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Radioactivity in building materials in Saudi Arabia: An overview of experimental method and Monte Carlo N-Particle (MCNP) calculation

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The aims of this work were clearly to assess the norms of radiation protection for building residents against natural radioactivity. This was done through measurement of natural radioactivity in building materials using gamma ray spectrometer. The annual effective dose (HR) linked to natural radioactivity was computed to estimate the radiation hazard in building materials. Obtained concentrations of these natural radionuclides and the calculated radiation hazard were compared with the national recommended values by natural limits by the Saudi standard code for radiation protection. The findings in this work of natural radioactivity levels were below the acceptable limits of 1 mSv/year which were found near the border of these limits. Therefore, it was found that the building materials may be safe to be used as construction materials. The annual effective doses were 0.8 ± 0.2 mSv/year for ceramics, 0.08 ± 0.02 mSv/year for adhesives, 0.6 ± 0.28 mSv/year for porcelains, 0.2 ± 0.1 mSv/year for marbles, 0.01 ± 0.01 mSv/year for paints, and 0.015 mSv/year for gypsum materials. The obtained results were compared with Monte Carlo N-Particle (MCNP) simulation. MCNP simulation was formulated to calculate the indoor gamma dose rate from the activity levels of the building materials which can take sample into very precise level. This computation was utilized to assess the uncertainty in the estimates. The results of MCNP were presented and an evaluation of the reported data shortly discussed. The radiation experimental values are in good agreement with the MCNP values, indicating that the obtained results are precise. Materials covered in the study are marbles, ceramics, adhesives, porcelain, paints, and locally produced cements.

Key words: Radioactivity, building materials, gamma ray spectrometer.

INTRODUCTION

Natural gamma radiations of indoor exposure owing to building materials, primitively quantified in building materials, is regarded as more significant than outdoor exposure. Natural radionuclides are always present in building materials but in various concentrations. Building materials often contain thorium and uranium decay series

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Table 1. The HPGe specifications.

Geometry	Co-axial open end closed end facing window
Diameter	74.7 mm
Length	92.9 mm
Active area window	11.6 mm
Operating voltage	4500V
Leakage current	0.01A
Amplifier gain	50
Amplifier ne	30-40
Pulse time	6 µs

radionuclides; therefore radiation exposure arises mostly from Th-$_{232}$, U-$_{238}$ series, and K-$_{40}$ (Dhanya et al., 2015; Usikalu et al., 2015; Mehra and Bala, 2014).

It is not only feasible but also more essential to assess the radiation hazard by computing indoor external dose by means of experimental and theoretical measurements. As the state-organization of King Abdualziz City for Science and Technology (KACST) is responsible for radiation protection by performing studies on natural radioactivity in dwellings, this work was mainly devoted and carried out to assess the contents of natural radioactivity in commercial building materials utilized in construction projects in Saudi Arabia. Also, the second aim of the work was to compare the obtained experimental data with Monte Carlo N-Particle (MCNP) mathematical model data.

Doses rates within buildings can only be detected with radiation measuring instruments like the high-resolution gamma-ray spectrometry system which consists of coaxial hyper-pure germanium (HPGe) or NaI detectors. It is also possible to quantify indoor exposure even before the building construction can take place. In this case, mathematical computations for example, MCNP, can be used to evaluate radiation doses from the reported data (Mehra and Bala, 2014; Abdo, 2010).

MCNP is the most widely-utilized method of trusted modeling of external radiation exposure in complex environments such as building materials. MCNP simulation can permit the regarding radiation transport with a very high value of precision. However, the main drawback of MCNP method is the requirement of having high performance computers (Al-Jundi et al., 2009). Koblinger (1978) was the first scientist who used MCNP method within a model to estimate dose rates in air at a point within model room. Although due to low performance of the 70's computers, the model is considered as standard and most highly appreciated model (Romano and Forget, 2013). Other researchers used different approaches. They used various methods of point-kernel integration over volume with analytical methods. The analytical methods can be easily applied for simple geometries whereas MCNP can be used for complex geometries (Zio, 2013).

EXPERIMENTAL WORK FOR GAMMA ANALYSIS

The collected samples were crushed and then homogenized. The homogenized samples were filled into 1000-ml Marinelli beakers which were later hermetically sealed with the help of commercial polyvinylchloride (PVC) to prevent the escape of airborne Rn-222 and Rn-220 from the samples. All the samples were accurately weighed and stored for a period of at least one month prior to determination in order to attain radioactive secular equilibrium between Ra-226 and Rn-222.

In this study, sample activities in building materials were measured using HPGe detector with highly passive shielding and low background located at KACST. The detector was cooled with liquid nitrogen cryostat to reduce the leakage current. To reduce the background radiation from natural sources, the detector was enclosed in a 10 cm thick cylindrical lead shield. The lead shield was graded with an inner layer of thick copper to reduce any influence of fluorescence. 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 8,000 channel multi-channel analyzer (MCA). The data acquisition, display, and analysis of spectra were carried out using Genie 2000 software.

The relationship between the channel numbers corresponding to absolute energies was determined. The specification of the used instrument is listed in Table 1. In this work, mixed gamma standard containing radionuclides were used for energy set of calibration. Gamma-ray energies covered the range from 50 to 1836 keV. The main gamma-ray energy lines of interest are shown in Table 2. The gamma energies used for Ra-226 was 186.2 keV and different energies of 295.2 and 351.9 keV were also used for Pb-214.

For gamma-ray spectrometry of unknown value, the detector efficiency measurement plays important role in gamma-counting. The full-energy peak efficiency can be computed through:

$$\varepsilon_f = \frac{N_p}{N_\gamma} \tag{1}$$

where "$_f$ 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 and can be calculated via:

$$N_\gamma = AP_\gamma \tag{2}$$

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

In order to remove interference between multi peaks, the calibration of energy efficiency was carefully carried. For every source, the energy efficiency was calculated using formula (1) and

Table 2. Gamma energies.

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

the energy channels was calculated.

MCNP calculations

The used standard living room, room with dimensions of 4 m × 4 m × 3 m (W x L x H), was defined for the proposed model as illustrated in Figure 1. In the room, the floor was covered with ceramic. Thermo-luminescent dosimeter (TLD) position was in the center of the room, precisely 2 m from each wall and 150 cm above the floor. The density of the used building materials was assumed to be 2325 kg per m^3. The thicknesses of the used building materials were assumed to be 0.20 m. The calculation of the dose rate conversion factors from our work was carried out based on MCNP code. The free-in air absorbed dose (nGy h^{-1}) value in the center of the room was obtained using MCNP in present study as:

$$D = 0.081A_{K-40} + 0.93A_{U-238} + 1.11A_{Th-232} \quad (3)$$

Where: D is the absorbed dose rate in the center of the room, 0.081, 0.93, and 1.1 nGy.kg.Bq are the dose conversion coefficients for K-40, U-238, and Th-232. The $A_{K\ 40}$, $A_{U\ 238}$, and $A_{Th\ 232}$ are the activity levels in unit of Bq/kg of K-40, U-238, and Th-232, respectively.

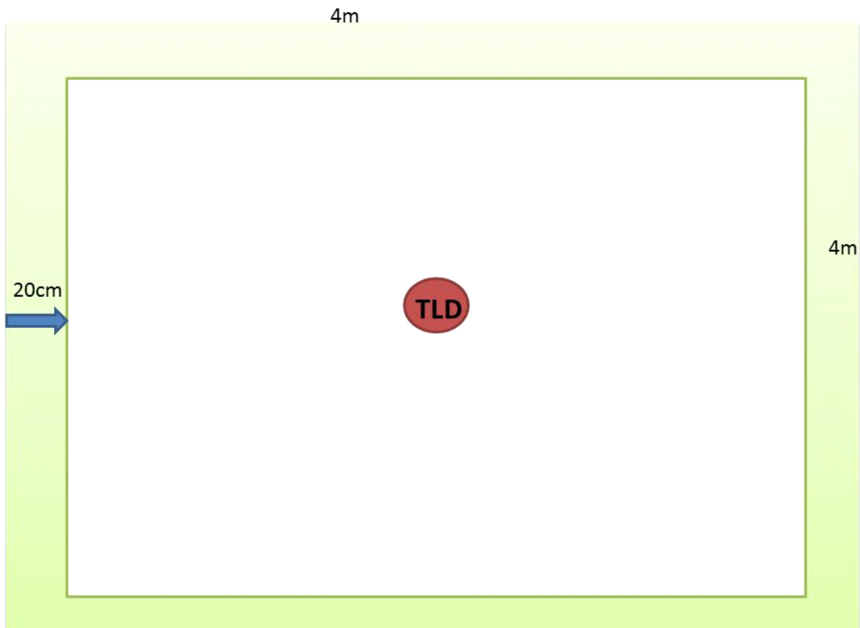

Figure 1. Room geometry model.

To compute the effective dose rate E in unit of mSv, the conversion factor 0.7 Sv/Gy is required for adult categories. The indoor occupancy factor used by UNSCEAR is 0.8 and the permittable indoor dose is 1 mSv/year. Therefore, the effective annual dose rate can be calculated via (Boda et al., 2013; Ravisankar et al., 2012; Atwood, 2013):

$$E(nSv) = D * 0.7(Sv/Gy) * 0.8 * 24 * 365(h/yr) \quad (4)$$

RESULTS AND DISCUSSION

Equation 3 obtained in the present work was compared with the ones reported in literature using MCNP method. Table 3 shows the dose evaluation was computed by direct measurement and from MCNP values reported by different authors. K-40 absorbed dose rate was experimentally 0.081 (nGy/h/Bq/kg) whereas the average reported values in literature was 0.079 (nGy/h/Bq/kg). Therefore, statistically the difference between the research's K-40 value and literature value was less than 2%. Similarly, U-238 absorbed dose rate was computed in this work to be 93 (nGy/h/Bq/ kg) and the literature value was on the average 0.75 (nGy/h/Bq/ kg). Moreover, Th-232 absorbed dose rate was 1.11 (nGy/h/Bq/kg) and the literature value reported in Table 3 was 1.09 (nGy/h/Bq/kg). Thus, the error in case of Th-232 absorbed dose rate between our reported results and literature values was less than 2%. It can be concluded that the present obtained results in Equation 3 are comparable to the values reported by different authors in Table 3. The induction of the present model and other well-known reported models in the literature implies that the present model and assumptions are in best agreement with other models.

In order to visualize the obtained simulated and experimental results, Figure 2 clearly indicated no difference between the simulated and experimental models owing to the fact that the regression line in Figure 2 is almost very close to a unit and implies that there is an excellent positive correlation between this experimental results and the simulated values. Therefore, MCNP simulation was found to be able to quantify the gamma ray and the absorbed dose in air for any marble materials. The gamma radiation due to natural radioactivity should be evaluated for room dimensions. The simulation results are in good agreement with experimental results.

For ceramic materials, the maximum annual effective dose was approximately 1.7 mSv/year which is slightly elevated while the min value was 0.9 mSv/year, as reported in Table 4. The ceramic materials were still in acceptable range of mode value of safe side although the max value was over a unit. As always expected, adhesive materials are made out of organic materials where no natural sedimentary materials are added. The annual effective dose value of max value was less than 0.23 mSv/year, which was less than the recommended values (1 mSv/year) by our national regulation and International Commission on Radiological Protection (ICRP).

The studied porcelain materials showed the maximum annual effective dose value of less than 1.41 mSv/year which is slightly above the recommended value of 1 mSv/year by ICRP, whereas the mode annual effective dose value was 0.62 mSv/year. Therefore, in considering

Table 3. Comparison of specific absorbed dose rates in publications and present work.

Absorbed dose rate in air (nGy/h.Bq/ kg)			Density (g/cm^3)	Wall thickness (cm)	Room dimension (mm^3)	Method used	Reference
Th-232	U-238	K-40					
1.11	0.93	0.081	2.32	20	4×4×3	MCNP	This Research
1.03	0.93	0.078	2.32	20	4×4×2.8	MCNP	Koblinger (1978)
1.11	0.92	0.078	2.35	20	4×4×2.8	Analytical	Stranden (1979)
1.05	0.89	0.078	2.35	20	4×4×2.8	Analytical	Mustonen (1984)
1.21	0.95	0.081	2.35	20	4×4×2.8	Analytical	Ahmad and Hussein (1998)
1.19	0.88	0.08	2.35	20	4×4×2.8	Analytical	Ademola and Farai (2005)
0.92	0.7	0.072	2.35	20	4×4×2.8	Analytical	Máduar and Hiromoto (2004)

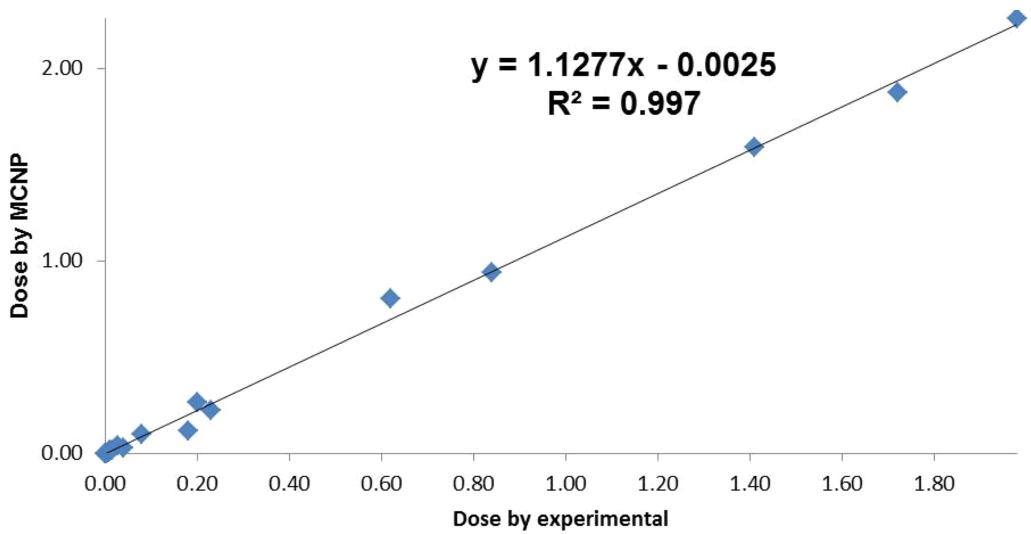

Figure 2. Experimental and simulation of gamma radiation e ective annual dose

the average value of annual effective dose, one can say that the porcelain materials are free of natural radioactive contaminations.

Similarly, the mode value of annual effective dose of marble materials was approximately 0.2 mSv/year which is slightly below the recommended value of 1 mSv/year by ICRP. Nevertheless, the max value of annual effective dose was about 2 mSv/year a result that is believed to be high. For paint materials, with similar procedures for adhesive materials, the max value of annual effective dose was less than 0.03 mSv/year and this value is far below the value of 1 mSv/year by ICRP. Thus, the paint materials are assumed to be safe against radiation hazard. For gypsum materials, the maximum reported value in this work was less than 1.1 mSv/year; hence they are regarded as safe materials for construction.

Table 4. Comparison of annual dose of experimental calculations and MCNP calculations.

Material	U-238 (Bq/kg)	SD	Th-232 (Bq/kg)	SD	Ra-226 (Bq/kg)	SD	K-40 (Bq/kg)	SD	Dose by MCNP (mSv/year)	Dose by experimental (mSv/year)
Ceramic										
Mean	65.15		71.3		81		636		0.939	0.84 0.28
Min	0	58	0	46.7	31.12	45	296	285	0.118	0.18
Max	148		136		150		1144		1.872	1.72
Adhesive										
Mean	8.69		7.1		10.49		44.3		0.096	0.08 0.02
Min	0	4.83	4.9	2.2	6.5	3	0	7.9	0.027	0.04
Max	17.4		12.4		18.1		183		0.22	0.23
Porcelain										
Mean	53		61		60		585		0.804	0.62 0.28
Min	0	31	0	32	0	33	43	252	0.017	0.01
Max	116		126		135		939		1.591	1.41
Marble										
Mean	12.1		21.77		13.5		220		0.261	0.2 0.3
Min	0	3.7	0	47.95	0.3	15.85	0	423	0	0
Max	53.8		254		57.7		1588		2.261	1.98
Paint										
Mean	2.27		0.1		2.75		3.2		0.012	0.01 0.01
Min	0	3.93	0	0.17	0.5	3.73	0	0.95	0	0
Max	6.8		0.3		7.05		8.9		0.036	0.029
Gypsum										
Mean	2.7		0.1		2.7		3.2		0.014	0.015
Min	-	10	0	0.01	0.7	4.5	0.7	-	0	0.004
Max	-		0.3		7.05		8		0.005	0.008

Conclusion

The estimated average annual effective doses were 0.8 mSv/year for ceramics, 0.08 mSv/year for adhesives, 0.6 mSv/year for porcelains, 0.2 mSv/year for marbles, 0.01 mSv/year for paints, and 0.015 mSv/year for gypsum materials. Fortunately, all of the reported annual effective dose values of the studied building materials were located within the safe limits of acceptable recommendation of less than 1 mSv/year in accordance with the national regulation and ICRP.

For the second part of this work, the radiation data reported by MCNP code and gamma laboratory showed that the radiation experimental data and radiation simulated data were comparable and matched because the drawn regression between both experimental and simulated data was 99.7%; implying that the matrix correlation between experimental and simulated data are excellently positive.

CONFLICT OF INTERESTS

The authors have not declared any conflict of interests.

REFERENCES

Abdo SYM (2010). The activity concentrations for some building materials in Yemen and simulation of the indoor gamma dose rate. The Degree of Master of Science Nuclear physics, Physics Department, Faculty of Science, Cairo University.

Ademola J, Farai I (2005). Annual effective dose due to natural radionuclides in building blocks in eight cities of southwestern Nigeria. Radiat. Prot. dosimetry 114(4):524-526.

Al-Jundi J, Ulanovsky A , Pröhl G (2009). Doses of external exposure in Jordan house due to gamma-emitting natural radionuclides in building materials. J. Environ. Radioact. 100(10):841-846.

Ahmad N, Hussein A (1998). Radiation doses in Jordanian dwellings due to natural radioactivity in construction materials and soil. J. Environ. Radioact. 41(2):127-136.

Atwood DA (2013). Radionuclides in the Environment, John Wiley & Sons.

Boda D, Csányi É, Gillespie D, Kristóf T (2013). Dynamic Monte Carlo simulation of coupled transport through a narrow multiply-occupied pore. J. Phys. Chem. C 118(1):700-707.

Dhanya B, Umadevi AG, Jose PA, JoJo PJ, Harikumar M, Sujata R (2015). Assessment of natural radioactivity level and radiation hazard parameters in the terrestrial environment of Eloor Island, Kerala. Int. J. Fund. Phys. Sci. 5(2):38-42.

Koblinger L (1978). Calculation of exposure rates from gamma sources in walls of dwelling rooms. Health Phys. 34(5):459-463.

Máduar MF, Hiromoto G (2004). Evaluation of indoor gamma radiation dose in dwellings. Radiat. Prot. Dosimetry 111(2):221-228.

Mehra R, Bala P (2014). Assessment of radiation hazards due to the concentration of natural radionuclides in the environment. Environ. Earth Sci. 71(2):901-909.

Mustonen R (1984). Methods for evaluation of radiation from building materials. Radiat. Prot. Dosimetry 7(1-4):235-238.

Ravisankar R, Vanasundari K, Chandrasekaran A, Rajalakshmi A, Suganya M, Vijayagopal P, Meenakshisundaram V (2012). Measurement of natural radioactivity in building materials of Namakkal, Tamil Nadu, India using gamma-ray spectrometry. Appl. Radiat. Isot. 70(4):699-704.

Romano PK, Forget B (2013). The OpenMC Monte Carlo particle transport code. Ann. Nucl. Energy 51:274-281.

Stranden E (1979). Radioactivity of building materials and the gamma radiation in dwellings. Phys. Med. Biol. 24(5):921.

Usikalu MR, Maleka PP, Malik M, Oyeyemi, KD (2015). Assessment of geogenic natural radionuclides contents of soil samples collected from Ogun State, Southwestern Nigeria. Int. J. Rad. Res. 13(4):355-361.

Zio E (2013). The Monte Carlo simulation method for system reliability and risk analysis, Springer. DOI: 10.1007/978-1-4471-4588-2.