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The Experimental Gamma Radiation Dose Rate for Radiation Hazard into Adhesive Building Materials in Saudi Arabia

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Abstract

The primary aim of this work was clearly to apply the norms of radiation protection to building residents against natural radioactivity. This was done through measurement of natural radioactivity in adhesive building materials using HPGe gamma ray spectrometer. The radium equivalent activity (Ra_{eq}), indoor gamma absorbed dose rate (D_R), and annual effective dose (H_R) associated with natural radioactivity were computed to assess the radiation hazards in adhesive building materials. The obtained specific activities of these natural radionuclides and the calculated radiation hazard indexes were compared with the international recommended values. The findings in this work of natural radioactivity levels were below the acceptable limits. Therefore, it was found the adhesive building materials were safe to be used as construction materials. Also, as a minor work, previous unpublished data of heavy metals in the same study adhesive materials were investigated by ICP-MS to figure out the correlation between heavy metal presence and natural radioactivity. The findings showed insignificant correlations between heavy metals and radioactivity.

Keywords

Building Material, Radioactivity, Adhesives

1. Introduction

The exposure of human to naturally occurring radiation comes primarily from two different origins. The first source, the main contributor is the terrestrial radioactive materials which shape from the formation of the earth crust. The second source comes directly from the cosmic radiation. The term of naturally occurring radionuclides is known as NORM. Only long-lived radionuclides, with half-lives comparable to the age of the earth, and their daughters, contribute to this natural radiation background in significant levels [1].

The majority of NORMs belong to the U-238, Th-232 decay series and K-40 as illustrated in **Figure 1**. NORMs emit alpha, beta particles and gamma ray as these radiations represent the primary sources of external exposure to the society [2].

These radionuclides (U-238, Th-232 decay series, and K-40) which emit either beta or alpha particles may be ingested or inhaled and surely can increase the internal exposures. Moreover, some radiation emitters may emit gamma radiation following their nuclear decay [3].

Terrestrial radionuclides occurred in all types of building materials, can give rise to external exposures owing to gamma rays. The specific activities of the radionuclides of various rocks and soils used as raw material in building materials are presented in **Table 1**. In **Table 1**, ignition rocks show higher levels of natural radionulcides than sedimentary rocks.

There have been so many studies concerning NORMs in soils, rocks, and construction materials which can furnish invaluable details on the nature and levels of radiation in any region and provide information in the change in radionuclide concentrations. All the studies of regional radionuclides in **Table 1** showed that most of building materials contain wide ranges of NORM levels.

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Determination of radioactivity in building materials used in them, shows that natural radionuclides of uranium (U-238) and thorium (Th-232) series, together

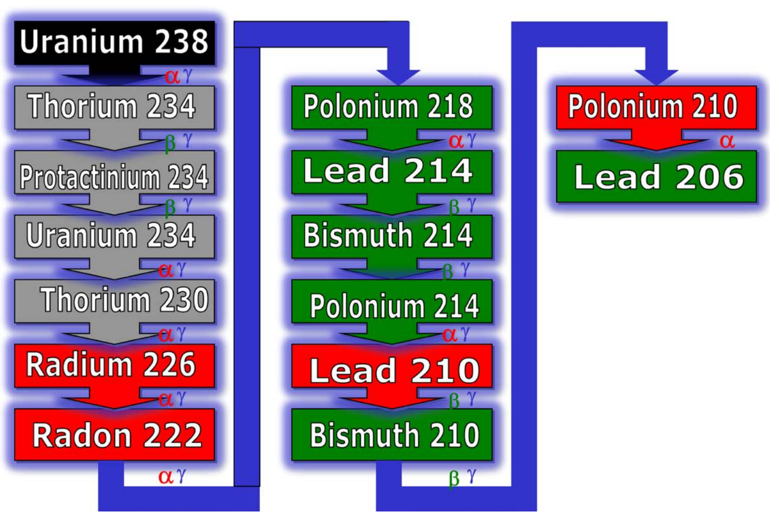

Figure 1. Uranium-238 decay series.

Table 1. Typical activities of U-238, Th-232, and K-40 in rocks and soils, data cited from [3].

Rock Type	Potassium-40		Thorium-232		Uranium-238	
	Total K (%)	$Bq \cdot kg^{-1}$	ppm	$Bq \cdot kg^{-1}$	ppm	$Bq \cdot kg^{-1}$
Igneous rocks						
Crustal average	0.8	300	3 to 4	10 to 15	0.5 to 1	7 to 10
Mafic	0.3 - 1.1	70 - 400	1.6 - 2.7	7	0.5 - 0.9	7
Salic	4.5	1100 - 1500	16 - 20	60	3.9 - 4.7	50
Granite (crustal aver.)	≤4	≤1000	17	70	3	40
Sedimentary rocks						
Shale, sandstones	2.7	800	12	50	3.7	40
Clean quartz	≤1	≤300	≤2	≤8	≤1	≤10
Dirty quartz	2	400	3 to 6	10 to 25	2 to 3	40
Arkose	2 to 3	600 - 900	2	≤8	1 to 2	10 to 25
Beach sands	≤1	≤300	6	25	3	40
Carbonate rocks	0.3	70	2	8	2	25
All rock (range)	**0.3 - 4.5**	**700 - 1500**	**1.6 - 20**	**7 to 80**	**0.5 - 4.7**	**7 to 60**
Continental crust (ave.)	2.8	850	10.7	44	2.8	36
Soil (ave.)	1.5	400	9	37	1.8	22

with the radioactive isotope of potassium (K-40), are presented. Limits of Ra-226 concentrations are established by different countries in order to control Rn-222 levels (200 Bq/m^3 in European Union and up to 1000 Bq/m^3 in Saudi Arabia). Potassium-40 and others gamma emitters of Ra-226 and Th-232 descendants, can cause an external dose. In European Union, a maximum value of 1 $mSv \cdot y^{-1}$ is recommended as well as in Saudi Arabia [4].

Merle Lust studied the NORM in building materials used in Estonia. During the Merle Lust investigation, 53 samples of commonly used raw materials and building products were collected and measured. The activity levels were determined by gamma ray spectrometry [5]. Their mean values were in the ranges 7 to 747 Bq/kg for K-40, 4.4 to 69 Bq/kg for Ra-226, and 0.8 to 86 Bq/kg for Th-232. The activity index I in the 53 different building materials varied from 0.02 to 0.74 and the radium equivalent, from 6 to 239. The average annual dose for the people, caused by the building materials of dwellings, was assessed for most commonly used materials. It was estimated to be in the range from 0.16 mSv to 0.44 mSv.

Adriana Etokov and Lenka Palakov [6] studied activities of Ra-226, Th-232 and K-40 and radiological parameters (radium equivalent activity, gamma and alpha indexes, the absorbed gamma dose rate and external and internal hazard indices) of cements and cement composites commonly used in the Slovak Republic. The cement samples of 8 types of cements from Slovak cement plants and five types of composites made from cement type CEM I were analyzed. The radionuclide activities in the cements ranged from 8.58 to 19.1 Bq/kg, 9.78 to 26.3 Bq/kg and 156.5 to 489.4 Bq/kg for Ra-226, Th-232 and K-40, respectively. The radiological parameters in cement samples were calculated as follows: mean radium equivalent activity $Ra_{eq} = 67.87$ Bq/kg, gamma index $I_\gamma = 0.256$, alpha index $I_\alpha = 0.067$, the absorbed gamma dose rate $D = 60.76$ nGy/h, external hazard index $H_{ex} = 0.182$ and internal hazard index H_{in} was 0.218. The radionuclide activity in composites ranged from 6.84 to 10.8 Bq/kg for Ra-226, 13.1 to 20.5 Bq/kg for Th-232 and 250.4 to 494.4 Bq/kg for K-40.

Singh [7] carried out radiation measurement of Indian building materials. The activity concentrations of Ra-226, Th-232 and K-40 have been determined by gamma-ray spectrometry. The measured activity in the selected building materials ranges from 3.2 to 151.7 Bq/kg, 14 to 63.7 Bq/kg and 24.3 to 121.5 Bq/kg for Ra-226, Th-232 and K-40 respectively. The activity concentration of U-238 were determined using fission track technique and the value ranges from 0.11 to 3.85 ppm.

W.R. Alharbi, J.H. AlZahrani [8] studied the radioactivity in some building materials in Saudi Arabia, the natural radionuclides (Ra-226, Th-232 and K-40) present in various building materials available in Saudi Arabia (Jeddah city) analyzed using Gamma-ray spectrometry. The results showed that the activity concentration of Ra-226, Th-232 and K-40 was between 12.6 Bq/kg (Brick-clay) to 31.5 Bq/kg, (Granite), 9.2 Bq/kg (Brick-clay) to 27.2 Bq/kg (Granite) and 114.4 Bq/kg (Brick-clay) to 534.7 Bq/kg (Granite), respectively. The radiological hazard parameters radium equivalent activity, gamma index, absorbed dose rate and the annual exposure rate, were calculated to assess the radiation hazards associated with Saudian buildings. All studied samples were lower than world average limits. The results were compared with the published data of other countries and with the world average limits. The measurements helped in the development of standards and guidelines for the use and management of building materials.

Therefore, this work dealt with assessing of natural radioactivity in adhesive materials used and sold in Riyadh city, Saudi Arabia.

2. Assessment of Radiation Hazard

The risk assessment of radiation doses can be given in form of radiation indexes. In literature, there has been tonnes of publications on how to evaluate the radiation hazards linked to presence of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K [9] [10].

In order to carry on such assessment, one needs to provide some terminologies associated with radiation hazard. Therefore, this section will explain them.

2.1. Absorbed Dose Rate

The direct link between radioactivity levels and their exposure is known to be the absorbed dose rate. The following equation can be used to calculate the absorbed dose rate [11] [12]:

$$D = 0.462A_{\text{Ra-226}} + 0.604A_{\text{Th-232}} + 0.0417A_{\text{K-40}} \quad (1)$$

where D is the adsorbed dose rate in nGy/h,

$A_{\text{Ra-226}}$, $A_{\text{Th-232}}$ and $A_{\text{K-40}}$ are the activities of Ra_{226}, Th_{232} and K_{40}, respectively. The equation above was taken directly from UNSCEAR.

2.2. Radium Equivalent Activity

This index is very commonly used in radiological hazard evaluation. The index was mainly introduced by UNSCEAR owing to uniform distribution of the mentioned-above radionuclide in environmental, geochemical, biological samples [12] [13] [14].

The next equation can be estimated through:

$$Ra_{eq} = A_{\text{Ra-226}} + 1.43A_{\text{Th-232}} + 0.077A_{\text{K-40}} \quad (2)$$

where $A_{\text{Ra-226}}$, $A_{\text{Th-232}}$ and $A_{\text{K-40}}$ are the activities levels of Ra-226, Th-232, and K-40, respectively.

The value of 370 Bq/kg is set to be permissible max level that corresponds to effective dose of 1 mSv for public [15] [16].

2.3. Annual Effective Dose Equivalent

It is well known that the absorbed dose rate in one meter in air above the earth surface can not provide the radiological risk to public [17]. So, the absorbed dose has be to converted to annual effective dose equivalent (AEDE) from outdoor regional gamma radiation. In order to calculate the annual effective dose equivalent, one can use the following equation [18]:

$$\text{AEDE} = D(\text{nGy/h}) \times 8760\ \text{hr} \times 0.2 \times 0.7(\text{Sv/Gy}) \times 10^{-3} \quad (3)$$

where D is absorbed dose,

0.7 (Sv/Gy) is conversion factor,

0.2 is outdoor occupancy factor.

2.4. External Hazard Index

Krieger proposed a model to introduce external hazard index (H_{ex}) owing to limitation of radiation attribute to natural radionuclide [19].

To calculate the external radiation hazard, one can use the following equation:

$$H_{ex} = \left[\frac{A_{\text{Ra}}}{370}\right] + \left[\frac{A_{\text{Th}}}{259}\right] + \left[\frac{A_{\text{K}}}{4810}\right] \leq 1 \quad (4)$$

The max value of H_{ex} equal to unity meets to the upper limit of Ra_{eq} 370 Bq/Kg Kg [20] [21].

3. Measurements of Natural Radioactivity in Building Materials in Saudi Arabia

The samples were crushed using crusher and then homogenized. The homogenized samples were filled into 1000 ml Marinelli beakers which were later hermetically sealed with the help of PVC (polyvinyl chloride) commercial to prevent the escape of air-borne of Rn-222 and Rn-220 from the samples. All the samples were accurately weighted and stored for period of at least one month prior to determination in order to attain radioactive secular equilibrium between Ra-226 and Rn-222 [9].

In this investigation, the sample activities in building materials were measured by using high-resolution gamma-ray spectrometry system consists of coaxial hyper-pure germanium (HPGe) detector with highly passive shielding and low background. The detector was cooled with liquid nitrogen cryostat to re-duce the leakage current. To reduce the background radiation from natural sources the detector was enclosed of 10 cm thick cylindrical lead shield. The lead shielding was graded with an inner layer of thick copper to reduce any influence fluorescences [22].

The detector was connected to a pre-amplifier, shaping amplifier and high voltage power supply which were used for conversion of the observed energy into a pulse height spectrum. The pulse amplitude was converted to a discrete number through more 8000 channel multi-channel analyser (MCA). The data acquisition, display, and analysis of γspectra were carried out using Genie 2000 software [23].

The relationship between the channel numbers corresponding to absolute γenergies was determined. The specification of the used instrument is listed in **Table 2** [22].

In this work, gamma reference sources containing mixed of radionuclide were used for energy set of calibration. These references emit a wide range of gamma-ray energies covering the entire energy range of interest. The main gamma-ray energy lines of the used references are shown in **Table 3**.

The gamma energies used for Ra-226 was at 186.2 keV and Pb-214 was also used at different energies at 295.2 and 351.9 keV.

For gamma-ray spectrometry of unknown, the detector efficiency measurement plays important role in gamma-counting. The full-energy peak efficiency

Table 2. The HPGe specifications.

Geomertry	Co-axial open end closed end faceing window
Diameter	74.7 mm
Length	92.9 mm
Active area window	11.6 mm
Operating Voltage	4500 V
Leakage Current	0.01 A
Amplifier gain	50
Amplifier fine	30 - 40
Pulse time	6 micro sec

Table 3. Gamma energies [22].

Identified radionuclide	Gamma-ray energy (KeV)	Gamma emission probability	Source of gamma ray transition
Th-234	92.58	0.0558 ± 0.0030	U-238 series-doublet peak
Ac-228	129.06	0.0242 ± 0.0009	Th-232 series
Ac-228	153.97	0.0072 ± 0.0002	Th-232 series
U-235	185.72	0.572 ± 0.0005	Primordial U-235
Ra-226	186.21	0.0359 ± 0.0019	U-238 series
Ac-228	209.25	0.0389 ± 0.0007	Th-232 series
Pb-212	238.63	0.4360 ± 0.0030	Th-232 series
Pb-214	241.99	0.0725 ± 0.0002	Th-238 series
Ac-228	270.24	0.0346 ± 0.0006	Th-232 series
Tl-208	277.35	0.0227 ± 0.0003	Th-232 series
Pb-214	295.22	0.1842 ± 0.0004	Th-238 series
Pb-214	300.08	0.0318 ± 0.0013	Th-232 series
Ac-228	328	0.0295 ± 0.0012	Th-232 series
Ac-229	338.32	0.1127 ± 0.0019	Th-232 series
Pb-214	351.93	0.3560 ± 0.0007	Th-238 series
Ac-228	463	0.0440 ± 0.0007	Th-232 series
Annihilation	511		Annihilation radiation
Tl-208	583.19	0.3055 ± 0.0017	Th-232 series
Bi-214	609.31	0.4549 ± 0.0016	U-238 series
Cs-137	661.65	0.8510 ± 0.0020	Man-made
Bi-212	727.33	0.0674 ± 0.0012	Th-232 series
Bi-214	768.35	0.0489 ± 0.0001	U-238 series
Ac-228	794.94	0.0425 ± 0.0007	Th-232 series
Tl-208	860.56	0.0448 ± 0.0004	Th-232 series
Ac-228	911.2	0.2580 ± 0.0040	Th-232 series
Bi-214	934.06	0.0311 ± 0.0001	U-238 series
Ac-228	964.76	0.0499 ± 0.0002	Th-232 series
Ac-228	968.97	0.1580 ± 0.0030	Th-232 series
Bi-214	1120.28	0.1492 ± 0.0003	U-238 series
Bi-214	1238.11	0.0583 ± 0.0015	U-238 series
Bi-214	1377.67	0.0399 ± 0.0001	U-238 series
Bi-214	1407.98	0.0239 ± 0.001	U-238 series
K-40	1460.83	0.1066 ± 0.0013	Primordial K-40
Ac-228	1588.19	0.0322 ± 0.0008	Th-232 series
Bi-212	1620.5	0.0151 ± 0.0003	Th-232 series
Bi-214	1729.59	0.0298 ± 0.0001	U-238 series
Bi-214	1764.49	0.1530 ± 0.0003	U-238 series
Bi-214	2204.21	0.0492 ± 0.0002	U-238 series
Tl-208	2614.5	0.3585 ± 0.0007	Th-232 series

can be computed through:

$$\varepsilon_f = \frac{N_p}{N_\gamma} \tag{5}$$

where ε_f is defined as the full-energy peak efficiency,

N_p is the net gamma-ray counting rate in the full-energy peak

N_γ is defined as the gamma-ray emission rate where it can be calculated via:

$$N_\gamma = AP_\gamma \tag{6}$$

where A is the activity in Bq of the reference and P_γ is the branching ratio of the radionuclide.

In order to removed interference between multi peaks, the calibration of energy efficiency was carried out carefully. For every source, the energy efficiency was calculated using formula (5) as shown in Figure 2 and the energy channels was calculated as shown in Figure 3 [21] [22].

The minimum detection activity (MDA) which is the performance of gamma-ray spectrometry is defined as the lowest quantity of radionuclide that can be measured for a certain measurement. MDA can be calculated via the following

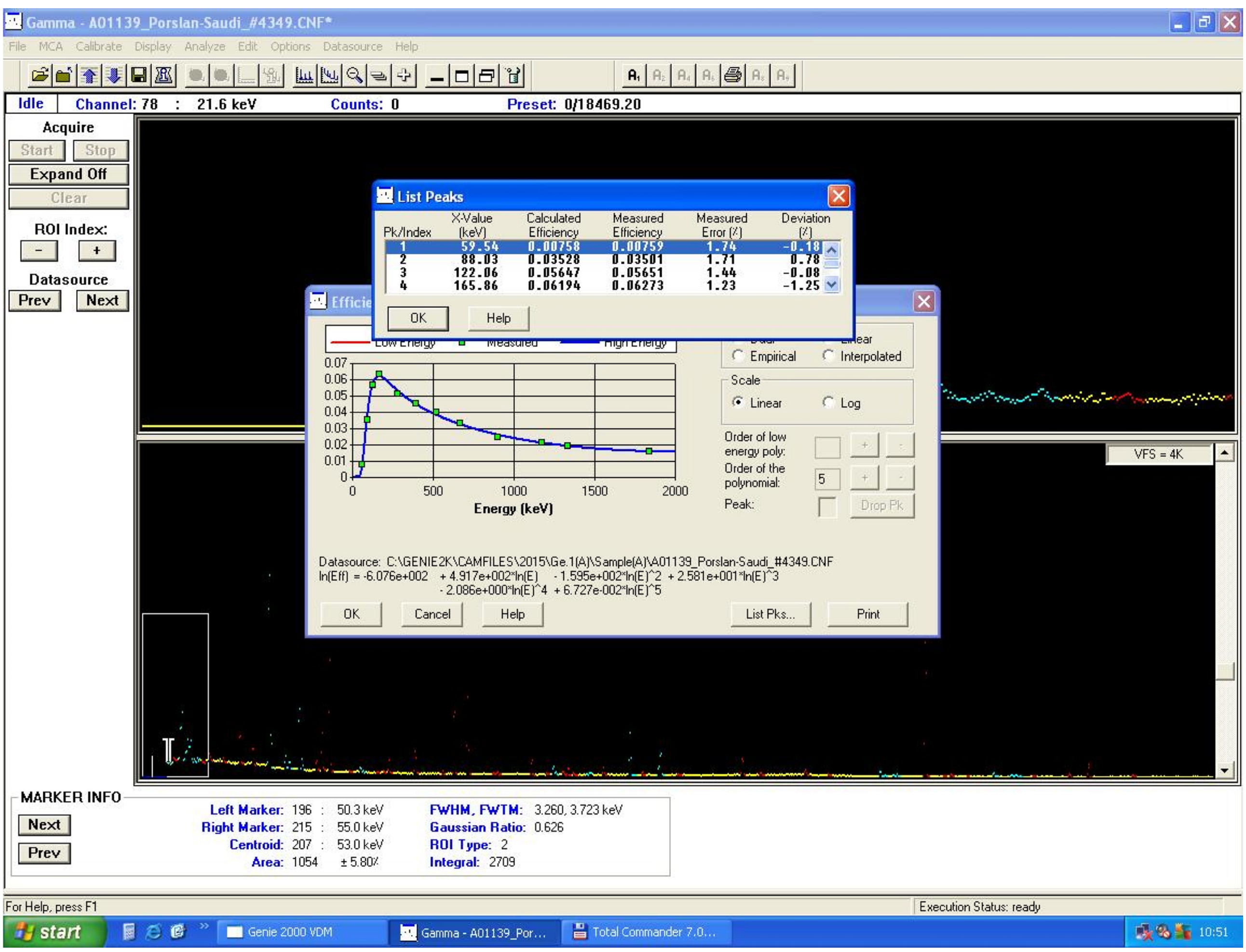

Figure 2. Absolute full-energy peak efficiency as function of γ energy for the HPGe detector used in our study.

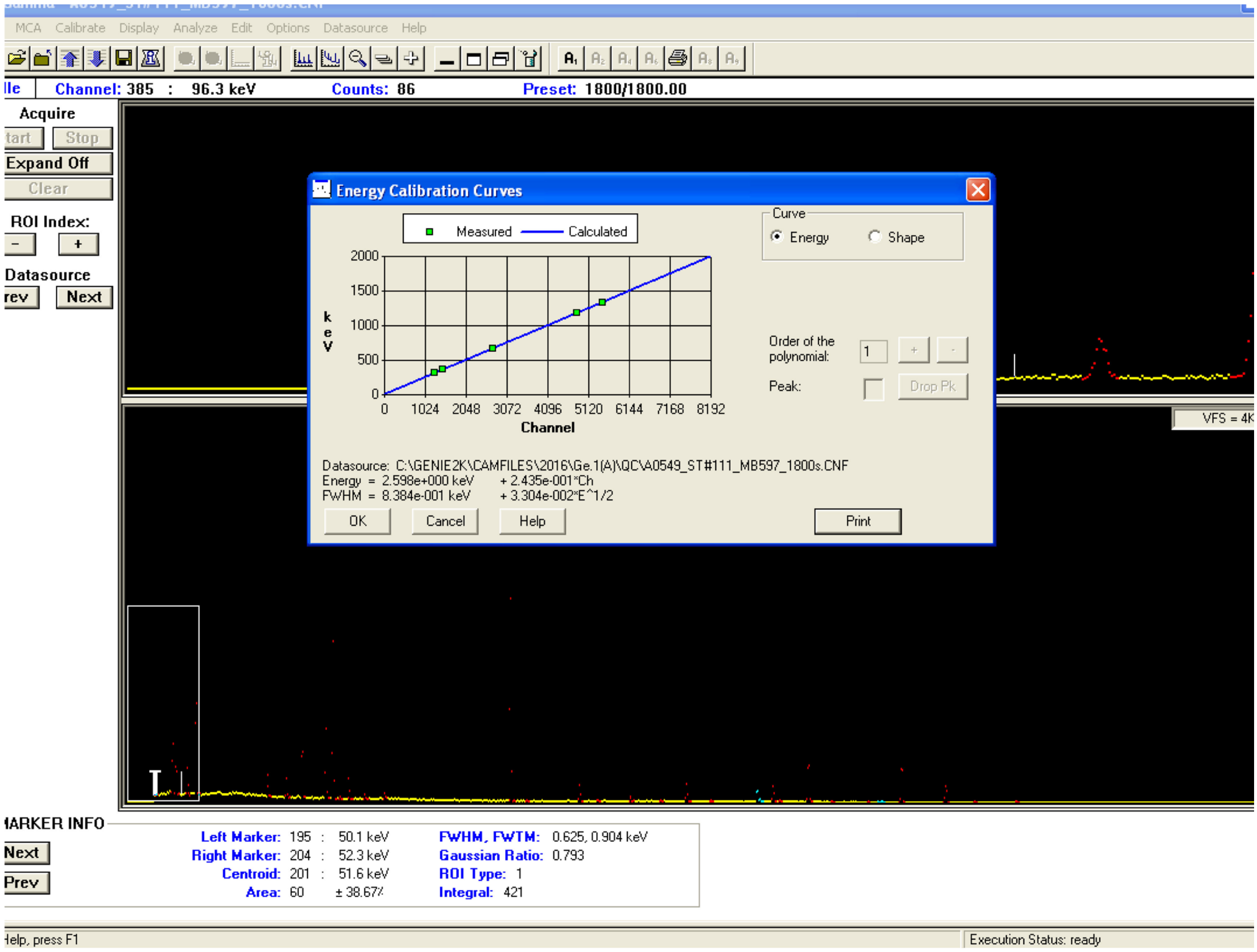

Figure 3. The relationship between gamma-ray energies and their channel number.

equation in unit of Bq/kg:

$$MDA = \frac{L_D}{\varepsilon_f P_\gamma TM} \tag{7}$$

where L_D is the detection limit,

ε_f is the absolute efficiency of the detector,

P_γ is the gamma branching ratio or gamma probability,

T is the counting time,

M is the sample mass in kg.

L_D can be expressed through the equation:

$$L_D = 2.71 + 4.65(\text{background})^{0.5} \tag{8}$$

L_D was measured for over 170,000 sec with no radiation and it was carried out with 1000 Marinelli beaker filled with tri-di-ionized water placed inside the detector using the same geometry.

The specific activity is defined as the activity per mass unit. The specific activity of individual radionuclide in the studied building material can be calculated using the following equation:

$$A = \frac{N}{\varepsilon_f P_\gamma TMK} \quad (9)$$

where ε_f is the efficiency of energy at the photopeak of interested radionuclide

T is counting time in second (86,400 sec)

M is the mass in kg of the analysed sample,

P_γ is the gamma branching ratio or gamma probability,

K is a correction factor,

N is the corrected net peak area

$$N = N_S - N_B \quad (10)$$

where N_S is the net peak area and N_B is the net peak area of the background [23].

4. Radiation Hazard in Adhesive Materials

The relevant radiological assessed values for adhesive materials are listed in Table 4. The highest reported value of U-238 in adhesive was 17.4 Bq/kg whereas the lowest value was 5.2 Bq/kg and the mean value was 8.7 Bq/kg. For Th-232, the lowest reported value in this study was 5.3 Bq/kg and the highest value was 12.4 Bq/kg. The average of Th-232, by this study, was 7.2 Bq/kg. For K-40, The highest reported value, by our study, was 183 Bq/kg and the lowest values was 0 Bq/kg which is normal as adhesive does not contain potassium.

To discuss the statistical evaluation, one can start with confidence limits test of Shawhart. The confidence limit test of Th-232 in Figure 4 indicated that Th-232 levels in adhesive materials were normal distributed and all the data were located within the max and min border of confidence limits.

The confidence limit test for U-238 is illustrated in Figure 5. The U-238 levels clearly proved that data can be treated as parametric due to normal distribution of the obtained data.

The shawhart confidence limit interval test showed K-40 results passed the test as illustrated in Figure 6.

Ra_{eq} mean value was 24 Bq/kg that is lower than set limit of 370 Bq/kg [24]. For H_{ex}, the lowest reported value was 0.05 and the highest value was 0.07 with mean value of 0.06 mSv/yr. The fixed limit of H_{ex} is set to be 1 mSv/yr. H_{in} lowest value for adhesive was 0.08 mSv/yr and highest value was 0.13 mSv/yr with mean of 0.09 mSv/yr. Lucky, the study adhesive materials were less than max permissible value of 1 mSv/yr. The annual effective does reported in this work was 0.08 mSv/kg in average where this values is less than max permissible value of 1 mSv/yr. Therefore, The reported radiological values were far below the permissible limits. Therefore, it is obvious that the adhesive did not posses any radiation hazard to residents.

Turhan, eref *et al.* (2008) reported natural radioactivity in adhesive materials. In their study, U-238 activities were 7.3 to 69.4 Bq/kg whereas this study showed the ranges were 0 to 17 Bq/kg. Thus, the study adhesives were located within the worldwide ranges. In Tuhan study, Th-232 activity was 2 to 57 Bq/kg in adhesives

Table 4. Radiation calculations for adhesive materials.

Sample code	K-40 Bq\Kg	Ra-226 Bq\Kg	Th-232 Bq\Kg	U-238 Bq\Kg	Ra eq	$H_{ex} \le 1$	$H_{in} \le 1$	α Concentration	α index	Outdoor dose	Annual Effective Dose (mSv/y)
A01129	49.22	14.11	6.24	15.76	26.82	0.07	0.11	0.09	0.07	12.45	0.09
A01121	0.00	12.20	12.40	15.80	29.93	0.08	0.11	0.10	0.06	13.34	0.09
A01140	93	8.1	6.8	8.7	24.99	0.07	0.09	0.09	0.04	11.84	0.08
A01144	183	11.4	7.1	0	35.64	0.10	0.13	0.13	0.06	17.31	0.12
B01047	13.7	9.6	6.4	6.9	19.81	0.05	0.08	0.07	0.05	8.98	0.06
B01057	40	9.6	6.4	4.06	21.83	0.06	0.08	0.08	0.05	10.08	0.07
C0533	12.3	9.89	9.61	9.29	24.58	0.07	0.09	0.09	0.05	11.05	0.08
B01059	81.8	7.6	5.7	7.3	22.05	0.06	0.08	0.08	0.04	10.46	0.07
B01061	69.8	6.5	5.07	5.2	19.12	0.05	0.07	0.07	0.03	9.06	0.06
C0543	18.05	12.7	4.87	11.5	21.05	0.06	0.09	0.07	0.06	9.64	0.07
B01044	36.4	18.1	5.03	17.4	28.10	0.08	0.12	0.10	0.09	13.00	0.09
C0548	0	9.8	6.8	9.4	20	0.05	0.08	0.07	0.05	8.75	0.06
C0554	9.6	11.1	7.9	5.8	23	0.06	0.09	0.08	0.06	10.43	0.07
C0555	32	6.85	5.3	8.89	17	0.05	0.06	0.06	0.03	7.79	0.05
C0558	25.2	9.8	10.3	4.3	26	0.07	0.10	0.09	0.05	11.97	0.08
Count	15	15	15	15	15	15.00	15.00	15.00	15.00	15.00	15.00
Mean	44.3	10.49	7.1	8.69	24	0.06	0.09	0.09	0.05	11.08	0.08
Stdev	47.9	3.00	2.2	4.83	5	0.01	0.02	0.02	0.02	2.38	0.02
Range	183.0	11.60	7.5	17.40	19	0.05	0.06	0.07	0.06	9.52	0.07
Minimum	0.0	6.50	4.9	0.00	16.89	0.05	0.06	0.06	0.03	7.79	0.05
25th Percentile (Q1)	12.3	8.10	5.3	5.20	19.81	0.05	0.08	0.07	0.04	9.06	0.06
50th Percentile (Median)	32.0	9.80	6.4	8.70	23.14	0.06	0.09	0.08	0.05	10.46	0.07
75th Percentile (Q3)	69.8	12.20	7.9	11.50	**26.82**	**0.07**	**0.11**	**0.09**	**0.06**	**12.45**	**0.09**
Maximum	183.0	18.10	12.4	17.40	35.64	0.10	0.13	0.13	0.09	17.31	0.12
95.0% CI Mean	17.7 to 70.7	8.8 to 12.1	5.8 to 8.2	6.1 to 11.3	21.3 to 26.7	0.06 to 0.07	0.08 to 0.1	0.07 to 0.1	0.04 to 0.06	9.7 to 12.4	0.07 to 0.08
95.0% CI Sigma	35.038 to 75.477	2.2 to 4.7	1.6 to 3.4	3.5 to 7.6	3.6 to 7.7	0.01 to 0.02	0.01 to 0.03	0.01 to 0.03	0.01 to 0.02	1.7 to 3.7	0.01 to 0.02
Anderson-Darling Normality Test	0.9	0.41	0.8	0.36	0.26	0.26	0.34	0.41	0.41	0.37	0.37
p-value (A-D Test)	**0.014**	0.30	**0.02**	0.41	0.65	0.65	0.44	0.30	0.30	0.37	0.37
Skewness	1.9	1.08	1.36	0.35	0.87	0.86	0.48	1.27	1.08	1.18	1.18
p-value (Skewness)	**0.003**	0.07	**0.02**	0.52	0.13	0.13	0.39	0.03	0.07	0.04	0.04
Kurtosis	4.4	1.77	1.39	−0.22	0.89	0.89	−0.49	2.51	1.77	2.17	2.17
p-value (Kurtosis)	**0.012**	0.14	0.20	1.00	0.34	0.34	0.76	0.07	0.14	0.09	0.09

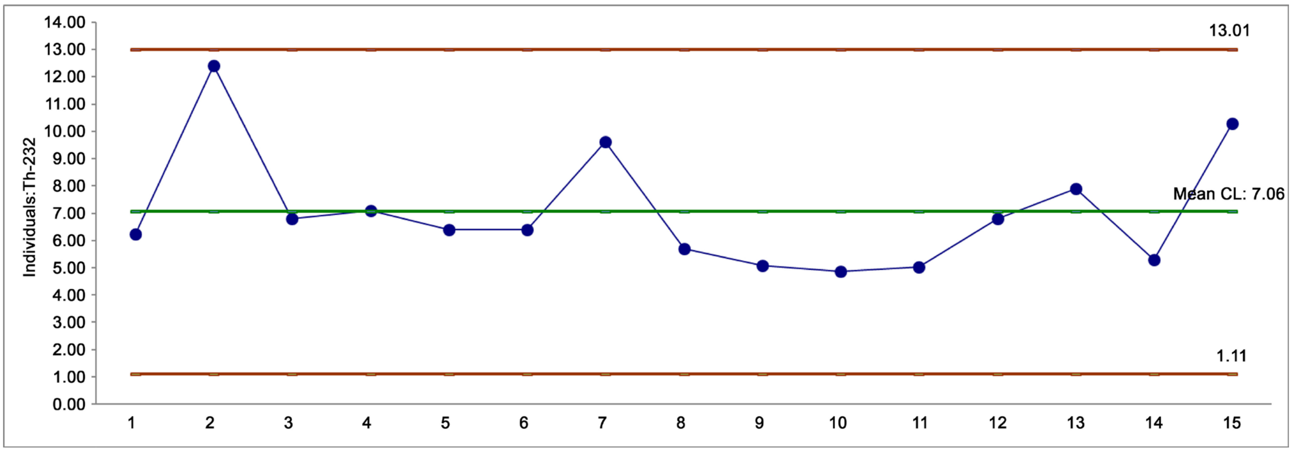

Figure 4. The Shawhart confidence limits of Th-232.

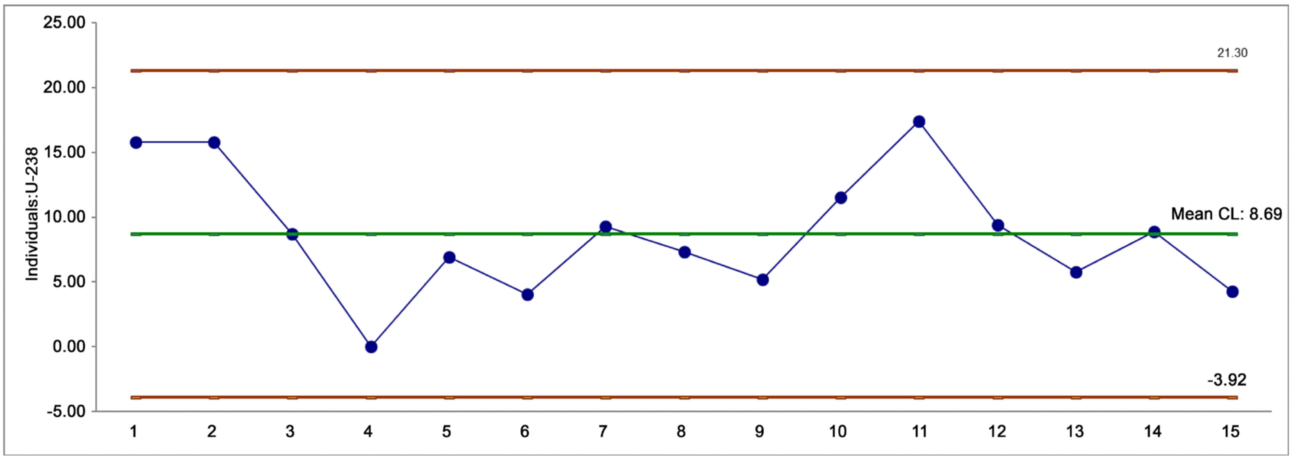

Figure 5. The Shawhart confidence limits of U-238.

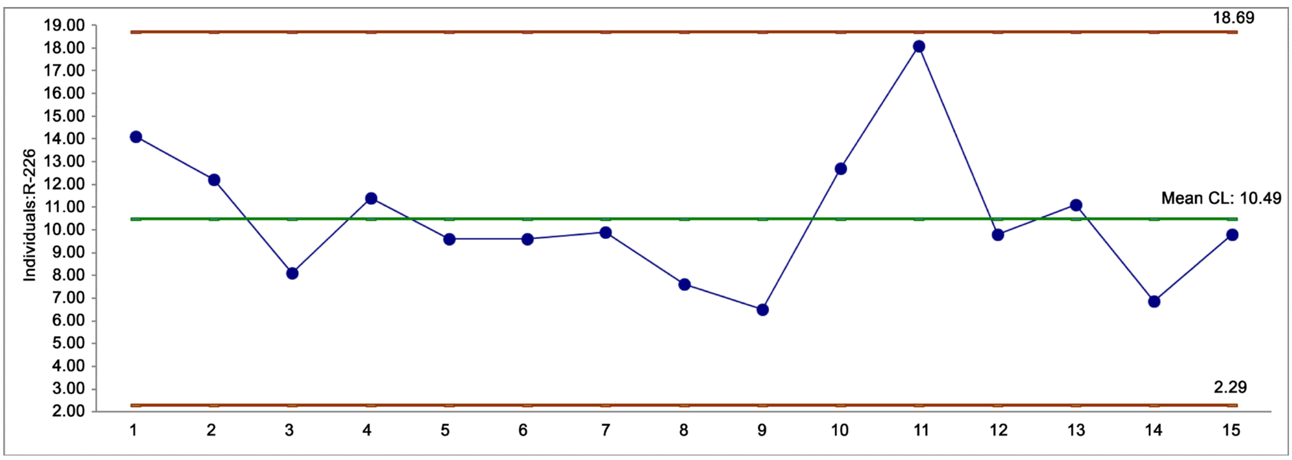

Figure 6. The Shawhart confidence limits of K-40.

while, this study, showed the range of Th-232 was 4.9 to 12.4 Bq/kg. So, it can be stated that the study adhesives were within the worldwide range. K-40, in Turhan study, was ranging 21 to 816 Bq/kg whereas in this study was 0 to 183 Bq/kg. Therefore, the natural radioactivity in adhesives, by this study, were less than the worldwide values.

5. Correlations of Heavy Metals and Radioactivity in Adhesive Materials

This section deals with previous unpublished data of heavy metals in adhesive materials and their correlation with radioactivity. It is a step to explore the relationship between them in form of matrix correlations and Mood's test (Monte Carlo).

Using Mood's Median Test, the obtained results showed there was different in the medians of the data as calculated in **Table 5** and can be shown in **Figure 7**. Thus, the obtained results of heavy metals levels and natural radioactivity may be treated as non-parametric data.

Table 6 and **Table 7** show the calculations of correlations of the studied adhesive materials between selected heavy metals and natural radioactivity.

K-40 was positively correlated with Ga, As, Mo, and Cd. Th-232 was also correlated with Ga, As, and Cd.

Using Mood's Median Test, the obtained results showed there was different in the medians of the data as calculated in **Table 5** and can be shown in **Figure 5**. Thus, the obtained results of heavy metals levels and natural radioactivity may be treated as non-parametric data.

6. Conclusions

In **Figure 8** and **Table 8**, the obtained results of Radium equivalent radiation hazard index showed that data were located below the max permissible limit of 370 Bq/kg. Therefore, the radiation hazard index of Ra_{eq} indicated the analysed adhesive material were not contaminated with NORM.

Table 5. Mood's median test for adhesive materials.

Test Information														
H0: Median 1 = Median 2 = … = Median k Ha: At least one pair Median i Median j														
Results:	**Cr**	**Zn**	**Ga**	**As**	**Sr**	**Mo**	**Cd**	**Ba**	**Pb**	**Bi**	**U 238**	**K-40**	**Ra-226**	**Th-232**
Count (N ≤ Overall Median)	2	5	14	15	0	14	15	0	12	11	15	2	0	0
Count (N > Overall Median)	13	10	1	0	15	1	0	15	3	4	0	13	15	15
Median	5.70	3.87	1.38	0.74	32.75	1.17	0.21	23.54	1.97	0.18	0.68	32.00	9.80	6.40
UC Median (2-sided, 95%)	8.98	4.39	1.77	0.97	47.45	1.55	0.27	31.29	3.26	4.11	0.83	62.11	11.90	7.60
LC Median (2-sided, 95%)	4.38	3.44	1.09	0.66	24.33	0.82	0.13	10.82	1.50	0.03	0.55	12.82	8.66	5.45
Overall Median	3.626													
Chi-Square	154													
DF	13													
P-Value (2-sided)	**0.0000**													

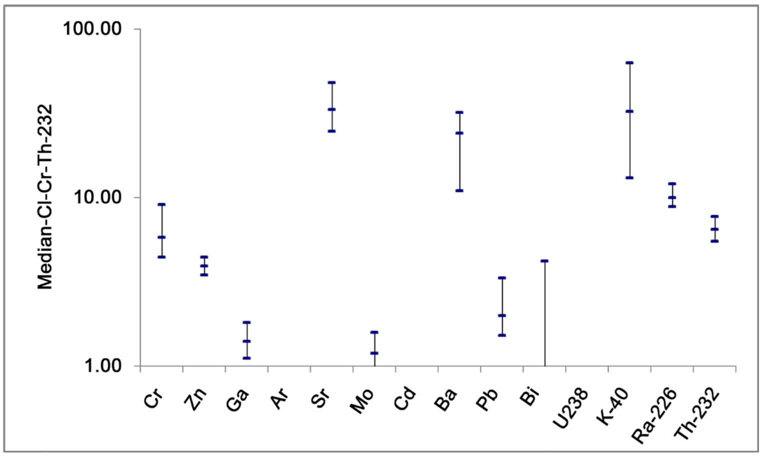

Figure 7. Medians (log scale) of adhesive materials for Mood's median test.

Table 6. Correlation calculations between chemical and radiation measurements using Pearson Methods for adhesive materials.

Pearson Correlations	Cr	Zn	Ga	As	Sr	Mo	Cd	Ba	Pb	Bi	K-40	Ra-226	Th-232	U-238
Cr	1	0.0184	0.1952	−0.2415	**0.7283**	−0.1013	−0.0168	0.0416	0.3076	0.3129	0.1172	−0.1953	−0.1649	−0.0044
Zn		1	0.4322	**0.5571**	−0.1850	**0.5997**	**0.5145**	0.4254	0.1814	0.2489	0.4857	0.1627	0.2882	−0.1536
Ga			1	**0.5967**	0.2276	0.2831	**0.6114**	**0.8990**	**0.5221**	0.0732	**0.5241**	−0.0345	**0.5740**	0.0352
As				1	−0.3862	**0.7136**	**0.7798**	**0.5659**	−0.1353	0.1225	**0.6512**	0.0787	**0.6002**	−0.3315
Sr					1	−0.2175	−0.3477	0.0303	**0.6765**	0.1157	0.1258	−0.3674	−0.0532	**0.5838**
Mo						1	**0.5305**	0.1477	−0.1616	**0.6229**	**0.7677**	0.0832	0.2417	−0.2492
Cd							1	**0.6037**	−0.1533	0.0450	**0.6774**	0.0753	0.5058	−0.3984
Ba								1	0.3562	−0.0944	0.3624	0.0347	**0.5495**	−0.0914
Pb									1	−0.0304	0.0455	−0.3514	0.3123	**0.6187**
Bi										1	0.3317	−0.0142	−0.1309	−0.3190
U 238											1	−0.1030	0.4035	0.0166
K-40												1	−0.1287	−0.2703
Ra-226													1	0.0320
Th-232														1
Pearson Probabilities	**Cr**	**Zn**	**Ga**	**As**	**Sr**	**Mo**	**Cd**	**Ba**	**Pb**	**Bi**	**K−40**	**Ra−226**	**Th−232**	**U−238**
Cr		0.9481	0.4856	0.3859	**0.0021**	0.7196	0.9526	0.8830	0.2648	0.2562	0.6775	0.4855	0.5570	0.9875
Zn			0.1076	**0.0310**	0.5093	**0.0181**	**0.0497**	0.1139	0.5177	0.3711	0.0664	0.5623	0.2976	0.5848
Ga				**0.0189**	0.4145	0.3065	**0.0155**	**0.0000**	**0.0459**	0.7955	**0.0449**	0.9029	**0.0252**	0.9010
As					0.1551	**0.0028**	**0.0006**	**0.0279**	0.6307	0.6636	**0.0086**	0.7804	**0.0180**	0.2274
Sr						0.4363	0.2042	0.9146	**0.0056**	0.6813	0.6551	0.1779	0.8506	**0.0223**
Mo							**0.0419**	0.5993	0.5649	**0.0131**	**0.0008**	0.7682	0.3856	0.3704
Cd								**0.0172**	0.5854	0.8734	**0.0055**	0.7897	0.0544	0.1414
Ba									0.1925	0.7379	0.1844	0.9024	**0.0339**	0.7458
Pb										0.9144	0.8721	0.1990	0.2571	**0.0139**
Bi											0.2272	0.9599	0.6419	0.2465
U-238												0.7148	0.1358	0.9533
K-40													0.6477	0.3299
Ra-226														0.9100
Th-232														

Table 7. Correlation calculations between chemical and radiation measurements using spearman Rank Correlations Methods for adhesive materials.

Spearman Rank Correlations	Cr	Zn	Ga	As	Sr	Mo	Cd	Ba	Pb	Bi	K−40	Ra−226	Th−232	U−238
Cr	1	−0.0750	0.3893	−0.2536	**0.7571**	−0.0679	−0.1179	0.3000	0.5036	0.3429	0.0107	−0.2359	−0.2111	0.1735
Zn		1	0.3107	0.4179	−0.2786	0.5000	0.3679	0.2321	0.0321	0.2464	0.3929	0.4433	−0.0233	−0.2147
Ga			1	0.3036	0.3679	**0.5393**	0.2714	**0.7286**	**0.5821**	**0.5464**	**0.5857**	0.1233	0.1771	0.0429
As				1	**−0.5500**	**0.6357**	0.4143	−0.0679	−0.3500	0.2464	0.4750	0.4629	0.0930	−0.3041
Sr					1	−0.1857	−0.2857	0.3607	**0.7893**	0.2143	0.0821	**−0.5612**	0.0984	**0.5295**
Mo						1	**0.6357**	0.1679	−0.0464	**0.7071**	**0.7857**	0.4486	0.2630	−0.2558
Cd							1	−0.1143	−0.1429	0.2393	**0.6607**	0.1305	0.1020	−0.1843
Ba								1	**0.5786**	0.1286	0.1357	0.1055	0.0501	0.0555
Pb									1	0.1429	0.1250	−0.4772	0.4168	0.3345
Bi										1	0.5107	0.3092	0.0519	−0.2075
U 238											1	0.0268	0.3059	0.1342
K-40												1	−0.2677	−0.3715
Ra-226													1	0.0995
Th-232														1
Spearman Rank Probabilities	**Cr**	**Zn**	**Ga**	**As**	**Sr**	**Mo**	**Cd**	**Ba**	**Pb**	**Bi**	**K−40**	**Ra−226**	**Th−232**	**U−238**
Cr		0.7905	0.1515	0.3618	**0.0011**	0.8101	0.6757	0.2773	0.0557	0.2109	0.9698	0.3973	0.4501	0.5363
Zn			0.2597	0.1212	0.3147	0.0577	0.1773	0.4051	0.9095	0.3760	0.1475	0.0980	0.9344	0.4423
Ga				0.2714	0.1773	**0.0380**	0.3278	**0.0021**	**0.0228**	**0.0351**	**0.0218**	0.6615	0.5278	0.8792
As					**0.0337**	**0.0109**	0.1247	0.8101	0.2009	0.3760	0.0736	0.0823	0.7416	0.2705
Sr						0.5075	0.3019	0.1866	**0.0005**	0.4431	0.7710	**0.0295**	0.7272	**0.0424**
Mo							**0.0109**	0.5499	0.8695	**0.0032**	**0.0005**	0.0935	0.3437	0.3574
Cd								0.6851	0.6115	0.3904	**0.0073**	0.6430	0.7176	0.5109
Ba									**0.0238**	0.6479	0.6296	0.7084	0.8593	0.8444
Pb										0.6115	0.6571	0.0721	0.1222	0.2230
Bi											0.0517	0.2621	0.8543	0.4580
U 238												0.9244	0.2675	0.6336
K-40													0.3348	0.1727
Ra-226														0.7243
Th-232														

Figure 9 shows the obtained results of external hazard values where all the reported data are located below 0.09. The average external radiation hazard was much more below the permissible limit of one mSv/yr. Thus, it can be stated that adhesive materials were free of natural radioactivity in term of external radiation hazard.

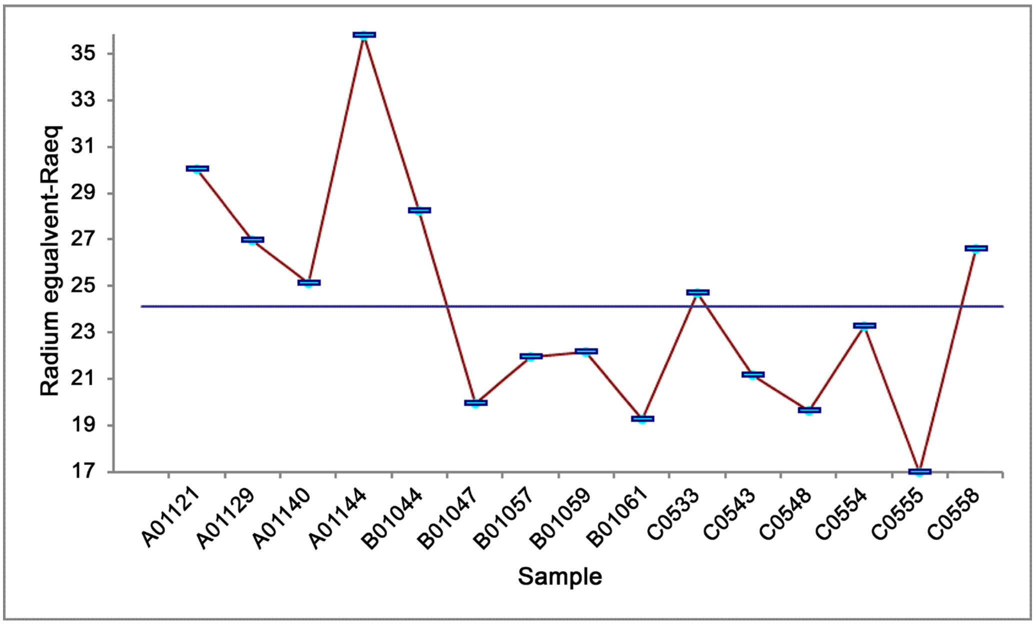

Figure 8. Radium equivalent values of the adhesive materials.

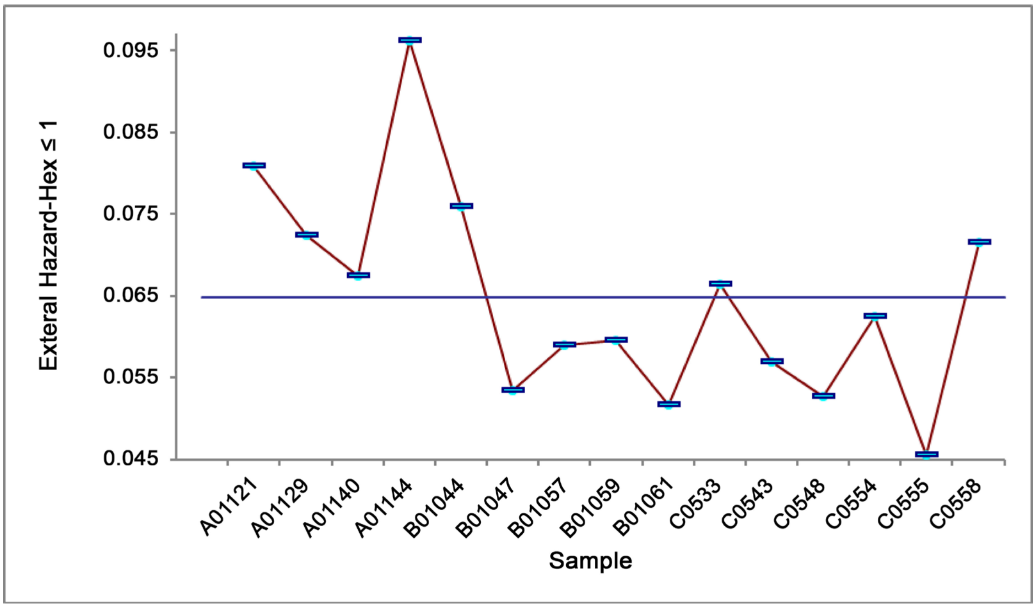

Figure 9. External Hazard values of the adhesive materials.

Similarly, the internal radiation hazard was computed as demonstrated in Figure 10. All the reported data of internal radiation hazard were in range of less than 0.1 whereas the max allowable limit is fixed by one.

The last radiation hazard index used in this study was annual effective dose. This index is the most important radiation index in any radiation risk assessment. Figure 11 shows the average valued of annual effective dose was less than 0.07 while the fixed value of this index is one mSv/yr.

Turhan, eref *et al.* [25] reported natural radioactivity in adhesive materials. In their study, U-238 activities were 7.3 to 69.4 Bq/kg whereas this study showed the ranges were 0 to 17 Bq/kg. Thus, the study adhesives were located within the worldwide ranges. In Turhan study, Th-232 activity was 2 to 57 Bq/kg in adhesives while, this study, showed the range of Th-232 was 4.9 to 12.4 Bq/kg. So, it can be stated that the study adhesives were within the worldwide range. K-40, in Turhan study, was ranging 21 to 816 Bq/kg whereas in this study was 0 to 183 Bq/kg. Therefore, the natural radioactivity in adhesives, by this study, were less than the worldwide demonstrated in **Table 9**. It can be stated that the study

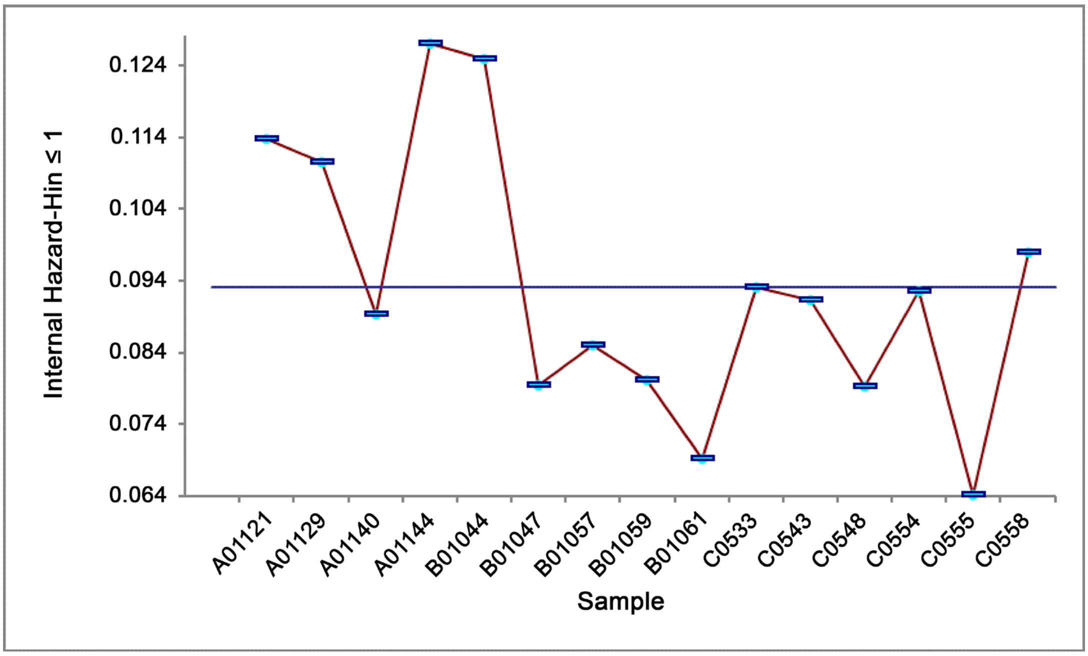

Figure 10. Internal Hazard values of the adhesive materials.

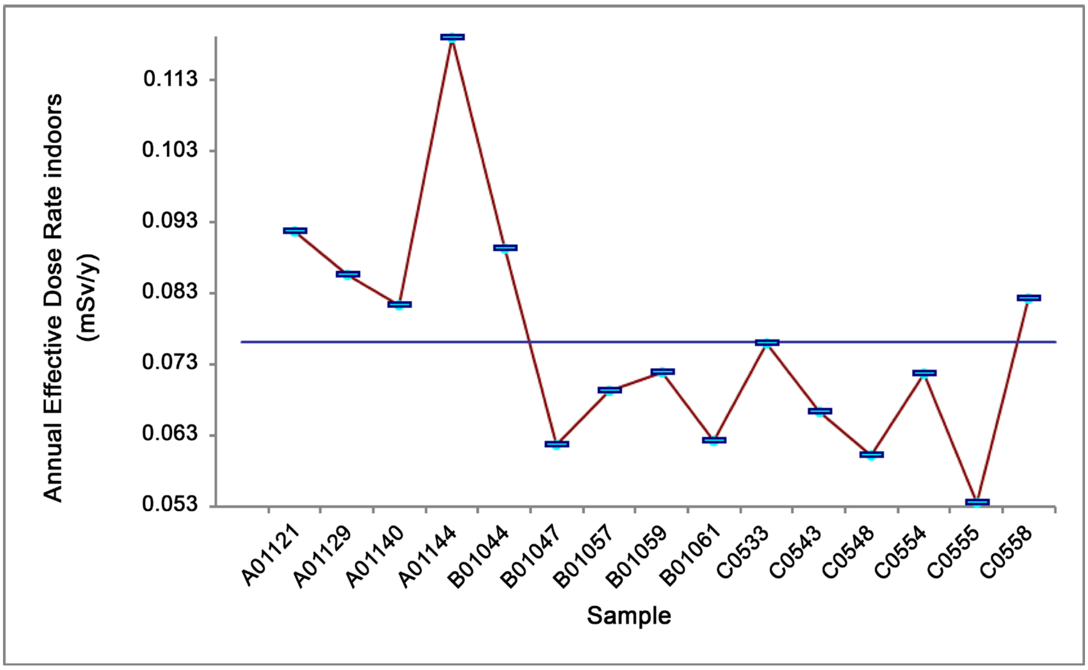

Figure 11. Radium equivalent values of the adhesive materials.

Table 8. Radiation calculations for Porcelain materials.

Sample code	K-40 Bq\Kg	Ra-226 Bq\Kg	Th-232 Bq\Kg	U-238 Bq\Kg	Ra eq	$H_{ex} \leq 1$	$H_{in} \leq 1$	α Concentration	α index	Outdoor dose	Annual Effective Dose (mSv/y)
A10118	904	86	100	71.3	299	0.81	1.04	1.09	0.43	140	0.96
A01119	567	62.4	67	61	202	0.55	0.71	0.73	0.31	94	0.65
A01125	323	46	45	0	135	0.37	0.49	0.49	0.23	63	0.43
B01034	811	86.8	101.1	100.3	294	0.79	1.03	1.07	0.43	137	0.94
B01037	42.5	0.1	0.1	0	3.52	0.01	0.01	0.02	0.00	2	0.01
B01040	514	43.2	77.3	53.6	193	0.52	0.64	0.70	0.22	89	0.61
A01131	226.7	12.9	8.8	6.06	42.94	0.12	0.15	0.16	0.06	21	0.14
A0133	621.7	48.4	51.3	55.4	170	0.46	0.59	0.63	0.24	80	0.55
A0134	914.3	55.00	57.40	57.30	207	0.56	0.71	0.78	0.28	99	0.68
A01136	938.5	57	58	37.9	212	0.57	0.73	0.79	0.29	102	0.70
A01139	678	44	49.0	40	166	0.45	0.57	0.62	0.22	79	0.54
A01141	618	84	47.7	60	200	0.54	0.77	1	0.42	94	0.65
B01048	831	57	55.8	45	200	0.54	0.69	0.74	0.28	95	0.66
B01050	352	32	27.6	38	98	0.27	0.35	0.36	0.16	47	0.32
C0540	682	72	88.9	76	252	0.68	0.87	0.91	0.36	117	0.80
C0545	497	135	126.4	116	354	0.96	1.32	1.25	0.68	162	1.11
C0547	485	39	42.8	37	137	0.37	0.47	0.50	0.19	65	0.44
C0549	787	123	93.9	92	318	0.86	1.19	1.14	0.62	148	1.02
C0552	321.2	59.1	57.2	51.2	166	0.45	0.61	0.59	0.30	76	0.52
Count	19	19	19	19	19	19.00	19.00	19.00	19.00	19.0	19.00
Mean	585	60	61	53	192	0.52	0.68	0.70	0.30	89.9	0.62
Stdev	252	**33**	32	31	89	0.24	0.33	0.32	0.17	41.1	0.28
Range	896	135	126	116	351	0.95	1.31	1.23	0.68	159.8	1.10
Minimum	43	0	0	0	**4**	0.01	**0.01**	0.02	**0.00**	1.9	**0.01**
25th Percentile (Q1)	352	43	45	38	137	0.37	0.49	0.50	0.22	64.6	0.44
50th Percentile (Median)	618	57	57	54	200	0.54	0.69	0.72	0.28	94.1	0.65
75th Percentile (Q3)	811	84	89	71	252	0.68	0.87	0.91	0.42	117.0	0.80
Maximum	939	135	126	116	354	0.96	1.32	1.25	0.68	161.6	1.11
95.0% CI Mean	463 to 706	44 to 76	45.4 to 76.1	37.5 to 67.6	149to 235	0.4 to 0.63	0.52 to 0.83	0.54 to 0.85	0.22 to 0.38	70.1 to 109	0.48 to 0.75
95.0% CI Sigma	190 to 373	25 to 48.9	24 to 47	23.5 to 46.14	67 to 131	0.18 to 0.35	0.24 to 0.48	0.24 to 0.47	0.12 to 0.24	31 to 60	0.21 to 0.41
Anderson-Darling Normality Test	0.24	0.45	0.35	0.36	0.32	0.32	0.28	0.27	0.45	0.27	0.27
P-Value (A-D Test)	0.75	0.25	0.44	0.42	0.51	0.51	0.60	0.64	0.25	0.63	0.63
Skewness	−0.42	0.61	0.11	0.09	−0.22	−0.22	−0.03	−0.31	0.61	−0.29	−0.29
P-Value (Skewness)	0.41	0.23	0.83	0.86	0.66	0.66	0.96	0.53	0.23	0.56	0.56
Kurtosis	−0.48	0.80	0.06	0.06	0.17	0.17	0.30	0.14	0.80	0.15	0.15
P-Value (Kurtosis)	0.72	0.34	0.77	0.76	0.68	0.68	0.60	0.71	0.34	0.70	0.70

Table 9. Comparison of activity concentrations and radium equivalent activities in tiles in the world [12].

Coutnry	Raeq	Dose	Annual effective		EAD	I	Hin	Hex
			indoor	outdoor				
China	306.40	141.92	0.70	0.17	1.33	1.11	1.13	0.83
China	332.76	152.16	0.75	0.19	1.41	1.20	1.16	0.90
Spain	171.49	80.69	0.40	0.10	0.75	0.64	0.59	0.46
U.A.E	179.77	81.20	0.40	0.10	0.75	0.65	0.57	0.49
Italy	243.06	108.03	0.53	0.13	1.01	0.84	0.93	0.66
Ave.	246.69	112.80	0.55	0.14	1.05	0.89	0.88	0.67
Min.	171.49	80.69	0.40	0.10	0.75	0.64	0.57	0.46
Max.	332.76	152.16	0.75	0.19	1.41	1.20	1.16	0.90

adhesive building materials were safe to be used in construction building materials in term of natural radioactivity.

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