REVIEW AND RANKING OF NDA TECHNIQUES TO DETERMINE PLUTONIUM CONTENT IN SPENT FUEL

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ABSTRACT
A number of efforts are under way to improve nondestructive assay (NDA) techniques for spent nuclear fuel (SNF) safeguard applications. These efforts have largely focused on advancing individual NDA approaches to assay plutonium content for the purpose of material accountability. Although significant improvements have been made in NDA techniques, relatively little work has been done to thoroughly and systematically compare the methods. While the content of SNF can be determined accurately with destructive assay, this technique is very costly and extremely impractical to perform at operating nuclear power facilities. NDA measurements have the potential to provide a quick alternative to destructive analysis when determining SNF isotope content for Material Control and Accountability (MC&A). To gauge the practicality and effectiveness of the various relevant NDA techniques, criteria and a ranking system have been developed. Each criterion has an associated numerical ranking to enable a quantifiable comparison of the NDA techniques relative strengths and weaknesses. The final results of this ranking process will be used to help guide the NDA research at the Oak Ridge National Laboratory.

INTRODUCTION
Fork detectors (Rinard and Bosler, 1988), the safeguards MOX python SMOPY (Lebrun, 2001), and a Cherenkov viewing device (Kuribara and Nemoto, 1994) constitute the NDA techniques that are currently used at operating nuclear power plants (NPPs). While these techniques can be used to confirm NPP burnup and assembly declarations, they do not have the capability to independently determine burnup, initial enrichment, and cooling time. These methods require assembly-specific calibration based on trusted assembly declarations for measurement interpretation. Because of these calibration requirements and the lack of independent verification, the accuracy of the NPP burnup declarations is paramount for accurate MC&A. While assembly burnup values provided by NPPs are typically accurate "to at least 5% of 'true' assembly burnup" in the United States (Bevard et al., 2009), burnup predictions have been in error by up to 16,000 MWD/MTU when batch average values for burnup were used (Bechtel SAIC Company, LLC, 2004). Large errors in the burnup declarations provided by the NPP – either unintentionally or intentionally – will significantly affect the accuracy of the MC&A calculations, illustrating the need for burnup measurements independent of the NPP records.

To address the needs for improved NDA methods, considerable resources are currently being invested to investigate a wide range of passive and active interrogation techniques (e.g., delayed gammas, delayed neutrons, differential die-away technique, lead slowing-down spectrometer, coincident neutron

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measurements, nuclear resonance fluorescence, passive gamma rays, passive neutron albedo reactivity, self-integration neutron resonance densitometry, total passive neutron counting (gross neutron), and X-ray fluorescence (Tobin et al., 2008). While each of these candidate techniques has potential advantages and disadvantages, to our knowledge a systematic comparison and evaluation of the relevant techniques to assess and rank their potential effectiveness and viability has not been performed. To separate out the relative strengths and weaknesses of the different NDA methods, a set of ranking criteria and a ranking system have been developed. The ranking criteria will gauge

1. obtrusiveness of the measurements to an NPP,
2. plutonium content verification within assay measurement range,
3. assay measurement range, defined as the distance between the detector face and the depth into the assay target where 90% of the detector response originates,
4. deployability of the technique (entails the movement of measurement equipment to a location),
5. sensitivity of the assay due to changes in material composition,
6. measurement time, and
7. NDA synergy (the effect of combining two different types of measurements to gain more information).

These criteria are intended to facilitate rankings of NDA techniques for different types of facilities, such as a recycling facility or an NPP, under different types of scenarios. Each criterion is ranked from (1) to (5) with (1) being the least attractive and (5) being the most attractive ranking. However, the importance of each criterion for choosing the most suitable technique will depend on the involved parties, the type of facility, and the specific scenario.

OBTRUSIVENESS OF THE MEASUREMENTS TO A FACILITY

The obtrusiveness of the spent fuel measurements to an NPP or other type of nuclear facitliy gages the incurred costs and risks the facility may experience from restricted operations as well as safety and liability concerns. More-obtrusive NDA techniques will be less appealing for those operating the nuclear facility, and more justification will be required for their use. In particular, any measurement that delays or impairs facility operations will cause significant loss of revenue.

In the case of an NPP, even if the measurements do not directly delay the refueling of the core during an outage, indirect factors can cause refueling delays. Facilities that have started a dry storage campaign typically have a spent fuel pool that is near or at the regulated capacity. Therefore, these nuclear power plants are susceptible to a shutdown if the NDA measurements delay a dry storage campaign.

If an NPP's operations are not impaired, then the time, cost, dose given to workers, risk of fuel movement accidents, etc., of the measurements will be the leading concern. For example, atypical fuel assembly movements will be more challenging for an NPP to implement. This includes moving dried spent fuel assemblies back into the spent fuel pool, operation and installation of new devices that have been approved by regulatory bodies but are new to the facility, disassembly of reactor assemblies to measure individual rods, etc. In these cases, unexpected delays and problems may occur that can increase the measurement times, increase the likelihood of assembly damage, and ultimately increase the likelihood of impacting normal facility operations.

NDA measurements that are performed within the spent fuel pool and require only traditional assembly movements are more likely to gain the acceptance of an NPP. Measurements that occur in the spent fuel pool minimize the chance that workers will receive any appreciable dose from the measurement campaign. There is also a precedence of assembly measurements in the spent fuel pool using the fork detector and SMOPY that could be extended to other measurements conducted "at the end of a pole."

Obtrusiveness of Measurement Criterion

(1) The facility is shut down because of the measurement requirements, and significant revenue is lost.
    a. Significant costs may be incurred due to retrofitting requirements for air or water.
    b. Measurements or retrofitting interferes with a planned or unplanned outage.
    c. Atypical operations are required with questions about safety, liability, and/or local regulations.
    d. Additional dose to workers may be incurred.
(2) The facility operates behind schedule as a result of the measurement campaign.
    a. Costs may be incurred due to retrofitting requirements.
    b. The dry storage campaign is delayed, potentially interfering with an outage or requiring significant overtime to proceed with work.
    c. Atypical operations are required with questions about safety, liability, and/or local regulations.
    d. Additional dose to workers may be incurred due to measurement procedures.
(3) The facility operates at normal capacity (potentially high cost).
    a. Atypical operations are required with questions about safety, liability, and local regulations.
    b. Additional dose to workers is likely due to decontamination or measurement configurations.
    c. The measurement campaign for a given number of assemblies is expected to account for a significant portion of the spent fuel pool operations.
    d. No significant delays that could affect facility operations are expected.
(4) The facility operates at normal capacity (potentially moderate cost).
    a. Personnel requirements at the facility are near normal operational requirements but may require more work from fuel pool operators.
    b. Only familiar facility procedures are required of workers.
    c. The measurement campaign for a given number of assemblies should be completed within a couple of months.
    d. No additional dose is expected beyond the normal expectations.
(5) The facility operates at normal capacity.
    a. No dose beyond normal operations is expected.
    b. A minimal number of personnel are required to aid the inspection, with no or few fuel assembly movements.
    c. The measurement campaign for a given number of assemblies should be completed in a few weeks.

PLUTONIUM CONTENT VERIFICATION WITHIN ASSAY RANGE

Plutonium verification is the ability to measure or infer plutonium content from an assay target within the measurement range of the technique. While significant correlation has been demonstrated between the $^{134}Cs/^{137}Cs$ ratio or ^{244}Cm neutron emissions with plutonium content, these measurements do not directly measure plutonium. Instead, plutonium content is inferred from established correlations and computer models.

A more direct measurement of plutonium is the fissile response of the assay target after interrogation by neutrons. The fissile response will imply the ^{239}Pu and ^{241}Pu combined isotope content if fissile atoms such as ^{235}U are known quantities in the spent fuel. An improvement to the total fissile response of the assay target would be the ability to measure a single plutonium isotope with great confidence. In both of the previous cases, the identification of plutonium within the spent fuel target is a strong indication that the target has not been tampered with. However, these measurements only imply the total plutonium content in the spent fuel and therefore, without additional information, would not be sufficient to determine diversions within a recycling facility.

If the mass of the plutonium in the SNF were known, then the differences between the input tank and the multiple outputs of a recycling facility could be measured to ensure a plutonium mass balance. This mass balance would make it a challenge to divert plutonium from the facility, regardless of the spent fuel isotopics.

Finally, measuring both the total mass of plutonium and its isotopes would distinguish between weapons-grade and reactor-grade plutonium.

Plutonium Content Verification Criterion

The criterion for plutonium content verification is satisfied by one or more of the following:

(1) Implied plutonium content by indirect sources (gamma ratios or gross neutron/gamma count).
(2) Combined fissile response of the SNF due to neutron interrogation. Plutonium fissile content is implied if the current enrichment of ^{235}U is known.
(3) One or two isotope measurements of plutonium and implied total mass.
(4) Total plutonium mass without isotopic knowledge.
(5) Total plutonium mass and relative isotopic concentrations.

THE ASSAY MEASUREMENT RANGE FOR PWR FUEL

Because of the high atomic number and density of SNF, gamma radiation is significantly attenuated in an assembly. Because of this, greater than 90% of the measured 661 keV gamma peak from ^{137}Cs comes from the outer three rows of a Westinghouse 15 × 15 assembly (Reilly et al., 1991). Neutron attenuation is a more complicated phenomenon as a result of both induced fissions in fissile atoms and neutron poisons that can change both the neutron population and spectrum. Neutron attenuation will also be significantly altered if the measurements occur in water instead of in air, particularly water with soluble boron present as is the case in PWR spent fuel pools. Reilly et al. note that the "neutron signal decreases by a factor of 10 in about 10 cm" of water (Reilly et al., 1991), making underwater neutron assay more challenging.

To compare the assay range of the different NDA techniques, rows of a fuel assembly are used. A row is defined as approximately the thickness of one fueled pin cell in an assembly. Therefore, a Westinghouse 17 × 17 assembly will be 17 rows thick, with each row containing 17 fuel rods. If the assay technique can assay nine rows, then an entire assembly can be measured using two appropriately placed detectors at the same time or via two separate measurements.

Assay Measurement Range Criterion for PWR Fuel

The assay measurement ranges are as follows: (1) one row or less, (2) three rows, (3) five rows, (4) seven rows, and (5) nine rows.

THE DEPLOYABILITY OF THE TECHNIQUE

Deployability rates the door-to-door shipping cost of the materials and installation cost required to conduct the NDA. This ranking criterion will not be applicable if the measurement equipment is a permanent part of a facility. While this approach is largely a bulk-shipping rate, there are meaningful secondary considerations. For example, International Atomic Energy Agency (IAEA) equipment may be subject to tampering and thus require constant monitoring or tamper-proof containers while in transit. Also, the shipping of radioactive material (should the need arise) requires a number of procedures to register the source and meet the transportation regulations that vary from country to country.

The transportation regulations examine both the surface radiation level of the parcel, the radiation at a specified distance from the parcel, and the total number of curies contained. Depending on the characteristics of the radioactivity, air transportation may be limited and special arrangements may have to be made. The possibility of transporting large radioactive sources could potentially be used as an argument, regardless of merit, to limit the deployment of a given NDA technique. Since the shipping of radioactive material depends on the transportation method, the countries being traversed, and the final destination, it is not a simple matter to estimate costs. Therefore, transportation cost estimation for radioactive material is outside the scope of this ranking.

The criterion price ranges listed below represents the total cost to transfer the necessary equipment for the NDA measurement per assembly assayed. Although it is beneficial to measure a larger number of assemblies for a given campaign, this approach will make the measurement campaign more obtrusive to an NPP. However, this criterion does not include the costs incurred by the facility in which the measurements take place. Because of time constraints, the measurement times may also limit the number of assemblies that can be measured in a campaign. To put this ranking into perspective, the budget for the IAEA safeguards program for the Democratic People's Republic of Korea was 2.2 million euros in 2008 (International Atomic Energy Agency, 2009). At $20,000 U.S. dollars per assembly, only 150 assemblies can be measured for that budget. Since some spent fuel pools can contain over 1000 assemblies, it may take multiple fiscal years to measure every assembly.

Deploy-ability Criterion

The cost associated with the deploy-ability criterion simply decreases in 5K increments for values between 5K and 25K: (1) >$20,000; (2) $20,000 < x < $15,000; (3) $15,000 < x < $10,000; (4) $10,000 < x < $5,000; and (5) < $5,000.

THE SENSITIVITY OF THE ASSAY

The term *sensitivity* encompasses the change of the assay signal strength due to a change in the material characteristic being perturbed. The larger the change in signal strength for a given material property change, the more sensitive and effective the assay is. For example, if the plutonium content is doubled and the resulting signal strength is also doubled, the technique would be ranked as a (3) for assaying changes in plutonium content.

While many characteristics can be explored, a list of the most important ones includes burnup, cooling time, initial fuel composition, fuel rod diversions, plutonium content, and stimulated signal response from active interrogation. A more sensitive technique will be able to discern the differences between small changes in the spent fuel content, e.g., individual rod diversion, while a less sensitive technique will require large changes to detect a statistically significant difference resulting from the change.

Sensitivity of Assay Criterion

The following ranks the correlation of change in property to change in signal strength:

(1) x% change in property results in 0.5*x% or less change in signal strength,
(2) x% change in property results in 0.75*x% change in signal strength,
(3) x% change in property results in 1.0*x% change in signal strength,
(4) x% change in property results in 1.25*x% change in signal strength, and
(5) x% change in property results in 1.5*x% or greater change in signal strength.

MEASUREMENTS TIME

The rate at which the NDA measurements can be conducted depends directly on the setup and measurement times for a spent fuel assembly. While measurement times are often stated to be the time a detector is actively counting a source, this ranking also includes the assembly movement time to the measurement location and back. In a spent fuel pool, it will take ~15 min to select an assembly, pick it up, move it to a new location, confirm the location, and then place the assembly in the new location. Most of the 15 min is spent identifying the spent fuel assembly and final destination, not the travel time within the spent fuel pool itself. Therefore, 20 to 30 min can be spent by simply moving the assembly into and out of the measurement system in addition to the actual detector measurement time.

Movement times can be dramatically increased if an assembly must be dry before it can be measured at an NPP. Drying an assembly from a spent fuel pool for a measurement will require an effort similar to that for a dry storage campaign. In a dry storage campaign, it typically takes 8 h to load 32 assemblies into the dry

storage cask. If fresh fuel could be placed into the cask, the number of assemblies that can be safely loaded decreases from 32. Additionally, sealing the airtight lid onto the cask takes 1–2 h; then, the cask decontamination takes another 2–4 h once it is removed from the spent fuel pool. Next, the cask must be vacuum dried, which will take ~24 h. A cask is vacuum dried because of the concern that pinhole fractures in the fuel rod may oxidize in air and further rupture the fuel. The cask is backfilled with a chemically inert gas such as helium to facilitate heat transfer away from the fuel assembly.

Once the fuel assemblies have been dried in the cask and surrounded by an inert gas, the actual NDA measurement can be conducted. The cask used for NDA measurements will probably need beam ports and other equipment built into it to avoid having to build facilities on site. Detector positioning and manipulation of the spent fuel would then occur within the modified dry storage cask. After the measurements have been made, the fuel will most likely have to be returned to the spent fuel pool. The process of flooding the dry storage cask containing hot spent fuel is an unpracticed, but planned for, technique. Because of concerns about steam pressure and the unfamiliarity with the approach, the flooding process may take more time than drying the assemblies.

If a spent fuel pool has 1000 assemblies to be assayed and each measurement (with averaged assembly movement times) requires 3 h, then it will take 375 eight-hour workdays to assay the entire pool. Therefore, NDA techniques with long measurement times could be time prohibitive to implement on more than a small number of assemblies. This clearly indicates that a staged approach of quick measurements followed by more-accurate NDA of suspicious assemblies has merit. The 10-min increments in the criterion listed below imply a change of 48 assembly measurements with associated SNF positioning per 8-h workday.

Measurement Time Criterion

The measurement and movement time periods (in minutes) per assembly are as follows: (1) >60, (2) $50 < x < 60$, (3) $40 < x < 50$, (4) $30 < x < 40$, and (5) <30.

NDA SYNERGY

NDA synergy examines how different measurements could potentially be combined to correctly identify plutonium content and isotopics within spent fuel and/or reduce or eliminate the need for a priori information about the SNF. Detailed knowledge of the initial fuel characteristics, operational history, and cooling time can be used to calculate a detailed isotopic composition for spent fuel. However, in the absence of the detailed history of an assembly, a fork detector measurement of gross gamma and neutron emissions cannot distinguish between assemblies with the same gross emissions but with different enrichments, operational history, and cooling times. Since plutonium production is largely dependent on initial enrichment and burnup, the fork detector alone cannot uniquely determine plutonium content in the absence of more information. However, the fork detector can be used to show how information from additional measurements can be combined to significantly improve burnup and cooling time estimations. While NDA Synergy does not yet have a numerical ranking criterion, an example of its use is being investigated.

To investigate this concept, a Westinghouse 17 × 17 assembly with 4% initial enrichment and 40 GWd/MTU burnup was defined as a reference assembly for examination. The total 40 GWd/MTU burnup is distributed equally over three cycles with 30 MW/MT power. After each burnup cycle is a cooling period of 60 days, 60 days, and 5 years for cycles 1, 2, and 3, respectively. Using 72,000 ORIGEN runs with three different initial enrichments (3, 4, and 5%) and randomly selected burnup and cooling times, we ask the following question: How many of these random solutions have the same measurable characteristics of the previously mentioned reference assembly? Solutions that are within a 5% relative error of the reference assembly are plotted as a point in a graph of cooling time vs burnup. Burnup in the following plots refers to the combined cycle (total) burnup. For example, burnups of 5, 17, and 18 GWd/MTU for cycles 1, 2, and 3, respectively, have a total burnup of 40 GWd/MTU. Another example is a burnup of 30, 2, and 8 GWd/MTU, which also gives a total burnup of 40 GWd/MTU. The total assembly cooling time is presented as a sum of the three reactor downtimes, similar to the way in which total burnup is calculated.

Figure 1 shows the total burnup and total cooling time for assemblies that are within 5% relative error of the reference assembly in terms of gross gamma production. Unfortunately, very little structure is associated with the solution space. Therefore, with the use of gross gamma counts alone, it is not possible to infer much about an assembly, unless additional information is available. Gross neutron production is compared with the reference assembly in Figure 2, which shows considerable structure associated with enrichment, burnup, and cooling time. For a given enrichment, a large clustering of solutions is noted around a burnup with a range of cooling times. However, without more-detailed information, it is still not possible to infer much about the reference assembly.

By combining the examination of gross neutron and gamma production as a fork detector would, a clearer picture of an assembly can be seen (Figure 3). While it is still impossible to distinctly identify the characteristics of the reference assembly by these two measurements alone, the 72,000 potential candidates have been narrowed to burnup ranges depending on an initial enrichment but unpredictable cooling times. This demonstrates that the current fork detector system (which uses gross gammas and neutrons) will have challenges inferring the burnup history and initial enrichment of an arbitrary assembly, even though the fork detector can be used to reject an NPP's incorrectly stated burnup history and initial enrichment.

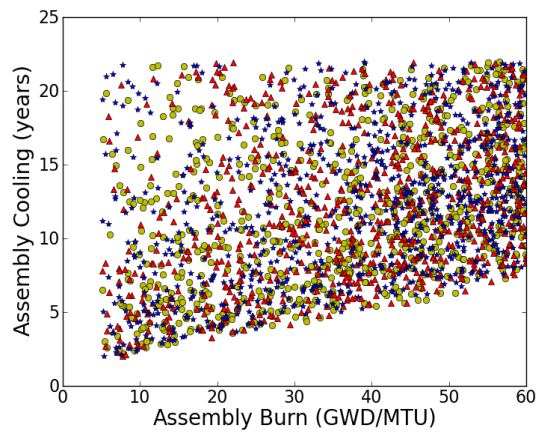

Figure 1: Possible solutions examining gross gamma production within 5% of the reference solution. Initial enrichments for the yellow circle, red triangle, and blue star symbols are 3, 4, and 5%, respectively.

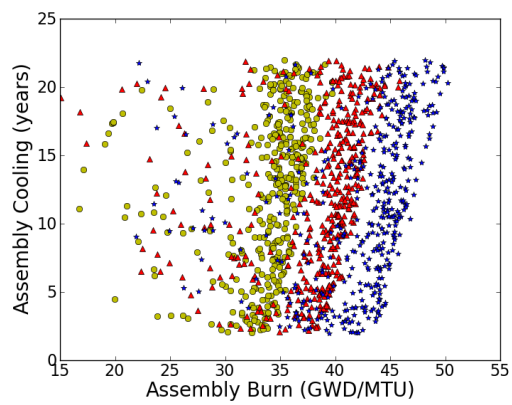

Figure 2: Possible solutions examining gross neutron production within 5% of the reference solution. Initial enrichments for the yellow circle, red triangle, and blue star symbols are 3, 4, and 5%, respectively.

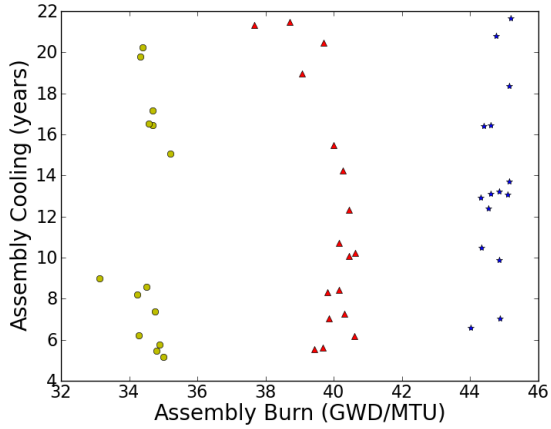

Figure 3: Possible solutions examining gross gamma and gross neutron production within 5% of the reference solution. Initial enrichments for the yellow circle, red triangle, and blue star symbols are 3, 4, and 5%, respectively.

CONCLUSIONS

The current NDA systems used at an NPP cannot be used to independently verify the initial enrichment, burnup, and cooling times of an assembly. Without this information, the plutonium content in an assembly cannot be accurately and/or reliably inferred. The next generation of NDA techniques is being designed to either directly measure the plutonium content or gain enough information to confidently infer the plutonium content in a given spent fuel assembly. To determine the relative merits of the different proposed techniques, a scenario-driven ranking system has been developed. This allows for the selection of the NDA technique or techniques that best fit the next generation of safeguards goals within the given scenario restrictions. Although this work is still ongoing, a preliminary example of the application of the ranking criterion can be seen in Appendix A of this paper.

APPENDIX A: Example of Ranking Criterion for a Measurement at a NPP in a Spent Fuel Pool

(Ranking is from (1) to (5) with (5) being the most desirable outcome.)

X-Ray Fluorescence (XRF) – Overall Ranking (TBD)

XRF is a passive self-interrogation technique that directly measures plutonium in spent nuclear fuel (SNF). The fuel is self-interrogated by the fission product radiation, exciting the K shell electrons in plutonium and uranium into emitting X-rays in the 100 keV energy range. Even though there is a large Compton background when measuring spent fuel, a small 103.7 keV plutonium X-ray peak can be detected using a coplanar high-purity germanium detector. With a known mass of uranium, the ratio of the plutonium and uranium peaks will give the mass of plutonium.

Obtrusiveness—(3 and 4)

Depending on the measurement implementation, the obtrusiveness of the measurement could change for an NPP. If the measurement uses an evacuated watertight pipe that starts outside of the spent fuel pool and ends at an underwater-spent fuel assembly, questions about shielding and radiation dose will need to be answered. If a specific watertight device is built to allow a germanium detector to take measurements in the spent fuel pool, the technique will rank higher.

Plutonium verification—(4)

This technique can directly measure plutonium in SNF.

Assay measurement volume—(1)
Because of the low energy of the fluoresced X-ray and the high photon attenuation of uranium, only the outer 200–300 microns of the fuel pin can actually be measured. Plutonium distribution in fuel pin models will have to be used to infer the total plutonium content within a single fuel pin.
Deployability—(4)
In addition to a coaxial germanium detector, the XRF technique requires collimation. This collimation in a spent fuel pool will come in one of two forms. The first is an evacuated watertight pipe roughly ~20 ft long with one end out of the spent fuel pool and the other near the surface of the assay target. The other form would be the coaxial high-purity germanium detector, most likely requiring a soundproof casing, with a pinhole collimator for use underwater.
Sensitivity of the assay
Burnup sensitivity—(3)
XRF appears to have a strong linear relationship with the measured plutonium and uranium ratios in low–enriched-uranium pressurized-water-reactor fuel.
Diversions of fuel rods—(1)
Because of the small assay volume of XRF, interior fuel rod diversions will not be detectable in an assembly.
Cooling time—(?)
Because the fuel is self-interrogating, there is a complicated relationship between cooling time and the induced fluorescence that has not yet been fully determined.
Measurement times—(1)
Measurement times of over an hour have been reported for accurate measurement of the plutonium peak. It is likely that this measurement time could be reduced if the Compton scattering is reduced in the measurement system.
NDA synergy—(TBD)
While X-ray fluorescence directly measures plutonium content, it can do so only in a 200–300 micron range. This short range may be sufficient if the SNF is measured in small quantities or homogenous mixtures, but XRF would not be sufficient to assay plutonium content in an entire assembly. Using a neutron technique that can assay an entire assembly via XRF measurements on the outside of the assembly as a boundary condition could prove to be a strong combination.

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