RADIATION HAZARD ASSESSMENT: NATURAL OCCURING RADIOACTIVE MATERIAL IN BRICKS

Nur Azna Mahmud[1], Zalina Laili[2], Ramlah Tajuddin[3]

[1]*Institute for Public Health, Ministry of Health Malaysia, Jalan Bangsar, 50590, KUALA LUMPUR.*

[2]*Malaysian Nuclear Agency, Ministry of Science, Technology & Innovation Malaysia,Bangi, 43000 Kajang, SELANGOR.*

[3] *Faculty of Engineering, University Technology MARA 40450 Shah Alam, SELANGOR.*

**Corresponding author: aznanur@yahoo.com*

ABSTRACT

This study focused on the activity concentration and radiation hazard assessment of radionuclide in bricks. The activity concentrations per unit of mass in the studied bricks ranged from 59.82 to 236.65 Bqkg^{-1} for Ra-226, from 66.29 to 185.4 Bqkg^{-1} for Th-232, and from 283.50 to 1599.67 Bqkg^{-1} for K-40. In terms of radiation hazard assessment, granite bricks exceeded the allowance limit stated by Nuclear Energy Agency -Organisation for Economic Co-operation & Development (NEA-OECD) in radium equilibrium activity (R_{eq}), External Hazard Index (H_{ex}), Internal Hazard Index (H_{in}) and Representative Level Index (I_{yr}). Thus, this has proof that the naturally occurring radionuclide materials present in the brick have a potential in effecting the dwellers health.

ABSTRAK

This study focused on the activity concentration and radiation hazard assessment of radionuclide in bricks. The activity concentrations per unit of mass in the studied bricks ranged from 59.82 to 236.65 Bqkg^{-1} for Ra-226, from 66.29 to 185.4 Bqkg^{-1} for Th-232, and from 283.50 to 1599.67 Bqkg^{-1} for K-40. In terms of radiation hazard assessment, granite bricks exceeded the allowance limit stated by Nuclear Energy Agency -Organisation for Economic Co-operation & Development (NEA-OECD) in radium equilibrium activity (R_{eq}), External Hazard Index (H_{ex}), Internal Hazard Index (H_{in}) and Representative Level Index (I_{yr}). Thus, this has proof that the naturally occurring radionuclide materials present in the brick have a potential in effecting the dwellers health.

Keywords: brick, naturally occurring radioactive material, radioactivity, and radiation hazard assessment

INTRODUCTION

Bricks are normally a block of ceramic material that has been used as one of the building materials in masonry construction. Bricks are still being used due to their great physical strength and engineering characteristics (Aeslina, and Abbas, 2011). Beside clay, other materials can be used to produce bricks such as cement, concrete, ash, granite, mud and sludge. Nowadays, industrial waste or by-products such as fly ash, coal slag, phosphogypsum, are extensively used in building materials (Gustavo Haquin, 2008). These materials are known to have natural radionuclide which result the bricks to become naturally radioactive. Naturally occurring radionuclide in brick can be the potential sources of external and internal radiation exposure in dwellings due to the gamma radiation emitted by the members of uranium and thorium decay series and isotope of K-40 (Ravisankar et al., 2011). Most of the dwellers spend 80% of their time indoors and have the possibility to be exposed to gamma rays and radon gas(IAEA, 2008).Chronic exposure to radium and thorium can induce bone cancer and damage the cell of the lung when it is inhaled and deposited in the mucous lining in the respiratory tract (USEPA, 2012).

The common calculation that is used in the radiation hazard assessment is by determination of radium equivalent activity, external and internal hazard index and representative level index. Radium equivalent activity calculation usually indicate the rate of effective equivalent dose one meter from the source surface while the calculation of the three index (external, internal and representative level) estimates the radiation hazard cause by material it selves(Flores et L., 2005). Typical world values which are 500, 50 and 50 $Bgkg^{-1}$ for K-40 , Ra-226, and Th-232 respectively and the allowance limits of the Internal Hazard Index (Hin) and External Hazard Index (Hex) less than one and representative level index less than 1.5, should be considered in minimizing the radiation hazard.(NEA-OECD, 1979).

This study was conducted to measure the radioactivity of Ra-226, Ra-228, Th-232 and K-40 present in the building materials commonly used in the construction industry in Malaysia. The activity concentration value of the common naturally occurring radioactive material from building materials collected are required in assessing the radiation index of the building. Therefore, the most suitable brick in term of low radiation hazard can be determined in order to minimize the radiation hazard. Moreover, study done in estimating the radioactivity level of materials can be used in benchmarking and formulating the compliance limit for the regulation regarding radiation protection since the baseline data is limited in Malaysia.

MATERIAL AND METHODS

Samples Selection

Five types of brick were sourced from local hardware shops. The selected bricks were clay, cement, fly ash, granite and pavers bricks. Each type of bricks was prepared for activity concentration measurement.

Activity Concentration Measurement

The brick samples were grounded by using Laboratory Ball Mill Grinder Machine (Retsch Model PM100) and sieved by using USA Standard Sieve No. 18 (equivalent to 1.0mm).Plastic container was filled with the brick dust and the weights of the samples were recorded. The samples were labelled accordingly with the detail of the sample types, date of preparation and weight. The samples were kept at room temperature for three weeks to reach secular equilibrium between Rn-222 and Ra-226. For the activity concentration of the radionuclide, the samples were counted for 24 hours by using a Canberra n-type high purity germanium (HPGe) gamma spectrometry system with 30% relative efficiency and 1.9 keV resolutions at 1332.5 keV of Co-60. calibration on the efficiency of the detector were performed by using certified multi-nuclide standard source in 350ml plastic container containing Am-241, Cd-109, Co-57, Te-123m, Cr-51, Sn-113, Sr-85, Cs-137, Y-88 and Co-60. The spectrum of the samples were analyzes by using Genie-2000 analysis software. The radionuclides were identified according to the energy level.

For solid sample, activity concentration of K-40 was determined at energy level of 1460 keV, Ra-226 at energy level of 295 keV (19.2%), 352 keV (37.%) from Pb-214 , 609 keV (46.50%) from Bi-214 and Th-232 at energy level 238.6 keV (43.6%) from Pb-212 , 338.4 keV (12%) , 911 keV (27.7%) , 968.9 keV (16.6%) from Ac-288 and 583 keV (30.60%) from TI-208. For samples in the liquid form, energy level of 295 keV (19.2%), 352 (37.1%) keV from Pb-214 and 609 keV (46.5%) from Bi-214 were used to determine Ra-226 while Ra-228 were determined at energy levels of 911 keV (27.7%). The activity concentrations of the radionuclides were reported as per unit weight or volume, $Bqkg^{-1}$ and BqL^{-1}.

RADIATION HAZARD ASSESSMENT

Radiation Hazard Assessment was performed by calculating the four indexes, i.e. Radium equivalent activity (Ra_{eq}), External hazard index (H_{ex}), Internal hazard index (H_{in}) and representative level index (I_{yr}).Radium equivalent index (Ra_{eq}) and representative level index (I_{yr}) are commonly used to estimate radiation hazard(Yasir et al., 2004).

Radium Equivalent Activity (Ra_{eq})

Ra_{eq} is a common index used to compare the specific activity of ^{226}Ra, ^{232}Th and ^{40}K contain in the samples since the distribution of the radionuclide is not uniform. Raeq is a weighted sum of activities of the three radionuclides based on the estimation value of 10 $Bqkg^{-1}$, 7 $Bqkg^{-1}$ and 130 $Bqkg^{-1}$ for ^{226}Ra, ^{232}Th and ^{40}K respectively. By using the estimated value, Raeq can be calculated by using an equation (Beretka J and Matthew PJ, 1985).

$$\mathbf{Raeq=CRa + 10/\ 7Cxh + 10/130C_K}$$

where

C_{Ra}= specific activities of ^{226}Ra, $Bqkg^{-1}$.

Cxh= specific activities of ^{232}Th, $Bqkg^{-1}$.

C_K = arespecific activities of ^{40}K, $Bqkg^{-1}$.

External Hazard index (H_{ex})

External hazard index or indoor radiation hazard index symbolized as H_{ex}. H_{ex} value can be calculated by an equation (Beretka J and Matthew PJ, 1985).

$$\mathbf{Hex=CRa/370+CTh/259+CK/4810 < 1}$$

where

C_{Ra}= specific activities of ^{226}Ra, $Bqkg^{-1}$.

Cxh= specific activities of ^{232}Th, $Bqkg^{-1}$.

C_K = arespecific activities of ^{40}K, $Bqkg^{-1}$.

Internal Hazard Index (H_{in})

Internal hazard index is due to exposure of radon and its daughter that have high carcinogenic effect. Internal hazard index is denoted as H_{in} andcan be calculated by an equation (Beretka J and Matthew PJ, 1985).

$$\mathbf{Hin=CRa/185+Cih/259 + Ck/4810 < 1}$$

where

C_{Ra}= specific activities of ^{226}Ra, $Bqkg^{-1}$.

C_{Th}= specific activities of ^{232}Th, $Bqkg^{-1}$.

C_K = arespecific activities of ^{40}K, $Bqkg^{-1}$.

Representative Level Index (1^)

The degree of gamma radiation hazard caused by natural radionuclides contain in building material is estimated by the representative level index, I_{yr}(Yasir et al., 2004). Representative level index is denoted as I_{yr} and can be calculated by an equation. (Beretka J and Matthew PJ,1985).

$$I_{T}r = CRa/150 + CTh/100 + Ck/1500 < 1$$

where

C_{Ra}= specific activities of ^{226}Ra, $Bqkg^{-1}$.

C_{Th}= specific activities of ^{232}Th, $Bqkg^{-1}$.

C_{K} = arespecific activities of ^{40}K, $Bqkg^{-1}$.

RESULTS AND DISCUSSION

Specific activity concentrations of Ra-228, Ra-226, Th-232 and K-40, measured in the studied bricks are listed in Table 1, the activity concentrations were compared with corresponding typical world value which is 50, 50 and 500 $Bqkg^{-1}$ for Ra-226, Th-232 and K-40 respectively (NEA-OECD, 1979). As listed in the table, the activity concentrations of the uranium daughters (Ra-226) in the studied samples ranged from 59.82 $Bqkg^{-1}$ in clay brick to 236.65 $Bqkg^{-1}$ in granite brick. The activity concentrations of Ra-226 in all the samples exceed the corresponding world average of 50 $Bqkg^{-1}$ (NEA-OECD, 1979).

Table 1: Specific activity concentration of Ra-228, Ra-226, Th-232 and K-40 of the samples

Radionuclide	Samples (Geometry 350ml) ,$BqKg^{-1}$					Typical Value
	Clay	Cement	Granite	Fly ash	Paver	
Ra-228	69.78	94.37	84.28	182.5	68.9	-
Ra-226	59.82	102.65	236.7	72.04	65.5	50
Th-232	94.1	86.73	185.4	70.82	66.3	50
K-40	389.9	1030.8	1600	283.5	383	500

This might be due to the aggregates use to manufactured bricks is predominantly high content of naturally occurring radioactive material. However, the activity concentration of Ra-226 and Ra-228 in all the bricks were still within the range of radium concentrations in peninsular granite studied is 120-490 $Bqkg^{-1}$ and 120-430$Bqkg^{-1}$, respectively (Omar & Hassan, 1999). Study by Gustavo (2008), showed that the content of natural occurring radioactive material (NORM) in the bricks depends on the origin of the raw material used to manufacture the bricks. Granite brick shows the highest activity concentration of Th-232 content at 185.4 $Bqkg^{-1}$ while the low activity concentration of Th-232 was detected in paver brick at 66.29 $Bqkg^{-1}$.The activity concentrations of Th-232 in all the samples exceeded the typical value of 50 $Bqkg^{-1}$for Th-232(NEA-OECD, 1979).The result had shown that granite brick had the highest activity concentrations of radium and thorium. Igneous rocks of granitic composition are rich with uranium and thorium due to partial melting and fractional crystallization of magma (Alharbiet, 2011). The activity concentration of K-40 in table 1 also shows that granite brick have the highest concentration with 1599.67 $Bqkg^{-1}$ while the lowest activity concentration is detected in fly ash bricks which are 283.50 $Bqkg^{-1}$. The activity concentration of K-40 in clay brick, fly ash brick and paver brick are less than the typical world average value of 500 Bq/kg except for granite brick and cement brick.

RADIATION HAZARD ASSESSMENT

The calculated value of Radium equivalent activity of the studied building material is listed in table 2 ranges from 189.62 to 625.56 Bqkg^{-1}. Radium equivalent activity for granite bricks is the highest at 625.56 while paver brick possessed the lowest Ra $_{eq}$ among the studied bricks. The Ra_{eq} for all the samples except for granite were found to be lower than the limit state by NEA-OECD which is 370Bqkg^{-1}.This indicate that, an extra consideration need to be taken if granite bricks is use as the building bricks in order to minimize the radiation hazard cause by the granite bricks. The value of external hazard index must be less than unity in order the external hazard index to be below the limit of 1.5 mSv yr^{-1} (Beretka J and Matthew PJ, 1985).H_{ex} of the studied sample is less than unity except for granite at 1.69. This shows that granite have the potential on causing the external hazard to the dwellers if it is use as the building material.

Beside external radiation hazard, the internal radiation hazard, Hin which mainly contributed by the radon and its shorts live progeny need to be taken into consideration in the radiation hazard assessment. The value of the Hinmust not exceed one. Calculated Hinvalue of the studied bricks shows that the H_{in}value for cement and granite bricks has exceed the limit at 1.10 and 2.33 respectively while for the other samples shows H_{in}value is less than 1.0. Based on the H_{in}value, cement and granite might contribute to the internal radiation especially to the dwellers. The internal radiation cause by cement bricks might due to the sand used as the aggregates. A study done by International Atomic Energy Agency shows minerals in the sands are deposited by gravity and can achieve to 4000 Bq/kg activity concentration of certain types of radionuclide (IAEA, (2003).

Table 2: Radium Equivalent Activity, external and internal hazard index and representative level index of the samples.

Radiation Hazard Assessment components	Samples (Geometry 350ml) ,BqKg^{-1}					Allowance Limit
	Clay	Cement	Granite	Fly ash	Paver	
Radium Equivalent Activity ,Bqkg^{-1}	224	305.8	625.56	195.02	189.62	<370
External hazard Index , H_{ex}	0.6	0.83	1.69	0.53	0.51	<1.0
Internal hazard index, Hin	0.8	1.1	2.33	0.72	0.69	<1.0
Representative level index, I_{Yr}	1.6	2.24	4.5	1.38	1.35	<1.5

As recommended by OECD's nuclear Energy Agency , the representative level index, Iy_r is used to estimate the level of gamma radiation hazard associated with the natural radionuclide in specific building materials such as bricks (NEA-OECD, 1979).The degree of gamma radiation hazard caused by natural radionuclides contains in building material is estimated by the representative level index, I_{yr} (Yasir et al., 2004). Based on the calculated value, the Iy_r for the samples is in the range of 1.35 up to 4.50.

The Iy_r for all the samples have exceeded the unity which is the upper limit for the representative level. The NORM can be presence in the soil of each region in the world depends primarily on the geological and geographical conditions at different level (UNSCEAR, 2000).

CONCLUSION

Generally, the activity concentration of Ra-226 and Th-232 for all the bricks has exceeded the international typical value, 50 Bqkg-1 and 50 Bqkg-1. Activity concentration of K-40 in clay brick, paver brick and fly ash brick is lower than international typical value, 500 Bqkg-1 except for granite brick and cement brick. Based on the radiation hazard indexes assessment, fly ash brick possess the lowest potential radiation hazard among the bricks followed by paver brick, clay brick, cement brick and granite brick. Therefore, radiation hazard assessment based on activity concentration of the radionuclide can be used as a benchmark in formulating the compliance limit for the regulation regarding radiation protection in Malaysia.

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