MONTE CARLO SIMULATION FOR THE EFFICIENCY OF THE RGU-1 STANDARD IN URANIUM MEASUREMENT IN SAMPLE DIFFERENT ENVIRONMENTAL MATRIX

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Received: 12-03-2020
Revision Received: 20-03-2020
Accepted: 15-08-2021

DOI :
10.17146/jstni.2022.23.2.6944

Keywords: Gamma ray, Spectrometer, RGU-1, Uranium, MCNP

Abstract *Preliminary Study of Plutonium Utilization in AP1000 Reactor Use SRAC 2006 and JENDL 3.3 has been conducted. Nuclear energy, especially for nuclear reactor, become important this day because the need of energy will increase along with the increasing of human population, the advanced technology and economic. The more nuclear reactor operated the more existence of plutonium stockpile. This study evaluated the standard of Westinghouse AP1000 reactor and ZrB2 as Integral Fuel Burnable Absorber (IFBA). Different fuel compositions of assembly type were analyzed in by using SRAC 2006 code system with JENDL 3.3 nuclear data library. This study aiming to compare the neutronics characteristics of an UO_2 and an $(U,Pu)O_2$ assembly designs. Some results of the study show that optimal criticality of the fuel assembly can be accomplished by using 5% enrichment of U-235 for UO_2 fuel and 9% plutonium fraction for $(U,Pu)O_2$ fuel assembly.*

INTRODUCTION

Uranium is a natural radionuclide so that it can be contained in various environmental samples with various concentrations. The average concentration of uranium in nature is 2 ppm, but there are some high and low depending on geological conditions. Uranium concentration in the environment will affect the high rate of radiation exposure in the region. The high rate of exposure will increase the risk of radiation especially if the material contained in uranium and high torium enters the human body through breathing or digestion (1).

The most uranium isotopes in nature are ^{238}U, 99.3%, while for ^{235}U around 0.7%. ^{238}U is a primordial radionukeuclide with a decay time of 4.46×10^9 years and produces a nuclear of a lot of daughters form its decay. ^{238}U daughter's is mostly radioactive so that it emits radiation with various types and energy and abundance of various gamma energy as shown in Table 1.

Radioisotope ^{238}U in environmental samples can be measured using various methods, one of which is by spectrometry gamma. Isotopes ^{238}U is an alpha transmitter and a little gamma, while the daughters are diverse, some are the transmitters of the alpha, beta, and also gamma. With the number of ^{238}U daughter who are gamma emitters, the Gamma spectrometry can be used effectively as a method in measurement ^{238}U. Gamma rays have a large translucent power and can be easily detected by the gamma detector so that the Gamma Spectrometry Method is quite competitive compared to other methods.

Table 1. List of Gamma Energy for Sample Radiation Sources (2, 3)

No.	Nuclide	Energy *(KeV)*	Abundance *(%)*
1	^{210}Pb	46	4.25
2	^{234}Th	63	3.7
3	^{234}Th	92	4.2
4	^{226}Ra	186	3.6
5	^{214}Pb	242	7
6	^{214}Pb	295	18
7	^{214}Pb	352	36
8	^{214}Bi	609	45
9	^{214}Bi	934	3.2
10	^{214}Bi	1120	15.3

The gamma spectrometry method is relatively easier to do because it does not require complicated sample preparation, simply by measuring the gamma radiation emitted from ^{238}U or the child is directly using the detector. To measure ^{238}U, a high -resolution gamma detector is needed such as High Purity Germanium (HPGe) and a double channel analyst (MCA). Because the efficiency of the HPGE detector is not too large, in this measurement a sample is needed with a large enough volume and generally uses a marinelli container with a size of 0.5 - 1 liter. The sample container is conditioned tightly closed, the ^{222}Rn gas does not escape out. If the sample is allowed to stand for 30 days, the radionuclide in the sample occurs secular equilibrium, where the number of each child will be the same as the number of parents, ^{238}U.

Determination of the number ^{238}U can be done by determining the number of daughters to decay in the sample. With a gamma spectrometer the number of daughters who emit gamma can be calculated. The calculation results are used to calculate the number of ^{238}U in the sample. In the spectrometry technique of Gamma Radioisotop ^{238}U is measured relatively, where a standard source containing isotopes ^{238}U is needed which is known to its activities. The standard source is used to determine the efficiency value of the HPGe in the gamma energy specified to measure ^{238}U. Standard sources that can be used for calibration efficiency must have conditions that are close to the environment to the environment to be measured. These conditions include volume, density, and gamma.

One of the standard sources used for efficiency calibration in ^{238}U measurements is IAEA RGU-1 which is Uranium ore in Silica Gamma. The problem is that the standard source of RGU-1 has silica gamma, while environmental sample material has different gamma such as water, soil, sand, clay, and others. Therefore, it should be proven that the standard source of RGU-1 can be used as a standard for uranium measurement in various environmental sample gamma. In this study conducted simulation using the Monte Carlo method with the MCNP computer program.

HPGe Detectors

The HPGE detector is the most widely used type of semiconductor detector in the gamma spectrometer. Although, the efficiency is not as big as other types of detectors such as NaI (Tl), but for the HPGE type detector resolution is still the highest. Because of this high resolution the HPGe detector is commonly used in the Gamma spectrometer for radionuclide analysis. PSTNT-BATAN Environmental Radiology Laboratory has a Gamma spectrometer with the Gr2519 HPGe detector which is a Canberra product rege type.

REGe (Reverse Electrode Coaxial Germanium detectors) Based on the model, Gr2519 is a germanium crystal detector with a density of 5,323 g.cm-3 in the form of coaxial, type-P-type, relative efficiency of 25% and gamma energy resolution of 1.9 KeV in 1,333 Mev. The cylindrical detector is a hollow with an outer diameter of 55.5 mm, a hole diameter of 10 mm and a height of 53 mm. The GR2519 HPGE detector design and the geometric model in MCNP are shown in Figure 1 with:

1. Crystal HPGE Model GR2519,
2. Window be 0.6 mm,
3. Outer Al 1 mm Casing,
4. Space in the detector,
5. Mylar 0.06 mm,
6. Dead layer ge 30 µm, and
7. Al Casing in 0.8 mm.

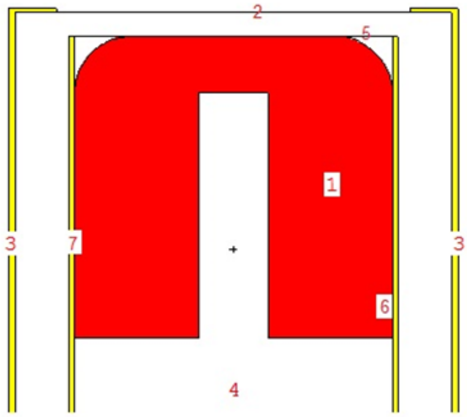


Figure 1. Display of the HPGE detector in McNP visited

Radiation source

The sample containing ^{238}U will produce a gamma ray with a lot of energy because it produces a child who also emits gamma rays, as shown in Figure 1. For that the entire gamma energy produced ^{238}U and the child is simulated so that the efficiency value can be obtained so that each efficiency value can be obtained energy. Gamma energy from ^{238}U and the decayed child used in the simulation is shown in Table 1. 46 keV gamma Energy is a characteristic emission of ^{214}Pb and 63 KeV characteristic emissions ^{214}Th, both of which are ^{238}U. Energy gamma 92 KEV is a combination of ^{214}Th and ^{235}U. The ^{214}Th isotope also contributed two gamma energy, 63.29 KeV and X-Ray Thorium, namely 93.8 KeV. The peak of 186 keV is a combination of gamma rays of ^{226}Rn namely 186 keV (3.6%) and ^{235}U 185 keV (54%). The peak 242 KeV and 352 KeV are ^{214}Pb Energy Gamma, while the peak of 609 KeV, 934 KeV, and 1120 KeV are gamma rays from ^{214}Bi. The sample was in the Marinelli volume container of 0.5 liters, with a diameter of 113 mm, a hole diameter of 79 mm and a height of 110 mm. Marinelli is made of polyethylene and polypropilin with a thickness of 2 mm. The sample did not fill the Marinelli room but only reached an altitude of 83 mm so that the sample volume was not up to 500 ml but 311 ml. The sample in the form of a standard source of IAEA RGU-1 which is a BL-5 Uranium ore mixed with silica gamma to have a density of 1,624 g.cm-3 [6, 7, 8]. The composition of the elements in RGU-1 includes 46.4% (Si), 0.04% (U), 0.1% (Al), 0.03% (Fe), 0.03% (Ca), 0, 02% (Na), 0.01% (C), 0.01% (mg), 0.008% (Pb), 0.002% (K), 0.008% (TI), 0.002% (s), <1 ppm (th) , and 53.3% (O) (7, 9).

Detector Efficiency

Simulation using MCNP is done to get efficiency in the detector. In its calculations, MCNP uses Tally F8: P to get the results of the photon pulse in the detector volume. MCNP will simulate the interaction of gamma radiation with materials starting from the sample gamma then the Marinelli container, Al Casing, and the HPGE detector. MCNP input is run using a computer with an Intel Core i5- 2.3 GHz processor, 4 GB RAM, and Windows 8.1 operating system. Latitude Data Photon interaction in the calculation of MCNP using the Endf/B-VI library (4).

RGU-1 Standard Sources

Validation of the simulation results is done by comparing the results of the RGU-1 standard source using the HPGq detector gamma detector spectrometer. The standard source of RGU-1 contains Uranium 400 ppm or contains ^{238}U activity 4940 ± 30 Bq/kg and ^{235}U with activity 231 Bq/Kg (7, 9, 10). In this experiment a sample will be chopped in the form of a standard source of RGU-1 with a mass of 410 g placed in a 500 ml Marinelli container. The HPGe detector used is the rege type of Gr2519 Canberra product. Analysis of the gamma spectrum is carried out using MCA and PCA II Nucleus software. RGU-1 standard sources are carried out for 8 hours to be able to identify the peaks of 46 Kev, 63 Kev, 92 KeV, 186 Kev, 242 Kev, 295 Kev, 352 Kev, 609 Kev, 934 Kev, and 1120 Kev in gamma spectrum clearly. Efficiency (ε) Each peak of the energy is calculated using the press. (1)

$$\varepsilon = \frac{cps}{A \cdot I} \qquad (1)$$

With CPS is the value of chopped per unit of enumeration time (1/s), A is the isotope activity (Bq), and I is an abundance of gamma emissions (%). The value of ^{214}Pb and ^{214}Bi isotopes is equal to ^{238}U, were 2025 Bq because in the standard source is considered to have occurred secular equilibrium so that the number of daughters of short age will be the same as the parent age. Gamma Energy 46 KEV (^{214}PB), 63 KeV and 92 KeV (^{214}Th) are quite good to be used for determining ^{238}U. It's just that the energy is very low, the measurement can only be done using the HPGE detector with window made from beryllium and carbon, while HPGE with aluminum window cannot be due to photon absorption problems. Energy 186 Kev (^{226}Ra) is quite good used to measure ^{238}U and can be done using HPGe with any window material. It's just that in the 186 Kev energy spectrum piled up between ^{226}Ra and ^{235}U so special calculations need. If the uranium in the sample is a natural uranium, the ^{238}U level is 99.28% and ^{235}U is 0.72% so that the activity ratio of ^{235}U/^{226}Ra is 0.0072 (11). With the abundance of 186 Kev Gamma from ^{226}RA is 3.6% and ^{235}U is 54%, the number of chopped at the peak of the 186 Kev energy is 41.2% owned by ^{235}U and 58.8% owned by ^{226}Ra. The use of ^{214}Pb and ^{214}Bi for measurement ^{238}U is very well done. Besides having gamma energy with a large amount, high energy and gamma abundance are also large.

METHODS

Monte Carlo N-Particle (MCNP) is a program package for the simulation of the traveling electron, photons and neutron travel simulations in three dimensions with a very broad energy range (4). Photon and electron interactions emitted from radioactive sources with detector materials are simulated to obtain output results in the form of detector efficiency. The use of MCNP for the simulation of the efficiency of the HPGE detector has been well tested (5). To simulate using MCNP, geometry inputs and detector materials, geometry and sample material and sample containers are needed, source defining, and tally to calculate efficiency. After obtaining MCNP input, it is run on a computer and an analysis of the output results is carried out. The efficiency value of the child's gamma energy is calculated using Tally Cultivation F8: P which is accompanied by the value of statistical errors.

Measurement of gamma from RGU-1 weighing 410 g in the Marinelli container using the HPGE detector is carried out in the chopped laboratory. Measurements were made for 40000 seconds. The chopped data are then processed using Microsoft Excell software. MCNP modeling is carried out for the RGU-1 measurement simulation weighing 410 g in the Marinelli container using the HPGE detect and the sample is simulated for 40000 seconds with the MCNP program. The results of RGU-1 chopped with a simulation are then processed using Microsoft Excell software. Furthermore, the two data are compared.

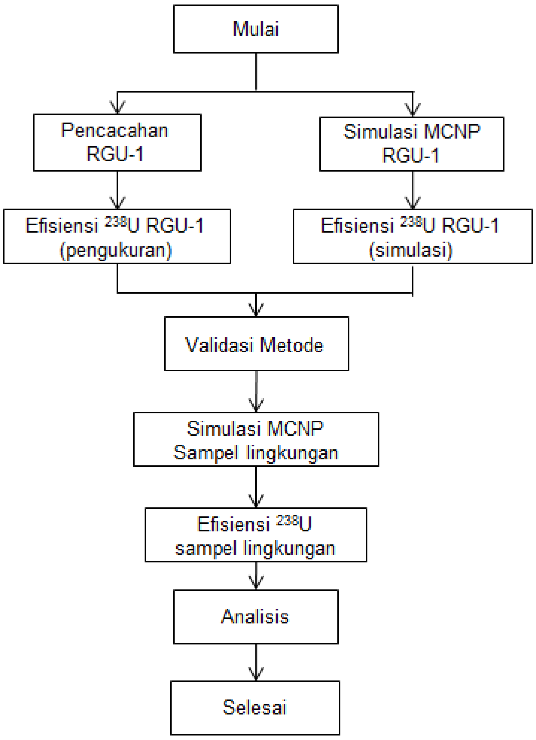
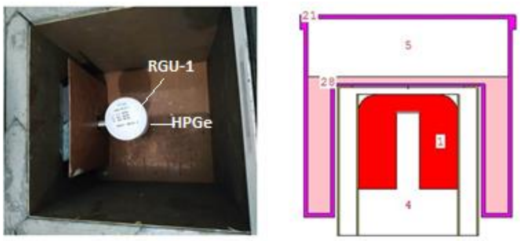


Figure 2. Research Flowchart

RESULTS AND DISCUSSION

MCNP input has been made for HPGE detectors and RGU-1 samples to simulate the efficiency of some gamma energy. Cacah pulses as an energy function are calculated in the HPGE detector position with 1%statistical error. The time required by the computer to run the input to get a 1% statistical error is 20 minutes. The display of the HPGE and RGU-1 geometry models to be simulated is shown in Figure 3 (B) with (28) RGU-1 samples, (21) Marinelli containers, and (5) air cavities.



a b

Figure 3. (a) RGU-1 enumeration with HPGE, and (b) MCNP simulation model.

Figure 3.a is shown the condition of RGU-1 measurement with the Gr2519 HPGe detector is equipped with a shield of lead material (Pb) and copper (Cu). The existence of PB shields has a major effect on reducing background radiation so that the display of the spectrum of gamma results of the sample enumeration will be much better because pure gamma radiation from the sample. The use of Cu material to reduce the occurrence of gaming gaming from the effects of beta radiation braking samples to the detector in this HPGE efficiency simulation is not modeled because there is no modeling of background radiation sources or beta radiation from the sample.

RGU-1 sample enumeration in Marinelli 500 ml for 8 hours obtained a spectrum of gamma as shown in Figure 4. In the spectrum of the gamma there are many peaks of the gamma energy. Some peaks are the gamma energy of ^{238}U, ^{235}U, and X-Ray from U and Th. The peak of the most gamma energy is the energy of the ^{238}U of the Daughter's Radiation which is primarily ^{214}Pb and ^{214}Bi. In determining the efficiency of the HPGe for RGU-1 is only determined at the ten peaks of the gamma energy with the position of the peaks it is shown in Figure 4. Because there has been a secular equilibrium in the RGU-1 material, it is assumed to be a concentration of ^{238}U and the decayed daughter are the same. Therefore, the efficiency value of each peak of the energy is only determined by the number of chopped and abundance of the gaming with the relationship as shown in the press. (1)

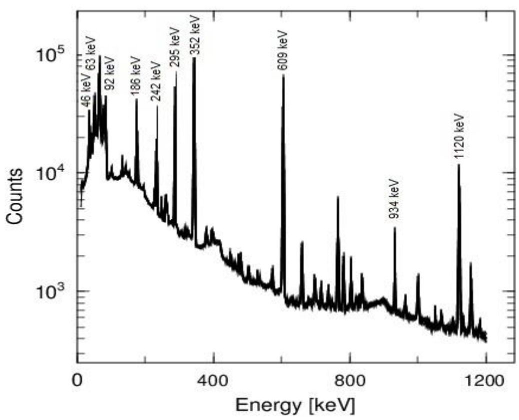


Figure 4. Gamma Spectrum of RGU-1 in Marinelli 500 ml.

Comparison of the efficiency value of each Uranium daughter's energy in the RGU-1 standard source from the simulation results with the measurement results shown in Figure 5. The difference is large enough to occur in low-energy-energy, namely 46 KeV, 63 Kev positions, and 92 KeV with a difference around 20%.

The efficiency value of simulation results in low energy is greater than the measurement results. Generally, this occurs due to material modeling where the effect of the density and composition of the material from the sample, sample container, and the detector component is large enough for the absorption of low energy photons.

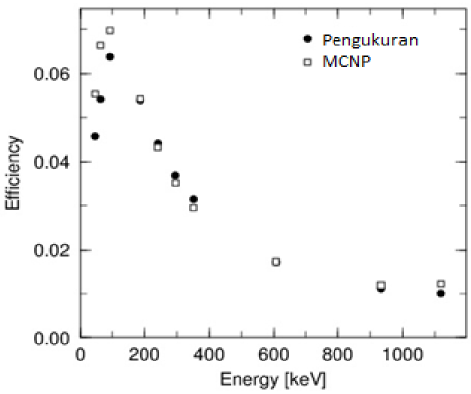


Figure 5. HPGE Efficiency for RGU-1 samples in Marinelli 500 ml

Therefore, the use of daughter with a low gamma energy is not recommended. Table 2 shows the results of the simulation efficiency for environmental samples with various gamma. Efficiency in low energy is 46 kev (^{214}Pb), 63 kev (^{214}Th), and 92 kev (^{214}Th) experienced significant changes with changes in the energy of the gamma from the sample, while the efficiency for energy greater than that is energy above 186 kev relatively fixed relatively, where the energy of the gamma is from ^{214}Pb and ^{214}Bi isotopes. This shows that the difference in sample density does not have a significant effect of self -absorption on high energy. So that the efficiency of the detector is relatively fixed.

Table 2 displays the results of the simulation of the energy efficiency of the Gamma Daughter's Energy ^{238}U as a function of environmental sample gamma. Gamma samples simulated are water, soil, sand, clay, and concrete and the standard source of RGU-1 is silica. The display in Table 2 is also sorted based on the sample density value of the smallest namely water (1.0 g.cm^{-3}) and the largest concrete (2,312 g.cm^{-3}). Data on the composition of water, soil, sand, clay, and concrete materials taken from McConn et al (12). The efficiency value of each gamma energy for a variety of environmental samples with variations in density and the gamma is close to the same. The value of efficiency with the smallest deviation of 0.002 is in the energy 609 kev and 934 kev in which both are gamma energy from ^{214}Bi isotopes. The efficiency value with other low deviations is 0.006, namely for 352 KeV energy and 0.007 for 295 KeV energy, both of which are ^{214}Pb isotopian gamma.063, in 46 KeV gamma energy (^{214}Pb). From these results show that ^{214}Pb and ^{214}Bi with relatively large gaming energy produce efficiency values in the HPGE detector are relatively the same for measuring environmental samples with various gamma.

Table 2. HPGE Efficiency of Various Matrices with Simulation Using MCNP

Energy (keV)	HPGe Efficiency					
	Water 1.0 g.cm^{-3}	Soil 1.52 g.cm^{-3}	RGU-1 1.62 g.cm^{-3}	Sand 1.7 g.cm^{-3}	Mud 2.2 g.cm^{-3}	Cemen 2.31 g.cm^{-3}
46	0.0672	0.0332	0.0458	0.0532	0.0442	0.0351
63	0.0744	0.0543	0.0542	0.066	0.0598	0.0535
92	0.0749	0.0667	0.0639	0.0693	0.0654	0.0626
186	0.0572	0.0538	0.0539	0.0543	0.0523	0.0515
242	0.0451	0.0442	0.0442	0.0431	0.0417	0.0411
295	0.0369	0.0363	0.0369	0.0354	0.0342	0.0338
352	0.0305	0.0298	0.0315	0.0293	0.0284	0.0281
609	0.0182	0.0178	0.0172	0.0176	0.0172	0.0171
934	0.0122	0.0122	0.0112	0.0119	0.0116	0.0115
1120	0.0123	0.0123	0.0101	0.012	0.0117	0.0118

CONCLUSION

Simulation with the MCNP program has shown that RGU-1 can be used as a standard source in ^{238}U measurements in environmental samples using ^{214}Pb and ^{214}Bi radionuclides as a reference. During the sample geometry with the same standard source, the gamma energy of the ^{214}Pb and ^{214}Bi nuclear energy has efficiency that is still close to the same even though the sample is different.

ACKNOWLEDGEMENTS

Author wants to thank to the entire PSTNT Radiology Laboratory Team for their assistance in the operation of the Gamma spectrometer and ARL laboratory for the use of RGU-1 standards. All researcher from BATAN were the main contributor of this paper.

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