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LEACHING OF SOME METALS FROM BROWN-COAL ASH

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The leaching characteristics of brown-coal ash from Sokolovo area were studied. Demineralized and acidified water was used as a leaching solution. Extraction experiments were performed both in a column and in a batch arrangement. Mobilization of eleven elements was investigated. Metal concentration in the leachate is relatively low in comparison with its original ash content, released contents range from $10^{-3}\%$ (Fe) to 3% (Zn) of the solid phase concentration. In general, chemical changes are most significant in the initial stage of leaching. Concentration of metals in the leachate then usually declines and reaches a steady state during a leaching process. This is in accordance with the fact, that the studied fly ash exhibits an extreme „buffering ability“. Various initial pH values of experimental solution are changing with time and they stabilize on a constant value. The sequence of relative mobility of metals (the ability to enter the liquid phase) during the leaching process is as follows: $Zn > Mn > Ni > Co > Be > Pb > Cd \gg Cu > Cr > As \gg Fe$. Our experiments suggest that the leaching of metals under natural conditions is low and the tested metals probably pose a little threat to terrestrial and aquatic environment in the studied locality, with the exception of beryllium. The concentration of Be in a leachate is up to two orders higher than the limit for surface water admits; therefore Be from the investigated disposal can be a source of contamination for local surface and underground waters.

coal fly ash; leaching; heavy metals; trace elements; migration; mobilization; environment

INTRODUCTION

Disposals of the ash material are one of the serious environmental problems associated with the existence of coal-fired power plants in our country. Fly ash and bottom ash as a „special waste“ are deposited to the disposal sites in suitable areas near the sources. The dry disposals have the advantage of the limiting amount of water that comes into contact with the ash; however, there are problems with the wind erosion and the stabilization of the ash piles. The coal ash is, therefore, transferred more frequently from the collection sites to

the final disposal site by sluicing with water. While the recent ash ponds are isolated from the environment, many disposal ponds constructed prior to 1980 are unlined and may allow the infiltration of leachate through permeable sediments into the underground aquifer.

This paper discusses the concentration of selected elements that can be expected to be released under ash disposal conditions, as simulated by laboratory experiments. The objectives of the presented study are: 1. evaluation of bulk physical changes within the sample caused by the acid water leaching, 2. assessment of the relation of the bulk ash composition (unleached sample) to the composition of leachate generated from the particular ash material, 3. determination of the relative mobility of Fe, Mn, Cr, Ni, Co, Cu, Zn, Pb, As, Cd and Be during the interaction of coal ash with acid water solutions. The partial results were presented in the publications of Minařík, Kvídová (1993) and Kvídová et al. (1994).

MATERIALS AND METHODS

Description of ash samples

The coal ash deposited either on dry or on wet disposal sites is a mixture of the fly ash from mechanic and electrostatic precipitators and bottom ash (material falling to the bottom of the boiler). The former component always dominates in the mixture. It is very difficult, if not impossible at most power plants, to obtain samples of bottom ash that have not been in contact with sluicing water. Samples for the experiments were taken from the collection tank of the boiler of glass factory Oloví. These samples represent the brown coal ash from the Sokolovo basin in the north-west part of Bohemia. The tested material contains fine sized particles of fly ash, most frequently ranging from 0.5 to 100 μm and coarse-grained particulate matter of the bottom ash (200–4 000 μm in diameter). X-ray powder diffraction study of our samples indicates the presence of α -quartz, haematite and magnetite only. The substantial amount of material is amorphous. The bulk of the fly ash matrix consists of an impure aluminosilicate glass. The mineralogy, morphology and bulk chemical composition of the samples indicate that the ash was produced at low temperature under incomplete combustion (the presence of primary quartz and clay, the large amount of organic constituents of the unburnt coal and the semicoked particles, relatively of high content of sulphur, the lack of smooth solid and hollow glass spheres). Mineralogy, chemism and some physical properties of the brown-coal ashes are summarized in the papers of Sulovský, Kühn (1990), Liberský (1990), Kühn, Sulovský (1991), Raclavská, Jiránek (1991) and Dirner et al. (1994).

Column leaching tests

The coal ash samples (20 g) were packed into glass columns and the leaching solutions with initial pH = 2, 3, 4 and 5.5 were passed through the material in downflow. The first and the fourth portions (volume 50 cm^3) of leachate were collected and analysed. This leaching procedure is described in more details by Kopsick (1980) and Fliszár-Baranyai et al. (1992).

Batch leaching tests

The fly ash (10 g) was weighted into the glass Erlenmeyer flask to which 100 cm^3 acidified bidistilled water was added, to set the ratio of leachant to the solid phase 10, for each individual step. The closed flasks were continuously agitated in a shaker. Twenty four experiments were performed for time intervals of 15 minutes, 1 hour, 7 hours, 24 hours, 7 days, 6 months with pH values of 2, 3, 4 and 5.5.

Nitric acid (suprapur grade) was used for preparation of leaching solutions.

Analytical methods

All analyses were performed in the Geological Institute of AS CR. Major components of original ash samples were analysed by classical methods of the silicate analysis (anal. Chalupský and Švec). Minor and trace element analyses of coal ash were performed by atomic absorption spectrometry after the digestion of the samples using HF + HClO₄ mixture. The metals in the leachates were determined using appropriate AAS methods depending on the element and its concentration level. Flame or graphite furnace AAS were used for Fe, Mn, Cr, Ni, Co, Zn, Cu, Pb, Cd and Be determination, hydride technique was employed for the As determination. The electron microprobe analysis was also used for studying the particulate coal ash matter before and after the leaching.

RESULTS AND DISCUSSION

The elemental composition of brown-coal ash is shown in Tab. I. The fly ash contains higher concentration of carbon, sulphur and several volatile minor and trace elements in comparison with those of the bottom ash. Both types of the material are extremely enriched with arsenic, beryllium and copper. The fly ash leachate in distilled water has an acid reaction (pH 4.5–4.7) while that of the bottom ash remains neutral (pH 7.2).

Fig. 1 shows the content of selected elements in fresh samples of both the fly and bottom ash in comparison with range values for the same elements

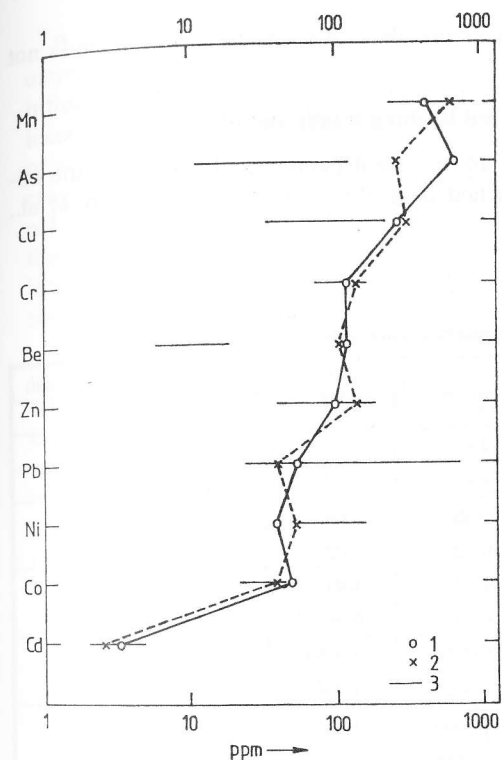
I. Element composition of brown-coal ash

	Fly ash	Bottom ash	12 disposals CR (Liberský, 1990)
Major elements (g/kg)			
Si	172.4	191.1	176.9–259.4
Al	92.8	104.4	74.1–149.1
Fe	73.7	80.0	33.5–100.0
Ca	38.3	45.2	4.5–45.1
Mg	9.6	10.9	3.3–13.3
Na	7.6	8.1	1.5–3.8
K	4.8	4.8	4.5–55.5
Ti	25.0	19.2	4.5–31.6
S	19.1	2.7	0.5–26.5
P	1.3	1.3	0.3–4.0
C	15.9	1.2	–
Minor elements (mg/kg)			
Mn	423.9	619.2	232–929
Cr	123.3	127.5	75–164
Ni	37.7	52.4	56–162
Co	44.4	40.3	22–46
Zn	99.6	136.9	41–196
Cu	272.6	300.5	34–230
Pb	55.1	33.1	24–826
Be	118.0	109.1	6–19
As	663.8	259.7	11–129
Cd	3.3	2.6	< 5

in the ash samples from twelve disposal ponds from the CR (Liberský, 1990). Extreme content of Be, As and Cu in our material is remarkable.

Physical alteration of ash during the leaching

In general, physical properties indirectly influence the geochemical behaviour of elements in fossil fuel combustion wastes. The elements that volatilize during the combustion of coal condense from the gaseous phase on fine



1. Content of some elements in brown-coal fly and bottom ash (Sokolov basin); 1 – content of elements in a fly ash, 2 – content of elements in a bottom ash, 3 – range of values for the same elements in a coal ash (samples from the boreholes in 12 disposal ponds – Liberský, 1990)

particle surfaces; these particles have large surface areas relative to their mass. As a result, ashes with large proportion of fine particles may release large amounts of these surface – condensed elements. The deposited ash with low permeability, however, may retard the movement of pore water, thus allowing longer time for equilibration between the solid and aqueous phases.

The physical changes of the examined fly and bottom ash samples were not found to be essential during the leaching. The permeability of both types of the tested ash was relatively high and the rate of flow of leachant remained approximately constant throughout the whole column leaching period. The samples did not exhibit a pozzolanic reaction. The measurement by electron microprobe reveals that the mean diameter of ash particles before and after leaching experiments is similar. The fly ash particles occur mainly in silt sizes, whereas bottom ash particles occur in sand and gravel sizes. Thin coatings were observed on the surface of some ash particles exposed to the leaching solutions. The character of these surface layers could not be identi-

fied since the depth resolution of the electron microprobe technique is not sufficient for this purpose.

Ash leachate composition, observed leaching trends and pH changes

The pH value of leachate is an important parameter that controls the migration ability of elements leached from the coal ash (Mattigod et al., 1990).

II. Composition of a leachate from laboratory column leaching tests (data in mg/l)

	Fly ash				Bottom ash
	2.06	3.1	4.05	5.55	
Initial pH	2.06	3.1	4.05	5.55	2.08
pH of 1st leach	4.55	4.65	4.67	4.82	7.25
pH of 4th leach	4.51	4.85	4.98	5.10	7.31
Fe	0.05	0.05	0.08	0.08	0.11
	0.03	0.02	0.02	0.01	0.02
Mn	3.96	4.32	4.44	4.28	0.53
	0.23	0.09	0.09	0.25	0.10
Cr	0.0067	0.0050	0.0054	0.0067	0.0044
	0.0034	0.0029	0.0045	0.0005	0.0025
Ni	0.0610	0.0660	0.0530	0.0680	0.0247
	0.0173	0.0148	0.0196	0.0086	0.0111
Co	0.0750	0.0800	0.0890	0.0700	0.0122
	0.0152	0.0100	0.0100	0.0051	0.0074
Zn	0.41	0.43	0.43	0.42	0.22
	0.06	0.04	0.04	0.21	0.03
Cu	0.0120	0.0199	0.0161	0.0155	0.0044
	0.0095	0.0050	0.0044	0.0019	0.0032
Pb	0.0429	0.0364	0.0377	0.0438	0.0120
	0.0071	0.0099	0.0089	0.0018	0.0085
As	0.0033	0.0035	0.0033	0.0040	0.0028
	0.0030	0.0020	0.0033	0.0025	0.0023
Cd	0.0026	0.0024	0.0029	0.0053	0.0010
	0.0006	0.0007	0.0005	0.0002	0.0009
Be	0.0310	0.0299	0.0283	0.0264	0.0004
	0.0112	0.0084	0.0094	0.0002	-

Both the column and batch leaching experiments (Tabs. II and III) show extreme „buffering ability“ of the coal ash. All the leaching solutions pH are influenced during their interaction with ash samples in a short time. The leaching solutions with initial pH = 2, 3, 4 and 5.5 passed through the column of a fly ash remain acidic; final pH value of all leachates varies in a narrow range from 4.4 to 4.7. The leachates of bottom ash are nearly neutral (final pH = 7.2) due to higher contents of Ca, Mg, Na and lower content of S in the original ash material (Tab. I). Similar results are valid for the batch leaching. The final pH of fly ash leachates varies in dependence on the initial solution pH and the leaching period (from 15 min to 6 months).

The observations from the column leaching study (Tab. II) confirm the maximum release of the tested elements in the first portion of leachate. The concentration of all metals are significantly lower in the subsequent leachates. In general, the elements are released in higher concentrations from the fly ash in comparison with the bottom ash, probably due to the different grain size and chemical composition.

The laboratory long term batch leaching of the fly ash (Tab. III) does not indicate simple and unambiguous trends. A majority of the studied elements has its concentration maximum in a leachate with the lowest initial pH at the beginning of the leaching process.

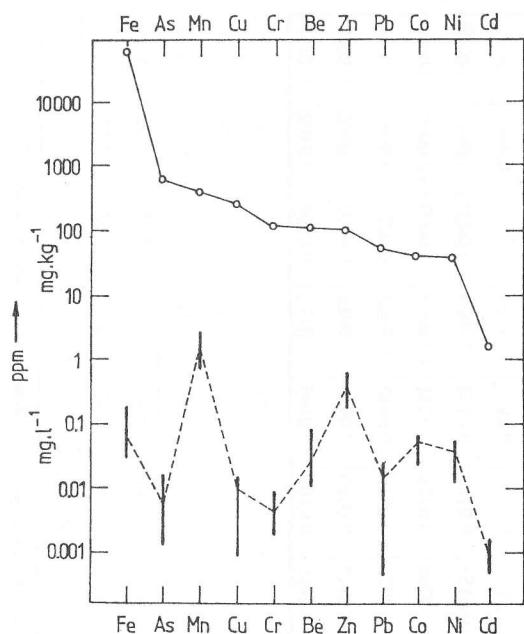
There are two factors which can affect the long term leaching in the opposite way: the time and the „buffering ability“ of the studied material. That is the reason, why the leaching trends are not unambiguous with increasing time periods. The pH value was found to be the determining factor for the leachate concentration of a majority of elements. With increasing pH in dependence on time concentration of elements in the leachate decreases, with notable exception of Mn, Zn and to a lesser extent of Ni and Co, too. In general, however, from environmental point of view the concentration of metals in leachates is relatively very low, except Be.

Extraction experiments show that the element composition of the leachate does not reflect proportionally the chemical composition of the original ash sample. The leaching ability of individual elements differs in several orders of magnitude as documented in Fig. 2, which exhibits high concentration of Fe, As for the ash sample and on the contrary, only minor amounts of these elements found in the leachates. The rate of mobilization of individual elements depends on their speciation (the form in which they are present in a solid phase) and their location, either within the ash matrix or adsorbed onto the ash particle surface. The elements present in a silicate matrix will not be readily available for leaching. On the contrary, the elements adsorbed onto the surface of ash particles will be leached much more easily. Iron and partly arsenic occur primarily in the matrix or as the extraneous sulphide

III. Results of batch leaching tests on brown-coal fly ash (locality Lomnice near Sokolov), concentration of elements in mg/l

	15 minutes				1 hour				7 hours				
	2.03	3.00	4.42	4.01	5.52	2.03	3.00	4.01	5.52	2.03	3.00	4.01	5.52
Initial pH													
pH of leachate	3.70	4.42	4.49	4.45	3.88	4.45	4.53	4.55	4.16	4.46	4.47	4.50	
Fe	0.2	0.07	0.04	—	0.17	0.05	0.04	0.03	0.16	0.05	0.05	0.07	
Mn	1.08	0.86	0.86	0.87	1.55	1.15	0.99	1.05	1.85	1.38	1.48	1.36	
Cr	0.0048	0.0031	0.0039	0.0045	0.0036	0.0034	0.0035	0.0036	0.005	0.0038	0.0038	0.0064	
Ni	0.049	0.018	0.022	0.015	0.056	0.03	0.02	0.02	0.066	0.037	0.036	0.034	
Co	0.0399	0.025	0.03	0.024	0.053	0.031	0.031	0.039	0.067	0.039	0.045	0.033	
Zn	0.31	0.17	0.19	0.21	0.6	0.3	0.25	0.24	0.46	0.28	0.33	0.32	
Cu	0.0164	0.0096	0.0099	0.0072	0.0164	0.0133	0.0069	0.0069	0.0121	0.0117	0.0119	0.0108	
Pb	0.0238	0.0223	0.0232	0.0232	0.0269	0.0224	0.0193	0.0183	0.0226	0.0213	0.0213	0.0195	
As	0.016	0.0033	0.0028	0.0048	0.0118	0.003	0.002	0.002	0.0055	0.003	0.003	0.0033	
Cd	0.0014	0.0007	0.0008	0.0009	0.0014	0.001	0.0009	0.001	0.002	0.001	0.0017	0.0011	
Be	0.067	0.0197	0.0128	0.0133	0.086	0.0195	0.0137	0.017	0.055	0.0186	0.0187	0.0163	

	24 hours				7 days				6 months				
	2.03	3	4.5	4.01	5.52	2.03	3	4.01	5.52	2.03	3	4.01	5.52
Initial pH													
pH of leachate	4.26	4.5	4.57	4.59	4.46	4.46	4.52	4.54	4.45	4.64	4.65	4.7	
Fe	0.06	0.05	0.05	—	0.06	0.1	0.06	0.07	0.09	0.06	0.04	0.05	
Mn	2.11	1.6	1.56	1.66	2.25	1.71	1.7	1.71	2.87	2.16	2.29	2.4	
Cr	0.0052	0.005	0.0047	0.004	0.0063	0.0092	0.006	0.0092	0.0025	0.0017	0.002	0.002	
Ni	0.067	0.047	0.037	0.03	0.053	0.047	0.038	0.038	0.048	0.046	0.034	0.032	
Co	0.072	0.047	0.042	0.056	0.075	0.07	0.062	0.066	0.073	0.064	0.054	0.055	
Zn	0.53	0.37	0.34	0.38	0.38	0.31	0.3	0.28	0.7	0.57	0.49	0.47	
Cu	0.0099	0.0108	0.0163	0.01	0.0155	0.0155	0.0162	0.0153	0.0023	0.0022	0.0012	0.0009	
Pb	0.0251	0.0231	0.0232	0.0248	0.0303	0.0268	0.0258	0.0231	<0.0005	0.0003	<0.0005	<0.0005	
As	0.003	0.0023	0.0023	0.002	0.0013	0.002	0.0018	0.0015	0.0027	0.0027	0.005	0.0048	
Cd	0.0018	0.0017	0.0013	0.0013	0.002	0.0018	0.0016	0.0014	0.0011	0.0007	0.0005	0.0007	
Be	0.044	0.0212	0.0204	0.0194	0.0318	0.0165	0.0165	0.0145	0.017	0.0161	0.0102	0.0112	



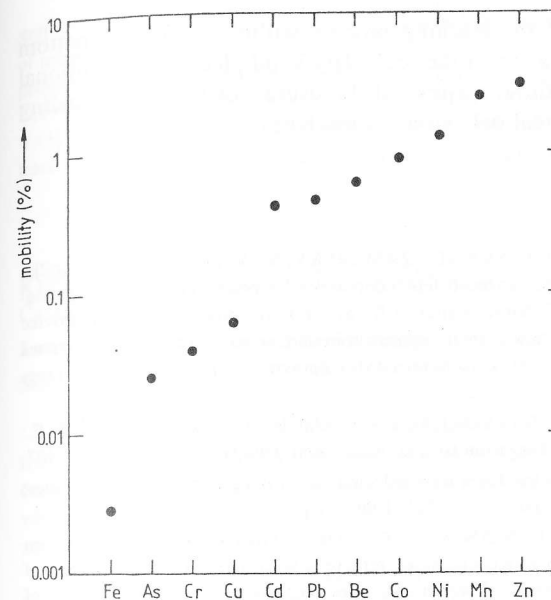
2. Bulk fly ash composition (above) and the maximum and minimum leachate composition (below) for selected elements in the brown-coal fly ash. Analytical data from the long-term laboratory batch leaching tests; dashed line – mean concentration value

material, that is not susceptible to leaching. But probably this is not a sufficient explanation for the low concentration of As, where the condensation of As, in the form of As oxide, on solid particles is suggested (Van der Sloot et al., 1985). The arsenious oxide is relatively well soluble. There is a possible hypothesis, that the dissolved arsenic, which reacts in aqueous acid environment to form As anions, is immediately bound again on the positively charged sites of an ash surface. It can be concluded, that Cd, Pb, Be, Co, Ni, Mn and Zn are associated with more soluble solid phases than Fe and As as follows from the obtained leaching data.

CONCLUSION

The ash samples from the studied locality generated slightly acidic up to neutral leachates. The fly ash leachates are more acidic than their bottom ash counterparts due to the higher concentration of sulphur in the fly ash. Both tested materials exhibit a considerably high „buffering ability“.

The alterations in morphology of samples were not significant during the leaching process in the interval from 15 minutes to 6 months. The relatively low concentration of elements in the leachate obtained in both types of leaching



3. Mobility of elements expressed as a soluble portion of a metal from its original content in the sample, after 15 minutes leaching (%)

techniques (column leaching tests and batch leaching tests) indicate that a considerable part of the tested elements is probably present in a glassy ash matrix.

The sequence of mobility (soluble portion of an element from its original content in sample – %) is the following (see Fig. 3): Zn (3.1), Mn (2.5), Ni (1.3), Co (0.90), Be (0.57), Pb (0.43), Cd (0.42), Cu ($6.0 \cdot 10^{-2}$), Cr ($3.8 \cdot 10^{-2}$), As ($2.4 \cdot 10^{-2}$), Fe ($2.7 \cdot 10^{-3}$). The difference between the marginal members of the set is four orders of magnitude. Generally, the concentrations of the tested metals in the examined brown-coal ash leachates from environmental point of view is low, with the exception of beryllium. Concentration of Be in the leachate is up to two orders higher than the limit for surface water admits; therefore Be from the investigated disposal can be a source of contamination for local surface and underground water. The other studied elements pose only a little threat to terrestrial and aquatic environment in the locality Lomnice.

The studied samples cannot, in any way, be considered as representative for a fly ash in general. Considerable variations undoubtedly exist among the individual disposal areas in dependence on a type of the solid fossil fuel waste in relation to the particular mining district. The results, however, are

instructive for the study of the leaching process within the fly and bottom ash disposal areas. Identification of the secondary solid phases and additional solubility data concerning these wastes will be useful for the understanding and predicting of the chemical behaviour of leachates.

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Loužení vybraných kovů z hnědouhelného popílku. *Scientia Agric. Bohem.*, 26, 1995 (3): 227–240.

Skládkování uhelných popelů patří do kategorie vážných problémů životního prostředí, které úzce souvisí s provozem uhelných elektráren v naší zemi. Zatímco současná úložiště se už budují tak, aby infiltrace do okolního prostředí nebyla možná, většina skládek založených před rokem 1980 dovoluje interakci s okolím a ohrožuje zejména vodní toky.

V práci byly sledovány charakteristiky vyluhování některých kovů z hnědouhelného popílku a škváry ze složiště v Lomnici v oblasti Sokolovské pánve. Studována byla mobilizace celkem 11 elementů v laboratorních podmínkách. Loužení bylo prováděno jednak v průtočném systému skleněných kolon, jednak vsázkovým způsobem. Počáteční pH loužicích roztoků bylo 2, 3, 4 a 5,5, doba experimentů 15 min až 6 měsíců. Testovaný materiál představovaly jemné částice popílku z odlučovačů a hrubší zrna (>200 μm) škváry. Oba typy materiálu tvořil převážně amorfní alumosilikát Fe s vysokým obsahem nespáleného uhlíku a síry. Krystalická fáze byla zastoupena pouze α -křemenem, hematitem a magnetitem.

Extrakční experimenty prokázaly velkou pufrální schopnost studovaného materiálu. Již v počáteční fázi loužení se pH různě kyselých výchozích roztoků stabilizovalo na hodnotě 4,5 pro popílek a 7,2 pro škváru. Koncentrace vodíkových iontů ve výlužích pak zůstala převážně konstantní po celou dobu experimentů; pro míru mobilizace většiny zkoumaných prvků se hodnota pH ukazovala jako určující.

Fyzikální změny popela během loužení byly nepodstatné, jak ukázalo studium povrchu, tvaru a velikosti zrn elektronovou mikroskopou. Propustnost materiálu byla dobrá a k výrazným změnám nedocházelo ani při dlouhodobých pokusech.

Obsah prvků v eluátu neodráží proporcionálně složení původního vzorku. Kovy, jejichž obsah je v popelu nejvyšší (Fe, As), se louží nejméně. Rozdíl mezi okrajovými členy souboru prvků, seřazeného podle mobility, je čtyřřádivý. Pevnost vazby kovů, resp. rozpustnost jejich forem v popelovém materiálu, je nejvýznamnějším faktorem ovlivňujícím stupeň mobilizace.

Pořadí relativní mobility kovů (rozpustný podíl kovu z jeho celkového obsahu v původním vzorku vyjádřený v procentech) je následující: Zn (3,1), Mn (2,5), Ni

(1,3), Co (0,90), Be (0,57), Pb (0,43), Cd (0,42), Cu ($6,0 \cdot 10^{-2}$), Cr ($3,8 \cdot 10^{-2}$), As ($2,4 \cdot 10^{-2}$), Fe ($2,7 \cdot 10^{-3}$).

Obecně jsou koncentrace testovaných kovů loužených ze skladovaného popílku nízké, a proto nepředstavují hrozbu pro terestrické ani vodní prostředí dané lokality, s výjimkou berylia. Berylium uvolňované do kapalné fáze v našem experimentálním uspořádání přesahuje povolené limity dané zákonem z roku 1992 pro tzv. „ostatní povrchové vody“ (nikoliv vodárenské toky) o jeden až dva řády. Je proto reálným nebezpečím pro místní vodoteče.

uhelný popílek; loužení; těžké kovy; stopové prvky; migrace; mobilizace; prostředí

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