NATURAL RADIOACTIVITY AND RADIOLOGICAL HAZARD OF RED-CLAY BRICK PRODUCED IN SHANGLUO, CHINA

by

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The radiological hazard of building materials originating from clay, rock and other mineral wastes has attracted more attention because they contain natural radionuclides (^{226}Ra, ^{232}Th, and ^{40}K). The activity concentration of radionuclides in red-clay brick samples obtained from three different brickyards in Shangluo, China was measured. Various indexes, including radium equivalent activities, Ra_{eq}, external hazard index, H_{ex}, internal hazard index, H_{in}, indoor air absorbed dose rate, D, and annual effective dose, *AED*, of the aforementioned radionuclides in the bricks were used to assess the radiation hazard for people. The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were respectively 34.5 1.9, 62.5 2.1, and 713.7 19.8 Bqkg^{-1} for the studied red-clay bricks. The Ra_{eq} values of the red-clay brick samples varied from 167.0 to 184.7 Bqkg^{-1}, which are lower than the limit of 370 Bqkg^{-1}. Moreover, the activity concentrations of natural radionuclides in unfired brick, clay and coal were also determined and the results were compared with that in the red-clay brick samples. This study shows that the red-clay bricks produced in Shangluo, China can be used safely in construction industries.

Key words: natural radioactivity, radium equivalent activity, radiological hazard, red-clay brick

INTRODUCTION

Building materials derived from the Earth's crust, contain varying amounts of natural radionuclides. These radionuclides contained in building materials can cause exposure to external and internal radiation of individuals living in those buildings [1-4]. Gamma radiation originating from primordial nuclides (^{226}Ra, ^{232}Th, ^{40}K and their decay products) can cause external radiation exposure, however, the internal radiation exposure is caused by the inhalation of radioactive noble gases radon (^{222}Rn, a daughter product of ^{226}Ra) and thoron (^{220}Rn, a daughter product of ^{224}Ra), and their short-lived decay products which are exhaled from construction materials into indoor air, especially where the ventilation is poor. As most of the residents spend more than 80 % of their lifetime indoors [5, 6], knowledge of the natural radioactivity in building materials is important for the determination of population exposure to radiation. Moreover, an understanding of this activity helps to develop standards and national guidelines for the use and management of these materials and to assess the radiation hazards to human health associated with them.

Natural radioactivity of various building materials has been reported by researchers in several areas of China [7-17] and in many other countries such as Egypt [18-23], Israel [24, 25], Italy [26], Pakistan [27], Lebanon [28], Greece [29], North Macedonia [30], Serbia [31, 32], Turkey [3, 33-36], Yemen [37, 38], Austria [39], Jordan [40], India [41], and Malaysia [2]. However, data retrieval indicates that the measurement of radioactivity of building materials in Shangluo, China has not been previously carried out. This study was carried out to measure the radioactivity of the red-clay bricks which are the dominant artificial construction materials used in Shangluo, China and to assess their associated radiation exposure to the residents.

MATERIALS AND METHODS

Sampling

Shangluo is located in the southeast of the Shaanxi Province, northwest China, between the longitudes of 33°2'30"-34°24'40" N and the latitudes of 108°34'20"-111°1'25" E with a population of 2.49 million. The water conservation land of the Middle Route Project of the national South-to-North Water Transfer,

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is a strategic project to solve the water shortage problem in northern China. In recent years, with the implementation of the South-to-North Water Transfer Project, urban and rural construction in Shangluo has developed rapidly. Large quantities of various materials are required in the building industry. Due to its low cost and durability, red-clay brick is one of the most commonly used building materials in the construction industry both in the urban district and rural area of Shangluo. Red-clay bricks used in Shangluo mainly came from three brickyards located in villages Zhaoyuan, Liuwan and Shahezi near the urban district.

The investigated red-clay brick samples in this study were collected from these three brickyards. Eighteen red-clay brick samples were collected from three brickyards (6 samples per brickyard) and 12 unfired brick samples were collected from the brickyards in Liuwan and Shahezi (6 samples per brickyard). The size of each red-clay brick is about 23 cm × 11 cm × 5 cm and weighs about 2 kg. Clay is the dominant materials for red-clay brick production, and its chemical composition is the main factor affecting the natural radioactivity level of red-clay brick. Three clay samples, each of which was about 2 kg, were collected from each brickyard. Coal which can increase the strength and reduce the apparent density of the red-clay brick constitutes a relatively small portion of the materials in red-clay brick production. Three coal samples, each of which was about 2 kg, were also gathered from every brickyard. All the gathered samples were cataloged and marked at the initial production site.

The collected samples were taken back to the laboratory, dried naturally at room temperature, broken into small pieces by a hammer for further crushing. All the samples were then crushed by a ball crusher to pass through a 100 mesh sieve. To minimize cross-contamination between samples, the ball crusher was completely cleaned after each sample was ground. The samples were then homogenized and dried at 110 °C for 48 hours to constant weight. After moisture is completely removed, the sample powder was cooled to a normal temperature in a moisture-free atmosphere. Powder samples were weighed and placed into airtight, radon impermeable, cylindrical polyethylene plastic containers (these containers were the same in size of 7.0 cm in height and 6.5 cm in diameter). These containers were kept in a laboratory for about one month in order to allow ^{226}Ra, ^{232}Th, and their short-lived decay products to reach radioactive equilibrium. One month later, the natural radionuclide concentrations of the samples were analyzed by γ-ray spectrometry.

Radioactivity measurement

A 3 × 3 inch NaI (Tl) detector with an energy resolution of 8 % (^{137}Cs 661.6 keV) was used to estimate the concentrations of natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in the investigated samples. In order to reduce the background effect, the detector was located in a cylindrical lead chamber which is 10.5 cm thick and 38 cm high. The inside diameter of the lead chamber is 21 cm and the height from the detector to the upper chamber is 17 cm. A 1024 microcomputer multi-channel pulse height analyzer was coupled to the detector, and the system was calibrated for the gamma-energy from 50 keV to 3.2 MeV [10, 11]. Photopeaks at several energies were used to average the activity concentrations. The gamma lines of 609.3 and 1764.5 keV, emitted from ^{214}Bi were used for ^{226}Ra. The 238.6 keV photopeak from ^{212}Pb and 2614 keV photopeak from ^{208}Tl were averaged for ^{232}Th. The 1460.8 keV photopeak was used directly to measure the concentration of ^{40}K.

Standard materials, purchased from the Beijing Research Institute of Uranium Geology, were used to calibrate the detection system. Certain activity content together with phthalic acid powder was used to prepare the standard sources for ^{226}Ra and ^{232}Th (in long-term equilibrium with ^{228}Th). The prepared standard sources were stored in sealed cylindrical polyethylene containers with a height of 7.0 cm and a diameter of 6.5 cm. Therefore, it is possible to prevent the loss of gaseous decay products of ^{226}Ra and ^{232}Th, which may result in interference with the radioactive balance. A known amount of analar grade potassium chloride of the same geometry was used as the standard source of ^{40}K. Each sample was measured for 300 min and counted two times, and then the results were averaged.

Radiological hazard assessment

In order to compare the radiological effects of the red-clay bricks, unfired bricks, clays and coals which contain ^{226}Ra, ^{232}Th, and ^{40}K, an index called radium equivalent activity, Ra_{eq}, has been introduced. The Ra_{eq} is expressed as [42]

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

where C_{Ra}, C_{Th}, and C_{K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K respectively in Bqkg^{-1}. Equation (1) is based on the assumption that 370 Bqkg^{-1} of ^{226}Ra, 259 Bqkg^{-1} of ^{232}Th and 4810 Bqkg^{-1} of ^{40}K cause the same gamma-ray dose equivalent. The maximum value of Ra_{eq} in building materials must be no more than the recommended limit of 370 Bqkg^{-1} in order to keep the external annual dose below 1.5 mSv [1].

According to [43], the superior limit of the annual radiation dose produced by building materials is 1.5 mSv. For limiting the external gamma annual radiation dose from the studied materials to the normal annual dose equivalent of 1.5 mSv, the external hazard index, H_{ex}, is defined as [42]

$$H_{\text{ex}} \quad \frac{C_{\text{Ra}}}{370} \quad \frac{C_{\text{Th}}}{259} \quad \frac{C_{\text{K}}}{4810} \tag{2}$$

where C_{Ra}, C_{Th}, and C_{K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bqkg^{-1}, respectively. The value of this index must be less than the unit for the radiation hazard to be negligible, *i. e.* the annual radiation exposure due to radioactivity in construction materials must be limited to 1.5 mSv.

In addition to the external hazard, inhalation and ingestion of terrestrial radionuclides can lead to internal exposure. Doses by inhalation are due to the presence of air dust particles containing radionuclides of the ^{238}U and ^{232}Th decay chains. The short-lived decay products of radon (^{222}Rn) contributed most to the inhalation exposure. The internal exposure to radon and its decay products is determined by the internal hazard index, H_{in}, which is given by [42]

$$H_{\text{in}} \quad \frac{C_{\text{Ra}}}{185} \quad \frac{C_{\text{Th}}}{259} \quad \frac{C_{\text{K}}}{4810} \tag{3}$$

where C_{Ra}, C_{Th}, and C_{K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in Bqkg^{-1} for the studied samples. In order to keep the radioactivity of building materials in the safe range, the value of H_{in} should be smaller than unit. Due to gamma rays released by natural radionuclides in the building materials, buildings will result in different radiation doses to the whole body. The indoor air absorbed dose rate, D [nGyh^{-1}], is evaluated according to [44]

$$D \quad 0.92C_{\text{Ra}} \quad 1.1C_{\text{Th}} \quad 0.08C_{\text{K}} \tag{4}$$

where C_{Ra}, C_{Th}, and C_{K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bqkg^{-1}, respectively. Equation (4) is used to calculate the absorbed dose rate in air at a height of 1.0 m above the ground from the measured radionuclide concentration in building materials.

To estimate the *AED*, a conversion factor (0.7 Sv Gy^{-1}) was applied for conversion from the air absorbed dose to the effective dose received by a person. The indoor occupancy factor (0.8) was also taken into account, since people spend about 80 % of their time indoors [1]. The *AED* [mSv], is estimated as

$$AED \quad D \; 8760 \; 0.8 \; 0.7 \; 10^{6} \tag{5}$$

where D (nGyh^{-1}) is the total absorbed dose rate in due to gamma radiation from materials containing radionuclides and 8760 is the number of hours every year. According to the European Commission on natural radiation protection principles of building materials, the *AED* should be within 1 mSv [44].

RESULTS AND DISCUSSION

Specific activity

The range, mean value and standard deviation (SD), of ^{226}Ra, ^{232}Th, and ^{40}K concentrations in the investigated samples has been measured and presented in tab. 1. From tab. 1 we can see that the activity concentrations of ^{226}Ra in red-clay brick samples vary from 30.2 to 36.7 Bqkg^{-1}. The average activity concentrations of ^{226}Ra in clay samples from brickyards in Zhaoyuan, Liuwan and Shahezi are 33.6, 33.8 and 36.0 Bqkg^{-1} respectively, which all slightly exceed that of the worldwide population-weighted average values (32 Bqkg^{-1}) for soil [1]. The mean activity concentrations of ^{232}Th in red-clay brick samples of the brickyards in Zhaoyuan, Liuwan and Shahezi are 61.6, 61.2 and 64.6 Bqkg^{-1} respectively. The minimum 59.3 Bqkg^{-1} appeared in two samples from the brickyards in Zhaoyan and Liuwan and the maximum 65.8 Bqkg^{-1} appeared in a sample from the brickyard in Shahezi. The average concentrations of ^{232}Th in red-clay brick samples of all

Table 1. Activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in red-clay brick and raw materials of Shangluo, China

Materials	Brickyard	Activity of concentration [Bqkg^{-1}]								
		^{226}Ra			^{232}Th			^{40}K		
		Range	Mean	SD	Range	Mean	SD	Range	Mean	SD
Clay	Zhaoyuan	29.7-33.2	31.3	1.4	57.7-61.6	59.2	1.5	699.5-715.1	705.4	6.0
	Liuwan	31.1-33.6	32.8	1.0	55.2-59.1	56.6	1.4	651.8-679.6	662.3	11.6
	Shahezi	26.0-28.2	27.2	0.7	58.5-60.4	59.6	0.9	647.1-678.8	600.6	12.2
Coal	Zhaoyuan	34.4-39.7	37.1	1.9	40.3-43.3	42.0	1.2	121.3-157.0	139.5	12.2
	Liuwan	36.1-40.1	38.2	1.5	39.7-43.3	41.6	1.4	122.8-148.5	138.3	10.7
	Shahezi	37.6-41.4	39.6	1.3	39.8-42.1	40.9	0.8	153.0-176.9	166.7	9.4
Unfired brick	Liuwan	25.2-27.4	26.2	1.0	57.8-60.4	58.8	0.9	682.4-725.2	694.0	16.6
	Shahezi	24.9-29.6	27.2	1.8	58.5-61.0	59.9	1.0	638.4-661.9	648.8	8.8
Red-clay brick	Zhaoyuan	30.2-36.4	33.6	2.6	59.3-63.3	61.6	1.3	692.2-755.1	721.0	20.7
	Liuwan	32.1-34.9	33.8	1.0	59.3-62.6	61.2	1.1	711.3-754.9	724.5	15.7
	Shahezi	35.5-36.7	36.0	0.5	60.8-65.8	64.6	1.9	688.9-710.3	695.5	7.7

brickyards exceed the worldwide population-weighted average values (45 Bqkg^{-1}) for soil [1]. As for ^{40}K, the mean activity concentrations are 721.0, 724.5, and 695.5 Bqkg^{-1} respectively from brickyards in Zhaoyuan, Liuwan and Shahezi. The activity concentrations of ^{40}K obtained for all red-clay brick samples exceeded the worldwide population-weighted average values (420 Bqkg^{-1}) for soil [1]. The 755.1 Bqkg^{-1} in a sample from the brickyard in Zhaoyuan is the maximum value and 688.9 Bqkg^{-1} in a sample from the brickyard in Shahezi is the minimum value.

Table 1 also showed the radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in clay, coal and unfired brick samples. The average concentration of ^{226}Ra in these studied samples decreases in the order of coal > red-clay brick > clay > unfired brick. For ^{232}Th the concentration decreases in the order of red-clay brick > unfired brick > clay > coal. As for ^{40}K, the order is the same as for ^{232}Th. Figure 1 shows that ^{40}K is the main contributor to the total activity for all samples. It contributed 88.1 % in clay and 65.0 % in coal. In unfired bricks and red-clay bricks it occupied 88.6 % and 88.0 %. For the ^{226}Ra, it accounts for 4.1 %, 3.5 % and 4.3 % in clay, unfired bricks and red-clay bricks. But in the coal, the ^{226}Ra contributed 16.8 % to the total activity, higher than in other samples. As for ^{232}Th, it contributed 18.2 % in coal, and 7.8 %, 7.8 %, 7.7 % in clay, unfired bricks and red-clay bricks.

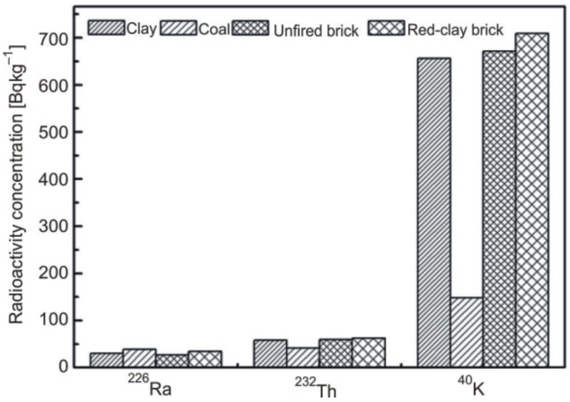

Figure 1. Radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in red-clay brick, unfired brick, clay and coal of Shangluo, China

Radiological hazard assessment results

The Ra_{eq}, H_{ex}, H_{in}, D, and AED for the red-clay bricks in Shangluo, China are showed in tab. 2. Table 2 indicates that the Ra_{eq} varies from 167.0 Bqkg^{-1} in the brickyard in Zhaoyuan to 184.7 Bqkg^{-1} in the brickyard in Shahezi. There are no obvious variations in the Ra_{eq} of the red-clay bricks from the three different brickyards. All of the Ra_{eq} are much lower than the recommended value of 370 Bqkg^{-1}. Table 3 shows the comparison of activity concentrations and radium equivalent activity of red-clay bricks with similar studies from some other cities or countries. The comparison result showed that the Ra_{eq} obtained in the present study is lower than that from most of Chinese cities except Baoji [9]. Compared with those from other countries, the result is lower than that from Algeria [45], Aden, Yemen [38], Bangladeshi [46], and Bitlis, Turkey [35], but higher than that from Isparta, Turkey [33], Egypt [18, 21], Punjab Province, Pakistan [27], Cuba [47], and Tamilnadu, India [41]. The diversity of radium equivalent activity in red-clay bricks may be related to the geological locations and geochemical properties of those materials.

The calculated values of H_{ex} for the red-clay bricks range from 0.46 in the brickyard in Zhaoyuan to 0.50 in the brickyard in Shahezi. It can be determined that all of them are less than unit and no significant difference between the three brickyards exists. The obtained results of H_{in} for the red-clay brick samples range 0.54 in the brickyard in Zhaoyuan to 0.60 in the brickyard in Shahezi with an average of 0.58. All these values are less than unit and in agreement with the recommended limit. The averaged values of D for the studied red-clay bricks are 156.32, 156.36, and 159.77 nGyh^{-1} in the brickyards in Zhaoyuan, Liuwan and Shahezi. The total average value of the indoor air absorbed dose rate from three brickyards is 1.87 times higher than the world population-weighted average indoor absorbed gamma dose rate of 84 nGyh^{-1} [1]. The calculated values of AED in the studied red-clay

Table 2. The calculated value of the radiological hazard index in the red-clay brick of Shangluo, China

Brickyard	Statistics	Ra_{eq} [Bqkg^{-1}]	H_{ex}	H_{in}	D [nGyh^{-1}]	AED [mSv]
Zhaoyuan	Min	167.0	0.46	0.54	150.14	0.74
	Max	180.1	0.49	0.58	158.87	0.78
	Mean SD	177.2 3.7	0.48 0.01	0.57 0.02	156.32 3.24	0.77 0.02
Liuwan	Min	175.0	0.47	0.57	154.83	0.76
	Max	179.7	0.49	0.57	158.76	0.78
	Mean SD	177.1 1.6	0.48 0.00	0.57 0.00	156.36 1.36	0.77 0.01
Shahezi	Min	175.5	0.47	0.57	154.66	0.76
	Max	184.7	0.50	0.60	162.98	0.80
	Mean SD	181.9 3.3	0.49 0.01	0.59 ± 0.01	159.77 2.64	0.78 0.01

Table 3. Comparison of the activity concentration and Ra_{eq} of red-clay brick with other cities or countries

City, country	Activity concentration [Bqkg^{-1}]			Ra_{eq} [Bqkg^{-1}]	References
	^{226}Ra	^{232}Th	^{40}K		
Xi'an, China	58.6	50.4	713.9	185.6	[8]
Baoji, China	37.9	46.5	697.4	158.0	[9]
Baotou, China	46	56	846	190	[12]
Xianyang, China	51.6	60.2	715.9	192.8	[10]
Urumqi, China	49.3	44.5	860.4	179	[13]
Yan'an, China	47.3	61.6	821.8	199	[6]
Weinan, China	124.7	28.9	390.2	196.1	[11]
Xiangyang, China	42.4	63.1	655.0	183.1	[16]
Xining, China	55.2	58.4	937.8	204.2	[15]
Anhui Province, China	56.2	63.8	535	188	[17]
Bangladeshi	57.5	75.8	1080.3	248.9	[46]
Egypt	31.5	25.5	298	90.8	[18]
Algeria	65	51	675	190	[45]
Cuba	57	12	857	140	[47]
Punjab Province, Pakistan	23	35	431	106	[27]
Isparta, Turkey	58.9	11.7	248.8	94.81	[33]
Aden, Yemen	54.8	37.32	1256.02	204.9	[38]
Egypt	23.06	23.11	447.84	90.59	[21]
Bitlis, Turkey	123.9	42.06	1160.4	273.4	[35]
Shangluo, China	34.5	62.5	713.7	178.7	This study

brick samples range from 0.74 mSv in the brickyard in Zhaoyuan to 0.80 mSv in the brickyard in Shahezi as shown in tab. 2. All of the effective doses are below the dose criterion of 1 mSv [44].

CONCLUSIONS

The specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in red-brick from Shangluo range from 30.2 to 36.7, 59.3 to 65.8 and 688.9 to 755.1 Bqkg^{-1}, respectively, which are found to be in the range of Chinese soil values. The radioactivity of unfired bricks, and the materials for brick-making, clay and coal have also been measured. These results are compared with that in red-clay bricks. The radium equivalent activity levels in all studied red-clay bricks are far below the limited value of 370 Bqkg^{-1}. The external and internal hazard indexes, the indoor air absorbed dose rate, the *AED* rate of red-clay bricks manufactured in Shangluo, China were determined. Calculations of external and internal hazard indices showed that no studied samples exceeded the recommended exemption levels. So, the red-clay bricks in Shangluo, China can be used as building materials in the construction industry safely.

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AUTHORS' CONTRIBUTIONS

The idea for this study was put forward by X. Lu. The brick sampling and sample experimental execution were carried out by S. Zhuang. Both of the authors contributed to the literature research, data analysis and the preparation of the manuscript.

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ПРИРОДНА РАДИОАКТИВНОСТ И РАДИОЛОШКА ОПАСНОСТ ОД ЦРВЕНО-ГЛИНЕНЕ ОПЕКЕ ПРОИЗВЕДЕНЕ У ШАНГЛУУ, КИНА

Радиолошка опасност грађевинских материјала начињених од глине, стена и другог минералног отпада привукла је већу пажњу због садржаја природних радионуклида (^{226}Ra, ^{232}Th, и ^{40}K). Измерена је концентрација активности радионуклида у узорцима опеке од црвене глине добијеним из три различите циглане у Шанглуу у Кини. За процену опасности од зрачења за људе коришћени су различити индекси, укључујући радијум еквивалентну активност, Ra_{eq}, индекс спољашње опасности, H_{ex}, индекс унутрашње опасности, H_{in}, јачину апсорбоване дозе ваздуха у затвореном простору, *D,* и годишњу ефективну дозу, *AED*. Просечне концентрације активности од ^{226}Ra, ^{232}Th и ^{40}K биле су 34.5 1.9 Bqkg^{-1}, 62.5 2.1 Bqkg^{-1}, и 713.7 19.8 Bqkg^{-1}, за испитиване црвено-глинене опеке. Вредности Ra_{eq} узорака црвене глинене опеке варирале су од 167.0 то 184.7 Bqkg^{-1}, што је ниже од границе од 370 Bqkg^{-1}. Штавише, утврђене су и концентрације активности природних радионуклида у непеченој цигли, глини и угљу и резултати су упоређени са онима у узорцима црвене глинене опеке. Ова истраживања показују да се опеке од црвене глине произведене у Шанглуу могу безбедно користити у грађевинској индустрији.

Кључне речи: природна радиоактивност, радијум еквивалента активност, радиолошка опасност, опека од црвене глине