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Determination of Plutonium Content in Spent Fuel with Nondestructive Assay

by

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ABSTRACT

There are a variety of reasons for quantifying plutonium (Pu) in spent fuel such as independently verifying the Pu content declared by a regulated facility, making shipper/receiver mass declarations, and quantifying the input mass at a reprocessing facility. As part of the Next Generation Safeguards Initiative, NA-241 has recently funded a multilab/university collaboration to determine the elemental Pu mass in spent fuel assemblies. This research effort is anticipated to be a five year effort: the first part is a two years Monte Carlo modeling effort to integrate and down-select among 13 nondestructive assay (NDA) technologies, followed by one year for fabricating instruments and then two years for measuring spent fuel. This paper gives a brief overview of the approach being taken for the Monte Carlo research effort. In addition, preliminary results for the first NDA instrument studied in detail, delayed neutron detection, will be presented. In order to cost effectively and robustly model the performance of several NDA techniques, an "assembly library" was created that contains a diverse range of pressurized water reactor spent fuel assemblies (burnup, enrichment, cooling time) similar to that which exists in spent pools today and in the future, diversion scenarios that capture a range of possible rod removal options, spatial and isotopic detail needed to accurately quantify the capability of all the NDA techniques so as to enable integration. Integration is being designed into this study from the beginning since it is expected that the best performance will be obtained by combining a few NDA techniques. The performance of each instrument will be quantified for the full assembly library in three different media: air, water and borated water. In this paper the preliminary capability of delayed neutron detection will be quantified for the spent fuel library for all three media. The 13 NDA techniques being researched are the following: Delayed Gamma, Delayed Neutrons, Differential Die-Away, Differential Die-Away Self-Interrogation, Lead Slowing Down Spectrometer, Neutron Multiplicity, Nuclear Resonance Fluorescence, Passive Prompt Gamma, Passive Neutron Albedo Reactivity, Self-integration Neutron Resonance Densitometry, Total Neutron (Gross Neutron), X-Ray Fluorescence, ^{252}Cf Interrogation with Prompt Neutron Detection.

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INTRODUCTION

Although the majority of plutonium (Pu) in the world is stored in commercial spent fuel assemblies, a measurement system for directly quantifying the Pu mass contained in these assemblies does not exist. The nondestructive assay systems in use today (Safeguards Mox Python Detector,[1] Fork Detector[2] and Cerenkov Viewing Device [3]) primarily measure indirect signatures from spent fuel such as gamma emission from fission fragments, or photons induced by radiation from fission fragment, or total neutron emission pre-dominantly emitted from curium. Calculation codes, known as burnup codes, can be used to infer plutonium mass from these measured signatures. In order to accurately use burnup codes to predict the Pu mass in a particular assembly, input from the operator is required. From an international safeguards perspective, this input is undesirable given the regulatory requirement of independent verification.

Below, five safeguards motivations and four non-safeguards motivations for improving on the status quo are listed. The safeguards reasons for designing a nondestructive assay (NDA) system that can quantify the Pu mass in spent fuel assemblies are the following: (1) Provide regulators with the capability to independently verify the mass of plutonium at any site that has spent fuel. (2) Enable regulators to accurately quantify the Pu mass leaving one facility and arriving at another facility ("shipper/receiver difference"). (3) Provide regulators with a tool for recovering continuity of knowledge at any site storing spent fuel. (4) Enable determination of the input accountability mass of an electro-chemical (pyro-chemical) processing facility. (5) Provide facility operators with a means for quantifying the Pu mass in spent fuel that is no longer considered "self-protecting."

The non-safeguards motivations are the following: (1) Provide confidence to the public that the shipment of spent fuel around the world is being undertaken in a rigorous manner; assure that material is not diverted during shipment. (2) Provide reactor operators with a tool enabling optimal reloading of reactor cores. (3) Enable any place that stores spent fuel, either for the short or long term, to do so more efficiently ("burnup credit"). (4) Promote cost savings by facilitating assembly selection at a reprocessing facility. Facility operators blend assemblies to obtain optimal compositions in reprocessing solutions. The blending is presently based on reactor history and burnup codes. The inaccuracy of the status quo decreases plant operational efficiency.

TECHNICAL GOALS AND THE NEED FOR INTEGRATION

There are two primary technical goals for this research effort: (1) Determine the absolute accuracy of an NDA system for determining Pu mass in an assembly. And (2) determine the sensitivity of each NDA system to the removal of pins.

Regarding the first goal of quantifying the Pu mass in spent fuel assemblies, researchers identified several NDA techniques that quantify various signatures from commercial spent fuel.[4] The approach for researching the capabilities of these techniques was shaped by two key factors: (1) None of the NDA techniques identified at that time was capable of determining elemental Pu mass as a standalone technique. And (2) more than one NDA systems will likely be needed to satisfy the range of motivations listed above; factors such as cost, accuracy, ability to work under water and portability will impact what system of techniques are best for a given motivation.

The 13 NDA techniques being researched are the following: Delayed Neutrons,[5] Differential Die-Away,[6] Differential Die-Away Self-Interrogation,[7] Lead Slowing Down Spectrometer,[8,9,10,11] Neutron Multiplicity,[12] Passive Neutron Albedo Reactivity,[13,14,15] Total Neutron (Gross Neutron),[16] X-Ray Fluorescence,[17,18, 19] ^{252}Cf Interrogation with Prompt Neutron Detection,[20] Delayed Gamma,[16] Nuclear Resonance Fluorescence,[21] Passive Prompt Gamma,[16] Self-integration Neutron Resonance Densitometry.[22,23]

MODELING APPROACH

The research plan to determine Pu mass in spent fuel is nominally a five year effort and is part of the Next Generation Safeguard Initiative.[24] The initial two years are focused primarily on Monte Carlo modeling while the later three years involve the fabrication of hardware and measuring spent fuel assemblies. The Monte Carlo effort has two main goals: (1) Quantify the expected capability of each technique as an independent instrument; the performance of each NDA technique will be documented in independent reports. (2) Determine how to integrate a few techniques together in order to determine elemental Pu mass. The result of the integration will be documented in one or more reports. In order to cost- effectively and robustly achieve these two goals, a library of assemblies was created.

In order to keep the assembly library manageable in size, one reactor type (pressurized water) was selected. The largest standard assembly size (17 by 17) was selected given that larger assemblies are generally more challenging than small ones. The differences among the assemblies emphasized isotopic, spatial and diversion variability as described in the next three paragraphs.

The isotopic variability among 64 assemblies was obtained by using the Monte Carlo N-Particle eXtended (MCNPX) transport code that recently had the CINDER burnup capability added.[25] Each assembly has a unique combination of burnup, enrichment, and cooling time. The burnup cases were 15, 30, 45 and 60 GWd/tU; the initial enrichments were 2, 3, 4, 5%; and the cooling times were 1, 5, 20, 80 years.

Since integration among techniques is an upfront requirement, it was necessary to assure that the assembly library contained technique-specific physical attributes. For example, the 100 keV photons measured with X-Ray Fluorescence come primarily from the outer 10% of the volume of each pin. Over this same dimension, the plutonium content can vary by over a factor of two. As such, it is necessary to burn the fuel with radial resolution in every pin. This was done by dividing the fuel into 4 radial cells with the greatest refinement in the exterior region of the pin in order to quantify the radial variation in Pu. Another example of including technique-specific properties in the spent fuel library involves the hydrogen sensitivity of the Lead Slowing Down Spectrometer. The level of hydrogen in the fuel cladding produces a noticeable effect; and so the variation in the hydrogen content in the cladding as a function of burnup was included.

Over 40 “diversion assemblies” were created from a few of the 64 “non-diversion” assemblies. The majority of the diversion assemblies involve replacing pins from the center, mid and outer regions with natural uranium pins. The details of the diversion assemblies are to be presented in a paper by Fensin et al.[26] The performance for most instruments will be quantified for the full 100+ assemblies in the library as if the measurements took place in three different media: air, water and borated water. In total, over 3,000 models will be run with the assemblies of the library.

MEASUREMENT APPROACH

The experimental phase of determining the Pu mass of spent fuel assemblies has two basic parts: (1) The measurement of spent fuel assemblies in an NDA system and (2) the determination of the Pu content in the spent fuel assembly by means other than NDA so that the accuracy of the NDA system can be quantified. The present declared Pu mass in "standard" commercial assemblies is generally thought to be accurate in the 5% to 10% range. The hope/expectation is that an NDA system will perform better than this. As such, the current target is to have a group of assemblies for which the Pu mass is known to better than 2%. The following list of five measurement scenarios was formulated to accommodate these needs:

- Measure assemblies for which the Pu mass is well known from past spent fuel characterization research. As part of other research programs, some assemblies have been studied in considerable detail. In some cases the goal of past research may have been burnup credit, in other it might have been to learn of reactor operation. This scenario is desirable since assemblies could be measured at relatively low cost given that the difficult task of characterizing the assemblies has already been done.

- Measure assemblies for which the Pu mass of an individual assembly is known by means of a very well benchmarked burnup codes. Through considerable effort, it is possible to make burnup codes that are accurate to better than 2% for some assemblies. Primarily this accuracy comes from iteration with reprocessing results and/or through diagnosing the core very well. This scenario is desirable since it would likely allow a significant number of assemblies to be measured at relatively low cost since the difficult task of creating the accurate burnup code has already been done.

- Measure all the assemblies that go into an input accountability tank (IAT) at an industrial reprocessing plant. This scenario would require the cooperation of one of the major reprocessing facilities. This scenario would require access to the IAT and leached hulls measurement data. Furthermore, consideration of the type of dissolver used (continuous or batch) would need to be accommodated.

- Measure assemblies from which a few pellets were removed for destructive analysis. The Pu mass of the entire assembly would be determined by extrapolation Pu mass of one pellet to the entire assembly using a burnup code. Preliminary research needs to be done to determine how accurately one can extrapolate from a single pellet to the entire assembly.

- Measure an assemblies, then reprocess the entire assembly in a small IAT. This is the ideal case. However, given there are few facilities capable of this research and given the large cost of such a research plan, the ability to perform such research is uncertain.

The five bullets listed above were formulated to address one of the two main technical priorities of this research that is the goal of determining the absolute accuracy of an NDA system for determining Pu mass in an assembly. The second technical goal is to determine the sensitivity of each NDA system to the removal of pins. It is expected that this second goal is complimentary with any of the primary goals. In fact, any "standard" assembly will suffice for the purpose of quantifying the sensitivity of any given technique to pin removal. Furthermore, if standard

assemblies were used, it would be possible to use one assembly as a standard and compare others assemblies to it so that the relative variation in Pu content among a group of assemblies could be estimated.

DELAYED NEUTRON DETECTION

One of the NDA techniques being investigated involves the detection of delayed neutrons. The basic concept for using delayed neutrons in the context of spent fuel is the following: (1) A neutron generator produces neutrons near the assembly – most likely from each side of the assembly; these neutrons induce fissions. (2) The neutron generator is turned off. (3) The total neutrons emitted are counted. The counted neutrons have two primary sources, delayed neutrons emitted from the fission fragments after induced fissions and spontaneous fission neutrons emitted primarily from ^{244}Cm.

A cross section of the geometry used to quantify the anticipated performance of a delayed neutron system is depicted in Fig. 1. The assembly is in the center with air, water or borated water within and around the assembly. The assembly and media are then surrounded by a void (white) region followed by a ring of material used for spectrum tailoring. The content of the spectrum tailoring material will be discussed later. The neutrons from the neutron generator were assumed to be released from the outermost of these concentric rings. A deuterium- tritium (DT) neutron generator was used in the research presented here. Exterior to the rings (white) is another void region. The outer most region is comprised of polyethylene wall in which fission chambers with a 43.5 cm long active region are located. The inside of the polyethylene walls was lined with cadmium. The fission chambers are coated with 93% enriched uranium. If the assembly was removed and a ^{252}Cf source placed in the center of the detector, the detection efficiency in air is 0.3%.

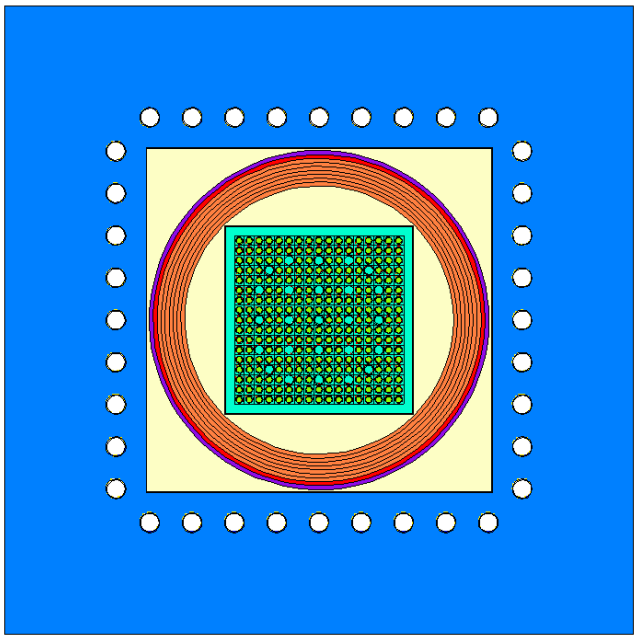

Fig. 1, Horizontal cross section of the delayed neutron detector – a description of the various materials is given in the text.

From discussions with subject matter experts, a source strength for the DT neutron generator of 1×10^{12} n/s was selected since this intensity is considered possible with present technology.[27] The

following timing was used for all calculations in this paper: (1) uniform irradiation for 0.9 s, (2) 0.1 s pause, (3) 1.0 s count time. For the data presented in this paper, this pattern was reproduced so that the total delayed neutron count time lasted 150 s. Hence, the irradiation time, pause time and count time was a total of 300 s.

In Fig. 2a, the delayed neutron signature is depicted for assemblies from the spent fuel library described earlier; sixteen cases were modeled: four burnup cases (15, 30, 45 and 60 GWd/tU) and four initial enrichments (2, 3, 4, 5%) for a cooling time of 5 years. The medium around the assembly is water. The uncertainty on each data points was very small and varies since the back ground signal, primarily from the spontaneous fission of Cm and subsequent multiplication, increases as the third or fourth power of the burnup. For the source strength of 1 x 10^{12} n/s, the statistical uncertainty in the delayed neutron count rate in all cases was less than 0.01%; in light of the inevitable systematic uncertainties, this uncertainty is much lower than will be obtainable. To make a preliminary estimate of the magnitude of the system effect of positioning the assembly in the detector, the delayed neutron count rate was measured for the assembly at various locations within the measurement chamber. There is a 0.5 cm gap on all sides of the assembly. In the case of water measurements, an uncertainty of ~1% was determined. The positioning uncertainty in air was not quantifiable given the uncertainties of the simulations. It is thought that the uncertainty in water can be reduced by staggering the fission chambers in the polyethylene, yet a systematic uncertainty of at least a fraction of 1% is anticipated.

For the low delayed neutron count rate case of 60 GWd/tU, 5% initial enrichments and 5 years cooling times, the delayed neutron signal was 85% of the total signal. Hence, the neutrons generator is much stronger than needed. If the neutron generator intensity were reduced by a factor of 10, the delayed neutron signal would be 36% of the total signal for the 60 GWd/tU case which is still stronger than is likely needed for typical count times.

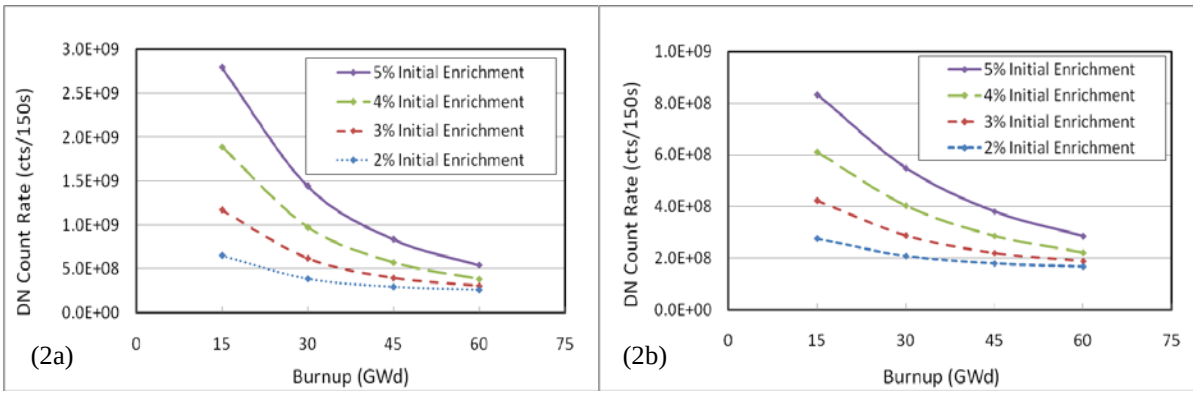

Fig. 2a, the delayed neutron counts in 150 s is illustrated as a function of burnup and initial enrichments for a cooling time of 5 years; the medium was water. Fig. 2b, the delayed neutron counts in 150 s is illustrated as a function of burnup and initial enrichments for a cooling time of 5 years; the medium was borated water.

In Fig 2b and 3, the delayed neutron signature is illustrated for the exact same assemblies as in Fig. 2a, however, in the case of Fig. 2b the medium about the assemblies was borated water (2200 ppm) while in Fig. 3 the medium was air. The main conclusions from the count rates determined for

water, borates water and air are the following: (1) A large dispersion in the delayed neutron count rate is obtained for both water and borated water since the neutron energy is nearly thermal. (2) The low dispersion obtained in air is due to the dominant delayed neutron signal from ^{238}U. When the neutron energy is above 1 MeV, ^{235}U and ^{238}U contribute nearly the same number of delayed neutrons per unit mass. Since the mass of ^{238}U is almost 100 times more than ^{235}U, the fission of ^{238}U dominates even though the cross section is low compared to the thermal fission of ^{235}U. (3) The source intensity of 1 x 10^{11} n/s will be strong enough in all cases. Given that the delayed neuron signal is stronger than the ^{244}Cm background for a very high burnup case, the intensity of the generator can be reduced from 1 x 10^{12} n/s. It may be possible to use a deuterium-deuterium (DD) generator which would reduce the low dispersion in the air case and eliminate the need for tritium handling.

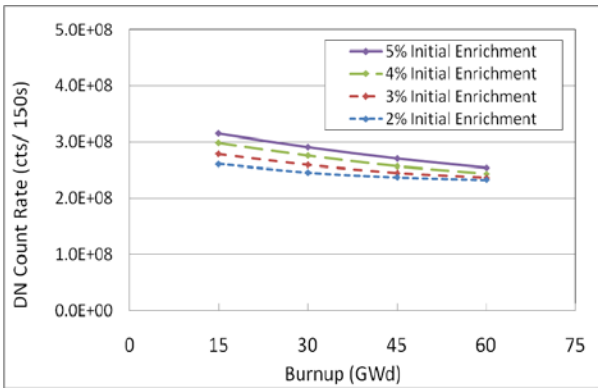

Fig. 3, the delayed neutron counts in 150 s is illustrated as a function of burnup and initial enrichments for a cooling time of 5 years; the medium was <u>air</u>.

The lack of dispersion in the case of measuring delayed neutrons in air will result in further research into spectrum tailoring. For the data depicted in Fig. 3, only tungsten and lead were used for spectrum tailoring to assess if (n, 2n) reactions would effectively lower the neutron energy as well as boosting the interrogating flux. As implemented, this concept was not effective. The next step is to utilize lower atomic number materials to reduce the neutron spectrum incident on the fuel to below the threshold of ^{238}U induced fission (~1 MeV) or to use a DD generator which produces 2 MeV neutrons instead of 14 MeV as with a DT generator.

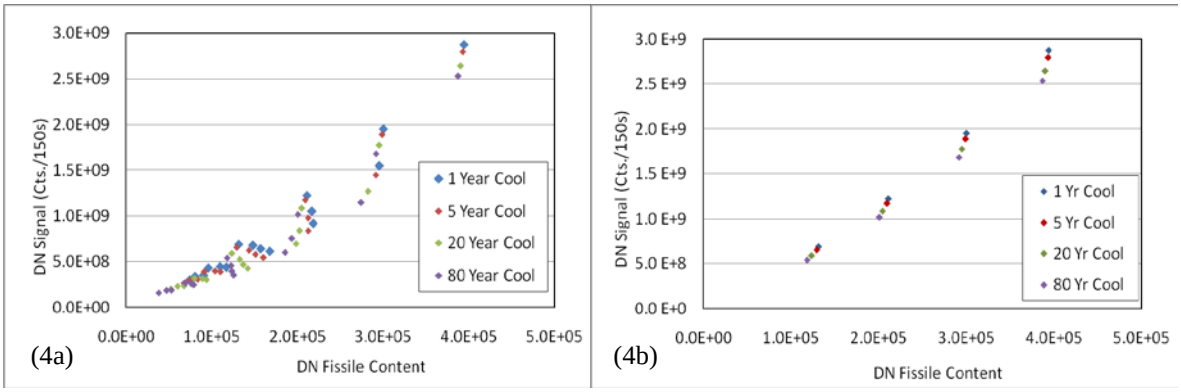

Fig. 4a, the delayed neutron counts in 150 s are illustrated as a function of fissile content and cooling time when all the assemblies in the spent fuel library were measured in <u>water</u>. Fig 4b only the delayed neutron count rates for a 15 GWd/tU case is illustrated.

The delayed neutron fissile content per assembly illustrated in Fig. 3b was determined by starting with the declared uranium and plutonium fissile mass and isotopic concentrations for ^{235}U, ^{239}Pu and ^{241}Pu. The declared mass of each fissile isotope was multiplied by the respective thermal cross sections (586 b, 748 b, 1,013 b, respectively) to approximate the fission rate of each isotope (assuming the same neutron flux to each isotope). In order to weight the fissile content by the number of neutrons produced per isotope, the normalized fission rate was multiplied by the number of neutrons produced per fission (2.41, 2.88, 2.80, respectively). Finally each fissile isotope was multiplied by the delayed neutron production fraction per fission (1.58%, 0.61% and 1.60%, respectively). This approximate fissile content is approximate with the major assumption being that all the reactions occur at thermal energies. Furthermore, the delayed neutron count rate is influenced by factors such as the build of poisons in the fuel with burnup. None the less, the primary signature of a delayed neutron instrument is illustrates. The delayed neutron signal varies for all the assemblies with fissile content. Note the error bars in the delayed neutron count rate is much less than a percent. Considerably more research is needed to determine the uncertainty in the fissile content.

It is instructive to segregate the data along the lines of what an inspector might do. For example, the burnup can be estimated from total neutrons or gamma line ratios. In Fig 4b, the data for the 15 GWd/tU case are graphed. Four clear data clumps are observed; these clumps progress from the highest to lowest delayed neutron count rate with initial enrichment (2%, 3%, 4% and 5%). With these clumps there is a clear trend with cooling time since ^{241}Pu has a 14 year half-life. In the context of IAEA inspections, the cooling time could be considered a known since inspectors are on site when a reactor begins operation and on site when fuel is unloaded. In Fig. 4b, 4 curves could be formed for each of the cooling time. The final step of quantifying Pu mass requires the determination of the relative amount of ^{235}U, ^{239}Pu to ^{241}Pu. Such information may be obtained from several of the other instruments in possible combination with Pu isotopic correlation with burnup; techniques that would integrate particularly well with a delayed neutron system are Delayed Gamma, Differential Die-Away, Self-integration Neutron Resonance Densitometry, ^{252}Cf Interrogation with Prompt Neutron Detection.

CONCLUSIONS

A nominally 5 year research effort has recently begun focused on quantifying (1) the elemental Pu mass in spent fuel assemblies with NDA and (2) the sensitivity of NDA techniques to detect missing pins. Nine motivations for undertaking this effort were described. Thirteen NDA techniques capable of quantify different signatures emitted by spent fuel were listed. A key aspect to the research approach being taken is that a few techniques will likely need to be integrated into one or two system; which techniques will be integrated together will depend strongly on which of the motivations is driving the effort. The first two years of the five year is primarily Monte Carlo based research to quantify how well each of the NDA techniques is expected to perform individually and in a system. At the end of two years, a system or two will be identified. In the later three years, instruments will be fabricated and spent fuel assemblies will be measured. Five scenarios for obtaining assemblies for the experimental phase of this research were described.

Preliminary results for the delayed neutron techniques were presented. The results indicated that the delayed neutron count rate can be quantified with excellent precision in water, air and borated water

but that air measurements would benefit considerably by interrogating with a lower neutron energy spectrum; spectrum tailoring or replacing the DT generator with a DD generator are two options. The variation in the delayed neutron count rate as a function of fissile content was illustrated as a function of initial enrichment, burnup, and cooling time. Speculations as to how a delayed neutron instrument might be used by a regulatory agency such as the IAEA were made. The need for input for other instruments is evident if the Pu mass is to be unfolding from the fissile.

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