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Natural radioactivity in building materials in Iran

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Abstract. This work presents a comprehensive study of natural radioactivity in building materials used in Iran. For this purpose, 177 samples of five types of building material, i.e. cement, gypsum, cement blocks, gravel and brick, were gathered from different regions of the country and analyzed by gamma spectroscopy to quantify radioactivity concentrations using a high purity germanium (HPGe) detector and a spectroscopy system. According to the results of this investigation, cement samples had maximum values of the mean Ra-226 and Th-232 concentrations, 39.6 and 28.9 Bq/kg, respectively, while the lowest value for mean concentration of these two radionuclides were found in gypsum samples 8.1 and 2.2 Bq/kg, respectively. The highest (851.4 Bq/kg) and lowest (116.2 Bq/kg) value of K-40 mean concentration were found in brick and gypsum samples, respectively. The absorbed dose rate and the annual effective dose were also calculated from the radioactivity content of the radionuclides. The results show that the maximum values of dose rate and annual effective dose equivalent were 53.72 nGy/h and 0.37 mSv/y in brick samples. The radium equivalent activities R_{eq} calculated were below the permissible level of 370 Bq/kg for all building materials. The values of hazard indexes were below the recommended levels, therefore, it is concluded that the buildings constructed from such materials are safe for the inhabitants. The results of this study are consistent with the results of other investigations in different parts of the world.

Key words: gamma spectroscopy • absorbed dose rate • annual effective dose • hazard index

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Introduction

All building materials are mostly composed of rock and soil and these two raw materials contain natural radioactive isotopes such as Th-232 and U-238 decay series and K-40 [12].

Determination of population exposure to radiation from building materials is of great importance, since people spend about 80% of their life inside the buildings. The activity concentration of natural radionuclides in building materials has been estimated in different countries and regions of the world such as Australia [7], Bangladesh [2], Pakistan [13, 18], Tanzania [24], Eastern Europe [20], Syria [26], Kuwait [8], China [34], Egypt [1, 4, 9, 16], and Cyprus [23]. Ra-226 is the most important radionuclide in the U-238 decay chain from radiobiological viewpoint, therefore, the measurements of Ra-226 concentration in building materials are considered as reference in all investigations. Natural radionuclides in building materials may cause both external exposure caused by their direct gamma radiation and also internal exposure from radon gas.

In this study, the activity concentration of natural radionuclides in five important types of material, which are mostly used in buildings of Iran, were measured using high resolution gamma spectroscopy. The amount of dose rates and hazard indexes were also calculated and finally a comparison between these values and the world median levels are presented.

Materials and methods

Sampling

A total of 177 samples of five common building materials in Iran, cement, gypsum, cement blocks, brick, and gravel, were gathered from different parts of the country. Thus the sampling regions were chosen according to their population and the amount of building material production in each region. Fourteen provinces of the country (containing about 64% of the population of Iran) were chosen for this investigation, i.e. Tehran, Alborz, Ghom, Ghazvin, North Khorasan, Central Khorasan, south Khorasan, eastern Azarbaijan, Isfahan, Fars, Mazandaran, Golestan, Gilan and Yazd. It should be noted that the raw materials of high natural background region of Ramsar are not used in the production of common building materials applied in constructing buildings in Iran, and the habitants of this region mostly use building materials from other parts of Iran. As this high background region is local, building materials of this small village are not included in this study.

The building material samples were directly taken from the manufactories in these provinces. First, the samples were crushed and milled to powder, and dried by heating to 100°C for 24 h to remove the moisture. Then, after dissolution, 300 ml of each sample were moved to a sealed cylindrical vial and kept for four weeks to reach seqular equilibrium between Ra-226 and its decay products.

Radioactivity measurement

The concentrations of the natural radionuclides such as Th-232, Ra-226, U-238, were measured using gamma spectroscopy with a p-type HPGe detector with a 20% relative efficiency and 2 keV resolution for the 1332 keV photons of Co-60. The activity concentrations of Cs-137 and K-40 were measured using the gamma lines of 661 and 1460 keV, respectively.

The gamma line 609 keV of Bi-214 was used as an indicator of Ra-226, and the activity concentration of Th-232 was measured using the gamma line 583 keV of Tl-208.

The values of minimum detectable activity (MDA) of the detection system for Cs-137, Th-232, Ra-226 and K-40 were 1, 3.40, 3.60 and 38 Bq/kg, respectively. The whole detecting system was shielded to reduce the background radiation. The calibrations of energy and efficiency of the spectrometer were performed using calibration sources with a known activity of the National Radiation Protection Department of the Atomic Energy Organization of Iran.

Absorbed dose rate measurement

The absorbed dose rate (D) in air at a height of 1.0 m above the ground from the radionuclides K-40 and also Th-232 and the U-238 decay series, were calculated using Eq. (1), if the naturally occurring radionuclides are uniformly distributed [5, 6, 22].

$$(1) \quad D = 0.52813C_{\mathrm{Th}} + 0.38919C_{\mathrm{Ra}} + 0.03861C_{\mathrm{K}}$$

where: C_{Ra}, C_{Th}, and C_{K} are the activity concentration of Ra-226, Th-232, and K-40 in (Bq/kg), respectively.

Annual effective dose rates

Values of the absorbed dose rate were used for the calculation of annual outdoor effective dose rate considering some correction factors. A conversion factor (CF = 0.7 Sv/Gy) was applied for conversion of the absorbed dose in air to the corresponding effective dose. The outdoor occupancy factor (OF = 0.8) was taken into account, since the people spend most of their time (80%) in buildings. Finally, the ratio of indoor to outdoor gamma dose rates (R = 1.4) was also applied as used by other investigators [30] to calculate the annual indoor effective gamma dose rate (D_E) in (Sv/y) as given follow:

$$(2) \quad D_E = D \times \mathrm{CF} \times \mathrm{OF} \times R \times t$$

where: D is the dose rate in (Gy/h) and t is the duration of the exposure (= 365 × 24 h).

Radium equivalent activity

The non-uniformity of natural nuclides concentration in building material samples would cause in definition of an index the so-called radium equivalent activity (R_{eq}), [7, 25, 31] for taking into account the radiation hazards of radioactive nuclides. This quantity is calculated as given below using the activity concentration of the radionuclides [32].

$$(3) \quad R_{\mathrm{eq}} = C_{\mathrm{Ra}} + 1.43C_{\mathrm{Th}} + 0.077C_{\mathrm{K}}$$

where: C_{Ra}, C_{Th}, and C_{K} are the activity concentration of Ra-226, Th-232, and K-40 in (Bq/kg), respectively.

This index (R_{eq}) is related to both internal dose due to radon and external gamma dose [10], and should have the highest value of 370 Bq/kg for safe use of the building materials.

Hazard indexes

Indexes for external gamma radiation

Two indexes are used in this paper for assessment of excess gamma radiation from the building materials in order to be sure of the safety of building material usage.

Beretka and Mathew introduced a hazard index for the external gamma radiation dose from building materials as given bellow [7].

$$(4) \quad H_{ex} = \frac{C_{\mathrm{Ra}}}{370} + \frac{C_{\mathrm{Th}}}{259} + \frac{C_{\mathrm{K}}}{4810} \le 1$$

The value of H_{ex} should be below one to be sure of the safe use of building materials, which corresponds to the upper limit of R_{eq} (370 Bq/kg).

The European Commission (EC) proposed an index called gamma index (I_γ) to verify whether the guidelines of EC for building material usage are met. I_γ is calculated using the following formula [11]:

$$I_\gamma = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \leq 1 \qquad (5)$$

EC introduces two dose criteria for the gamma dose of building materials, an exemption criterion of 0.3 mSv/y, and an upper limit of 1 mSv/y. Most of the countries apply their control on the upper limit (1 mSv/y). If the exemption level of 0.3 mSv/y is considered, the values of I_γ should be below 0.5 for materials used in bulk (i.e. brick and cement), but if the upper level of 1 mSv/y is considered, the values of I_γ should be below 1 for such materials. For superficial building materials with restricted use (i.e. tiles, boards), I_γ should be below 2 and 6, supposing the control value of 0.3 and 1 mSv/y, respectively.

Hazard indexes for alpha radiation

Some indexes have been proposed for assessment of excess α radiation due to radon gas from the building materials. In this study, we have assessed two indexes called internal hazard index (H_{in}) and alpha index (I_α).

H_{in} can be used for considering the excess internal radiation due to inhalation of Rn-222 and its short-lived decay products from building materials, which is defined as [10].

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \qquad (6)$$

This quantity should be less than unity for the safe use of materials in construction of buildings.

The I_α has been proposed by Krieger [19] and Stoulos *et al*. [29] as given below

$$I_\alpha = \frac{C_{Ra}}{200} \qquad (7)$$

The recommended values of I_α, like I_γ are below (0.5, and 1) [11].

Results and discussions

The concentration of natural radionuclides

The activity concentrations of Ra-226, Th-232 and K-40 have been measured in five types of building materials in Iran. Table 1, presents the ranges and mean values of activity of each radionuclide. As it is seen from table the highest mean value of Ra-226 and Th-232 were 39.6 and 28.8 Bq/kg in cement samples, while the brick samples were found to have the highest mean value of K-40 concentration, 851.4 Bq/kg. According to the results of Table 1, the maximum concentrations were found for K-40 in all types of building materials. The lowest mean values of K-40, Th-232 and Ra-226 were in gypsum samples 8.51, 2.22, and 116.2 Bq/kg.

Table 1. Range and mean value of natural radioactivity in building materials in Iran

Building material	Number of samples	C_{Ra} (Bq/kg)		C_{Th} (Bq/kg)		C_K (Bq/kg)	
		Mean	Range	Mean	Range	Mean	Range
Brick	77	37.0 ± 1.5	(18.9 ± 0.9)–(56.2 ± 1.9)	12.2 ± 0.7	(8.1 ± 0.5)–(20.4 ± 1.1)	851 ± 15	(511.0 ± 11)–(1800 ± 32)
Cement block	21	20.7 ± 1.1	(10.7 ± 0.8)–(36.3 ± 1.2)	3.0 ± 0.4	(1.5 ± 0.2)–(12.6 ± 0.8)	436 ± 14	(146.0 ± 5.5)–(1140 ± 19)
Gravel	32	20.4 ± 1.2	(8.1 ± 0.4)–(32.2 ± 1.4)	6.3 ± 0.3	(0.7 ± 0.1)–(13.7 ± 0.9)	451 ± 14	(83.3 ± 9.1)–(1235 ± 21)
Gypsum	30	8.1 ± 0.1	(1.9 ± 0.1)–(42.6 ± 0.9)	2.2 ± 0.1	(0.4 ± 0.1)–(4.1 ± 0.2)	116 ± 11	(37.7 ± 4.2)–(437 ± 12)
Cement	17	39.6 ± 1.4	(7.4 ± 0.4)–(78.4 ± 1.1)	28.9 ± 0.9	(10.0 ± 0.6)–(107 ± 3.1)	291 ± 12	(263.1 ± 11.1)–(614 ± 16)

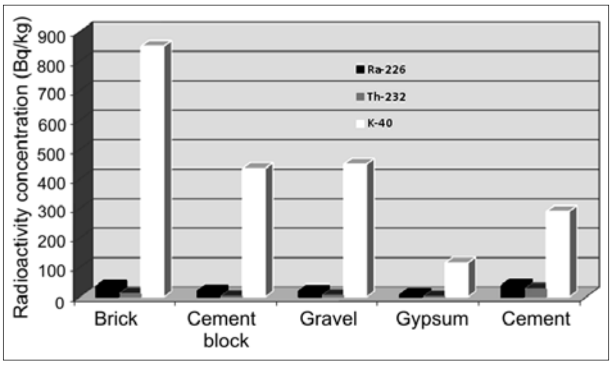

Fig. 1. Mean values of natural radionuclide concentrations in five type of building materials in Iran.

Figure 1, compares the mean values of natural radionuclides in different types of building materials. A comparison between the mean values of radioactive materials in Iran and the values in other countries is shown in Table 2. As it is observed from table, the ranges of mean values of natural radionucides concentration in building materials differ from one country to another depending on the soil and raw materials used for their formation.

Absorbed gamma dose rates (D) and annual effective dose rates (D_E)

Mean value of gamma dose rate in air at the distance of 1 m from the ground were estimated using Eq. (1) for different kinds of building materials. As can be seen from Table 3, the maximum gamma dose rate was 53.72 nGy/h in brick, while the minimum value was found for gypsum – about 8.80 nGy/h.

The annual outdoor effective dose of each building material, as obtained by Eq. (2) are also shown in Table 3. The values of D_E varied from 0.08 for gypsum to 0.37 for brick.

Radium equivalent activity

The estimated mean values of radium equivalent activities for all types of building materials are summarized in Table 3. The highest mean value of R_{eq} is estimated in brick samples, 120.00 Bq/kg which is significantly less than the upper limit of 370 Bq/kg.

Table 2. A comparison between radioactivity content of building materials in Iran and other parts of the world

Building material	Radioactivity concentration (Bq/kg)			Country/Reference
	Ra-226	Th-232	K-40	
Cement	39.6 ± 1.4	28.9 ± 0.9	290.8 ± 12	Iran (this study)
	26.1	28.6	272.9	Pakistan [18]
	41.0	27.0	422.0	Brazil [22]
	20.0	13.0	241.0	Grece [29]
	37.0	24.1	432.2	India [21]
	31.1	12.4	121.0	Iran [14]
Gypsum	8.1 ± 0.9	2.2 ± 0.2	116.2 ± 12	Iran (this study)
	9.4	3.9	40.7	Turkey [31]
	14.0	2.0	31.0	Syria [26]
	6.2	13.3	173.7	Pakistan [18]
	13.3	5.3	39.5	Bulgaria [20]
Gravel	20.4 ± 1.2	6.3 ± 0.3	450.7 ± 14	Iran (this study)
	13.9	14.8	171	Australia [28]
	10.3	Not detected	933	Brazil [22]
	33.3	33.3	14.8	USA [17]
Brick	37.0 ± 1.5	12.2 ± 0.7	851.4 ± 15	Iran (this study)
	65	51	675	Algeria [3]
	12	7	332	Kuwait [8]
	35	45	710	Greece [27]
	23	35	431	Pakistan [13]
	35	72	585	Sri Lanka [15]
	59	50	714	China [33]

Table 3. Mean values of gamma dose rate, annual effective dose rate and hazard indexes for building materials

Building material	D_γ (nGy/h)	D_E (mSv/y)	R_{eq}	H_{ex}	H_{in}	I_α	I_γ
Brick	53.72	0.37	120.00	0.32	0.42	0.19	0.47
Cement block	26.48	0.18	58.58	0.16	0.21	0.10	0.23
Gravel	28.67	0.20	64.11	0.17	0.23	0.10	0.25
Gypsum	8.80	0.06	20.19	0.05	0.08	0.04	0.077
Cement	41.90	0.29	103.32	0.28	0.39	0.20	0.37

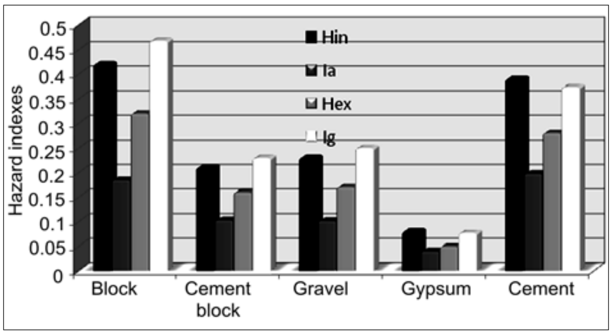

Fig. 2. A comparison between internal and external gamma indexes.

Hazard indexes

The values of external and internal hazards (H_{ex} and H_{in}) along with the alpha and gamma indexes (I_γ and I_α) are also shown in Table 3. As can be seen, the maximum value of H_{ex} and H_{in} are found in brick samples, 0.32 and 0.42, respectively. The highest values of I_γ and I_α are 0.2 and 0.47 were found in cement and brick, respectively.

A comparison between the four indexes is shown in Fig. 2. As it is observed from the figure, all the indexes are below the recommended levels. The values of H_{ex} and H_{in} are all bellow unity, and the values of I_γ and I_α are below the recommended levels of 0.5 and 1. Therefore, the materials included in this study can be safely used for construction of buildings.

Conclusion

The gamma spectroscopy method was used for assessment of the U-238 and Th-232 series and K-40 concentration in common building materials of Iran. The lowest mean value of the three radionuclides were found in gypsum, while the brick samples contained the highest values of K-40, and cement values contained the maximum value of Ra-226 and Th-232. The values of dose rate at a distance of 1 m in air (D) and annual effective outdoor dose equivalents (D_E) were calculated using the activity concentration of radionuclides. The maximum values of dose rate (53.72 nGy/h) and annual effective dose equivalent (0.72 mSv/h) were calculated in brick samples. The radium equivalent activities obtained using the concentration of these nuclides, were below the allowable level of 370 Bq/kg in all five types of building materials. The calculated values of H_{ex} and H_{in} corresponding to the activity concentration of natural radioisotopes were lower than unity, and the values of (I_γ and I_α) were also below the recommended values. Therefore, the use of these materials in the construction of buildings is safe for people.

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