ash	%	Tailing	Ash
Na	<0.01	<0.01	Mo	0.001	0.001	Pr	<0.0002	0.005	Te	<0.0003	<0.0003
Ag	<0.0002	<0.0002	Nb	0.005	0.0001	Cs	0.014	<0.0004	Sr	0.053	0.09
Al	**11**	0.82	Nd	0.029	0.01	Cu	0.03	0.007	Ta	0.01	0.008
As	0.02	0.0001	Ni	0.03	0.01	Fe	**25**	1.2	Sn	0.0008	0.0001
Ba	0.68	0.15	P	0.3	2.3	Ga	0.009	0.001	Th	0.008	0.001
Bi	<0.0001	<0.0001	Pb	0.03	0.001	Ge	0.0002	0.0002	Ti	1.3	0.13
Br	0.008	0.004	Cr	0.04	0.005	Hf	0.002	0.0001	Tl	0.0004	0.0001
Ca	0.68	**20**	Rb	0.054	0.02	Hg	0.006	<0.0001	U	0.002	<0.0001
Cd	0.0005	0.0001	S	3.5	2.3	I	0.06	<0.0003	V	0.05	0.002
Ce	0.01	0.02	Sb	0.0004	<0.0003	K	**5.5**	**25**	W	0.002	<0.0001
Cl	0.49	1.3	Se	0.0006	0.0001	La	0.01	0.01	Y	0.01	0.001
Co	0.005	0.0003	Si	**27**	**28**	Mg	0.2	1.4	Zn	0.05	0.03

Note: Values of elements exceeding 5% are highlighted in bold.

During the biomass combustion process, organic matter decomposes into various components, including carbon oxides, water vapor, nitrogen oxides, sulfur oxides, and other gases, which are then emitted from the combustion process. Ash is formed, which contains nutrients and micronutrients. For example, ash from wood combustion is rich in calcium, while ash from straw and grains is rich in potassium. Magnesium, calcium, and potassium are present in the form of carbonates, which are formed during the mineralization of organic compounds by the conversion of cations to oxides, which then hydrate and convert to carbonates under atmospheric conditions [27]. The nitrogen content in ash is very low and practically negligible because at higher temperatures during the combustion process, it transforms into oxides and escapes into the atmosphere. Risky metals can also be present in ash, but their occurrence is closely related to the origin of the burned biomass and the temperature during combustion. Generally, ash from biomass combustion exhibits alkaline properties.

The high content of alkaline metals and their form are the main factors contributing to these properties. The pH value of biomass ash increases primarily due to the loss of organic acids during the combustion process and the formation of soluble oxides, hydroxides, carbonates, and bicarbonates of calcium, magnesium, potassium, and sodium. The pH value is further influenced by the combustion temperature and storage time, with alkalinity decreasing as both factors increase. During prolonged storage, the hydroxides in the ash can convert to carbonates. For example, ash from woody biomass generally has a higher pH value due to a higher calcium content and lower sulfur content compared to ash from straw and grains [28]. Based on elemental analysis, the bottom ash from biomass combustion also exhibits low levels of heavy metals. The silicon content in bottom ash (28%) is practically the same as in the original material (27%).

The samples of ash and overburden were mixed in ratios of 1:6, 1:7, and 1:8 to achieve a pH value within the neutral range of 6.1–7.0, considering the research objectives. One of the parameters monitored in the samples was their active acidity, which is determined by hydrogen ions present in the solution. These hydrogen ions originate from dissociated mineral and organic acids. The values of active acidity (pH/H_2O) provide information about the current state of acidity and alkalinity of the material [29]. Passive acidity ($pH/CaCl_2$) is formed by adsorbed H^+ and Al^{3+} (Fe^{3+}) ions, which can be released into the solution through the exchange with basic cations from neutral salt solutions in the soil. Passive acidity is considered a more important and commonly used indicator for evaluating acidity, as it undergoes less significant changes compared to active acidity over time. The values of passive acidity are generally lower than active acidity, ranging from a pH difference of 0.2–1.0 [30]. If the mixture of overburden and ash is to be used in the biological remediation of the dump, particularly in combination with the importation of soil substrate, the active acidity becomes significant as it fundamentally affects the biochemical processes occurring in the soil and the nutrient uptake processes by autotrophic organisms [15,30,31]. The values of active and passive acidity, along with the evaluation, are presented in Table 4.

Table 4. Active and passive acidity in the studied samples.

Sample	Active Acidity		Passive Acidity	
	pH/H_2O	Interpretation	pH/$CaCl_2$	Interpretation
Mixture of Ash/Tailing 1:6	8.2	Moderately alkaline	6.7	Moderately acidic to slightly alkaline
Mixture of Ash/Tailing 1:7	7.1	Slightly alkaline	5.8	Moderately acidic to slightly alkaline
Mixture of Ash/Tailing 1:8	6.6	Slightly acidic	5.7	Moderately acidic to slightly alkaline
Tailing	4.5	Strongly acidic	4.6	Strongly acidic
Ash	11.2	Strongly alkaline	10.2	Moderately to strongly alkaline

Acidic tailings can limit microbial activity, reduce the availability of essential nutrients, and cause aluminum formation in the subsurface, slowing root growth and limiting access to water and nutrients [15]. By combining strongly alkaline grate ash with strongly acidic tailings, the expected significant modification of tailing acidity occurred. Based on the determined values of both active and passive acidity, the mixture of grate ash and tailings in a ratio of 1:6 can be considered the most suitable. It is a realistic assumption that the use of this mixture for remediation will be much more effective than if the grate ash was applied to the tailings in layers.

The evaluation of soluble salts in the soil based on electrical conductivity (EC) is used to determine the amount of salts in the samples. Excessive concentration of ions from any group of salts can lead to the weakening of plant growth or even their complete death. The values of electrical conductivity, along with the evaluation, are presented in Table 5.

Table 5. Values of specific conductivity of the studied material.

Sample	Specific Conductivity	
	mS cm^{-1}	Interpretation
Mixture of Ash/Tailing 1:6	3.5	Salty
Mixture of Ash/Tailing 1:7	3.0	Salty
Mixture of Ash/Tailing 1:8	3.0	Salty
Tailing	0.2	Unsalted
Ash	4.6	Salty

Based on the values of specific conductivity in all samples of the mixture as well as in the ash sample alone, it can be concluded that these are saline samples. Considering the composition of the ash, it primarily consists of calcium, silicon, and iron salts, which are not environmentally hazardous. Conversely, the content of calcium salts will contribute to increasing the pH value of the strongly acidic tailings.

4. Conclusions

The mixing of alkaline ash with unburned surface tailings from the thermally active Heřmanice dump significantly influenced its acidity, which had a positive effect on increasing the active and exchange acidity. The ash is also a rich source of various basic cations that can enrich the tailings. This can ultimately accelerate and positively impact the reclamation of landscapes affected by deep mining. The use of a mixture of tailings and ash will certainly be more effective in the remediation of the thermally active dump and its surroundings than the mere layering of these materials.

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