# Plutonium Content, Uranium Content, and Subcriticality Data of Spent Fuels *

*I. Kobayashi, T. Suzaki, H. Okuno, and Y. Naito*

Department of Fuel Safety Research, Nuclear Safety Research Center
Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken 319-11, JAPAN

## 1. Introduction

It is well known that the design concepts of transportation systems, storage facilities, reprocessing processes, etc., of spent nuclear fuels can be economized due to the significant decrease in the reactivity of these fuels as a result of burnup. The fissile isotopic content of spent fuels should be known to estimate the subcriticality of these fuel assemblies in the corresponding systems and processes. However, the isotopic content of plutonium and uranium and the other fission products (FPs) is not systematically known, and this isotopic content depends on the type of reactor (BWR, PWR), burnup history, and the position of fuel rods in the fuel assembly. Furthermore, the accuracy of the calculated results not have been verified in comparison with destructive data, especially since systematic data have not been measured for BWR's and PWR's. At the Japan Atomic Energy Research Institute (JAERI), studies were performed to review burnup data. These studies focused on the isotopic content of plutonium and uranium in spent fuels. The information gained will be used in corresponding subcriticality calculations. In addition, isotopic ratio of $^{235}U$ to total uranium and $^{239}Pu$, $^{240}Pu$, $^{241}Pu$, $^{242}Pu$ to total plutonium respectively, which depend on the initial $^{235}U$ enrichment and burnup ratio, was determined for use in the criticality safety evaluation of PWR spent fuels.

However, the criticality and subcriticality data of spent fuels are not sufficient to verify criticality calculation results. These data must be systematically measured as a function of the burnup ratio, burnup history, etc.

Therefore, an experimental program to measure subcriticality data of spent fuel assemblies in a storage pool has begun at JAERI. The objectives of this program are to apply an exponential experimental technique to obtain

---

* Part of this work was performed under the auspices of the Science and Technology Agency by the Japan Atomic Energy Research Institute.

benchmark data for verification of criticality calculation codes, to demonstrate this technique as a viable method for the pre-loading burnup measurement, and to present systematic data for fissile isotopic content, burnup history, etc., that are needed for the verification of burnup calculation codes.

## 2. Plutonium and Uranium Content in LWR spent fuels

The reactivity of the PWR and BWR fuels decreases as a function of burnup. This is due to changes in actinide compositions and the creation of FPs. The actinides are mainly induced by fissions of $^{235}U$, $^{239}Pu$, $^{241}Pu$, and the successive (n,$\gamma$) reactions and $\beta$-decays. This chapter addresses the changes in isotopic compositions of U and Pu and the methodology used to calculate their quantities together with the lower-limit of the critical mass of arrayed $UO_2$ rods in water as a function of burnup.

### 2. 1 Computer Code COMRAD

A computer code COMRAD (Naito et al. 1986) was developed to estimate the isotopic compositions of LWR spent fuels because isotopic compositions are considered important in the design and safety of nuclear fuel cycle facilities. The COMRAD code identifies the decay chains and calculates the neutron flux, burnup quantities, isotopic compositions, neutron emission rates by ($\alpha$, n) reactions of light nuclides and spontaneous fission, as well as the radiation source intensities. The code requires four data libraries: a decay data library, a burnup dependent one-group neutron cross-section library, a ($\alpha$,n) reaction library, and a spontaneous fission library.

Of the four data libraries, the burnup dependent one-group neutron cross-section library, ONEGL, is the most important in estimating the isotopic composition of uranium and plutonium in spent fuels. As a first step in using ONEGL, a burnup calculation is performed with the cell burnup code UNITBURN (Naito et al. 1990) to obtain the burnup dependent neutron energy spectrum, $\phi_g$ (B), and the burnup dependent one-group neutron cross sections, $\sigma^i$ (B), for the nuclides in the core. The isotopic compositions $N^i$ (B) of uranium and plutonium in the spent fuels of the PWR are determined in the cell burnup calculation using UNITBURN. These compositions are then compared with measured data. As shown in Fig.1 , the calculated results agree well with the measured data. This confirms the neutron energy spectrum used to obtain the one-group neutron cross-section constants is accurate.

The burnup dependent one-group infinite-dilution neutron cross sections, $\sigma^j_{\infty,g}$, of the nuclides, except uranium and plutonium, are collapsed into one group using the burnup dependent neutron energy flux, $\phi_g$ (B), as the weighting function. In this way, the one-group constants for the standard PWR are prepared and stored in the ONEGL library. The code COMRAD then calculates one-point burnup of the fuel with the data from the ONEGL.

## 2. 2 Estimation of Isotopic Compositions

Isotopic compositions of spent fuels from domestic PWRs were measured at JAERI. The plutonium to uranium ratios of these fuels and spent OBRIGHEIM reactor fuel (Barbero et al. 1989) are shown in Fig.2 as a function of burnup. The curves shown in Fig.2 were fitted using COMRAD calculated data. The curve corresponding to an initial enrichment of 3 wt% shows excellent agreement with the data of the OBRIGHEIM reactor, marked Z in Fig.2, whose initial enrichment was 3.1 wt%. In the other cases, however, i.e. the spent fuels of the domestic PWRs, the agreement is not as good. Figure 3 shows the relationship between the $^{235}U$ to U ratio and the Pu to U ratio for the same fuel as in Fig.2. The $^{235}U$ to U ratio is proportional to the initial enrichment since it decreases almost linearly with the increasing Pu to U ratio. Similarly, each ratio of $^{239}Pu$, $^{240}Pu$, $^{241}Pu$, and $^{242}Pu$ to Pu is expressed as a second or higher order polynomial of the Pu to U ratio. All measured data are within the range of three standard deviations from the fitting curve. The empirical equations of the curves from Figs. 1 through 3 are:

$$\frac{Pu}{U} \times 100 = \frac{a(E) \cdot B}{b(E) + B} \equiv x \qquad (1)$$

$$^{235}U/_{U} = E\left(1.0 - \frac{x}{1.25}\right), \qquad (2)$$

$$^{239}Pu/_{Pu} = 1.0 - 0.49x, \qquad (3)$$

In Eq. (1), B is the fuel burnup [GWd/MtU] ; the coefficients a and b are dependent on the initial enrichment, E, and their values are given in Table 1.

## 2. 3 Conservative Isotopic Compositions

From these studies above written, a conservative isotopic composition for PWR fuel can be recommended as shown in Table 2. The composition is determined by selecting the maximum value for the fissile nuclides, $^{235}U$, $^{239}Pu$ and $^{241}Pu$, and the minimum value for the fertile nuclides, $^{240}Pu$ and $^{242}Pu$. This recommended composition is referred to the nuclear criticality safety handbook of Japan (STA 1988) .

## 2. 4 Lower-Limit of Critical Mass

Considering the changes in the Pu/U ratio according to Eq. (1) and the isotopic compositions given in Table 2, the lower-limit of the critical mass for $UO_2$ fuel rod lattices immersed in water are calculated using the nuclear criticality safety evaluation code system JACS (Katakura et al. 1982). The results, shown in Fig.4 as a function of fuel rod lattice pitch, show that the lower-limit of the critical mass increases with fuel burnup.

### 3. Subcriticality data of spent fuels

To validate calculational codes of fuel depletion and criticality analyses, criticality or subcriticality data of spent fuels is needed with known three-dimensional isotopic contents for a reactor. Variables for this are burnup history, cooling time and reactor type (BWR, PWR). For this reason, JAERI is conducting an experimental program to develop a subcriticality measuring technique to provide subcriticality benchmark data of spent fuels. Data will include the destructively measured isotopic content of the fissile nuclides, burnup history, etc.

In late 1992, the subcriticalities of two PWR spent fuel assemblies of 14×14 and 17×17 types are to be measured by the exponential experiment method. These assemblies have been irradiated to about 35GWd/t, on the average. Their initial $^{235}U$ enrichment was 3.4%. Figure 5 shows an example of the exponential distribution of thermal neutron flux in the axial (z) direction of the 14×14 type assembly calculated by the multigroup neutron diffusion code CITATION (Fowler et al. 1969) with extraneous neutron sources. Curve A is for the case of only inherent sources of spontaneous fissions and ($\alpha$, n) reactions from several actinide nuclides. When a $^{252}Cf$ neutron source of about 20 mCi is positioned at some height, the axial distribution is given by curve B. By subtracting curve A from curve B, the exponential distribution $\exp(-\gamma z)$, curve C, is obtained. Based on the theory of the exponential experiment, the fundamental mode spatial decay constant, $\gamma$, increases when the neutron absorption rate in the assembly and the radial leakage rate of neutrons from the assembly become large compared to the fission rate. This means that $\gamma$ is directly related to the effective neutron multiplication factor $k_{eff}$ for the infinitely long assembly as:

$$1 - 1/k_{eff} = -K\gamma^2 \qquad (4)$$

where K is the buckling coefficient of reactivity (Suzaki 1991).

The measured value of $\gamma$ is considered to be usable as benchmark data for the criticality calculation when used in conjunction with precise data of fuel compositions obtained from the post-irradiation examination. Furthermore, the value of K can be evaluated using the neutron flux of the spatially decaying state and the static adjoint flux, which defines $k_{eff}$ as the weight. The measured $\gamma$ can then be converted into $k_{eff}$ by Eq.(4) with the use of K, and thus be provided for the rather indirect calculation-to-experiment comparison of $k_{eff}$.

In JAERI's program for burnup credit, the subcriticality data to be measured for the actual spent fuel assemblies and the corresponding data of actinide and FP nuclides will be used to validate the criticality calculation codes. Moreover, fuel composition data of various burnup rates will be accumulated from the

existing destructive assay data. Through these activities, it is intended to establish the fundamental safety margins for the effective use of burnup credit.

**References**


Barbero, P. et al., "Post Irradiation Analysis of the OBRIGHEIM PWR Spent Fuel," Eur. Appl. Res. Rep.-Nucl. Sci. Technol. Sect. EUR-6589, 2 (1989) 129.

Fowler, T. B. et al.: ORNL-TM-2496, (1969).

Katakura, J. et al., "Development of the Computer Code System JACS for Criticality Safety," ANS Trans. 41 (1982) 329.

Naito Y. et al., "Development of Nuclear Decay Data Library JDDL, and Nuclear Generation and Decay Calculation Code COMRAD," JAERI-M86-121, 1986.

Naito Y. et al., "UNITBURN: A Computer Code for Burnup Calculation of a Unit Fuel Cell," JAERI-M91-019, 1990.

Science and Technology Agency (ed.), "Nuclear Criticality Safety Handbook," Nikkan-Shobo, 1988.

Suzaki, T. : J. Nucl. Sci. Technol., 28(12), p.1067, (1991).

Table 1 Coefficients a and b in Eq. (1) for each initial $^{235}U$-enrichment

| Enrichment [wt%] | a | b |
|---|---|---|
| 2.0 | 1.711 | 22.69 |
| 3.0 | 1.975 | 36.85 |
| 3.5 | 2.119 | 45.49 |
| 4.0 | 2.273 | 55.47 |
| 5.0 | 2.592 | 78.87 |
| 6.0 | 2.900 | 106.0 |

Table 2 Recommended value of the isotopic composition of PWR spent fuel

| Burnup [GWd/MtU] | $\frac{^{235}U/U}{(^{235}U/U)_{Init}}$ | $\frac{^{239}Pu}{Pu}$ | $\frac{^{240}Pu}{Pu}$ | $\frac{^{241}Pu}{Pu}$ | $\frac{^{242}Pu}{Pu}$ |
|---|---|---|---|---|---|
| 0 | 1 | 1 | 1 | 0 | 0 |
| 5 | 0.88 | 0.95 | 0.05 | 0 | 0 |
| 15 | 0.70 | 0.80 | 0.15 | 0.05 | 0 |
| 30 | 0.40 | 0.60 | 0.25 | 0.15 | 0 |
| 50 | 0.20 | 0.50 | 0.25 | 0.15 | 0.10 |

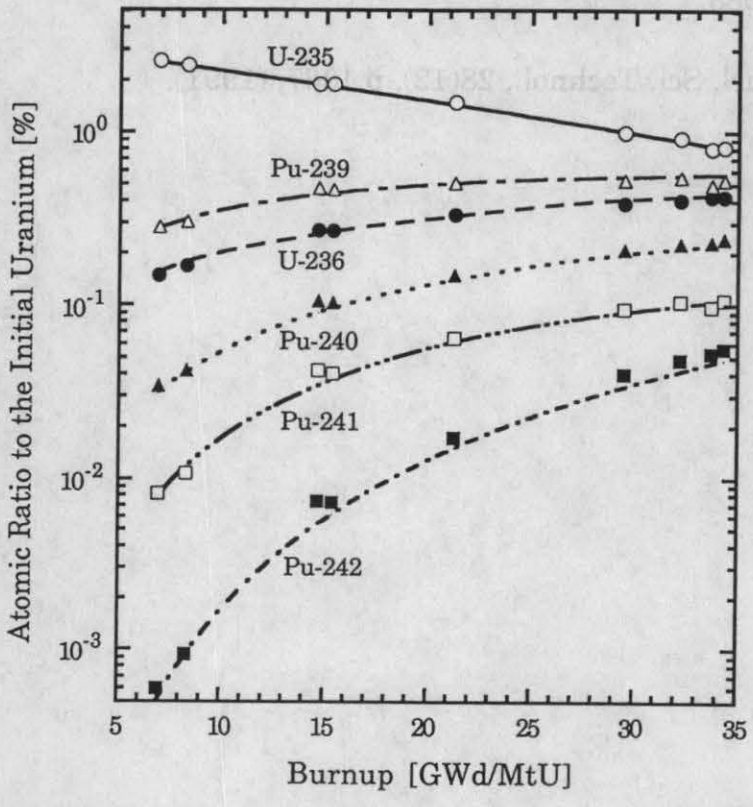


Fig. 1. Comparison of calculated and measured isotopic compositions of U and Pu in PWR spent fuels[2)]

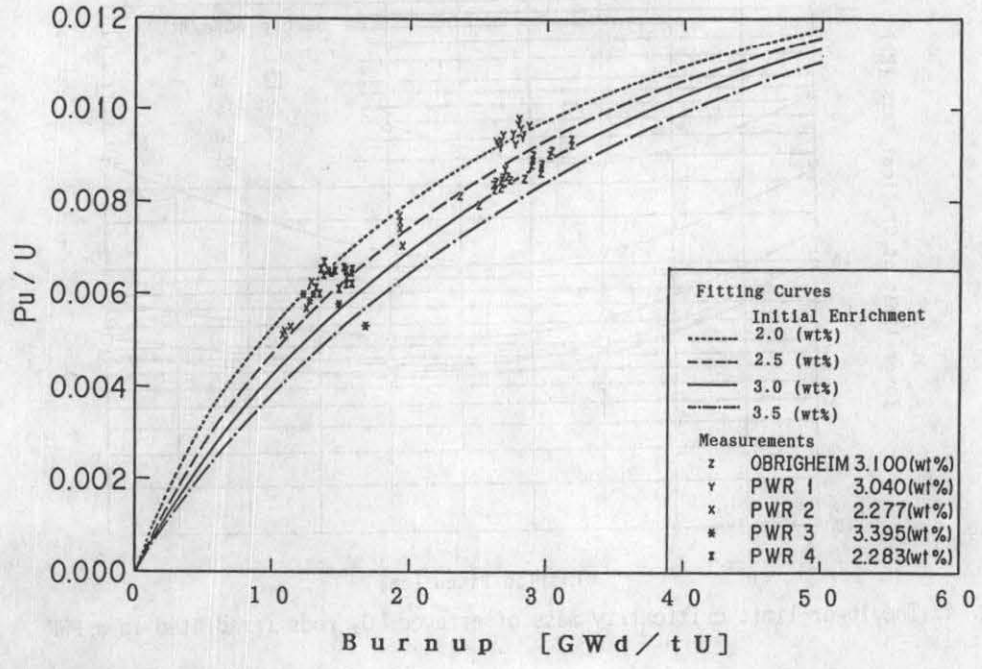


Fig. 2 Burnup dependence of Pu/U ratio
The symbols indicate measurements; the lines are expressed as $100 \cdot Pu/U = a(E) \cdot B / [b(E)+B]$, where B is fuel burnup and E is initial enrichment. The values of a and b corresponding to E are shown in Table 1.

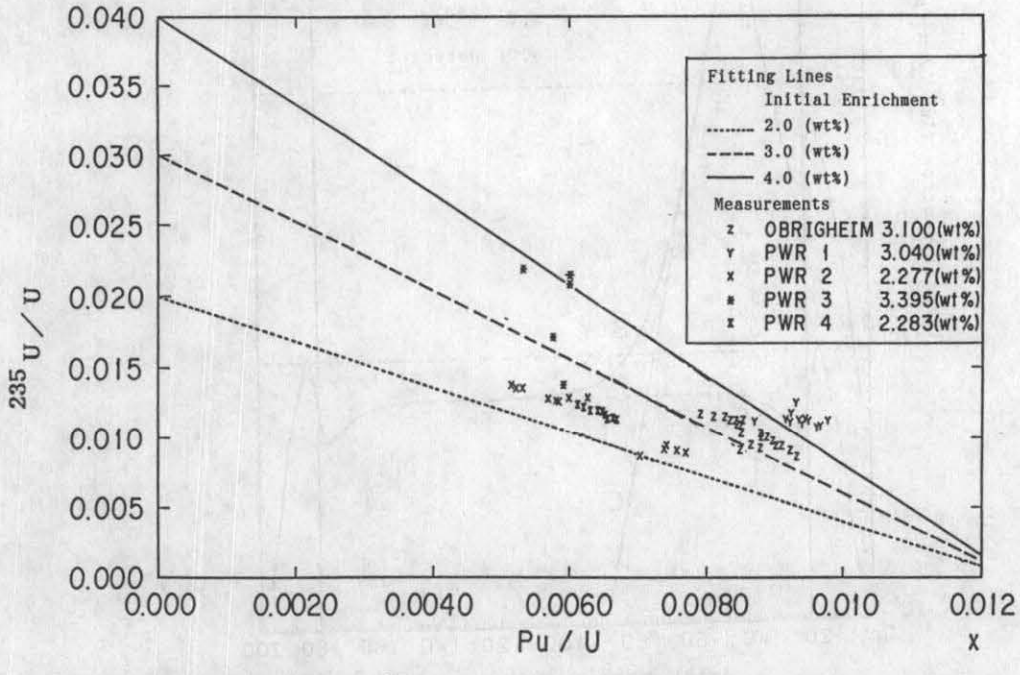


Fig. 3 The relation between $^{235}U/U$ and Pu/U ratios
The symbols indicate measurements; the lines are fitted by COMRAD calculations and expressed as $^{235}U/U = E \cdot (1-0.8x)$, where E is initial enrichment and x is Pu/U in percentage.

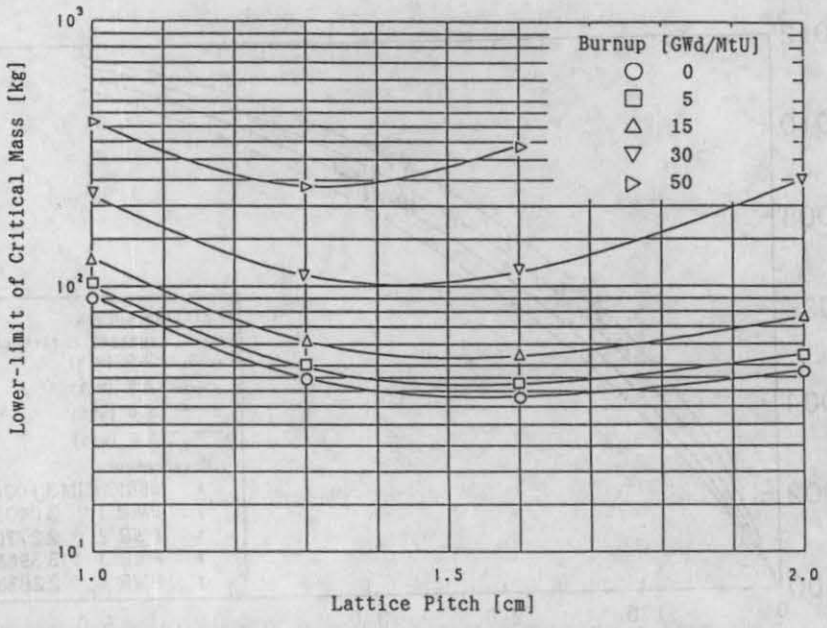


Fig. 4. The lower-limit criticality mass of arrayed $UO_2$ rods irradiated in a PWR

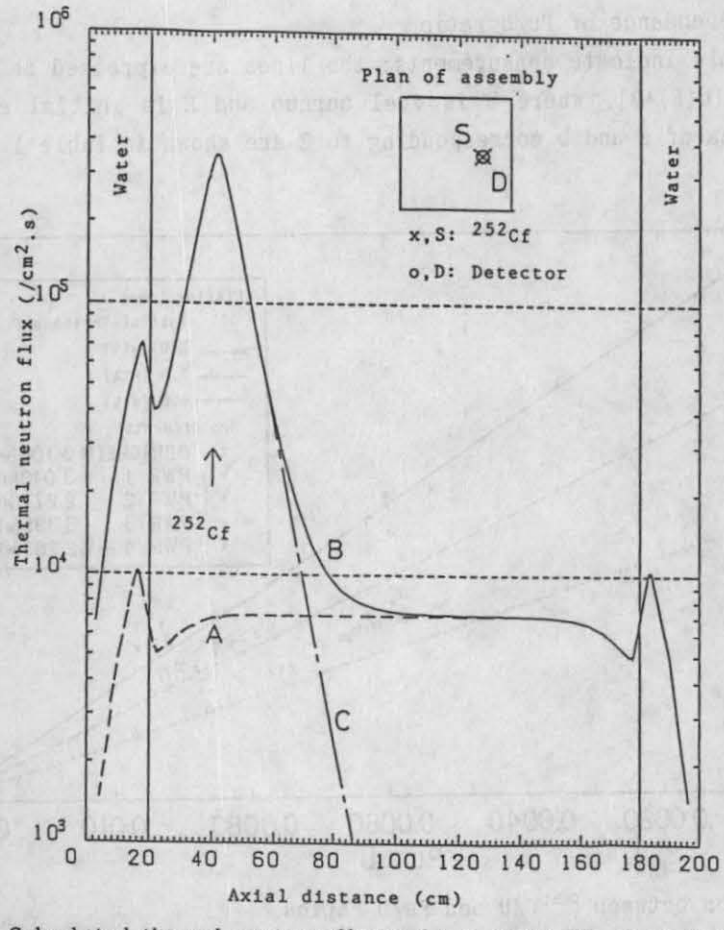


Fig. 5. Calculated thermal neutron fluxes for a 14x14 PWR spent fuel assembly of hypothetically shortened length (ini. enr. = 3.4%, burnup ~40GWd/t and cooling time ~13.5 yr). A: with inherent neutron source, B: with inherent and $^{252}Cf$ (20 mCi) neutron sources, and C: the result of A subtracted from B.