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Radon emanation from backfilled mill tailings in underground uranium mine

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ABSTRACT

Coarser mill tailings used as backfill to stabilize the stoped out areas in underground uranium mines is a potential source of radon contamination. This paper presents the quantitative assessment of radon emanation from the backfilled tailings in Jaduguda mine, India using a cylindrical accumulator. Some of the important parameters such as ^{226}Ra activity concentration, bulk density, bulk porosity, moisture content and radon emanation factor of the tailings affecting radon emanation were determined in the laboratory. The study revealed that the radon emanation rate of the tailings varied in the range of 0.12–7.03 Bq m^{-2} s^{-1} with geometric mean of 1.01 Bq m^{-2} s^{-1} and geometric standard deviation of 3.39. An increase in radon emanation rate was noticed up to a moisture saturation of 0.09 in the tailings, after which the emanation rate gradually started declining with saturation due to low diffusion coefficient of radon in the saturated tailings. Radon emanation factor of the tailings varied in the range of 0.08–0.23 with the mean value of 0.21. The emanation factor of the tailings with moisture saturation level over 0.09 was found to be about three times higher than that of the absolutely dry tailings. The empirical relationship obtained between ^{222}Rn emanation rate and ^{226}Ra activity concentration of the tailings indicated a significant positive linear correlation ($r = 0.95$, $p < 0.001$). This relationship may be useful for quick prediction of radon emanation rate from the backfill material of similar nature.

1. Introduction

The uranium content of Indian ores is low. The existing uranium processing technology in India is based on leaching of uraninite ore in sulphuric acid medium in presence of pyrolusite oxidant followed by filtration, ion exchange separation and product recovery in the form of magnesium di-uranate (MgU_2O_7). As a result of the low uranium content, large quantities of solid and liquid wastes are produced during the production of uranium concentrate, which are mixed together, neutralized at elevated pH (>9.5) for precipitation of radionuclides and heavy metals. The slurried tailings is separated into coarse and fine fractions, the former is sent back to underground uranium mines for backfilling the stoped out areas and later is discharged into a geologically stable structure with proper embankment and barriers called tailings pond (Tripathi et al., 2008). Depending on the ore processing and waste immobilization technology, these tailings may contain elevated amounts of ^{226}Ra.

In mine atmosphere, the concentration of radon gas (^{222}Rn, $t_{1/2} = 3.82$ d) which is the immediate decay product of ^{226}Ra, largely depends on activity concentration of ^{226}Ra and ^{222}Rn emanation rate from the tailings used as backfill material (Raghavayya and Khan, 1973). The main source of internal dose for miners and members of public is due to exposure of ^{222}Rn and its short-lived decay products. Prolonged exposure of short-lived radon progeny at elevated level is carcinogenic and may lead to lung carcinoma (Field et al., 2000; Gulson et al., 2005; Al-Zoughool and Krewski, 2009; ICRP, 1993, 1989; UNSCEAR, 2006). In view of the nature of hazard associated with radon progeny, monitoring of activity concentration of ^{222}Rn in uranium mines is required to minimize the internal exposure of miners within the limits stipulated by regulatory agencies (ICRP, 1993; IAEA, 1996).

The ^{222}Rn gas emanation from the tailings into mine opening is caused by its diffusion and transport through the matrices. In diffusion, the ^{222}Rn gas moves through the fluid (air and water) filling the pore spaces of the associated matrices, whereas in transport, the fluid carries the radon gas through these pores. ^{222}Rn

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emanation is primarily governed by the factors such as porosity (Tanner, 1964, 1978; Evans, 1969; Lawrence et al., 2009), distribution of ^{226}Ra and grain size (Garver and Baskaran, 2004; Sakoda et al., 2011; Somlai et al., 2008) and moisture content of the matrix (Bossew, 2003; Adler and Perrier, 2009; Breitner et al., 2010). Apart from this, temperature (Iskandar et al., 2004; Girault and Perrier, 2011) and pressure (Schroeder, 1966; Pohl-Rueling and Pohl, 1969; IAEA, 1981) also have important bearing in deciding the fate of radon emanation. Despite low radium content, emanation potential of radon from the tailings may be high as compared to the bulk ore from which it is originated due to the increased porosity and surface area (Khan and Raghavayya, 1973; Thompkins, 1982). The presence of low moisture content in pore spaces of the tailings enhances radon emanation (Strong and Levins, 1982). Washington and Rose (1990) reported that temperature changes in the porous materials have less effect on radon emanation in dry materials than in moist materials. Clements and Wilkening (1974) reported that the change of pressure in pore spaces of the materials from 1 to 2% changes the radon emanation rate from 20 to 60%. 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. The radon atoms leaving the tailing matrices, within which they are formed, may directly enter into the interstitial flowing water and transported along with it.

Earlier, in Elliot Lake mine (Wheeler, 1982; Archibald and Nantel, 1984) and Saskatchewan mine (Cheng, 1981) of Canada, the radon emanation rates from the tailings materials were estimated based on diffusion theory which does not consider some important mechanisms such as adsorption and advection. Under very dry condition, radon diffusion from the tailings decreases due to the effect of re-adsorption of the recoiled radon atoms on the surfaces of the pores (IAEA, 1981). On the other hand, when these pores are saturated with water, the recoiling ions escaping into the pores encounter a dense absorber and have a greater probability of remaining in the pores (Meslin et al., 2010; Hassan et al., 2011). However, when the interstices of the tailings are filled with water, radon is transported along with flowing water. The concentration of radon in water may approach a much higher level than the pores filled with air (Andrews and Wood, 1972). Due to the aforementioned reasons, there may be uncertainty in the estimated radon emanation rate. Raghavayya and Khan (1973) used a 25 L capacity steel drum from which air samples were drawn after a time lapse varying from 7 to 65 h for measurement of radon emanation rate from backfill material in the mine. However, this technique has also some limitations like occurrence of back diffusion and leakage of air due to longer experiment period giving rise to uncertainty in the results and inconvenience of carrying the large size drum for radon emanation studies at different locations inside the mine.

In the present study, we measured the radon emanation rate from *in situ* backfilled tailings in Jaduguda mine based on accumulation technique using an accumulator of approximately 7 L capacity. The experiments were carried out within a smaller accumulation period of 1 h to minimize the errors due to alterations in the parameters affecting radon emanation, back diffusion of radon and air leakage (Jha et al., 2000; Mayya, 2004). Radon emanation factor of the tailings was determined using both *in situ* measurement of radon emanation rate and empirical function developed by Rogers and Nielson (1991) for predicting the effective diffusion coefficient. This study intended to investigate the effects of tailings parameters such as moisture content, bulk density, bulk porosity and ^{226}Ra activity concentration on emanation potential of radon. It also aimed at developing an empirical relationship using the primary data of backfilled tailings for quick prediction of radon emanation rate from backfill material of similar nature.

2. Materials and methods

2.1. Study area

This study was carried out at Jaduguda uranium mine (Latitude 22° 39′ and Longitude 86° 22′) located in the Singhbhum Thrust Belt of Jharkhand, India. The geological map of Singhbhum Thrust Belt is shown in Fig. 1 (Sarangi, 2003). Uranium bearing minerals in Jaduguda mine occur in the Precambrian meta-sedimentary rocks. The host rocks are autoclastic conglomerate (brecciated quartzite) and quartz-chlorite-biotite-magnetite schist. The uranium minerals are associated with a wide variety of sulphides of copper, nickel, cobalt, molybdenum, arsenic and bismuth (Krishnamurthy et al., 2004).

The mine has two parallel lodes starting from the surface and extending to a depth of 905 m. The strike of the main ore body dipping towards north is in the east–west direction. Horizontal cut-and-fill method using de-slimed coarser fraction mill tailings as backfill is the principal stoping method adopted in Jaduguda mine. The tailings are pumped into the mine in the form of aqueous slurry for backfilling, which settle and consolidate by its own weight to stabilise the excavations after draining out of water.

2.2. Methodology

The radon emanation rate from *in situ* backfilled tailings was measured at 16 locations of the backfilled stopes using a cylindrical accumulator. Thereafter, the settled core samples of backfill tailings were collected from the locations of radon emanation study by inserting a stainless steel cylinder of 36 mm diameter and 5 cm length. The samples were kept in plastic vials marked with proper identification of the location and date of collection. The desired physical and radioactive properties of the tailings were determined in the laboratory.

2.2.1. Measurement of physical properties of tailings

The moisture content (dry weight basis) of backfilled tailings was determined by heating the samples at 105 ± 5 °C for 24 h as per standard ASTM D2216. An undisturbed core of backfill material was collected inserting a stainless steel cylinder for determination of bulk density. The mass of sample oven dried at the aforementioned temperature for 24 h was noted. The bulk density was determined by dividing the mass by the cylinder volume. The specific gravity of the tailings was determined using water pycnometer as per standard IS: 2386 (Part III). The bulk porosity (φ) of the tailings was estimated using Eq. (1)

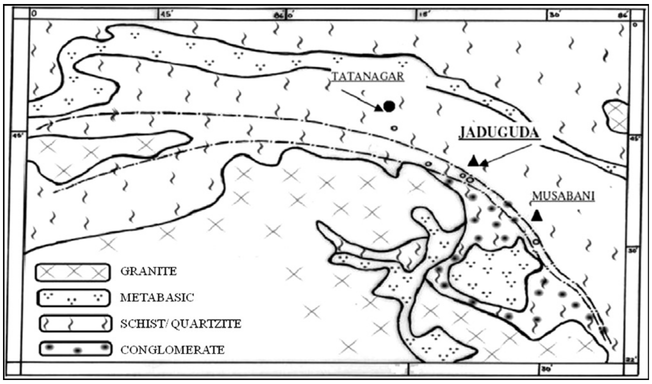

Fig. 1. Geological map of Singhbhum Thrust Belt (Sarangi, 2003).