RADON/THORON EXHALATION CHARACTERISTIC OF HEAT TREATED RED MUD, RELATIONSHIP BETWEEN INTERNAL STRUCTURE FEATURES, POSSIBILITIES OF REDUCTION

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Abstract

Inhaled Rn-222, Rn-220, and their progenies augment the risk of the evolution of lung cancer. Depending on the porosity, the radon isotopes can exhale from materials, e.g. building materials and can concentrate in enclosed spaces. In this study the effect of heat-treatment was surveyed on NORM origin red mud. The heat-treatment was performed in different temperatures (100-1000 °C). The exhalation were measured with accumulation chamber technique. Owing to the treatment the initial (100 °C) 41±5 mBqkg^{-1}h^{-1} Rn-222 exhalation reduced under 3±2 mBqkg^{-1}h^{-1} (1000 °C), whilst the initial 34±5 mBqkg^{-1}s^{-1} Rn-222 exhalation decreased under 6±2 mBqkg^{-s}s^{-1}.

Introduction

The environmental factors that pose a risk to human health, furthermore the accurate knowledge of their mechanism has become one of the most important task of present days.[1]

In building material production sector natural origin raw and industrial origin by-products can be used. In several cases these materials contain terrestrial radionuclides and also their decay products in significantly higher concentration than the natural world average value, which can cause elevated radiological risk on residents after inbuilt.[2,3]

The production and design of new types of synthetic building materials based on NORM by-products is raising concerns among authorities, public and scientists. It is incumbent on professional engineers and scientists to demonstrate that this material does not pose significant risks to human health and the environment.[4]

The natural radionuclide content of building material products contributes to natural background radiation in two ways. On the one hand the gamma radiation of the primordial radionuclides (K-40; and daughter elements of U-238, Th-232) increases the external gamma dose rate. On the other hand the radon isotopes are the major contributors to ionising radiation dose received by most of the population. The inhaled radon (Rn-222), thoron (Rn-220) and their progenies augment the risk of the evolution of pulmonary cancer. More than 50 % of the radiation dose received by individuals from natural radiation sources. In case of indoor conditions the elevated radon and thoron levels can cause significant dose contribution on residents.[1]

The EU BSS[5] Member States shall establish national reference levels for indoor radon concentrations. The reference levels for the annual average activity concentration in indoor air of workplaces (buildings, underground workplaces e.g. mines, caves, etc.) and residential buildings shall not be higher than 300 Bq m^{-3}.

During the alpha decay of mother elements (Ra-226, Ra-224) the newly formed Rn-222 and Rn-220 atoms are recoiled with 1-2 % of the total decay energy. Depending on the containing matrix (grain size, density, radium distribution, shape, moisture content) the recoiled radon can reach the surface of the grain and it can be slowed down in the pore space. The ratio between the amount of radon released into pore space and the parent isotope called emanation factor. Only the emanated radon and thoron have a chance to exhale from the matrix. The radon exhalation rate – defined in NEN 5699:2001 EN Standard[6] – is the radon activity that diffuses per unit of time from a material to the air surrounding the material, in Bq s^{-1}. By dividing the radon exhalation rate by either the area of the exhaling surfaces or by the mass of the sample, the areic (radon flux Bq m^{-2} s^{-1}) and massic radon exhalation rates (Bq kg^{-1} s^{-1}) can be calculated.

If the thickness of the investigated sample is remarkably smaller than the diffusion length of radon or thoron all the emanated radon and thoron has a chance to exhale i.e. the amount of the sample determine the exhalation rate, which can be related to unit mass. In the case of porous materials the diffusion length of radon[7] is higher than 0.4 m (e.g. for porous soil, limestone, brick it is 0.4 m, for limestone and sandstone 0.85 m, for aerated concrete 1.0, for gypsum 1.1 m) whilst the granulose materials has even higher diffusion length e.g. sand with approximately 2.0 m.

In case of thoron former surveys proved that in case of humid clay the thoron exhalation does not depend on the thickness if the sample is under 0.5 cm. Owing to that fact, the determination of massic exhalation rate can provide great possibility to compare exhalation features of samples in granular form.[8]

Materials and Methods

Sampling and sample preparation

The investigated red mud sample was collected from a 1 to 2 m depth of red mud reservoirs in Ajka (Hungary). The samples were heated to a constant mass at a temperature of 105 ± 3 °C, grinded under 0.63 mm and homogenised. In order to obtain Ra-226 and Th-232 activity concentration of the sample the powdered red mud was put into aluminium Marinelli vessel, weighted, sealed and stored during 27 days to reach the secular equilibrium between Rn-222 and its progenies. The gamma spectrometry of investigated sample was published in former study.[9]

Radon and thoron exhalation measurements

For massic exhalation measurement spherical-shaped red mud samples were prepared. The size of the samples was < 0.5 cm diameter to ensure exhalation condition which was not dependent from sample thickness.

The radon concentration was obtained with the help of accumulation chamber technique described in recent study.[9] After the accumulation time the radon and thoron content were measured together. After the sampling had been finished the thoron decayed in the detector chamber and only the radon was inside. The thoron concentration was obtained from the measured activity concentration difference. To get accurate result the decay correction factor of thoron in the air flow was determined with the help of thoron source. The thoron exhalation was calculated with the following formula:

$$E_{Tn\_Mass} = \frac{A_{eq} \cdot F_C}{0{,}2045 \cdot m \cdot t_{eq}}$$

- A_{eq} = decayed activity [Bq/m^3]
- E_{TnMass} = massic Tn exhalation rate [Bqkg^{-1} h^{-1}]
- t_{eq} = secular equilibrium time [389 s]
- F_C = decay correction factor [0-1] (decay of Tn in airflow)
- 0.2045 = decay correction factor under equilibrium state
- m = mass of the sample [kg]

Porosity measurements

The porosity features of the samples were measured using a combined method. The micro- (< 2 nm) and mezo-porosity (2 to 300 nm) were investigated with a Micromeritics ASAP 2000 device. The macro-porosity interval (above 300 nm) was investigated with the help of a SMH6 mercury poremeter. The applied method explained earlier.[9]

Results and Discussion

The Ra-226 and the Th-232 content of investigated red mud sample were 182 ± 18 Bq/kg and 136 ± 32 Bq/kg respectively, which is approximately six times higher than the world average of soils' (32 Bq/kg)' published in UNSCEAR 2008 Report.[3]

The radon and thoron exhalation were measured with widespread used ALPHAGUARD 2000 radon monitor connected to closed loop accumulation kit. It was found that the exhalation capacity reduced greatly as a result of heat-treatment. The initial radon and thoron exhalation reduced in case of the red mud (Figure 2):

- Radon exhalation: 75±10 $mBqkg^{-1}h^{-1}$ (100 °C treated) to 7±4 $mBqkg^{-1}h^{-1}$ (1000 °C treated)
- Thoron exhalation: 312±19 $Bqkg^{-1}h^{-1}$ (100 °C treated) to 80±7 $Bqkg^{-1}h^{-1}$ (1000 °C treated).

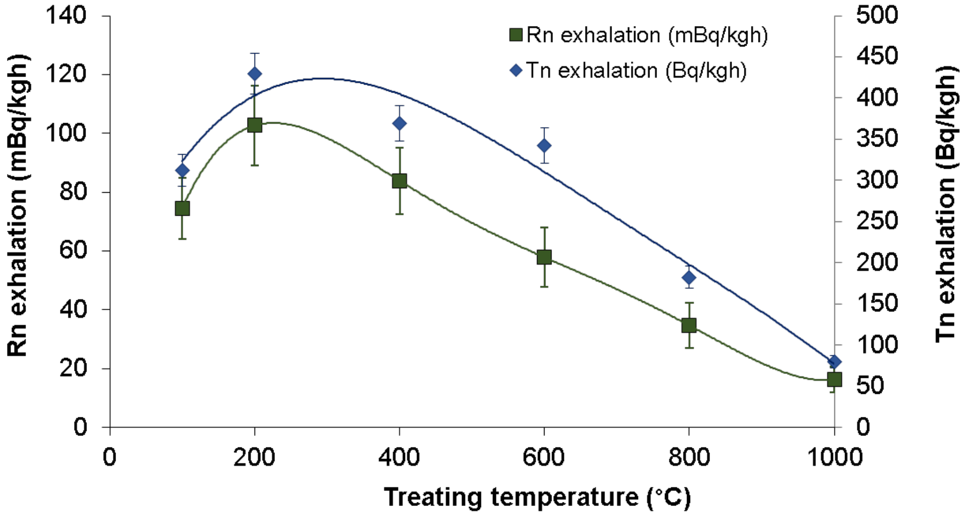

Figure 1: Radon/thoron exhalation characteristic of red mud in the function of the applied heat-treatment

The cumulative pore size distribution of the heat-treated samples can be seen in Figure 2. The obtained results were previously published in previous paper.[9]

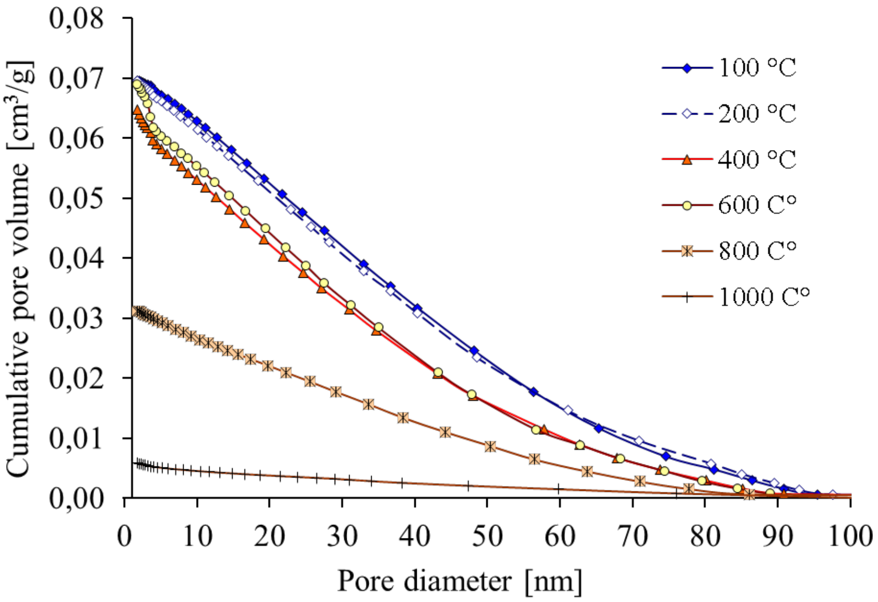

Figure 2: Cumulative pore volume in function of pore size between 1-100 nm

The thoron exhalation was investigated in function of the nano porosity. The curves clearly prove that significant differences were found between the porosity features of the investigated samples. The pores size distribution shows that in the lower case range (< 100 nm), the frequency of the pores was very low. This was assumed to be the reason why the massic exhalation capacities were very low in the high temperatures treatments in the case of radon and thoron exhalation as well.

Conclusions

On the basis of the obtained results it can be clearly stated that the radon and thoron exhalation can be effectively reduced (under ~10% of the initial) with the help of high temperature heat-treatment. In case of building material production the risk can originated from radon and thoron exhalation of building material can be reduced significantly with ~1000 °C heat-treatment.

As such, the heat-treatment had a beneficial effect on massic exhalation from the perspective of the red mud case highlighted previously. A strong correlation was found between the micro porosity and the radon emanation and massic exhalation features. This correlation was based on the obtained pore volume distribution in the function of pore size and the comparison of pore volume in micro porosity range (1.7-300 nm). It was found that pores under 100 nm were primarily responsible for the elevated radon emanation and as such, for massic exhalation, too.

Despite these promising results, the mixtures composed by red mud and other matrix components that are used in building material factories must be investigated, since certain components can have an effect on one another, which in turn can cause a potentially harmful final structure.

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