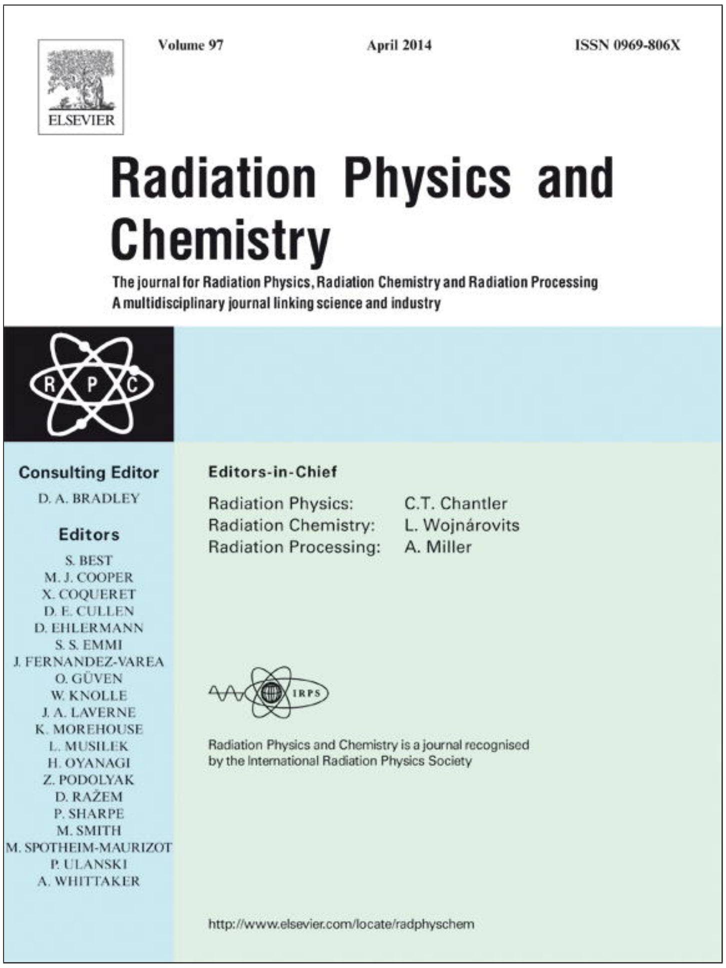
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Gamma spectrometric characterization of refractory products used in Turkey

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HIGHLIGHTS

- The study determines the radiometric characterization of refractory products.
- The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in refractory products are measured by the gamma-ray spectrometer.
- Refractory magnesite brick samples contain low levels of ^{226}Ra ^{232}Th and ^{40}K.
- Refractory high alumina brick samples contain high levels of ^{226}Ra and ^{232}Th depending on the raw materials.
- Refractory concrete samples contain high levels of ^{226}Ra and ^{232}Th depending on the raw materials.

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ABSTRACT

This is the first detailed study related to the radiometric characterization of refractory materials produced for use in the industries of iron–steel, cement and glass manufacturing in Turkey. In this study, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K naturally occurring in 52 refractory material samples collected from Turkey's leading refractory factories were measured by using a gamma spectrometer with HPGe detector. Results show that the natural radioactivity of refractory material samples varies depending on raw material. The values of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the refractory brick and concrete samples varied from 1.3 to 384.2 Bq kg^{-1} (average: 173.5 $\pm$ 20.2 Bq kg^{-1}), 2.9 to 392.9 Bq kg^{-1} (average: 176.7 $\pm$ 20.8 Bq kg^{-1}) and 18.9 to 679.6 Bq kg^{-1} (average: 130.4 $\pm$ 23.9 Bq kg^{-1}) and 24.0 to 158.2 Bq kg^{-1} (average: 73.3 $\pm$ 10.5 Bq kg^{-1}), 25.0 to 134.4 Bq kg^{-1} (average: 76.3 $\pm$ 10.8 Bq kg^{-1}) and 31.0 to 480.0 Bq kg^{-1} (average: 172.8 $\pm$ 40.5 Bq kg^{-1}), respectively. The activity concentrations of ^{226}Ra and ^{232}Th measured in fireclay brick and high alumina brick samples are significantly higher than the average values of earth's crust.

1. Introduction

Some non-nuclear industrial activities (fossil fuel combustion, oil and gas extraction, cement production, fertilizer production, mining and metal processing, refractory products and brick manufacture, etc.) include the use of naturally occurring radioactive materials (NORM). All NORMs contain trace amounts of radionuclides in the ^{238}U, ^{235}U and ^{232}Th decay series and from ^{40}K. When these materials are processed, concentrations of these radionuclides may be much higher than those usually present in earth's crust. Therefore, working with these materials or storing them can lead to a significant increase in the exposure of workers and members of the public (Cazala et al., 2009). The two main pathways of exposure for workers and the public are external exposure to gamma radiation emitted from the radionuclides and internal exposure of the respiratory tract to alpha and beta particles due to the inhalation of the radioactive inert gas radon and the inhalation and/or ingestion of dust particles (Turhan et al., 2010). In the last ten years, the production of large amounts of NORM and the potential radiological hazards has been extensively described in studies related to the measurement of the activity concentrations of the radionuclides in NORM for the assessment of the radiological impact on workers and members of the public (Omar et al., 2004; Beddow et al., 2006; Desideri et al., 2006; IAEA, 2008; Akinci and Artir, 2008; Somlai et al., 2008; Gazineu and Hazin, 2008; El Afifi et al., 2009; Bakr 2010; Turhan et al., 2010; Chang et al., 2011; Ruyters et al., 2011; Turhan et al., 2011; Iwaoka

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and Yonehara, 2012; Gezer et al., 2012; Uğur et al., 2013; Hrichi et al., 2013).

Refractory materials are able to maintain strength, dimensional stability and chemical resistance at high temperature. Refractory materials have been produced in the form of bricks, and grouts by the use of raw materials such as alumina, silica, magnesia, zirconia (or zircon), andalusite, bauxite, carbide, clays, dolomite, kaolin, graphite, etc. Refractory materials are used in kilns, annealing and induction melting furnaces used primarily by industries manufacturing iron–steel, cement, copper, metallurgy, glass, etc. In Turkey, consumption of refractory material is approximately 295,000 t, and is expected to exceed 350,000 t in the next ten years (Erez, 2010). So far, there are several studies on the wear characteristics, thermal shock behavior and mechanical properties of refractory materials and production of different types of refractory materials in the literature (İnel, 1997; Üstünbaş et al., 1997; İnel et al., 2001; Köksal et al., 2003; Köksal and Ünlü, 2004; Yıldırım and Yılmaz, 2006; Köksal, 2009), but there has not been study for the radiometric characterization of refractory materials. The purpose of this study is to measure the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in refractory brick and concrete samples manufactured in Turkey by using the gamma-ray spectrometer with a high purity germanium detector for determining radiometric characterization of refractory materials missing in the literature.

2. Materials and method

2.1. Collection and preparation of samples

A total of 52 refractory material samples (16 fireclay bricks, 2 magnesia–chrome bricks, 4 magnesia-spinel bricks, 2 magnesia bricks, 13 high alumina bricks, 4 andalusite bricks and 11 refractory concretes) were supplied from Turkey's leading refractory factories. Refractory brick and concrete samples brought to Sample Preparation Laboratory were properly coded, taking into account the business concerns of refractory firm, as REFBRICK and REFCONC, respectively. Then each sample was crushed into small pieces by a crusher, powdered with the aid of grinding and passed through the sieve in order to bring the geometry of the samples similar to the geometry of the standard calibration source. Each powdered sample was subjected to the drying process in an oven set 110 °C for a period of 15 h to obtain net mass. Each sample was transferred to 1 L of Marinelli container and weighed to determine the net mass. Sample containers were hermitically sealed and allowed to stand for at least 4 weeks to ensure secular equilibrium between ^{226}Ra and its short-lived decay products.

2.2. Radiometric measurements

Each refractory material sample was subjected to gamma spectrometric analysis by using a high-resolution gamma-ray spectrometer with a coaxial p-type HPGe detector (Canberra GX3018) at the Radioactivity Measurement and Analysis Unit of Çekmece Nuclear Research and Training Center. The energy resolution of the HPGe detector is 1.8 keV at 1332.5 keV of ^{60}Co gamma energy with a relative efficiency of 30%. The detector was shielded with 100 mm thick lead inserted into 9.5 mm thick steel frame to minimize natural background radiation from the environment. The certificated standard calibration source of 1 L Marinelli beaker which contains multinuclide distributed in 1.0 g cm^{-3} epoxy (Eckert&Ziegler Isotope Products) was used for the energy and absolute efficiency calibration of the system in the energy range from 122 keV to 1836 keV. Each sample was counted in the same source-to-detector geometry as used for the efficiency calibration. The gamma spectrum for each sample acquired for 20,000–86,000 s was evaluated using Genie-2000 software. The gamma-ray photopeak of the 609.3 keV from ^{214}Bi, the 583.2 keV from ^{208}Tl and the 1460.8 keV were used to determine the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively. Detection limit (D_L, in term of Bq kg^{-1}) of the system with 95% confidence limit was calculated as follows (Currie, 1968):

$$D_L = \frac{1.64\sigma_B}{\varepsilon PtM} \tag{1}$$

where $\sigma_B$is the standard deviation of the background in the region of interest of gamma photopeak, ε is the absolute efficiency of the system, P is the gamma-ray probability per decay, t is the counting time and M is the mass of the sample. The detection limit average values calculated for ^{226}Ra, ^{232}Th and ^{40}K are 0.8, 2.1, 9.8 Bq kg^{-1} for refractory brick samples and 0.6, 1.2 and 8.7 Bq kg^{-1} for refractory concrete samples, respectively.

3. Results and discussion

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the refractory bricks samples and the refractory concrete samples are presented in Tables 1 and 2, respectively. Skewness values, which are a measure of symmetry, kurtosis values which are a measure of whether the data are peaked or flat relative to a normal distribution, and some statistical data of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in all the refractory samples is given in Table 3. It can be seen from Tables 1 and 2 that distribution of ^{226}Ra, ^{232}Th and ^{40}K in refractory brick and concrete samples concentration is not homogeneous and the activity concentration values vary depending on raw materials. The average values of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the refractory bricks and the refractory concretes are 173.5 ± 20.2 Bq kg^{-1} (range: 1.3–384.2 Bq kg^{-1}), 176.7 ± 20.8 Bq kg^{-1} (range: 2.9–392.9 Bq kg^{-1}) and 130.4 ± 23.9 Bq kg^{-1} (range: 18.9–679.6 Bq kg^{-1}) and 73.3 ± 10.5 Bq kg^{-1} (range: 24.0–158.2 Bq kg^{-1}), 76.3 ± 10.8 Bq kg^{-1} (range: 25.0–134.4 Bq kg^{-1}) and 172.8 ± 40.5 Bq kg^{-1} (range: 31.0–480.0 Bq kg^{-1}), respectively. From Table 1, the highest values of the activity concentration of ^{226}Ra and ^{232}Th were measured in the high alumina brick samples while the highest value of the activity concentration of ^{40}K was measured in the fireclay brick samples. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in earth's crust are determined as 32, 45 and 412 Bq kg^{-1}, respectively (UNSCEAR, 2008). Comparison of the average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the refractory material samples with the average values of the activity concentrations measured in earth's crust is given in Fig. 1. The ^{226}Ra concentration in the high alumina brick samples is significantly (8–12 times) higher than the average value for earth's crust. Also, the ^{232}Th concentration in the high alumina brick samples is 5–9 times higher than the average value for earth's crust. The average activity concentration of ^{226}Ra in the fireclay brick samples is 4 times higher than the average value for earth's crust while the average value in the refractory concrete samples is twice higher than the quoted average value. The average activity concentrations of ^{232}Th in the fireclay brick and concrete samples are twice higher than the average value for earth's crust. However, the ^{226}Ra and ^{232}Th activity concentrations measured for the magnesite brick samples are significantly lower than the average values for earth's crust. Also, all values of the activity concentrations of ^{40}K, except for REFRRICK6 and REFBRICK41 are lower than the mean value in earth's crust.

4. Conclusion

Natural radionuclide characterization of the refractory product samples manufactured in Turkey was determined by gamma-ray

Table 1
The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the refractory bricks.

Refractory material	Sample code	Activity concentration (Bq kg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
Fireclay brick	REFBRICK1	103.3 ± 6.9	125.9 ± 14.2	61.0 ± 6.1
	REFBRICK2	157.1 ± 20.8	133.1 ± 8.3	25.4 ± 5.3
	REFBRICK3	116.7 ± 13.6	107.7 ± 7.3	64.6 ± 5.9
	REFBRICK4	140.3 ± 15.6	117.5 ± 9.5	385.8 ± 24.4
	REFBRICK5	172.4 ± 11.4	206.8 ± 14.7	169.8 ± 14.0
	REFBRICK6	105.7 ± 3.4	124.8 ± 6.3	679.6 ± 34.0
	REFBRICK7	151.2 ± 8.4	232.2 ± 12.0	92.9 ± 21.6
	REFBRICK8	154.7 ± 15.6	152.7 ± 8.5	44.0 ± 6.0
	REFBRICK9	171.5 ± 11.0	156.6 ± 8.4	45.0 ± 7.0
	REFBRICK10	108.6 ± 8.4	129.8 ± 5.4	120.0 ± 23.0
	REFBRICK11	151.7 ± 9.5	169.5 ± 6.3	253.0 ± 6.0
	REFBRICK12	192.7 ± 4.4	243.9 ± 5.3	86.1 ± 9.6
	REFBRICK13	195.0 ± 13.1	197.5 ± 8.7	280.0 ± 21.0
	REFBRICK14	190.1 ± 4.2	196.8 ± 8.0	57.9 ± 15.7
	REFBRICK15	195.8 ± 9.9	153.8 ± 8.1	237.0 ± 24.0
	REFBRICK16	194.0 ± 10.0	151.0 ± 11.0	247.0 ± 21.2
Magnesite–chrome brick	REFBRICK17	10.5 ± 3.4	4.1 ± 0.3	54.3 ± 3.1
	REFBRICK18	12.7 ± 3.1	3.5 ± 0.2	49.0 ± 0.4
Magnesite-spinel brick	REFBRICK19	2.7 ± 0.6	9.2 ± 1.4	53.5 ± 3.8
	REFBRICK20	1.3 ± 0.5	3.4 ± 0.5	58.8 ± 3.6
	REFBRICK21	1.4 ± 0.4	3.1 ± 0.5	49.7 ± 3.8
	REFBRICK22	2.4 ± 0.5	3.2 ± 0.4	58.2 ± 3.9
Magnesite brick	REFBRICK23	6.2 ± 0.6	3.3 ± 0.4	50.1 ± 3.0
	REFBRICK24	6.6 ± 0.6	2.9 ± 0.6	49.3 ± 2.8
High alumina brick	REFBRICK25	331.7 ± 20.8	252.8 ± 14.3	18.9 ± 4.0
	REFBRICK26	315.8 ± 23.9	245.7 ± 14.1	19.0 ± 4.2
	REFBRICK27	315.3 ± 11.2	321.1 ± 13.4	62.9 ± 8.2
	REFBRICK28	384.2 ± 16.1	379.2 ± 17.4	138.2 ± 20.0
	REFBRICK29	369.9 ± 15.0	392.9 ± 9.9	81.4 ± 23.2
	REFBRICK30	360.8 ± 11.2	385.2 ± 11.4	95.0 ± 24.0
	REFBRICK31	342.2 ± 10.2	351.4 ± 15.1	61.1 ± 3.2
	REFBRICK32	351.7 ± 8.2	370.4 ± 7.4	56.1 ± 11.1
	REFBRICK33	361.4 ± 10.7	384.6 ± 11.2	92.0 ± 24.0
	REFBRICK34	342.2 ± 10.2	351.4 ± 15.1	61.1 ± 3.2
	REFBRICK35	269.9 ± 12.4	308.7 ± 13.2	84.0 ± 15.0
	REFBRICK36	301.8 ± 4.4	318.9 ± 9.7	122.5 ± 11.4
	REFBRICK37	321.8 ± 3.3	354.5 ± 6.4	84.8 ± 7.2
Andalusite brick	REFBRICK38	27.0 ± 2.3	27.8 ± 6.5	27.4 ± 5.0
	REFBRICK39	20.3 ± 4.0	28.8 ± 6.1	21.4 ± 5.0
	REFBRICK40	64.6 ± 4.0	59.9 ± 3.0	403.0 ± 38.0
	REFBRICK41	88.4 ± 10.8	77.6 ± 6.4	643.8 ± 59.2

Table 2
The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the refractory concretes.

Refractory material	Sample code	Activity concentration (Bq kg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
Concrete	REFCONC1	50.0 ± 17.0	27.0 ± 5.0	480.0 ± 24.0
	REFCONC2	52.1 ± 4.1	78.5 ± 2.8	88.0 ± 7.0
	REFCONC3	67.3 ± 3.8	127.0 ± 3.8	31.0 ± 2.3
	REFCONC4	91.2 ± 2.3	81.6 ± 3.9	132.1 ± 8.3
	REFCONC5	98.2 ± 5.3	95.2 ± 4.1	90.2 ± 7.9
	REFCONC6	158.2 ± 9.0	134.4 ± 5.7	31.5 ± 2.2
	REFCONC7	60.6 ± 2.4	50.2 ± 2.1	240.4 ± 12.2
	REFCONC8	55.6 ± 3.5	62.9 ± 3.3	311.1 ± 14.2
	REFCONC9	80.6 ± 4.9	60.3 ± 3.5	217.1 ± 10.2
	REFCONC10	68.7 ± 5.3	97.1 ± 7.8	174.2 ± 33.4
	REFCONC11	24.0 ± 1.0	25.0 ± 2.0	105.0 ± 16.0

spectrometric technique. The ^{226}Ra, ^{232}Th and ^{40}K activity concentrations measured for the examined refractory samples were tabulated and compared with the average values for earth's crust. This study can be used as a reference for more extensive studies of the NORM. From the results of the activity concentrations, the following outcomes may come into prominence:

Table 3
Statistical data of ^{226}Ra, ^{232}Th and ^{40}K measured in all refractory material samples.

	Refractory brick samples			Refractory concrete samples		
	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K
Average	173.5	176.7	130.4	73.3	76.3	172.8
Standard error	20.2	20.8	23.9	10.5	10.8	40.5
Median	157.1	153.8	62.9	67.3	78.5	132.1
Standard deviation	129.1	133.1	153.1	34.9	36.0	134.3
Kurtosis	−1.3	−1.2	5.9	3.1	−0.7	1.6
Skewness	0.2	0.2	2.4	1.4	0.2	1.3
Min	1.3	2.9	18.9	24.0	25.0	31.0
Max	384.2	392.9	679.6	158.2	134.4	480.0

– magnesite brick samples contain low levels of ^{226}Ra, ^{232}Th and ^{40}K;
– high alumina brick, fireclay brick and concrete samples contain high levels of ^{226}Ra and ^{232}Th depending on the raw materials.

Therefore the production, handling and use of these refractory materials shall be subject to the national safety regulation. In order to reduce the inhalation exposure to workers of these radionuclides, dust control should be implemented.

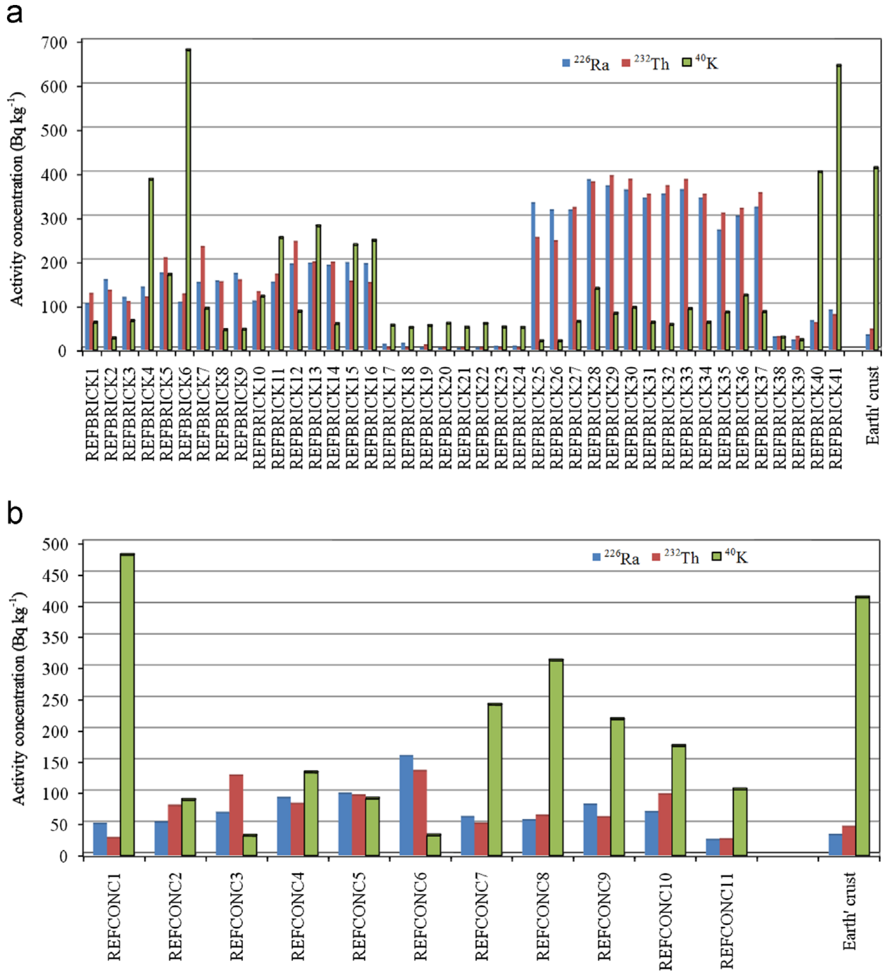

Fig. 1. Comparison of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the refractory brick (a) and refractory concrete (b) samples with the average values measured in earth's crust.

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