PATITAPABAN SAHU
IMRAN ATHAR BEG
and
DURGA CHARAN PANIGRAHI

Mathematical modelling of radon (^{222}Rn) exposure of underground mine workers: a comprehensive review

The primary purpose of this study is to establish the methods for determination of total growth of ^{222}Rn daughters and ^{222}Rn exposure of the workers at desired places of mines. This paper presents a comprehensive review of the types of radiation hazards associated with underground uranium mining. The mathematical modelling of the growth of ^{222}Rn daughters for a horizontal tunnel is also derived in this paper. Further, an example of the determination of inhalation exposure of miners in a cut-and-fill stope mine is provided in this paper. A critical analysis of the literature review revealed that the radiological hazards associated with low-grade uranium ore are primarily owing to inhalation of ^{222}Rn and its daughters. The total growth of ^{222}Rn daughters are found to be 0.468 $\mu J/m^3$ that are calculated based on the reported values of ^{222}Rn release rate from mine walls, blasting materials, fill materials, and the percolating water. Further, it also revealed that the radon contamination of intake air was the significant contributory factor in the radiation exposure of uranium mine workers.

Keywords: *Depletion, modelling, radon (^{222}Rn) daughters, contamination, radiation, hazards, porousness*

1.0 Introduction

Rapid depletion of the conventional sources of energy and growing demands has resulted in a renewed search for alternative resources, and uranium has been recognized as a potential source of abundant energy. In order to supply natural uranium for the nuclear power programmes, mining of uranium ore is given more importance which contains higher abundance of ^{38}U. Radon (^{222}Rn), which is a daughter product of radium in ^{238}U series, is an inert gas with a half-life of about 3.82 days. Its concentration may be significant in the confined places such as underground mines (Mudd, 2008). ^{222}Rn gas is continuously exhaled into the mine air through the parent ore. The diffusion of ^{222}Rn from the materials into mine air is mainly owing to the pressure, radium content and porousness of the materials. The significant sources of radon are mine walls, blasting materials, filling materials, and percolating water in the mine (Sahu et al., 2013 and 2016).

More than 50% of the total radiation exposure of uranium mine workers is owing to the inhalation and ingestion of ^{222}Rn gas and its daughters, respectively (UNSCEAR, 2000). From epidemiological studies, it has been found that exposure of uranium mine workers to ^{222}Rn and its daughters for longer periods may give rise to lung cancer due to the ionizing alpha radiation associated with their decay (ICRP, 2012; WHO, 2009). Keeping these in view, the inhalation of the radon is mainly responsible for causing lung cancer. Different regulatory bodies have stipulated several guiding principles to reduce the radon exposure of the underground mine workers (IAEA, 2004; UNSCEAR, 2008). International Commission on Radiological Protection (ICRP) has suggested the system of dose limitation against the health effects of radiation exposure with the basic aim to have the exposure as low as reasonably achievable (ALARA). To achieve the ALARA principle, all mining methods and operations must be optimised and the main focus should be given on controlling the exposures by engineering means such as providing adequate ventilation, adopting safe equipment layout and work procedures.

This paper aimed at understanding the growth of radon daughters and to establish the method for determination of inhalation exposure of the underground mine workers. A comprehensive literature review of the types of radiation hazards and radioactive aerosol particles in the underground mine environment is presented in this paper. It also comprehensively described the derivation of mathematical modelling for the determination of growth of radon daughters in the underground mine environment.

2.0 Types of radiation hazards

Since the percentage of ^{238}U in natural uranium is 99.2739%, its contribution to radiological health hazards in underground uranium mines may be significant. The health hazards can be categorized into external and internal hazards.

Dr. Patitpaban Sahu, Assistant Professor, Mr. Imran Athar Beg, PhD Research Scholar and Prof. D.C. Panigrahi, Professor, Department of Mining Engineering, Indian Institute of Technology/ISM), Dhanbad. Corresponding Author: Dr. Patitapaban Sahu, Email: patitapaban@iitism.ac.in

2.1. EXTERNAL HAZARD

The external hazard is owing to the exposure of uranium mine workers to β and γ radiations produced from the uranium-bearing minerals. Since uranium (^{238}U) has half-life period of 45×10^8 years, equilibrium ratio between ^{238}U and its daughters may occur in the mine rocks. Raghavayya (2005) established the following relationship between the external exposure level 'D' (μGy/h) and the percentage of U_3O_8 in the mine rocks:

$$D = 50 \times G \qquad \text{... (1)}$$

From Eq. (1), it may be concluded that external exposures of uranium mine workers are not important if grade of uranium ore is < 0.2% U_3O_8.

2.2. INTERNAL HAZARDS

The internal hazard is owing to inhalation of long-lived α – emitters, ^{222}Rn and its daughters.

2.2.1. Long-lived α – emitters

Ore dust generated during the different mining activities contain the long-lived α-emitters (LLα - emitters). Inhalation of long-lived decay products is a source of internal radiation dose to the workers. IAEA (1989) reported that in low ore grade uranium mines (<1%U_3O_8), the contribution of long-lived daughter products to internal exposure is less significant. The epidemiological studies also revealed that the possible contribution from the inhalation of LLα - emitters to lung cancer is very uncertain (ICRP, 1993).

2.2.2. Radon and its daughters

The most significant source of radiation exposure of mine workers is ^{222}Rn and its daughters such as ^{218}Po (RaA), ^{214}Pb (RaB), ^{214}Bi (RaC) and ^{214}Po (RaC′). Air-borne short-lived ^{222}Rn daughters can be categorized into unattached and attached radon daughters. The unattached ^{222}Rn daughters form the clusters with a particle size of <10 nm for a smaller period of $10^{-2} - 10^{-1}$ s with moisture and oxygen in the mine air (Porstendoerfer, 2001). These radon daughters can rapidly attach to the prevailing aerosol particles in the mine air due to their high diffusion rate. The size of attached ^{222}Rn daughters in the aerosols varies from 10nm to 1000nm (Dankelmann et al., 2001).

The characteristics of radon progeny aerosols in atmospheres shown in Table 1 are very important for the evaluation of the radiation exposure of the workers. From the table, it may be observed that the equilibrium ratio between ^{222}Rn and its daughters increases with decreasing the size of aerosols in terms of Activity Median Aerodynamic Diameter (AMAD). The AMAD for radon progeny varies with ambient conditions, but it is always less than 1μm. Liu et al. (2014) reported that the AMAD of aerosols in Schwartzwalder uranium mine of USA varies in the range of 34.2 – 72.7nm with an average value of approximately 55.3nm.

TABLE 1: CHARACTERISTICS OF RADON PROGENY AEROSOLS (UNSCEAR1988)

Atmosphere	Concentration (cm^{-3})	AMAD (nm)	Equilibrium factor
Mines	10^4 - 10^5	90 - 300	0.3
Indoor	10^3 - 10^5	5 - 150	0.4
Outdoor	10^3 - 10^5	30 - 500	0.7 - 0.8

3.0 Exposure of uranium mine workers to short-lived decay products of ^{222}Rn

The US Public Health Service (US PHS) introduced a unit called Working Level 'WL' for routine evaluation of radiation exposure of uranium mine workers. WL is the concentration produced by the combination of ^{222}Rn daughters such that the total energy potential of the daughters released on ultimate decay to $^{210}Pb$is 2.08×10^{-5} J/m^3. This particular value corresponds to Potential Alpha Energy (PAE) associated with the ^{222}Rn daughters in equilibrium with 3700 Bq m^{-3} of radon (IAEA, 1996). ICRP (1993) defines the WL as 1.3×10^8 MeV/m^3 that would correspond to 2.08×10^{-5} J/m^3.

The dose to the workers can be estimated by multiplying the radiation exposure with a conversion factor. The dose conversion factor 'D_{cf}' mainly depends on the unattached and attached progeny clusters in underground uranium mines (Cavallo, 2000). Porstendorfe *r*(1996) used the following equations for the calculation of the dose conversion factor in different breathing modes. The dose conversion factor 'D_{cfm}' (mSv/WLM) in case of mouth breathing can be computed using the following relation

$$D_{cfm} = 101f_{un} + 6.7(1\text{-}f_{un}) \qquad \text{... (3)}$$

In case of nasal breathing 'D_{cfn}' (mSv WLM^{-1}), it is given by

$$D_{cfn} = 23f_{un} + 6.2(1\text{-}f_{un}) \qquad \text{... (4)}$$

Where f_{un} is the unattached fraction of ^{222}Rn progeny and is expressed as (Knutson 1988)

$$f_{un} = (PAEC)_{un} / [(PAEC)_{un} + (PAEC)_{att}] \qquad \text{... (5)}$$

where $(PAEC)_{un}$ and $(PAEC)_{att}$ are the potential alpha energy concentration of the unattached and attached ^{222}Rn daughters (J/m^3). The unattached fractions of radon progeny reported for underground uranium mines located in different countries are presented in Table 2. The dose conversion coefficient for the inhalation of ^{222}Rn daughters is directly proportional to the unattached fraction. UNSCEAR (2000) reported that the dose conversion coefficient became doubled for an increase in the unattached fraction from 0.05 to 0.21 for the median aerosol diameter of above 200nm.

Bennett et al. (2003) reported the following equation for the calculation of combined dose conversion factor 'D_{cfc}' (mSv/WLM) for miners during heavy physical work in which mouth and nasal breathings were assumed to be 60% and 40%, respectively.

TABLE 2: UNATTACHED FRACTIONS OF RADON PROGENY IN DIFFERENT UNDERGROUND MINES

Mine	Min – Max (Median)	Reference
Germany uranium mine	0.001 – 0.025 (0.007)	Butterweck et al. (1992)
Canada uranium mines	0.01 – 0.06	Cavallo et al. (1999)
Jaduguda Uranium Mine, India	0.02 – 0.16 (0.06)	Khan (1979)
American uranium mines	0.04 – 0.33 (0.05)	Raghavayya and Jones (1974)
French uranium mines	< 0.01	Boulaud and Chouard (1992)

$$D_{cfc} = 0.6\, D_{cfm} + 0.4\, D_{cfn} \quad \text{... (6)}$$

Thus, the effective dose 'E_f' (mSv year^{-1}) can be computed using the relation

$$E_f = (WLM)\, D_{cfc} \quad \text{... (7)}$$

As discussed above, the contribution of uranium mine workers to ^{222}Rn daughters is significant. However, this research work only concerns on the diffusion of ^{222}Rn into the mine atmosphere. Because radon daughters are solid and hence, are not able to diffuse from the materials. The concentration of ^{222}Rn daughters is directly proportional to the concentration of ^{222}Rn in the atmosphere. Thus, the ^{222}Rn concentration in air is a sign of the inhalation exposure of the miners.

4.0 Modelling of growth of working level

The understanding of the growth of ^{222}Rn daughters in the underground tunnel atmosphere is the essential factor for a uranium mine ventilation engineer. The "age of air" concept becomes apparent based on the radon decay chain. At radioactive equilibrium, the number of atoms of each daughter product present in the decay chain is directly proportional to the half period of that daughter product. Generally, longer is the half-life period, greater will be the number of atoms and greater is the contribution to the working level '*WL*'. The growth of radon daughter concentrations as the radon-laden air moves through an underground tunnel as shown in Fig.1 is caused by different radioactive components: (i) initial concentrations of ^{222}Rn 'C_0' and its short-lived decay products 'WL_0' at inlet of the tunnel, and (ii) diffusion of ^{222}Rn from materials.

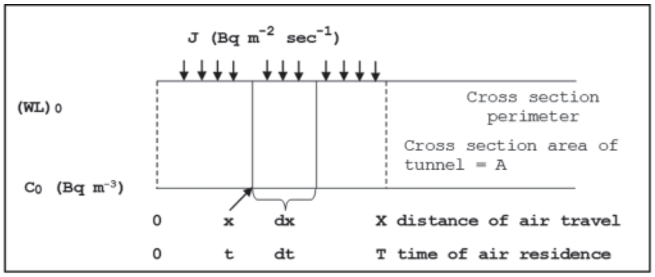

Fig.1 Plan view of a tunnel

To calculate the growth of working levels '*WLs*' in initially present pure ^{222}Rn, the concentrations of radon daughters have to be computed for different ages of the mine atmosphere. It involves the determination of the number of atoms of each daughter product per unit volume, along with their corresponding activities. Such calculations may give rise to the characteristic working level growth curve as shown in Fig.2. It showed that the growth of RaC (^{214}Bi) and RaC' (^{214}Po) did not become significant until 20 minutes of aging and about 80% equilibrium would be achieved in 90 min.

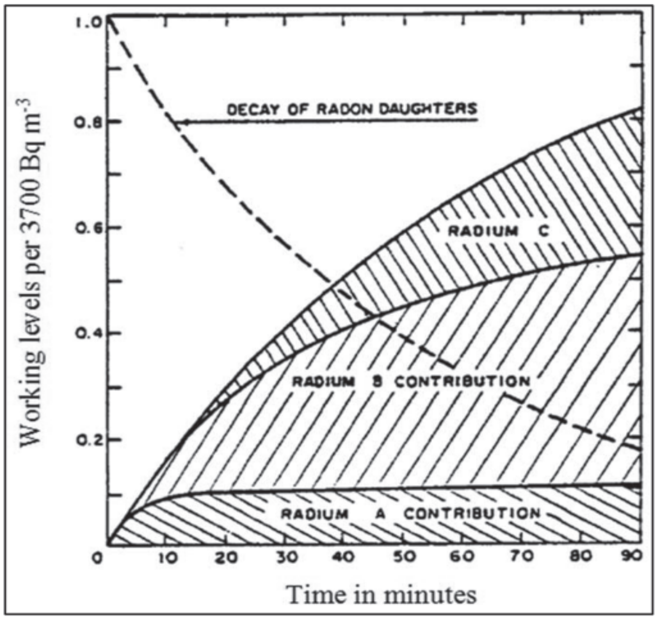

Fig.2 Growth of *WL* in mine atmosphere (Holaday, 1974)

The *WL* growth between 1 and 40 min can be approximately determined using Eq. (8)

$$WL = 62.16 \times 10^{-7} \times C_0 \times t^{0.85} \quad \text{... (8)}$$

where 62.16×10^{-7} WL m^3/Bq/min$^{0.85}$ is the constant derived from Fig.2, C_0 is initial pure concentration of radon (Bq/m^3) and t is time of air residence at a distance '*x*' from the starting point (min).

The initial radon daughter concentration 'WL_0' decreases at a rate related to the mixture of nuclides present. At the starting point, the equilibrium ratio 'ER' between ^{222}Rn and its daughters is expressed as

$$ER = {}^{(3700 \times WL_0)}/_{C_0} \quad \text{... (9)}$$

If '*G*' represents the radioactive age of the air in minutes at a given starting point and may be substituted for '*t*'. By combining Eqs. (8) and (9), it becomes

$$G = (43.48\, ER)^{1.1765} \quad \text{... (10)}$$

The fractional decrease 'WL_f' in the initial radon daughter concentration 'WL_0' after air residence time '*T*' is given by:

$$WL_f = {}^{(WL_{G+T} - WL_T)} / {}_{WL_G} \quad \ldots (11)$$

where WL_T is growth of WL in the air at time 'T'.

or

$$WLf = {}^{[(G+T)^{0.85} - \mathrm{T}^{0.85}]} / {}_{G^{0.85}} \quad \ldots (12)$$

Thus, the first growth of WL component 'WL_1'due to the existing ^{222}Rn daughter concentration is expressed as:

$$WL_1 = WL_f \times WL_0 \quad \ldots (13)$$

The second growth of WL component 'WL_2'owing to the initially pure ^{222}Rn is given by:

$$WL_2 = 62.16 \times 10^{-7} \times C_0 \times T^{0.85} \quad \ldots (14)$$

The third component 'WL_3' due to diffusion of radon into the mine air can be determined with reference to Fig.2. The ^{222}Rn exhalation rate 'J' is assumed to be uniform over the rock surfaces. Over an interval of distance 'dx', radon diffuses from a tunnel surface '$O \times dx$' where 'O' represents the surface perimeter of the tunnel. Since this flow continues for a time interval 'dt', the added ^{222}Rn activity is given by:

$$J \times (O \times dx) \times dt \quad \ldots (15)$$

This additional radon concentration dilutes into a volume 'A×dx', where A is the tunnel cross-sectional area. Increase in radon concentration over the distance 'dx' is expressed as

$$dc_{Rn} = {}^{(J \times (O \times dx) \times dt)} / {}_{A \times dx} \quad \ldots (16)$$

The ultimate radon daughter concentration 'WL_3' due to diffusion of^{222}Rn can be expressed as:

$$WL_3 = {}^{(J \times 0 \times 62.16 \times 10^{-7} \times T^{1.85})} / {}_{(1.85 \times A)} \quad \ldots (17)$$

where J is ^{222}Rn exhalation rate (Bq/m^2/s) and A is cross-sectional area of the tunnel (m^2).

If there are number of sources of ^{222}Rn present in the tunnel, the total ^{222}Rn production rate 'E_s' (Bq/s) can be computed using the following relation

$$Es = \Sigma^{n}_{i-1} J_i a_i \quad \ldots (18)$$

where J_i is ^{222}Rn exhalation rate of sources 'i' (Bq/m^2/s) and a_i is surface area of source 'i' (m^2).

If the distance of air travel inside the tunnel is 'X' then the ^{222}Rn exhalation rate can be expressed as

$$\mathrm{J} = {}^{\mathrm{E_s}} / {}_{(0 \times X)} \quad \ldots (19)$$

$$WL_3 = {}^{(E_s \times 62.16 \times 10^{-7} \times T^{1.85})} / {}_{(1.85 \times A \times X)} \quad \ldots (20)$$

or

$$WL_3 = {}^{(E_s \times 62.16 \times 10^{-7} \times T^{1.85})} / {}_{(1.85 \times V)} \quad \ldots (21)$$

Total radon daughter concentration 'WL_F' at a particular end point is given by:

$$WL_F = WL_1 + WL_2 + WL_3 \quad \ldots (22)$$

Thus total potential alpha energy concentration '$(PAEC)_F$' (J/m^3) at that point can be calculated by multiplying the total growth of WL with the factor of 2.08×10^{-5}. Thus, it can be expressed as

$$(PAEC)_F = WL_F \times 2.08 \times 10^{-5} \quad \ldots (23)$$

5.0 An example for the determination of total growth of radon daughters in an Indian underground uranium mine

The experimental set up for determination of the concentrations of radon and its daughters with scintillation cell and alpha guard in a cut-and-fill stope of an Indian underground uranium mine is presented in Fig.3. The dimensions of stope are 130m length, 5m width, and 4.5m height. The time required to pass the air from inlet to outlet of the stope is determined to be 2 min. Concentrations of radon at the inlet and outlet airways of the stope are found to be 182 Bq/m^3 and 255 Bq/m^3, respectively. The concentration of radon daughter was found to be 4.16×10^{-7} J/m^3 (0.02WL). The release rates of ^{222}Rn from mine wall, broken materials, fill materials and mine water in the stope were reported to be 1.01×10^{3}Bq/min, 8.46×10^{3}Bq/min, 138.45×10^{3}Bq/min and 149.74×10^{3}Bq/min, respectively (Panigrahi et al., 2015). Equilibrium ratio between the ^{222}Rn and its daughters in the underground uranium mine was assumed to be 0.4 (ICRP, 1993). Thus, the radioactive age of the air was determined to be 29 min. The fractional decrease in the initial radon daughter concentration after an air travel time of 2 min was worked to be 0.96.

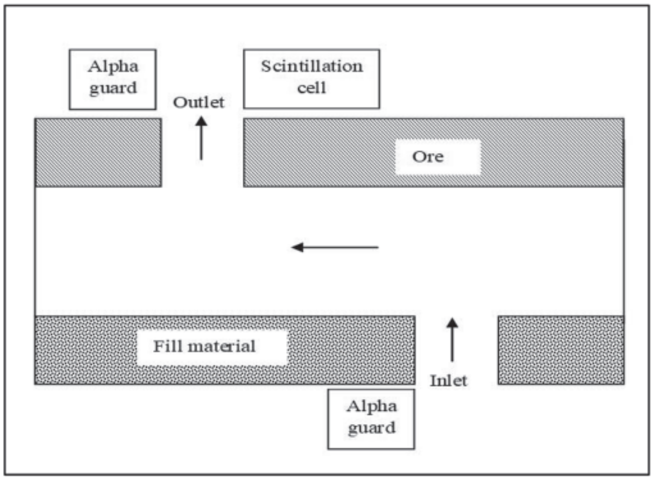

Fig.3 The layout of a cut-and-fill stope.

The first, second, third and total growths of radon daughters [Eqs. (13), (15), (28) and (29)] are found to be 0.4 µJ/m^3, 0.0424 µJ/m^3, 0.0256 µJ/m^3, and 0.468 µJ/m^3, respectively. From the results, it is observed that the initial concentration of ^{222}Rn at the inlet airway of the stope are the significant contributory factors in the total PAEC. Further, radon contaminated intake air may result in more ventilation requirement to accomplish the desired radiation level in the underground mine environment (Panigrahi et al., 2015). In view of the above, it is recommended to supply the radon-free air into the underground mines. The total PAEC is worked out to be 8% of the derived limit value of 6.2 µJ/m^3, which is equivalent to 0.30 WL stipulated by ICRP (1993). It might be owing to improved ventilation conditions in the mine. Panigrahi et al. (2005) observed that the radiation exposure in a stope decreased by 30% when the airflow rate in an Indian underground uranium mine increased from 21m^3/s to 100m^3/s. Skubacz et al. (2019) reported that a 22% decrease in airflow quantity increased radiation exposure by 60%. In view of the above, ventilation is one of the most important parameres in reducing the radaion exposure of the underground mine workers.

6.0 Summary and conclusions

A critical review of the literature reveals that the radiation exposure of uranium mine workers during the mining of low grade of uranium ore is primarily owing to contributions of ^{222}Rn and its daughter products. The modelling of the growth of radon daughters is derived based on the initial concentrations of ^{222}Rn and its daughter products at the inlet of the horizontal tunnel and radon production rate from the different sources. This model is helpful in the evaluation of ^{222}Rn exposure of the uranium mine workers. An example is also explained for determination of growth of radon daughters in an Indian underground uranium mine based on the reported values of radon production rate from mine walls, blasting materials, fill materials and percolating water. The first, second, third and total growths of radon daughters are worked out to be 0.4 µJ/m^3, 0.0424 µJ/m^3, 0.0256 µJ/m^3, and 0.468 µJ/m^3, respectively. It is observed that the significant contribution to growth of radon daughters is primarily due to the initial concentration of ^{222}Rn in the intake airway of the stope. When the inlet air is contaminated with ^{222}Rn, more air quantity is required to accomplish the radiation exposure as low as reasonably achievable. The total PAEC is worked out to be 8% of the derived limit value of 6.2 µJ/m^3 stipulated by ICRP. Therefore, radiological health problems of the persons working in underground uranium mines are observed to be negligible. It might be owing to low-grade uranium ore, negligible porosity of mine rocks and good ventilation condition of the mine. However, the total PAEC may exceed the derived limit stipulated by regulatory agency if suitable control measures are not implemented during mining operation. Since ventilation plays a vital role in reducing the ambient concentrations of ^{222}Rn and its daughters in underground uranium mines, effective management of ventilation and operating practices should be implemented periodically to reduce the ^{222}Rn exposure of underground mine workers.

Acknowledgements

The support of the Science and Engineering Research Board (SERB), Government of India (Grant No. ECR/2016/000723, 2016) is gratefully acknowledged.

References

1. Bennett, W.D., Zeman, K.L., Jarabek, A.M. (2003): Nasal contribution to breathing with exercise: effect of race and gender. *J. Appl. Physiol*. 95, 497-503. https://doi.org/10.1152/japplphysiol.00718.2002
2. Boulaud, D., Chouard, J.C. (1992): Submicron sized aerosol and radon progeny measurements in a uranium mine. *Radiat. Prot.Dosim*. 45, 91-94. https://doi.org/10.1093/rpd/45.1-4.91
3. Butterweck, G., Porstendorfer, J., Reineking, A., Kesten, J.(1992): Unattached fraction and the aerosol size distribution of the radon progeny in a natural cave and mine atmospheres. *RadiatProtDosim* 45, 167-170. https://doi.org/10.1093/rpd/45.1-4.167
4. Cavallo, A., Hutter, A., Shebell, P., (1999): Radon progeny unattached fraction in an atmosphere far from radioactive equilibrium. *Health Phys.* 76, 532-536. https://doi.org/10.1097/00004032-199905000-00010
5. Cavallo, A.J.,(2000): Understanding mine aerosols for radon risk assessment. *J. Environ.Radioact.* 51, 99-119. https://doi.org/10.1016/S0265-931X(00)00047-3
6. Dankelmann, V., Reineking, A., Porstendorfer, J., (2001): Determination of neutralization rates of ^{218}Po ions in air. Radiat. *Prot. Dosim.* 94, 353 - 357. https://doi.org/10.1093/oxfordjournals.rpd.a006510
7. Holaday, D.A., (1974): Evaluation and control of radon hazards in uranium mines. National Institute Occupational Health and Safety, NIOSH 75-117, Rockville, Md., USA, pp.64.
8. IAEA (1989): Radiation monitoring in the mining and milling of radioactive ores. International Atomic Energy Agency, IAEA Safety Series No.95, Vienna.
9. IAEA, (1996): International basic safety standards for protection against ionising radiation and for the safety of radiation sources. *International Atomic Energy Agency,* Vienna.
10. IAEA (2004): Occupational radiation protection in the mining and processing of raw materials. Safety Standards Series No. RS-G-1.6, STI/PUB/1183, *International Atomic Energy Agency,* Vienna.
11. ICRP (1993): Protection against radon at home and at work. International Commission on Radiological Protection, ICRP Publication No. 65, Vol. 23 (2).
12. ICRP (2012): Compendium of Dose Coefficients based on ICRP Publication 60. ICRP Publication 119, Ann. ICRP 41(Suppl.). https://doi.org/10.1016/j.icrp. 2012.06.038
13. Khan, A.H. (1979): A study on the factors affecting the build-up of radon-222 and its progeny in uranium mines. M.Sc. Thesis, University of Bombay, Mumbai, India.
14. Knutson, E.O.(1988): Modelling indoor concentrations of radon's decay products. In: Nazaroff, W.W., Nero Jr., A.V. (Eds.), Radon and Its Decay Products in Indoor Air. Wiley, New York, pp. 161-199.
15. Liu, X., Doerges, J.E., Volckens, J., Johnson, T.E. (2014): Aerosol size distribution in the Schwartzwalder uranium mine. *Health Phys.* 106, S20-S24. https://doi.org/10.1097/HP.0000000000000000

(Continued on page 371)