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Abstract

Active interrogation, a measurement technique which uses a radiation source to probe materials and generate unique signatures useful for characterizing those materials, is a powerful tool for assaying special nuclear material. The most commonly used technique for performing active interrogation is to use an electronic neutron generator as the probe radiation source. Exploiting the unique operating characteristics of these devices, including their monoenergetic neutron emissions and their ability to operate in pulsed modes, presents a number of options for performing prompt and delayed signature analyses using both photon and neutron sensors. A review of literature in this area shows multiple applications of the active neutron interrogation technique for performing nuclear nonproliferation measurements. Some examples include measuring the plutonium content of spent fuel, assaying plutonium residue in spent fuel hull claddings, assaying plutonium in aqueous fuel reprocessing process streams, and assaying nuclear fuel reprocessing facility waste streams to detect and quantify fissile material. This paper discusses the historical use of this technique and examines its context within the scope and challenges of next-generation nuclear fuel cycles and advanced concept nuclear fuel cycle facilities.

Keywords: Electronic Neutron Generator, Safeguards, Reprocessing

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Introduction

For over 15 years Idaho National Laboratory (INL) has supported research and development programs focusing on active interrogation to assess objects and containers and identify the presence of certain materials including chemical warfare agents, special nuclear material, and high explosives.[1 -5] This work has used both neutron and photon (bremsstrahlung) sources and has ranged from small portable systems to large fixed installations. Nuclear instrumentation used in these programs has included traditional radiation measurement equipment including organic and inorganic scintillators, gas filled detectors, and solid-state detectors as well as more advanced equipment and techniques including neutron imaging systems, high-speed waveform digitization and signal processing, and multiplicity analysis. With the intention of leveraging this knowledge, research at INL is now underway to explore opportunities where active interrogation might be of use in support of research initiatives into next-generation nuclear fuel cycles and advanced concept nuclear fuel cycle facilities, including facilities for proliferation-resistant nuclear fuel reprocessing.

These initiatives involve exploratory research related to nuclear power in the areas of advanced nuclear fuel and involve the development of research demonstration facilities related to advanced nuclear fuel cycles. These facilities include an advanced reactor, a fuel treatment facility, and a fuel cycle research and development facility. Critical to the success of this program will be the ability to demonstrate that effective and affordable solutions can be found to meet the nuclear nonproliferation and nuclear safeguards monitoring instrumentation requirements. Challenges in this area will be more difficult than previously encountered in fuel reprocessing facilities built around the world because the advanced fuels to be used in these initiatives will be hybridized mixtures of not only uranium and plutonium but also other transuranic elements including americium, neptunium, and curium. The presence of these extra materials adds complexity to the safeguards problem because to varying degrees these materials possess inherent nuclear attributes similar to plutonium including undergoing spontaneous fission, their susceptibility to undergo fission following neutron absorption, the emission of high energy alpha particles (which can generate neutrons through (α,n) reactions), and the emission of both low and higher energy gamma rays. Historically, many of the tools used to safeguard fuel reprocessing facilities have relied on measuring one or more of these properties to detect and quantify the presence of plutonium in samples. The presence of interferences that also contribute to these signatures means that many of the historical tools and techniques used in nuclear safeguards must be modified or redesigned to be able to provide plutonium specificity in the presence of Am, Np, and Cm. Active interrogation

techniques, which use external radiation sources to excite nuclear processes in materials, may help in addressing these challenges because inherently weak signatures are amplified under active interrogation and, by using pulsed radiation fields, active interrogation can investigate time-correlated signatures which passive signature techniques generally cannot.

In addition to nuclear safeguards a second important challenge in the next generation fuel cycle facilities will be to extend current industrial process monitoring instrumentation and controls technologies for use in high radiation and restricted access environments. In many cases there is a significant overlap between measurements useful for nuclear safeguards and measurements useful for monitoring the processes in nuclear fuel reprocessing; combining redundant plant measurements useful for both safeguards and process monitoring purposes may result in significant savings in labor and hardware costs. Also, by integrating process measurements into the safeguards process following the 'safeguards by design' philosophy a more integrated and more complete assessment of the compliance of a facility with its safeguard requirements can be ascertained.[6]

Today's industrial nuclear fuel reprocessing facilities at Sellafield, UK, Cadarache, France, and Rokkasho, Japan are large and complicated installations; next generation fuel reprocessing facilities may also be similar in size and complexity to these installations. In order to efficiently operate these plants a large quantity of information is needed to control different process streams and to monitor the performance of processes to make sure they are performing in accordance with their design specifications. Monitoring these processes can be both labor and instrumentation intensive; however, active interrogation can also play a role in support of process monitoring in future facilities. Because active interrogation measurements can be performed without contacting material process streams active interrogation is an ideal solution for performing on-line monitoring in areas where the process streams are hazardous or difficult to contain, and where the consequences of spills and contamination are high. Also, since active interrogation measurements can be made without contacting process streams directly, instrumentation maintenance and repair can be performed without the need to break into these process streams or impact plant operations. Finally, since active interrogation measurements and techniques are inherently designed for operating in high radiation fields they are particularly well-suited for implementation in nuclear fuel reprocessing environments.

Active Interrogation in Nuclear Process Monitoring and Safeguards

Active interrogation as a tool for nuclear safeguards and process monitoring in the nuclear fuel cycle has been the subject of many research projects and has been explored in nearly every aspect of the reprocessing cycle.[7] A simple flow sheet showing the major steps involved in spent nuclear fuel reprocessing is shown in Figure 1 together with examples of where active interrogation has been used for process monitoring and for safeguards measurements. However, active interrogation monitoring techniques have not seen widespread application in today's fuel reprocessing facilities, most likely because alternate technologies have been found to achieve the same goals and because active techniques are typically more complicated and more expensive than comparable passive techniques. In order to meet the demanding needs of next generation reprocessing facilities though active interrogation techniques may be the only means of providing process monitoring and safeguards answers in some cases. The following paragraphs provide some historical examples of the use of active interrogation in spent nuclear fuel reprocessing.

Irradiated Fuel Assay – A critical safeguards variable that must be known in a fuel reprocessing facility is the quantity of plutonium entering the facility in the incoming spent nuclear fuel. This is a difficult parameter to measure and in many instances it is determined analytically based upon numerical estimates of fuel burn-up from the originating facility where the fuel was used. In contrast to these numerical estimates active interrogation has been explored as a means of directly measuring the special nuclear material content of spent nuclear fuel.[8,9,10,11,12] One particularly promising active interrogation technique for assaying spent nuclear fuel is the lead slowing-down spectrometry (LSDS) technique. LSDS takes advantage of the fact that there is a difference in the fission cross-sections of ^{235}U and ^{239}Pu at 0.3 eV due to a plutonium neutron fission resonance. The LSDS technique uses a large assembly of lead to slowly moderate a monoenergetic neutron ensemble, generated using an accelerator (either a neutron generator or an electron accelerator that produces neutrons via photonuclear reactions), from fast to thermal energies. Neutron detectors embedded within the lead assembly are used measure the neutron flux inside the assembly as the ensemble passes through the 0.3 eV range. Quantifying this value and then normalizing it to measurements taken when the neutron ensemble reaches the thermal neutron energy range (0.025 eV) can then be used to infer the uranium and plutonium contained within the assembly. A measurement precision of better then 5% has been reported with this technique for measurements of 30 minutes duration.

Leached Cladding Hulls Analysis – An early step in fuel reprocessing is the mechanical separation (shearing) of the incoming fuel assemblies and the dissolution of the spent fuel contained within the metal fuel assembly rod pieces. The shearing process is typically done with a large mechanical chopper which cuts the fuel assembly rods into short lengths of a few inches or less. The dissolution process is typically carried out using strong nitric acid (or electrochemically). When the process is complete the dissolved spent fuel/nitric acid solution proceeds to the radiochemical separations process. The metal fuel rod casings, or hulls, which do not dissolve in this acid are then dried and melted and cast into waste ingots for permanent disposal. However, there is a possibility that not all of the spent fuel has been removed from these hulls and safeguards measurement must be made to quantify the residual plutonium still in the hulls; since the hulls themselves are highly radioactive after their residence in a nuclear reactor this is a difficult measurement. Typically spent fuel hulls are assayed by small-lot grab sampling of the inventory of chopped hulls. In contrast to this approach, active interrogation techniques have been developed to perform complete assays of the entire inventory of leeched hulls.[13,14,15,16,17] Results at the THORP plant in the UK have demonstrated this technique to be capable of achieving good measurement precisions over a range of from 20 – 350 g of residual fissile material quantities in leached hull.

Process Monitoring – In addition to monitoring and assessing plutonium and fissile material content entering and exiting a reprocessing facility there is also utility in measuring a) the fissile material content in process streams within a reprocessing plant and b) the chemistry and material content in support process streams within a reprocessing plant. Process stream information can be used to identify if non-standard operating conditions exist within a plant that may be indicative of proliferation activities. In addition, process monitoring can also be used to control in-plant activities to increase plant efficiency, increase material throughput, and reduce operating costs. A few trial experiments have been reported of using active interrogation for process monitoring.[18,19,20] Typically these instruments use neutron sources as the probe radiation and then, for fissile material analysis, use delayed neutron counting to infer fissile material masses. These instruments have been shown to be capable of measuring uranium or plutonium concentrations of ~10 grams per Liter in just a few minutes with measurement uncertainties of ~10%; they have also been used to measure lower fissile concentrations with a similar precision but with longer counting times.

Active Interrogation at INL

As a first step towards developing a safeguards focused active interrogation research capability at INL, scoping experiments were performed in December of 2007 to investigate a technique for receipt inspections to confirm the presence of SNM in a Department of Transportation (DOT) 6M drum.[a] These experiments served a dual role within INL's active interrogation program and were also used to examine interrogation techniques useful for shielded SNM detection. For these experiments three shipping drums were used; one contained 30 kg of depleted uranium (DU); one contained 4 kg of highly enriched uranium (HEU) (oxide fuel rodlets, 40% enriched ^{235}U), and the third contained 8 kg of the same HEU material.[b]

The detectors used in these experiments included a 2 m^2 array of helium-3 filled proportional counters. The proportional counters were located within polyethylene moderators that were encased within cadmium and boron to shield the gas tubes against thermal neutrons generated outside of the detector assemblies. The detector arrays were assembled in two sections and placed on opposite sides of the 6M drums. The active interrogation source for these experiments was a Thermo Electron MP320 electronic neutron generator (ENG) that produced a monoenergetic neutron spectrum of 2.5 MeV neutrons with an intensity of roughly 2 x 10^6 neutrons per second. The 6M drums were positioned with their axis of symmetry parallel to the ground. The neutron generator was roughly centered on the top of the drums for active interrogation; the generator was pulsed at a rate of 300 Hz with a pulse width of 166 microseconds. Measurements were taken for a duration of approximately 40 minutes, measuring the die-away neutron intensity in between neutron pulses, presented in Figure 2. The two top plots in this figure show the standard die-away neutron signature following the DD ENG pulse (left) and a cumulative counts plot starting at 350 microseconds after the NG pulse start time (right). Below these plots are corresponding plots showing the measured signature of counting pairs-of-counts in the He-3 array when pairs occur less then 650 microseconds apart. The signal from 0 – 350 microseconds is essentially a result of fast neutrons from the neutron generator slowing down within the detector's moderating structure and then being counted by the He-3 tubes. Neutrons from 350 – 2000 microseconds are a combination of fission neutrons from the test object still being generated as neutrons are undergoing moderation in the assembly, and delayed neutrons from the fission product beta-delayed neutron emitters (the die-away region). At times greater then 2000 microseconds the neutron signature is dominated by the

[a] These experiments were performed with researchers from Oak Ridge National Laboratory ORNL). INL collaborators included B. Blackburn, S. Watson, J. Johnson, E. Seabury, D. Norman, K. Haskell, B. Bennett, and B. Brush. ORNL collaborators included P. Hausladen, J. Mihalczo, and S. McConchie.
[b] These fuel rodlets were from INL's zero power physics reactor (ZPPR).

beta delayed neutron emitters fission products and the detectors inherent background count rate (the delayed neutron region).

Reviewing these plots there is a clear signature that is observable in the die-away region from both DU and HEU that is above the background signature. In the delayed neutron region the HEU signature presents a significant change over the background signature but it is not nearly as large as that seen during the die-away region. Taking advantage of the inherent background rejection achieved through the use of paired pulse counting, as seen in the lower plots of Figure 2, a further improvement in signal-to-noise is seen in the die-away of this data versus the single pulse counts. Placing a signal requirement of $+3\sigma$ over the background to determine the lower detection limit (LDL), the single pulse counting technique would have a LDL of 564 g DU and 20.5 g HEU (40% enriched ^{235}U). The paired pulse counting technique would have a LDL of 401 g DU and 4.2 g HEU (40% enriched ^{235}U). These values could be further improved through the use of a more intense ENG, or perhaps a deuterium-tritium ENG producing 14.1 MeV neutrons, or by counting for a longer period. Also, a custom designed detector system and improved irradiation geometry would serve to improve the detection efficiency of these techniques.

Summary

There are many cases where active interrogation may be used to help answer material control and accountability questions within advanced nuclear fuel cycle facilities. At INL work is proceeding towards applying active neutron interrogation technology to find measurement solutions for multiple questions related to advanced fuel cycle facilities.[21] In particular, work is investigating solutions related to spent fuel receipt and inventory, cold and hot stream process monitoring, and waste form fissile material assessment. These investigations will be carried out in consultation with experts in aqueous and electrochemical reprocessing in order to identify unique situations associated with advanced fuel cycle facilities (which will incorporate transuranic elements in the final fuel forms) that have not been previously encountered in other aqueous reprocessing facilities associated with uranium fuel remanufacturing or mixed-oxide fuel manufacturing.

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Figure 1

Major steps in the fuel recycling process and areas where active interrogation has been used for process monitoring and safeguards measurements.

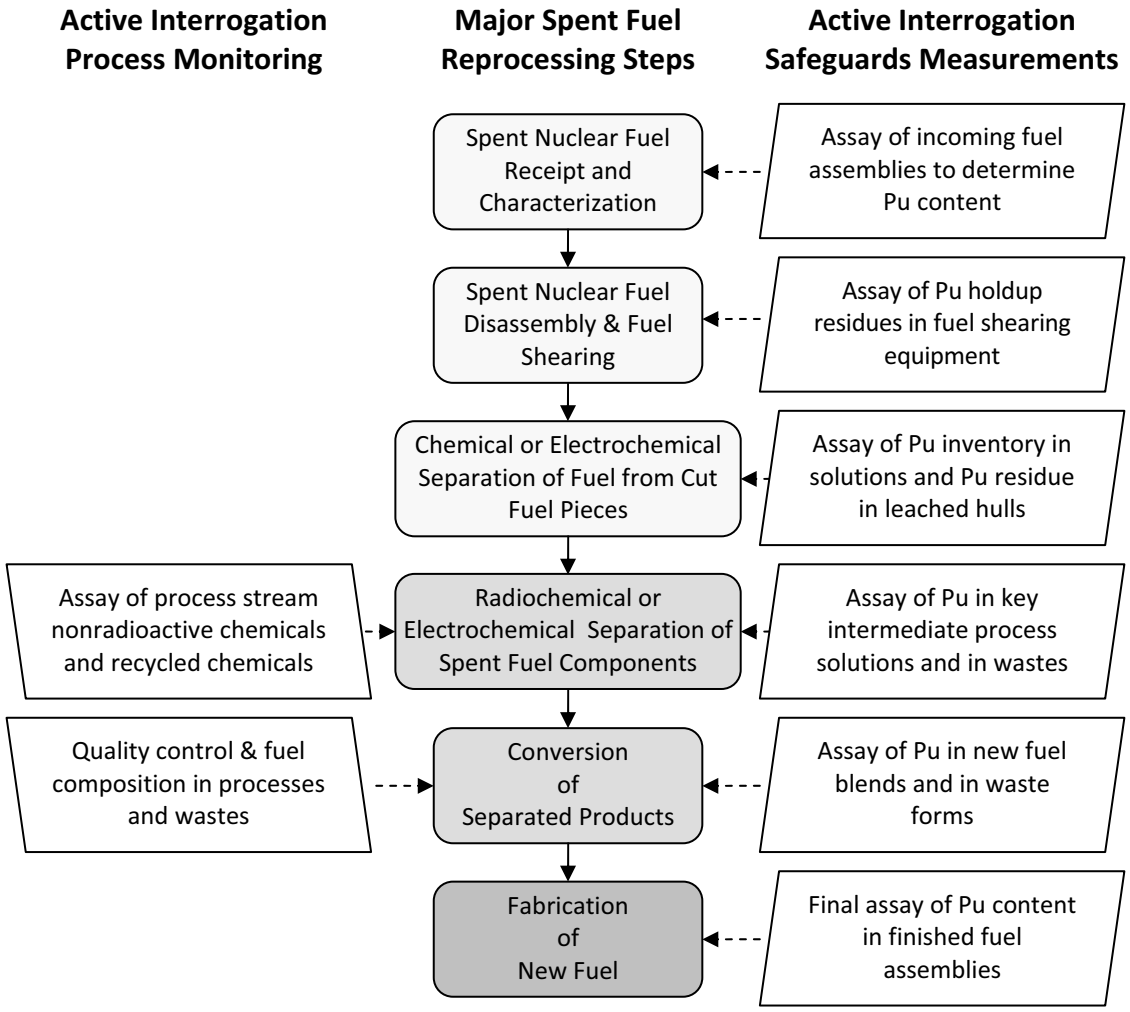

Figure 2 Die-away time spectra and cumulative event spectra (beginning 350 microseconds after the start of the neutron pulses) from the He-3 array for both single count events and paired counting events (events separated by less than 650 microseconds) measuring the active background, the 6M Drum signature containing 30 kg DU, and the 6M Drum signature while containing 8 kg of HEU.