Neutron Source Term Evaluation for the Material Accounting of Advanced Spent Fuel Conditioning Process

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1. Introduction

Advanced Spent Fuel Conditioning Process (ACP) is a pyrochemical processing technique to convert oxide-type spent nuclear fuel into a metallic form. Korea Atomic Energy Research Institute (KAERI) has been developing this technology for the purpose of spent fuel management, and is planning to perform a lab-scale demonstration in 2006.

In this study, source term analyses on the ACP materials were performed to drive the inputs for the development of an effective non-destruction material accounting system based on coincidence neutron measurement. The goals are to produce the (α,n), and spontaneous fission neutron spectrum, and to investigate the fission multiplicity distributions, which may be variable across the samples in the process.

2. Methods and Results

The calculations were performed by ORIGEN-S of SCALE5 for the reference spent fuel and the material balance for the process of ACP. The reference spent fuel was the Korean YoungGwang 17×17 PWR spent fuel with 3.5 wt% enrichment, 10-year cooling time, and 43 GWD/MTU burn-up with 37.5 MW/MTU average power [1]. Lacking specific data for the ACP facility, the features such as material flow pattern, lithium reduction rate, and recovery yields of elements in the products were assumed based on the conceptual design [2]. Many assumptions necessary to estimate the physical properties of the materials were also made. Based on these source term data, Monte Carlo calculations were performed to investigate the variable property of neutron multiplicity distributions of samples.

2.1 Neutron Energy Spectrum

Neutron coincidence counting has been a valuable tool for assaying plutonium bearing materials that are pure or whose matrix constituents are well-known even in spent nuclear fuel. The multiplicity analysis equations are derived on the assumptions that all neutrons are detected with the same efficiency. However, the detector efficiency is energy dependent, so neutron counters are designed to have the detection efficiency as independent of energy as reasonably possible. This is very important to eliminate detection efficiency as one of the potential unknown parameters in multiplicity assay. In this regards, the neutron energy distribution of sample material is valuable information for the optimal design of neutron counter.

The neutron spectra of ACP materials derived by ORIGEN-S are shown in Figure 1. It was verified that there is no significant difference in total neutron spectrum between feed material (UO_2) and products (U-metal) because there is no process for isotopic changes of actinides in ACP. It also could be seen that the major contribution to the energy distributions of neutron in ACP materials are ranged between 0.1 and 5 MeV, and it means the NDA system for these samples should be designed to have same response in this energy range.

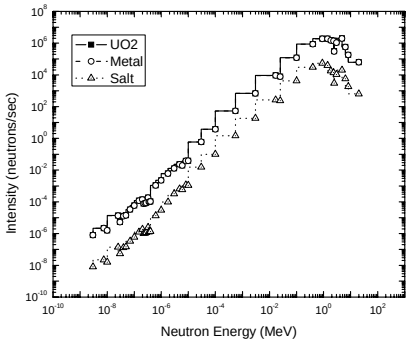

Figure 1. Neutron energy distribution of ACP materials (20kgHM basis).

2.2 (α,n) Neutron Energy Distribution

While the major target material for (α,n) reaction in feed material of ACP is oxygen, the salt waste has a lot of lithium and chlorine as target element for the reaction. The ratio of (α,n) to spontaneous fission neutrons (the quantity "α") is an important factor to be considered to design an optimum NDA system. Passive neutron counting usually shows significantly lower accuracy for such samples and assay precision is degraded to the point that long count times are required for acceptable results. Therefore, one of the specific goals for neutron counter design is to minimize the effects of variations in the sample's emitted neutron energy spectrum due to (α,n) reactions or sample moderators [3].

Based on the batch mode calculations with blending option of ORIGEN-S code, the quantity "α" of the

samples in ACP were derived as summarized in Table 1. The results were calculated from (α,n) spectra of process materials. According to these results, it was known that the process materials have similar (α,n) neutron spectra and "α" value. However, salt waste material shows different spectrum in energy range of 1~5 MeV, and the "α" value was calculated as about 0.7. It was also found that the major contribution to the (α,n) reaction of salt waste is conducted in Li-7 by Cm-244. Even though the "α" value of salt waste is 40 times higher than feed material, it could be considered as relatively low "α" value regarding other impure matrices of high "α" value [4]. For the ACP application, therefore, neutron coincidence counting using the known-alpha analysis method is most appropriate because of the negligibly low "α" value of the samples.

Table 1. Neutron characteristics of ACP materials.

Sample	(α,n) neutron		SF neutron		α
	Intensity (n/s)	Avg. Energy (MeV)	Intensity (n/s)	Avg. Energy (MeV)	
UO_2	2.04E+05	2.40	1.18E+07	2.10	0.017
U_3O_8	2.41E+05	2.40	1.18E+07	2.10	0.020
Hull	4.08E+02	2.40	2.36E+04	2.10	0.017
U Metal	6.39E-02	4.95	1.17E+07	2.10	0.000
Salt	8.24E+04	0.66	1.18E+05	2.10	0.697

2.3 Fission Multiplicity

The neutron emission multiplicity distribution P_ν, the probability that a fission will result in ν neutrons, is essential information in the development of methods of standardization of instrumentation [3]. It is also important to develop as accurate and complete a P_ν data base for ACP as possible in order to support the development or evaluation of empirical or theoretical calculations of neutron coincidence counting. Such calculations or correlations in turn can be used to predict neutron counting statistics or related values for samples of ACP for which experimental approaches are difficult.

The nuclear parameters P_ν, and $<\nu>$ that are of particular safeguards interest, have been evaluated for sample materials of ACP. The values presented in Table 2 were derived from the latest coincidence capability of the MCNPX code that provides the Singles, Doubles (and Triples), and the factorial moments of fission multiplicity. As shown in Table 2, multiplicity distributions from the spontaneous fission of ACP material show quite similar values to the canonical "consensus" set of safeguards community [5]. The multiplicity distributions from the thermal neutron induced fission were also derived for various process materials of ACP, and some of which may not be available to the safeguards community at large. It was found that the induced fission multiplicities of ACP materials are slightly larger than typical U-235 case, and it's rather similar to the prompt induced fission multiplicity of Pu-239 [6].

Table 2. Neutron multiplicity distributions of ACP materials.

Probability Distribution	Spontaneous Fission	Induced Fission			
	ACP	UO_2	U_3O_8	U-Metal	Salt
P(0)	0.015	0.037	0.037	0.037	0.045
P(1)	0.117	0.124	0.123	0.121	0.123
P(2)	0.300	0.259	0.259	0.258	0.266
P(3)	0.332	0.303	0.303	0.303	0.266
P(4)	0.183	0.195	0.194	0.195	0.195
P(5)	0.044	0.066	0.068	0.069	0.097
P(6)	0.009	0.014	0.015	0.014	0.006
P(7)	-	0.002	0.002	0.001	-
P(8)		0.000	0.000	0.000	-
$<\nu>$	2.717	2.759	2.765	2.769	2.760
$<\nu(\nu-1)>$	5.928	6.500	6.541	6.554	6.610

3. Conclusion

In this study, neutron source terms were analyzed to provide background data for the development of effective NDA system for ACP. The results of this study would be useful for the development of an optimized nuclear safeguards system that could meet IAEA safeguards criteria.

Acknowledgement

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