Request from Nuclear Fuel Cycle and Criticality Safety Design

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The quality and reliability of criticality safety design of nuclear fuel cycle systems such as fuel fabrication facilities, fuel reprocessing facilities, storage systems of various forms of nuclear materials or transportation casks have been largely dependent on the quality of criticality safety analyses using qualified criticality calculation code systems and reliable nuclear data sets. In this report, we summarize the characteristics of the nuclear fuel cycle systems and the perspective of the requirements for the nuclear data, with brief comments on the recent issue about spent fuel disposal.

1. Introduction

Figure 1 shows the schematic flow diagram of LWR fuel cycle. We find variety of fissile materials there. Table 1 summarizes their variety. They range from gaseous form to solid and their mixture and include many indeterminate forms, resulting in demanding requirements for the modeling capability of criticality calculation codes.

Fissile materials are sometimes dry



Fig.1 Schematic Flow of the LWR Nuclear Fuel Cycle

and sometimes wet. In some processes they are in the form of aqueous solution. It means that there are variety of neutron moderation conditions according to the hydrogen content. So, variety of neutron spectra, from the fast reactor-like hardest one to the fully thermalized one must be covered. Also, they are composed of variety of resonance nuclides and their mixture, which requires appropriate treatment of resonance self-shielding and reliable resonance data.

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Physical / Chemical form		Material
Gaseous	Hexa-Fluoride	UF ₆
Fluidal	Fluoride Solution	UO_2F_2aq
	Nitric Solution	$UO_2(NO_3)_2aq$, $Pu(NO_3)_4aq$
	Homo. Powder	UO ₂ / UO ₃ -H ₂ O, MOX-H ₂ O
	Slurry	ADU
Solid & Mixture	Pellets/Rods/Assemblies	UO ₂ -H ₂ O, MOX-H ₂ O
	Dissolving Mixture	Spent UO ₂ – Nitric Solution

Table 1 Variety of Fissile Materials Found in the Nuclear Fuel Cycle

2. Criticality safety design practices in Japanese industries

According to the aforementioned requirements, Japanese industries have been using the standardized criticality calculation code systems with multi-group constants library and Monte Carlo codes to apply for the licensing of many nuclear fuel cycle systems.

The JACS system^[1] whose main tools are KENO-IV^[2] Monte Carlo code and MGCL (Multi-Group Constants Library)^[3] based on ENDF/B-IV was developed by JAERI. The MGCL has 137group multi-purpose version and 26 group condensed version. JACS has been used for many licensing application.

The well-known SCALE system has been also used commonly, particularly for the design of fuel casks. The recent version of the system^[4] has KENO-V.a^[5] Monte Carlo code and its CSRL (Criticality Safety Reference Library) based on ENDF/B-V. The CSRL has 238 group version and condensed 44 group version. Formerly, ENDF/B-IV based 218 group and 27 group libraries were widely used.

For the design of fuel storage at power plant site, reactor core design codes (ex. PHOENIX-P/HIDRA^[6]) are used. Our PHOENIX-P code is attached with ENDF-B/V based 42-group library and is used for fuel storage rack systems at PWR site for the consistency with reactor core design.

Nowadays, continuous energy Monte Carlo codes such as MCNP^[7] or MVP^[8] are widely used for the studies. The latest nuclear data ENDF-B/VI or JENDL-3.3 are both used.

3. Criticality safety design criteria

Japanese Criticality Safety Handbook ^[9] lays out some subcriticality criteria and associated conditions. The most commonly applied criterion for nuclear fuel systems is k off < 0.05

 $k\text{-}eff \leq 0.95,$

where *k-eff* shall be calculated with "well qualified system". The 0.95 criterion is a rather heaven-sent one that has been used since very old days. The Handbook suggests that one can rationalize it, if he/she can show the validity by quantitative data. In this validation, the quality of nuclear data is of large importance. The integral validation by some series of criticality experiments simulating the systems of interest is directly useful for this purpose. The Handbook introduced the concept of the "estimated critical lower limit *k-eff*", which includes the minimum bias to be considered for computational uncertainty and is specific to the combination of nuclear data, criticality code and fissile systems. Theoretically, we could

apply as high *k-eff* criterion as the estimated critical lower limit value, provided that sufficient backup by integral validation through criticality benchmarks is available.

4. Logistics in nuclear fuel cycle – fuel casks

The fuel casks are playing key roles in the logistics in the nuclear fuel cycle. They are versatile components that are used for many purposes, such as transportation, interim or long-term storage of fuels.

Since transportation or storage costs are not small among total fuel cycle cost, the economy of casks affects the total economy of fuel cycle. Furthermore, the market of fuel casks is rather open to the world. So, there has been the continuous pressure for the streamlining and sophistication of the cask design.

The economy of casks can be measured by the;

- payloads number of fuel assemblies to be loaded,
- cost for specific materials, such as neutron absorber, gamma/neutron shielding,
- material and assembly cost to assure mechanical strength, heat resistance and radiation resistance, and
- light weight for easy handling.

The current practice of criticality safety design of spent fuel casks is often with built in neutron absorber such as boronated stainless steel, boronated aluminum etc. Usually in the criticality safety design analyses, unirradiated fuel with initial ²³⁵U enrichment is still assumed and the 0.95 criterion is still applied.

5. Challenges for Burnup Credit

The front of nuclear criticality safety designs has been seeking for the application of "Burnup Credit". In Japan, "A guide introducing burnup credit, preliminary version" was issued in 2001^[10].In this field, it is regarded to be prudent to take the credit of the limited nuclides seen in the spent fuel. In the level-1 burnup credit, the actinides in the spent fuel are solely considered, while more ambitious level-2 burnup credit assumes the absorption by fission products (FPs)

In the criticality safety design taking credit of burnup, two categories of uncertainty are to be considered. One is the uncertainty of the assumed isotopic composition of spent fuels. Since the isotopic composition of spent fuel is obtained through depletion analysis of the power reactor, its uncertainty originates from nuclear data used in the depletion analysis, depletion code, depletion environment, as well as cooling time after reactor shutdown. And the other is the uncertainty of criticality prediction of the systems containing spent fuels. This uncertainty originates from the nuclear data used in the criticality analysis, criticality code as well as the aforementioned uncertainty of isotopic composition itself.

For the validation against the isotopic composition, the destructive analysis data of spent fuels obtained in the post-irradiation examination (PIE) are useful. As the public PIE database, the SFCOMPO^[11] is available.

Although the criticality experiments using actual spent fuel are considered useful for the validation against the criticality prediction of spent fuel systems, very few numbers of such

experiments have been run because of the difficulty to handle the actual spent fuels in the experimental facility. The REBUS international project^[12] is one of the few examples. This approach has intrinsic difficulty in handling the highly radioactive spent fuels and in covering the whole spectrum of depletion environment.

More strong-arm approach is the validation through the direct simulation of the power reactor with criticality codes, which is coming to be realistic these days.

6. Level-1: Actinide burnup credit

The level 1 burnup credit has been already applied to many examples. In Japan, Rokkasho Reprocessing Plant has the spent fuel storage pool designed for the fuel whose residual ²³⁵U enrichment is lower than certain limit, and its head end process including the dissolver will be operated with or without gadolinium poison, according to the burnup of the fuel to be processed. The transport casks in France, Germany, Netherlands, Switzerland and USA are also the examples ^[13].

7. Level-2: FP credit

The level 2 burnup credit has been also applied to some examples. The spent fuel pit of the US nuclear power plant is an example.

The FP nuclides to be considered are preferred to be long-lived chemically stable and of course have large neutron absorption capability. Table 2 shows the examples of the selected FP nuclides. Also we have to be aware of the cooling time of interest, because the relative importance of each nuclide is not fixed with time^[10].

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Case	Nuclides	
6 FPs ^[14]	Sm-149, Rh-103, Gd-155, Nd-143, Cs-133, Sm-152	
(CEA at early stage)		
12 FPs ^[10]	Sm-149, Rh-103, Gd-155, Nd-143, Cs-133, Sm-152, Tc-99, Eu-153, Nd-145,	
(JAERI-Tech 001-055)	Sm-147, Mo-95, Sm-150	
15 FPs ^[14]	Sm-149, Rh-103, Gd-155, Nd-143, Cs-133, Sm-152, Tc-99, Eu-153, Nd-145,	
(OECD BUC W.G.)	Sm-147, Mo-95, Sm-150, Sm-151, Ag-109, Ru-101	
13 FPs for Casks ^[15]	Tc-99, Rh-103, Xe-131, Cs-133, Nd-143, Nd-145, Pm-147, Sm-147, Sm-149,	
(SAND87-0151)	Sm-151, Sm-152, Eu-153, Gd-155	

Table 2 The examples of selected FP nuclides for the level 2 burnup credit

8. Burnup credit case study – Spent fuel cask

To demonstrate the nature of the FP credit, we applied the levels of burnup credit to the spent fuel cask model designed for the intact fuel with 235 U enrichment of 4.1wt%.

Figure 2 shows the considered cask model. It has boronated aluminum spacers and flux traps. The boron in the spacer is assumed to be enriched in ${}^{10}B$.

The required width of flux trap and ¹⁰B content for the 4.8wt% ²³⁵U fuel with various burnup were surveyed. The considered levels of burnup credit are level 1, level 2A where the SAND87-0151 (13) FP nuclides are considered and the level 2B where the virtually all FP

nuclides are considered. Also the uncertainties of FP and actinide amounts were treated as the parameters.

Figure 3 shows the surveyed results of the flux trap width under various conditions. FP

credit is quite attractive even with only 13 nuclides. On the other hand, in the cases with the assumed uncertainties of $\pm 5\%$ for actinides and/or -20% for FPs, the merits of burnup credit are significantly reduced.

Figure 4 shows the results for the ¹⁰B content. The uncertainties reduce the merit of burnup credit here, too.



Fig.2 Spent Fuel Cask Model for the Study



Fig.3 Required flux trap width versus burnup



9. Recent issue: spent fuel disposal

In the course of the periodical revision of the "Long-Term Program for Research, Development and Utilization of Nuclear Energy of Japan", the economic study on the direct disposal of spent fuels was performed ^[16]. In this study, major technical and non-technical challenges were studied and assessed. In the conclusion, criticality issue was identified as one of the major uncertainties of the study, since no safety evaluation criteria to prevent criticality by plutonium etc had been established yet.

Besides the safety criteria, modeling, scenario or phenomenology to be considered have not been well established. From the viewpoint of nuclear data, very long term ($\sim 10^3$ y) transient of isotopes would increase the uncertainty. Also, integral validation would be more difficult than for the postulated burnup credit design of fuel cycle systems.

10. Conclusions

- (1) Nuclear fuel cycle consists of wide spectrum of fissile systems. Variety of resonance nuclides and neutron spectra are to be covered.
- (2) Better-qualified codes and nuclear data could improve criticality safety design criteria, and give more competitive edge to nuclear fuel cycle.
- (3) Burnup credit is the major front of criticality safety design of spent fuel systems.
- (4) Level-2 burnup or FP credit is promising, whose efficiency depends on uncertainty of

spent fuel characteristics and their prediction.

- (5) Integral tests for spent fuel systems are difficult by nature, microscopic validation would be of more importance.
- (6) In the spent fuel disposal study, criticality issue was identified as one of the major uncertainties. If its reduction were necessitated, improvement of FP and actinides data would play a certain role.

References

- J.Katakura, Y.Naito and Y.Komuro,"Development of Computer Code System JACS for Criticality Safety", Trans. Am. Nucl. Soc., 41, 329 (1982)
- [2] L.M. Petrie and N.F. Cross, "KENO-IV An Improved Monte Carlo Criticality Program", ORNL-4938 (1975).
- [3] Y.Naito et al., "MGCL-Processor: A Computer Code System for Processing Multi Group Constants Library MGCL", JAERI-M9396(1981)
- [4] ORNL SCALE Home Page, http://www.ornl.gov/sci/scale/
- [5] L.M. Petrie and N.F. Landers, "KENO-V.a An Improved Monte Carlo Criticality Program with Supergrouping", VOL.2, SEC. F11 ORNL/NUREG/CSD-2/V1/R2 (1984)
- [6] "On the Criticality Safety Analysis for the Spent Fuel Storage at LWR Power Plant Site", MHI-NES-1003 rev.3, Mitsubishi Heavy Industries, Ltd. (1998) (in Japanese)
- [7] LANL MCNP Home Page, http://laws.lanl.gov/x5/MCNP/index.html
- [8] T. Mori, M. Nakagawa, JAERI-Data/Code 94-007 (1994) (in Japanese)
- [9] Working group on Nuclear Criticality Safety Data, "Criticality Safety Handbook Version 2", JAERI-1340 (1999) (in Japanese)
- [10] Nuclear Fuel Cycle Safety Research Committee, " A Guide Introducing Burnup Credit, Preliminary Version (Contract Research)", JAERI-Tech 2001-055 (2001) (in Japanese)
- [11] M. Kurosawa, Y. Naito and T. Kaneko, "Isotopic Composition of Spent Fuels for Criticality Safety Evaluation and Isotopic Composition Data Base (SFCOMPO)", Proc. Fifth International Conference on Nuclear Criticality Safety (ICNC'95), Vol.1, P.2.11, Albuquerque, NM, USA (1995)
- [12] P. D'hondt et al., " The REBUS Experimental Programme for Burn-Up Credit", ICAPP'03, Cordoba, Spain, http://www3.inspi.ufl.edu/icapp03/program/abstracts/3036.pdf, (2003)
- [13] International Atomic Energy Agency, "Practices and Developments in Spent Fuel Burnup Credit Applications", Proc. Technical Committee Meeting, Madrid, Spain, IAEA-TECDOC-1378 (2002)
- [14] J. Raby et al., "Current Studies Related to the Use of Burnup Credit in France", Proc. ICNC2003, p.627, JAERI-Conf 2003-019(Part II)
- [15] T. L. Sanders, R. M. Westfall and R. H. Jones, "Feasibility and Incentives for the Consideration of Spent Fuel Operating Histories in the Criticality Analysis of Spent Fuel Shipping Casks", SAND87-0151, UC71, TTC-0713 (1987)
- [16] Atomic Energy Commission of Japan HP, http://aec.jst.go.jp/jicst/NC/tyoki/indexh2.htm, materials for 2004.10.7 technical subcommittee meeting (in Japanese)