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Radioactivity analysis of commonly available building and flooring material in Kerala, India

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ABSTRACT

Building materials are primary sources of indoor gamma radiation. Therefore, we have made a detailed study of building materials which are being used extensively in the region. Primordial radionuclides namely uranium (^{238}U), Thorium (^{232}Th) and potassium (^{40}K) were studied in the building materials. As of the results acquired, we can arrive at the following inferences: We have estimated specific activities of radionuclides, the radium equivalent and radiological risk parameters resulting from the gamma originating from building materials. The highest Radium equivalent activity was obtained for red clay brick and the minimum value obtained was for M-sand. M-sand is processed sand being used widely now a days in the region. Most of the building materials were found to have very low levels of Radium. As of the results we obtained, none of the building materials were found to pose any notable radiological health hazard.

Key words : *Building materials, Primordial radionuclides, Gamma ray spectroscopy dose rate, Radiation hazard, Activity concetration index*

Introduction

A major source term of the indoor gamma activity is building materials. As individuals spend more than 80% of their time indoors, the internal and external radiation exposure from building materials creates prolonged exposure situations (ICRP, 1999). The worldwide average indoor effective dose due to gamma rays from building materials is estimated to be about 0.4 mSv per year (UNSCEAR, 2000). In many parts of the world, building materials containing radioactive material have been used for generations. Most building materials of terrestrial origin contain small amounts of NORM, mainly radionuclides from the Uranium-238 (238U) and Thorium-232 (232Th) decay chains and the radioactive isotope of Potassium-40 (40K). The external radiation exposure is caused by the gamma emitting radionuclides, which in the uranium series mainly belong to the decay chain segment starting with Radium-226 (226Ra). The internal (inhalation) radiation exposure is due to Radon-222 (222Rn), and marginally to Radon-220 (220Rn), andtheir short lived decay products, exhaled from building materials into the room air (Papastefanou *et al*., 2005). Coal ash, produced as waste in the combustion of coal, issued as an additive to cement, in concrete and in some countries bricks are made from fly ash. Coal slag is used in floor structures as insulating filling material. Phosphogypsum, a by-product in the production of phosphorous fertilizers is used as building material, and red mud, a waste from primary aluminum production, is used in bricks, ceramics and tiles.

Although the building materials absorb the radiation that originates outside the building, exposure within the building is more than compensated by the presence of radionuclides in the materials of construction.

Materials and Methods

Sampling and Sample Preparation

Samples for this study, building materials being used for construction and used for flooring of dwellings, were collected from the Normal Background Radiation Area in different locations in Thiruvananthapuram and Kollam districts. Commonly used brands of cements, joint filler and granites were selected for the study. Locally available bricks (two types), sand, m sand and rock powder were also taken for study. Materials like white oxide, yellow oxide, red oxide, and black oxide used for flooring were also selected.

Collected samples were dried at 110^0 C for 24 h to remove moisture. All the selected samples were made into fine powder and were stored in the polyethylene cans of specific size (70 mm diameter and 80mm height) for analyzing using Gamma Ray Spectrometer. The hermetically sealed airtight containers were kept for about four weeks to ensure the secular equilibrium between ^{226}Ra (of the ^{238}U) and ^{232}Th and with their radioactive progenies.

Methodology

Convenient and non-destructive method analysis of the sample for determining the levels of ^{238}U, ^{232}Th and ^{40}K were done using a 5"×4" NaI (Tl) detector based on Gamma ray spectrometry , housed in a 3"thick graded lead shield., PC coupled 8 K MCA. The energy resolution of the detector was 1.95 keV at 1332 keV of a ^{60}Co source. A cylindrical source was placed coaxially with the detector for determining efficiency and the same procedure was applied for sample measurements. The measurement was carried out in three steps: energy calibration, sensitivity calibration and gamma-ray analysis. The energy calibration was carried out by two radioactive calibration sources, ^{137}Cs and ^{57}Co. The sensitivity calibration was achieved by using three artificial standard sources of Ra, Th and K. 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 10000s. Background counts were deducted for obtaining the net activity. The output of the detector is coupled by an 8 K Multi Channel Analyzer. The spectra obtained were analyzed using the WINTMCA software.

Results and Discussion

Uranium, Thorium and Potassium concentrations of the sample

Specific activity analyses of the collected building materials are presented in the Table 1. The table shows the activity concentrations of 2 samples Black oxide, Yellow oxide, Red oxide and White oxides,4 samples of Cement, 2 samples of Granite, 3 samples of Sand, M sand and Rock powder and 2 samples of Red clay bricks. The worldwide average concentrations of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K reported by UNSCEAR (2000) are 35, 30 and 400 Bqkg^{-1} are respectively. Table 2 shows the comparative study of the assessment of building materials worldwide. Below Detectable Value(BDL) of ^{40}K, ^{226}Ra and ^{232}Th are 27.18 Bqkg^{-1}, 4.7 Bqkg^{-1}and 14.3 Bqkg^{-1} respectively. Activity concentrations of ^{232}Th ranged from 14.3 to 160Bqkg^{-1}, of^{226}Ra from 4.7 to 85Bqkg^{-1} and of ^{40}K from 27.18 to 204.85 Bqkg^{-1}were observed from the investigated material. Activity concentrations of ^{232}Th are within the range of figures reported in UNSCAR. But the concentration of ^{226}Ra in brick sample, the value is slightly greater than the world average. However the concentration of ^{40}K in most of the samples shows below detectable value. The distribution of radionuclides in building material is not uniform. Therefore, a common index namely Radium Equivalent Activity has been introduced to represent the radioactivity levels of radium, thorium and potassium in the samples, which takes into account the radiation hazards associated with them. It is based on the assumption that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40} K produce the same gamma ray dose rate equivalent. Thus, the radium equivalent activities may be calculated using the formula (Viruthagiri *et al.*, 2013).

$$R_{eq} = 370\left(\frac{ARa}{370} + \frac{ATh}{259} + \frac{Ak}{4810}\right)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations in Bq kg-1 for $_{226}$Ra, $_{232}$ Th and $_{40}$ K, respectively.

Figure 1 represents the graphical analysis of the radium equivalent of the building material collected as sample. The recommended limits of the radium equivalent activity for building materials must be less than 370Bq kg^{-1} to the dwellings or homes[6]. In the study, observed values of radium equivalent was less than 370 Bq/Kg. The radium equivalent activity (Ra_{eq}) values for all building materials under investigation ranged from 34.55 to 324.75 Bq kg^{-1}, which are safe to use.

Estimation of dose rate

UNSCEAR (1988) has given the dose conversion factors for converting the activity concentrations of $_{226}$Ra, $_{232}$Th and $_{40}$K into doses (nGy.h^{-1} per Bq.kg^{-1}) as 0.427, 0.662 and 0.043, respectively.

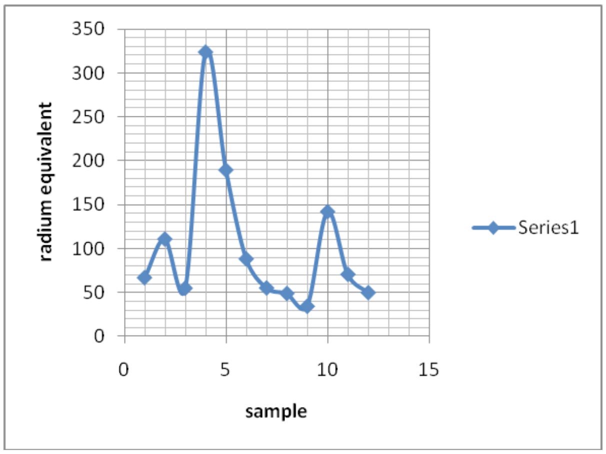

Fig. 1. Analysis of radioactivity equivalent

The gamma radiation population doses of those living m the area are given as:

$$D = 0.427A_{Ra} + 0.662\ A_{Th} + 0.043\ A_K$$

Where D is the dose rate in nGy.h^{-1} and A_{Ra}, A_{Th}, A_K are the concentrations of Radium, Thorium and Potassium, respectively (Viruthagiri *et al.,* 2013).

The annual effective dose rate outdoors in units of $mSvy^{-1}$ is calculated by the following formula:

Annual Effective Dose Rate = D × T× F

Where D is the calculated dose rate (in nGy h^{-1}), T is the indoor occupancy time (0.8 × 24 h × 365.25 days =1753 h y^{-1}) and F is the conversion factor (0.7 × 1026 $SvGy^{-1}$)

The worldwide average annual effective dose from natural sources is estimated to be 2.4 mSv of which about 1.1 mSv is due to the basic background radiation and 1.3 mSv is due to the basic background radiation is due to exposure to radon (UNSCEAR 1993). Annual effective dose observed in the building material under study ranges with in the world average.

Assessment of radiation hazard from building materials

There important parameter to evaluate the hazard of natural gamma radiation, External hazard index H_{Ex}, Internal hazard index H_{In} and Gamma index $I_{a.}$

External Hazard index is defined as

$$H_{Ex} = \frac{ARa}{370} + \frac{ATh}{259} + \frac{AK}{4810}$$

Where A_{Ra}, A_{Th} and A_K be the activity concentrations in Bq $kg^{-1}$of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

The value of this index must be less than unity in order to keep the radiation hazard to be insignificant. The maximum value of H_{ex} equal to unity corresponds to the upper limit of Ra_{eq} (370 Bq kg^{-1}) (UNSCEAR, 1993). The calculated values of external hazard index obtained in this study ranged from 0.09 to 0.88.

Table 1. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of the building materials

No	Name	Activity of ^{226}RaBqkg^{-1}	Range	Activity of ^{232}ThBqkg^{-1}	Range	Activity of ^{40}KBqkg^{-1}	Range
1	Black oxide	19.03±13	BDL-19.03	32.35±0.5	14.13-5.04	BDL	BDL
2	Granite	BDL	BDL	**70.22±1.45**	65-74	84.81±13.96	72-97
3	Red Oxide	32.69±0.54	30-35	BDL		BDL	BDL
4	Choolakkata	**83.24±5.8**	80-85	**158.23±13.89**	155-160	204.85±108.77	200-250
5	Kambikatta	**83.13±2.61**	81-84	**73.52±2.7**	72-75	BDL	BDL
6	Cement	34.83±0.87	12-54	36.22±1.665	30-50	BDL	BDL
7	White Oxide	9.82±0.17	8-12	30.56±0.64	25-45	BDL	BDL
8	M Sand A	14.72±0.31	10-15	22.65±0.57	20-25	BDL	BDL
9	Rock powder	12.11±0.42	10-15	BDL		BDL	BDL
10	Sand A	34.22±1.005	30-40	**74.5±2.665**	70-80	BDL	BDL
11	Yellow Oxide	BDL		45.04±0.75	40-50	BDL	BDL
12	Joint filler	BDL		30.4±0.55	25-40	BDL	BDL

Internal Hazard Index is defined as

$$H_{In} = \frac{ARa}{185} + \frac{ATh}{259} + \frac{Ak}{4810}$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations in Bq kg^{-1} for $_{226}Ra$, $_{232}$ Th and $_{40}$ K, respectively (Masitah Alias *et al.*, 2008). For the safe use of a material in the construction of dwellings, index (H_{in}) should be less than unity and the values obtained from the study is also less than unity.

Activity concentration index

Activity concentration index is used for the monitoring of the material which is used for building purpose. It is calculated by using the following equation (European union commission Report, 1999).

$$I\gamma = \left(\frac{ARa}{300\,Bq\,kg-1,} + \frac{ATh}{200\,Bq\,kg-1,} + \frac{Ak}{3000\,Bq\,kg-1}\right)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations in Bq kg^{-1} of $_{226}$ Ra, $_{232}$ Th and $_{40}$K respectively. It is recommended that $I\gamma \leq 0.5$ for concrete materials and $I\gamma \leq 2$ for other materials like tiles etc in the absorbed dose rate of 0.3 $msva^{-1}$. If absorbed dose rate criteria is $1msva^{-1}$ $I\gamma \leq 1$ for concrete materials and $I\gamma \leq$ 6 for other materials like tiles etc (European union commission Report, 1999).

Table 2. Comparative study of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of the building materials

Sample	Country/Place	Activity concentration (Bq/Kg)			Reference
		^{226}Ra	^{232}Th	^{40}K	
Cement	Algeria	41±7	27±3	422±3	Amrania *et al*., 2001
	Cyprus	4.4-60.3	0.6-12.3	4.1-289.2	Michael *et al*., 2010
	Iran	31.1	12.4	121	Fathivand *et al*., 2007
	China	39.7	34.3	189	Xinwei *et al*., 2008
	Pakisthan	25-55	10-30	15-300	Mujahid *et al*., 2008
	Egypt	48	22	220	Medhat *et al*., 2010
	Iraq	67-223.7	15-53	0.5-231.9	Kamal *et al*., 2010
	Turkey	12-41	12-44	9-254	Atayatufan *et al*., 2012
	Albenea	49.8-58	15.1-19.8	157-230	Xhixha *et al*., 2013
	Vietnam	30-110	20-90	60-300	LeNhusien *et al*., 2009
	Present Study	**12-54**	**30-50**	**BDL**	
Red Clay Brick	Algeria	65±7	51±5	675±4	Amrania *et al*., 2001
	China	37.9	46.5	697.4	Xinwei *et al*., 2008
	Egypt	30	21	289	Medhat *et al*., 2010
	Punjab	21-47	22-58	299-918	Asghan *et al*., 2010
	Iraq	24.4-69.6	10.8-11.7	119-180	Kamal *et al*., 2010
	Turkey	15-25	10-40	350-500	Atayatufan *et al*., 2012
	India	19 -25	20-32	318-348	Rohit Mehru *et al*., 2009
	Present Study	**80-85**	**70-158**	**BDL-205**	
Sand	Algeria	12±1	7±1	74±7	Amrania *et al*., 2001
	China	43.7	64.4	455.8	Xinwei *et al*., 2008
	Egypt	33	27	385	Medhat *et al*., 2009
Iraq	10-52	0.5-21	40-220	Kamal *et al* .,2010	
	Turkey	10-20	10-30	9-235	Atayatufan *et al*., 2012
	Present Study	**10-15**	**20-25**	**BDL**	
Granite	Cyprus	0.2-81	5.7-260	920.7-1576	Michael *et al*., 2010
	Turkey	15.85	33.76	359	Ahmet *et al*., 2006
	Egypt	65	60	920	Medhat *et al*., 2009
	Present study	**BDL**	**65-74**	**72-97**	
Rock Powder	Cyprus	0.1-40.2	0.3-6.4	4.6-147.3	Michael *et al*., 2010
	Yemen	10-80	4-125	30-2222	Al.hayadrei*et al*., 2012
	Rom	12-53.8	1.5-92	9-675	Trevisil*et al*., 2005
	Present Study	**10-15**	**BDL**	**BDL**	
White Cement	Pakisthan	25-35	10-20	17-32	Mujahid *et al*., 2008
	Egypt	21±0.64	9.90±0.30	16.22±0.49	Medhat *et al*., 2009
	Present Study	**25-45**	**5-15**	**BDL**	

Annual effective dose rate (AED)

By using the conversion factor 0.7 svGy^{-1} the gamma absorbed doses in nGy h^{-1}were converted to annual effective dose in mSvy^{-1} as proposed by (UNSCEAR, 2000). The annual effective dose rate (AED) was calculated by using the following equation.

AED=D × T × F

D is the dose rate nGy^{-1} T is the indoor occupancy factor (0.8 × 24h× 365.25 =7013Gy^{-1}) and conversion factor 0.7 SvGy^{-1}.The calculated AED values were vary from 14.23 to 130.84 µSvy^{-1} the values are under the recommended limit 0.07mSvy^{-1} reported by UNSCEAR, 2000.

Excess life time cancer risk (ELCR)

Excess life time cancer risk is evaluated by using the equation –

ELCR= AED × DL × RF

Where DL is duration of life (65.8 year) and RF is risk factor (Sv^{-1}), it is fatal cancer risk per Sievert. For stochastic effects from low dose background radiation, suggested the value of 0.057 for the public exposure (Amrania *et al.*, 2001). Table 3 shows the calculated values of absorbed dose rate, external hazard index, Activity concentration index, Annual effective dose (AED) Excess life time cancer risk (ELCR). The highest value of ELCR value is 4.588 × 10^{-3} lower than the average value 2.9 × 10^{-4} recommended by ICRP.

Conclusion

NORMS and its related radiation hazards in building materials along Kerala have been studied using gamma spectrometry. The activity concentration levels of various radionuclides were also determined. The ^{226}Ra content have the activity ranges from 9.82 ± 0.17 to 83 ± 5.8. The highest value showed by Red clay bricks found locally. The highest value of ^{232}Th and ^{40}K were also showed by the same. The radium equivalent (Ra_{eq}) varied from 34.55 to 324.17 Bqkg^{-1}. The highest value occurring in brick sample while the lowest value occurring in Rock Powder. The absorbed dose rate in air ranged from 15.37 to 142.57 nGyh^{-1} with the maximum dose rate resulting in bricks. Therefore, the uses of above material which are under study are safe for the construction of inhabitants. The average value of the radioactive indices of the collected samples like hazards indices and excess life time risk shows admissible limit recommended by UNCEAR 2000.

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Table 3. Estimation of hazard indices and excess lifetime cancer risk

No.	Name	External gamma dose Bqkg^{-1}	External hazard index	Absorbed dose Bqkg^{-1}	Activity concentration index	Annuel effective dose µ svy^{-1}	Excess life time cancer risk
1	Blackoxide A	33.75	0.21	36.54	0.28	44.8	1.57 ×10^{-4}
2	Blackoxide B	18.56	0.11	18.76	0.14	23.0	8.06×10^{-5}
3	Granite A	50.40	0.32	54.67	0.41	67.1	2.35×10^{-4}
4	Granite B	45.85	0.29	49.61	0.38	60.9	2.13×10^{-4}
5	Red Oxide A	24.02	0.14	23.81	0.18	29.2	1.02×10^{-4}
6	Red OxideB	25.72	0.15	25.38	0.20	31.1	1.09×10^{-4}
7	Choolakkata	142.57	0.88	149.10	1.14	183.0	6.4×10^{-4}
8	Kambikatta	83.95	0.51	85.34	0.65	104.7	3.67×10^{-4}
9	Dalmia Cement A	30.89	0.19	31.89	0.24	39.1	1.37×10^{-4}
10	Dalmia Cement B	27.98	0.17	29.63	0.23	36.4	1.27×10^{-4}
11	White Oxide	24.13	0.15	25.59	0.19	31.4	1.1×10^{-4}
12	M Sand A	21.61	0.13	22.45	0.17	27.5	9.64×10^{-5}
13	Rock powder	15.37	0.09	15.81	0.12	19.4	6.79×10^{-5}
14	Sand A	62.77	0.39	66.28	0.50	81.3	2.85×10^{-4}
15	Sand B	61.70	0.38	64.57	0.49	79.2	2.77×10^{-4}
16	Yellow Oxide	30.51	0.19	32.99	0.25	40.5	1. 42 ×10^{-4}
17	Joint filller	21.67	0.14	23.30	0.18	28.6	1×10^{-4}
18	Sanker Cement A	47.40	0.29	47.56	0.37	58.4	2.04×10^{-4}
19	Sanker Cement B	50.15	0.31	51.02	0.39	62.6	2.19×10^{-4}

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