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Radiological aspects of the usability of red mud as building material additive

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Abstract

Several researchers have examined and achieved favourable results in connection with the building industry's use of red mud extracted in large quantities from the processing of bauxite. These days more and more precedence is being given to limiting the radiological dose to the population. In this study carried out in Hungary, the use of red mud, bauxite, and clay additives recommended for the production of special cements, were examined from a radiological aspect.

^{226}Ra and ^{232}Th activity concentrations measured in Hungarian bauxite, red mud and clay samples were significantly similar with the levels for such raw materials mentioned in international literature.

Taking radiation protection aspects into consideration, none of these products can be directly used for building construction. Taking Hungarian and international values into consideration, a small amount of red mud, not exceeding 15% could be used for brick production, for example as a colouring material. However, beyond this amount the standards for building materials would not be met.

For the production of cements an even stricter limit needs to be determined when both bauxite and red mud are used.

Keywords: Red mud; Radioactivity; Building material

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1. Introduction

The Bayer process is used for refining bauxite to smelting grade alumina, the precursor to aluminium. The process was developed and patented by Karl Josef Bayer more than 100 years ago, and has become the cornerstone of the aluminium production industry worldwide. The Bayer process involves the digestion of crushed bauxite in a concentrated sodium hydroxide solution at temperatures up to 270 °C. Under these conditions, the majority of the aluminium is dissolved, leaving an insoluble residue, that is called red mud in the alumina refining industry. This red mud is removed by settling/filtration. The red mud is composed primarily of iron oxides, quartz, sodium aluminosilicates, calcium carbonate/aluminate, titanium dioxide and sodium hydroxide (pH 10–12), so it is relatively toxic and can pose a serious pollution hazard. In addition to this, the amount produced as a result of the refining process is significant. Red mud is thus of interest to researchers for a number of reasons and has been the subject of many detailed investigations. Of particular interest is the influence of red mud on red mud disposal methods and their influence on the environment, and the application of this abundant waste product in other unrelated areas [1].

Since the recycling of waste by-products is gaining more and more importance nowadays, such research and/or production is being carried out in several countries where these materials are used as additives for building materials. For example, in Turkey bricks are being made with fly ash [2]. In Brazil phosphogypsum is produced during the production of phosphoric fertilizer being used as building material [3], and in Hungary, coal slag is being used in floor structures as insulating filling material [4].

The clay like structure of red mud has been noted by a number of researchers, who have demonstrated that, when fired it becomes a useful ceramic material. Red mud can be used in the manufacture of tiles, bricks and insulating materials [5,6].

Thus, nowadays several studies are being carried out [7–9] on the possible use of red mud from bauxite processing for use in building material additives (e.g. special cement or brick

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production). Some researchers [10–12] have assessed the mechanical and aesthetical characteristics of building ceramics (bricks, tiles) produced by mixing clay with different quantities (0–50%) of red mud with positive results.

Several researchers studied the possible production methods of cement through the use of lime + red mud + fly ash, lime + red mud + bauxite or lime + red mud + bauxite + gypsum, and their experience showed that cements made of lime + red mud + bauxite or lime + red mud + bauxite + gypsum exhibit strengths comparable or superior to ordinary Portland cement [13,14].

All rocks and minerals contain naturally occurring radioactive material (NORM) of terrestrial origin [15–17]. From these, ^{238}U and its daughter products, ^{232}Th and its daughter products, and ^{40}K are the most important. Population weighted averages in soils are: $^{40}\mathrm{K} = 420\,\mathrm{Bq\,kg^{-1}}$, $^{238}\mathrm{U} = 33\,\mathrm{Bq\,kg^{-1}}$, $^{232}\mathrm{Th} = 45\,\mathrm{Bq\,kg^{-1}}$ [18]. The radiobiological and epidemiological effects of ionizing radiation from naturally occurring radioactive materials (NORM) on man, especially in areas with elevated NORM, are becoming prime concerns in the area of radiation protection [19].

In some cases the radiation dose originating from minerals, rocks (uranium ore, coal, phosphate, monazite, bauxite, etc.) in the mine, and from the rocks brought to the surface after mining, and from the technologically enhanced naturally occurring radioactive material (TENORM) concentrated in by-products (uranium slime, phosphogypsum, fly ash and bottom ash, red mud, etc.) during their processing, may be significant [20].

As radiation of terrestrial origin in buildings does not only originate from the soil, but also from the used building materials [21], a greater absorbed dose rate can be measured within buildings (world average $84\,\mathrm{nGy\,h^{-1}}$) than outdoors ($59\,\mathrm{nGy\,h^{-1}}$) [18].

In houses where materials with elevated NORM or TENORM are used in building materials or additives, an absorbed dose rate highly exceeding the world average can be measured [22,23]. Therefore, the different national and international recommendations on the radionuclide concentration of building materials have proposed limit values [24,25].

The EU details Radiation Protection principles in issue 112 of radiation protection. Controls should be based on dose criterion, which was established considering the overall national circumstances within the EU [24]. It is recommended that controls should be based on a dose in the range of $0.3–1\,\mathrm{mSva^{-1}}$. Doses exceeding these values should be taken into account from a radiation protection point of view. This is the excess gamma dose to that received outdoors.

Table 1
The activity concentration index ("*I*")

Dose criterion	$0.3\,\mathrm{mSvy^{-1}}$	$1\,\mathrm{mSvy^{-1}}$
Materials used in bulk amounts, e.g. concrete	$I \leq 0.5$	$I \leq 1$
Superficial and other materials with restricted use: tiles, boards, etc.	$I \leq 2$	$I \leq 6$

Because more than one radionuclide contributes to the dose, it is practical to present investigation levels in the form of an activity concentration index. The activity concentration index (I) is calculated by the next formula:

$$I = \frac{C_{\mathrm{Ra}}}{300\,\mathrm{Bq\,kg^{-1}}} + \frac{C_{\mathrm{Th}}}{200\,\mathrm{Bq\,kg^{-1}}} + \frac{C_{\mathrm{K}}}{3000\,\mathrm{Bq\,kg^{-1}}}$$

where C_{Ra}, C_{Th}, C_{K} are the radium, thorium and potassium activity concentrations ($\mathrm{Bq\,kg^{-1}}$) in the building material. The activity concentration index should not exceed the values presented in Table 1.

When industrial by-products are incorporated in building materials and there is reason to suspect that these contain enhanced levels of natural radionuclides, the activity concentrations of these nuclides in the final product should be measured or assessed reliably from the activities of all the component materials [24].

Typical and maximum concentrations in common building materials and industrial by-products used for building material in the EU are presented in issue 112 of the Radiation Protection of the EU. Some data are presented in Table 2.

As mentioned above, according to international literature, red mud mixed with bauxite, lime, fly ash and gypsum are suitable for manufacturing aesthetic ceramics (bricks, tiles) as well as cement with a good bonding capacity. In several cases, scientific literature deals with the increase of the possible radiation dose of the population [26–29] resulting from using slag, fly ash, phosphogypsum, etc. but the expected dose through the use of red muds and bauxites has not yet been dealt with. Therefore, prior to possible use preliminary radiological tests should be carried out in order to reliably assess the radiation dose related to the use of red muds. Based on the findings it will be possible to decide whether these materials can be used for the given purposes from a radiological point of view.

The radionuclide concentrations of natural origin contained in bauxite and red mud found in the various scientific literatures are summarized in Table 3.

It is apparent that the radionuclide concentration of natural origin in bauxites and red mud significantly exceeds the world average for building materials ($^{232}\mathrm{Th} = 50\,\mathrm{Bq\,kg^{-1}}$,

Table 2
Typical and maximum activity concentrations in common building materials and industrial by-products used for building materials in the EU

Material	Typical activity concentration ($\mathrm{Bq\,kg^{-1}}$)			Maximum activity concentration ($\mathrm{Bq\,kg^{-1}}$)		
	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K
Clay	50	50	670	200	200	2000
Gypsum (by-product)	390	20	60	1100	160	300
Coal fly ash	180	100	650	1100	300	1500

Table 3
Radionuclide concentration of natural origin of bauxite and red mud [18,30]

Material	Activity concentration (kBq kg^{-1})		
	^{238}U	^{226}Ra	^{232}Th
Bauxite	0.25		0.20
ore	0.4–0.6		0.3–0.4
Bauxite from China	0.46	0.31	0.37
Bauxite from Guyana	0.08	0.05	0.23
Bauxite (typical)	0.5	0.4	0.4
Red mud	0.28		0.18
Red mud	0.26–0.54	0.122–0.335	0.34–0.50

^{226}Ra = 50 Bq kg^{-1}, ^{40}K = 500 Bq kg^{-1}) [30]. From this it can be assumed that if radiological controls on these materials are not carried out before their use, the building materials produced will in some cases not comply with regulations.

Between 1940 and 1990, intensive bauxite mining and alumina production was carried out in Hungary.

In the process of alumina production, approximately 1.0–1.6 t of red mud is generated per ton of alumina. It is estimated that over 40 Mt of this waste remains in total in Hungary. The amount of residue generated, per tonne of alumina produced, varies greatly depending on the type of bauxite used, from 0.3 t for high grade bauxite to 2.5 t for very low grade.

During the ore processing of 1 t of bauxite, an amount of red mud approximately equal to it or greater is generated (1–1.5 t).

Due to bauxite processing, a significant amount of red mud accumulated in Hungary. At this time, most of the red mud has dried in the areas it has been stored. Based on international recommendations the possible use of red mud and bauxite as a building material is under consideration.

According to previous investigations the level of radioactivity in Hungarian bauxites is almost identical to the values found in international literature. Analyses of Hungarian radiological investigations so far indicate that further radiological examinations may be necessary, not only in Hungary but in other countries as well, if the material will be used by the building industry.

During our work the radionuclide concentration of red mud, and other materials mentioned in the international literature were investigated. Taking into consideration the radionuclide concentration values obtained, and those given in the international literature as well as the mixing ratios given in the mentioned literature, the effects of these materials were assessed from a radiological point of view.

2. Methods of measurements and calculation

2.1. Sampling and sample preparations

Concerning bauxites, samples collected from several Hungarian bauxite mines (Csordakút, Fenyőfő, Halimba) were examined. A total of 46 bauxite samples were taken from the exploited bauxite heaps, and the samples were milled and homogenized.

Red mud samples (a total of 58) came from the deposits of two alumina plants (Ajka and Almásfüzitő), which were filled in between 1974 and 1990, and therefore at the present time are already dry. Samples from the deposits were taken from the surface and from different depths (100–200 cm).

Several clay samples used during brick production were also collected for analysis.

Samples were first dried in the air, then at a temperature of 105 °C until mass constancy was achieved.

The dried samples were stored for 30 days in air-tight aluminium Marinelli beakers with a volume of 600 cm^3 in order to attain a radioactive equilibrium for ^{226}Ra with its progeny.

2.2. Gamma spectrometric analyses of samples

The concentrations of natural radionuclides were determined by high-resolution gamma ray spectrometry, using an Eurisys EGNC 20-190-R n-type HPGe detector with an efficiency of 20% and an energy resolution of 1.8 keV at 1332.5 keV. The gamma spectra were recorded by a Tennelec PCA-MR 8192-channel analyser. Samples were measured for 60,000–80,000 s. The system was calibrated using a red mud reference material certified by the Hungarian National Office of Measures.

The ^{226}Ra concentrations were determined by measuring the activities of its decay product ^{214}Pb (352 keV) that were in secular equilibrium with ^{226}Ra following the 30-day storage. The activity of ^{40}K was measured directly through its 1461 keV peak. In the case of ^{232}Th, the 911 keV peak of ^{228}Ac as well as the 2614 keV peak of ^{208}Tl were measured [31,32]. This was made possible by the fact that in the case of thorium, a shift in radioactive equilibrium is not expected during the processing due to the outstandingly high pH value (pH ≈ 14) of the red mud. Thorium and radium in an alkaline medium are practically insoluble and therefore remain the red mud.

Industrial by-products like red mud are mainly used as additives in the production of building materials. This has to be taken

Table 4
Recommended mixing ratios for the production of bricks (B), and special cements (C)

Sign	Compositions (wt%) of different bricks	
	Red mud	Clay
B_1	5	95
B_2	10	90
B_3	15	85
B_4	20	80
B_5	50	50
B_6	100	–

Sign	Compositions (wt%) of different cements				
	Lime	Red mud	Bauxite	Gypsum	Fly ash
C_1	65	10	–	–	25
C_2	65	25	–	–	10
C_3	50	15	35	–	–
C_4	50	35	15	–	–
C_5	47.5	15	30	7.5	–
C_6	47.5	30	15	7.5	–

Table 5
Hungarian and international average (minimum–maximum) values of ^{226}Ra, ^{232}Th and ^{40}K concentration of bauxite, red mud, lime, clay and gypsum

Sample	Number of samples	Activity concentration (Bq kg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
Hungarian samples				
Bauxite	46	419 (132–791)	256 (118–472)	47 (10–82)
Red mud	58	347 (225–568)	283 (219–392)	48 (4.9–101)
Clay	7	39 (31.8–52.5)	59 (40.6–75.3)	688 (518–843)
International data				
Bauxite		400	400	
Red mud		122–335	341–496	
Gypsum (world average)		10	10	80
Clay (world average)		50	50	670
Lime (world average)		50	50	500
Fly ash (Hungarian average)		760	117	441

into consideration, with the qualification that the recommended limit values apply to the final products.

The recommended and applied mixing ratios, in this case, for bricks and special cements, as given in different literature are presented in Table 4.

3. Results and discussion

3.1. Radionuclides in the samples

The minimum, maximum and average values of the ^{226}Ra, ^{232}Th and ^{40}K activity concentration of bauxite samples, the red mud, as well as the clay samples determined during the processing, as well as the international values taken into consideration by the calculations are summarized in Table 5. The variation coefficients of the concentration measurements were 3–8% depending on the radionuclide and the activities measured. The lower limit of detection (LLD) for the radionuclides were: $^{226}\text{Ra} = 1.5\ \text{Bq kg}^{-1}$, $^{232}\text{Th} = 2.4\ \text{Bq kg}^{-1}$ and $^{40}\text{K} = 3.3\ \text{Bq kg}^{-1}$.

In accordance with international data, the activity concentration of radionuclides in the examined Hungarian bauxite samples is relatively high. Radionuclides of natural origin found in bauxite get mixed into the red mud during processing. No significant differences were found between the activity concentration of samples taken from the surface or at different depths.

The ^{232}Th and ^{226}Ra concentration measured in the red mud samples significantly (5–7 times) exceeds the world average (50–50 Bq kg^{-1}) for radionuclide concentration in building materials [30]. At the same time their ^{40}K concentration remains significantly below the world average of ^{40}K 500 Bq kg^{-1}.

The activity concentration of ^{232}Th and ^{40}K in the examined clay samples slightly exceeds the world average values for clays.

3.2. Characterization of samples for use as building elements

Index values "I", calculated by taking the Hungarian and international radionuclide concentrations into consideration, are presented in next tables.

Table 6
Values of activity index "I" of brick mixtures taking the activity concentrations measured by us, and those given in the international literature, into consideration

Sign	Activity index "I"			
	Hungarian			World
	Minimum	Maximum	Average	Average
B_1	0.55	0.78	0.75	0.74
B_2	0.62	0.95	0.85	0.83
B_3	0.69	1.11	0.94	0.93
B_4	0.75	1.27	1.04	1.03
B_5	1.16	2.25	1.62	1.61
B_6	1.85	3.89	2.59	2.59

As the international and Hungarian data are similar, the results from Hungary can be applied to radiological risk estimation in other countries as well.

Taking into consideration the dosage recommended for the production of bricks and the measured radionuclide concentrations, the activity index "I" presented in Table 6 was obtained.

Based on the above, for brick production in Hungary, up to 15% of red mud can be used. If a greater mixing ratio is used, the criteria $I \leq 1$ would not be complied with. Calculating the activity index with the radionuclide concentrations mentioned in international literature gives similar values.

In case of cements which are used in relatively small amounts in building work, limits of $I \leq 2$ and $I \leq 6$ must be complied with.

Table 7
Activity index "I" values of cement mixtures

Sign	Activity index "I"			
	Hungarian			World
	Minimum	Maximum	Average	Average
C_1	2.42	7.94	0.98	0.97
C_2	1.85	7.67	1.18	1.16
C_3	1.27	7.06	1.64	1.89
C_4	1.43	6.83	1.64	1.72
C_5	1.19	6.63	1.50	1.81
C_6	1.31	6.46	1.50	1.68

Activity indices for "I" for cements based on the average values of examined samples are presented in Table 7.

If these guidelines are followed, all the end products would be in accordance with the stricter 0.3 mSvy^{-1} regulation.

4. Conclusions

On the whole, it can be stated that when either Hungarian or international values are taken into consideration, looking at their average radionuclide content, red muds could be used in small amounts for brick production (e.g. as colouring agents). However, in case of amounts exceeding 15% the criteria $I \leq 1$ for building materials would not be complied with.

For the production of cement, the dose limit of 0.3 mSvy^{-1} can be met in all cases. Therefore, the use of red mud and bauxite in the production of special cements seems to be effective from a radiological aspect. In the case of brick production, the use of the materials is only possible with a limited blending ratio. As the maximum radionuclide concentration of the base materials may significantly differ from the average value, it would be reasonable to check the components or the products from a radiological aspect in all cases.

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