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Variation of natural radionuclides in non-ferrous fayalite slags during a one-month production period

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ABSTRACT

The European Basic Safety Standards (EU-BSS) describes a set of NORM (Naturally Occurring Radioactive Material)-processing industries which produce residues known to be possibly enriched in NORs (Naturally Occurring Radionuclides). These residues can be used as a component in building materials aimed for public usage. The industrial processes, in which the residues are produced, are often complex and total monitoring can be challenging especially when the origin of the used raw materials varies. In this study the NORs present in non-ferrous fayalite slags of a secondary smelter facility, a NORM-processing industry according to the EU-BSS, were monitored daily during a one-month production period. In addition flue dust samples and feedstock samples, known to contain elevated levels of NORs, of the same period were measured. The survey involved the gamma-ray spectrometric analysis of the decay products from the ^{238}U and ^{232}Th decay chains, ^{235}U and ^{40}K using HPGe detectors. Secular equilibrium was observed for the slags, flue dust and feedstock samples in the ^{232}Th decay chain, in contrast to the ^{238}U decay chain. During the month in question the ratios of maximum over minimum activity concentration were 3.1 ± 0.5 for ^{40}K, 4 ± 1 for ^{238}U, 6 ± 1 for ^{226}Ra, 13 ± 7 for ^{210}Pb, 4.5 ± 0.6 for ^{228}Ra and 4.7 ± 0.7 for ^{228}Th for the slags. Even with the activity concentration of the feedstock material ranging up to 2.1 ± 0.3 kBq/kg for ^{238}U, 1.6 ± 0.2 kBq for ^{226}Ra, 22 ± 7 kBq/kg for ^{210}Pb, 2.1 ± 0.2 kBq/kg for ^{228}Ra and 2.0 ± 0.4 kBq/kg for ^{228}Th, none of the slag samples exceeded the exemption/clearance levels of the EU-BSS and RP-122 part II, which can respectively provide guidance under equilibrium and in absence of equilibrium. As each NORM-processing industry has its own complexity and variability, the observed variations point out that one should approach one-time measurements or low frequency monitoring methods cautiously. Low frequency measurements should be optimised depending on the discharge of the batches. A follow up of the industrial process and its output can provide important insights to assure a limited public exposure upon application of these industrial residues.

Finally a comparison is made with reported data on other metallurgical slags and the use of the slags in building materials is evaluated using the Activity Concentration Index (ACI) proposed by the EU-BSS.

1. Introduction

Each year over 400 million tons of metallurgical slags, a by-product from metal producing industries, are produced worldwide (van Oss, 2013). Due to the presence of Naturally Occurring Radionuclides (NORs) in the raw materials used by the metallurgical industries the produced metallurgical slag can contain enhanced concentrations of NORs. In Table 1 an overview is given on the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for different types of metallurgical slags reported in the literature. These activity concentrations are found in the intervals 0.004–69 kBq/kg, 0.002–130 kBq/kg and 0.002–23 kBq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively. It must be noted that this list is not exhaustive and some data originate from samples of the 1980's. In several cases the number of measured samples is not specified and not all considered NORs (^{226}Ra, ^{232}Th and ^{40}K) are reported. The data and nomenclature are shown as mentioned in the corresponding reference. Data on other radionuclides mentioned in the corresponding

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Table 1
Overview of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in metallurgical slags in Bq/kg. N is the number of samples.

Slag type	N	^{226}Ra	^{232}Th	^{40}K	Slag origin	Reference
Tin slag[a]	/	1100	/	/	Germany	Leopold and Weiss, 2003
Tin slag[b]	/	/	11 000		United Kingdom - The Netherlands	Ryan et al., 2004
Tin slag	/	2000	/	/	Malaysia	Omar, 2000
Tin slag	12	492–1153	720–1532	11 230–23 183	Malaysia	Ismail et al., 2011
Tin slag	4	/	12 100–14 200	/	United Kingdom	Gilmore and Jackson, 1992
Tin slag	3	500–3400	800–7300	/	Malaysia	Omar et al., 2008
Tin slag	/	4570	420	/	Malaysia	Kontol et al., 2007
Tin slag	3	20 000–69 000	34 000–130 000	/	Brazil	Garcia, 2009
Tin slag	/	1000	4000	/	United Kingdom	European Commission, 1997
Tin slag	/	1000–1200	230–340	330	Germany	Lehmann, 1996
Tin melting slag[a]	/	5500	15 000	/	Germany	Leopold and Weiss, 2003
Nickel smelt slag	3	16.7–364	7.9–82	78.1–888	Poland	Żak et al., 2008
Nickel slag	/	52	78	76	Germany	Lehmann, 1996
Niobium slag	/	/	80 000	/	/	European Commission, 1997
Niobium slag	/	3300–5000	17 000–118 000	/	Brazil	Pires do Rio et al., 2002
Lead slag	/	270	36	200	Germany	Lehmann, 1996
Copper slag[a]	/	2000	/	/	Germany	Leopold and Weiss, 2003
Copper slag	8	287–401	44–73	674–900	Poland	Zak, 1995
Copper smelt slag[d]	80	236.6–517.8	25.7–183	615.4–1250.6	Poland	Żak et al., 2008
Copper slag[a]	/	530	183	1459	Poland	Skowronek and Dulewski, 2005
Copper slag	23	237–336	26–76	615–1251	Poland	Zak et al., 1993
Copper slag (old production)	/	861–2100	18–78	300–730	Germany	Lehmann, 1996
Copper slag (new production	/	490–940	41–60	530–760	Germany	Lehmann, 1996
Copper slag primary process	/	/	13	/	/	RP-122 part II
Copper slag secondary process	/	17	15	/	/	RP-122 part II
Steel slag	/	88	49	/	United Kingdom	Crockett et al., 2003; Hughes and Harvey, 2008
Steel Slag	1	8.62	3.73	5.14	Romania	Ene and Pantelica, 2011
Steel slag	/	/	150	/	The Netherlands	Van Der Steen, 2004
Steel slag	1	62	21	51	China	Wendling et al., 2013
Steel slag	3	184–213	156–182	<17–25	Qatar	Taha et al., 2014
Steel slag	10	15.2–21.4	12.9–15.1	45.3–62.9	Croatiä	Sofilic et al., 2011a,b
Steel slag	/	5–31	0–5	/	/	RP-122 part II
Steel slag	/	100–600	/	/	Slovenia	Smodis et al., 2006
Steel and Iron slag	/	150	150	/	/	European Commission, 1997
Steel slag[c]	/	196	29.6	148	Romania	Tanase and Tanase, 2003
Steel slag	5	51–114	28.6–35.5	118–145	Romania	Sahagia et al., 2014
Non-Iron slag	/	20–30	10–15	20	South Korea	Jeong et al., 2014
Iron slag	2	107.4–113.9	95.2–109.6	2.27–18.9	Saudi Arabia	ALamoudi and ALmehmadi, 2013
Iron slag	/	10–220	10–90	10–150	South Korea	Jeong et al., 2014
Iron slag	12	15–22	/	/	Scandinavia	Brodén et al., 2001
Iron slag	/	64–380	30–98	/	/	RP-122 part II
Iron slag	6	4–234	2–196	8–105	Slovenia	NORM4Building, 2016
EAF slag	3	14.6–17.1	6.7–13.1	15.3–36.9	Croatiä	Sofilić et al., 2010a,b
EAF slag	12	18.3–21.6	12.3–15.4	45.3–63.8	Croatia	Sofilić et al., 2010a,b
EAF slag	/	25	5	10	Greece	Xirouchakis and Manolakou, 2011
Blast furnace slag	/	160–165	35–40	/	Belgium	Vanmarcke et al., 2010
Blast furnace slag	40	13.7–310.1	3.8–330	18.1–290.1	Turkey	Ugur et al., 2013
Blast furnace slag	1	166	47.6	232.3	Spain	Chinchón-payá et al., 2011
Blast furnace (boiler) slag	368	12.4–351.1	2.2–115	18.0–1400	Poland	Żak et al., 2008
Blast furnace slag	4	116–223	83–141	136–196	China	Wendling et al., 2013
Blast furnace slag	12	8–308	1.6–337.3	18.4–388.9	Turkey	Turhan, 2008
Blast furnace slag	/	251.2	24.8	361.7	Croatia	Sofilic et al., 2011a,b
Blast furnace slag	2	143.4–150.9	45.6–45.8	75.7–76.8	/	Puertas et al., 2015
Blast furnace slag	5	105–129	32.4–102	97.2–209	Finland	Mustonen, 1984
Blast furnace slag	5	323 ± 18.6	39.8 ± 7.2	158 ± 16	Egypt	Sharaf et al., 1999
Blast furnace slag	/	88.3–142.0	26.8–46.0	188–269	Hungary	Gallyas and Torok, 1984
Blast furnace slag	/	186.69 ± 2.38	35.87 ± 1.67	295.91 ± 9.08	Turkey	Baltas et al., 2014
Blast furnace slag[e]	42	18.5–458.8	/	225.7–2227.4	Poland	Pensko et al., 1980
Blast furnace slag[a]	/	2100	340	1000	/	RP-112
Blast furnace slag[b]	/	270	70	240	/	RP-112
Blast furnace slag	/	131–139	4	157–177	Ukraine	NORM4Building, 2016
Metallurgical slag	/	251	115	1400	Poland	Skowronek and Dulewski, 2005
Metallurgical slag	6	41–124	41–106	166–395	Slovakia	Cabanekova, 1996
Metallurgical slag	2	162–173	25–52	179–219	Romania	Muntean et al., 2014
Metallurgical slag	40	13–341	2–115	36–889	Poland	Zak, 1995
Metallurgical slag	8	10.8–38.8	2.7–21.8	7.3–63.3	/	Sofilic et al., 2004
Metallurgical slag	160	33–351	12–102	14–825	Poland	Zak et al., 1993
Metallurgical slag + dross[f]	43	10.0–436.0	9.5–55.6	75.7–649.6	Slovakia	Cabáneková, 2008

[a] Mentioned as maximum value.
[b] Mentioned as typical values.
[c] Mentioned as average value.
[d] Could contain samples reported by Zak et al., 1993 and Zak 1995.
[e] Could contain boiler slag data and is converted from pCi/g.
[f] Could contain doubles with Cabanekova 1996.