การจัดประชุมเสนอผลงานวิจัยระดับบัณฑิตศึกษา มหาวิทยาลัยสุโขทัยธรรมาธิราช ครั้งที่ 5

The 5th STOU Graduate Research Conference

Determination of Radioactive Isotopes in Ceiling Paint Samples by Gamma-Ray Spectrometry

ลีเฮง ตัน (Lyheng Tan) [1] นเรศร์ จันทน์ขาว (Nares Chankow) [2] สมยศ ศรีสถิตย์ (Somyot Srisatit) [3]

Abstract

Some paint samples may contain radioactive materials that are intentionally added to give some special properties such as antifungus and ion generator but not showing in the compositions. The amount of added radioactive substance needs to be strictly controlled because the emitted radiation can be harmful to persons in the room. Our preliminary measurement with a simple survey meter showed that radiation can be detected from outside some ceiling paint cans. The main objectives of this research are to identify the radioactive isotopes and to determine the amount of each. Four ceiling paint samples are collected locally to analyze by using a gamma-ray spectrometer equipped with high purity germanium (HPGe) detector of 30% relative efficiency. Radioactive isotopes including potassium-40 (^{40}K), uranium daughters such as lead-214 (^{214}Pb) and bismuth-214 (^{214}Bi) as well as thorium daughters such as actinium-228 (^{228}Ac) and thallium-208 (^{208}Tl) were found in all samples. The specific activities of ^{40}K, ^{238}U, and ^{232}Th were found to be 26.48±18.87 to 39.67±21.29, 144.61±3.28 to 299.76±4.87, and 1773.59±23.66 to 2121.11±26.52 Bq/kg respectively. The estimated total dose rate from gamma-rays received by a person living in the room was found to be 0.13 to 0.16 μSv/h.

Keywords: Gamma-Ray Spectrometry, Specific activity, Radioactive Isotope, Ceiling paint

1 Student, M.Sc. in Nuclear Technology, Department of Nuclear Engineering, Chulalongkorn University: lyhengtan@yahoo.com

2 Associate Professor, Department of Nuclear Engineering, Chulalongkorn University: nares.c@chula.ac.th

3 Associate Professor, Department of Nuclear Engineering, Chulalongkorn University: somyot.s@chula.ac.th

Introduction

Radionuclides in Naturally Occurring Radioactive Materials (NORMs) consist primarily of material containing ^{40}K and isotopes belonging to the primordial series such as the long-lived isotopes ^{238}U in uranium series and ^{232}Th in thorium series which are present since the creation of the earth (4.5 billion years). Human beings can be exposed to ionizing radiation through external sources such as terrestrial radiation and cosmic radiation which irradiate the body with gamma photon, whereas the internal hazard requires the incorporation of radioactive materials into the body through ingestion or inhalation. Radon-222 (^{222}Rn) is a daughter product of radium-226 (^{226}Ra), which in turn is derived from uranium-238 (^{238}U). Thoron (^{220}Rn) is the daughter of thorium-232 (^{232}Th). ^{222}Rn and ^{220}Rn are naturally occurring radioactive gas which can cause lung cancer.

Nowadays ceiling paint may contain these radionuclides which can cause harmful to people and animals. According to the United State Environmental Protection Agency as well as the Scientific and Medical Communities recognized radon as a class A carcinogen (Schmidt, 2011, p. 3). Furthermore, The Environmental Protection Agency estimated that 14,000 American die every year from radon-related lung cancer, but this number could range from 7,000 to 30,000 deaths per year (EPA, 1993, p. 3). The data regarding the specific activities of ^{238}U, ^{232}Th, and ^{40}K in ceiling paint belonging to Thailand is not available in literature. Knowledge of radioactivity present in ceiling paint samples enables one to assess any possible radiological risk to human health (Kumar et al, 2003, p. 465). Therefore, the specific activity of these radionuclides were firstly determined in our low background HPGe gamma-ray spectrometer NucMAT, Nuclear Tech at CU with a CANBERRA lead shield (coated inside by the thin layer of copper). This semiconductor detector is the best choice for this research because of its superior energy resolution. Moreover, the gamma-ray spectrometry of this detector is a powerful method that can identify and quantify the radionuclides for determining the specific activity of gamma emitting isotopes in a variety of matrices. According to the rule that all exposure to radiation should be kept “as low as reasonably achievable” (ALARA), and the results obtained in the present study are compared with the relevant results available in some other countries of the world.

Objective and scope of this research

1. To identify the radioactive isotopes and to determine the amount of each radioactive isotope in ceiling paint samples by using a gamma-ray spectrometer equipped with High Purity Germanium (HPGe) detector.
2. To assess gamma-ray dose rate from the room ceiling painted with the paint containing radioactive isotopes.

Experimental Procedure

Sampling and Samples Preparation

From our preliminary survey using a simple GM pancake survey meter to measure radiation outside the paint cans, it was found that two kinds of paint samples are radioactive. They were ceiling paints of the same brand having different colors i.e. ash grey and white. Firsty, two samples were bought, one for each color weighing approximately 3.8 kg. Furthermore, two more samples were later bought from another shop to obtain samples from different lots. Therefore, there were four samples for the measurements. The samples W1 and W2 were the white ceiling paints while the samples G1 and G2 were the grey ceiling paints. An interior paint sample was also bought to be used as our blank because it was not radioactive. Furthermore, 312, 314, 313, 336, and 276 g of samples W1, W2, G1, G2, and blank respectively were taken from the cans to fill in a 3”diameter-2” height plastic container. Then they were sealed to prevent escape of radioactive radon gas and left for at least 30 days to obtain radioactive equilibrium between uranium-238 (^{238}U) and its daughters as well as thorium-232 (^{232}Th) and its daughters (Abudlkarim et al, 2013, p. 867).

Measurement of gamma-rays

A gamma spectrometer equipped with a high purity germanium (HPGe) semiconductor detector Canberra model GC3021 was used for the measurements. It had a relative efficiency of 30%. To reduce the natural gamma-ray background level, the detector was installed inside a low background lead shield with 10 cm thickness as illustrated in Figures 1 and 2. The detector was connected to a digital spectrum analyzer (DSA) which was connected with a notebook computer. Canberra Genie-2000 Gamma Analysis Software run on the notebook computer was used to control the measurement and to perform data acquisition and spectrum analysis. The Standard Reference Materials including RGK-1 (potassium standard), RGU-1 (uranium ore standard), and RGTh-1 (thorium ore standard) obtained from the International Atomic Energy Agency (IAEA) were used in this research.The specific activities of RGK-1, RGU-1, and RGTh-1 were 2928.55 Bq/kg with the weight of 225.87 g, 4903.47 Bq/kg with the weight of 324.15 g, and 3257.89 Bq/kg with the weight of 313 g respectively. These Standard Reference Materials were filled in a 3"diameter-2" height plastic container and then were kept as the same samples. Three Standard Reference Materials, four samples, and a blank (an interior paint sample) were used for the measuremnts, and they were placed on top of the HPGe detector for measurement one after the other for 7,200 seconds. All spectra were then analysed to identify radioactive isotopes from their energy peaks and to calculate the amount of potassium, uranium and thorium from the net peak intensities.

Figure 1: Lead shield to lower gamma background level

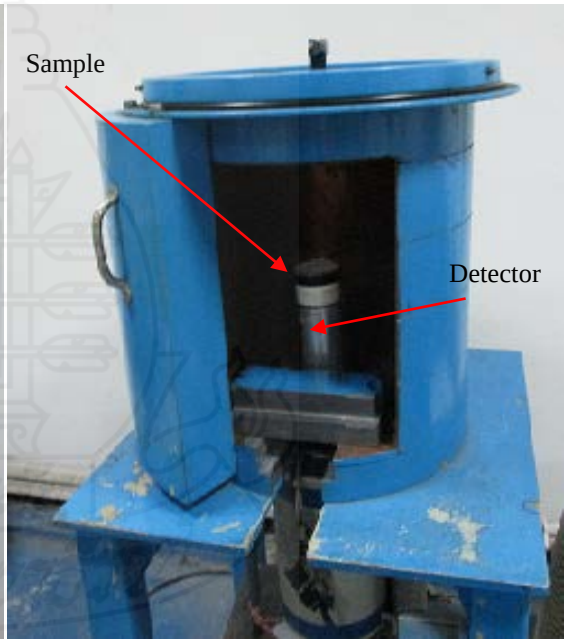

Figure 2: Showing HPGe detector indisde lead shield

Data Analysis and Results

The obtained spectra of all four ceiling paint samples showed peaks of potassium-40 (^{40}K), uranium daughters such as lead-214 (^{214}Pb) and bisbmuth-214 (^{214}Bi) as well as thorium daughters such as actinium-228(^{228}Ac) and thallium-208 (^{208}Tl). The spectra indicated that there were potassium, uranium and thorium in the ceiling paint samples. ^{40}K is the only natural radioactive isotope of potassium. There is 0.0118 percent by atom of ^{40}K in natural potassium with a half-life of 1.3 x 10^{13} years. ^{40}K decays mainly (~90%) to ^{40}Ca by beta emission and about 10% to ^{40}Ar by electron captures followed by emission of 1.46 MeV gamma-rays to ^{40}Ar at ground state . Potassium presents in soil, fertilizer, wood, food and even in human body.The specific activity of ^{40}K in 1 g of natural potassium is

about 30 becquerrel (Bq) or 30,000 Bq per kg. Uranium and thorium mainly present in soil and rock. Products having raw materials from soil and rock always contain uranium and thorium such as construction materials, ceramic tiles, fertilizers, coals, etc. Normally paint samples should not contain uranium and thorium at a detectable level except they are intentionally added.

From our measurement, the amount of potassium, uranium and thorium could be obtained from the following relationships.

Sample Activity = (Std Activity × Sample Area × Mass Std)/ (Std Area × Mass Sample) (1)

Where,

- Std and Sample Area are the net peak area per second of the standard and sample respectively
- Mass Std and Sample are the mass in gram of the standard and sample respectively
- Std Activity is the specific activity of standard expressed in (Bq/kg)
- Sample Activity is the specific activity of sample expressed in (Bq/kg)

Specific Activity

The uncertainties of these specific activities were determined using the error propagation formula (Ahmad. 1997, p. 13; Knoll. G. F., 2000. p. 89).The specific activities of the radionuclides ranged from 26.48±18.87 to 2124.55±26.30 Bq/kg for all samples as be shown in Table I below.

TABLE I: The Specific Activity (Bq/kg±1σ) of Some Natural Radionuclides (Counting Time 7,200 s)

Samples (Activity)	^{40}K (E=1458 keV)	^{214}Pb (E=352 keV)	^{214}Bi (E=609 keV)	^{228}Ac (E=338 keV)	^{228}Ac (E=911 keV)
W1	39.67±21.29	173.43±3.24	176.87±3.86	2117.66±26.73	2124.55±26.30
W2	29.49±19.43	169.35±3.22	171.88±3.82	2035.09±26.11	2018.70±25.50
G1	26.48±18.87	140.00±2.94	149.22±3.58	1754.73±23.70	1792.45±23.61
G2	35.22±20.97	293.45±4.40	306.07±5.30	2013.57±27.17	2116.97±27.64

The highest specific activity of these radionuclides was found in sample W1 (2124.55±26.30 Bq/kg) in ^{232}Th series of ^{228}Ac (E=911 keV), but the lowest specific activity was found in sample G1 (26.48±18.87 Bq/kg) in ^{40}K (E=1458 keV). The specific activity of ^{40}K (E=1458 keV) in all samples ranged from 26. 48±18.87 to 39.67±21.29 Bq/kg. Moreover, in ^{238}U series, the specific activity of ^{214}Pb (352 keV) and ^{214}Bi (609 keV) in all samples ranged from 140.00±2.94 to 293.45±4.40 Bq/kg and 149.22±3.58 to 306.07±5.30 Bq/kg respectively. Furthermore, in ^{232}Th series, the specific activity of ^{228}Ac (338 keV) and ^{228}Ac (911 keV) ranged from 1754.73±23.70 to 2117.66±26.73 Bq/kg and 1792.45±23.61 to 2124.55±26.30 Bq/kg respectively.

The gamma-rays spectra of the standardard reference materials of RGK-1, RGU-1, and RGTh-1 were be shown below. There was only one peak intensity of RUK-1 at E=1458 keV, so the specific activity of ^{40}K was calculated based on this energy directly. For RGU-1 and RGTh-1, there are several peak intensities emitting from several radionuclides. However, the specific activity of ^{238}U was calculated based on the radionuclides of ^{214}Pb (E=352keV) and ^{214}Bi (E=609 keV), and the specific activity of ^{232}Th was calculated based on the radionuclides of ^{228}Ac (E=338 keV) and ^{228}Ac (E=911 keV). Their peak intensities represented the emitted radiation from the radionuclides with their energy in keV.

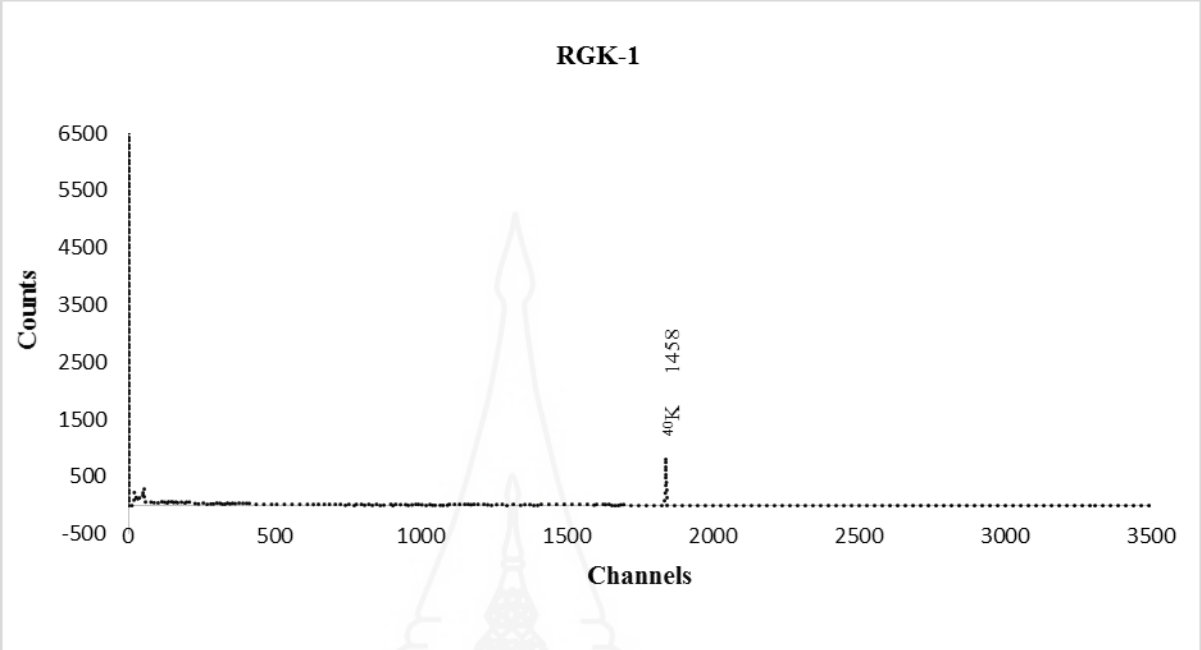

Fig. 3: Characteristic Gamma Spectrum of RGK-1 with Counting Time 7,200 s

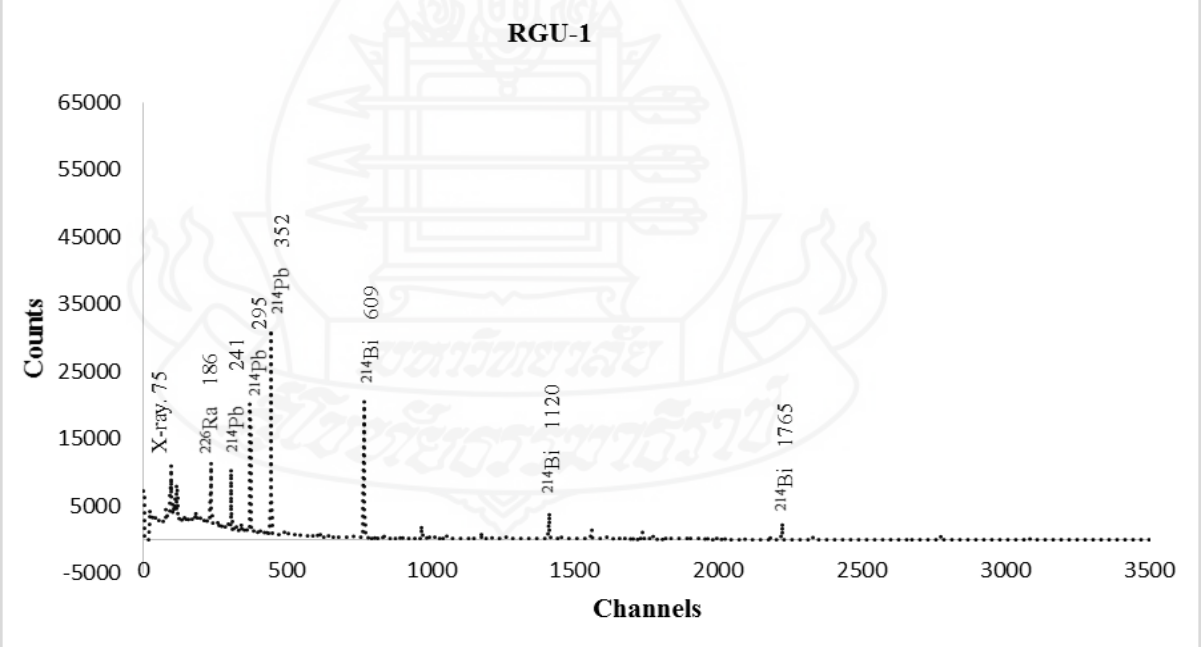

Fig. 4: Characteristic Gamma Spectrum of RGU-1 with Counting Time 7,200 s

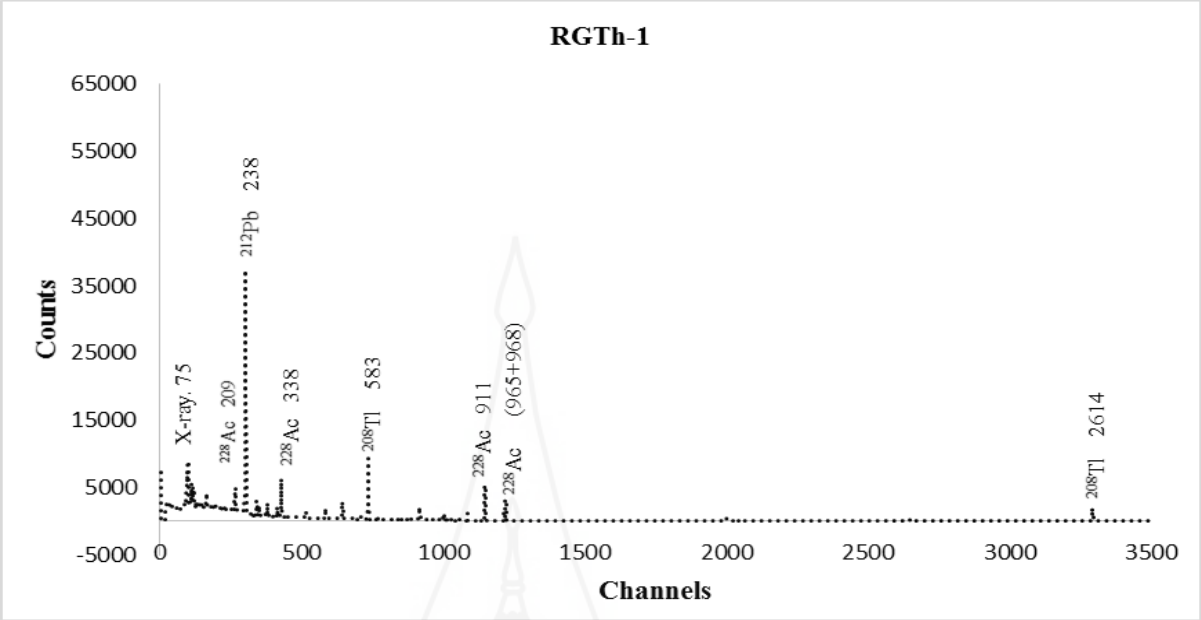

Fig. 5: Characteristic Gamma Spectrum of RGTh-1 with Counting Time 7,200 s

The gamma spectra of four samples and a blank were shown below. The gamma spectrum of the blank showed very low peak intensities of radionuclides while the spectra of four samples showed several energy peaks with high peak intensities. Their peak intensities represented the emitted radiation from the radionuclides with their energy in keV.

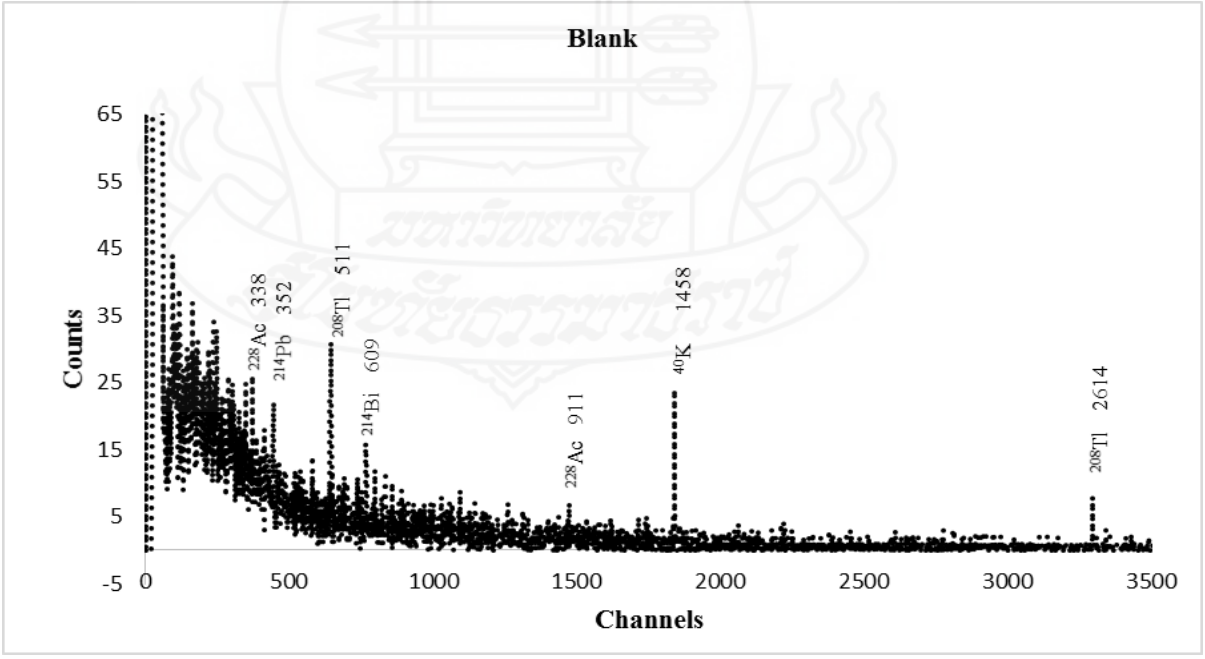

Fig. 6: Characteristic Gamma Spectrum of blank (interior paint sample) with Counting Time 7,200 s

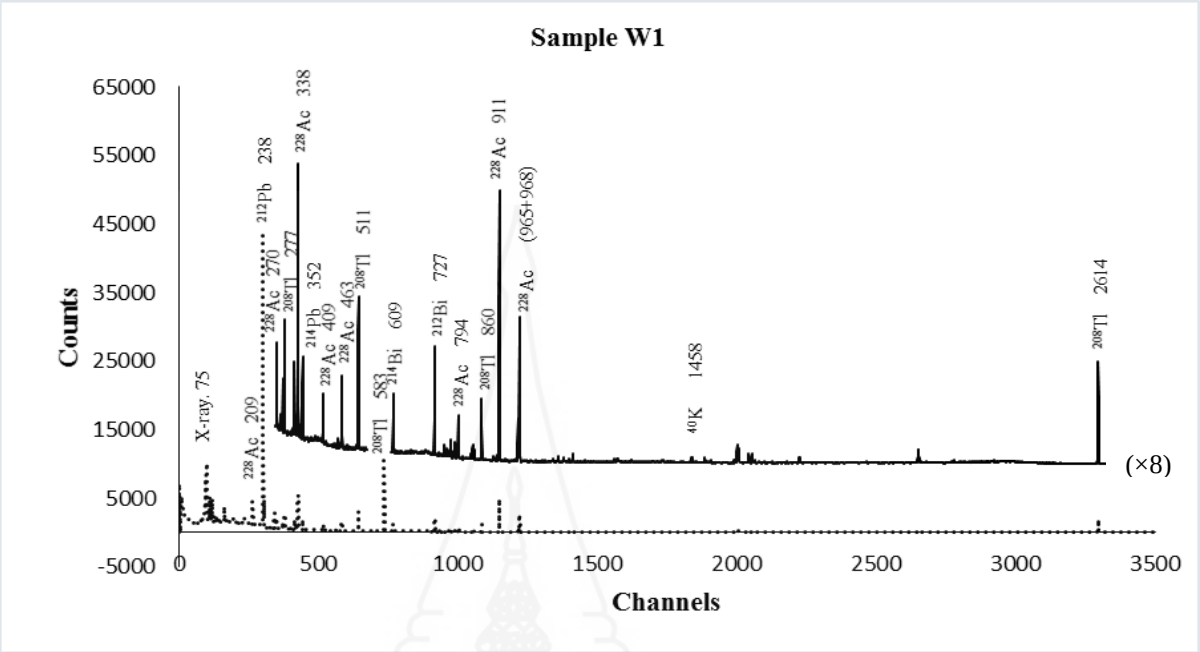

Fig. 7: Characteristic Gamma Spectrum of Ceiling Paint Sample W1 with Counting Time 7,200 s
(The solid line spectrum above the dotted spectrum was increased eight times)

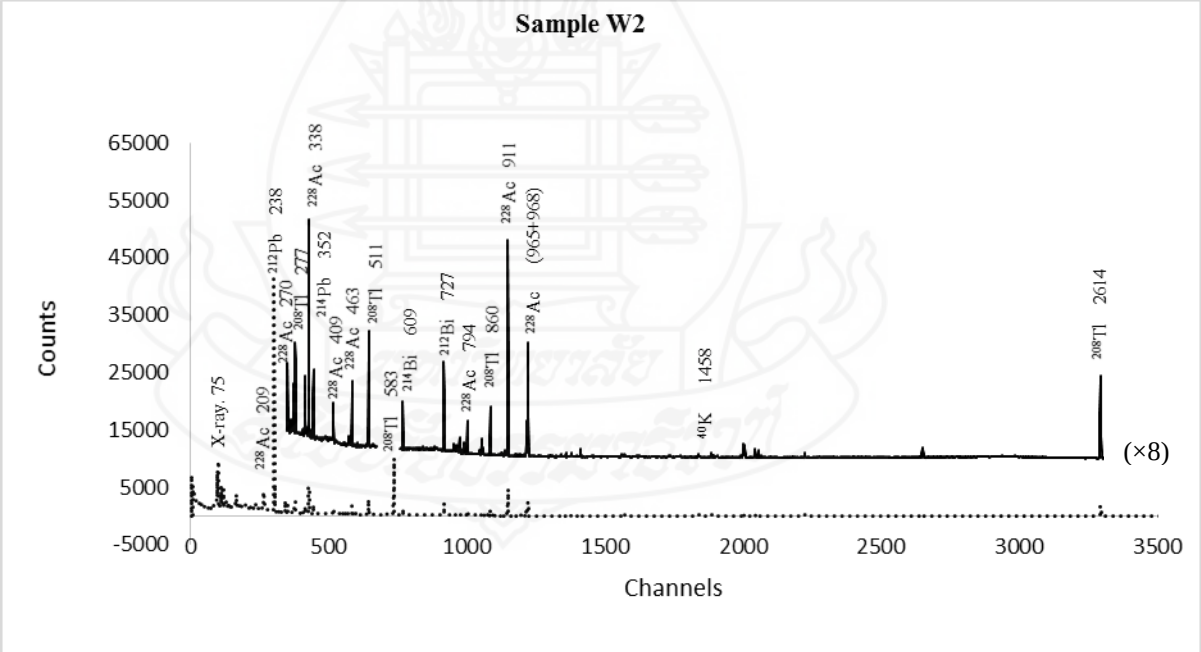

Fig. 8: Characteristic Gamma Spectrum of Ceiling Paint Sample W2 with Counting Time 7,200 s
(The solid line spectrum above the dotted spectrum was increased eight times)

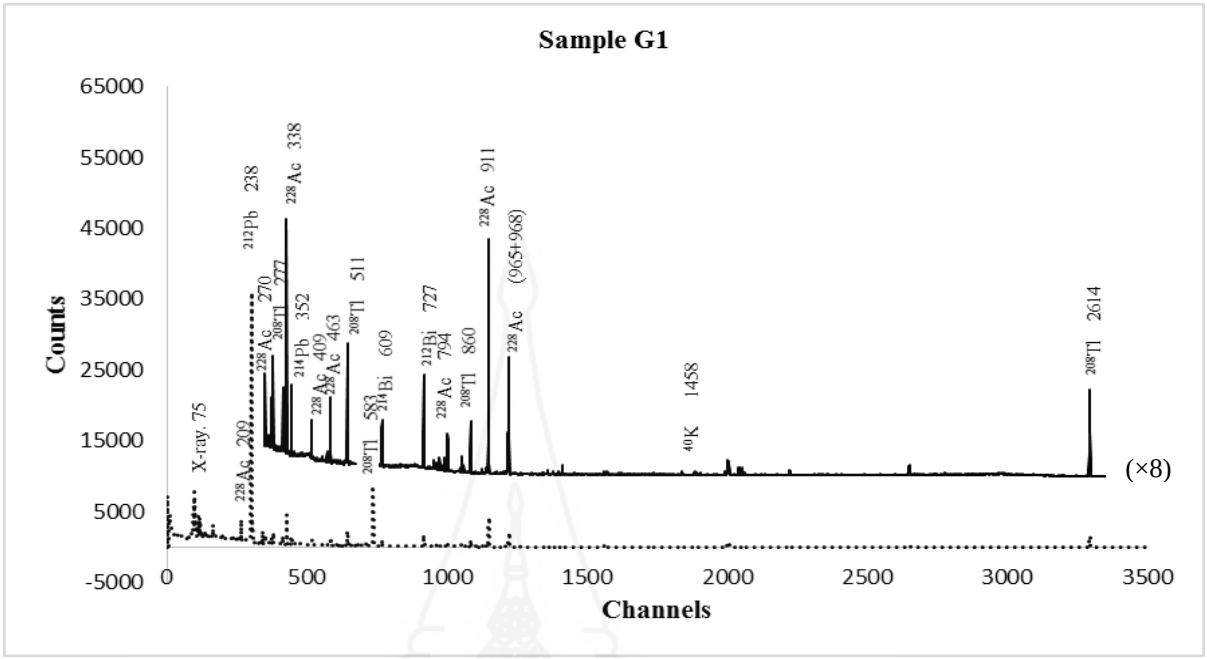

Fig. 9: Characteristic Gamma Spectrum of Ceiling Paint Sample G1 with Counting Time 7,200 s
(The solid line spectrum above the dotted spectrum was increased eight times)

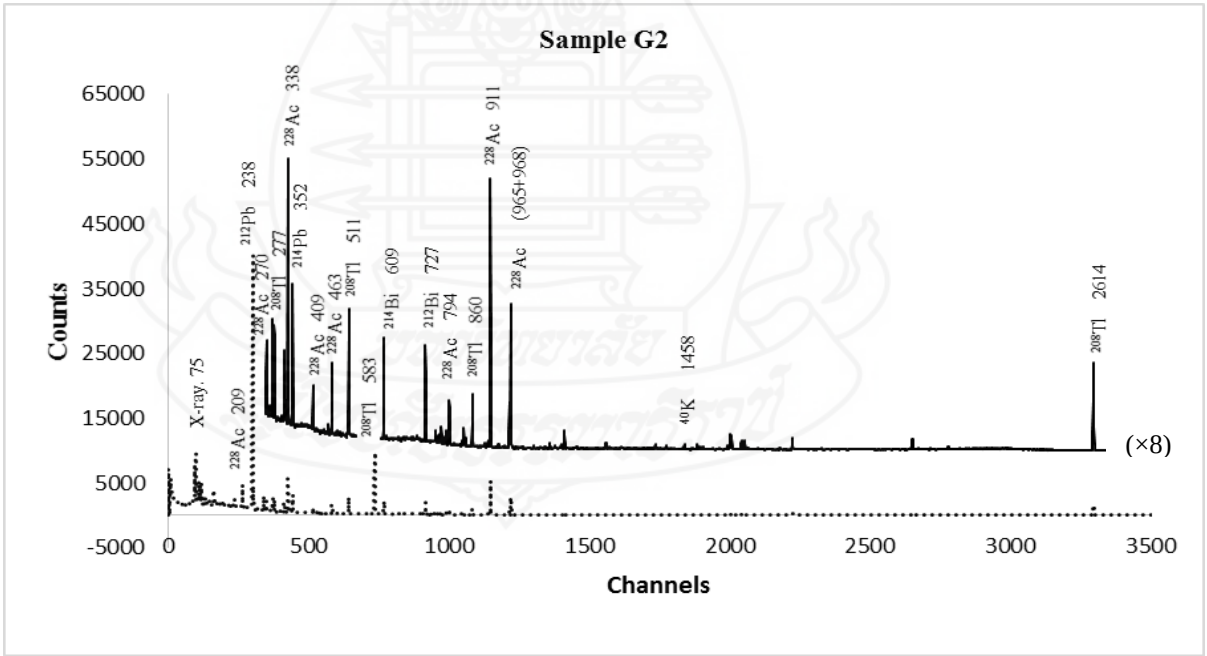

Fig. 10: Characteristic Gamma Spectrum of Ceiling Paint Sample G2 with Counting Time 7,200 s
(The solid line spectrum above the dotted spectrum was increased eight times)

The peak intensities of ^{214}Pb (E=352 keV) and ^{214}Bi (E=609 keV) in uranium ore standard were higher than the speak intensities of ^{214}Pb (E=352 keV) and ^{214}Bi (E=609 keV) in all samples, and the peak intensity of ^{40}K in potassium standard was higher than the peak intensity of ^{40}K in all samples. Moreover, the peak intensities of ^{228}Ac (E=338 keV) and ^{228}Ac (911 keV) in thorium ore standard were slightly higher than the peak intensities of ^{228}Ac (E=338 keV) and ^{228}Ac (911 keV) in all samples.

The gamma spectrum of the blank in Fig. 6 showed that the backround was very low. This means that this blank, the interior paint sample, was not radioactive. However, the spectra of four samples such as W1, W2, G1, and G2 were shown that there were several peak intensities emitting from different radiounclides with their individual energy in keV. The peak intensities of those radionuclides were found out that they were the daughters of ^{238}U, and ^{232}Th series because the energy peaks of them were the same as the energy peaks of uranium and thorium standards. Moreover, the peak intensity of ^{40}K for all samples was very low in comparision with other peak intensities.

In this study, the specific activity of ^{238}U was calculated as ^{226}Ra (The parent of ^{222}Rn) activity based on the arithmetic mean values of its decay products (^{214}Pb and ^{214}Bi) in radioactive equilibrium with their parent. In the ^{238}U decay series, both ^{214}Pb and ^{214}Bi are consider as ^{226}Ra indicators corresponding with radium mobility in geological times because of geochemical reasons (Corte et al., 2005, p. 590). In secular equilibrium, the specific activity of ^{220}Rn was equal to the specific activity of its parent, ^{232}Th.

The uncertainties of these specific activities were determined using the error propagation formula. The specific activity and mean specific activity of all radionulcides for all samples ranged from 26.48±18.87 to 2121.11±26.52 Bq/kg and 32.71±20.17 to 1996.72±25.88 respectively with the counting time of 7,200 s as be shown in Table II below. Moreover, the mean specific activity of these radionuclides for all samples was also calculated.

TABLE II: The Specific Activity and Mean Specific Activity (Bq/kg±1σ) of ^{238}U or ^{226}Ra, ^{232}Th, and ^{40}K

Samples (Activity)	^{40}K	^{238}Uor ^{226}Ra	^{232}Th
W1	39.67±21.29	175.15±3.56	2121.11±26.52
W2	29.49±19.43	170.62±3.53	2026.90±25.81
G1	26.48±18.87	144.61±3.28	1773.59±23.66
G2	35.22±20.97	299.76±4.87	2065.27±27.41
Mean Specific Activity	32.71±20.17	197.53±3.86	1996.72±25.88

In secular equilibrium, the specific activity of ^{238}U is equal to the specific activity of ^{226}Ra as be shown in Table II. The highest specific activities of ^{232}Th and ^{40}K were found in sample W1 (2121.11±26.52 and 39.67±21.29 Bq/kg respectively), and the highest specific activity of ^{238}U was found in sample G2 (299.76±4.87 Bq/kg). The lowest specific activities of ^{40}K, ^{238}U, and ^{232}Th were found in sample G1 (26.48±18.87, 144.61±3.28, and 1773.59±23.66 Bq/kg respectively. The mean specific activities of ^{238}U and ^{232}Th (197.53±3.86 and 1996.72±25.88 Bq/kg respectively) of these four samples were higher than the corresponding world mean values for building materials (50 and 50 Bq/kg respectively), but the mean specific activity of ^{40}K (32.71±20.17 Bq/kg) of these four samples was lower than the corresponding world mean values for building mateials (500 Bq/kg) (UNSCEAR, 1993, p. 41).

Estimation of the Gamma Dose Rate

The gamma dose rate was calculated due to the gamma ray emission from the radionuclides such as ^{40}K, ^{238}U series, and ^{232}Th series in the ceiling paint. It is estimated that the ceiling areas were 4 m × 4 m = 16m^2 (Radius = 2 m) with the height 1.5 m between the ceilings and people. Thus the dose rate was estimated using the following formula:

$$X = 0.0659 \times I \times E \times (\mu_a/\rho)^{air} \quad (3)$$

Where, X is the exposure rate expressed in (mR/h), I is the gamma ray intensity expressed in (photon/cm^2-sec), E is the gamma ray energy expressed in (MeV), and $(\mu_a/\rho)^{air}$ is the mass absorption coefficient of air at the energy E expressed in (cm^2/g) (Larmarsh and Baratta, 2001, pp. 511-539).

The gamma ray intensity was calculated using the flux or gamma ray intensity from a planar disc source.

$$I = (S/4) \times \ln[1 + (R^2/X^2)] \quad (4)$$

Where, S is the activity expressed in gamma-ray isotropically per cm^2-sec, R is the radius of the ceiling expressed in (m) and X is the distance between source and people expressed in (m). The exposure rate (mR/h) can be converted to the dose rate (μSv/h).

According to the experiment, 0.005 kg/cm^2 the ceiling paint was used. Therefore, the activity (Bq/cm^2) for samples W1, W2, G1, and G2 were be shown in Table III.

TABLE III: The Specific Activities (Bq/cm^2±1σ) with Their Uncertainties of the Samples W1, W2, G1, and G2

Samples (Activity)	^{40}K	^{238}U or ^{226}Ra	^{232}Th
W1	0.20±0.11	0.88±0.018	10.61±0.13
W2	0.15±0.0.097	0.85±0.018	10.13±0.13
G1	0.13±0.094	0.72±0.016	8.87±0.12
G2	0.18±0.10	1.499±0.024	10.33±0.14

Due to the activities (in Bq/cm^2) in Table III, the gamma intensity can be calculated using the formula in equation (4), and the dose rate can be estimated using the formula in equation (3).

TABLE IV: The Gamma Dose Rate (μSv/h±1σ) with Their Uncertainties of Samples W1, W2, G1, and G2

Samples	Dose Rate (μSv/h)
W1	0.16±0.0012
W2	0.15±0.0011
G1	0.13±0.0010
G2	0.15±0.0012

The dose rate of these four samples ranged from 0.13±0.0010 to 0.16±0.0012 μSv/h. The highest gamma dose rate was found in sample W1 (0.16±0.0012 μSv/h), while the lowest gamma dose rate was found in sample G1 (0.13±0.0010 μSv/h). The mean of these estimated gamma dose rate (0.15±0.0011 μSv/h) was about two times higher than the world average (populated weighted) indoor absorbed gamma dose rate of 0.084 μSv/h (Turhan and Gunduz, 2007, p. 341). All the samples exceeded the world average.

Conclusion

In these experiments, the low level background lead shield was used, and the measurements were performed with an HPGe detector in order to acquire the gamma spectra for the measurement samples. By knowing the specific activities of the standards, we can determine the specific activities of the samples using the relationship in equation (1). The highest specific activity of ^{238}U was found in sample G2 (299.76±4.87 Bq/kg), and the highest specific activities of ^{232}Th and ^{40}K were found in sample W1 (2121.11±26.52 and 39.67±21.29 Bq/kg respectively). The lowest specific activities of ^{238}U, ^{232}Th, and ^{40}K were found in sample G1. The estimated total dose rate from gamma-rays received by a person living in the room was found to be 0.13 to 0.16 μSv/h. The mean of these estimated gamma dose rate (0.15±0.0011 μSv/h) was about two times higher than the world average (populated weighted) indoor absorbed gamma dose rate of 0.084 μSv/h. Moreover, the mean specific activity of ^{40}K (32.72±20.17 Bq/kg) was lower than the corresponding world mean values (500 Bq/kg), while the mean specific activities of ^{238}U and ^{232}Th (197.53±3.86 and 1996.72±25.88 Bq/kg respectively) were higher than the corresponding world mean values (50 and 50 Bq/kg respectively). However, the contribution of ceiling paints to the total specific radioactivity of the ceiling becomes relatively small because of using the small portions. In conclusion, the use of paints needs some careful regulations to reduce the hazardous radiation into the environment.

Suggestion

It is suggested that measurements of radionuclides in ceiling paint samples should be conducted for the further research, and the results can be considered as the base values to be used as reference information.

References

1. Schmidt, K. (2011). Commercial radon survey report. Radon detection Sspecialists, Inc. Burr Ridge, Illinois. Searched on September 9, 2015, from http://www.d107.org/media/Administration/FOIA/2014-2015/Cooper/ES%20May%202006%20Radon%20Results.pdf
2. EPA. (1993). Radon measurement in schools. United State environmental protection agency. Searched on August 10, 2015, from http://www.epa.gov/radon/pdfs/radon_measurement_in_schools.pdf
3. Kumar, A., Kumar, M., Singh, B., & Singh, S. (2003). Natural activity of ^{238}U, ^{232}Th, and ^{40}K in some Indian building materials. Elsevier science Ltd. Science direct. Paper no 36. Retrieved 3 August 2015 from http://www.sciencedirect.com
4. Abduldarim, M. S., Sadiq. U., & Zakari. I. Y. Determination of radionuclides in building blocks made in Kaduna Metropolis. IJST explore. Centre of professional research publications. 2. Paper no 12 (December). Retrieved June 21, 2015 from http://www.journalofsciences-technology.org
5. Turhan, S., & Gunduz. L. (2007). Determination of specific activity of ^{226}Ra, ^{232}Th, and ^{40}K for assessment of radiation hazards from Turkish pumice samples. Elsevier Science Ltd. Science Direct. Paper no. 99. Retrieved August 15, 2015 from http://www.sciencedirect.com
6. Ahmad, N., Matiullah & Hussein, A. J. A. (1997). Natural radioactivity in Jordanian soil and building materials and the associated radiation hazards. Elsevier science Ltd. Science direct. 39. Paper no 1. Retrieved 10 August, 2015 from http://www.sciencedirect.com
7. Knoll. G. F. (2000). Radiation detection and measurement, (3rd ed). Counting statistics and error prediction. John Wily and Sons. New York. USA: Quebecor Printing.
8. De Corte, F., Umans, H. De Wispelaere, A. D., & Van den haute. P. (2005). Direct gamma-spectrometric measurement of the ^{226}Ra 186.2 keV line for detecting $^{238}U/^{226}Ra$ disequilibrium in determining the environmental dose rate for the luminescence dating of sediment. Elsevier science Ltd. Science direct. Paper no 63. Retrieved 10 August, 2015 from http://www.sciencedirect.com
9. UNSCEAR. (1993). Sources and effects of ionizing radiation. United Nations scientific committee on the effects of atomic radiation. United Nations Publication, New York, USA.
10. Lamarsh, R. J., & Baratta, J. A. (2001). Introduction to nuclear engineering, (3rd ed). Radiation protection. New Jersey, USA: Prentice-Hall.