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Investigation on ^{226}Ra, ^{238}Th, ^{40}K and ^{137}Cs Concentrations in Common Polishing Materials Consumed by Inhabitants in Saudi Arabia

Wedad Rayif Alharbi

Department of Physics, Faculty Science, King Abdulaziz University, Jeddah, Saudi Arabia
Email: walharbi@kau.edu.sa

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Abstract

Knowledge of radioactivity present in polishing materials enables one to assess any possible radiological risks to human health. In this work, the radioactivity due to the presence of ^{226}Ra, ^{232}Th, ^{40}K, and ^{37}Cs has been measured in polishing materials consumed in Saudi Arabia using gamma spectrometry with HPGe. The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs ranged from 13.61 ± 0.005 to 0.60 ± 0.002, 18.43 ± 0.003 to 0.78 ± 0.001, 342.59 ± 0.009 to 2.47 ± 0.001 and 1.47 ± 0.001 to 0.55 ± 0.001 Bq/l, respectively. For ^{226}Ra and ^{40}K, the highest values are measured in stainless steel polisher. The highest values also measured in metal polisher (copper-chrome) and disinfectant cleaner and polisher sample for ^{232}Th and ^{137}Cs. Radium equivalent activity due to the natural radioactivity of the investigated samples ranged from 51.37 to 1.24 Bq/l. This value is less than the recommended values in the established standards. The evaluated data were compared with the literature data. Our results indicate that no significant radiological hazards arise from using investigated samples hence quite safe to be used as polishing materials.

Keywords

Gamma Spectrometry, Radiation Hazard, Polishing Materials, Natural Radioactivity

1. Introduction

Naturally occurring radioisotopes (^{238}Th, ^{232}U series and ^{40}K) are the main source of both external and internal radiation exposure in humans, whence the radiation finds its way into building materials, air, water, food, and eventually the human body. Human beings are exposed to external radiation from cosmic rays and terrestrial radiation. These isotopes enter the human body by intake from food and inhalation and result in exposure to internal radiation [1] [2]. According to the report by UNSCEAR, the total exposure per person resulting from ingestion of terrestrial radioisotopes was 0.3 mSv, of which 0.17 mSv comes from ^{40}K and 0.12 mSv comes from thorium and uranium series. Exposure from inhalation of terrestrial radioisotopes contributes another 0.01 mSv. Thus, the internal dose of terrestrial radioisotopes can be estimated from the concentrations of natural radioisotopes in foods [3]. Internal radiation exposure, mainly affecting various levels of the respiratory tract, is due to the deposition of short lived progeny products of radon, which are exhaled from building material into indoor air [4] [5]. Radon (^{222}Rn), a gaseous product of decay of ^{226}Ra, is extremely important in an indoor exposure. Radon easily diffuses from the ground and building materials to indoor air and is the source of exposing bronchus and lungs [6]-[8]. The isotope ^{137}Cs is produced anthropogenically by several types of nuclear activity, including past testing of nuclear weapons, accidents in nuclear facilities, reprocessing of spent nuclear fuel and nuclear power reactors. All these materials contain some amount of natural radionuclides that cause exposure of people to ionizing radiation. The measurement of activity concentrations of radionuclides in polishing materials is important in the assessment of population exposures, as most individuals spend 80% of their time indoors [9]. Long exposures to low levels of ionizing radiation and poor ventilation of residential buildings can seriously increase health risks to polishing materials [10]. Polishing materials have a variety of chemical natures and some of them are toxic to humans and may cause sickness including cancers and death [11]. Even nontoxic polishing materials can pollute the land or water. Therefore, care needs to be exercised when using and disposing such materials [12]. The present study was undertaken with the purpose of determining natural radioactivity in some polishing materials. The data obtained are essential for the development of standards and guidelines concerning the use and management of polishing materials.

2. Sampling and Measurements

Twenty-two samples representing commercial species of the most commonly consumed polishing materials were collected from Saudi Arabia local market. They were being used for cleaning and polishing purposes of marble, granite, ceramic, tiles, stucco, brick, porcelain, chrome and metal, houses and vehicles. Weighed samples were stored in standard polyethylene Marinelli beakers of 650 cm^3 volume each. The beakers were tightly sealed for 4 - 5 weeks to reach secular equilibrium where the rate of decay of the progeny becomes equal to that of the parent (radium and thorium) [13]. **Table 1** shows samples description.

3. Gamma Spectrometric Measurements

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were measured with gamma-spectrometric system based on HPGe. The samples were counted using a gamma spectrometry (Canberra coaxial-type Model GC2520) with relative efficiency of 25% and resolution FWHM of 0.2 keV for 1332.5 keV gamma-ray peak of ^{60}Co and a peak to

Table 1. Sample code and their description.

Sample code	Sample description	Country
P1	Marble and ceramic cleaner	Saudi Arabia
P2	Super thick cleaner with Bleach	Saudi Arabia
P3	Ceramic cleaner, disinfectant and polisher	Saudi Arabia
P4	Eliminator bad odors cleaner and polisher	Italy
P5	Stainless steel cleaner and polisher	U.S.A
P6	Metal polisher (copper-chrome)	Nederland
P7	Antiseptic disinfectant	U.A.E
P8	Disinfectant, cleaner and polisher	Saudi Arabia
P9	Super polisher for all paint types including metallic	England
P10	High glosser, protective paintwork sealant for the perfectionist	England
P11	Rapidly remover for intensive tar, adhesive, grease, wax, oil and silicone	Britain
P12	Silver polish	Italy
P13	Tiles and surface cleaner(cement, gypsum, rust and dirt remover	Saudi Arabia
P14	Instant disinfectant, cleaner and polisher pine scent	U.A.E
P15	Instant disinfectant, cleaner and polisher pine scent	Saudi Arabia
P16	Cleaner and polisher	U.S.A
P17	Advanced cleaner and polisher	U.S.A
P18	Multi-surface cleaner and disinfectant—powerful scent	U.S.A
P19	Windows, mirrors, appliances, counters, Tiles and other smooth surface cleaner and polisher	Saudi Arabia
P20	Lime solvent from pots, tube and sinks rust remover, stains from stucco, brick, porcelain, chrome and metal-blasts calcium from glassware and tile	U.S.A
P21	Glass and household cleaner and polisher	Saudi Arabia
P22	Concentration cleaner and polisher of marble, granite, ceramic and tiles	Spain

Compton ratio of 50:1. The gamma-spectrometry system consists of HPGe connected to a desk top computer provided with a Canberra multichannel analyzer (MCA) in conjunction with a configuration software for spectrum acquisition and evaluation [14] [15].

The specific activities were averaged from gamma-ray photo peaks at several energies. The gamma-ray lines at 295.2 and 351.9 keV from ^{214}Pb and at 609.3 and 1764.5 keV from ^{214}Bi were used to determine the specific activity of ^{226}Ra. The gamma ray lines of 338.4 keV; 911.2 keV and 968.97 keV from ^{228}Ac, the 238.58 keV and 727.3 keV from ^{212}Bi and 583.2 keV from ^{208}Tl were used to determine the specific activity of ^{232}Th. The specific activities of ^{40}K and ^{137}Cs were measured directly by its own gamma-ray lines at 1460.8 and 661.66 keV, respectively [13]. The minimum detectable amount (BDLs) for ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were 0.312 ± 0.16, 0.340 ± 0.16, 1.660 ± 1.10 and

0.45 ± 0.06 Bq/l respectively. Radioactivity concentrations of each sample were measured for about 23 h. The data acquisition, display and spectrum analysis were carried out using a dedicated software program [16], which enabled also the concentrations of ^{226}Ra, ^{232}Th and ^{40}K to be calculated. The degree of secular equilibrium reached between ^{226}Ra and ^{232}Th and their decay products was taken into account during the concentration calculations.

4. Results and Discussion

The specific activity of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs, in the samples are shown in Table 2. The highest average concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs (13.60 ± 0.0015, 18.43 ± 0.0031, 342.59 ± 0.0091and 1.47 ± 0.0005 Bq/l respectively) were found in samples P5,

Table 2. Average of ^{226}Ra,^{232}Th, ^{40}K and Ra_{eq} of polishing materials in the current study.

Sample code	Activity concentration (Bq·l^{-1})				Ra_{eq} (Bq·l^{-1})
	^{226}Ra	^{238}Th	^{40}K	^{137}Cs	
P1	1.55 ± 0.001	1.56 ± 0.004	61.46 ± 0.001	1.06 ± 0.001	8.52
P2	1.23 ± 0.001	5.19 ± 0.006	39.26 ± 0.001	ND	11.67
P3	0.60 ± 0.002	2.10 ± 0.005	ND	0.96 ± 0.001	3.71
P4	ND	1.70 ± 0.003	ND	0.72 ± 0.001	2.46
P5	13.61 ± 0.005	17.48 ± 0.011	342.59 ± 0.009	0.55 ± 0.001	51.37
P6	9.50 ± 0.003	18.43 ± 0.003	36.34 ± 0.001	0.98 ± 0.001	38.66
P7	2.82 ± 0.003	2.60 ± 0.001	40.64 ± 0.001	0.75 ± 0.001	9.65
P8	1.02 ± 0.002	5.24 ± 0.007	10.04 ± 0.001	1.47 ± 0.001	9.28
P9	4.22 ± 0.001	11.23 ± 0.005	11.74 ± 0.001	1.19 ± 0.001	21.18
P10	2.96 ± 0.002	2.24 ± 0.001	6.13 ± 0.001	0.63 ± 0.001	6.64
P11	2.27 ± 0.003	1.90 ± 0.003	5.63 ± 0.001	ND	5.43
P12	11.30 ± 0.001	3.92 ± 0.004	31.93 ± 0.001	ND	19.36
P13	0.67 ± 0.009	3.15 ± 0.005	ND	ND	5.17
P14	2.12 ± 0.006	ND	4.05 ± 0.001	ND	2.93
P15	2.27 ± 0.006	3.28 ± 0.005	2.47 ± 0.001	ND	7.16
P16	4.09 ± 0.005	2.84 ± 0.003	ND	ND	8.15
P17	ND	0.78 ± 0.001	ND	ND	1.24
P18	1.77 ± 0.005	1.60 ± 0.004	ND	ND	5.58
P19	0.79 ± 0.002	1.16 ± 0.001	ND	ND	2.46
p20	1.35 ± 0.004	1.33 ± 0.004	ND	ND	3.25
P21	1.94 ± 0.002	2.25 ± 0.003	48.88 ± 0.001	ND	8.92
P22	2.60 ± 0.002	3.70 ± 0.004	57.14 ± 0.007	ND	12.30

ND: Not detected.

P6, P5 and P8 respectively, whereas the lowest average concentrations ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs (0.60 ± 0.0015, 0.78 ± 0.0011, 2.47 ± 0.0005, 0.55 ± 0.0008 Bq/l respectively) were found in samples P3, P17, P15 and P5. The ^{40}K activity concentration dominated over that of the ^{226}Ra and ^{232}Th, as normally happens in samples, while the highest activity level of ^{40}K (342 Bq/l) which was recorded in sample P5 was lower than that of the global average of 400 Bq·kg^{-1} [17].

The variation among the activity levels in different samples may be attributed to the pH, and chemical composition of the raw materials from which they derived [18]. Some studies present the concentration values of ^{226}Ra only and do not consider the ^{238}U presence taking into account the fact that 98.5% of the radiological effects of the uranium series are produced by radium and its daughters [19].

The activity concentrations of Ra, presented here, fall within the range reported by other authors. Both Th and K in this work revealed higher concentration levels compared to those level for painting oxides and paints. However, their levels were lower than in the case of titanium enamel frits (**Table 3**). According to the recommended reference level of 30, 25 and 370 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively, for the world average concentrations published by UNSCEAR [1], it was noted that the obtained results in all samples are lower than the recommended reference level.

5. Radiological Parameters

Radium Equivalent Activities (Ra_{eq})

Uniformity of the distribution of ^{226}Ra, ^{232}Th and ^{40}K in materials with respect to radiation exposure has been described by a common index. This index is called the radium equivalent (Ra_{eq}) activity and is defined as a weighted sum of the activity concentrations of the above three radionuclides. Ra_{eq} is given in Bq·l^{-1} to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K. It was assumed that 370 Bq·kg^{-1} of ^{226}Ra, 259 Bq·kg^{-1} of ^{232}Th or 4810 Bq·kg^{-1} of ^{40}K produce the same γ-ray dose rates [20]; Ra_{eq} is calculated through the following relationship:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (1)$$

where C_{Ra}, C_{Th}, and C_K are the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, expressed in Bq·l^{-1}. The maximum value of Ra_{eq} in building materials must be less than 370 Bq·kg^{-1} for safe use of materials in the construction of buildings [1], to keep the external dose below 1.5 mSv·y^{-1} [21]. The results obtained by us showed that the lowest

Table 3. Comparison of concentrations range of ^{226}Ra, ^{232}Th, and ^{40}K (Bq·kg^{-1}) recorded in various polishing materials of several studies.

Materials	Activity concentration			References
	^{226}Ra	^{232}Th	^{40}K	
Titanium enamel frits	1.05 - 1018.57	1.05 - 1164.09	942.29 - 16,679.19	[22]
Painting oxides	2.35 - 72.96	1.76 - 12.88	2.26 - 200	[23]
Paits	10.5 - 43.0	0.5 - 0.5	0.5 - 7.7	[24]
Polishing	0.60 - 13.61	0.78 - 18.43	2.47 - 342.59	Present work

Ra_{eq} was equal to 1.24 Bq·l^{-1} in sample P17, while the highest value reached 51.37 Bq·l^{-1} in in sample P5 (Table 2). All the studied samples showed lower values than the recommended worldwide mean value 370 Bq kg^{-1}. Thus, every material used to clean and polish of marble, granite, ceramic and tile can be considered to does not reveal a significant radiological hazard.

6. Conclusion

The activity levels of the natural terrestrial radionuclides as ^{226}Ra, ^{232}Th, ^{40}K and artificial ^{137}Cs were determined using a gamma-ray spectrometry in 22 samples of polishing materials collected from local stores. The obtained values were, in general, comparable to the corresponding ones obtained from other studies, and they all fell within the average worldwide ranges. Basing on the measurement results, the values of the radium equivalent activities Ra_{eq} were calculated to assess the radiological hazard for inhabitants of Jeddah. The obtained values displayed to be below the permissible limiting values.

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