Natural Radioactivity in Commonly Building Materials Used in Vietnam - 11255

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ABSTRACT

Naturally occurring radionuclides in building materials contribute to external and internal exposure and that is necessary to be investigated. In the research, 213 samples of 11 different kinds of commonly used structural and covering building materials were collected from housing and other building construction sites and from suppliers in Vietnam to measure the natural radioactivity of Ra-226, Th-232 and K-40. The measurements were carried out using low background gamma-ray spectrometry with HPGe detector. The specific activities of the different building materials varied from 0.18 - 395.28, 0.10 - 266.52 and 0.76 - 2006.78 Bq/kg with the average values of 52.09, 55.70 and 593.5 Bq/kg for Ra-226, Th-232 and K-40, respectively. The obtained data were compared with the corresponding reported data of other countries.

The activity concentration index and the annual effective dose were evaluated to assess the potential radiological hazard associated with these building materials. The results showed that the enhanced concentration values were sometimes felling into granite tiles, especially imported granite tiles. These obtained data aimed to enlarge the database on natural radioactivity in commonly building materials used in Vietnam and to support technical aspects in hazard exposure reduction.

Keywords: Natural radioactivity; building materials; activity concentration index.

INTRODUCTION

All materials contain various amounts of natural radionuclides. Materials derived from rocks and soils contain mainly natural radionuclides of the U and Th series, and K-40. In the uranium series, the decay chain segment starting from radium is radiologically the most important and, therefore, reference is often made to radium instead of uranium. The world-wide average concentrations of radium, thorium and potassium in the earth's crust are about 40Bq/kg, 40Bq/kg and 400Bq/kg, respectively (UNSCEAR, 2000).

Naturally occurring radioactive materials (NORMs) are the major sources which cause exposure to people by ionizing radiation of about 2.3mSv/year in the average (UNSCEAR, 1993). These radionuclides pose exposure risks externally due to their gamma-ray emissions and internally due to radon and its progeny which emit alpha particles with the contributions of 46% and 54%, respectively to the total of annual effective dose caused by the NORMs.

Building materials are commonly originated from rocks and soils and also contain the NORMs. In order to be able to assess radiological risk linking to standards and regulatories on natural radioactivity in building materials, it is important to study the levels of radiation emitted from them.

Results of radioactivity measurement in some imported and domestic building materials, like cement, sand, red-clay brick, gypsum, gravel aggregate, lime/limestone, glazed tile, granite, marble are presented in this paper. These particular materials were considered because they are dominant used in construction of dwellings in Vietnam. The results of the natural radionuclides activities, activity concentration indexes and annual effective doses associated with the usage of 11 kinds over 213 building material samples which collected from the whole country were obtained in the study and they were compared with the findings of other studies.

In majority of the reports, the enhanced concentration values were sometimes fell into granite tiles, especially imported granite tiles. The activity concentration indexes of some samples are high up to 2.63. The external dose rate and annual effective doses that are due to natural radionuclides and their progenies, could achieve up to several hundreds of nGy/h and several of mSv, respectively, in the conservative case of all structures in a building.

The obtained data in this research aimed to enlarge the database on natural radioactivity in commonly building materials used in Vietnam and to support technical bases in hazard exposure reduction.

MATERIALS AND METHODS

Sample collection, preparation and counting

The building materials investigated are cement, sand, red-clay brick, gypsum, gravel aggregate, lime/limestone, glazed tile, granite, marble which are the dominant building materials in Vietnam with about 5-10kg of weight per each sample.

The sand samples originating from major rivers of Vietnam were collected from restores of local building materials companies (30 samples). The red-clay brick samples originating from local clay mines of different provinces over the whole country were collected from brick manufactures (23 samples). The lime/limestone and gravel aggregate samples were collected from major mines being exploited (11 lime/limestone samples, 40 gravel aggregate samples). The local & imported granite, marble, and glazed tile samples were purchased from different dominant trading brands (10 of imported granites, 9 of imported marbles, 25 of local granites, 5 of local marbles, and 35 glazed tile samples). The cement and gypsum samples were collected from some high capacity cement plants (17 cement samples, 8 gypsum samples).

About 3kg of each sample with large grain size was ground to a fine powder, except for cement, gypsum & sand samples; and then was homogenized and air dried. An approximation of 140g fine powder drawn out from each sample was mixed carefully with ~70g polyester resin and then was cast by a cylindrical steel container (40mm height and 60mm diameter) in order to seal in gas tight, impermeable radon. The technique aims to approach that all of the samples get approximately a matrix density (1.224 ± 0.122 g/cm^3) and the same measuring configuration. This measuring configuration with 40mm in height and 60mm in diameter for building materials matrix has been investigated on the estimation of self-attenuation corrections over the depth of materials matrix, and it is supposed that one is the most suitable to be used in this research. The samples after casting were then stored for 25–30 days before counting in order to reach radioactive equilibrium.

The radioactivity of the samples was measured on gamma spectrometry with the HpGe detector model GX3019, which has a relative efficiency of 30%, an energy resolution of 1.90keV at 1333keV and a peak-to-Compton ratio of 56:1. Computer software MAESTRO-32 was used to analyze obtained spectrums.

Detector calibration was done by soil reference materials (IAEA-CU-2006-03 containing Mn-54, Co-60, Zn-65, Cd-109, Cs-134, Cs-137, Pb-210, Am-241 radionuclides). The counting time for each measurement was around 90000s, in order to obtain good counting statistics.

To measure Ra-226, peaks from Pb-214 at energies of 295.2keV (18.53%) & 351.9keV (35.85%); and peak from Bi-214 at energy of 1764.5keV (15.36%) were used. To measure Th-232, peaks from Ac-228 at energies of 911.2keV (26.61%) and 338.3keV (11.25%) were used; and it was checked with peaks from Tl-208 at energies of 583.2keV (84.55%) and 2614.5keV (99.16%). Activity of K-40 was measured from its intensive line at 1460.8keV (10.67%).

Activity concentration index, I

Activity concentration index, or shortly, gamma index, *I* is defined in the following way:

$$I = \frac{C_{Ra}}{300\,Bq.kg^{-1}} + \frac{C_{Th}}{200\,Bq.kg^{-1}} + \frac{C_K}{3000\,Bq.kg^{-1}} \qquad [1]$$

Where C_{Ra}, C_{Th} *and* C_K are the radium, thorium and potassium activity concentrations (Bq/kg) in the building material, respectively. The activity concentration index is derived to identify whether a dose criterion is met. The gamma index should not exceed limit values depending on the dose criterion (EC 1999) and the amount material used in a building.

Investigation levels can be derived for practical monitoring purposes. Because more than one radionuclide contribute to the dose, it is practical to present investigation levels in the form of a gamma index, *I*. The gamma index also takes into account typical ways and quantities in which the material is used in a building. A methodology which can be used to derive such indexes is described in Annex I of Report 112 (EC 1999).

The gamma index should be used only as a screening tool for identifying materials which might be of concern. Any actual decision on restricting the use of a material should be based on a separate dose assessment. Such assessment should be based on scenarios where the material is used in a typical way for the type of material in question. Scenarios resulting in theoretical, most unlikely maximum doses should be avoided.

Dose rate and annual effective dose

The absorbed dose rates in air in a room can be calculated by using the specific dose rates given in (EC, 1999). The specific dose rates (in unit's nGy h^{-1} per Bq kg^{-1}*)* for Ra-226, Th-232 and K-40 are given for different screening tool of identifying materials which might be of concern. Indoor dose rates for a model room (the dimensions of 4m × 5m × 2.8m, thickness of 20cm, density of 2.35g/cm^3, and the background of 50nGy/h) are calculated with different structures in a building causing the irradiation as follows:

- All structures: $D^{\cdot} = 0.92\times C_{Ra} + 1.1\times C_{Th} + 0.08\times C_K$ [2]

- Floor and walls: $D^{\cdot} = 0.67\times C_{Ra} + 0.78\times C_{Th} + 0.057\times C_K$ [3]

- Floor only: $D^{\cdot} = 0.24\times C_{Ra} + 0.28\times C_{Th} + 0.02\times C_K$ [4]

- Superficial material, tile/stone on all walls: $D^{\cdot} = 0.12\times C_{Ra} + 0.14\times C_{Th} + 0.0096\times C_K$ [5]

The annual effective dose, D_E (mSv), due to gamma radiation from building materials with the annual exposure time of 7000h (EC, 1999) was calculated as follows:

$D_E = 0.7\ (Sv.Gy^{-1}) \times 7000\ (h) \times 10^{-6} \times D^{\cdot}\ (nGy.h^{-1})$, [6]

RESULTS AND DISCUSSION

The mean and range of specific radioactivities of Ra-226, Th-232 and K-40 in 11 kinds of building material samples are shown in Table I and Fig.1. The corresponding data over all of 213 samples are 52.09, 55.70 and 593.5 Bq/kg in the averages and varied in the ranges of 0.18 - 395.28, 0.10 - 266.52 and 0.76 - 2006.78 Bq/kg for Ra-226, Th-232 and K-40, respectively. The obtained results show that the average specific radioactivities in investigated building materials are higher compared with the world-wide average concentrations of radium, thorium and potassium in the earth's crust about of 1.30, 1.39, 1.48 times, respectively. The specific radioactivities enhance fell into granite tiles, especially imported granite tiles.

The activity concentration index, *I*, was calculated as the formula [1] which was proposed by Ministry of Construction of Socialist Republic of Vietnam's Regulatory: TCXDVN 397:2007 (adopted by the European Commission 1999). The results are shown in Tables II and Fig.2. In ascending, the average gamma indexes of 11 kinds of building materials are of the sequence: local & imported marble, gypsum, lime/limestone, cement, sand, gravel aggregate, red-clay brick, glazed tile, local granite, and the highest belonging to imported granite, which is exceeds the dose criterion of 1mSv. It was found that values of *I* being larger than unit fell into: 2 sand samples; 8 gravel aggregate samples; 3 red-clay brick samples; 12 glazed tile samples; 14 local granites; and 5 imported granites. In particularly, the *I* values of some imported granites are significant to be considered in the assessment of radiological risk linking to standards and regulatories on natural radioactivity in building materials. In several recent papers, gamma index *I* was calculated also and reported values of *I* ranged from 0.1 to 0.98 in building materials used in bulk, and from 0.01 to 2.92 in materials used in restricted quantities (Righi and Bruzzi, 2006).

The mean & range of dose rates and annual effective doses depending on the structures in a building causing the irradiation in 11 kinds of building material samples are also shown in Table II. The obtained data shown that there are many building materials with annual effective doses exceeding the criterion of 1mSv in the conservative case of all structures in a building, for an example, imported granites approaches up to 3.24mSv. To reduce the hazard exposure, therefore, it is necessary to consider to using restrictively the enhanced NORMs in building construction. In this situation, for some materials where the value of *I* exceeds the recommended value, the annual effective dose can be below the upper limit. Obviously, if the above highest imported granite is used with structure of superficial material only on all walls, the annual effective dose could reduce down to 0.41mSv.

Table I. Mean and Range of The Specific Radioactivities of Ra-226, Th-232 and K-40 in Building Materials Used in Vietnam.

No.	Building materials	Abb.	No. of samples	Specific radioactivities, Bq/kg		
				Ra-226	Th-232	K-40
				Mean ± Std. Range	Mean ± Std. Range	Mean ± Std. Range
1	Sand	SA	30	26.74 ± 30.30 2.10 ÷ 162.46	39.77 ± 61.70 1.79 ÷ 266.52	506.9 ± 317.1 2.5 ÷ 1096.8
2	Gravel aggregate	GA	40	47.18 ± 53.19 4.59 ÷ 224.93	51.53 ± 50.48 0.10 ÷ 196.93	720.6 ± 473.0 0.8 ÷ 1440.3

3	Imported granite	IG	10	79.15 ± 133.49 1.82 ÷ 395.28	86.19 ± 67.21 10.56 ÷ 197.26	1126.0 ± 584.8 164.1 ÷ 2006.6
4	Imported marble	IM	9	11.29 ± 14.20 0.67 ÷ 40.23	0.76 ± 0.51 0.18 ÷ 1.45	5.1 ± 5.9 1.2 ÷ 11.9
5	Local granite	LG	25	59.48 ± 43.40 0.77 ÷ 162.48	85.17 ± 55.89 0.18 ÷ 190.67	992.0 ± 443.0 162.1 ÷ 1593.0
6	Local marble	LM	5	12.37 ± 12.97 0.72 ÷ 26.40	2.30 ± 0.97 1.33 ÷ 3.64	6.6 ± 4.8 2.3 ÷ 12.5
7	Glazed tile	GT	35	95.14 ± 47.27 15.26 ÷ 270.85	74.64 ± 28.14 0.50 ÷ 147.57	658.7 ± 273.9 0.8 ÷ 1409.1
8	Lime stone	LI	11	14.32 ± 14.39 4.46 ÷ 55.64	8.79 ± 19.25 0.96 ÷ 60.04	39.0 ± 59.6 4.2 ÷ 211.5
9	Clay brick	BR	23	64.35 ± 23.78 31.34 ÷ 113.15	77.57 ± 32.49 33.29 ÷ 169.81	589.0 ± 171.8 329.0 ÷ 868.8
10	Gypsum	GY	8	8.80 ± 9.52 0.18 ÷ 28.80	6.46 ± 4.87 0.39 ÷ 16.24	74.6 ± 72.8 1.9 ÷ 195.2
11	Cement	CE	17	39.86 ± 17.43 23.54 ÷ 67.46	25.46 ± 4.69 16.27 ÷ 32.22	243.5 ± 62.2 83.7 ÷ 308.9

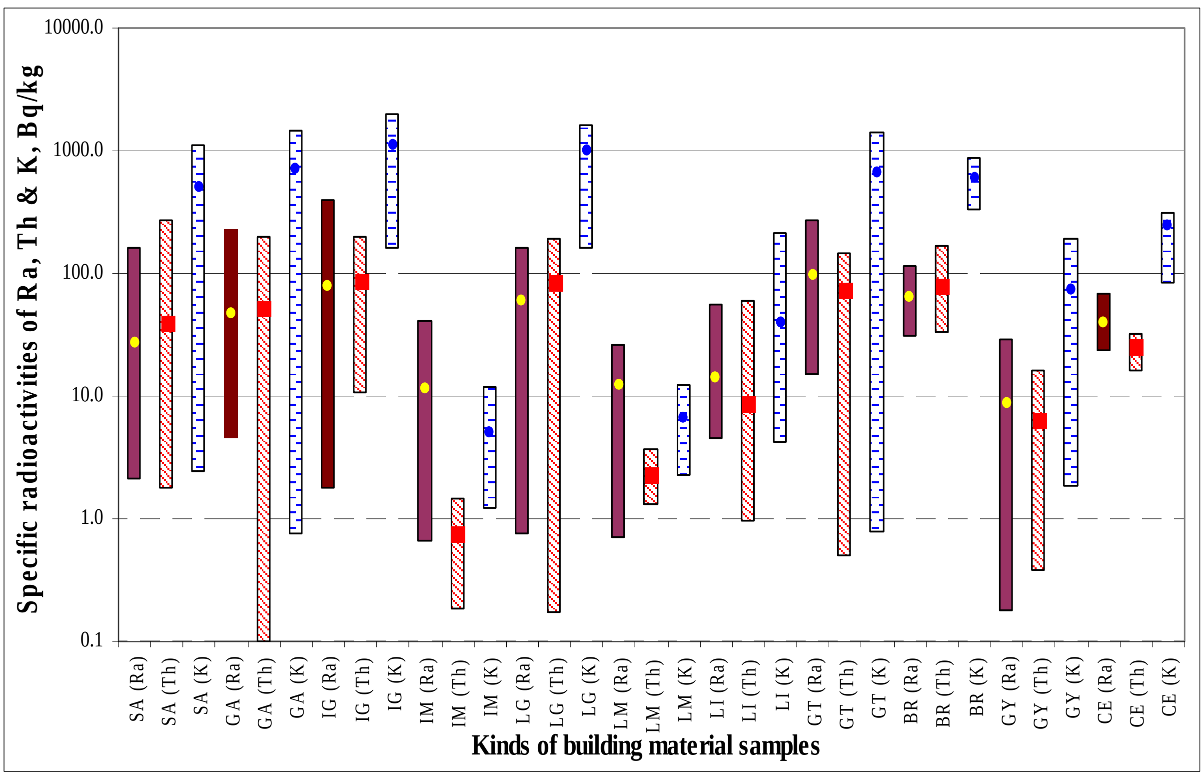

Fig.1. Mean and range of Ra-226, Th-232 and K-40 radioactivities in 11 kinds of building materials over 213 samples used in Vietnam.

Table II. Mean and Range of Activity Concentration Indexes, Dose Rates and Annual Effective Doses in Building Materials Used in Vietnam.

No.	**Building materials**	**Abb.**	**I**	**Structures in a building causing the irradiation**	$\dot{D}$ **, nSv.h^{-1}**	**D_E, mSv**
			Mean Range		**Mean Range**	**Mean Range**
1	Sand	SA	0.45 0.02 ÷ **1.99**	All structures	107.6 5.6 ÷ 472.6	0.53 0.03 ÷ **2.32**
2	Gravel aggregate	GA	0.64 0.02 ÷ 2.18		154.9 4.3 ÷ 530.6	0.76 0.02 ÷ **2.60**
3	Imported granite	IG	**1.07** 0.14 ÷ **2.63**	All structures	257.7 33.0 ÷ 661.4	**1.26** 0.16 ÷ **3.24**
				Floor and walls	184.4 23.6 ÷ 476.3	0.90 0.12 ÷ **2.33**
				Floor only	65.6 8.4 ÷ 170.2	0.32 0.04 ÷ 0.83
				Superficial material: material on all walls	32.4 4.1 ÷ 84.5	0.16 0.02 ÷ 0.41
4	Imported marble	IM	0.04 0.00 ÷ 0.13	All structures	9.8 0.0 ÷ 37.0	0.05 0.00 ÷ 0.18
				Floor and walls	7.1 0.0 ÷ 27.0	0.04 0.00 ÷ 0.13
				Floor only	2.6 0.0 ÷ 9.7	0.01 0.00 ÷ 0.05
				Superficial material: material on all walls	1.3 0.0 ÷ 4.8	0.01 0.00 ÷ 0.02
5	Local granite	LG	0.94 0.00 ÷ **1.73**	All structures	224.6 0.9 ÷ 409.0	**1.10** 0.00 ÷ **2.00**
				Floor and walls	160.6 0.7 ÷ 293.2	0.79 0.00 ÷ **1.44**
				Floor only	57.2 0.2 ÷ 104.6	0.28 0.00 ÷ 0.51
				Superficial material: material on all walls	28.2 0.1 ÷ 51.8	0.14 0.00 ÷ 0.25
6	Local marble	LM	0.04 0.01 ÷ 0.10	All structures	11.6 1.8 ÷ 27.3	0.06 0.01 ÷ 0.13
				Floor and walls	8.4 1.3 ÷ 19.8	0.04 0.01 ÷ 0.10
				Floor only	3.0 0.5 ÷ 7.1	0.01 0.00 ÷ 0.03
				Superficial material: material on all walls	1.5 0.2 ÷ 3.5	0.01 0.00 ÷ 0.02

7	Glazed tile	GT	0.91 0.05 ÷ **1.57**	All structures	222.3 14.7 ÷ 404.4	**1.09** 0.07 ÷ **1.98**
				Floor and walls	159.5 10.7 ÷ 291.9	0.78 0.05 ÷ **1.43**
				Floor only	56.9 3.8 ÷ 104.0	0.28 0.02 ÷ 0.51
				Superficial material: material on all walls	28.2 1.9 ÷ 51.4	0.14 0.01 ÷ 0.25
8	Lime stone	LI	0.10 0.03 ÷ 0.56	All structures	24.2 7.8 ÷ 134.2	0.12 0.04 ÷ 0.66
9	Clay brick	BR	0.80 0.41 ÷ **1.46**		191.7 100.0 ÷ 347.3	0.94 0.49 ÷ **1.70**
10	Gypsum	GY	0.09 0.00 ÷ 0.22		20.2 1.1 ÷ 48.6	0.10 0.01 ÷ 0.24
11	Cement	CE	0.34 0.20 ÷ 0.46		84.2 49.6 ÷ 114.5	0.41 0.24 ÷ 0.56

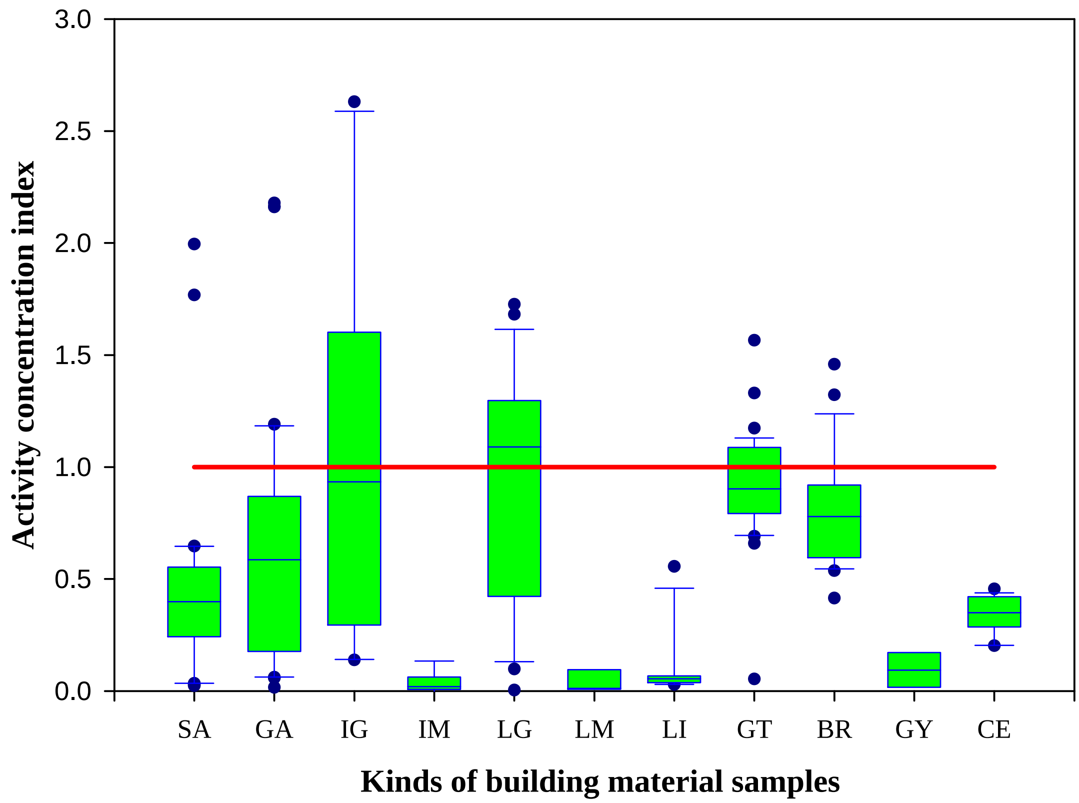

Fig. 2. Box-and-Whisker plot of activity concentration index calculated from specific radioactivities of 11 kinds of building materials over 213 samples used in Vietnam.

CONCLUSION

The natural radionuclide contents, activity concentration indexes, dose rates and annual effective doses from 11 different kinds' imported and domestic building materials over 213 samples used in Vietnam were determined and explained.

The results showed that the enhanced concentration values were sometimes felling into granite tiles, especially imported granite tiles. The gamma indexes and annual effective doses determined show that some of samples exceeded the values recommended by TCXDVN: 397:2007. However, the radiation exposure and attributed risk could be reduced by careful choice of building materials during construction.

The obtained data in this research aimed to enlarge the database on natural radioactivity in commonly building materials used in Vietnam and to support technical aspects in hazard exposure reduction.

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