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Radiological assessment of cement and clay based building materials from southern coastal region of Kerala

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Building materials are one of the potential sources of indoor radioactivity because of the naturally occurring radio nuclides in them. External as well as internal exposures are the two pathways of radiation dose imparted to the human beings from the building materials. Natural clay is the main raw material for the production of bricks and tiles. In the present study, samples of cement, bricks, floor and wall tiles have been analyzed for the primordial radio nuclides namely, uranium, thorium and potassium using gamma ray spectrometry. Radon exhalation rates of these materials were also measured. Among the samples analyzed, the minimum radium equivalent activity was found in bricks (66.08 $\mathrm{Bqkg^{-1}}$) and the maximum was found in cement (225.46 $\mathrm{Bqkg^{-1}}$). External gamma dose, effective dose equivalent, internal and external hazard indexes resulting from the radio nuclides were also determined. The radon exhalation rates from building materials were found to vary between 10.5±2.8 and 82.8±8.2 $\mathrm{mBqh^{-1}m^{-2}}$.

Keywords: Uranium, Thorium, Radon, Building materials, Hazard index, Radon exhalation

1 Introduction

Naturally occurring radioactive materials (NORM) are found everywhere including soil, rock, vegetation, air, and water in varying levels. The most commonly found primordial radio nuclides in soil are ^{238}U and ^{232}Th, their subsequent radioactive decay products and ^{40}K. Building materials are either part of the environment or derived from naturally occurring materials. Cement, Bricks and other clay based building materials like tiles are used extensively for the construction of buildings. Use of industrial waste materials like fly ash for the manufacture of cement and cement bricks are increased manifold in the recent years resulting in Technologically Enhanced Naturally Occurring Radioactive Materials[1] (TENORM). The reasons for the use of industrial wastes products as admixtures in building materials are cost reduction, saving of natural resources and recycling. Fly ash is known to contain elevated levels of long lived radio nuclides like uranium[1]. Owing to its relatively small size and hence, large surface area, fly ash has a greater tendency to absorb trace elements that are transferred from coal to waste products during combustion[2]. The naturally occurring radio nuclides constitute the source of indoor and outdoor radiation exposure. Assessment of NORM in building materials and radon exhalation rate from them are of radiological importance since they contribute more than 50% of the total dose received by human beings[3]. The south west coastal region of India is known to have high natural background radiation mainly because of the thorium rich monazite sand available in plenty in the region. In the present study, samples of cement, bricks, floor and wall tiles have been analysed for their radiological characterization using gamma ray spectroscopy and the radon exhalation measurements have been done using well accepted passive measurement using SSNTDs.

2 Experimental Details

2.1 Radiometric analysis (for uranium, thorium and potassium)

There are a large variety of building materials being used for construction of dwellings in the region. Most commonly used brands of cements and ceramic tiles in India were selected for the study. Samples of cement, bricks and tiles were collected from the construction sites situated in the coastal region of Kollam district in the state of Kerala. For making bricks, clay is collected from local fields. There are several brick factories in the region and therefore, it was difficult to analyze all brands of bricks available.

Hence, the selection of brick samples was at random from the construction sites. The samples include six brands of most commonly used cement (C1-C6), four brands of clay based wall tiles (WT1-WT4) and floor tiles (FT1-FT4), and eight brands of bricks (B1-B8). Two to four samples of each brand and category were collected for the analyses. Altogether sixty samples were collected and analyzed. Cement samples were dried in hot air oven, sieved and stored in the polyethylene cans of specific size (70 mm diameter and 80 mm height). The tiles and bricks were crushed to small pieces and ground to fine grain size and sieved in order to homogenize the sample. The samples were then dried at 110°C for 24 h to ensure that moisture is completely removed. The samples were then transferred to standard polyethylene cans and sealed hermetically with adhesive tape and left for 6 weeks, in order to ensure that the daughter products of ^{226}Ra up to ^{210}Pb and those of ^{232}Th up to ^{208}Pb are in secular equilibrium with their respective parent radio nuclides before analyzing by gamma spectrometry. The amount of samples taken in the sealed cans for analysis varied from 242 to 268 g in the case of cements, from 265 to 278 g for floor tiles, from 258 to 274 g for wall tiles and from 188 to 222 g for bricks.

Analysis of the sample for determining the levels of ^{238}U, ^{232}Th and ^{40}K were done using a flat type 3″×3″ NaI(Tl) detector, housed in a 3″ thick graded lead shield. The detector was coupled to a PC through an 8 K MCA. The energy resolution of the detector was 1.95 keV at 1332 keV of a standard ^{60}Co source. A cylindrical source (having homogeneously distributed activity with constant volume and shape) was placed coaxially with the detector for efficiency determination and the same procedure was applied for sample measurements. The activity of ^{40}K was evaluated from the 1460 keV photo peak of its own gamma, the activity of ^{238}U from 1764 keV gamma ray of ^{214}Bi and that of ^{232}Th from 2614 keV gamma ray of ^{208}Tl. Each sample was counted for 18000s. Background counts were deducted for obtaining the net activity. In the uranium series, the decay chain segment starting from radium (^{226}Ra) is radiologically the most important and, therefore, reference is often made to radium instead of uranium. Along with the assessment of ^{238}U, ^{232}Th, and ^{40}K levels, radiological parameters like the radium equivalent activity (Ra_{eq}), the external hazard index (H_{ex}), the internal hazard index (H_{in}), the indoor gamma absorbed dose (D) and the corresponding annual effective dose equivalent (EDE) were also determined. These parameters are important in radiation risk assessment.

2.2 Measurement of radon exhalation rates

Radon exhalation rates from the building materials were determined using the well established ‘Can Technique’ described elsewhere[4–6]. The diameter of the dosimeter cup was 7 cm and the height was 4.5 cm. Two samples each of bricks, cements, wall tiles and floor tiles were selected for the exhalation measurements. Dosimeter cups equipped with LR-115 Type II pelliculable films of 2cm × 2cm size were sealed firmly on to the surface of bricks and tiles were left for exposure for six weeks for exhalation measurements. For measurement of radon exhalation rate from cements, specific amount of samples were filled in the dosimeter cup. In every case, the distance of the films from the sample was kept 4.5 cm. The radon gas exhaled from the building material into the dosimeter cup will reach equilibrium with its progeny typically after two weeks or more. After exposure, the detectors were chemically etched in 2.5 N NaOH solution at 60±1°C for 90 min in a constant temperature water bath for the revelation of the tracks through preferential chemical etching. The sensitive part of the LR 115 film was peeled off and scanned with a spark counter to determine the track density on the detector films. Using the track density, the equilibrium activity concentration of the radon inside the cup can be obtained using the calibration factor for the open cup[6] (0.056 Bqm^{-3} per tracks cm^{-2} day^{-1}). The area specific exhalation rate of radon can be calculated using the relation:

$$E_x = (CV\lambda/A) \,/\, [T + 1/\lambda(e^{-\lambda t} - 1)] \qquad \dots (1)$$

where E_x is the radon exhalation rate (Bqh^{-1}m^{-2}), C the integrated radon exposure (Bqm^{-3}h), V the volume of the Can (173 cm^3), A the area covered by the Can (38.5 cm^2), λ the decay constant of radon (0.0075 h^{-1}) and T is the exposure time (~1000 h).

3 Results and Discussion

3.1 Uranium, thorium and potassium concentrations in the samples

The results of radiometric analysis of the samples are presented Tables 1-3. Table 1 shows the measured specific activity concentrations of the ^{238}U, ^{232}Th, and ^{40}K in the nineteen cement samples, fourteen floor tiles, eleven wall tiles and sixteen bricks. Distribution of ^{238}U, ^{232}Th and ^{40}K in the samples analyzed has a heterogeneous distribution with no specific relation between them. Therefore, with respect to exposure to radiation, the radioactivity may be defined in terms of radium equivalent activity (Ra_{eq}) to compare the specific activity of materials containing different amounts of ^{238}U, ^{232}Th and ^{40}K in different combinations. The parameter Ra_{eq} has been defined on the fact that 370 Bqkg^{-1} of ^{226}Ra, 260 Bqkg^{-1} of ^{232}Th and 4810 Bqkg^{-1} of ^{40}K produce the same gamma ray dose. Thus, the radium equivalent activities (Ra_{eq}) were estimated using the equation[3,7,8].

Table 1 — Average specific activity concentrations of the ^{238}U, ^{232}Th, and ^{40}K in the samples

Sample Code	Number of samples	Uranium-238 (Bqkg^{-1})	Thorium-232 (Bqkg^{-1})	Potassium-40 (Bqkg^{-1})
C1	3	56.76	67.89	567.33
C2	3	77.45	68.24	612.67
C3	3	48.62	80.43	438.84
C4	3	62.42	72.75	566.24
C5	3	44.24	62.22	410.22
C6	4	34.48	44.65	348.68
FT1	3	45.33	62.44	448.24
FT2	4	37.65	58.46	496.43
FT3	3	60.52	65.34	472.98
FT4	4	44.26	56.88	512.65
WT1	3	42.88	52.66	523.88
WT2	2	36.89	59.6	468.74
WT3	3	58.66	60.33	542.64
WT4	3	43.26	54.77	442.23
B1	2	20.18	20.68	283.14
B2	2	18.45	20.44	264.22
B3	2	25.45	28.34	276.54
B4	2	15.67	19.45	320.69
B5	2	27.33	30.11	289.45
B6	2	23.56	27.45	318.32
B7	2	15.32	21.25	277.45
B8	2	17.66	20.23	286.28

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_K \qquad \ldots (2)$$

where C_U, C_{Th} and C_K are activity concentration in Bqkg^{-1} of ^{238}U, ^{232}Th and ^{40}K, respectively.

Table 2 presents the statistics of the results of analyses of the building materials. The specific activity concentration of ^{238}U, ^{232}Th, ^{40}K and radium equivalent activity (Ra_{eq}) in cement samples were found to have mean values of 54±15.06, 66.03±12.10, 490.66±105.63 and 186.20±35.50 Bqkg^{-1}, respectively. In floor tiles, these quantities were found to have mean values of 46.94±9.67, 60.78±3.84, 482.58±28.09 and 171.01±13.50 Bqkg^{-1}, respectively whereas for wall tiles they were 45.42±9.3, 56.84±3.72, 494.37±46.82 and 164.77±14.69 Bqkg^{-1}, respectively. For bricks, these quantities were 20.54±4.52, 23.49±4.35, 289.51±20.02 and 76.34±10.76 Bqkg^{-1}, respectively. The uncertainties quoted are the standard deviation from the mean values. The Ra_{eq} values obtained for all types of building materials are much below the limit of 370 Bqkg^{-1} raising no radiological hazard to the public.

3.2 Dose and hazard indices determination

With the measured activities in building materials, external gamma dose rate (D) in air has been calculated using the equation[3]:

$$D = 0.427\, C_U + 0.662\, C_{Th} + 0.043\, C_K \qquad \ldots (3)$$

where 0.427, 0.662 and 0. 043 (nGyh^{-1} per Bqkg^{-1}) are the dose conversion factors for ^{238}U, ^{232}Th and

Table 2 — Maximum, minimum, mean and standard deviation of the levels of ^{238}U, ^{232}Th, ^{40}K and the Radium equivalent activity in the building materials

		Uranium-238 (Bqkg^{-1})	Thorium-232 (Bqkg^{-1})	Potassium-40 (Bqkg^{-1})	Ra_{eq} (Bqkg^{-1})
	Maximum	84.02	82.43	614.66	225.46
Cement	Minimum	30.08	40.67	346.22	123.47
19 samples	Mean	54	66.03	490.66	186.20
	S D	15.06	12.10	105.63	35.50
	Maximum	68.54	66.64	510.06	192.28
Floor tiles	Minimum	34.66	55.26	444.82	154.46
14 samples	Mean	46.94	60.78	482.58	171.01
	S D	9.67	3.84	28.09	13.50
	Maximum	62.08	61.42	544.68	187.62
Wall Tiles	Minimum	34.86	52.48	440.62	153.42
11 samples	Mean	45.42	56.84	494.37	164.77
	S D	9.3	3.72	46.82	14.69
	Maximum	28.88	31.12	324.42	93.87
Bricks	Minimum	13.68	18.84	260.88	66.08
16 samples	Mean	20.54	23.49	289.51	76.34
	S D	4.52	4.35	20.02	10.76

^{40}K, respectively[7]. In order to estimate the annual effective dose rate from absorbed dose, the conversion coefficient of 0.7 SvGy^{-1} and the indoor occupancy factor of 0.8 has been used as suggested[3] by UNSCEAR 2008. The external hazard index (H_{ex}) resulting from the exposure to gamma rays of the primordial nuclei has been evaluated as an estimate of radiation risk if a building is constructed using these materials using the relation[6,7] The value of this index must be less than unity in order to keep the radiation hazard to be insignificant.

$$H_{ex} = C_U(370)^{-1} + C_{Th}(259)^{-1} + C_K(4810)^{-1} \quad ...(4)$$

A similar hazard index called the internal hazard index (H_{in}) for the internal exposure due to the inhalation of ^{222}Rn and its radioactive progeny has also been evaluated using the equation[7,8]. For the safe use of a material in construction of dwellings, index should be less than unity

$$H_{in} = C_{Ra}(185)^{-1} + C_{Th}(259)^{-1} + C_K(4810)^{-1} \quad ...(5)$$

Results of these analyses are presented in Table 3. All the calculated hazard indices are well within the limit of unity. Contributions of building materials towards the total annual effective dose are presented in Table 3. Table 2 indicates that the concentrations of uranium and thorium are slightly more than the world average of these isotopes in soil samples. The worldwide concentrations of the radionuclides ^{238}U, ^{232}Th and ^{40}K in soil have averages 40, 40 and 580 Bqkg^{-1}, respectively[3].

Table 3 — Calculated values of radium equivalent activity (Ra_{eq}) external hazard index (H_{ex}), internal hazard index (H_{in}), gamma dose (D) and effective dose equivalent (EDE) of the samples

Sample Code	Ra_{eq} (Bqkg^{-1})	H_{ex}	H_{in}	D (nGyh^{-1})	EDE (mSvy^{-1})
C1	197.53	0.53	0.69	93.57	0.46
C2	222.21	0.60	0.81	104.59	0.51
C3	197.43	0.53	0.66	92.88	0.46
C4	210.05	0.57	0.74	99.16	0.49
C5	164.80	0.45	0.56	77.72	0.38
C6	125.18	0.34	0.43	59.27	0.29
FT1	169.13	0.46	0.58	79.97	0.39
FT2	159.47	0.43	0.53	76.12	0.37
FT3	190.38	0.51	0.68	89.44	0.44
FT4	165.07	0.45	0.57	78.60	0.39
WT1	158.52	0.43	0.54	75.70	0.37
WT2	158.21	0.43	0.53	75.36	0.37
WT3	186.72	0.50	0.66	88.32	0.43
WT4	155.63	0.42	0.54	73.75	0.36
B1	71.55	0.19	0.25	34.48	0.17
B2	68.02	0.18	0.23	32.77	0.16
B3	87.27	0.24	0.30	41.52	0.20
B4	68.18	0.18	0.23	33.36	0.16
B5	92.67	0.25	0.32	44.05	0.22
B6	87.32	0.24	0.30	41.92	0.21
B7	67.07	0.18	0.22	32.54	0.16
B8	68.63	0.19	0.23	33.24	0.16

3.3 Radon exhalation rates from building materials

The results of radon exhalation rate measurements from building materials were found to be in the range 11.5± 2-17.8± 3.1 mBqh^{-1}m^{-2} for floor tiles, 10.5± 2.8-21.8± 2.9 mBqh^{-1}m^{-2} for wall tiles, 44.4±2-77.8± 4.1 mBqh^{-1}m^{-2} for bricks and 38.5±7.2-82.8±8.2 mBqh^{-1}m^{-2} for cement. The variations found in the exhalation rate measurements may be because of the different diffusion coefficients and diffusion lengths of radon in these materials. Figure 1 compares the results of analysis of cements carried out in Hungary[9], Iran[10] and Turkey[11]. Figure 2 compares the reported values for bricks from Sri Lanka[12], Pakistan[13] and Jalandhar in India[14]. Figure 3 shows the values obtained for ceramic tiles from China[15] and another study held in Tamilnadu, India[16]. A recent study of radon exhalation rate in another high background radiation area in India through the same methodology was found to vary between 423.2 to

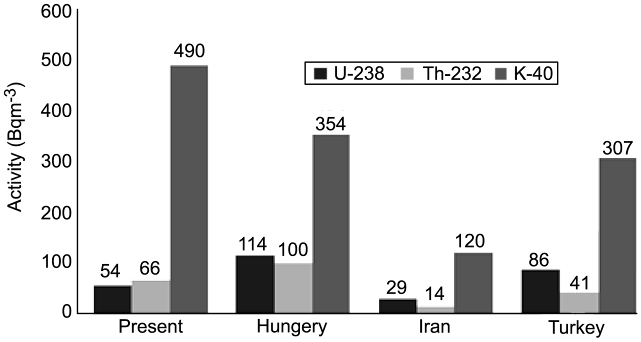

Fig. 1 — Comparison of radioactivity (Bqkg^{-1}) in cements of different countries

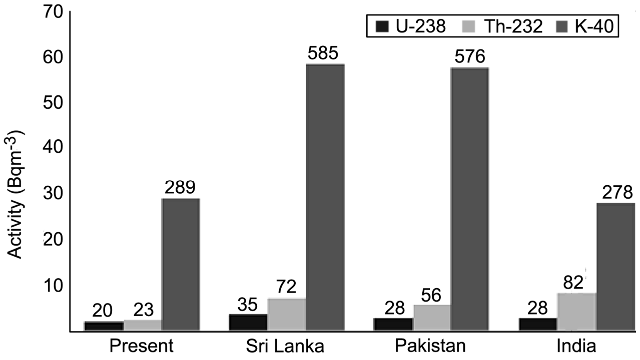

Fig. 2 — Comparison of radioactivity (Bqkg^{-1}) in bricks of different countries

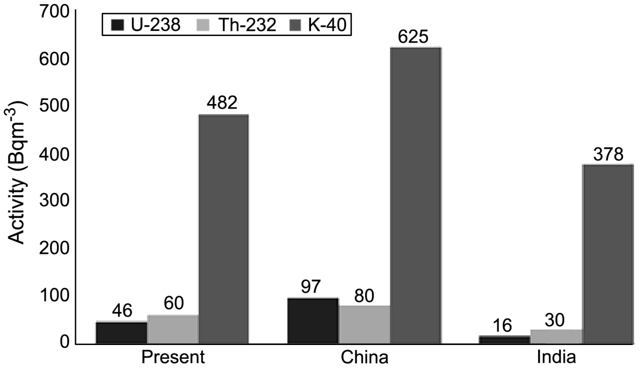

Fig. 3 — Comparison of radioactivity (Bqkg^{-1}) in tiles of different studies

1636.3 mBqh^{-1}m^{-2} with an average value of 763.9 mBqh^{-1}m^{-2} (Ref. 17). In general, the radioactive levels of primordial radio nuclides show an exorbitant range in the building materials indicating the necessity of regionwise assay of samples and radiometric parameters for a meaningful estimation of population dose.

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