

1. Introduction

The countries with oil and gas industries, uranium ores extraction and other resource extraction industries produce enormous amounts of residues containing radionuclides of the uranium or thorium chain and natural occurring radioactive materials (NORM). One important pathway for radiological impact arising from radioactive waste and NORM residues is the release of radon isotopes into the atmosphere. The principal isotopes of radon, ^{222}Rn and ^{220}Rn , are generated from the radioactive decay of ^{226}Ra and ^{224}Ra , respectively, which are the decay daughters in ^{238}U and ^{232}Th decay series. The half-life of ^{220}Rn is 55 s, which is not included in the safety considerations due to its very short half-life. The half-life of ^{222}Rn is 3.8 days that allows it to move long distances before decay. Furthermore, the generation rate of ^{222}Rn continues for many thousands of years due to the long half-life of ^{226}Ra which is present in the tailings.

Atmospheric radon (^{222}Rn) and its decay products have potential radiological impacts on public health. Therefore, prediction/evaluation of radon flux and effectiveness of different covers are the major elements in radiation protection, long-term safety aspects, and to model radon release to the environment for final assessment of radiological impacts and required remediation actions [1,2]. The effectiveness of a cover material in attenuation of radon release depends on the material thickness and ability to control the diffusion of radon and trapped its solid daughters before it exhaled from the cover. Thus, radon attenuation, including the effect of various covers is an important element in rehabilitation planning. As a result, the radon cover design is one of the basic issues in waste management to reduce radon releases to the atmosphere and soil erosion in repositories. The key parameter that characterizes the degree of radon flux reduction is the diffusion coefficient.

The aim of this study is to evaluate experimentally radon exhalation from soil samples and effectiveness of the cover layer for optimal design of the radon cover. A measurement system was designed to measure the radon diffusion coefficients and soil radon flux density by short-time accumulation technique based on the transient-diffusion method. The validity of the theoretical equations governing radon release from the soil and of the laboratory model to quickly estimate the radon release from soils, diffusion coefficient, and the effect of covers was investigated [3]. Comparison of theoretical and experimental results show that both theoretical model and presented method to measure radon flux have sufficient accuracy and the results of the radon cover designed in this device can be generalized to real conditions.

2. Radon exhalation from porous materials

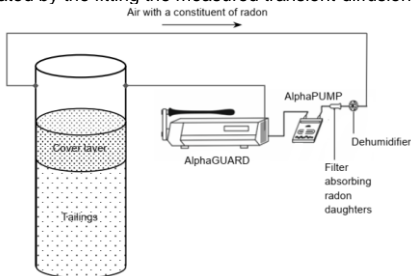
In this study, a simple and reliable accumulator method with a system of continuous radon detector (consisted of the AlphaGUARD™ PQ2000 radon monitor, the AlphaPUMP with 1 l min^{-1} flow rate, the dehumidifier, and the filter for absorbing radon daughters) was used to measure radon exhalation rate from bare and covered tailings.

Soil samples with different thickness were analyzed for radon exhalation rate. A plexiglass transparent chamber (28 cm diameter and 120 cm height) was used as the air-tight accumulator device for transient-diffusion measurements. The soil sample was placed on the tailings layer and the contact between the tailings and sample was covered with a fine metal mesh. The detection system was sealed to the container to measure the radon concentration in the air above the soil sample. The exhalation rate was determined through the measured radon concentration buildup. Once the measurement system was equilibrated, the time-dependent radon concentrations were recorded over 2-3 hours along with pressure, humidity and air temperature.

At the closed system, the radon detector collects variable concentrations with the growth curve given by

$$C(t) = C_0 + \frac{JA}{V\lambda_e}(1 - e^{-\lambda_e t}) \quad (1)$$

where J is the unbounded radon flux ($\text{Bq} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$), A is the cross-section area of the accumulator (m^2), λ_e is the effective time constant (s^{-1}), and V is the volume of the accumulator (i.e., air volume above the soil) (m^3). With no radon leakage, the effective time can estimate the back diffusion effect. The radon flux density was calculated by the fitting the measured transient-diffusion concentrations to Eq. (1).



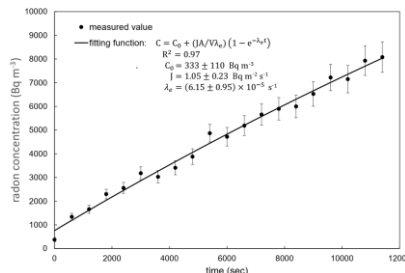
Set-up for short-time measurement of radon exhalation rate and diffusion coefficient

4. Conclusions and Acknowledgements

The radon release from bare and covered tailings model were estimated using diffusion theory with measured diffusion coefficient to calculate the thickness of an adequate cover system. It was observed that after 0.5 m and 1 m clay cover layer with diffusion coefficient $(1.78 \pm 0.24) \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$, the measured radon flux density from bare waste, $1.05 \pm 0.23 \text{ Bq m}^{-2} \text{ s}^{-1}$, decreases by a factor of 1.7 and 2.8, respectively, to $0.61 \pm 0.12 \text{ Bq m}^{-2} \text{ s}^{-1}$ and $0.37 \pm 0.06 \text{ m}^{-2} \text{ s}^{-1}$. Concerning to the measured radon diffusion length, the radon flux reduction factor increases to 10 for 1.6 m clay cover layer. The results show that the effectiveness of the studied cover layer is 3, which is similar to theoretical and experimental results in uranium tailings pond [4].

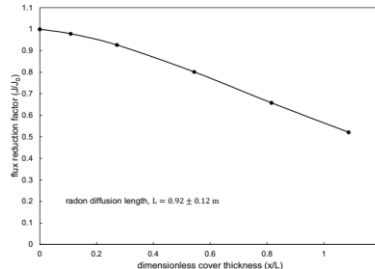
3. Evaluated radon exhalation rates

The regression of measured values gives the radon flux density from bare tailings with 95% confidence and close fit ($R^2 = 0.97$). The exhalation rate was $1.05 \pm 0.23 \text{ Bq} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ for the bare tailings. It is expected that the radon exhalation rate should be dependent on cover layer thickness and the effective diffusion coefficient of radon for the material used. For the chosen clay soil the radon diffusion coefficient was measured in previous studies and was $(1.78 \pm 0.24) \times 10^{-6} \text{ m}^2 \cdot \text{s}^{-1}$ [3].



Measured transient-diffusion radon concentration for bare tailings

Measurement procedure was performed for cover layer with different sample thicknesses. The flux reduction factor of radon exhalation rate as a function of cover thickness with effective radon diffusion length of $0.92 \pm 0.12 \text{ m}$ was determined.

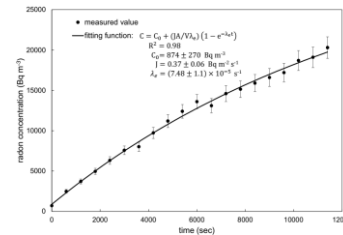
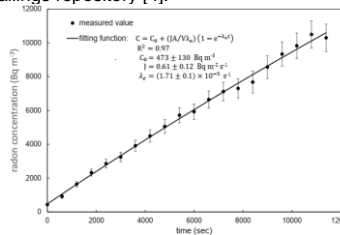


Radon flux reduction factor for various cover thickness

Fitting the time variation of radon concentrations for the covered tailings by clay sample with thickness of 0.5 m determined the flux density as

$0.61 \pm 0.12 \text{ Bq} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ with 95% confidence and $R^2 = 0.97$. For the cover layer of 1 m thickness, the best fitting ($R^2 = 0.98$) of the radon concentration values to Eq.

(1) gives the flux density $0.37 \pm 0.06 \text{ Bq} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$. The results showed that after 0.5 m and 1 m cover layer the radon exhalation rate decreases by a factor of 1.7 and 2.8, respectively. The reduction factor increases to 10 for 1.6 m clay cover layer with radon diffusion length of 0.92 m. Further, the effectiveness of the cover layer is 3, which is in consistent with the theoretical and experimental results in uranium tailings repository [4].



Measured transient-diffusion radon concentration and evaluated fitting parameters for tailings with cover layer thickness of 0.5 m (left), 1m (right)

References

- [1] International Atomic Energy Organization, TECDOC- 1403, IAEA, Vienna, 2004.
- [2] Ishimori Y., et. al., TRS-474, IAEA, Vienna, 2013.
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- [4] Ferry C., Richon P., Benetio A., Robe M.C., Journal of Environmental Radioactivity **63**, 49, 2002.