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An Improved Characterization Method for International Accountancy Measurements of Fresh and Irradiated Mixed Oxide (MOX) Fuel: Helping Achieve Continual Monitoring and Safeguards through the Fuel Cycle

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Abstract

Nuclear fuel accountancy measurements are conducted at several points through the nuclear fuel cycle to ensure continuity of knowledge (CofK) of special nuclear material (SNM). Non-destructive assay (NDA) measurements are performed on fresh fuel (prior to irradiation in a reactor) and spent nuclear fuel (SNF) post-irradiation. We have developed a fuel assembly characterization system, based on the novel concept of "neutron fingerprinting" with multiplicity signatures to ensure detailed CofK of nuclear fuel through the entire fuel cycle. The neutron fingerprint in this case is determined by the measurement of the various correlated neutron signatures, specific to fuel isotopic composition, and therefore offers greater sensitivity to variations in fissile content among fuel assemblies than other techniques such as gross neutron counting. This neutron fingerprint could be measured at the point of fuel dispatch (e.g. from a fuel fabrication plant prior to irradiation, or from a reactor site post-irradiation), monitored during transportation of the fuel assembly, and measured at a subsequent receiving site (e.g. at the reactor site prior to irradiation, or reprocessing facility post-irradiation); this would confirm that no unexpected changes to the fuel composition or amount have taken place during transportation and/ or reactor operations. Changes may indicate an attempt to divert material for example. Here, we present the current state of the practice of fuel measurements for both fresh mixed oxide (MOX) fuel and SNF (both MOX and uranium dioxide). This is presented in the framework of international safeguards perspectives from the US and UK. We also postulate as to how the neutron fingerprinting concept could lead to improved fuel characterization (both fresh MOX and SNF) resulting in: (a) assured CofK of fuel across the nuclear fuel cycle, (b) improved detection of SNM diversion, and (c) greater confidence in safeguards of SNF transportation.

Introduction

Safeguarding the Commercial Nuclear Fuel Cycle

Preventing the proliferation of nuclear weapons is a complex mission. One facet of meeting this mission and thus a task of nuclear safeguards is reducing the probability of misuse of the commercial nuclear fuel cycle. It should be noted however that there has been no historical record of a case where the material used in nuclear weapons production has been obtained by the diversion of material from the civil nuclear fuel cycle [1]; nevertheless the potential for this occurrence should be mitigated and addressed in a timely manner pending the expansion of nuclear energy around the world. International safeguards form the institutional, legal and technical frameworks which ensure a synergy between the peaceful expansion of nuclear technology and the prevention of the spread of nuclear weapons.

This paper discusses one implementation of international safeguards at the working level; safeguarding the fissile material content of nuclear fuel assemblies – both fresh (in the case of mixed oxide (MOX) fuels) and irradiated fuels. The neutron fingerprinting concept and technique presented in this paper can be used to verify that the fissile content of nuclear fuel assemblies has not (within tolerances) been diverted from peaceful purposes, and can be applied at any stage of the commercial nuclear fuel cycle prior to reprocessing of those assemblies i.e. where the nuclear fuel assemblies remain intact. Today the majority of the Pu inventory in the world exists at the back-end fuel cycle in the form of spent nuclear fuel (SNF) [2], or separated Pu following the reprocessing of SNF assemblies [3]. Since almost any grade of commercial Pu can be made in to a nuclear explosive (or alternatively a radiation dispersal device), maintaining an inventory of and securing Pu in this form is essential to maintaining global security. In the next section we present estimates for Pu arisings to illustrate the magnitude of the challenge. The sections that follow present the concept of neutron fingerprinting using neutron multiplicity signatures and the instrument design that could be used to implement this technique.

Nuclear Renaissance and Impact on Future Safeguards

A worldwide renaissance of nuclear energy and the expansion of the currently operating light water reactor (LWR) fleet inevitably lead to the growth of the global SNF inventory and thereby an increase in the total Pu inventory in the world in future years. Let us first consider the U.S. and then the U.K. by way of examples. Although not a comprehensive evaluation, consideration of these two nations raises some interesting considerations for the future use of nuclear energy and the demand placed on nuclear fuel safeguards for the future.

The U.S. currently employs an open, or once-through, nuclear fuel cycle whereby SNF is stored pending disposal in a long-term repository, although alternative options, including recycle and re-use of Pu in thermal reactors is now being considered. Table 1 contains our estimates of the historic and future arisings in the amount of SNF and the fissile Pu inventory in the U.S. reactor fleet. Based on the historic fuel arisings of 62,000 tHM of SNF as of 2010 (with an average discharge burnup of 33 GWd/tHM) there are approximately 500 t of Pu contained in the fuel with a fissile quality of approximately 68 wt % Pu-fissile (60 wt % ^{239}Pu + 8 wt % ^{241}Pu). There will be approximately 0.8 wt % ^{235}U remaining of the average initial ^{235}U enrichment of 3.2 wt %.

Pending reactor new build in the U.S., for the "future fleet" of U.S. next generation LWRs we have assumed the 22 units "proposed" (equating to 32 GWe) and 9 units "planned" (equating to 11.6 GWe) due to start in the next 10 years, according to the World Nuclear Association [4]. In addition, there are several scenarios as to how much "Phase 2" new build there will be after these units. For example, the 2003 MIT study "The Future of Nuclear Power" [5] states that 300 GWe of nuclear are projected for the US by 2050. Previous work on GNEP by one of the co-authors has independently derived a figure of around 200+ GWe. It therefore follows that the projected capacity of nuclear energy in the U.S. by 2050 is expected to be of the order of 200-300 GWe, using LWR technology.

On the basis of this number of GWye to be generated, and assuming 15-20 tHM/GWye of spent fuel is discharged, then approximately 5000 tHM/yr of SNF will be generated by the future U.S. fleet. Therefore the new build of reactors, with an expected lifetime of 60 years, will produce approximately 300,000 tonnes of SNF (in addition to the 62,000 tonnes already produced). Based on reactor physics inventory calculations, the Pu composition of SNF will be approx 1.2% total Pu. On this basis, the future fuel will contain 3,600 tonnes of Pu (total Pu content). At the higher average burnups appropriate to the future reactors, the fissile compositions will be approximately 65 wt % Pu-fissile (50 wt % ^{239}Pu + 15% ^{241}Pu) and there will be approximately 0.8 wt % ^{235}U remaining of the average initial ^{235}U enrichment of 4.8 wt % assumed.

The notable increase in the future SNF compared with historic fuel reflects the difference in GWe produced; 101 GWe historic c.f. 200-300 GWe future generating capacity (upper limit assumed above). Plus, the next generation of LWRs will operate for 60 years compared with nearer 40 years for the historic fleet. The higher Pu content in the future fuels (1.2 wt % c.f. 0.9 wt %) reflects the accumulation of Pu expected with burnup i.e. the continued breeding of Pu from ^{238}U captures.

The fissile content of the lower burnup material is as expected i.e. higher than the future fuels (68 wt % c.f. 65%). This reflects the fact that in the higher burnup fuel, the

higher total Pu composition is contributing more to the overall energy at higher burnups and as such is being depleted and at the same time leads to greater production of the higher Pu isotopes. The final ^{235}U enrichments (historic and future) are almost identical (0.8 wt %) - this reflects the fact that the fuels have been taken to their natural "end of life".

	Electrical Output (GWe)	Discharge Burnup (GWd/tHM)	Fuel initial Enrichment (average wt% ^{235}U)	SNF (tU)	Total Pu content of SNF (t Pu)	Fissile Quality Pu (wt % Pu-fissile)
Historic Fleet (as of 2010)	101	33	3.2	62,000	500	68
Future Fleet	200-300	60	4.8	300,000	3,600	65

Table 1. Estimates of the historic and future arisings in the amount of SNF and the fissile Pu inventory in the U.S. reactor fleet

The 300,000 tHM of fuel required for such a scenario (which is assumed in many U.S. studies) will require a little over 3,000,000 tU ore. Based on World Nuclear Association predictions [6], the amount of U ore that is "reasonably assured" and based on a reasonable economic price of U ore (<130 $/kgU) is around 5-6 million tU ore. Based on these assumptions, the future U.S. fleet alone would require approximately 50 % of the world's U ore. Experts have therefore concluded that, for reasons of sustainability of U ore and because of the volumes of SNF arising, the U.S. will ultimately need to consider recycling SNF and move from an open to a modified open (using recycle of Pu as MOX in thermal reactors), and then to finally a fully closed fuel cycle using fast reactors.

In a similar vein, but with different drivers, the UK has adopted a modified open fuel cycle for many decades where the SNF from the gas reactor fleet (Magnox and Advanced Gas Cooled Reactors (AGRs)) is reprocessed. This strategy was adopted both for technical reasons (as the Magnox fuel can not be stored indefinitely under water or in air) and for economic considerations (expecting U ore prices to escalate and fast reactors to be required). As such, the UK will have approximately 100 tonnes of separated Pu currently described as a "zero value asset" awaiting consideration of the credible options by UK Government, which includes the potential re-use of Pu as MOX fuel in future LWRs [7]. This means that the UK finds itself in a similar situation to the U.S. in terms of future fuel cycle and thus safeguards needs.

The recent MIT study "The Future of the Nuclear Fuel Cycle" [8] recommends planning for SNF management as an integral part of nuclear fuel cycle design. Furthermore, the study also recommends that the U.S. should move toward centralized spent fuel storage sites. This in turn will increase the need for fuel transportation and material movements requiring safeguards. Any transition toward a more sustainable fuel cycle, such as the modified open fuel cycle anywhere in the world would mean that Pu would be separated from SNF for a program of reactor re-use using a mixed oxide (MOX) of Pu and uranium (U). The use of MOX, with its increased Pu content along with increased material transportation and separated Pu, will require rigorous technical safeguards measures throughout the fuel cycle. It is therefore anticipated that accountancy measurements and monitoring will be required at the following stages: transport of spent uranium fuels, separation of Pu, storage of Pu, production of MOX fuel, transportation of MOX fuel and irradiation of MOX fuel.

If the issue of spent fuel management is a challenge for the U.S. and the UK, then it is clear that as nuclear energy expands around the world, this challenge will also need to be addressed in those existing nations expanding their nuclear programs as well as those new nuclear nations looking to adopt nuclear. In short, this means potentially treating Pu as an energy resource rather than as a waste form. This places a greater emphasis on safeguarding MOX fuel and a need for instrumentation to be designed to meet the requirements of MOX fuel safeguards as well as U oxide fuels. This paper suggests a technology for addressing this challenge.

Role of Instrumentation in SNF Safeguards

Instrumentation serves the following purposes in SNF safeguards:

(1) Determining shipper-receiver difference [9]
(2) Establishing and maintaining continuity of knowledge (CofK) of the fuel as it discharged from the reactor core and either reprocessed or packaged and stored at a storage facility [10]
(3) Recovering CofK in the event of a failure or significant gap in the continuous containment and surveillance (C/S) [11]
(4) Independently confirming the presence and content of Pu in the SNF via measurements [10]
(5) Detecting material diversion and thus functioning as a deterrent to those attempting to divert

Neutron Fingerprinting using Multiplicity Signatures

Concept

Nuclear fuel presents a complex measurement challenge. Fuel is a highly multiplying measurement item, further the fuel and detection system are closely coupled, which are departures from the scenarios traditionally encountered in safeguards measurements. To date it is difficult to measure the mass of plutonium in spent fuel directly from the detected count rates. The traditional approach has been to rely on inventory calculations and operator declarations verified using observable signals such as gross neutron or gross gamma counting rates. Even relative to a reference item it is difficult to make quantitative measurements without assuming and applying additional correction factors for cooling time to account for the in-growth of neutron absorbers within the fuel assembly which reduce the detected count rates. In addition, an accurate calibration scheme over a representative range of initial enrichments, burnup values and cooling times would prove cumbersome.

Neutron fingerprinting alleviates the need for representative calibration standards. It also has the advantage that it is not therefore dependent on fuel type. Neutron fingerprinting is based on the unique neutron emission characteristics of nuclear fuel assemblies with varying isotopic composition. A single measurement can be used to characterize a nuclear fuel assembly by its unique neutron signature. A repeat of this measurement *in situ* (e.g. fuel cask monitoring), or following transportation of the fuel assembly to another fuel cycle facility, enables the second result to confirm that no nuclear material had been diverted from the storage site or during material movements. If the measurement was repeated within a short time period, the neutron counting rates characterizing the fingerprint would be expected to be identical within statistical variation. If the measurement was repeated after a longer time period, decay corrections would need to be applied to the measured counting rates. The time-dependence of the neutron emission rate from a spent nuclear fuel assembly varies in a well-known manner as a function of the time of the removal from the power reactor [12]. Curium-242 decays to Curium-244 with a 162.8 day half-life. Curium represents ~99% of the primary neutron emission from a fuel assembly and is therefore the source term that drives subsequent multiplication within the fuel assembly itself. Fissile Plutonium-241 also decays as a function of cooling time to Am-241 with a 14.3 year half-life.

Traditionally, neutron fingerprinting measurements are performed by comparing total neutron counting rates. In principle, material could be diverted from the fuel assembly and replaced in such a manner as to artificially boost the total neutron counting rate to the same value. Here, we propose the extension to correlated counting rates and ratios for a "harder to spoof" signature.

The neutron fingerprinting technique presented here can be used to address points: (1), (2) [with the exception of reprocessing], (3), (4) [although used to measure signatures not Pu content] and (5).

Neutron Multiplicity Counting

Spontaneous and induced fission result in the emission of groups of neutrons closely correlated in time. The detection of these temporally correlated neutrons and the analysis of detected events within timing gates in electronics enable correlated neutron counting rates to be obtained. The number of neutrons emitted per fission event is known as the neutron multiplicity and is unique to each fuel assembly in the sense that the Cm content is very sensitive to burnup and the burnup profile is characteristic of the assembly.

Neutron multiplicity counting yields three primary signatures, with Doubles and Triples counting rates arising from temporally correlated neutrons:

- Singles (totals) counting rate
- Doubles (pairs) counting rate
- Triples counting rate

The three counting rates can be obtained using a number of different timing gates for counting precision and the discrimination of induced to spontaneous fission events (which exhibit different temporal behavior). Ratios of counting rates provide additional signatures and are harder to spoof:

- Doubles to Singles ratio
- Triples to Singles ratio
- Triples to Doubles ratio

Further, the inclusion of a Cadmium (Cd) liner in the detector assembly provides an additional measurement condition which yields a further five signatures from the fuel assembly. An axial scan of a fuel assembly during a measurement enables a burnup profile to be constructed and rotation of a fuel assembly during a measurement may be performed to counteract spatial inhomogeneity. A typical SNF assembly from a LWR is ~ 3.5 m in height. The detection system would be ~ 0.5 m in height, enabling 7 axial scans of the assembly to be performed. This results in a total of 14 scans both with and without the Cd liner in place.

The composition of a fuel assembly depends on the following parameters:

- Neutron spectrum and thus reactor type (e.g. PWR, BWR, Candu etc)
- Initial heavy metal enrichment (wt % HM)
- Burnup achieved in a power reactor (GWd/tHM)
- Time since discharge from the reactor until the measurement, or cooling time (years)

The number of neutrons available for detection is dependent on the leakage multiplication which represents the number of neutrons that leak out from the fuel assembly. In turn this quantity is dependent on the neutron capture probability within the fuel assembly (without fission) and the probability of induced fission (leading to neutron multiplication). Neutron emission will therefore be dependent on the following parameters:

- Presence of fissionable isotopes
- Presence of fissile isotopes
- Incident neutron energy spectrum
- Moderation and multiplication within the fuel assembly (fast and thermal fission)

The apparent signature can therefore be altered by the surrounding storage medium and coupling with the detector assembly. These factors can be controlled at the measurement station.

Characterization System

Los Alamos National Laboratory is part of an international safeguards program for the research and development of instruments to non-destructively assay Pu content in spent fuel. All neutron instruments being developed under this program can be applied to implement neutron fingerprinting. The highest efficiency instrument that is being developed is Differential Die-Away Self-Interrogation (DDSI) detection system, proposed and developed by Menlove, *et al.* [13]. Figure 1 shows the MCNPX design of the detection geometry. A single SNF assembly is centered in the detector assembly. The system has 58 ^{3}He gas-filled proportional counters, or tubes, available for neutron detection.

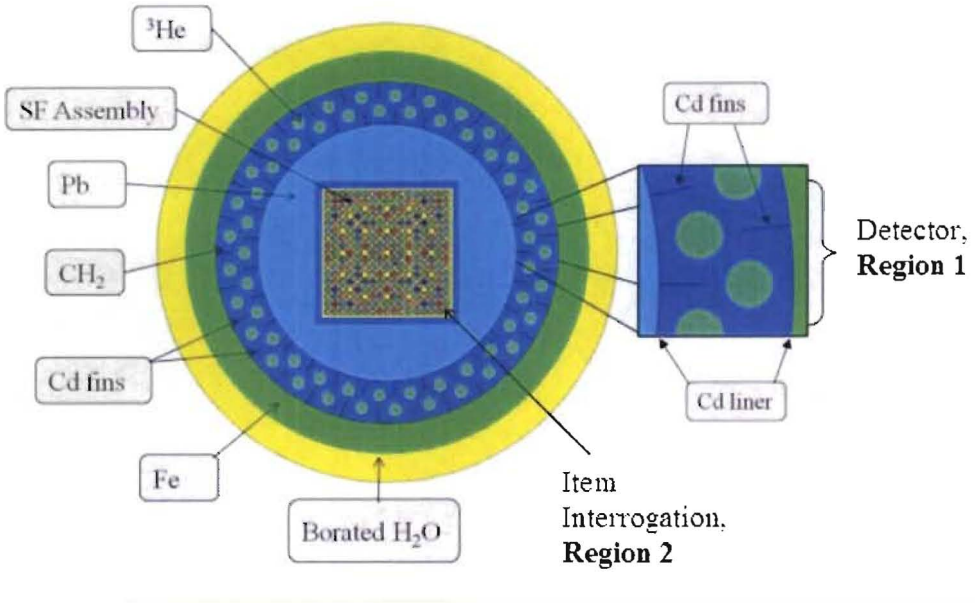

Figure 1. DDSI MCNPX Detector Geometry, developed by M. Schear [14].

Passive Neutron Albedo Reactivity implemented with ^{3}He gas-filled proportional counters (PNAR-^{3}He) is a more straightforward technique that can be implemented using the DDSI-designed system. The method of Passive Neutron Albedo Reactivity (PNAR) was first proposed by Menlove and Beddingfield in 1997 and applied to neutron multiplicity measurements of uranium fuel rods [15]. PNAR utilizes the self-interrogation of an assay item via reflection of neutrons born in the item (e.g. SNF assembly) back in to the item. The neutrons originate primarily from spontaneous fission events (e.g. ^{244}Cm) and (α, n) reactions (e.g. oxides) within the item itself but are amplified by multiplication. The presence and removal of a Cd liner (~1mm thick) between the reflecting boundary and the item provides two measurement conditions with different neutron energy spectra and therefore different interrogating neutron characteristics. In the case with the Cd liner removed, reflected low energy neutrons (thermal neutron albedo) are incident on the fuel assembly and the number of induced fissions, hence neutron multiplication within the fuel, are increased. This amplifies the original spontaneous neutron emission from the fuel. Cd has a high cross-section of absorption for low energy neutrons (< 0.5 eV) therefore the presence of the Cd liner greatly reduces the number of low energy (primarily thermal) neutrons returning to the fuel. PNAR is used to assay fissile content in an item by detecting the change in multiplication between these two measurement conditions.

The detection system design shown in Figure 1 therefore features a 1mm Cd liner in the item interrogation region; which can be present or removed in order to perform two measurements and obtain a Cd ratio. A second Cd liner is also in place in the detector region. Without this additional Cd layer surrounding the neutron detection medium, detection efficiency, ε, is also increased when the inner Cd liner is removed since slow neutrons from the water stored fuel assembly can emerge to reach the detector surrounding the fuel assembly.

Conclusion

In conclusion, neutron fingerprinting via neutron multiplicity signatures is an improved SNF characterization method over neutron fingerprinting via gross neutron or gross gamma counting. The PNAR-^{3}He instrument is an ideal candidate for this technique. The measurement of three counting rates and three counting rate ratios at seven axial positions of a SNF assembly, both with and without Cd, leads to 84 neutron signatures from a single assembly. This technique therefore has the potential to produce a tight identification on a SNF assembly.

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