ANRCP-1998-15
October 1998

Amarillo National Resource Center for Plutonium

A Higher Education Consortium of The Texas A&M University System,
Texas Tech University, and The University of Texas System

Non-Destructive Control of the Plutonium Content of Spent Fuel Under Uncertainties on Irradiation History

V. Savander and V. Glebov
Department of Radiation Physics and Ecology
Moscow Engineering Physics Institute

This report was prepared with the support of the U.S. Department of Energy (DOE), Cooperative Agreement No. DE-FC04-95AL85832. However, any opinions, findings, conclusions, or recommendations expressed herein are those of the author(s) and do not necessarily reflect the views of DOE. This work was conducted through the Amarillo National Resource Center for Plutonium.

Edited by

Angela L. Woods
Technical Editor

MASTER

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600 South Tyler • Suite 800 • Amarillo, TX 79101
(806) 376-5533 • Fax: (806) 376-5561
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AMARILLO NATIONAL RESOURCE CENTER FOR PLUTONIUM/
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A Report on

**Non-Destructive Control of the Plutonium Content of Spent Fuel Under
Uncertainties on Irradiation History**

V. Savander and V. Glebov
Department of Radiation Physics and Ecology
Moscow Engineering Physics Institute
Moscow, Russia 115409

Submitted for publication to

Amarillo National Resource Center for Plutonium

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V. Savander and V. Glebov
Department of Radiation Physics and Ecology
Moscow Engineering Physics Institute
Moscow, Russia 115409

Abstract

It is apparent that serious attention must be given to the use of weapons-grade plutonium in fuel cycles in nuclear power plants. It is becoming increasingly necessary to organize a mixed fuel cycle which would be presumed to have ordinary, low-enriched uranium fuel and weapons-grade plutonium fuel in a one-time reactor core loading.

This research examined the problem of how to detect the possible substitution of a low-enriched uranium bundle for a bundle containing weapons-grade plutonium. Final determination of fuel assembly substitution was made using a non-destructive control methodology, which was based on gamma-spectra analysis of spent fuel for initial fuel composition identification.

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1. NON-DESTRUCTIVE CONTROL OF THE PLUTONIUM CONTENT OF SPENT FUEL UNDER UNCERTAINTIES ON IRRADIATION HISTORY

During recent years, questions related to weapons-grade plutonium utilization in fuel cycles of nuclear power plants have required serious attention. First, the VVER-1000 pressurized water reactors have been considered as an option because they are the basis of recent Russian Nuclear Energy. In the case of the fuel cycle of the previously mentioned reactors, calculational estimations of the possible weapons-grade plutonium involvement give 30% of total fuel loading without significant design and technology changes for the reactor core. This fact maintains the necessity to organize a mixed fuel cycle which presumes to have ordinary low enriched uranium fuel and plutonium-uranium fuel, based on weapons-grade plutonium, in a one-time reactor core loading.

Under the condition to keep a three-cycle refueling scheme within one year of fuel irradiation, enrichment of plutonium fuel should be about the same as uranium fuel. Therefore, the amount of weapons-grade plutonium per one fuel assembly will be significantly higher than the permitted quantity (PQ). As a result, it creates a problem of non-proliferation for weapons-grade plutonium that will be involved in a VVER-1000 fuel cycle.

This problem has many different aspects. In this report, we will examine how to detect the possible substitution of a low-enriched uranium bundle for a bundle containing weapons-grade plutonium. As a hypothetical situation that may be created at a nuclear power plant, we will consider substitution of plutonium assembly by uranium assembly. In this case, first of all, it is necessary to use an in-core control system for detection of such a substitution. It is possible to perform a first verification of a declared loading map and to find questionable situations.

Final determination of the fuel assembly substitution is planned to be made with non-destruction control methodology which is based on gamma-spectra analysis of a

spent fuel for initial fuel composition identification.

1.1 Statement of the Problem

Reactor core design and internal fuel cycle parameters for the VVER-1000 type of reactor were considered as the basis variant. It is assumed that weapons-grade plutonium involvement in the fuel cycles of these reactors will be made within the frame of the following existing design and technology limitations:

- Application of the VVER-1000 design for fuel element and fuel assembly;
- Conservation of a three-cycle refueling scheme and period of one cycle;
- Regular systems for control and safety.

One reactor core fuel loading contains ordinary uranium assemblies together with weapons-grade plutonium assemblies. It is assumed that plutonium fuel has been obtained by mixing weapons-grade plutonium with depleted uranium. In depleted uranium, the contents of U-235 is equal to 0.3% and isotopic composition of weapon-grade plutonium is a mixture of Pu-239 and Pu-240, or 93.0% and 7.0%, respectively.

Three different enrichments were taken into consideration during the calculational study: 3.6%, 4.0% and 4.4% (Table 1). For each uranium fuel enrichment determination of the plutonium fuel enrichment was performed under condition to obtain the same specific energy production.

It is assumed that during the refueling process, one plutonium assembly has been substituted by a uranium assembly in such a way that total assembly burnup remains unchanged. Presume that the in-core control system is able to select, with some degree of certainty, a fuel assembly which creates suspicion in conformity to a declared loading map. In this case, identification of selected fuel assemblies is required in order to determine conformity to a declared loading map. Particularly, it should be determined that the assembly was initially fueled with uranium or plutonium.

Table 1: Initial Nuclides Composition of Fuel for Uranium and Plutonium Assemblies

Variant	Uranium Fuel	Plutonium Fuel
1	3.6% - U-235 96.4% - U-238	0.3% U-235 96.4% U-238 3.1% Pu-239 0.2% Pu-240
2	4.0% - U-235 96.0% - U-238	0.3% U-235 96.0% U-238 3.4% Pu-239 0.3% Pu-240
3	4.4% - U-235 95.6% - U-238	0.3% U-235 95.6% U-238 3.8% Pu-239 0.3% Pu-240

It is reasonable to assume that such an identification can be performed during regular refueling by non-destructive control methodology based on gamma-spectra analysis. It should be noted that this identification has some delay, and that cooling time has influence on the accuracy of related gamma-spectra measurements.

Basis for such an identification is that for some fission products, release per one fission is considerably dependent on the fission nuclide. In the practice of gamma-spectra analysis of irradiated fuel, the following set of fission products is most frequently used: Zr-95, Nb-95, Ru-106, Cs-134, Cs-144 and Eu-156. Data on cumulative releases for these fission products are given below for fission of U-235, Pu-239, and Pu-241 by thermal neutrons (Table 2).

The data in Table 2 shows that Ru-106 and Eu-156 are the most suitable nuclides for

identification purposes. For these fission products, releases under fission of uranium and plutonium have about one order of difference. In this report we will consider the possibility of identification using only Ru-106 because the cross section of neutron absorption by Ru-106 is small for the thermal and resonance energy range, and hence, its accumulation in irradiated fuel is the result of two factors: fission and radioactive decay. In this case, the equation for Ru-106 accumulation is:

$$\frac{dp}{dt} + \lambda\rho = q(t); \quad \rho(t) = e^{-\mu} * \int_0^t q(\tau) * e^{\lambda\tau} d\tau$$

under zero initial conditions, where

$$q(t) = \sum_i \omega_i * \Sigma_{fi} * \Phi(t).$$

Table 2: Cumulative Releases and Half-Time for Some Fission Products of U-235, Pu-239 and Pu-241

Element	U-235	Pu-239	Pu-241
Zr-95, $T_{1/2}=64.0$ d	6.2	5.03	4.58
Nb-95, $T_{1/2}=35.1$ d	$6.0 * 10^{-4}$	$3.75 * 10^{-5}$	$1.28 * 10^{-2}$
Ru-106, $T_{1/2}=368.0$ d	0.400	4.31	6.18
Cs-134; $T_{1/2}=2.06$ y	$1.35 * 10^{-5}$	$9.88 * 10^{-4}$	$1.75 * 10^{-5}$
Cs-137, $T_{1/2}=30.2$ y	6.21	6.52	6.83
Ce-144, $T_{1/2}=284.3$ y	5.48	3.73	4.39
Eu-156, $T_{1/2}=4.06$ y	0.0132	0.118	0.179

In a cell approximation, the neutron flux $\Phi(t)$, is related to the specific reactor core power Q_0 by:

$$Q_0 = E_f * \sum_i \bar{\Sigma}_{fi} * \Phi(t).$$

After simple transformations we obtain

$$q(t) = \frac{Q_0}{E_f} * (\omega_s * \Delta_f^U + \omega_{Pu} * \Delta_f^{Pu}), \text{ where}$$

$\Delta_f^U, \Delta_f^{Pu}$ are the partial contributions of uranium and plutonium nuclides in the fission process, and $\omega_{Pu} = \frac{\omega_9 * \Sigma_{f9} + \omega_1 * \Sigma_{f1}}{\Sigma_{f9} + \Sigma_{f1}},$

which gives the release of Ru-106 per one fission of plutonium.

As a result, Ru-106 accumulation has a directly proportional dependence from the partial contributions of plutonium and uranium isotopes in the fission process. Because Ru-106 release from the fission of plutonium is one order of magnitude higher than in the case of its release from the fission of uranium, variation of the partial contributions of uranium and plutonium isotopes in the fission process (Figure 1) is an important factor. Data

was obtained for uranium fuel with 4.4% of enrichment ($x=4.4\%$) and corresponding plutonium fuel. It should be noted that for the plutonium assembly, the fraction of plutonium fission is practically independent from the period of irradiation, and is equal to about 90% during the life of the fuel. Therefore, all Ru-106 will have plutonium lineage. For a uranium assembly, the amount of Ru-106 that results from the contribution of plutonium isotopes as well as uranium isotopes, will be reached at 1/6 of the full fuel life-time.

As previously mentioned, the identification process of suspicious assemblies has been planned to be performed during regular refueling. In such a situation, the assembly to be verified and a trusted assembly will be allocated in experimental channels for gamma-spectra measurements after some cooling time. These gamma-spectra will be related to characteristic lines of Ru-106. Intensity relationships for those lines will be proportional to the concentrations of Ru-106 at the refueling moment. This value can be approximately described by the following formula:

Figure 1: Time Behavior of Partial Contribution of Pu Isotopes in Fission Process

$$\rho = \text{const} * PT * \left(\omega_s * \Delta_f^{\bar{U}} + \omega_{Pu} * \Delta_f^{\bar{Pu}} \right),$$

where $\Delta_f^{\bar{U}}, \Delta_f^{\bar{Pu}}$ partial contributions in fission for the related isotopes averaged through the whole irradiated period.

For the plutonium assembly this value is time-independent and for uranium assembly time dependence of this value is given in Figure 2.

Figure 2: Time Behavior of Partial Contribution of U and Pu Isotopes in Fission Process

Because the fuel will reach about 1/4 of the total burn-up after the first irradiation cycle, Ru release of the uranium assembly will also be determined by fission of plutonium. This conclusion is fully realized when the end of fuel lifetime has been reached. Therefore, it can finally be concluded that the ratio of intensities for gamma-lines of Ru-106 will be equal to the ratio of partial contributions to the total power production (PT) of plutonium isotopes in the case of uranium and plutonium assemblies.

For final estimations, we assume that we have obtained measured values of Ru-106 gamma-line intensities for the two assemblies to be compared (the "suspected" assembly and the "trusted" assembly). If intervals of these values do not cross each other when deviations are taken into account, identification of the fuel assemblies can be performed fully. We introduce a value named D, which is

the maximum deviation of measurements when it is still possible to perform full identification of uranium and plutonium assemblies. This value can be defined by the relationship of Ru-106 concentrations in these assemblies:

$$D = \frac{\rho_2 - \rho_1}{\rho_2 + \rho_1},$$

where ρ_2 and ρ_1 are the Ru-106 concentrations in the plutonium uranium assemblies, respectively.

Figure 3 represents behavior of this value as a function of irradiation period for these assemblies.

It is clear from Figure 3, that the maximum deviation in the measurements must be less than 60% when fresh, and 30% after the fuel is fully burned. It is known that recent deviation of measurements is not more than 10%, and thus, this is enough for identification purposes.

Figure 3: Uranium and Plutonium Fuel Burnup

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