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White sand potentially suppresses radon emission from uranium tailings

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ABSTRACT

Uranium tailings represent a huge radioactive waste contaminant, where radon emanation is considered a major health hazard. Many trials have been conducted to minimize radon exhalation rate by using different covering materials. In the present work, three covering materials, commonly available in the local environment, (kaolin, white sand and bentonite) have been used with different thickness 10, 15, and 20 mm). ^{238}U, ^{232}Th, ^{40}K and the radon exhalation rate were measured by using gamma spectrometry with a Hyper Pure Germanium (HPGe) detector and solid state nuclear track detectors (CR-39). Radon exhalation rate, calculated before and after covering, ranged from 2.80 ± 0.14 to 4.20 ± 0.21 Bq m^{-2} h^{-1}, and from 0.30 ± 0.01 to 4.00 ± 0.20 Bq m^{-2} h^{-1}, respectively. Also, the attenuation coefficients of different covering materials and radon emanation were calculated. The obtained results demonstrate that covering of uranium tailings by kaolin, white sand and bentonite has potentially minimized both the radon exhalation rate and the corresponding internal doses.

1. Introduction

Extraction of uranium from its ore is accompanied with production of a huge amount of uranium tailings that represent large masses of low level radioactive contaminants (Landa, 2003; Tan et al., 2012a). These tailing are the major source of radon-222, which decay to polonium, lead and bismuth. Such radionuclides are usually adsorbed to atmospheric particulates creating radioactive aerosol that inhaled by man leading to a major health hazard (Beck, 2017). This scenario is aggravated by the low solubility of ^{226}Ra, emerged from ore minerals in both sulfuric acid and sodium carbonate. Previous studies have demonstrated that, many of the physical characteristics of both tailings and covering materials (such as particle size, moisture, density, thickness, porosity and permeability) largely affect the radon emanation from uranium tailings (Strong and Levins, 1982; Zhou and Deng, 2004). Radon has short half life of 3.8 days and it releases from the tailings and large quantities may reach the atmosphere and the ground water as well. Many epidemiological studies have confirmed that the inhalation of significant activities of radon and its progeny is related to elevated incidences of lung cancer, especially in those working in uranium-related activities. The fraction of radon which is released relative to its total production is known as the emanation coefficient, and can range from 0 to 1 but is generally between 0.2 and 0.5 (Mudd, 2008). Radon emanation is one of the most important parameters influencing indoor and outdoor radon concentrations in the surrounding residential areas. It is enhanced by increasing the moisture, temperature and specific surface area (Bossew, 2003). Releasing of radon from uranium tailings takes place through a sequential processes including: its emanation from the decay of radium into the interstitial space between the grains; its transport to the ground surface then exhalation to the atmospheric air Fig. 1 (Sasaki et al., 2004; Alharbi and Abbady, 2013). Studies of tailings management take many approaches including assessment of its environmental radioactivity and their radiological risk in both atmospheric air, above the tailings surface and the underground water in the vicinity of tailings ponds (Sahoo et al., 2007). Also, covering of tailings is widely applied to reduce radon exhalation rate (Ferry et al., 2002). The present work is designated to investigate the potential of effective covering of tailings using locally available materials (kaolin, white sand and bentonite) to minimize the radon emanation and exhalation rate and subsequently reduce the cost of covering of tailings.

2. Experimental procedures

2.1. Sample preparation

Three different kinds of sedimentary rocks (sandy dolostone, siltstone and claystone) were collected from southwestern Sinai Egypt. These rocks were crushed, ground to approximately −60 mesh (0.250 μm), mixed together and prepared for acid (H_2SO_4) agitation leaching process under the following factors; 25% H_2SO_4, at room

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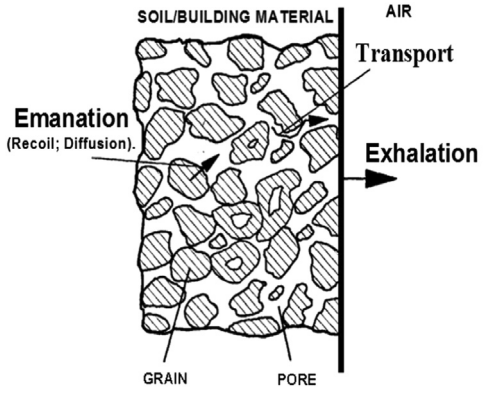

Fig. 1. Mechanisms show the difference between Radon exhalation and emanation (Alharbi and Abbady, 2013).

temperature, 1:3 Solid/Liquid ratio and one hour stirring time (El Aassy et al., 2012). The separation of leach liquor from the residual (waste) carried out by filtration and drying the residual at 120 °C. The three wastes obtained from leaching process (coded: Waste 1, Waste 2 and Waste 3) are investigated to be measured by high purity germanium detector and solid-state nuclear track detector. To eliminate the role of particles size and moisture, both the waste samples and the covering materials were sieved in a 60 mesh (0.250 μm), dried in a hot air oven at a temperature of 110 ± 0.1 °C for 24 h then kept in plastic containers with volume 100 ml (El Aassy et al., 2016). Also, to minimize the permeability of the covering layer, they were compressed on the top of the waste samples. The thicknesses were determined after compression.

2.2. Activity measurements

The system used for activity measurement consists of ORTEC closed-end coaxial HPGe detector (n-type) coupled with a PC-based 8096 channel analyzer and has a relative efficiency 60%. The detector is shielded to reduce the gamma-ray background by a lead cylinder with a fixed bottom and a movable cover to suppress the soft component of cosmic rays, consisting of photons and electrons, to very low level. A concentric copper cylinder is used to suppress the X-ray (73.9 keV) emitted from lead by its interaction with external radiation. To minimize the effect of the scattered radiation from the shield, the detector was mounted in the center of the chamber. The full width at half maximum (FWHM) was 2.3 keV for ^{60}Co-1332 keV gamma-ray line. The energy calibration was applied to determine the energies of any unrecognized gamma rays. The system was calibrated in the mode of keV/Ch. The gamma-ray energies emitted due to ^{60}Co (1173.2 and 1332.5 keV) and ^{137}Cs (661.9 keV) sources were used for the calibration. Also, efficiency calibration was made by using different energy peaks covering the range up to 2600 keV, where it was performed by using three reference materials (provided by the International Atomic Energy Agency) for U, Th and K activity measurements: RGU-1, RGTh-1 and RGK-1 (IAEA, 1973). Absolute efficiency calibration of the gamma spectrometry system was carried out using the radionuclide specific efficiency method in order to reduce the uncertainty in gamma-ray intensities, as well as the influence of coincidence summation and self-absorption effects of the emitting gamma photons. The IAEA reference materials RGU-1, RGTh-1 and RGK-1, have densities similar to the samples to be measured after pulverization. The sample containers were placed on top of the detector for counting. The same geometry and size were applied for both the samples and the reference materials.

To eliminate the background measurements, an empty bottle with the same geometry was measured and its value was subtracted from samples measurements. The data acquisition, display and spectrum analysis were carried out using Gamma vision software, where the concentrations of ^{238}U, ^{232}Th and ^{40}K were calculated. Calculations of count rates for each detected radionuclides depend on the establishment of secular equilibrium reached between ^{238}U and ^{232}Th and for their decay products. Radioactivity concentrations of each sample were measured for about 20 h.

The specific activities, in Bq kg^{-1} for the last radionuclides in the measured samples were computed using the following equation (Baykara et al., 2010; Lakehal et al., 2010; El-Taher et al., 2010; Bakr, 2010)

$$A_s = \frac{N}{\varepsilon.\ p.\ t.\ m} \quad (1)$$

where

N: Net Area of the peak at specified energy
ε: Absolute efficiency of the HPGe detector at this energy
p: Branching ratio of the gamma transition
t: Counting time (s)
m: Mass of the sample (kg)

Uranium-238 activity was indirectly calculated from the gamma rays emitted by its daughter products (^{234}Th and ^{234m}Pa) whose activities were determined from the 63.3 and 1001 keV photopeaks, respectively according to Sutherland and de Jong (1990). Also, ^{226}Ra specific activity was measured using the 186.1 keV photopeak from its own gamma-ray (after the subtraction of the 185.7 keV of ^{235}U). The specific activity of ^{214}Pb was calculated using the 241.9, 295.2 keV and 351.9 keV while the specific activities of ^{214}Bi and ^{210}Pb were measured using 609.3 and 46.5 keV, photopeaks respectively. The ^{235}U activity was determined directly by its gamma ray peaks 143.8, 163.4, 185.7, and 205.3 keV (Yücel et al., 1998; Ramebäck et al., 2010).

The specific activity of ^{232}Th was measured using the 338.4 keV and 911.2 keV from ^{228}Ac and 583 keV and 2614.4 keV from ^{208}Tl. The specific activity of ^{40}K was measured directly by its own gamma-ray at 1460.8 keV. The minimum detectable activity for ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K were 1.52, 0.64, 0.15 and 0.72 Bq kg^{-1}, respectively.

2.3. Radon measurements

CR-39 nuclear track detector of 500 μm thickness (Intercast, Italy) was used in this work. Measurements were made in three different wastes samples. Equal amounts of waste samples were placed in plastic containers (cups) with 7.5 cm height and 3.22 cm average diameter. In this work we used a thick-walled plastic cans. After sample mounting, their covers were tightly sealed to prevent any possible leakage of radon. Radon and its daughters are known to reach an equilibrium concentration after a week or more. Hence, the equilibrium activity of the emergent radon could be obtained from the geometry of the can and exposure time (Rawat et al., 1991). A piece of CR-39 detector of area 1 × 1 cm^2 was held at the top of the container (Fig. 2). Measurements were made, in triplicates, for the investigated samples without and with

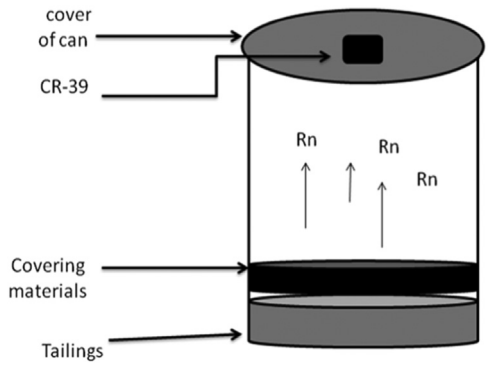

Fig. 2. Sealed-cup technique for radon measurement.