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Heavy Metals and Metalloids Leachability from Composite Ground Materials Peat – Fly Ash – Lime

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Abstract: This publication presents the test results of heavy metals and metalloids leachability from ground composite materials. The components of the obtained composites are peat and stabilizing binders in the form of fly ash and hydrated lime. The composites were designed to be used to stabilize low-bearing organic soil in the Lake Druzno basin in the area of Żuławy Elbląskie. The results of the leachability studies show that as the lime hydrated in the composition of the composite increases, the leachability of heavy metals and metalloids decreases. The decrease in the leachability of these elements is also influenced by the increase in the pH value. The results of heavy metals and metalloids leachability from composites as well as the obtained pH values make it possible to conclude that they are neutral to the ground environment and can be used in engineering practice under specific conditions in the area of Żuławy Elbląskie.

Keywords: Leachability, heavy metals, metalloids, peat, fly ash, hydrated lime, pH value

1. Introduction

Heavy metals play a major role in pollution and environmental degradation. The type, concentration and distribution of them in the soil depends on many natural factors such as granulometric composition, hydrogen ion content, organic matter content, soil type as well as geological and soil processes (Kończak-Konarkowska & Kuziak 2000). Environment is also affected by anthropogenic factors such as industrial contaminants and agrotechnical activities (Baran 2000, Kanakaraju et al. 2019, Skwaryło-Bednarz et al. 2014, Zorluer 2020).

Reinforcing the substrate with various stabilizers can increase the content of heavy metals in the ground. The ground composite material stabilizing the substrate should be selected so that it has established physical and mechani-

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cal properties. It is a common practice to use easily available materials in the vicinity of the construction site as components of composites. Cement, lime and gypsum, blast furnace slag, fly ash or mixtures of these materials are used as binders (Hayashi et al. 2005, Rathore et al. 2018, Timoney et al. 2012). This has a direct impact on the technologies of execution and, above all, on the economic effect (Afrin 2017, Al-Tabbaa 2005, Cortellazzo et al. 1999).

The purpose of the research is to select the optimal stabilizer in such a way that the resulting ground composite material has the assumed physical and mechanical properties.

The basic criterion for the applicability of composites should be their neutral environmental impact. This is particularly important in areas that are protected e.g., under EU or national programmes and in areas bordering national or landscape parks.

Important factors influencing solubility, adsorption and the presence of heavy metals in the porous solution are the content of hydrogen ions and organic matter. As the pH value decreases, their mobility increases (Wójcik 2009). Therefore, it is important to choose a stabilizer that will simultaneously stabilize the soil and increase its pH value (Jonczy et al. 2014, Kopańska & Dudziak 2015, Sybilski & Kraszewski 2004).

The purpose of the work is to assess the possibility of using ground composite materials, peat – fly ash – lime, in the basin of Lake Druzno in the area of Żuławy Elbląskie in terms of the content of heavy metals and metalloids and assessing their impact on environmental pollution.

The parameter assessing the degree of risk will be the leachability of heavy metals and metalloids from ground composite materials.

The phenomenon of leachability is the process by which soluble components change from solid material to liquid as a result of percolation or diffusion (Makowska et al. 2018).

2. Methods and materials

Composites made from the base material in the form of peat taken from the basin of Lake Druzno, Żuławy Elbląskie, and stabilizing materials in the form of fly ash and hydrated lime, were used for the study.

The selection of stabilizing binders was carried out on the basis of preliminary tests aimed at determining the amount of stabilizer in terms of binding properties and with a view to economic viability.

Peat (P) was taken by means of a drill from the depth of 0-2 m and additionally with mechanical equipment directly at the site of the excavation. The collected peat was mixed and then subjected to detailed laboratory tests.

A thermogravimetric method with the temperature range 105-650°C was used to determine the organic matter content of the base material. The formulas in PN-88/B-04481 were used to calculate natural humidity, density and porosity. The organic matter content was used to calculate the density of the ground skeleton using empirically established correlation relations (Borys 1993). The potentiometric method determined the concentration of hydrogen ions contained in the peat solution using an Elmetron CX-701 electronic pH meter. The degree of decomposition was determined on the Van Post scale (Post 1922). The results of the tests can be found in Table 1.

Table 1. Physical and chemical properties of peat and organic matter content

Physical and chemical properties	Humidity, W_n [%]	Bulk density, ρ [kg/m^3]	Specific density of the soil skeleton, ρ_s [kg/m^3]	Organic matter content, I_{OM} [%]	pH value	Porosity, n	Degree of decomposition, H
	350.0	$9 \cdot 10^2$	$1.8 \cdot 10^3$	62.9	6.5	0.88	H4-H5

Fly ash (FA) from the Elbląg Power Plant was used to make ground composite materials. It is obtained by gravity using fans blowing it out from under the furnace. Fly ash is formed by burning coal dust of type 32 MIIA in dust boilers. The tests of the chemical composition of fly ash carried out by means of the Thermo iCAP 6500 Duo ICP plasma spectrometer is shown in Table 2. The calculated specific density was 2298.32 kg/m^3 and the specific area was 364.78 m^2/kg. Le Chatelier method and Blaine method (Żygadło & Wozniak 2009) were used in the calculations, respectively. On the basis of the data obtained, the tested fly ash can be classified as silicate ash and the category A due to roasting losses of 3.71%. A radioactivity determination was also made with background radiation of 0.14 µSv/h. The ash showed 0.21 µSv/h, i.e., contains only trace quantities of radioactive elements.

Table 2. Chemical composition of fly ash

Ingredient	SiO_2	Fe_2O_3	Al_2O_3	Mn_3O_4	TiO_2	CaO	MgO	SO_3	P_2O_5	Na_2O	K_2O	BaO	SrO
Quantity [%]	56.5	6.9	18.6	0.12	0.86	4.74	2.81	0.48	0.41	0.72	3.08	0.16	0.07

Dry-slaked hydrated lime (HL), commercially named "Bielik", was also used to stabilize peat and is referred to as building lime EN 459-1 CL 90-S

(Karta charakterystyki 2018). Hydrated lime consists of CaO – min 92%, MgO – max 1%, SO_3 – max 0.5% and CO_2 – max 2.5%.

15 mixtures were designed, taking the ratio of fly ash mass to peat (FA/P) and hydrated lime to peat (HL/P) as shown in Table 3. The ingredients were mixed using a mechanical mixer which mixed the ground with the stabilizer for about two to three minutes to obtain a homogeneous mass. The mixed ingredients were placed in cubic forms measuring 150x150x150 mm. The formed samples were cured in a tightly closed container with access to water, where the relative vapour pressure p/po ≈ 1.

Table 3. Composition of ground composite, peat – fly ash – lime hydrate in mass ratio

Composite	FA/P	HL/P
C1	0.25	0.0125
C2	0.25	0.025
C3	0.25	0.0375
C4	0.25	0.05
C5	0.25	0.0625
C6	0.50	0.025
C7	0.50	0.05
C8	0.50	0.075
C9	0.50	0.1
C10	0.50	0.125
C11	0.75	0.0375
C12	0.75	0.075
C13	0.75	0.1125
C14	0.75	0.15
C15	0.75	0.1875

The test of heavy metals leachability from ground composite material and fly ash was carried out in the accredited Laboratory of Advanced Environmental Analysis of the Elbląg Technology Park.

The test material was taken from samples which were cured for 28 days under laboratory conditions in a sealed container with access to water, where the relative vapour pressure p/po ≈ 1.

According to the instruction, the preparation of the test water extract (RRM 2000) consisted in taking from each sample approximately 1 kg of composite which was dried at 105°C and then sifted through a standard woven sieve with square meshes No. 10. The sample of fly ash was taken and prepared in the same way. 0,1 kg samples were weighed from the sifted mass of composites and ash, placed in a flask and then flooded with distilled water in a mass ratio of 1:10. The tightly closed flask was shaken on the laboratory shaker for 4 hours. After 12 hours, the shaking process was repeated for 4 hours and then the suspension was left for 6 hours. The prepared suspension was filtered through a membrane filter with pores of 0.45 μm using a pressure-reduced filtration apparatus. The obtained (water extract) 10 ml of filtrate was mineralised with 7 ml of nitric acid and 3 ml of hydrogen peroxide. The process of mineralisation was carried out in a 1800 W Milestone Ethos One microwave mineraliser for 20 minutes at 205°C.

The concentration of the following metals and metalloids in the samples was determined: arsenic, cadmium, general chromium, copper, lead, molybdenum, nickel, vanadium, zinc, selenium, cobalt, iron, manganese. The test was performed in accordance with PN-EN ISO 17294-2:2016 by means of a mass spectrometer with inductively aroused plasma ICP-MS Agilent Technologies 7700x. Certified solutions and formulas conforming to the above standard were used during the test.

The prepared water extracts were also used to carry out tests on the hydrogen ion content by means of the Elmetron CX-701 pehameter with a measuring range of 3 to 20 pH and a measurement accuracy of up to 0.001 pH. The electrode used in the test was Elmetron EPP-1 designed to test both pure and contaminated liquids. Before each subsequent test of the individual composites, the device was calibrated using calibration solutions.

3. Test results

Strengthening soil with fly ash can pose a risk of environmental pollution from heavy metals. The permissible content of heavy metals, whether in waste material itself or in surface water, soil and ash itself resulting from the combustion of coal, is regulated by standards and regulations (DSO 1999, RMG 2015, RMOŚZNL 1995, RRM 2016, Sybilski & Kraszewski 2004). Coal-burning ashes are not classified as hazardous waste but, at the same time, due to their heavy metal content, cannot be considered neutral (RRM 2014). Table 5 shows the limit values of heavy metal content for surface water, non-hazardous waste and fly ashes (DSO 1999, RMG 2015, RMOŚZNL 1995, Sybilski & Kraszewski 2004). The test of heavy metal leachability from ground composite materials was carried out twice for each composite and for fly ash, and the obtained results are presented in Table 4 as the arithmetic means.

Table 4. Heavy metals and metalloids content in water extracts from ground composite materials and fly ash

Element / Composite	C 1 [mg/l]	C 2 [mg/l]	C 3 [mg/l]	C 4 [mg/l]	C 5 [mg/l]	C 6 [mg/l]	C 7 [mg/l]	C 8 [mg/l]	C 9 [mg/l]	C 10 [mg/l]	C 11 [mg/l]	C 12 [mg/l]	C 13 [mg/l]	C 14 [mg/l]	C 15 [mg/l]	Fly ash [mg/l]
Total chromium [Cr]	0.1122	0.1147	0.0806	0.0816	0.0826	0.0961	0.0732	0.0384	0.0970	0.0604	0.0731	0.1059	0.0991	0.0773	0.0768	0.2835
Nickel [Ni]	0.0333	0.0425	0.0259	0.0478	0.0694	0.0312	0.0429	0.1078	0.1824	0.1535	0.0276	0.0673	0.0944	0.0931	0.0851	0.0000
Copper [Cu]	0.0759	0.0936	0.0730	0.1714	0.1451	0.1160	0.1055	0.1865	0.2944	0.2963	0.1974	0.2278	0.2484	0.2320	0.2260	0.0122
Zinc [Zn]	0.0214	0.0645	0.0016	0.0706	0.0231	0.0260	0.0000	0.1410	0.0000	0.0000	0.0388	0.0000	0.0000	0.0000	0.0000	0.0000
Arsen [As]	0.0095	0.0162	0.0216	0.0293	0.0305	0.0113	0.0143	0.0267	0.0131	0.0132	0.0122	0.0163	0.0161	0.0145	0.0043	0.0012
Selenium [Se]	0.0150	0.0184	0.0174	0.0214	0.0260	0.0222	0.0276	0.0323	0.0321	0.0284	0.0286	0.0353	0.0317	0.0274	0.0282	0.0195
Molybdenum [Mo]	0.1036	0.1201	0.1008	0.1207	0.1222	0.1110	0.1445	0.1401	0.1661	0.1287	0.1262	0.1514	0.1271	0.1165	0.0965	0.1598
Cadmium [Cd]	0.0002	0.0053	0.0001	0.0002	0.0003	0.0001	0.0003	0.0001	0.0002	0.0001	0.0002	0.0001	0.0001	0.0002	0.0000	0.0001
Antimony [Sb]	0.0049	0.0098	0.0091	0.0107	0.0143	0.0100	0.0136	0.0049	0.0062	0.0013	0.0115	0.0326	0.0032	0.0024	0.0007	0.0002
Barium [Ba]	0.3148	0.3909	0.0944	0.0853	0.0563	0.1250	0.1032	0.1234	0.3722	0.5048	0.1578	0.0964	0.1860	0.4669	0.8899	1.0076
Mercury [Hg]	0.0013	0.0011	0.0009	0.0007	0.0006	0.0005	0.0005	0.0005	0.0004	0.0004	0.0003	0.0004	0.0004	0.0003	0.0002	0.0004
Lead [Pb]	0.0122	0.0242	0.0044	0.0051	0.0028	0.0030	0.0044	0.0022	0.0042	0.0007	0.0072	0.0117	0.0016	0.0008	0.0008	0.0004

Table 5. Acceptable concentrations of heavy metals for selected environmental conditions (DSO 1999, RMG 2015, RMOŚZNL 1995, Sybilski & Kraszewski 2004)

Element	Limit value for waste materials (non-hazardous) [mg/l]	Values of surface water pollution indicators (purity classes) [mg/l]			Maximum acceptable content for ashes [mg/l]
		I	II	III	
Total Chromium [Cr]	1	–	–	–	0.1
Nickel [Ni]	1	<1.0			0.1
Copper [Cu]	5	<0.05			1.00
Zinc [Zn]	5	–	–	–	3.0
Arsenic [As]	0.2	<0.05		<0.2	0.1
Selenium [Se]	0.05	<0.01			0.05
Molybdenum [Mo]	1	–	–	–	–
Cadmium [Cd]	0.1	<0.05	<0.05	<0.05	–
Antimony [Sb]	–	–	–	–	–
Barium [Ba]	10	–	–	–	1.00
Mercury [Hg]	0.02	<0.001	<0.05	<0.01	0.005
Lead [Pb]	1	<0.05			0.1

In the test samples of all ground composite materials and fly ash, the content of heavy metals such as cadmium, total chromium, lead, molybdenum, nickel, vanadium, zinc, cobalt, iron and metalloids such as arsenic, selenium and antimony was mostly lower than acceptable concentrations for selected environmental conditions, as shown in Table 4. As research by the Research Institute of Roads and Bridges (Sybilski & Kraszewski 2004) shows, the laboratory conditions in which samples are prepared and experiments conducted are more conducive to leachability than under natural environmental conditions.

The amount of selenium and copper in the composite samples tested slightly exceeds the limit values for surface water contamination indicators. The copper content of the fly ash itself is lower than in the ready-made composites. This might be due to the fact that copper is a compound strongly bonded by an organic substance in the ground (Konarkowska & Kuziak 2000). The same is true of selenium whose larger amounts are found in soils rich in organic matter (Niedzielski et al. 2000).

No nickel and zinc compounds were detected in the fly ash. However, trace amounts of zinc can be found in selected composites. The zinc content, which occurs naturally in the ground, changes together with the organic matter content in the composites, as shown in Table 4. In composites with the smallest amount of organic matter zinc was not detected.

In the all tested composite materials the pH value after 28 days of curing was above 9. The lowest value, i.e., 9.93, was obtained by the C1 composite with the smallest amount of fly ash and hydrated lime. The highest pH value, i.e., 12.88, was obtained by the C15 composite, where the stabilizer content was the highest. The pH value of the tested peat was 6.5, fly ash 12.30 and the hydrated lime 13.02. The increasing pH values are undoubtedly related to the addition of increasing amounts of stabilizer which minimizes the acidic reaction of peat. The pH values of all composites are shown in Table 6.

Table 6. PH values of ground composite material, peat – fly ash – hydrate lime

Composite	pH value
C1	9.93
C2	10.74
C3	11.63
C4	11.97
C5	12.31
C6	12.23
C7	12.50
C8	12.59
C9	12.68
C10	12.81
C11	12.30
C12	12.47
C13	12.69
C14	12.81
C15	12.88

It can be seen that in composites with the highest content of hydrated lime, the content of heavy metals is significantly lower. This fact has been confirmed by the research conducted in the Institute for Road and Bridge Research (Sybilski & Kraszewski 2004). It has shown that adding lime, which increases the pH value, also influences the reduction of heavy metal leachability from fly ash. This is particularly important while building roads where there is a possibility of high hydration of the substrate (Sybilski & Kraszewski 2004).

Based on the results of studies as well as literature of the subject, it can be concluded that the low leachability of heavy metals and metalloids is related to the high pH value (Desfitri et al. 2020, Leelarungroj et al. 2018). The reduction of heavy metals can also be the result of a pucolanic reaction and the formation of the CSH phase and ettryngite (Leelarungroj et al. 2018, Liu et al. 2018).

Therefore, it can be assumed that environmental pollution while strengthening organic soil with fly ash and lime, in the area of Elbląg Żuławy, and the Druzno Lake Basin in particular, might be of no practical importance. The use of these stabilising materials does not pose any threat to the environment, cultivated soils and humans. As a result, they can be used to strengthen organic soil (Filipiak 2013) for road construction and low volume structures in the area.

4. Conclusions

On the basis of the presented results of the studies, the following conclusions can be drawn:

- The low heavy metal and metalloids leachability from composites makes it neutral for the ground environment, which is important because these sites are protected under the Nature 2000 Programme (Szablon projektu 2013).
- The amount of leachable heavy metals and metalloids decreases with an increased amount of stabilizer. The smallest amounts of leachable metals and metalloids were obtained for composite C15, which is the optimal ratio of fly ash to peat FA/P = 0.75 and lime hydrated to peat HL/P = 0.1875. This is evident in case of mercury, where its quantity in composite C1 is 0.00129 mg/l and in composite C15 is 0.00023 mg/l.
- The addition of hydrated lime increases the pH value, which is associated with a decrease in the leachability of heavy metals and metalloids.
- The decrease in the amount of heavy metals and metalloids in compo-sites can also be the result of a pucolanic reaction and the formation of the CSH phase as well as ettryngite.
- The use of fly ash for strengthening the organic ground substrate in the area of the Lake Druzno basin might be one of the ways of managing the energy products of coal combustion from the Elbląg Power Plant. It can also contribute to reducing environmental pollution with stored material.
- Further work concerning the analyzed issue should consist in testing the water permeability of composite materials, which might be one of the parameters assessing the flow rate of heavy metals.

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