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Dependence of radon emanation of red mud bauxite processing wastes on heat treatment

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ABSTRACT

Natural radioactivity content, radon emanation and some other physical characteristics of red mud were investigated, so that to identify the possibilities of the safe utilization of such material as a building material additive. Based on the radionuclide concentration, red mud is not permitted to be used directly as a building material, however, mixing of a maximum 20% red mud and 80% clay meets the requirements. The main aim of this work was to determine the dependence of the emanation factor of red mud firing temperature and some other parameters. The relevant experimental procedure was carried out in two different ways: without any additional material, and by adding a known amount of sawdust (5–35 wt%) then firing the sample at a given temperature (100–1000 °C). The average emanation factor of the untreated dry red mud was estimated to 20%, which decreased to about 5% at a certain heat treatment. Even lower values were found using semi-reductive atmosphere. It has been concluded that all emanation measurements results correlate well to the firing temperature, the specific surface and the pore volume.

1. Introduction

Recently, there has been much focus on the reduction or the remediation of the environmental damage caused by industrial by-products containing elevated naturally occurring radioactive material (NORM) concentration, for instance red mud, which originates from bauxite processing. NORMs generally contain radionuclides found in nature, i.e., thorium, uranium, and their progeny. Once this NORM becomes concentrated through human activity such as mineral extraction, it may become a radioactive contamination hazard.

The use of industrial by-products (e.g. slag, fly-ash, and red mud) is widespread in the building industry, where they are primarily used as additives to concrete [1–4].

However, if materials with a high ^{226}Ra concentration are used in buildings, the gamma-dose rate may increases due to radium and its progeny, which in turn result to higher external radiation dose for the inhabitants [5,6]. Furthermore, ^{222}Rn could emanate from such NORM materials and accumulate in closed rooms; thus, increasing the internal radiation dose through the inhalation of ^{222}Rn and its daughter elements [7,8]. Therefore, it seems not enough to examine only the activity concentration of materials used for building construction; information should also be collected about their radon emanation potential. Within fired clay products and in different NORM materials, the emanation coefficient varies widely [9,10].

Several researchers [11–17] have pointed out that the emanation coefficient can be significantly influenced by many parameters, such as pore size, specific surface, grain size distribution, grain density, homogeneity of ^{226}Ra distribution within the grain, and humidity. Red mud is generally considered to be a NORM material with elevated ^{226}Ra concentration, and a potential of increased radon emanation. Therefore, if red mud is utilized as a component of a building material, serious consequences may result not only from external radiation doses but also from doses due to radon emanation.

The main goal of this work is to identify the possibilities of safe utilization of such bauxite processing wastes in the building industry in terms of radiation safety. To this end the activity concentration of ^{226}Ra, ^{40}K, and ^{232}Th, of red mud was determined and the European Union (EU) index classification concerning building materials was first used from the perspective of initially evaluating red mud applicability as building material additive [18].

Subsequently, the emanation coefficient of "pure" red mud samples and also red mud samples mixed with sawdust were fired at different temperatures, in order to examine any decrease in emanation coefficient. Heat dependence of the emanation coefficient was determined in the temperature range of 100–1000 °C in both sample cases.

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2. Materials and methods

2.1. Sampling

Red mud samples were collected from the deposition ponds of two alumina plants in Hungary (Fig. 1). (The Almásfüzitő plant was closed several years ago, but the Ajka plant is still operational. The technology in both places was the same, based on leaching with soda.) Samples from Ajka were taken from the settling ponds and from Dorr settler as well. Samples from Almásfüzitő were taken settling pond Number 7 (it is located between Dunaalmás and Almásfüzitő town) and 8 (it can be found in the vicinity of Neszmély village) close to Danube river. Approximately 3–5 kg of red mud samples were collected from each site, from the Dorr settler 20 l of sludge was collected. Samples were dried to constant weight at a temperature of 105 ± 0.5 °C for 24–48 h. The original moisture content varied in a range of 5–60% obviously the highest moisture content sample is a sludge from the Dorr settler. The dried samples were sieved according to grain size fractions using an Edmund Bühler KS 15-B type shaker. Sieving to different granulations is necessary for the samples specific surface, and pore volume measurements. It should be done in order to remove different parts of the vegetation/plants and separate different fractions if relevant.

Samples were measured using gamma-ray spectroscopy and radon emanation coefficient was determined.

2.2. Determination of ^{226}Ra, ^{232}Th and ^{40}K activity concentration

The dried red mud samples were stored for 30 days in airtight aluminum Marinelli beakers with a volume of 600 cm^3 to reach the secular equilibrium between ^{226}Ra and its progeny.

The activity concentrations of natural radionuclides were determined by high-resolution gamma-ray spectrometry using Eurisys EGNC 20-190-R n-type HPGe detector with an efficiency of 20% and 1.8 keV energy resolution at the energy peak of 1333 keV of ^{60}Co isotope. Gamma spectra were recorded by Tennelec PCA-MR 8192 multichannel analyzer. Data collection time changed from 15,000 to 80,000 s. The system was calibrated using reference material – homogenized red mud, from a settling pond of Ajka Alumina Plant – of the same geometry certified by the Hungarian National Authority of Measures [19], the reference material density is close to our samples (within 2% deviation). ^{226}Ra concentrations were determined by measuring the activities of its decay products, ^{214}Pb (295 and 352 keV) and ^{214}Bi (609 and 1120 keV), which were in secular equilibrium with ^{226}Ra following the 30 day storage. The activity of ^{40}K was measured by the 1461 keV gamma ray, ^{232}Th by the 911 keV gamma ray of ^{228}Ac, and the 2614 keV gamma ray of ^{208}Tl [20].

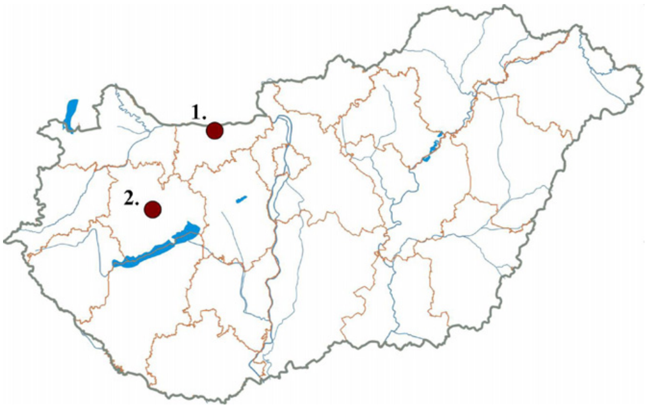

Fig. 1. Origins of red mud samples (1. Almásfüzitő and 2. Ajka).

2.3. Measurements of ^{222}Rn emanation

A precise mass weight of dried red mud sample (15 ± 0.1 g) was placed in a 50 cm^3 glass ampoule and dried in a vacuum dryer chamber for 3 h at 60 °C, then sealed. After 30 days' storage, the ampoule was broken inside a special metal receptacle. Radon gas was pumped into a 1 dm^3 Lucas cell through a filter using N_2. Pumping was repeated into a second Lucas cell, so that the efficiency of transporting ampoule air to the Lucas cells was higher than 99.7%. Measurements were performed with an EMI photomultiplier for 1000 s after, so that equilibrium of radon with its shortlived progeny could be reached [21]. The Lucas cells were calibrated using a PYLON RN 2000A type passive radon source with an activity of 105.7 ± 0.4% kBq in a Genitron EV 03209 calibration chamber having a volume of 210.5 dm^3. The radon emanation coefficient (ε) was determined as the quotient of activity of radon grabbed from the ampoule and the ^{226}Ra activity of red mud in the ampoule. The overall relative standard uncertainty was less than 12%.

2.4. Thermogravimetric measurements

When carrying out thermogravimetric measurements, 300–700 mg red mud was placed in a corundum sample holder with high chemical resistance. In the selection of the sample holder, the rather strong alkaline nature/reaction of the sample had to be taken into account. Derivatographic records were taken using a MOM Derivatograph in static air atmosphere. To heat-up the red mad samples evenly and gradually, the speed of heating was set to $10\,^{\circ}\text{C}\,\text{min}^{-1}$.

2.5. Specific surface, pore volume measurements

A Micromeritics ASAP 2000 device was used to measure pores under 100 nm. Samples (1–2 g) of different granulations were put in a vacuum ($P < 100$ Pa) at 100 °C to remove gases linked to the surface. Then, adsorption and desorption isotherms for nitrogen gas were measured at the temperature of liquid nitrogen. Specific surface was calculated according to the BET theory.

Pores above 100 nm were measured by SMH6 type mercury poremeter device. Samples (1–5 g) were put into vacuum ($P < 0.1$ mmHg) at room temperature. The measuring receptacle was then filled with mercury and the change of Hg level in the capillary was recorded against pressure (0–1000 bar). The distribution of pore volume was calculated using the abovementioned results.

2.6. Red mud and sawdust mixing

In most technological processes for building materials, additives like sawdust, polypropylene pellets in different mixing ratios (5–35%) are used to achieve better brick quality (i.e. enhanced porosity, strength, heat insulating property). For this reason, red mud samples were mixed with sawdust in different ratios. It results a semi-reductive atmosphere.

2.7. Red mud samples heat treatment

All prepared red mud samples, both pure and mixed with sawdust were fired in temperatures between 300 and 1000 °C.

3. Characterization of samples for use as building materials

Two major problems arise in the course of red mud utilization in the building industry. The first is the excess external gamma dose invoked, and the other is associated with the emanated radon

Table 1
The activity concentration index ("*I*") [RP 112].

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

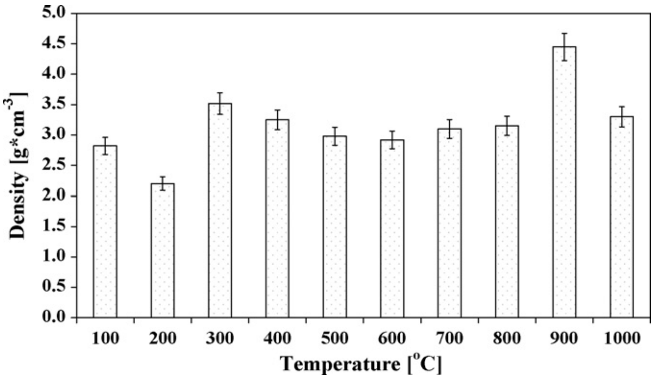

Fig. 2. Changes in the density of red mud as a function of burning temperature.

and thoron because of the relatively high ^{226}Ra, ^{232}Th activities. The potential risk due to ^{222}Rn could be estimated using measurements of ^{226}Ra activity concentration and radon emanation power.

At present (2009) the radiological evaluation of building materials is mainly based on the concentration of ^{226}Ra, ^{232}Th, and ^{40}K. To this end it is suggested to calculate a value for the activity concentration index according to the EU (and some other countries like Finland and Norway) [18] following the equation:

$$I = \frac{C_{\text{Ra-226}}}{300\,\text{Bq kg}^{-1}} + \frac{C_{\text{Th-232}}}{200\,\text{Bq kg}^{-1}} + \frac{C_{\text{K-40}}}{3000\,\text{Bq kg}^{-1}}$$

where I is the activity concentration index; C_X is the measured activity concentration of the radioisotope X (Bq kg^{-1}).

The EU recommended values for this index (Table 1) were taken into account in the course of the red mud characterization.

4. Results

4.1. Density and particle size distribution

The average density of air-dried red mud samples was 2.82 g cm^{-3} (2.80–2.91 g cm^{-3}), a value which is comparable to values given in literature [22].

Following the samples heat treatment and their consequent material structure changes the density changes too (Fig. 2). These density changes can be explained, according to literature data, by the fact that the phase composition of red mud changes with the firing temperature and so does the crystal structure within the different grains [23,24].

As more than 99% of the particles were within the range of 0.090–0.063 μm, all the measurements were only carried out with this fraction.

Table 3
"*I*" activity concentration index of the samples.

Samples	Activity concentration index (*I*)		
	Average	Minimum	Maximum
Ajka(100%)	2.58	2.09	3.85
Almásfüzitő(100%)	2.08	1.06	4.42
Ajka(20%)	1.04	0.75	1.27
Almásfüzitő(20%)	0.89	0.68	1.35

4.2. Activity concentration

Activity concentrations of different isotopes in the red mud samples of the present work have been reported in [25] and can be summarized in Table 2.

It can be clearly observed that the measured activities in our samples are higher than the world average ^{226}Ra and ^{232}Th activity concentrations in building materials. In fact, the activity concentration of these samples is between 2 and 14 times higher (e.g. ^{226}Ra: 105–700 Bq kg^{-1}) when compared with the world average radionuclide concentration of the building materials (^{226}Ra: 50 Bq kg^{-1}, ^{226}Th: 50 Bq kg^{-1}, ^{40}K: 500 Bq kg^{-1}) [26]. Activity concentration indexes "*I*" were calculated from the above mentioned values and introduced in Table 3.

On the grounds of their "*I*" activity concentration index, red mud of different origins are to be used with restrictions, for example, as an additional material at a maximum of a 20% mixing ratio.

4.3. Emanation coefficient (ε)

The radon emanation coefficient (ε) was determined for red mud samples as a function of certain parameters. As it can be observed in Table 4, the emanation coefficient of untreated dry samples varies – in a relatively wide range – between 6 and 22%. This variation depends on the material, grain size, moisture, and origin of sample. As a comparison between radon emanation coefficient of red mud in this study with other different building materials of various countries and some NORMs is presented in Table 4.

The variation of pure red mud samples emanation coefficient as a function of firing temperature in the range between 300 and 1000 °C, can be seen in Fig. 3.

The emanation coefficient changes with the temperature of burning and follows a specific pattern. It can be safely stated that firing temperature has an effect on the emanation. In Fig. 4 the variation of emanation coefficient of red mud mixed with sawdust as a function of the mixing ratio is presented.

The emanation coefficient similarly changed – as in the case of heat treatment – when sawdust was added to the red mud in different ratios (5–35%). When the amount of sawdust and burning temperature are increased, the emanation coefficient clearly decreases.

In order to provide an explanation for the dependence of the measured emanation power on the firing temperature, thermal (DTG) specific surface and pore volume examinations were carried out. Results of thermogravimetric measurements can be found in the following (Figs. 5 and 6).

Table 2
^{40}K, ^{226}Ra and ^{232}Th activity concentration of the samples.

Red mud samples	Average radionuclide concentrations (min–max) [Bq kg^{-1}]		
	^{40}K	^{232}Th	^{226}Ra
Ajka	48 (5–101)	292 (285–380)	360 (150–700)
Almásfüzitő	103 (47–212)	229 (87–545)	294 (102–506)
World average of building materials [26]	500	50	50

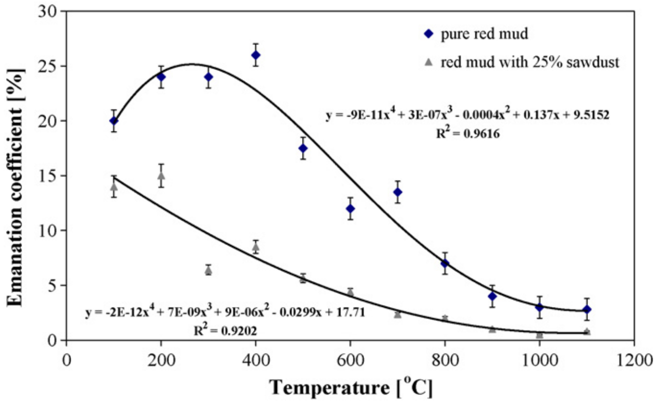

Fig. 3. Variation of emanation coefficient as a function of burning temperature.

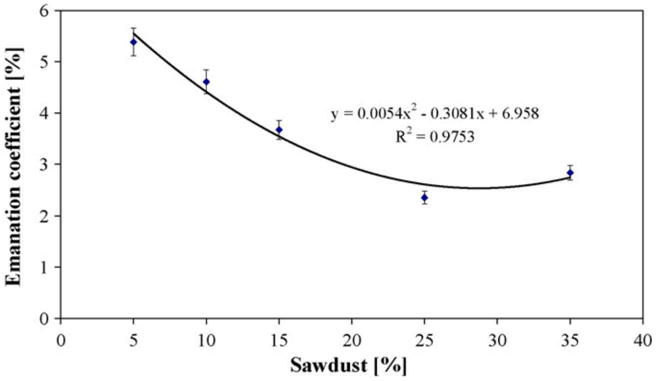

Fig. 4. Correlation between emanation coefficient as a function of mixing ratio of sawdust (at 800 °C).

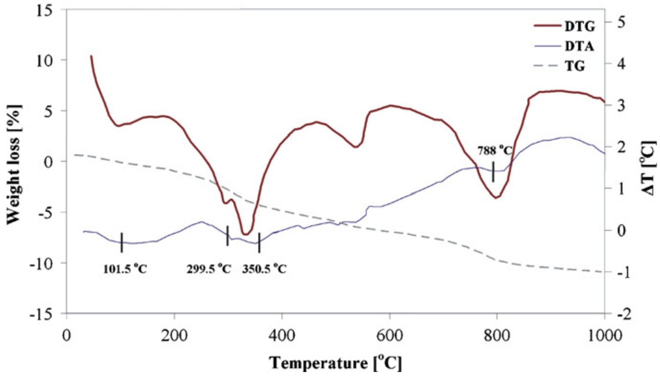

Fig. 5. TG–DTG–DTA diagram of dried “pure” red mud.

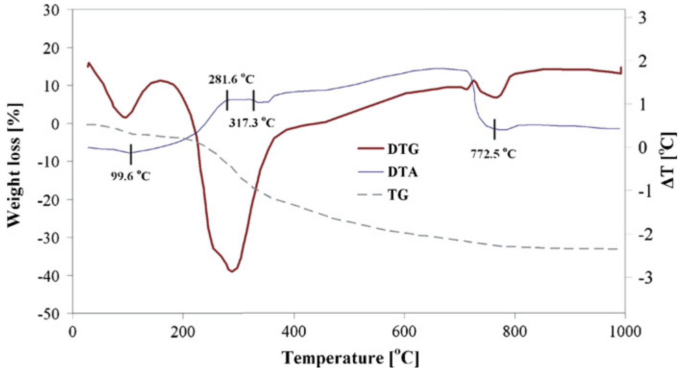

Fig. 6. TG–DTG–DTA diagram of dried red mud with 15% sawdust.

When the typical points of the TG–DTG–DTA curve are compared with the graph Fig. 3, the dependence of the measured emanation power on the firing temperature can be explained, since several structural changes within the material can be differentiated, based on the curves of the thermal examination, where temperature changes take place.

The following phases occur with the change in temperature.

- In the first phase (20–105 °C) less than 1% (0.83%) of the total volume of red mud is lost. In this phase, the physically bound pore water removes from the material. As a consequence, radon effuses more easily from the pores as it is not absorbed in pore water, but slows down in capillary water and gets into the air by diffusion. This explains the increase in the emanation coefficient value.
- The second significant loss of weight can be detected between 105 and 350 °C, when the transmitted heat is enough to dissipate chemically bound water (volume loss = 2.14%). In the next temperature range (100–300 °C), capillary water departs as steam; therefore, pore volume increases, which results in more radon diffusing into interpore space.
- In the third phase (350–790 °C), the loss of volume is most likely to be due to the departure of CO_2 that is discharged as a result of the decay of carbonate –mainly calcite – component (volume loss = 3.74%).
- In the last phase (over 800 °C), the emanation value significantly decreases. Over this temperature, the glass transition of the material commences. As a result, free pores are closed, and there is less chance for the radon to escape into pore space. It is notable that the emanation coefficient over 1000 °C is set on a constant level, most likely because the radon generated from radium on the grain surface is capable of getting into air-space.

The effect of firing temperature on the variation of pore volume, specific surface of red mud (0% sawdust) can be seen in Table 5.

The findings reveal that both specific surface and pore volume are reciprocally proportional to firing temperature above 400 °C and that pore volume changes with the amount of additional material (Fig. 7.).

Table 4
Emanation coefficient (*e*) of the red mud samples, various building materials and NORMs.

Origin of sample	Average emanation coefficient [%]	Range [%]	Reference
Ajka	18.5	12–22	
Almásfüzitő	12	6–14	
Slag (Ajka-Hungary)	14.3		[17]
Slag (Tatabánya-Hungary)	1.4		[17]
Soil	20	1–80	[27]
Cement, Egypt	17.9 ± 2.2		[10]
Ceramics, Egypt	14 ± 3.2		[10]
Gypsum A, Egypt	8.5 ± 0.9		[10]
Gypsum B, Egypt	0.5 ± 0.1		[10]
Clay, grain size < 0.05 mm, Spain	3.2		[28]
Clay grain size < 0.01 mm, Sweden		13.9–22.1	[28]
Slag, Austria	0.8	0.3–4.6	[29]
Gypsum board, Austria	29.5	28.2–30.9	[29]
Bricks, Austria	2.5	1.3–7.8	[29]
Portland cement, Greece	8	4–10	[30]
Fly-ash, Greece	< 1		[30]
Phosphogypsum, Greece	23	20–26	[30]
Brick, Italy		1.65–18.3	[31]
Concrete, Italy		6.6–16	[31]
Limestone, Italy		5.8–7.4	[31]
Phosphate rock-Cyprus	13		[32]
Phosphogypsum-Cyprus		22–28	[32]

Table 5
Variation of pore volume, specific surface of red mud (0% sawdust) as a function of temperature.

Temperature [°C]	F_{BET} [$m^2 g^{-1}$]	V_p [$cm^3 g^{-1}$]
200	13.9	0.09213
300	20.6	0.09797
400	20.3	0.09835
500	17.7	0.08663
600	16.3	0.08778
700	13.2	0.08233
800	6.96	0.02386
900	3.51	0.01273
1000	1.56	0.00468

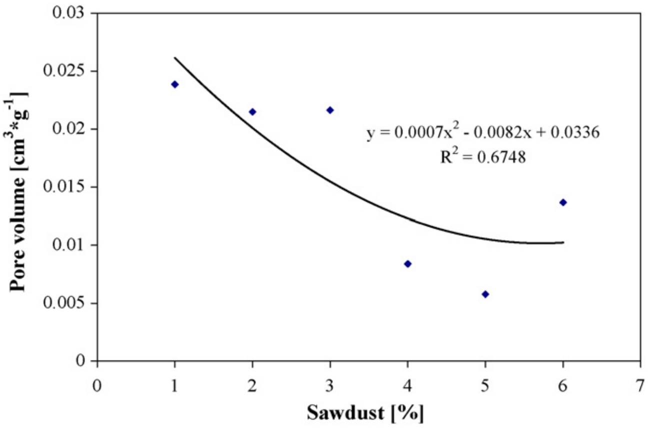

Fig. 7. Correlation between additional material (sawdust) and pore volume.

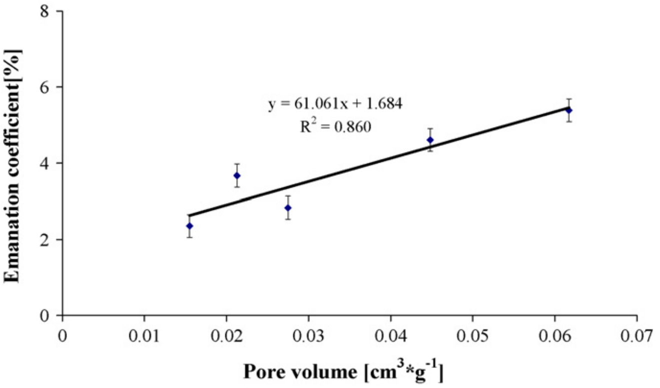

Fig. 8. Correlation between emanation coefficient and pore volume (15% sawdust).

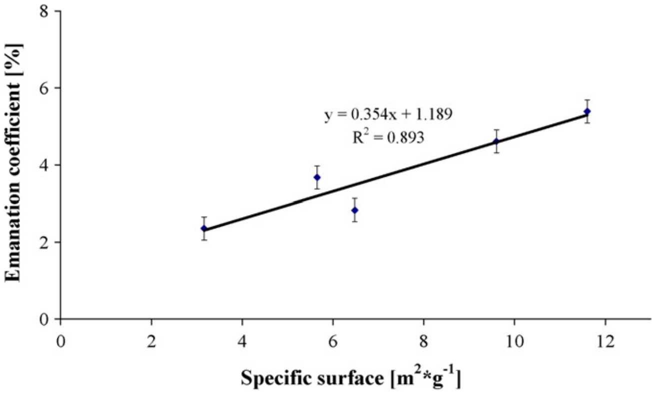

Fig. 9. Correlation between emanation coefficient and specific surface (15% sawdust).

The effects of pore volume and specific surface on emanation coefficient were also examined. For example, in Figs. 8 and 9, the correlations specific surface–emanation coefficient and, pore volume–emanation coefficient, respectively, are presented.

The results indicate that changes in pore volume and the specific surface may have a considerable effect on the emanation coefficient. This can be very important from the point of view of radiation protection for future applications.

Density changes may also have some effect on the radon emanating capacity of red mud; in this case, however, no close correlation was found between the two parameters.

5. Conclusions

The activity of natural radionuclides of the examined samples, and of Hungarian red mud in general is higher than the world average of clays. Hungarian red mud is allowed to be used with restrictions as an additive (maximum 20 wt%), in building materials, so that the resulting activity concentration index would not exceed EU recommendations.

Nevertheless, on the basis of the present emanation measurements, increased radiation exposure in dwellings could be expected due to red mud. The experimental evidence collected support that the emanation coefficient of red mud (a) is high and it varies within a wide range (6–22%), (b) it can be decreased significantly (may be up to 80%) by increasing the firing temperature ($T > 800\,°C$) and by optimizing the additives percentage (e.g. sawdust = 15–25 wt%), and (c) it depends on specific surface and pore volume. Following these observations, it seems advisable to radiologically qualify building materials containing red mud, prior to use in terms of both *I*-indices and emanation coefficient in order to safeguard against negative health effects.

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