

Neutron Multiplicity and Gamma Ray Correlation in the U/TRU/RE Ingot processed from Pyroprocessing

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1. Introduction

The ingot produced from pyroprocessing is a metal mixture of uranium, transuranic and rare earth isotopes. In the perspective of international safeguards, it is important to accurately and precisely estimate the amount of uranium and plutonium in the ingot. Among various nondestructive assay methods, neutron multiplicity counting has merits in its applicability to heterogeneous samples and no flux attenuation within samples when compared to gamma-ray analysis. The technique has been mainly used to assay MOX power and plutonium scrap, not spent fuel [1]. This study is to explore the feasibility of using neutron multiplicity counting to estimate the fissile materials in the pyroprocessed ingot and using gamma-ray spectroscopy to overcome the limitation.

2. Methods and Results

2.1 Simulation Configuration

We modeled eight 20 cm diameter \times 8 cm height cylindrical U/TRU/RE ingots with density 19.1 g/cm³. The ingots were prepared with SCALE6.1 with different initial enrichment, burn-up and cooling time as shown in Table I.

sample ID	enrichment (wt%)	burn-up (MWD/MTU)	cooling time (year)
C1	4	55,000	10
C2	4	55,000	5
C3	3.5	55,000	10
C4	3.5	55,000	5
C5	4.5	55,000	10
C6	4.5	55,000	5
C7	4	60,000	10
C8	4	50,000	10

Table 1 Conditions for spent fuel ingot

Neutrons emitted from the ingot were calculated with eight 2 inch diameter \times 2 inch height cylindrical stilbene detectors with density 1.15 g/cm³. The detectors were placed 25 cm away from the round surface of the ingot as shown in Fig. 1.

MCNP6.2. [2] simulation was conducted with PTRAC to analyze coincidence counting by proton recoil at the eight stilbene detectors and FMULT card to appropriately model neutron multiplicity sampling.

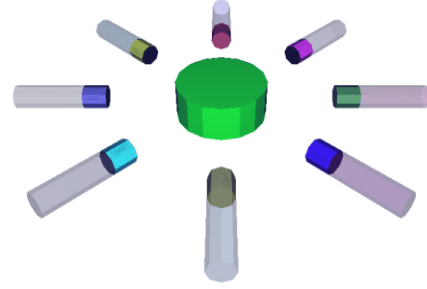


Fig. 1. Configuration of neutron detectors and U/TRU/RE ingot

2.2 Calculation Analysis

The point model [3, 4] has been commonly used to analyze the pulse train of neutron coincidence counting in passive method (without active interrogation). From the model, singles (S) and doubles (D) rate are as follows:

$$S = F\varepsilon M v_{s1}$$

$$D = \frac{F\varepsilon^2 f_d M^2}{2} \left[v_{s2} + \left(\frac{M-1}{v_{i1}-1} \right) v_{s1} \cdot v_{i2} \right]$$

- F: spontaneous fission rate
- ε : detection efficiency
- f_d : doubles gate fraction
- v_{s1}, v_{s2} : 1st and 2nd moments of spontaneous fission neutron distribution
- v_{i1}, v_{i2} : 1st and 2nd moments of induced fission neutron distribution

The ratio of doubles to the square of singles (D/S^2) can serve as a parameter to provide a reference curve to an unknown result since from the point model the ratio is in the linear relationship with multiplication factor (M).

$$D/S^2 = \beta[1+(M-1)\kappa(1+a)] = aM + b$$

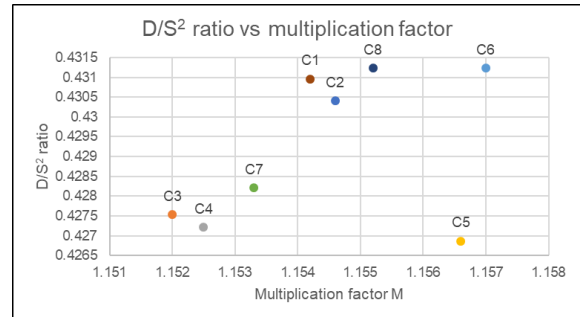


Fig. 2. D/S^2 ratio dependence on multiplication factor (M)

Sample C5 showed the highest deviation from the line. It was also difficult to relate the trend to three properties of ingots, initial enrichment, burn-up, and cooling time. Thus, it was necessary to examine where these neutrons came from.

2.3 Neutron Analysis and Limitation

From spontaneous fission neutron yield ($n/s \cdot g$) we can assume the number of neutrons from specific fissile isotopes. Among isotopes with high spontaneous fission rate, the ratio of neutron yields of Cm-244 to that of Pu-240 is greater than order of four [3]. Even samples contain more Pu-240 (around 14.0 wt%) than Cm-244 (around 0.4 wt%), but the high neutron yield of Cm-244 consists more than 95% of all the neutrons from the ingot.

Since only uranium and plutonium are the isotopes of interest in international safeguards perspective, neutrons from curium should be discriminated from those from uranium and plutonium. To separate them, the gamma-ray spectroscopy is inevitable. Thus, it is important to understand the correlation between neutrons and photons emitted from fission events and accurately simulate them.

Before MCNP6.2. is released, MCNP (until version 6.1.) uses the library to generate secondary particles independently from incident particles. Thus, it produces inelastic scattering gamma rays even in case of a neutron elastic scattering event. In addition, the library provides total gamma-ray multiplicity, not the multiplicity distribution, which impedes precise simulation [5, 6].

MCNP6.2. has a feature to correlate neutrons and gamma rays of fission events in FMULT card, enabling to use FREYA and CGMF fission model. FREYA samples neutron emission from Weisskopf spectrum. After neutrons are done emitting, gamma rays are emitted from residual energy. CGMF uses Hauser-Feshbach statistical theory for neutron and photon sampling [7].

2.4 Correlated Simulation

To estimate both neutrons and gamma rays, a simulation model was re-designed with addition of an NaI gamma-ray detector 25 cm away from top surface of the ingot, as shown in Fig. 3.

Even though stilbene is known for its ability to discriminate neutron and gamma-ray pulses, but it is difficult to conduct gamma-ray spectroscopy since it does not show photoelectric peaks. From the pyroprocess ingots, uranium and plutonium isotopes combined with other transuranic isotopes are expected to exhibit a myriad of photoelectric peaks. Estimating gamma-ray peaks with Compton edges in this case will be of a great challenge and therefore, an additional NaI gamma-ray detector was deployed.

From simulations, we could collect gamma-ray spectroscopy results with pulse height tally and neutron coincidence counting with PTRAC. Since gamma-ray yields correspond to the amount of the decaying isotopes,

possible radioactive isotopes were matched for each gamma-ray peaks and their relative quantity could be estimated. This relative spectral yield can be converted to absolute isotopic concentration when combined with neutron multiplicity results.

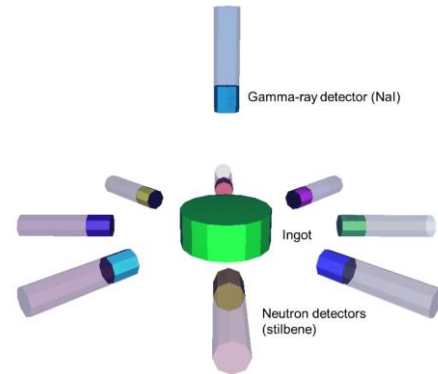


Fig. 3. Configuration of neutron and gamma-ray detectors and U/TRU/RE ingot

3. Conclusions

In this study, we explored the feasibility of using neutron multiplicity counting in calculating quantity of fissile material in pyroprocessed ingot. We found that neutrons from Cm-244 isotope made the analysis difficult and concluded that gamma-ray spectroscopy is inevitable. To do so, FREYA and CGMF fission models were suggested to correlate fission neutrons and gamma rays in monte carlo simulations. With neutron multiplicity results and gamma-ray spectroscopy results, this study can provide a method to assay spent fuel and other highly radioactive (fissionable) materials in non-destructive manner.

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