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## Measurement of natural radioactivity, radon exhalation rate and radiation hazard assessment in Indian cement samples

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### Abstract

Building materials are assumed to be the second source of Radon inside buildings. Due to low level of radon emanation from these materials, long term measurements are needed. Radiation doses from the building materials vary depending upon the natural radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and their daughter products and  $^{40}\text{K}$  present in them. Cement is the main and important component used in the construction of buildings in many countries. These radio nuclides pose exposure risk due to their gamma ray emission and internally due to radon and its progeny that emit alpha particles. In the present study radon exhalation rate and the activity concentration of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  radionuclides in cement samples used in Aligarh region (U.P.), India have been measured by “Sealed Can technique” using LR-115 type II detectors and a low level NaI (TI) based gamma ray spectrometer, respectively. From the measurements activity concentration of radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  vary from 9 to 28  $\text{Bq kg}^{-1}$ , 21 to 43  $\text{Bq kg}^{-1}$  and 280 to 554  $\text{Bq kg}^{-1}$  with overall average value of 19  $\text{Bq kg}^{-1}$ , 35  $\text{Bq kg}^{-1}$  and 406.7  $\text{Bq kg}^{-1}$  respectively. Radon equivalent activity ( $\text{Ra}_{\text{eq}}$ ) and external hazard index ( $\text{H}_{\text{ex}}$ ) have been found to vary from 60.8 to 121  $\text{Bq kg}^{-1}$  and 0.16 to 0.30 respectively. The values are lower than the recommended safe values.

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**Keywords:** Natural radioactivity; exhalation rate; radium equivalent activity; Gamma ray spectroscopy

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## 1. Introduction

Natural radioactivity content ( $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ ) in construction material are very important for the assessment of radiation exposure to human being since building materials act as a source of radiation and also shield against outdoor radiation. Indoor absorbed dose rate mainly depends on the activity concentration of natural radionuclides in building materials. Building material are made from natural sources (e.g., rock, soil) and products and also from industry (e.g., power plants, phosphate fertilizer and oil industry) products. Radionuclide content in different building materials depend on the chemical composition corresponding to their geological source. In uranium series, radon, a daughter product of radium is radiologically important to assess the radiation level in building materials. Radon ( $^{222}\text{Rn}$ ) and Thoron ( $^{220}\text{Rn}$ ) gases are present indoors due to the presence of their parents in soil and building materials. Other radon sources that they are in some cases relevant are water and natural gas. These gases contribute more than 50% of radiation doses received by individuals from natural radiation sources (UNSCEAR, 2000). Cement used as the material may be considered a source of radon and thoron in indoor environment has been studied for radon exhalation rate. Assessments of radiation exposure to the population and radiological parameters from the building materials are important as most people spend about 80% of their life inside houses and offices. This knowledge is also essential for the development of standards and guidelines for the safe use of these materials (Damla D et.al, 2010). In the present study radon exhalation rate and concentration of natural radionuclides have been measured in cement samples of various brands used in Aligarh (Uttar Pradesh) region of India. Radon exhalation rate is measured using Sealed Can Technique. Radionuclide contents  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  were measured using a low level gamma ray spectrometer. Different radiological parameters such as radium equivalent activity, absorbed dose, internal and external hazard index etc. were determined to find the radiological implication of the population.

## 2. Experimental Techniques

### 2.1. Radon exhalation rate

Radon exhalations rates are measured by using “Can techniques” (Singh A.K.et.al., 1997) Cement samples used in this study were collected from different manufacturers. For the measurement of Radon exhalation rate, fine quality (grain size  $150\mu\text{m}$ ) of each sample of 100 grams has been dried at  $110^{\circ}\text{C}$  in oven for twenty four hours for removing the moisture and then packed in air tight PVC container to prevent radon and thoron gases. LR-115 type II plastic track detector of size  $2.0\text{ cm} \times 2.0\text{ cm}$  was fixed at top inside of the Can. Thus the sensitive lower surface of the detector is freely exposed to the emergent radon so that it is capable of recording of alpha particles. Radon and its daughters reach an equilibrium concentration after 4 hours and thus the equilibrium activity of emergent radon could be obtained from geometry of the Can and time of exposure. Detectors have been fixed to expose for a period of 90 days, same were retrieved and etched in 2.5N NaOH solution for developing tracks at  $60^{\circ}\text{C}$  for the period of ninety minutes. Developed tracks were counted by using Spark counting systems.

### 2.2. Concentration of radium, thorium and potassium

A low level gamma spectrometric set up was used for measurement of the concentration of natural radionuclides, radium ( $^{226}\text{Ra}$ ), thorium ( $^{232}\text{Th}$ ) and potassium ( $^{40}\text{K}$ ). Each sample of 250 grams thus prepared was packed in plastic containers and sealed for four weeks (Shanbhag et.al, 2005) for the establishment of secular equilibrium between  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and decay products and to prevent  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  loss. The prepared cement samples were placed one by one in a shielded gamma ray spectrometry unit for 3 hours for counting (Ramola et.al, 2008). It consists of NaI (TI) gamma radiation detector of size  $63\text{ mm} \times 63\text{ mm}$  with a multichannel analyzer. The activity of  $^{40}\text{K}$  was evaluated from the 1460 KeV photo peak, the activity of  $^{226}\text{Ra}$  from the 1764 KeV Gamma lines of  $^{214}\text{Bi}$ , and that of  $^{232}\text{Th}$  from the 2610 KeV gamma lines of  $^{208}\text{Tl}$ . This spectral analysis was performed with the help of computer software SPTR-ATC (AT-1315).The peak energy of gamma spectra was measured in reference to the 661 KeV photo peak of  $^{137}\text{Cs}$ .The activity concentration of cement samples were calculated from the intensity of each line in the spectrum, considering the mass, the counting time, the geometry of samples and efficiency of the detector.

### 3. Results and Discussion

Surface exhalation rate and mass exhalation rate are obtained from the following expressions (Mahur et. al., 2013).

$$E_x = \frac{CV\lambda}{A T + \frac{1}{\lambda} e^{-\lambda T}} \quad (\text{Bq m}^{-2} \text{ h}^{-1}) \quad \text{----- (1)}$$

$$E_M = \frac{CV\lambda}{M T + \frac{1}{\lambda} e^{-\lambda T}} \quad (\text{Bq kg}^{-1} \text{ h}^{-1}) \quad \text{----- (2)}$$

Where,  $E_x$ , exhalation rate ( $\text{Bq m}^{-2} \text{ h}^{-1}$ );  $E_M$ , mass exhalation rate ( $\text{Bq kg}^{-1} \text{ h}^{-1}$ ); C, integrated radon exposure V is effective volume of Can;  $\lambda$  is decay constant of Radon is exposure ( $\text{h}^{-1}$ ); T is exposure time (h); A is covered area of Can ( $\text{m}^2$ ); M is mass of sample in Can.

Table 1. Radon activity, exhalation rate and radon effective dose in different cement samples

| Samples make | Radon Activity<br>( $\text{Bq m}^{-3}$ ) | Radon Exhalation<br>( $\text{mBq m}^{-2} \text{ h}^{-1}$ ) | Mass Exhalation<br>( $\text{mBq kg}^{-1} \text{ h}^{-1}$ ) | Radone ffective dose<br>( $\text{mSv y}^{-1}$ ) |
|--------------|--|--|--|---|
| BHIWANI      | 274 ± 23                                 | 99 ± 8   | 4 ± 0.3  | 12 ± 1  |
| JKSUPER      | 229 ± 21                                 | 82 ± 7   | 3 ± 0.2  | 10 ± 1  |
| ACC          | 86 ± 13                                  | 31 ± 5   | 1 ± 0.1  | 4 ± 1   |
| BANGUR       | 200 ± 10                                 | 72 ± 7   | 3 ± 0.2  | 8 ± 1   |
| AMBUJA       | 214 ± 20                                 | 77 ± 7   | 3 ± 0.2  | 9 ± 1   |
| SHREEULTRA   | 194 ± 19                                 | 70 ± 7   | 3 ± 0.2  | 8 ± 1   |
| ULTRATACH    | 320 ± 25                                 | 115 ± 9  | 4 ± 0.3  | 14 ± 1  |
| ROCKSTONE    | 171 ± 18                                 | 61 ± 6   | 2 ± 0.2  | 7 ± 1   |
| BIRLASAME    | 80 ± 12                                  | 29 ± 4   | 1 ± 0.1  | 3 ± 1   |
| JP           | 243 ± 21                                 | 87 ± 8   | 3 ± 0.2  | 10 ± 1  |

Table 2 Activity concentration values for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  and absorbed gamma dose rate, annual effective dose rate, Radium equivalent activity, external hazard index in cement samples

| Sample Make | $^{226}\text{Ra}$<br>( $\text{Bq kg}^{-1}$ ) | $^{232}\text{Th}$<br>( $\text{Bq kg}^{-1}$ ) | $^{40}\text{K}$<br>( $\text{Bq kg}^{-1}$ ) | ( $\text{Ra}_{\text{eq}}$ )<br>$\text{Bq kg}^{-1}$ | (D)<br>( $\text{nGy h}^{-1}$ ) | E<br>$\text{mSv y}^{-1}$ | Hex  |
|-------------|--|--|--|--|--------------------------------|--------------------------|------|
| BHIWANI     | 28 ± 10                                      | 40 ± 12                                      | 341 ± 10                                   | 111.4  | 51.3                           | 0.13                     | 0.30 |
| JKSUPER     | 10 ± 4                                       | 34 ± 10                                      | 430 ± 12                                   | 91.7   | 43.0                           | 0.11                     | 0.24 |
| ACC         | 9 ± 4  | 21 ± 9                                       | 284 ± 8                                    | 60.8   | 28.6                           | 0.07                     | 0.16 |
| BANGUR      | 24 ± 10                                      | 37 ± 11                                      | 573 ± 14                                   | 121.0  | 57.3                           | 0.14                     | 0.32 |
| AMBUJA      | 18 ± 9                                       | 33 ± 10                                      | 457 ± 12                                   | 100.3  | 47.3                           | 0.12                     | 0.27 |
| SHREEULTRA  | 19 ± 9                                       | 36 ± 11                                      | 443 ± 12                                   | 104.5  | 49.0                           | 0.12                     | 0.28 |
| ULTRATACH   | 25 ± 10                                      | 43 ± 13                                      | 280 ± 8                                    | 108.0  | 49.2                           | 0.13                     | 0.29 |
| ROCKSTONE   | 16 ± 8                                       | 39 ± 12                                      | 554 ± 13                                   | 114.4  | 54.0                           | 0.14                     | 0.30 |
| BIRLASAME   | 13 ± 6                                       | 32 ± 10                                      | 299 ± 9                                    | 81.7   | 37.8                           | 0.10                     | 0.22 |

Radon exhalation rate and mass exhalation rate are found to vary from  $28.7 \pm 4.4$  to  $115.0 \pm 8.8 \text{ mBq m}^{-2} \text{ h}^{-1}$  and  $1.1 \pm 0.1$  to  $4.4 \pm 0.3 \text{ mBq kg}^{-1} \text{ h}^{-1}$  respectively. The values are found minimum for BIRLASAME and maximum for ULTRATECH brand. Radon activities vary from  $4.4 \pm 0.6$  to  $17.9 \pm 1.3 \text{ Bq m}^{-3}$ . Activity is found minimum for BIRLASAME and maximum for ULTRATECH brand. Whereas calculated value of indoor inhalation exposure (Radon) and effective dose are found to vary from  $3.3 \pm 0.5$  to  $13.5 \pm 0.8 \text{ } \mu\text{Sv y}^{-1}$ . Effective dose have been found minimum for BIRLASANE and maximum for ULTRATECH also.results are shown in Table 1.

#### 3.1. Radium Thorium and Potassium in Samples

The activity concentration of  $^{226}\text{Ra}$  varies from  $9.0 \text{ Bq kg}^{-1}$  to  $28 \text{ Bq kg}^{-1}$  and  $^{232}\text{Th}$  varies from  $21 \text{ Bq kg}^{-1}$  to  $43 \text{ Bq kg}^{-1}$  whereas  $^{40}\text{K}$  varies from  $280 \text{ Bq kg}^{-1}$  to  $573 \text{ Bq kg}^{-1}$ . Cement samples used in this study, the activity of  $^{226}\text{Ra}$  is found minimum for ACC and maximum for Bhawani,  $^{232}\text{Th}$  concentrations are minimum for ACC and maximum for Ultratech and activity of  $^{40}\text{K}$  is found minimum for Ultratech and maximum for Bangur. Results are shown in Table 2.

### 3.2. Measurements of dose rate

Radiation risk to the human and other is a main factor of considering absorbed dose rate (UNSCEAR 2000) and radium equivalent activity (Yu et. al., 1992) Measurement of gamma radiation risk to the human by the cement used in building construction is done by using the following equations:

$$Ra_{eq} = C_{Ra} + 1.43 C_{Th} + 0.07 C_K \quad (3)$$

$$D = (0.462C_{Ra} + 0.604 C_{Th} + 0.0417 C_K) \quad (4)$$

To estimate the annual effective dose rate E, the conversion coefficient from absorbed dose in air to effective dose ( $0.7 \text{ Sv Gy}^{-1}$ ) and outdoor occupancy factor (0.2) proposed by (UNSCEAR, 2000) were used. The effective dose rate was calculated by the following relation:

$$E (\text{mSv y}^{-1}) = \text{Dose rate (nGy h}^{-1}) \times 8760\text{h} \times 0.8 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \quad (5)$$

$$E (\text{mSv y}^{-1}) = \text{Dose rate (nGy h}^{-1}) \times 8760\text{h} \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \quad (6)$$

Radium equivalent activities in cement samples vary from 60.8 to 121 Bq kg<sup>-1</sup>. The minimum was found for ACC and maximum for Bangur. The absorbed gamma dose rates (D) and corresponding outdoor annual effective dose varies from 28.6 to 57.3 nGy h<sup>-1</sup> and 0.07 to 0.14 mSv y<sup>-1</sup> respectively. The minimum dose was found for ACC while maximum was for Rockstone cement. Calculated results are given in Table-2. The annual effective dose rate calculated in this study is lower than the recommended value 1 mSv y<sup>-1</sup>.

### 3.3. External hazard index

$$H_{ex} = \frac{C_{Ra}}{370(\text{Bqkg}^{-1})} + \frac{C_{Th}}{259(\text{Bqkg}^{-1})} + \frac{C_K}{4810(\text{Bqkg}^{-1})} \leq 1 \quad (7)$$

The calculated values of H<sub>ex</sub> for these cement samples studies in this work vary from 0.16 to 0.32. The minimum value of H<sub>ex</sub> was found for ACC and maximum was for Bangur.

## 4. Conclusion

The value of external hazard index H<sub>ex</sub> is less than unity and is less than the recommended safe value. The values of radium equivalent activity of cement samples are less than 370 Bqm<sup>-3</sup> recommended level (UNSCEAR, 2000). The Natural radioactivity of the samples used in the present study has been found to be lower than the permissible level of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K respectively (UNSCEAR, 2000). The values of absorbed dose, annual effective dose were also found to be below the permissible limits. Hence we conclude that results for the cement samples used in present study indicate that cement can be used safely as building construction material.

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