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Review

A comprehensive review of radon emanation measurements for mineral, rock, soil, mill tailing and fly ash

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ABSTRACT

To our knowledge, this paper is the most comprehensive review to cover most studies, published in the past three decades at least, of radon emanation measurements. The radon emanation fraction, a possibility of radon atoms generated in a material escaping from its grains, has been widely measured for a variety of materials. The aim of this review is to organize a huge number of such data accumulated. The representative values of the emanation fraction for minerals, rocks, soils, mill tailings and fly ashes were derived to be 0.03, 0.13, 0.20, 0.17 and 0.03, respectively. Current knowledge of the emanation processes was also summarized to discuss their affected factors.
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Contents

1. Introduction

Radon (^{222}Rn) is the radioactive inert gas, which is produced by the alpha decay of radium (^{226}Ra) in the uranium series. All terrestrial materials have natural radionuclides such as uranium isotopes, and emit radon gas more or less. Radon atoms emanated from materials migrate with a half-life of 3.8 days to be distributed in indoor and outdoor airs. It is well known that radon and its short-lived progeny in the atmosphere are the most significant contributors to human exposure from natural sources (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2000). Recent epidemiological studies in North America (Krewski et al., 2005) and Europe (Darby et al., 2004) have made a strong impact, implying an association between indoor radon exposure and lung cancer (World Health Organization (WHO), 2009).

Radon emanation can be defined as escape of a radon atom from a Ra-bearing grain into pore spaces. The number of radon atoms released per the number of radon atoms generated is known as the emanation fraction, emanation coefficient or emanation power, etc. The term "radon emanation fraction" is used in this paper.

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Analytical models have been developed and improved to predict the flux of radon from the earth's surface into the atmosphere and building (UNSCEAR, 2000). All model expressions need several parameters, one of which is the radon emanation fraction. Many researchers have experimentally measured the radon emanation fraction for various natural samples, and demonstrated the influences of environmental factors. Over twenty years ago, Nazaroff et al. (1988, 1992) summarized emanation data from fifteen references, indicating an approximate range of 0.05–0.7 for soil. UNSCEAR (2000) reported a radon emanation fraction of 0.2 as a representative value for soil. Much data have been steadily accumulated since the last review by Nazaroff et al. (1988, 1992). Thus, updating this review should be now attempted to newly provide representative emanation fractions of radon from natural sources. In addition, it would be useful to organize the measurement results from the standpoint of experimental (environmental) conditions.

An extensive literature review of radon emanation measurements, especially in the last three decades, was done in the present paper. First, the current knowledge of the emanation processes and their affected factors was summarized for discussion. We then attempted to estimate the representative values of the radon emanation fraction for the following five materials, which is the main aim of this review. Measured samples were grouped according to material type: (1) mineral, (2) rock, (3) soil, (4) mill tailing (mostly uranium mill tailing), and (5) fly ash. Moreover, we discussed the difference of the radon emanation fractions among such materials and the influences of some factors.

2. Radon emanation phenomenon

2.1. Emanation processes

Current information on the radon emanation phenomenon and its related factors is referred to in this section, which is summarized in "Emanation process" of Table 1 and Fig. 1. The radon emanation is considered to consist of two components: alpha recoil and diffusion. Because of the very low diffusion coefficient (10^{-31}–10^{-69} $m^2\,s^{-1}$) of radon in the solid grain (Nazaroff et al., 1988; Nazaroff, 1992), which corresponds to the diffusion length (10^{-13}–10^{-32} m), the main component is believed to be the alpha recoil. Radon atoms, generated by the alpha decay of its parent nuclide (radium), recoil with an initial energy of 86 keV. This energy can be calculated on the basis of the law of conservation of linear momentum. The birthplace of radon in a grain, recoil direction, etc. determine whether the newly formed radon can escape to pore spaces (emanation: points A, B, E and F in Fig. 1) or stay in the grain (not emanation: points C, D and G in Fig. 1). The distance that recoil radon can travel in a grain relies on density and composition of the material. The range of radon is 34 nm in quartz (common mineral), 77 nm in water, and 53 μm in air, which the present authors calculated using a SRIM-2006 code (Ziegler et al., 1985). Only radium atoms within the recoil range from the grain surface can produce radon atoms that have any possibility of being emanated. Even if radon was released from a radium-bearing grain, it can penetrate the fluid-filled pore space, depending on its residual energy, to collide with a neighboring grain. In this case, radon can be embedded with the threshold energy (Semkow, 1991). After the embedding, one possible fate of radon is the migration from the pocket created by its recoil passage into pore (point E in Fig. 1); the other is the radioactive decay after the molecular diffusion in the grain (points D and G in Fig. 1). The former contributes to the emanation, but the latter not. On the other hand, radon completely escaping into inner pore space in the grain must diffuse to outer pore (point F in Fig. 1). For the emanation, the radon atoms that cannot diffuse out into the outer pore or are adsorbed on the inner surface of the grain should be regarded as being not emanated. Based on a part of the above considerations, radon emanation models have been developed to explain the effects of environmental factors on the

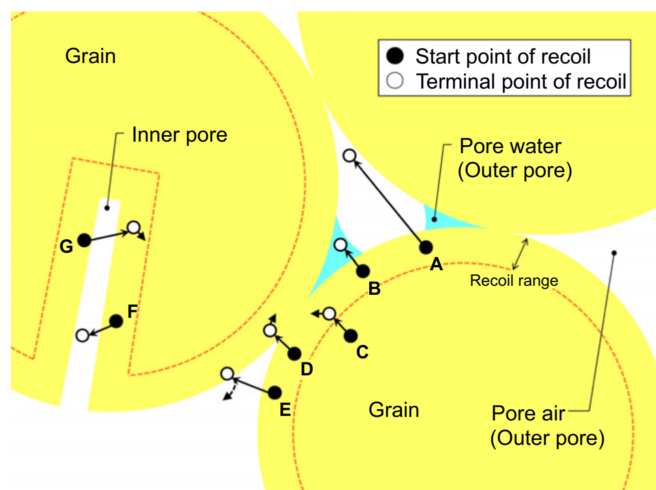

Fig. 1. Scheme of radon emanation phenomenon. Emanation: (A), (B), (E) and (F). Not emanation: (C), (D) and (G). If radon cannot diffuse out from inner pore into outer, radon in point (F) should not be regarded as being emanated. Arrows following terminal points of recoil represent diffusion process, which are not to scale.

Table 1
Radon emanation processes and their affected physical and experimental factors.

Emanation process	Physical factor	Experimental factor
Direct component • Alpha recoil from the outer surfaces of grains • Alpha recoil from the inner surfaces of grains • Diffusion in grains Indirect component • Diffusion in the inner pores of grains • Adsorption on the inner surfaces of grains • Embedding into an adjacent grain • Diffusion-based release after the embedding	• Radium distribution in grains • Grain size and shape • Moisture content • Temperature • Atmospheric pressure • Outer pore size • Inner pore size • Radiation damage • Solid density (crystal structure and elements)	• Instrument properties (Calibration, linearity, etc.) • Instrument environment (temperature, humidity, atmospheric pressure, etc.) • Sample properties (fracturing, sieving, single- or aggregate-grain structure, etc.) • Sample environment (moisture, temperature, vacuum, helium atmosphere, acid or alkaline leaching, etc.) • Sample packing thickness • Definition of radon emanation

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