SIMULATION OF THE RADON FLUX ATTENUATION IN URANIUM TAILINGS PILES

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Abstract. Tailings wastes are generated during the milling of certain ores to extract uranium and thorium. In the recent past uranium mill tailings consisted of fine-grained sand and silt materials, usually disposed in large piles in an open air area. Radium is probably the most hazardous constituent of uranium tailings. It produces radon, a radioactive gas which can easily spread into the environment. Airborne radon decays into a series of short half-life products that are hazardous if inhaled. Tailings also emit gamma radiation which can increase the incidence of cancer and genetic risks. Post closure and site rehabilitation involves, among other situations, controlling and estimating radon release from the surface of the tailings pile. Generally the primary cleanup method consists of enclosing the tailings with compacted clay or native soil to prevent the release of radon and then covering this layer with rocks and vegetation. This implies a cover design and placement which will give long term stability and control to acceptable levels of radon emission and gamma radiation, preventing also erosion and water infiltration into the tailings.

An algorithm based on the theoretical approach of diffusion was developed to estimate radon attenuation originated by a cover system placed over the tailings pile and subsequently the resulting concentration in the breathing atmosphere. The one dimensional steady-state radon diffusion equation was applied to a porous and multiphase system to estimate the radon flux from the tailings to the surface. The thickness of a cover that limits the radon flux to a stipulated value was performed for a particular contaminated site. The efficiency of the cover attenuation was evaluated from the comparison with the resulting radon concentration in the absence of any cover system.

Keywords: radon, cover system, tailings, radium, flux, diffusion

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1. Introduction

Because uranium mills use chemical processes to selectively remove uranium from the ore, contaminants in uranium mill tailings include in most cases radium and thorium, which are the dominant radioactive materials in mill tailings. Tailings also contain small residual amounts of uranium that were not extracted during the milling process. Uranium mill tailings were normally dumped as sludge in special ponds or piles, where they were abandoned. Most of the radioactive contaminants are still present in these materials which have been brought by mechanical and chemical processes to a condition where the contaminants are now much more mobile and thus susceptible to migrate into the environment.

Radon-222 gas emanates from the tailings being continuously produced from the radioactive decay of radium-226. Some of this radon escapes from the interior of the pile and may be quickly spread with the wind being dispersed in the environment. To avoid environmental contamination, a rehabilitation plan should initially promote the confinement of the radioactive wastes, followed by a geotechnical stabilization of the pile and then by a multilayer covering system placed over the surface of the tailings disposal.

This work is based in a mathematical model for simulating the radon flux attenuation by designing a multilayer cover system and monitoring its effect by predicting the respective concentration at a defined mixing height. The model estimates the radon flux released from the radium content in the tailings and the radon concentration at the breathing or mixing height. The efficiency of the cover system designed is evaluated by comparing the resulting radon concentration at the breathing height considering an inexistent cover system.

2. Methods and Materials

A methodology is proposed to describe the radon exhalation attenuation from a uranium tailings disposal and it may be divided into two main sub-models:

1. The estimation of the flux released from the tailings to the air breathing zone, considered with or without a cover system, estimated by the diffusion equation applied to a multiphase media.
2. The estimation of the corresponding radon concentration predicted by a box model formulation.

Radon emanation has been addressed mostly as diffusive transport from the soil. The main soil properties and cover materials control the radon migration from the subsoil to the surface. In order to optimize a potential efficient cover design, soil and air transport processes in the tailings and in the cover layers were investigated.

2.1. RADON EXHALATION

The movement of radon in the soil or in the tailings can be described by the diffusion coefficient, D ($cm^2.s^{-1}$), which can be estimated by an empirical correlation with the fraction of saturation, m (Rogers and Nielson, 1984):

$$D = 0{,}07\,e^{\left[-4\left(m - m\,\varepsilon^2 + m^5\right)\right]} \tag{1}$$

The radon migration from the subsoil to the surface is controlled by the tailings and cover materials properties such as porosity (ε), moisture (θ), and the degree of saturation (m). This last parameter depends on the commonly measured moisture and may be estimated with the dry bulk density, ρ ($g.cm^{-3}$), and the specific gravity, g ($g.cm^{-3}$) of the diffusing media (tailings or cover) (Rogers and Nielson, 1984):

$$m = 10^{-2} \cdot \frac{\theta}{\frac{1}{\rho} - \frac{1}{g}} \tag{2}$$

The release mechanism from the soil is based in the principles of radon diffusion across a porous medium which may be estimated with the one-dimensional steady-state radon diffusion equation:

$$D\frac{\partial^2 C}{\partial x^2} - \lambda C + \frac{R\rho\lambda E}{\varepsilon} = 0 \tag{3}$$

The first term of this equation defines the diffusion transport given by the Fick's law in a one-dimensional form, which means that the diffusing substances are proportional to the concentration gradient ($J = -D.\partial C/\partial x$). The second term represents a first order decay kinetics ($dJ/dx = -\lambda.C$) and the last term represents the radon generation from the radium decay ($R.\rho.\lambda.E/\varepsilon$).

The necessary parameters for solving this equation are the radon diffusion coefficient, D ($m^2.s^{-1}$), the radon decay constant, λ (s^{-1}), the radium concentration in the pores space, C ($Bq.m^{-3}$), the radium concentration in the tailings, R ($Bq.kg^{-1}$), the bulk density of the dry material, ρ ($kg.m^{-3}$), the radon emanation coefficient, E (dimensionless), the total porosity, (ε) (dimensionless) and the moisture, (θ) (dimensionless).

The generic solution of the diffusion equation (3) gives the radon flux released, J ($Bq.m^{-2}.s^{-1}$), which may be applied in 3 main situations (figure 1): (i) the flux to the atmosphere without cover trapping, J_t; (ii) the flux through a simple cover system, $J_{C(1)}$ and (iii) the flux through a multilayer cover system, $J_{C(2)}$.

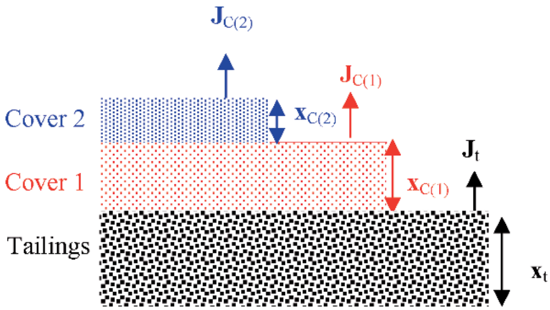

Figure 1. Parameters for a multilayer cover system, (NUREG, 1984).

The solution of the generic diffusion equation for a homogeneous medium represents the flux release, J_t (Bq.m^{-2}.s^{-1}), from the tailings with a thickness of x_t (m) without a cover system and is given by (Rogers and Nielson, 1984):

$$J_t = R\rho E\sqrt{\lambda D_t}\tanh\left(\sqrt{\frac{\lambda}{D_t}}x_t\right) \qquad (4)$$

For a two media problem, tailings (t) and a homogeneous cover material (c), the solution for the generic equation, J_c, is given by the equation (5) with $b_i = \sqrt{\lambda / D_i}$ (m) (i = c or t) and $a_i = \varepsilon_i^{\,2} D_i [1 - 0{,}74\, m_i]^2$ (m^2.s^{-1}) (Rogers and Nielson, 1984):

$$J_c(x_c) = \frac{2 \cdot J_t e^{-b_c x_c}}{\left[1 + \sqrt{a_t / a_c}\,\tanh(b_t x_t)\right] + \left[1 - \sqrt{a_t / a_c}\,\tanh(b_t x_t)\right] e^{-2 \cdot b_c x_c}} \qquad (5)$$

And the cover thickness, x_c, for a stipulated flux is obtained by rearranging the equation (5) for the tailings and the cover parameters (Rogers and Nielson, 1984):

$$x_c = \sqrt{\frac{D_c}{\lambda}}\ln\left[\frac{2 \cdot J_t / J_c}{\left(1 + \sqrt{a_t / a_c}\,\tanh(b_t x_t)\right) + \left(1 - \sqrt{a_t / a_c}\,\tanh(b_t x_t)\right)\left(Jc / Jt\right)^2}\right] \qquad (6)$$

2.2. RADON CONCENTRATION

For the estimation of the radon release to the breathing zone and the corresponding concentrations, a box model is used (figure 2). In a box model formulation the contamination source is defined by an emission area generating a constant emission rate (ϕ).

The box volume is defined by its length (L), width (w) and the mixing height (h) and inside, the concentration (C) is spatially homogeneous and constant in time resulting from the assumption of a complete mixing inside the compartmental box. Radon is directly diluted into the local air existing in the breathing zone above the contaminated source, and is carried away by the atmospheric circulation across it. The value for the wind speed (u) used in the calculation of radon dilution is matched by the average annual values through the mixing zone.

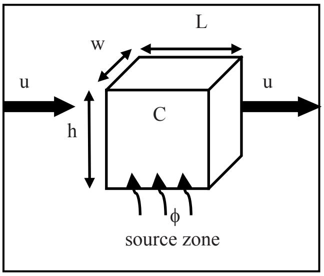

Figure 2. Box model for the radon concentration estimative at the breathing height, h.

In this type of model formulation, a mass balance concept is implicit:

$$V\frac{dC}{dt} = \phi A - uSC \tag{7}$$

and as consequence of a steady state assumption, we have that the pollutant concentration (C) is constant in time:

$$V\frac{dC}{dt} = 0 \tag{8}$$

that the mass flow rate entering (ϕA) into the box is equal to the flow rate leaving the box (uSC):

$$\phi A = uSC \tag{9}$$

and that the residence time (τ) is defined as the average amount of time that a particle of material remains in the box:

$$\tau = \frac{CV}{\phi A} \tag{10}$$

3. Case Study

3.1. PROBLEM DESCRIPTION

The intensive and extensive uranium mining and processing operations in Portugal has left a legacy of considerable environmental contamination. The uranium exploitation in Portugal began in 1913 and ended in 2000. During this period the uranium was mined in 62 different places. One particular place is Urgeiriça where the extensive exploitation and treatment of the uranium ore in this mine and others from the same region, has led to an accumulation of large amounts of solid wastes: about 4 million tons of rock material was routed into natural depressions confined by dams that cover an area of about 13.3 hectares. The radon exhalation from the tailings is potentially one of the main sources of contamination for nearby areas.

The Urgeiriça tailings pile was considered as a case study. The area and the average thickness of the mil tailings deposit were estimated at 12 hectares and 14 m, respectively. The radium concentration in the mill tailings was measured and the resulted value for ^{226}Ra was 12,900 Bq/kg. A value of 0.24 was assumed for radon emanation coefficient. A total porosity of 0.37 was considered for the tailings materials and a value of 1,67 g.cm^{-3} was used for bulk density (Pereira et al., 2004).

The radon flux in the surface of the pile depends on the diffusion coefficient, which is estimated with the moisture content measured in the tailings, the total porosity and the ^{226}Ra activity in the tailings. An average value of 9,2 $\times$ 10^{-7} m^2.s^{-1} was obtained for radon diffusion coefficient in the tailings (Pereira et al., 2004).

A cover design of 5.15 m average thickness composed by sand, clay and gravel was proposed on the basis of the plan rehabilitation for the site (Pereira et al., 2004). An average value of 2.2 g.cm^{-3} was considered for the cover bulk density. A total porosity of 0.30 was considered for the cover material and the diffusion coefficient was estimated at 5×10^{-7} m^2.s^{-1}.

3.2. RESULTS

The methodology used has as outputs: i) the radon flux without cover and the resulting concentration at one m above the ground considered the breathing height; ii) the radon flux attenuation produced by the cover system proposed and iii) the estimation of a cover thickness that allows a radon flux less than to a value stipulated as safe or permissible.

The value obtained for the average radon flux without cover was $J_t = 7.19$ $Bq.m^{-2}.s^{-1}$ and the resulting concentration at 1 m above the ground at the site was $C_0 = 547.4$ $Bq.m^{-3}$. The average value measured in the Urgeiriça tailings pile for the radon concentration was 557 $Bq.m^{-3}$ (Exmin, 2003). This corresponds to a dose of 5.2 mSv/year, resulting from the exposure to radon in outdoor air, assuming that the receptor spends 1760 h outdoor in a year (a 20% outdoor residence time), with an equilibrium factor between ^{222}Rn and its decay products of 0.6 and for a dose conversion factor of 9 $nSv.h^{-1}$ per $Bq.m^{-3}$ (Grasty and LaMarre, 2004).

The radon flux attenuation originated by the cover system proposed with a thickness of 5,15 m, a porosity of 0,30 and a diffusion coefficient of $5,0 \times 10^{-7}$ $m^2.s^{-1}$ is equal to $J_c = 187$ $\mu Bq.m^{-2}.s^{-1}$ and the corresponding concentration at the breathing height is equal to $C_0 = 0,0142$ $Bq.m^{-3}$, which is negligible.

We tested the same materials efficiency considering that the tailings pile will be covered with 0,5 m of clay plus a layer of overburden to achieve a surface flux less than the permissible one, which was considered to be 0.74 $Bq.m^{-2}.s^{-1}$ or 20 $pCi.m^{-2}.s^{-1}$ (EPA, 1983). We considered the clay cover layer and the same diffusion coefficient used in the previous example. For the overburden layer we considered a diffusion coefficient of 2.2×10^{-6} $m^2.s^{-1}$ and a porosity equal to 0.37.

The radon flux attenuation through the clay component cover is $J_{c1} = 2.63$ $Bq.m^{-2}.s^{-1}$ with a concentration of 200,23 $Bq.m^{-3}$. The diffusion coefficient for the new source term (tailings plus clay layer) is estimated at 6.98×10^{-7} $m^2.s^{-1}$ and this value is used to estimate the second layer cover thickness which only allows the exhalation of the stipulated flux. The value obtained for the second layer is $x_{c2} = 1.52$ m which gives a total cover thickness of 2.02 m. The resulting concentration at the breathing height is 56.34 $Bq.m^{-3}$ which corresponds to a dose, in the same previous conditions, of 0.53 mSv/year. This dose is less than 1 mSv/year, the limit derived from the European guidelines concerning the exposure of the general public to artificial radionuclides.

4. Conclusions

The placement of an engineered cover designed to isolate the tailings and any other material is efficient in attenuating radon emanation to a safe level. It also acts as a barrier preventing rainwater infiltration into the tailings pile as well as wind and water erosion. The optimized cover construction proposed to be built reduces the radon exhalation rate, and subsequently the radon concentration at the site, to negligible values once the natural background is less than 30 Bq.m^{-3}. If the desired performance criterion is to achieve the radon standard for ^{222}Rn emission rate from the surface of inactive uranium mill tailings piles (EPA, 1983), a lower thickness cover could be used. In the example given, the two layers of cover produces reduction of the radon flux and concentration at the breathing height by a factor of nearly 10.

When the rehabilitation plan is implemented, in particular the cover system proposed, monitoring and maintenance actions should be carried out to achieve long term efficiency.

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