Sensitivity Analysis of the Criticality Evaluation Concerning Pyroprocess

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1. Introduction

In regard to the specific neutron characteristics of the nuclear fuelstreated by the pyroprocess, criticality evaluation plays a promising role not only in offering the key data on determination of the size of each of thoseapparatus but also in the fulfillment of the security requirements of the operating regulations. Criticality evaluation of the materials concerning the pyroprocess has been performed employing Monte Carlo techniques. Fresh UO_2 fuel and spent PWR fuel have been employed as the evaluation objects, respectively. It is proposed that there is no criticality risk in the voloxidation process, electro-reduce process and electro-refining in dealing with fresh UO_2 or PWR spent fuel without water intrusion. However, if water mixes with the fuels, the subcritical could be obtained. In the water intrusion cases, it is difficult to determine the exact contribution of each component nuclide to the multiplication factors because there are many nuclides involved and every nuclide influences each other; however, it is meaningful to compare the importance of each nuclide in affecting the criticality and to determine the relationship between water concentration and multiplication factors as well, which could be realized by sensitivity analysis employing TSUNAMI code of SCALE 6. In this paper, the sensitivity of the system criticality to the nuclide compositions of the evaluation object has therefore been carried out to determine quantitatively the importance of each element on the criticality.

2. Evaluation Method

2.1. Sensitivity analysis method

TSUNAMI is a SCALE control module that facilitates the application of sensitivity and uncertainty analysis theory to criticality safety analysis. TSUNAMI predicts the change in k_{eff} due to perturbations in cross-sections or densities by the following Eq. (1). These values of the TSUNAMI outcome are equivalent to the sensitivity of $k_{eff}$to the number density of each nuclide.

$$S_{k,\alpha} = \frac{dk/k}{d\alpha/\alpha} = \frac{\alpha}{k}\times\frac{k_{\alpha^+}-k_{\alpha^-}}{\alpha^+-\alpha^-} \qquad (1)$$

Where α^+ and α^- represent the increased and decreased values, respectively, of the input quantity α nd k_{α^+} and k_{α^-} represent the corresponding values of k_{eff}. The values obtained therefore represent the percentage change in k_{eff} that would be observed by making a 1% increase or decrease in the atom density of each nuclide. These values of the TSUNAMI outcome are equivalent to the sensitivity of k_{eff} to the number density of each nuclide.

2.2. Evaluation objects

PWR spent fuel with the initial enrichment of 4.5 w% ^{235}U and a burnup of 55GWd was adopted as the evaluation object, and the cooling time is 10 years. The irradiation and decay calculations of the PWR fuel were carried out by employing ORIGEN-ARP. The library used in this paper is 27GrpENDF4 of the SCALE 6 system for uranium enriched PWR fuel. The density of the fuel water mixture was calculated and listed in table1.

Table 1. Calculated density data used in the input of TSUNAMI

	H2O w %	Mixture mass density (g/cm3)
The homogeneous mixture of UO_2 & H_2O	35	2.445
	40	2.201
	45	2.001
	50	1.834
The homogeneous mixture of SF & H_2O	20	3.656
	25	3.136
	30	2.745
	35	2.441

2.3. Geometry of evaluation objects

The practical geometries of pyroprocessing facilities are complicated. However, for the criticality evaluations, conservative geometries are usually preferred. Cylindrical geometry is widely applied in the pyroprocessing facilities such as the reducer cathode, refiner anode, and winner cathode etc. In this paper the evaluation geometry therefore employs a cylinderwith diameter equal to height. Cylinder geometry also applies to the reflectors.

3. Results and discussion

3.1. Sensitivity of nuclides composition to the criticality

As shown in Figure 1, ^{235}U, ^{1}H and ^{16}O show positive values; however, ^{238}U shows negative values. ^{235}U is fissile so exhibits positive values. H^1 acts as moderation so the values are positive. The increase of ^{16}O density contributes to the increase of k_{eff}. However, ^{238}U exhibits negative effects due to its high amount present in the fuel and the neutron capture. The sensitivity coefficient of ^{235}U increases with the increase of water content in the mixture and surpass the sensitivity coefficient of ^{1}H due to the improved performance in capture thermal neutrons. However, the sensitivity coefficient of ^{1}H decreases with the decrease of fissile concentration, which indicates that at the low hydrogen concentration scale hydrogen plays more important role in k_{eff}. due to its moderation effect with high fissile concentration. As shown in Figure 9, actinides ^{239}Pu, ^{235}U, ^{241}Pu, and ^{243}Cm exhibit positive values. The moderator ^{1}H exhibit positive value due to moderation effect before the mass fraction of spent fuel is bigger than 70% and the dilution effect of ^{1}H surpass its moderation effect after the mass fraction of spent fuel is less than 70%. The other neutron poisons shows negative effect due to the high thermal cross section for neutron capture.

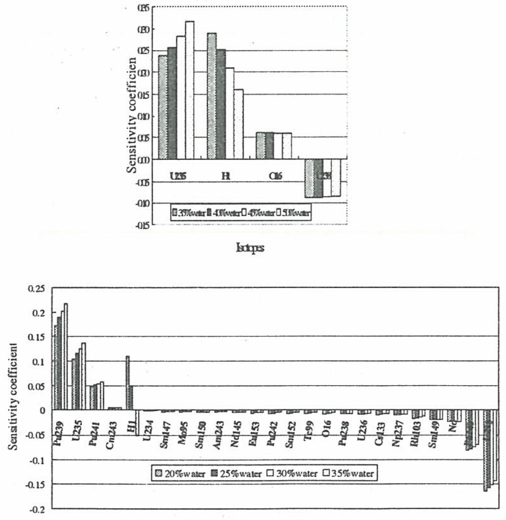

Fig. 1. Dependence of sensitivity coefficient of UO_2&H_2O and SF&H_2O on the H_2O weight percent.

3.2. Sensitivity analyses of water concentration to criticality

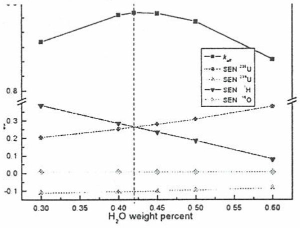

Fig. 2. Dependence of k_{eff} & sensitivity of U&H2O on the H_2O weight percent.

There is a crosspoint between the sensitivity plots of ^{235}U and ^{1}H near 42w% of water as shown in Fig.2. k_{eff} increases before the concentration of water reaches approximately 42w%, due to the contribution of ^{1}H is more than that of ^{235}U indicated by higher sensitivity values. Near the 42w% of water point, the importance of ^{235}U in determining the $k_{eff}$is seemly equal to that of ^{1}H indicated by the crosspoint of sensitivity of ^{235}U and that of ^{1}H. After the crosspoint k_{eff} therefore begins to decrease, because at this region main contribution comes from the ^{235}U, however, the concentration of ^{235}U decreases leading to the decline of the k_{eff} plot.

4. Summary

Sensitivity analysis by TSUNAMI clarifies the complex effects of key nuclides on the criticality probability quantitatively. As discussed above, the k_{eff} of UO_2 fuel reaches the maximum value with 42w% concentration of intrusion water. The concentration of hydrogen affects the complexity of reaching criticality by its competition between the concentrations of ^{235}U. Approximately if the weight percent of H_2O in the mixture is less than 42%, the moderation effect of hydrogen surpasses its dilution effect on ^{235}U. However, the importance of ^{235}U increases dramatically when the weight percent of water is bigger than 42%. In the sensitivity evaluation of UO_2 fuel employing TSUMAMI, there is a similar crosspoint of the sensitivity of ^{235}U and the sensitivity of ^{1}H where the criticality reaches summit. And the optimal water weight percent is determined to be 50%.

5. Reference

1) B. T. Rearden, *TSUNAMI-3D: Control Module for Three-dimensional Cross-section Sensitivity and Uncertainty Analysis for Criticality.* ORNL/TM-2005/39 Version 6 Vol. I, Sect. C9. (2009).