Radiation Measurements 68 (2014) 1–6

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Review

Inverse method for determining radon diffusion coefficient and free radon production rate of fragmented uranium ore

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HIGHLIGHTS

- Inverse method for determining two transport parameters of radon is proposed.
- A self-made experimental apparatus is used to simulate radon diffusion process.
- Sampling volume and position for measuring radon concentration are optimized.
- The inverse results of an experimental sample are verified.

ARTICLE INFO

Article history:
Received 21 August 2013
Received in revised form
22 June 2014
Accepted 24 June 2014
Available online 2 July 2014

Keywords:
Radon
Diffusion coefficient
Production rate
Fragmented uranium ore
Numerical inverse method

ABSTRACT

The radon diffusion coefficient and the free radon production rate are important parameters for describing radon migration in the fragmented uranium ore. In order to determine the two parameters, the pure diffusion migration equation for radon was firstly established and its analytic solution with the two parameters to be determined was derived. Then, a self manufactured experimental column was used to simulate the pure diffusion of the radon, the improved scintillation cell method was used to measure the pore radon concentrations at different depths of the column loaded with the fragmented uranium ore, and the nonlinear least square algorithm was used to inversely determine the radon diffusion coefficient and the free radon production rate. Finally, the solution with the two inversely determined parameters was used to predict the pore radon concentrations at some depths of the column, and the predicted results were compared with the measured results. The results show that the predicted results are in good agreement with the measured results and the numerical inverse method is applicable to the determination of the radon diffusion coefficient and the free radon production rate for the fragmented uranium ore.

1. Introduction

Radon is a radioactive gas, which is colorless, odorless, tasteless and chemically inert, and is 7.5 times heavier than air. It is a powerful natural alpha-emitter with a 3.8-day half-life. Many studies have shown that people who inhale radon and its progeny in high concentration for a long time are subject to lung cancer (Tomášek, 1993; Kusiak et al., 1993; Tomášek et al., 1994; Lubin et al., 1995; Darby et al., 2005; Krewski et al., 2005). Therefore, the concentration of radon and its progeny has been widely measured in the air of the ground floor, caves, tunnels, uranium mines and other underground cavities (Chen et al., 2010; Lespukh et al., 2013; Ishimori et al., 2013). Soil, building materials, and fragmented uranium ore such as uranium ore heaps for heap leaching, blasted ore heaps in underground stope, waste-rock piles and uranium tailings piles are the main radon sources. In recent years, these materials from uranium metal mining and extraction processes have become increasingly interesting from the point of view of radiological impact (Ishimori et al., 2013). In order to control radon exhalation flux of these media, the mathematical modeling of the radon migration through porous media is often used. In general, the fragmented uranium ore is also treated as a porous media, and the diffusion and convection are the main driving force for radon migration. Consequently, the diffusion and diffusion–convection models are the most well known ones. Their practical applications are complicated due to the main modeling parameters such as the radon diffusion coefficient and the free

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http://dx.doi.org/10.1016/j.radmeas.2014.06.009
1350-4487/

radon production rate, which is the activity of radon from the decay of radium that escape the solid phase and become free to migrate in unit volume porous medium per unit time (Zhang, 1982). Because the radon diffusion coefficient of porous media is mainly dominanted by porosity, tortuosity, moisture content and temperature, and because the free radon production rate is mainly dependable on uranium ore grade, particle size, moisture content and the uranium–radium equilibrium coefficient, it is difficult to determine their values for a specific porous media. So far, many studies have been conducted for determining the radon diffusion coefficient (Cohen et al., 1986; Keller and Hoffmann, 2000; Cozmuta and van der Graaf, 2001; Mujahid et al., 2005; Jiránek and Fronka, 2008; Jiránek and Svoboda, 2009; Rovenská and Jiránek, 2012) and the free radon production rate (Zhang et al., 2010; Sakoda et al., 2010a, 2010b,; Sahu et al., 2013) by theoretical and experimental methods.

In order to meet the needs for uranium, China has enlarged its production scale. As a result, heap leaching for uranium has been widely used and the uranium tailings from heap leaching have been piled in the uranium tailings impoundment. For evaluating the radiological impact from the heap leaching and the tailing impoundment and reducing the radon exhalation rate to an acceptable level, the radon diffusion coefficient and the free radon production rate for the fragmented uranium ore for heap leaching have to be determined. The two physical parameters are currently determined separately, and this takes relatively long time. In this paper, the radon diffusion transport theory for porous media and the numerical inverse theory for parameters (Marquardt, 1963; Kool et al., 1987; Hollenbeck and Jensen, 1998) were used to establish the inverse method for simultaneously determining the radon diffusion coefficient and free radon production rate of the fragmented uranium ore. In order to verify the applicability of this method, the fragmented uranium ore for heap leaching was used as the experimental sample, and a self-manufactured experimental column was used to simulate the diffusion process of radon in the fragmented uranium ore and to measure the pore radon concentrations at certain depths of the column. Some measured results were used to inversely determine the radon diffusion coefficient and the free radon production rate, and others were used to test the accuracy of predicted values of the pore radon concentrations.

2. Theoretical model

2.1. Radon diffusion transport in fragmented uranium ore

Based on the radon diffusion transport theory for porous media (Cozmuta and van der Graaf, 2001; Zhang et al., 2010), radon generation and transport within the fragmented uranium ore for heap leaching can be described using the equation of balance for each phase of the soil, water and interstitial air phases in the ore. The complexity of the problem can be reduced by transforming this equation into the one for the simple phase of the air. In order to obtain this analytical equation, it is necessary to make the following assumptions: Radon transports along the axial direction of a column; Pores are homogeneously distributed in the sample; ^{226}Ra content is homogeneously distributed in the sample; Pressure is constant within the column, and diffusive transport is only considered; Radon exhalation only occurs in the interface between the sample and the atmosphere. The radon transport equation for the fragmented uranium ore is as follows:

$$\frac{\partial C(x)}{\partial t} = D\frac{\partial^2 C(x)}{\partial x^2} - \lambda C(x) + \frac{\alpha}{\eta} \tag{1}$$

where $C(x)$ is the radon concentration in the pore of the fragmented uranium ore at depth of x (m) (Bq m^{-3}), D is the diffusion coefficient of the fragmented uranium ore (m^2 s^{-1}), λ is the radon decay constant (s^{-1}), α is the free radon production rate (Bq m^{-3} s^{-1}), and η is the porosity of fragmented uranium ore.

If the distribution of the radon concentration in the fragmented uranium ore comes to stable, then Eq. (1) can be expressed as follows:

$$D\frac{\partial^2 C(x)}{\partial x^2} - \lambda C(x) + \frac{\alpha}{\eta} = 0 \tag{2}$$

For the fragmented uranium ore with thickness L, the boundary conditions for Eq. (2) can be expressed as follows:

$$x = 0,\ C(0) = C_0$$

$$x = L, \frac{\partial C(L)}{\partial x} = 0$$

Then, the solution for Eq. (3) can be expressed as follows:

$$C(x) = \frac{\left(\lambda C_0 - \frac{\alpha}{\eta}\right)\left[e^{\sqrt{\lambda/D}(L-x)} + e^{\sqrt{\lambda/D}(x-L)}\right]}{\lambda\left(e^{\sqrt{\lambda/D}L} + e^{-\sqrt{\lambda/D}L}\right)} + \frac{\alpha}{\eta\lambda} \tag{3}$$

When the environment near the surface of fragmented uranium ore is well ventilated, the atmospheric radon concentration can be ignored. As a result, $C_0 = 0$ Bq m^{-3}, and Eq. (3) can be expressed as follows:

$$C(x) = \frac{\alpha}{\eta\lambda}\left[1 - \frac{e^{\sqrt{\lambda/D}(L-x)} + e^{\sqrt{\lambda/D}(x-L)}}{e^{\sqrt{\lambda/D}L} + e^{-\sqrt{\lambda/D}L}}\right] \tag{4}$$

Radon decay constant λ is 2.1×10^{-6} s^{-1}, and the porosity η of the fragmented uranium ore is fixed and can be obtained trough a simple test, so pore radon concentrations at different depths depend on the diffusion coefficient D and the free radon production rate α for the fragmented uranium ore.

2.2. Inverse method for parameter estimation

Inverse methods are typically based upon the minimization of an objective function which expresses the discrepancy between the observed values and the predicted values by Eq. (4). According to Eq. (4), pore radon concentrations in the fragmented uranium ore are observed as a function of the depths. The inverse method for parameter estimation involves the minimization of an objective function (OF) that sums all the squared deviations between the measured data and the predicted data at some observed depths, with the predicted results controlled by the adjustable parameters to be optimized. The objective function OF (b) for pore radon concentrations at different depths can be expressed as follows:

$$\mathrm{OF}(b) = \sum_{i=1}^{n}\left[C_m(x_i) - C_P(x_i, b)\right]^2 \tag{5}$$

where n is the number of the observations for pore radon concentration, b is the vector for the optimized parameters, $b = \{\alpha, D\}$. $C_m(x_i)$, and $C_p(x_i, b)$ are the measured value and the predicted value by Eq. (4) at depth x_i, respectively.

According to Eq. (4), the maximum pore radon concentration $C_{\max}$ of the fragmented uranium ore can be expressed as follows:

$$C_{\max} = \frac{\alpha}{\eta\lambda} \tag{6}$$

The influence of soil moisture content on the diffusion coefficient of radon in porous media has been investigated by Rogers and