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Radon emanation from low-grade uranium ore

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ABSTRACT

Estimation of radon emanation in uranium mines is given top priority to minimize the risk of inhalation exposure due to short-lived radon progeny. This paper describes the radon emanation studies conducted in the laboratory as well as inside an operating underground uranium mine at Jaduguda, India. Some of the important parameters, such as grade/^{226}Ra activity, moisture content, bulk density, porosity and emanation fraction of ore, governing the migration of radon through the ore were determined. Emanation from the ore samples in terms of emanation rate and emanation fraction was measured in the laboratory under airtight condition in glass jar. The *in situ* radon emanation rate inside the mine was measured from drill holes made in the ore body. The *in situ* ^{222}Rn emanation rate from the mine walls varied in the range of 0.22–51.84 $\times$ 10^{-3} Bq m^{-2} s^{-1} with the geometric mean of 8.68 $\times$ 10^{-3} Bq m^{-2} s^{-1}. A significant positive linear correlation ($r = 0.99$, $p < 0.001$) between *in situ* ^{222}Rn emanation rate and the ore grade was observed. The emanation fraction of the ore samples, which varied in the range of 0.004 –0.089 with mean value of 0.025 $\pm$ 0.02, showed poor correlation with ore grade and porosity. Empirical relationships between radon emanation rate and the ore grade/^{226}Ra were also established for quick prediction of radon emanation rate from the ore body.

1. Introduction

Uranium ore contains natural uranium comprising of 99.275% of ^{238}U, 0.715% of ^{235}U and 0.005% of ^{234}U. From radiation protection point of view, ^{238}U and its decay products are of major concern for uranium mining industry due to the large abundance of ^{238}U in natural uranium. The entire spectrum of decay products of ^{238}U can be found in the ore depending on the age of the deposit, which has important bearing on secular equilibrium status of the ore (Levinson et al., 1984). External gamma level and inhalation exposure due to radon (^{222}Rn), its short-lived progeny and long-lived alpha activity associated with ore dust constitute the major source of radiological hazard in uranium mines. However, in low-grade uranium mines (<0.1% U_3O_8), the hazards due to external exposure and long-lived activity are insignificant. Thus large fraction of occupational exposure is attributed to the potential alpha activity/energy of short-lived radon progeny (^{218}Po, ^{214}Bi, ^{214}Pb and ^{214}Po). Although concentration and activity of ^{214}Bi and ^{214}Pb are often used for assessment of the PAEC (Potential Alpha Energy Concentration), the PAEC is usually attributed to alpha emitters such as ^{214}Po and ^{218}Po. The isotopes ^{220}Rn and ^{219}Rn having half-lives of 54.5 s and 3.92 s respectively can be eliminated from the monitoring system by introducing filters or other delay techniques (Thompkins, 1982). ^{222}Rn ($t_{1/2}$ = 3.82 days) is found relatively in high concentration in mine atmosphere and can move a substantial distance from its point of origin (Nazaroff and Nero, 1988; Mudd, 2008). The increased risk of lung cancer due to the exposure of short-lived decay products of ^{222}Rn has been reported elsewhere (Field et al., 2000; Gulson et al., 2005; Al-Zoughool and Krewski, 2009). Monitoring of radon concentration inside uranium mines and in the environment has been a matter of concern since last several decades to minimize the extent of inhalation exposure of occupational workers and the public (IAEA, 1992; ICRP, 1993, 2010).

The radon emanation and concentration profile in mine air due to exhalation of the gas from ore body, underground water coming out through cracks and fissures, backfill material and broken ore pile (Raghavayya, 1968; Raghavayya and Khan, 1973; Panigrahi et al., 2005; Gherghel and De Souza, 2008; El-Fawal, 2011) depends primarily on the radon emanation rate from the grains and afterwards on the microstructure of the material. In addition, it depends on the parameters affecting physical processes such as diffusion, advection, absorption and adsorption. The amount of radon produced from the grains that finally enters into the pore space by recoil effect and diffusion process in the porous system of the material is defined as

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'effective radium', and the radon escape to production ratio is called 'radon emanation factor' (Tanner, 1980; Rogers and Nielson, 1991; Stoulos et al., 2004; Girault and Perrier, 2012). The rate of radon emanation is proportional to the rate at which radon is produced within the host material, which is a function of the ore grade (uranium content of the ore), the radon concentration gradient in the host pores, barometric pressure and diffusion properties such as porosity and emanation fraction of the material. It has been observed that change in barometric pressure affects the radon gas concentration in pores of the materials (Schroeder, 1966; Pohl-Rueling and Pohl, 1969; Clements and Wilkening, 1974; IAEA, 1981; Zhu and Zhang, 1984). When there is a pressure drop in mine environment, the radon laden air filling the pores moves out into the mine opening carrying the accumulated radon along with it. Besides, radon emanation depends on the bulk properties of rock, such as distribution of the mineral grains, size and specific surface area of the grains, degree of fracturing and fissuring and presence of water in the cracks (Bochiolo et al., 2012). Radon emanation, which is the fraction of radon-222 atoms released in the connected pore space of a porous material, increases with the water content due to low recoil range of radon atom in water compared with air (Semkow, 1991; Ferry et al., 2001; Barillon et al., 2005; Adler and Perrier, 2009). Choubey et al. (1999) reported that the presence of discontinuities (fractures) in the rock mass provides potential pathways for radon migration and favours air and water circulation resulting in higher radon exhalation. It has also been reported that the radon emanation rate in porous rock is less affected by ^{226}Ra content variations than the non-porous rock (Thompkins, 1982; Righi and Bruzzi, 2006). Therefore, high porosity and micro-fracture are the dominant factors that affect the rate of radon gas emanation from rock surfaces in mine openings unless the ore grades are high.

Numerous studies pertaining to radon emanation have been carried out in uranium mining and ore processing facilities in USA, Australia, Canada, China, India and Japan (Barretto, 1973; Rakotoson et al., 1983; Ferry et al., 2001; Zhuo et al., 2006; Griffiths et al., 2010; Sahoo et al., 2010; Hosoda et al., 2011; Khan and Puranik, 2011; Tan et al., 2012). Several researchers have described different methods for estimation of the radon emanation rate in mines (Khan and Raghavayya, 1973; Archibald and Nantel, 1979; Nantel and Archibald, 1981; Panigrahi et al., 2005; Bochiolo et al., 2012). A technique by determining the increase of radon concentration in air between two points in ventilation passage in Japanese and Canadian uranium mines has previously been studied (Fusamura and Misawa, 1963; Thompkins and Rajhans, 1967; Keshvani, 1970). Thompkins and Cheng (1969) have described a method in which a steel chamber provided with several valves was cemented on the walls of mine and radon samples were drawn from the chamber at intervals of several hours up to 50 h for computation of the emanation rate. A similar technique was used by Archibald and Nantel (1984) for the radon emanation measurements in Canadian uranium mines. However, the aforementioned techniques have drawbacks. The former may give high uncertainties in the results due to various mining operation conditions, contamination of intake air and air leakage, whereas, the later technique is very complex, expensive and time consuming for making the arrangement of experimental setup. Dwaikat et al. (2010) investigated the specific radon exhalation from the mine rocks based on the radon measurements by means of CR-39 detectors, in which the uncertainty of the measurements depends on several factors, such as exposure period, etching process and calibration. Keeping these in view, comparatively a simple, quick and less expensive technique giving low uncertainties in the results was used in the present study to overcome the aforementioned problems.

In the present study, we investigate the radon emanation rate from uranium ore samples in the laboratory and from *in situ* measurements in a uranium mine to obtain relationships between radon emanation and physical properties of the ore body that may be generalized to other low-grade uranium mines. Based on field and laboratory data, this study also aims at developing empirical relationships for quick prediction of the radon emanation rate from uranium ore body of similar nature in any low-grade uranium mine.

2. Materials and methods

2.1. Ore samples

We investigated the radon emanation from laboratory and *in situ* measurements on the ore body of Jaduguda uranium mine located in the Singhbhum shear zone in the eastern part of India. The location of Singhbhum shear zone in India and the transverse section showing the ore body and different lithological units around Jaduguda are shown in Fig. 1. Jaduguda mine has two parallel mineable lodes starting from the surface and lying up to a depth of about 905 m. The lodes dipping towards north with an average inclination of about 40° are separated from each other by a distance of about 80 m. The footwall and hangwall rocks of both the lodes are quite competent from geotechnical point of view. Uranium-bearing minerals in Jaduguda mine occur in the Precambrian meta-sedimentary rocks, which are highly folded and sheared. The principal lithological rock units are autoclastic conglomerate (brecciated quartzite), quartz–chlorite–biotite–magnetite schist, biotite–chlorite schist and epidiorite, of which first two rock units host the mineralisation. The primary uranium minerals of Jaduguda ore are uraninite and pitchblende and most common secondary mineral is autunite. The uranium minerals are associated with a wide variety of sulphides of copper, nickel, cobalt, molybdenum, arsenic and bismuth. Some prominent ore minerals are magnetite, ilmenite, uraninite, rutile, chalcopyrite, pyrhotite, marcasite, mackinawite, violarite, tellurobismuthite, tetradymite, cubanite and molybdenite (Sarangi and Singh, 2006).

Horizontal cut-and-fill using de-slimed mill tailing as backfill is the principal stoping method adopted in Jaduguda mine. Twenty-one ore samples were collected from different stopes of the mine. The samples were oven dried to determine their physical properties and the activity concentration. The *in situ* radon emanation rate was determined from drill holes made within the ore body. Samples of the drill cuttings were collected from these holes to determine the grade of ore in the laboratory.

2.2. Theoretical model for measuring radon emanation rate

The emanation of radon into an enclosed chamber, initially free from radon, may be assumed as a steady-state process. The radon concentration in the chamber will follow an exponential growth up to a certain build-up period. Thereafter, it reaches a constant value as a balance of the increase due to emanation and decrease due to radioactive decay (Thompkins and Cheng, 1969; Khan and Raghavayya, 1973; Girault and Perrier, 2012). The radon emanation rate can be determined from this build-up pattern. The activity concentration of radon is estimated by collecting the air sample in scintillation cells and using the following equation (Raghavayya, 1981)

$$C = \frac{6.967 \times 10^{-5} c}{E V_s e^{-\lambda\tau}\left(1 - e^{-\lambda T}\right)} \quad (1)$$

where c is the total counts during the counting duration "T", E is the efficiency of the system (%), V_s is the volume of scintillation cell (m^3), τ is the delay time after end of the sampling (s) and T is the counting duration (s). Since the volume of scintillation cell is small