Fissile Mass Estimation of the U/TRU/RE Ingot produced from Pyroprocessing by Neutron Multiplicity Measurement

Jihye Jeon [a], Chang Je Park [a], Geehyun Kim [a*]
[a]*Dept. of Nuclear Engineering, Sejong Uni., 209 Neungdong-ro, Gwangjin-gu, Seoul 05006, Korea*
[*]*Corresponding author: gkim01@sejong.ac.kr*

1. Introduction

The neutron multiplicity measurement has a strong advantage for measuring mass of special fissionable materials because it helps improve measurement for moist or impure nuclear material such as plutonium oxide, oxidized metal, scrap and residues [1]. This coincidence measurement can be useful when accounting for mass of the fissile material in a U/TRU/RE-blended ingot produced from pyroprocessing to fulfill safeguards obligations. Unlike the conventional He-3 coincidence counter for neutron multiplicity measurement, this study suggests stilbene scintillators as a prospective system for the very same purpose, due to their ability to detect fast neutrons as well as discriminate neutron and gamma-ray pulses. The feasibility of this concept is evaluated herein, roughly estimating the plutonium mass of the ingot from neutron multiplicity measurement with Monte Carlo simulation.

2. Methods and Results

The U/TRU/RE ingot and stilbene detectors are modelled, and neutron multiplicity distribution is measured with MCNP6.1 [2]. The passive method (without interrogation) is used for measuring neutrons from spontaneous fission, while active method (with interrogation) for induced fission neutrons.

2.1 Neutron Coincidence Measurement Model

Four cylindrical (2 in. diameter and 2 in. height) stilbene detectors with density of 1.15 g/cm^3 (Inrad Optics) are placed 5 cm away from the ingot, as shown in Fig. 1. For active method, a Am-Be point source is placed 10 cm below the target.

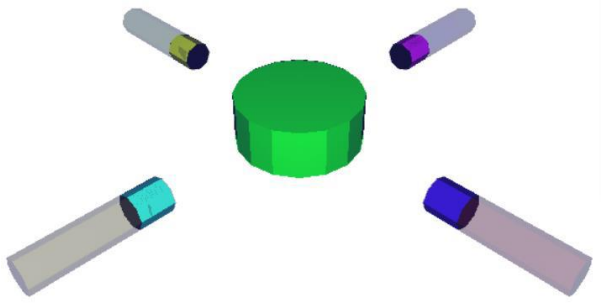

Fig. 1. Configuration of stilbene scintillators and U/TRU/RE ingot

The ingot, a conventional spent fuel with 4.5 wt% U-235 initial enrichment, burn-up 55,000 MWD/MTU, and 10 years of cooling time, is designed as a form of cylinder (20 cm diameter and 8 cm height) with density of 19.1 g/cm^3 [3].

For scoring, MCNP PTRAC card is used with f6 and f8 tally to count time-correlated neutrons which go through scattering reaction with hydrogen atoms in stilbene detectors. In addition, a time-dependent neutron sources for passive and active method are modelled with TME card of SDEF card.

2.2 Multiplicity Distribution

The Rossi-alpha distribution is used to determine the correlated and random events, which assumes that the number of correlated events from fission ('reals') attenuates exponentially while the distribution of random and uncorrelated events ('accidentals') is constant in time [1]. The coincidence resolving time or 'gate width' should be set to discriminate the reals and accidentals, as shown in Fig. 2. In this study, the gate width (G) for reals and accidentals (R+A) and accidentals (A) is set 10 ns (1 shakes) while predelay (P) 30 ns (3 shakes) and delay (D) 50 ns (5 shakes).

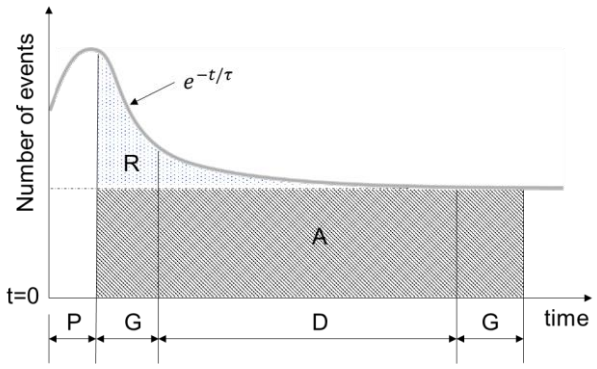

Fig. 2. A Rossi-alpha distribution. R = real coincidence events, A = accidental coincidence events, P = predelay, G = prompt and delayed gates, D= long delay and τ = die-away time. Adapted from Ref [5]

2.3 Multiplicity Calculations

The "point model", assuming the uniform neutron detector efficiency and the fission probability over the sample, is described in detail in chapter V of the Ref [1]. This model relates the statistical multiplicity calculation to physical properties of sample. The multiplicity data analysis has developed based on factorial moments of the

multiplicity distribution, using single, double and triple as the first, second, and third factorial moment of the distribution. Table I shows calculated values from multiplicity distribution measurement, using the point model.

Table I: Calculation of parameters

Parameters and description		Value
S	Single	6.853×10^{7}
D	Double	9.512×10^{7}
T	Triple	9.629×10^{7}
f_d	Doubles gate fraction	8.672×10^{-2}
f_t	Triples gate fraction	7.521×10^{-3}
v_{s1}, v_{s2}, v_{s3}	1st, 2nd and 3rd moments of spontaneous fission neutron distribution	2.836 10.025 39.828
v_{i1}, v_{i2}, v_{i3}	1st, 2nd and 3rd moments of induced fission neutron distribution	2.796 9.064 34.497

2.4 Mass Estimation

Using equations 5-48 through 5-51 in Ref [1] and calculated values in Table I, sample self-multiplication M can be determined. Neutron detection efficiency ε can be obtained by additional simulation with a californium reference source. Using these values, spontaneous fission rate F and effective mass of Pu-240 can be estimated with the equations 5-52 and 5-53 in Ref [1]. Those calculated values are shown in Table II.

Table II: Calculation of properties

Properties and description		Value
M	Self-multiplication	15.399
ε	Detection efficiency *(from Cf-252 measurement)*	1.025×10^{-2}
F	Spontaneous fission rate	5.637×10^{9}
m_{240}	Effective mass (kg)	1.192×10^{4}

The effective Pu-240 mass is that mass of Pu-240 giving the same neutron coincidence response as that obtained from all the even isotopes in the sample. It can be calculated from the spontaneous fission neutron yields of the isotopes, using the equation below which is modified from Ref [1]. The isotopes with high spontaneous fission yield more than 10^3 neutron/s·g, plutonium and curium, are only taken into account.

$$\textit{Effective mass of Pu-240} = 2.539 \times \textit{Pu-238} + \textit{Pu-240} + 1.686 \times \textit{Pu-242} + (2.059\times10^{4}) \times \textit{Cm-242} + (1.059\times10^{4}) \times \textit{Cm-244}$$

The actual effective mass of the Pu-240 ingot is 9.724×10^{3} kg.

2.4 Assessment

The self-multiplication factor (M) of the ingot metal is relatively high compared to the pure Pu-240 metal (M=1). The ingot has a substantial amount of fissile materials, especially Pu-239, and high M value implies a large amount of induced fission neutrons from those materials. In this respect, a correction factor called 'leakage multiplication M_L' is required, which considers neutron leakage, absorption, fission and reflection [5].

There is a difference between the calculated mass (1.192×10^{4} kg) and the original mass (9.724×10^{3} kg) in the scale of effective Pu-240 mass. This difference accounts for about 22% of the real mass. When the neutron response is multiplied, the sample appears to indicate more nuclear material than is actually present. Given that the metal ingot lacks the low-Z materials suitable for (alpha, n) reaction, it is assumed the difference is mainly caused by induced fission. If the corrected multiplication factor yields slightly higher value (~16.384) than M, similar mass to the original (~9.723×10^{3} kg) can be acquired.

From calculation of effective Pu-240 mass, Cm-242 and Cm-244 contribute to a large proportion of the estimated fissile mass due to its spontaneous fission yields three orders of magnitude higher than that of even-numbered plutonium isotopes. Conversion to the effective Pu-240 mass can give a rough approximation and cause a considerable degree of uncertainty to total Pu-240 mass.

3. Conclusions

In this research, fissile mass of pyroprocessed ingot is roughly estimated with MCNP simulation and statistical calculation with the point model. There is about 22% difference between the estimated mass from calculation and the original mass. Even with high self-multiplication, it is necessary to consider the correction factor in more sophisticated manner to account for induced neutrons from a large amount of fissile materials. The neutron multiplicity measurement, supplemented by other characterizing methods such as gamma-ray spectroscopy or densitometry, will allow more precise and detailed analysis of the material.

REFERENCES

[1] N. Ensslin et al, Application Guide to Neutron Multiplicity Counting, Los Alamos National Laboratory (LA-13422-M), 1998.
[2] T. Goorley et al., Initial MCNP6 Release Overview, Nuclear Technology, Vol. 180, pp. 298-318, 2012.
[3] C. J. Park, K. H. Kang, S. H. Na, and H. J. Ryu, Feasibility Study for Recycling Uranium from the Pyroprocessing in PHWR, Korea Atomic Energy Research Institute (KAERI/RR-3048/2009), 2009.
[4] J. Jeon, C. J. Park, G. Kim, Neutron Multiplicitiy Measurement of the U/TRU/RE Ingot produced from Pyroprocessing, Proceeding of 2018 IEEE Nuclear Science Symposium and Medical Imaging Conference, 2018.
[5] D. Reily, N. Ensslin, H. Smith Jr., S. Kreiner, Passive Nondestructive Assay of Nuclear Materials, Los Alamos National Laboratory (NUREG/CR-5550, LA-UR-90-732), 1991.