CONTRIBUTION OF SELANGOR AND JOHOR CLAY BRICKS TOWARDS THE NATURAL RADIOACTIVITY EXPOSURE TO DWELLER

Na'im Syauqi Hamzah*, Redzuwan Yahaya, Amran Ab. Majid, Muhammad Samudi Yasir, Ismail Bahari

Nuclear Science Programme, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor Darul Ehsan
e-mail: qi_luffy@yahoo.com

ABSTRACT

At present, soil and mineral based building material such as bricks are one of the main components in building construction in Malaysia. This building material is a direct source of radiation exposure since it contains naturally occurring radioactive materials (NORM). In this study, clay brick samples used were obtained from 7 factories in Selangor and Johore, Malaysia. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in these samples of clay bricks were determined using a comparative method and was analysed using gamma spectrometry with HPGe detector. The mean values of activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K were found to be in the range of 39.04 ± 0.88 $Bqkg^{-1}$ - 73.61 ± 5.32 $Bqkg^{-1}$, 43.38 ± 2.60 $Bqkg^{-1}$ - 73.45 ± 1.51 $Bqkg^{-1}$, and 381.54 ± 11.39 $Bqkg^{-1}$ - 699.63 ± 15.82 $Bqkg^{-1}$, respectively. The radiation hazard of NORM in the samples was estimated by calculating the radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}) and internal hazard index (H_{in}). Radium equivalent activity (Ra_{eq}) determined was in the range of 151.90 $Bqkg^{-1}$ - 194.22 $Bqkg^{-1}$ which is lower than the limit of 370 $Bqkg^{-1}$ (equivalent to 1.5 $mSvyr^{-1}$) recommended in the NEA-OECD report in 1979, whereas external hazard index (H_{ex}) and internal hazard index (H_{in}) were between 0.20 – 0.26 and 0.52 - 0.71 respectively. The annual effective dose rate exposure to a dweller received from the clay bricks was calculated to be in the range of 0.35 ± 0.18 $mSvy^{-1}$ - 0.43 ± 0.09 $mSvy^{-1}$.

ABSTRAK

Bahan binaan berasaskan tanah dan mineral seperti batu bata kini telah menjadi sebagai komponen utama pembinaan bangunan di Malaysia.Bahan binaan ini merupakan sumber langsung dedahan sinaran kerana ia mengandungi bahan radioaktif tabii (NORM). Kajian ini menggunakan batu bata tanah liat yang diperolehi dari 7 kilang di Selangor dan Johor. Kepekatan aktiviti ^{226}Ra, ^{232}Th dan ^{40}K dalam sampel batu bata tanah liat ditentukan menggunakan kaedah bandingan dan dianalisis mengggunakan spektrometri gama dan pengesan HPGe. Nilai min kepekatan aktiviti bagi masing-masing ^{226}Ra, ^{232}Th dan ^{40}K berada dalam julat 39.04 ± 0.88 $Bqkg^{-1}$ - 73.61 ± 5.32 $Bqkg^{-1}$, 43.38 ± 2.60 $Bqkg^{-1}$ - 73.45 ± 1.51 $Bqkg^{-1}$, dan 381.54 ± 11.39 $Bqkg^{-1}$ - 699.63 ± 15.82 $Bqkg^{-1}$. Hazard sinaran NORM dalam sampel dianggarkan dengan mengira aktiviti setara radium (Ra_{eq}), indeks hazard luaran (H_{ex}) dan indeks hazard dalaman (H_{in}). Aktiviti setara radium (Ra_{eq}) didapati dalam julat 151.90 $Bqkg^{-1}$ - 194.22 $Bqkg^{-1}$ yang mana lebih rendah daripada had yang disarankan dalam laporan NEA-OECD pada tahun 1979 iaitu 370 $Bqkg^{-1}$ (setara dengan 1.5 $mSvth^{-1}$), manakala bagi indeks hazard luaran (H_{ex}) dan dalaman (H_{in}) pula masing-masing didapati antara 0.20 – 0.26 dan 0.52 - 0.71. Kiraan menunjukkan bahawa kadar dos dedahan *berkesan tahunan yang diterima oleh penghuni bangunan dari batu bata tanah liat berada dalam julat 0.35 ± 0.18 mSvth^{-1}- 0.43 ± 0.09 mSvth^{-1}.*

Key words : Natural radioactivity, hazard index, clay bricks, effective dose

INTRODUCTION

In Malaysia, before the independence in 1957, wood and plant based materials were widely used as building materials particularly in rural area. At present, the scenario has changed along with the introduction of Vision 2020 to encourage national development. The choice of building material component based on soil and rock such as clay brick, cement, granite, tiles and concrete has been growing and has becoming more popular. Naturally occurring radionuclide materials (NORM) are radioactive elements that have always been present in the soil. Radiation exposure to members of the public increased appreciably by the used of building materials containing above normal levels of natural radioactivity (NEA-OECD, 1979). With the requirement, that exposure should be "as low as reasonably achievable", studies of natural radiation background has been of great importance especially in the area where soil or geological source of the building materials are obtained. Knowledge of ionising radiation levels in buildings is important in the assessment of population exposure, as the majority of individuals spend most of the time indoors (Righi & Bruzzi, 2006). The naturally occurring radionuclides in building materials can contribute to radiation exposure in two ways. Firstly, the external radiation those originate from γ-radiation of ^{238}U and ^{232}Th decay series and of ^{40}K. Secondly, is the internal α-radiation due to radon inhalation, leading to deposition of its decay products in the respiratory tract (Ngacin et. al, 2007).

In this work, activity concentration of NORM, the radium equivalent, the internal hazard index, the external hazard index**,** and the annual effective dose were assessed. The result was then compared with other studies and with the worldwide average value reported in the United Nations Scientific Committee on the Effects of Atomic Radiation report (UNSCEAR, 2000). Besides the clay bricks samples, activities in two samples of cement brick were also monitored.

STUDIES AREA

Samples of clay bricks (ClB) and cement bricks (CmB) were collected from several factories in Selangor and Johore, Malaysia to measure their natural radioactivity content. Four clay bricks samples were collected from Johore while five clay bricks samples from Selangor. For samples obtained in Johore, two samples were collected from Air Hitam (A.H. 1 and A.H. 2) and one each from Yong Peng (Y.P) and Kulai (KUL). Clay bricks samples obtained in Selangor were collected from Banting (BTN), Kepong (KPG) and Serendah (SRN1, SRN2, and SRN3). Two cement brick samples were also analysed in this work, were collected from factory in Rawang (RWG) and a hardware store in Dengkil (DNG).

MATERIALS AND METHODS

Each of bricks fabricated by the factories was cleaned, and then crushed with a hammer into a smaller pieces. In order to eliminate any water content, the samples were dried at temperature 105^{o}C in an oven until the weights are constant. Then, the samples were pulverized using a ball mill machine for around 5-10 minutes and sieved to less than 500 µm particle size. The homogenized samples were transferred to cylindrical plastic bottle (8.5 cm diameter and 7.6 cm height) and weighted. Each sample was triplicate to get the mean measurement reading. The samples were sealed and stored for at least 30 days to capture ^{222}Rn gas ($T_{1/2}$ = 3.8 days) and ensure secular equilibrium between ^{226}Ra ($T_{1/2}$ = 1.60 x 10^3 years) and the measured daughters ^{214}Bi ($T_{1/2}$ = 20 minute). HPGe detector (PCA, Tennelec) was used to determine the NORM activity in the samples. Counting time is between 12 hours to 24 hours. Activity concentrations for ^{226}Ra and ^{232}Th were determined through 1764.5 keV and 2614.5 keV gamma energies of ^{214}Bi and ^{208}Tl, respectively. For ^{40}K, its own photopeak energy (1460.3 keV) was used. Prior to measurement, mixture source consisting 3 radionuclides (^{137}Cs, ^{60}Co and ^{22}Na) with 5 peak energies (511 keV, 661.6 keV, 1173.2 keV, 1274.5 keV and 1332.5 keV) were used for the detector calibration.

RESULTS AND DISCUSSIONS

Natural Radioactivity

The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K were determined in 9 samples of clay bricks. Besides that, for comparison purpose, activity concentrations in 2 samples of cement bricks were also measured. The average activity values, together with their respective standard deviations are presented in Table 1. Comparative method was used to determine the activity concentration (Yasir et. al, 2007) following the Equation (1):

$$W_s = \frac{A_s \times M_r}{A_r \times M_s} \times W_r \qquad (1)$$

where W_s and W_r are activity concentration (Bqkg^{-1}), A_s and A_r are γ-ray emitted per second and M_s and M_r are mass (gram) in the brick samples and reference material, respectively. Reference material of IAEA soil-375 was used in this work. As shown in Table 1, the measured activity concentrations in clay bricks samples varies from 39.04 ± 0.88 Bqkg^{-1} - 73.61 ± 5.32 Bqkg^{-1} for ^{226}Ra, from 43.38 ± 2.60 Bqkg^{-1} - 73.45 ± 1.51 Bqkg^{-1} for ^{232}Th and from 381.54 ± 11.39 Bqkg^{-1} - 699.63 ± 15.82 Bqkg^{-1} for ^{40}K. Each value ranges were exceeded the world mean activity concentration values in soil which are 35 Bqkg^{-1}, 30 Bqkg^{-1} and 400 Bqkg^{-1} as reported in UNSCEAR (2000) for ^{226}Ra, ^{232}Th and ^{40}K, respectively. As a comparison, activity from another research works is tabulated in Table 2. Measurement was also conducted on 3 samples of clay bricks collected from the same factory in Serendah (SRN1, SRN2 and SRN3). The raw materials are the same but each sample has gone through different periods of burning process. From the result, it was found that period does not affect significantly the activity concentration values.

Table 1: Natural radioactivity concentration (NORM), radium equivalent (Ra_{eq}), and annual effective dose values in clay bricks (ClB) and cement bricks (CmB) samples in negeri Johor and Selangor, Malaysia.

Location	Activity concentration ($Bqkg^{-1}$)			Ra_{eq} ($Bqkg^{-1}$)	Annual effective dose ($mSvyr^{-1}$)
	^{226}Ra	^{232}Th	^{40}K		
A.H 1 (ClB)	47.69 - 52.04 *	52.33 - 57.24	669.99 - 682.98	178.69 - 185.01	0.41 – 0.42
	(50.34 ± 2.33) **	(55.26 ± 2.59)	(678.56 ± 7.42)	181.61	0.42 ± 0.07
A.H 2 (ClB)	42.16 - 51.78	55.24 - 58.93	420.83 - 433.95	158.40 - 166.89	0.36 - 0.37
	(48.05 ± 5.16)	(57.46 ± 1.95)	(427.82 ± 6.60)	163.16	0.37 ± 0.10
Y.P (ClB)	40.91 - 43.64	42.09 - 45.24	682.83 - 714.24	153.68 - 161.34	0.36 - 0.37
	(41.93 ± 1.49)	(43.95 ± 1.65)	(699.63 ± 15.82)	158.65	0.37 ± 0.10
KUL (ClB)	56.42 - 60.20	72.23 - 75.14	402.79 - 409.51	191.12 - 199.18	0.43 - 0.44
	(57.87 ± 2.03)	(73.45 ± 1.51)	(406.74 ± 3.52)	194.22	0.43 ± 0.09
BTN (ClB)	68.36 - 78.99	58.79 - 61.26	404.39 - 407.44	184.83 - 197.73	0.41 - 0.44
	(73.61 ± 5.32)	(59.91 ± 1.25)	(405.41 ± 1.76)	190.5	0.42 ± 0.14
KPG (ClB)	38.92 - 45.99	40.48 - 45.51	594.71 - 621.93	142.60 - 157.03	0.33 - 0.36
	(43.05 ± 3.69)	(43.38 ± 2.60)	(608.04 ± 13.62)	151.9	0.35 ± 0.18
SRN1 (ClB)	38.04 - 39.72	54.01 - 54.79	458.31 - 499.09	151.67 -155.10	0.34 - 0.35
	(39.04 ± 0.88)	(54.43 ± 0.40)	(481.31 ± 21.01)	153.94	0.35 ± 0.05
SRN2 (ClB)	43.10 - 50.00	60.16 - 62.08	369.72 - 392.45	160.48 - 166.73	0.36 - 0.37
	(45.78 ± 3.70)	(61.32 ± 1.02)	(381.54 ± 11.39)	162.85	0.36 ± 0.07
SRN3 (ClB)	37.35 - 45.72	59.78 - 62.71	377.48 - 405.70	151.90 - 166.63	0.34 - 0.37
	(42.93 ± 4.83)	(61.72 ± 1.69)	(396.26 ± 16.26)	161.72	0.36 ± 0.19
RWG (CmB)	200.17 - 215.78	39.04 - 40.96	1031.56 - 1062.66	338.38 - 353.78	0.79 - 0.82
	(209.80 ± 8.43)	(39.81 ± 1.01)	(1048.84 ± 15.84)	347.49	0.81 ± 0.18
DNG (CmB)	141.92 - 158.95	92.66 - 94.34	305.59 - 330.96	302.31 - 317.07	0.67 - 0.70
	(149.92 ± 8.56)	(93.71 ± 0.91)	(318.21 ± 12.69)	308.43	0.68 ± 0.17

* Range values
**Mean reading

Table 2: Comparison of mean activity concentration and radium equivalent values (Ra_{eq}) in Malaysia's clay bricks samples with another country.

Country	Natural radioactivity concentration ($Bqkg^{-1}$)			Ra_{eq} ($Bqkg^{-1}$)	References
	^{226}Ra	^{232}Th	^{40}K		
Algeria	65	51	675	190	Amrani et al. (2001)
Bangladesh	29.47	52.50	292.25	127.14	Mantazul et al. (1998)
Brazil	46.8	119.9	322	247.7	Malanca et al. (1995)
Cameroon	49.6	91	172	193.34	Ngachin et al. (2007)
China	58.6	50.4	713.9	178.3	Lu Xinwei (2005)
Egypt	24.0	24.1	258	78	El-Tahawy & Higgy (1995)
India	18.03	33.33	44.8	69.15	Kumar et al. (2003)
Kuwait	11.9	6.6	332	41.6	Bau-rabee and Bem (1996)
Mesir	33	37	511	-	Nour Khalifa. (2005)
Zambia	32	81	412	180	Hayambu et al. (1995)
Malaysia	32.3	30.9	487.7	114	Yasir et al. (2007)
World mean average (Soil)	35	30	400	-	UNSCEAR (2000)
Malaysia	49.18	56.76	498.37	168.73	This Work

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in building materials particularly in clay brick is not uniform. Thus, in order to represent the activities by a single quantity (taking into account the radiation hazard associated with them) the radium equivalent activity (Ra_{eq}) index has been used. It is defined by the following expression (Beretka & Mathew, 1985):

$$Ra_{eq} = A_{Ra} + 1.43 A_{Th} + 0.077 A_K \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the mean activity in $Bqkg^{-1}$ of ^{226}Ra, ^{232}Th and ^{40}K, respectively. Equation 2 is based on the fact that 370 $Bqkg^{-1}$ of ^{226}Ra, 259 $Bqkg^{-1}$ of ^{232}Th and 4810 $Bqkg^{-1}$ of ^{40}K produce the same γ-ray dose equivalent (El-Tahawy & Higgy, 1995). The Ra_{eq} estimation for all clay brick samples studied are included in Table 1. From the results, Ra_{eq} varies even in the same type of building material (clay bricks) collected from different areas. However, these mean Ra_{eq}

values ranging from 151.90 $Bqkg^{-1}$ - 194.22 $Bqkg^{-1}$ were less than the maximum admissible value of 370 $Bqkg^{-1}$ set in UNSCEAR report (1988), which is equivalent to an external dose of 1.5 $mSvy^{-1}$ (Ngacin, 2007; NEA-OECD, 1979). Ra_{eq} values in cement bricks sample have nearly reach the recommended maximum limit where the mean value are 347.49 $Bqkg^{-1}$ and 308.43 $Bqkg^{-1}$ for sample obtained in Rawang (RWG) and Dengkil (DNG), respectively. Commonly, cement brick in Malaysia are produced from 3 basic substances which are granite, sand and portland cement.

Previous works by Ruixiang (1986) founded that granite activity concentration were above the proposed acceptable level. Some works supporting findings such as Nour Khalifa Ahmed (2005) have reported that granite and marble activities give a great contribution to radiation exposure since the activity were found to be high.

Annual Effective Dose

Absorbed dose rate ($nGyh^{-1}$) are defined with the formula proposed by UNSCEAR (1988):

$$D = \sum_{X} A_x \times C_x \tag{3}$$

where A_x is the mean activity of ^{226}Ra, ^{232}Th and ^{40}K in the samples while C_x is the corresponding dose conversion factor. C_x values reported by UNSCEAR (2000) as 0.462 nGy h^{-1} per $Bqkg^{-1}$, 0.604 nGy h^{-1} per $Bqkg^{-1}$ and 0.0417 nGy h^{-1} per $Bqkg^{-1}$ for ^{226}Ra, ^{232}Th and ^{40}K, respectively. It's also reported that in order to estimate annual effective doses, the conversion coefficient from the absorbed dose in air to effective dose (C_f) and the indoor occupancy factor (O_f) must also be taken into account. So, the annual effective dose is calculated following this equation:

$$E = D \times C_f \times O_f \tag{4}$$

where D is given in equation (3), C_f is conversion coefficient and O_f is indoor occupancy factor for a year. In the UNSCEAR (2000) report, the committee used 0.7 $SvGy^{-1}$ for conversion coefficient from absorbed dose in air to effective dose (C_f) and 0.8 for indoor occupancy factor (O_f) since most people spend 80% of their daily time in a building (so, O_f for a year = 0.8 x 365 d x 24 hr). As indicated in Table 1, mean values of annual effective dose rate for clay brick is in a range 0.35 ± 0.18 $mSvy^{-1}$ - 0.43 ± 0.09 $mSvy^{-1}$. These values are less than 0.48 $mSvy^{-1}$ which is the worldwide average value reported in UNSCEAR (2000). Mean annual effective dose values in cement bricks samples were in the range of 0.68 $mSvy^{-1}$ – 0.81 $mSvy^{-1}$ which is higher than worldwide average. However this values is still under the recommended maximum level.

Hazard Index Assessment

There are two common hazard indexes that have been used in the previous studies which are external hazard index and internal hazard index (Kriger, 1981; Beretka & Mathew, 1985; Hayumbu et. al, 1995; Liu Xinmei, 2005). Determination of the external and internal hazard indexes in the bricks samples are shown in Figure 1. For external hazard index, the following equation proposed by Hewamma et. al, (2001) is used:

$$H_{ex} = \frac{C_{Ra}}{740} + \frac{C_{Th}}{520} + \frac{C_K}{9620} \tag{5}$$

where C_{Ra}, C_{Th} and C_K are referred to the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively. Radiation hazard are assume negligible if the value is less than unity.

The calculated values of external hazard index (H_{ex}) ranges from 0.20 – 0.26 in clay bricks samples. All the external hazard index (H_{ex}) values are under the recommended level, while values of external hazard index (H_{ex}) for cement bricks samples are found to be much higher compared to the clay bricks samples.

In addition to external hazard, radon and its short lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter product is quantified by the internal hazard index (H_{in}).

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

as in the equation (5) C_{Ra}, C_{Th} and C_K are referred to the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K respectively. The pattern of the result is similar to some previous works by Nour Khalifa Ahmad (2005) and Muhammad Iqbal et. al, (2000) where all the calculated internal hazard index (H_{in}) values in the building materials samples were higher than external hazard index (H_{ex}) values. All internal hazard index (H_{in}) values recorded in 9 samples of clay bricks are less than unity. However, we can see that hazard index (H_{in}) values in two samples of cement bricks are above unity. So, choosing clay bricks as a building materials component are more recommended while caution must be taken when using cement bricks.

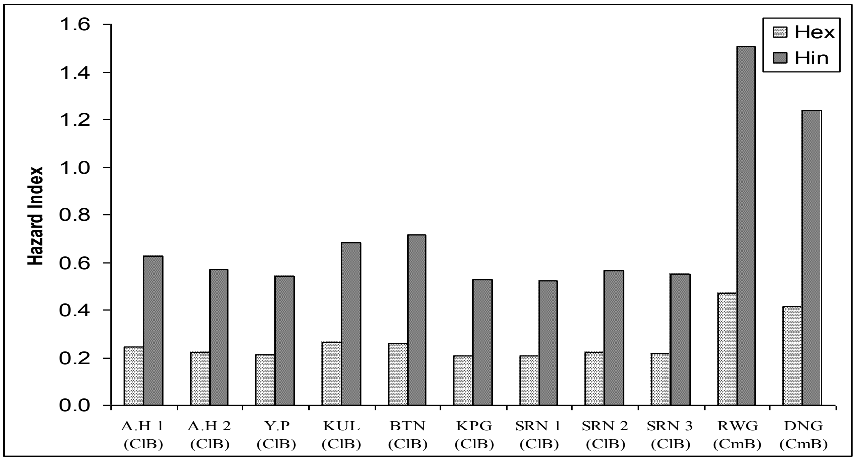

Figure 1: Internal and external hazard index values for clay brick (ClB) and cement brick (CmB) samples in Selangor and Johor, Malaysia.

CONCLUSIONS

The measurement of the radioactivity in some of Selangor and Johor bricks have lead to the following conclusions.

1. Activity concentration and radium equivalent activity (Ra_{eq}) varies even in the same type of building material collected from different areas. However, radium equivalent activity

(Ra_{eq}), annual effective dose rate, both internal and external hazard index in clay bricks samples indicated a measurement, less than the recommended values.

2. Different period of burning process of clay bricks have not affected significantly the activity concentration values.

3. Caution must be taken when using cement bricks since the activity concentration, radium equivalent activity (Ra_{eq}), internal and external hazard index have shown a significantly higher value compared to clay bricks samples.

ACKNOWLEDGEMENT

The authors sincere thanks to Ministry of Science, Technology and Innovation (MOSTI) for the research grant, committee members of research project 06-01-02-SF0129 and nuclear science staff for their encouragement and guidance provided throughout this work.

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