SHORT COMMUNICATION

Journal of Radioanalytical and Nuclear Chemistry, Vol. 262, No. 3 (2004) 775–777

Natural radioactivity in some building materials and by-products of Shaanxi, China

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(Received June 23, 2004)

The concentration of ^{226}Ra, ^{232}Th and ^{40}K has been determined by γ-ray spectrometry. The measured activity in the selected building materials and by-products of coal fired power plants ranges from 21.5 to 91.3 Bq·kg^{-1}, 15.4 to 63.2 Bq·kg^{-1} and 83.2 to 683.9 Bq·kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The results were compared with the reported data of other countries and with the world average activity of soil. The radium equivalent activities values of all building materials are lower than the limit of 370 Bq·kg^{-1}, equivalent to a γ-dose of 1.5 mSv·y^{-1}. The values of the external hazard index and the internal radiation hazard index are less than unity.

Introduction

The naturally occurring radionuclides present in building materials, recycled industrial waste products and soil are ^{226}Ra, ^{232}Th and ^{40}K,[1,2] which are the main sources of radiation of building materials. These radionuclides present external exposure risks due to their γ-ray emissions and internal ones due to radon and its progeny which emit alpha particles.[3]

Several building materials and those derived from industrial wastes and by-products have been shown to have high levels of radioactivity in many countries.[4–6] Fly ash, the particulate by-product of coal combustion, has been found to have a higher concentration of radionuclides than the coal itself.[6–8] In recent years, fly ash has been used as a replacement of sand and cement in premixed concrete, manufacture of blended fly ash Portland cement, aerated concrete, fly ash clay bricks and blocks and for filling of underground cavities, etc.[9]

The natural radioactivity of building materials in some Chinese areas and many countries have been reported.[9–17] However, detailed information of Shaanxi's building materials and by-products were not available. Therefore, systematic studies were performed and the activities of ^{226}Ra, ^{232}Th and ^{40}K in these materials and by-products have been determined.

In this paper, the concentration of ^{226}Ra, ^{232}Th and ^{40}K content of building materials has been estimated using γ-ray spectroscopy. Common building materials and the by-products of coal power plants that are being used, or have potential for use, in building construction were selected for the study. The paper presents the results for the measured activities, the radium equivalent concentrations and the values of both the internal and external hazard indices associated with the usage of building materials and by-products collected from Shaanxi, China. The results obtained in the present study are compared with the findings of other studies.

Experimental

The samples investigated were some common building materials and by-products of a coal power plant having the potential for use in large quantities in the building industry. The building materials measured were cement, cement plaster, cement brick, red-clay brick, sand, gravel aggregate, lime/limestone, and roof asbestos. Sand and gravel aggregate samples were collected from construction sites, and other building materials from various agencies supplying raw materials for building construction. Fly ash and slag samples were obtained from three large thermal power stations (Baiji, western suburb of Xi'an and Baqiao) of Shaanxi, China. Samples with large grain-size were crushed and milled to a fine powder with a particle size less than 0.16 mm, except for the cement, cement plaster, fly ash and part sand samples. Each of the materials was homogenized and air-dried and approximately 500 g was filled into cylindrical plastic containers (7.5 cm height and 10.5 cm diameter). Their respective net weights were recorded. These samples were then left for 4 weeks to allow for Ra and its short-lived progeny to reach radioactive equilibrium.

The concentration of the natural radioactivity (^{226}Ra, ^{232}Th and ^{40}K) in the material samples were determined using a ∅12.5×10 cm^3 NaI(Tl) γ-ray spectrometric system with excel 8% energy resolution (^{137}Cs) and 20% counting efficiency. The detector was maintained in a vertical position in a lead cylindrical shield of 10 cm thickness and 55 cm height. The internal dimensions of

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0236–5731/2004/USD 20.00

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the free surface within the shielding enclosure are 13 cm in diameter and 15 cm in deep. The detector was coupled to a 256 multi-channel pulse-height analyzer and the system was calibrated for the γ-energy range 80 keV to 3.2 MeV. The energy region for ^{40}K, 1.46 MeV γ-rays, ^{226}Ra, 1.76 MeV γ-rays and ^{232}Th, 2.62 MeV γ-rays were chosen as 1.30–1.60; 1.62–2.00 and 2.45–2.90 MeV, respectively. Standard sources for ^{226}Ra and ^{232}Th (in secular equilibrium with ^{228}Th) were prepared using known activity contents and mixing with the matrix material of phthalic acid powder. In order to avoid the loss of gaseous daughter products of ^{226}Ra and ^{232}Th which may lead to disturbance in radioactive equilibrium, the prepared standard sources were kept in sealed plastic containers (7.5 cm height and 10.5 cm diameter). Analar grade potassium chloride (KCl) of a known amount of the same geometry was used as the standard source of ^{40}K. The samples were counted for about 200 to 400 minutes. Each sample was counted twice before an average was taken.

Results and discussion

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in various building materials and by-products investigated in this work are given in Table 1. As shown in Table 1, the activity concentrations of ^{226}Ra in all building materials and by-products exceeded that of the world average for soil (25 Bq·kg^{-1}),[18] except for lime/limestone (21.5 Bq·kg^{-1}). The activity concentrations of ^{232}Th in cement, cement brick, cement plaster, red-clay brick, fly ash and slag exceeded that of the world average of 25 Bq·kg^{-1} for soil.[13] The activity levels of ^{232}Th in other materials were lower than the world average value of soil. Similar for ^{40}K activities, the activity concentrations obtained for all building materials, except for red-clay brick (683.9 Bq·kg^{-1}), were lower than the average value for soil (370 Bq·kg^{-1}).[13]

To assess the radiological hazard of the building materials used, it is useful to calculate an index called the radium equivalent activity (Ra_{eq}),[19] defined according to the estimation that 1 Bq·kg^{-1} of ^{226}Ra, 0.7 Bq·kg^{-1} of ^{232}Th and 13 Bq·kg^{-1} of ^{40}K produce the same γ-ray dose.[19,20] This index Ra_{eq} is given as:

$$Ra_{eq} = C_{\mathrm{Ra}} + 1.43C_{\mathrm{Th}} + 0.077\ C_{\mathrm{K}} \tag{1}$$

where C_{Ra}, C_{Th} and C_{K} are the activity concentrations in Bq·kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The calculated values of the radium equivalent Ra_{eq} for the studied building materials are given in Table 1. These values range from 49.9 to 206.6 Bq·kg^{-1}.

To limit the external gamma-radiation dose from building materials to 1.5 mSv·y^{-1}, the external hazard index (H_{ex}) is defined as:[10,19]

$$H_{ex} = (C_{\mathrm{Ra}}/370) + (C_{\mathrm{Th}}/259) + (C_{\mathrm{K}}/4810) \tag{2}$$

where C_{Ra}, C_{Th} and C_{K} are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The value of this index must be less than unity for the radiation hazard to be negligible. For the maximum value of H_{ex} to be less than unity, the maximum value of Ra_{eq} must be less than 370 Bq·kg^{-1}. The calculated values of H_{ex} for the studied building materials range from 0.13 to 0.56, values which indeed are less than unity.

In addition to the external hazard, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index (H_{in}) which is given by:[19]

$$H_{in} = (C_{\mathrm{Ra}}/185) + (C_{\mathrm{Th}}/259) + (C_{\mathrm{K}}/4810) \tag{3}$$

If the maximum concentration of radium is half that of the normal acceptable limit, then H_{in} will be less than 1.0.[19] For the safe use of a material in the construction of dwellings, H_{in} should be less than unity. The calculated values of H_{in} range from 0.19 to 0.80.

Table 2 compares the reported values of radium equivalent activities for selected building materials and by-products from other countries. As shown the values of radium equivalent activities obtained for lime/limestone and red-clay brick in this work are similar to those obtained for the German and Algerian samples, respectively. In the case of cement, the result of this study is of the same order as the Algerian, Australian, and Indian samples. The radioactivity in building materials varied from one country to another or even there are variations in the radium equivalent activities of different materials within the same type of materials. The results may be important from the point of view of selecting suitable materials for use in building construction. Large variations in radium equivalent activities may suggest that it is advisable to monitor the radioactivity levels of materials from a new source, before adopting it for using as a building material.

Table 1. Activity concentrations (in Bq·kg^{-1}) and radium equivalent activities of building materials under study

Material	Number of samples	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}, Bq·kg^{-1}
Cement	26	58.5 ± 7.5	38.6 ± 5.4	181.4 ± 38.6	127.7
Cement brick	6	46.2 ± 12.5	28.4 ± 6.2	137.4 ± 36.6	97.4
Cement plaster	8	64.7 ± 12.7	48.7 ± 9.3	161.3 ± 31.4	146.8
Gravel aggregate	12	28.5 ± 3.5	19.5 ± 6.7	286.7 ± 17.6	78.5
Red-clay brick	20	68.6 ± 13.4	56.4 ± 10.5	683.9 ± 46.6	201.9
Sand	9	40.7 ± 4.3	21.5 ± 5.6	302.6 ± 31.4	94.7
Lime/limestone	13	21.5 ± 8.3	15.4 ± 3.6	83.2 ± 15.4	49.9
Roof asbestos	6	31.3 ± 6.4	16.7 ± 8.2	156.8 ± 27.3	67.3
Fly ash	12	86.7 ± 36.2	56.4 ± 24.6	356.8 ± 67.3	194.8
Slag	6	91.3 ± 13.6	63.2 ± 18.2	323.8 ± 56.4	206.6

Table 2. Comparison of mean radium equivalent activity, Ra_{eq} (in Bq·kg^{-1})

Material	Shaanxi, China	Algeria[15]	Australia[19]	Germany[5]	India[9]	Malaysia[13]
Cement	128	112	115	70	105	188
Cement plaster	147	101	15	41	135	138
Gravel aggregate	79	58	115	322		
Red-clay brick	202	190	883	640		895
Sand	95	28	70	59	171	136
Lime/limestone	50	37	115	48		
Fly ash	195		355	451	109	
Slag	207		340	421	190	

Conclusions

The natural radioactivity of some building materials and by-products of coal fired power plants of Shaanxi, China has been measured using γ-ray spectroscopy. The Ra_{eq} values of all building materials and by-products were lower than the limit of 370 Bq·kg^{-1}, equivalent to a γ-dose of 1.5 mSv·y^{-1}. The values of both internal and external hazard indices associated with the usage of building materials and by-products collected from Shaanxi, China were less than unity. Therefore, the use of these materials in construction of dwellings is considered to be safe for inhabitants.

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This work was supported by the Provincial Natural Sciences Foundation of Shaanxi through Grants 2003D04 and by the Science Foundation of Shaanxi Normal University. Gratitude is expressed to J. XIAOWEI, W. XIAOLEI, L. ZHENDONG for assisting in sampling.

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