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Title:

SAFEGUARDING THE DIRECT USE OF PLUTONIUM
IN CANDU REACTORS (DUPIC)

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SAFEGUARDING THE DIRECT USE OF SPENT PWR FUEL IN CANDU REACTORS (DUPIC)

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ABSTRACT

The fuel cycle options under consideration for the Direct Use of PWR Spent Fuel in CANDU Reactors (DUPIC) satisfy the need to maximize the energy output of nuclear fuel while minimizing the potential for proliferation of separated plutonium. Several options have been considered including repackaging options and thermal processing. Los Alamos has completed preliminary investigations of the safeguards aspects of the different processing options and the need for measurement development. Future safeguards activities will focus on the thermal-mechanical process selected for further development.

INTRODUCTION

Pressurized-water reactors (PWRs) use fuel manufactured from low-enriched uranium. Only a small fraction of the available energy content of the fuel is used in the once-through fuel cycle. Significant amounts of fissionable material remain in spent fuel. The traditional closed fuel cycle assumes that spent fuel will be processed to recover the remaining fissionable material for eventual reuse as reactor fuel. Alternatives to reprocessing have been investigated as a means to reduce proliferation of separated pure plutonium while still using the fissionable material remaining in spent fuel. DUPIC provides such a fuel cycle by reusing PWR spent fuel in Canadian deuterium uranium (CANDU) reactors without removing uranium. This fuel cycle presents a number of safeguards challenges.

SYSTEM STUDIES

Earlier work on safeguards for the DUPIC fuel cycle has been reported.¹ In this work, several different options in the DUPIC fuel cycle were investigated. These options include several processes to declad, cut, or re-clad PWR fuel elements to construct long or short CANDU fuel elements. Also, two thermal-mechanical processing options were studied.

The repackaging options all involve "item rebatching" in that none of these options involve dismantling fuel rods to the level of pellets. The safeguards systems for these options

would have similar features. Item accounting approaches are appropriate for these options. Maintaining continuity of knowledge for each item will be the key to safeguards for these processes.

Of the two thermal-mechanical processes, the OREOX (oxidation/reduction of oxide fuel) process is the more complex. In the OREOX process, PWR cladding is removed and the fuel is subjected to a series of high-temperature oxidation and reduction reactions. During the chemical changes, volatile fission products including xenon, krypton, iodine, technetium, and some molybdenum and ruthenium are removed. The product is then fabricated into new fuel pellets for CANDU rods. Both options involve bulk-handling operations where item integrity and identification are lost. Safeguards requirements for these processes are much more complex and necessitate detailed understanding of the flows of materials, measurements of bulk process materials, and detailed materials accounting systems. The safeguarding of the OREOX process was studied in more detail in the earlier work.

The preliminary safeguardability assessment of the DUPIC options was based on assumptions about the attributes of the fuel cycle. A complete assessment requires detailed information on process and material characteristics that are not yet available. The preliminary assessment is that both types of options are less prone to diversion than conventional aqueous reprocessing of spent nuclear fuels. With appropriate combinations of measurements, containment and surveillance, and materials accounting, it should be possible to provide adequate safeguards for the DUPIC fuel cycle.

NONDESTRUCTIVE ASSAY (NDA) INSTRUMENTS FOR DUPIC SAFEGUARDS

For NDA measurements, the DUPIC fuel cycle has much in common with conventional spent PWR fuel. Over the past decade, various NDA instruments and techniques have been developed for the safeguards verification of mixed oxide (MOX) and spent-fuel assemblies, and some of these techniques are in current use by the International Atomic Energy Agency (IAEA). These techniques are discussed below.

*Fork Detector.*² This combined neutron and gamma-ray detector includes a pair of ²³⁵U fission chambers and an ion chamber in a fork-shaped holder to position the detectors on opposite sides of a fuel assembly. The measurements are performed in the spent-fuel storage pond under about 6 m of water. The 5-minute measurement requires that the fuel assembly be raised about halfway out of the storage grid. The combination of the neutron count and the gamma-ray count is used to verify the declared burnup.

*GRAND Electronics.*³ The GRAND electronics unit, which was developed at Los Alamos, is now commercially available from the D. S. Davidson Company. The computer-based package provides the voltage and power supplies to operate the multiple neutron and gamma-ray detectors. On-board computer hardware and software provide the capability to collect and store data over extended time periods. This unit is now in

extensive use by the IAEA and EURATOM inspectorates for remote NDA operations. The on-board GRAND firmware and software have been developed for remote and unattended operation by the IAEA.

High-Resolution Gamma-Ray Spectroscopy. Information concerning the spent-fuel burnup level and cooling time can be derived from high-resolution gamma-ray spectroscopy measurements. The ratios of particular isotopes such as $^{134}\text{Cs}/^{137}\text{Cs}$ can be determined from gamma spectroscopy line ratios. This information is useful in checking the consistency of the reactor operator's fuel declarations. However, the data are limited by the lack of penetrability of the gamma rays and the potential change in the result as the volatile fission products are released in the DUPIC fuel cycle process.

*Core Discharge Monitor.*⁴ The core discharge monitor system monitors the presence of radioactive materials outside the reactor containment shielding of CANDU reactors. The system is able to detect the neutron and gamma-ray activity present when irradiated fuel bundles are removed from the reactor. The measurement subsystem consists of the fission and ionization chambers, preamplifiers for the fission chambers, and a GRAND electronics package. The application required the detectors to be placed on a wall in each refueling machine room close to the reactor face.

*CANDU MOX Bundle NDA System.*⁵ A neutron coincidence counter is being used by the IAEA to verify plutonium content in fresh CANDU fuel bundles. For CANDU bundles containing spent fuel, this system would have to be modified with lead shielding for gamma-ray attenuation. The addition of multiplicity counting has the potential of separating the ^{244}Cm neutrons; however, research and development would be required to establish the feasibility of this technique.

*Continuous Operation Neutron and Gamma-Ray Monitors.*⁶ Because of the high level of radiation from the DUPIC fuel, it will be necessary to use remotely operated NDA systems. To provide continuity of knowledge during the remote operation of the instruments, it is usually desirable to operate the instruments in the continuous mode. This type of operation requires special highly reliable software for data storage and review and a total system that can be authenticated by the IAEA. Systems that satisfy these stringent reliability and authentication requirements for remote operation are used by the IAEA at the Power Reactor and Nuclear Fuel Development Corporation's MOX fabrication plant in Japan as well as the JOYO and MONJU reactors in Japan and a CANDU reactor in Canada.

*Plutonium-Scrap Multiplicity Counter.*⁷ During the past several years, neutron coincidence counting has been upgraded to neutron multiplicity counting. The higher multiplicity in the neutron counting makes it possible to separate the ^{244}Cm and plutonium components in the spent fuel. However, the error associated with this technique has not yet been established. A new plutonium-scrap multiplicity counter is being evaluated by the IAEA at a MOX fuel fabrication facility in Japan.

The extremely high neutron and gamma-ray radiation levels that are inherent in the DUPIC fuel cycle make this fuel resistant to diversion attempts. Unfortunately, many of the conventional methods of measuring special nuclear material cannot be used because of the high radiation levels. However, it should be possible to take advantage of the intense neutron and gamma-ray signals associated with the DUPIC fuel to quantitatively verify the amount of fuel for safeguards purposes.

Because no chemical reprocessing is involved in the DUPIC fuel cycle, the ratio of ^{244}Cm to plutonium is constant at the input, output, and process steps, and we can establish the associated plutonium inventory. The ^{244}Cm isotope is a prolific source of spontaneous fission neutrons, and we can separate background neutrons by means of coincidence or time-correlation neutron counting. Over 95% of the neutron emission from spent PWR fuel originates from ^{244}Cm ($T_{1/2} = 18.1$ y) after a few years of cooling time to allow the decay of the short-lived isotopes such as ^{242}Cm ($T_{1/2} = 163$ d).

NDA instrumentation presently used by the IAEA for spent-fuel verification includes gamma-ray detectors such as ion chambers, sodium iodide, and germanium; these should also be considered for safeguarding the DUPIC fuel cycle. However, some of the mechanical steps in the DUPIC process will remove volatile fission products, so gamma-ray spectrometry would probably be required to isolate specific non-volatile fission products.

The extremely high levels of neutron and gamma-ray emissions from the DUPIC fuel will make traditional NDA techniques difficult to apply. On the other hand, the strong spontaneous-fission-neutron signal from ^{244}Cm can be measured quantitatively at all steps in the DUPIC fuel cycle.

FUTURE ACTIVITIES

In Phase II of the project, Los Alamos will assist the Korea Atomic Energy Research Institute (KAERI) in the development, design, and evaluation of safeguards approaches for the OREOX process. Los Alamos will also be tasked with development, design, and evaluation of NDA measurement techniques and hardware necessary for measuring nuclear material in the DUPIC fuel cycle. KAERI and Atomic Energy of Canada Limited are planning fuel fabrication experiments in Phase II that could be used for testing NDA measurement hardware.

In Phase II of the DUPIC process, Canada and Korea are planning to fabricate CANDU fuel pins and a fuel bundle from spent PWR fuel. The first experiment will take place at Chalk River, Canada, where US-origin spent PWR fuel will be reconstituted into fuel pins. This will provide an opportunity for proof-of-principle tests of NDA equipment. The second experiment will take place in hot cells at the KAERI test reactor in Taejeon. Los Alamos and KAERI will develop a safeguards approach for this experiment in which spent Korean PWR fuel will be reconstituted into CANDU fuel pins using the OREOX process. Los Alamos also will develop and install NDA equipment for measuring a spent fuel assembly transferred into the hot cells and CANDU assemblies fabricated in the process.

REFERENCES

1. K. K. S. PILLAY, H. O. MENLOVE, and R. R. PICARD, "Safeguardability of Direct Use of Spent PWR Fuels in CANDU Reactors," LA-12432-MS, Los Alamos National Laboratory (October 1992).
2. P. M. RINARD and G. E. BOSLER, "Safeguarding PWR Spent Fuel with the Fork Detector," LA-11096-MS, Los Alamos National Laboratory (March 1988).
3. J. K. HALBIG, G. E. BOSLER, S. F. KLOSTERBUER, and P. M. RINARD, "Applications of a Transportable Spent-Fuel Measurement System," LA-UR-85-3735, Los Alamos National Laboratory (1985).
4. J. K. HALBIG and A. C. MONTICONE, "Proof-of-Principle for an NDA-Based Core Discharge Monitor," *Nucl. Mater. Manage. XIX*, 847-852 (1990).
5. H. O. MENLOVE, et al., "CANDU MOX (CMOX) Counter Design and Operation Manual," LA-12101-M (ISPO-336), Los Alamos National Laboratory (October 1991).
6. J. K. HALBIG, et al., "Applications for Unattended, Remote NDA Instrumentation," LA-UR-91-3542, Los Alamos National Laboratory (1991).
7. D. G. LANGNER, M. S. KRICK, and N. ENNSLIN, "Pyrochemical Multiplicity Counter Design," *Nucl. Mater. Manage. XIX*, 411-415 (1990).

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