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Radiometric analysis of raw materials and end products in the Turkish ceramics industry

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ABSTRACT

This study presents the findings of radiometric analysis carried out to determine the activity concentrations of natural radionuclides in raw materials (clay, kaolin, quartz, feldspar, dolomite, alumina, bauxite, zirconium minerals, red mud and frit) and end products (glazed ceramic wall and floor tiles) in the Turkish ceramics industry. Hundred forty-six samples were obtained from various manufacturers and suppliers throughout the country and analyzed using gamma-ray spectrometer with HPGe detectors. Radiological parameters such as radium equivalent activity, activity concentration index and alpha index were calculated to assess the radiological aspects of the use of the ceramic end products as decorative or covering materials in construction sector. Results obtained were examined in the light of the relevant national and international legislation and guidance and compared with the results of similar studies reported in different countries. The results suggest that the use of ceramic end product samples examined in the construction of dwellings, workplaces and industrial buildings in Turkey is unlikely to give rise to any significant radiation exposure to the occupants.

1. Introduction

Building materials originating from rock and soil contain variable quantities of natural radionuclides in the uranium–radium (^{238}U–^{226}Ra) and thorium (^{232}Th) decay series and the radioactive isotope of potassium (^{40}K), with concentrations depending upon the geologic and geochemical characteristics of the source materials. These natural radionuclides in building materials are sources of external and internal exposure in dwellings, workplaces and industrial buildings. External radiation exposure is caused by the gamma radiation. Internal radiation exposure, mainly affecting the respiratory tract, is due to the short-lived decay products of radon (^{222}Rn, a daughter product of ^{226}Ra), which are exhaled from building materials into room air. The precursors of ^{226}Ra in the ^{238}U series are generally ignored because 98.5% of the radiological effects of the ^{238}U series are produced by the ^{226}Ra and its daughter products (Beretka and Mathew, 1985). Measurement of the activity concentrations of the radionuclides in building materials is important to assess the possible radiological hazards to human health and to develop standards and guidelines for the use and management of these materials.

Ceramics can be defined as inorganic, non-metallic materials and are made of mixtures of raw materials that are crushed to powder, press molded and calcined at high temperature (up to 1250 °C) to form a ceramic. Local raw materials are sufficient to satisfy the demand of the Turkish ceramic industry, except that zircon and baddeleyite (zirconia) used as opacifier and pigment in glazes (to 5–13% of the finished products) are mainly imported from Italy. Zirconia is also used when the material must withstand extremely high temperatures.

Ceramic wall and floor tiles are commonly used as coverings or decorative building materials in bathrooms, toilets and kitchens in Turkey. The Turkish ceramic industry has the 5th largest production of ceramic tiles in the world, the major part of which is ceramic wall and floor tiles (DPT, 2001).

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in ceramic raw materials and end products have been reported in the literature (Beretka and Mathew, 1985; Chowdhury et al., 1998; Bruzzi et al., 2000; Yahong et al., 2002; Xinwei, 2004; Ballesteros et al., 2008; TAEK TR 2008-7, 2008; Righi et al., 2009; Turhan, 2009). However the detailed information about the natural radioactivity of the ceramic raw materials and end products in Turkey is not available in the literature. In this study, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the samples of ceramic raw materials (clay, kaolin, quartz, feldspar, dolomite, alumina, bauxite, zircon, baddeleyite, red mud and frit) and end products (ceramic wall and floor tiles) were determined using gamma

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spectrometry with HPGe detectors. Radium equivalent activity, activity concentration index (for the external exposure) and alpha index (for the internal exposure) were also calculated to assess radiological hazard from the ceramic end products used in construction sector. The results were presented and discussed on the basis of relevant national and international legislation and guidance and also compared with the corresponding results of ceramic raw materials end products of different national origin in the literature.

2. Material and methods

2.1. Sample preparation for gamma ray spectrometry

Samples weighing from 1 to 2 kg were obtained from manufacturers and suppliers in Turkey. A total of 146 samples were examined: (a) 98 ceramic raw materials (36 clay, 17 kaolin, 13 quartz, 9 sodium feldspar, 3 potassium feldspar, 4 dolomite, 2 alumina, 5 bauxite, 3 zircon, 3 baddeleyite, 2 red mud and 1 frit samples) and (b) 48 ceramic end products (25 wall tiles and 23 floor tiles).

The samples were pulverized and then dried in a temperature-controlled furnace at 110 °C for 10–15 h to remove moisture. The samples were filled into cylindrical plastic containers or 1 L Marinelli beakers, weighted, hermetically sealed and stored for four weeks before counting to allow reach the secular equilibrium between ^{226}Ra and ^{222}Rn and its short-lived decay products.

2.2. Gamma ray spectrometer and counting

Two high-resolution gamma ray spectrometers connected with p-type coaxial HPGe detectors were used to measure the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the samples of ceramic raw materials and end products. One of the HPGe detectors has a relative efficiency of 50% and an energy resolution of 1.8 keV at 1332.5 keV of ^{60}Co, while the other detector has a relative efficiency of 11.4% and an energy resolution of 1.96 keV. The detectors were shielded to reduce gamma-ray background. Full energy peak efficiency calibration of the gamma spectrometry systems has been determined using a solid nuclide mixture of gamma reference materials sealed in a standard Marinelli beakers with an active volume of 1000 mL, average densities of 1 and 1.7 g cm^{-3} and IAEA reference materials RGU-1 (U-ore), RGTh-1 (Th-ore) and RGK-1 (K_2SO_4). Each sample was measured for an accumulating time between 5 and 24 h. Background measurements were taken under the same conditions of sample measurements and subtracted in order to get net counts for the sample.

The activity concentrations were averaged from gamma-ray peaks at several energies assuming secular equilibrium in the ^{232}Th decay series and between ^{226}Ra and its short live decay products. The gamma-ray peak of the 351.9 keV from ^{214}Pb and the 609.3 keV from ^{214}Bi were used to determine the activity concentration of ^{226}Ra. The gamma-ray peaks of the 911.2 keV from ^{228}Ac and the 583.2 keV from ^{208}Tl were used to determine the activity concentration of ^{232}Th. The activity concentration of ^{40}K was measured directly by its own gamma-ray peak at 1460.8 keV.

3. Results and discussion

3.1. Natural activity concentration in raw materials and end products

The values of the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K measured in the samples together with the total statistical uncertainty are presented in Table 1 (ceramic raw materials) and Table 2 (ceramic end products). The total statistical uncertainty of the radioactivity measurements was calculated by taking into consideration the systematic uncertainties in the efficiency calibration and counting statistical uncertainty. Table 3 gives the reported activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for ceramic raw materials and end products compiled from the literature. The mean activity concentrations measured in the

Table 1
The range (maximum and minimum) and mean of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the samples of ceramic raw materials.

Raw materials	N		Activity concentration (Bq kg^{-1} ± 1σ)		
			^{226}Ra	^{232}Th	^{40}K
Clay	36	Range	8.6 ± 0.5–141.0 ± 11.2	4.0 ± 0.9–176.3 ± 11.8	63.3 ± 7.5–1103.6 ± 66.7
		Mean ± SE[a]	45.2 ± 5.4	51.6 ± 5.7	571.9 ± 40.9
Kaolin	17	Range	17.5 ± 1.6–130.5 ± 9.3	23.4 ± 2.2–180.8 ± 13.3	17.1 ± 1.8–1948.7 ± 163.4
		Mean ± SE	80.3 ± 9.8	89.2 ± 11.1	494.8 ± 152.1
Quartz	13	Range	3.1 ± 0.3–43.4 ± 4.1	2.9 ± 0.2–44.8 ± 4.2	49.5 ± 5.9–485.8 ± 62.4
		Mean ± SE	13.7 ± 3.1	11.3 ± 3.2	221.7 ± 47.6
Na-Feldspar	9	Range	1.7 ± 0.3–41.5 ± 7.3	9.0 ± 1.1–66.9 ± 5.6	11.5 ± 3.7–661.0 ± 55.7
		Mean ± SE	22.5 ± 3.9	46.4 ± 7.8	160.0 ± 66.0
K-Feldspar	3	Range	3.4 ± 0.6–27.8 ± 2.4	0.9 ± 0.2–6.3 ± 0.9	1766.0 ± 151.5–3633.9 ± 324.6
		Mean ± SE	13.0 ± 7.5	3.8 ± 1.6	2457.8 ± 591.1
Dolomite	4	Range	4.8 ± 0.5–15.0 ± 1.5	2.0 ± 0.2–4.7 ± 0.4	27.0 ± 2.3–78.9 ± 8.5
		Mean ± SE	10.2 ± 2.5	3.3 ± 0.6	51.4 ± 11.4
Alumina	2	Range	271.8 ± 16.4–327.5 ± 21.8	296.4 ± 21.1–307.3 ± 22.1	44.9 ± 4.9–45.0 ± 5.3
		Mean ± SE	299.6 ± 27.8	301.8 ± 5.5	44.9 ± 0.1
Bauxite	5	Range	15.5 ± 1.2–405.0 ± 33.6	19.9 ± 1.7–414.0 ± 34.8	12.8 ± 1.0–111.6 ± 13.0
		Mean ± SE	235.3 ± 89.7	243.4 ± 91.2	74.3 ± 16.7
Zircon	3	Range	1667.1 ± 127.7–2312.0 ± 199.5	257.4 ± 23.7–417.1 ± 34.8	57.0 ± 7.5–269.8 ± 30.5
		Mean ± SE	1973.4 ± 186.9	354.2 ± 49.1	131.9 ± 69.0
Baddeleyite	3	Range	3203.0 ± 262.6–5563.0 ± 447.5	425.0 ± 29.8–657.0 ± 58.4	46.0 ± 5.1–108.0 ± 11.2
		Mean ± SE	4037.0 ± 764.1	578.7 ± 76.8	77.3 ± 17.9
Red mud	2	Range	128.1 ± 7.3–285.0 ± 48.7	342.0 ± 23.4–357.4 ± 24.3	94.0 ± 8.4–110.4 ± 10.4
		Mean ± SE	206.6 ± 78.5	349.7 ± 7.7	102.2 ± 8.2
Frit	1		394.1 ± 30.8	84.9 ± 10.1	13.9 ± 4.0

[a] SE: Standard error.