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INL Active Interrogation Testing in Support of the GNEP Safeguards Campaign

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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. Work at Idaho National Laboratory (INL) in the area of active interrogation, using neutron and photon sources, has been under way for many years to develop methods for detecting and quantifying nuclear material for national and homeland security research areas. This research knowledge base is now being extended to address nuclear safeguards and process monitoring issues related to the Global Nuclear Energy Partnership (GNEP). As a first step in this area preliminary scoping studies have been performed to investigate the usefulness of using active neutron interrogation, with a low-power electronic neutron generator, to assay Department of Transportation 6M shipping drums containing uranium oxide fuel rodlets from INL's zero power physics reactor. Using the paired-counting technique during the die-away time period of interrogation, a lower detection limit of approximately 4.2 grams of enriched uranium (40% ^{235}U) was calculated for a 40 minute measurement using a field portable 2.5 MeV neutron source and an array of 16 moderated helium-3 neutron tubes. Future work in this area, including the use of a more powerful neutron source and a better tailored detector array, would likely improve this limit to a much lower level. Further development work at INL will explore the applicability of active interrogation in association with the nuclear safeguards and process monitoring needs of the advanced GNEP facilities under consideration. This work, which will include both analyses and field demonstrations, will be performed in collaboration with colleagues at INL and elsewhere that have expertise in nuclear fuel reprocessing as well as active interrogation and its use for nuclear material analyses.

Introduction

For over 15 years Idaho National Laboratory 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.[1,2,3,4,5] This work has used both neutron and photon (bremsstrahlung) radiation 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 the development of customized detectors and their associated electronics. With the hope of leveraging this knowledge base a work package was recently created within the Global Nuclear Energy Partnership (GNEP) Safeguards

Campaign to identify and explore opportunities where active interrogation might be of use in support of the GNEP program.

Active Interrogation and GNEP

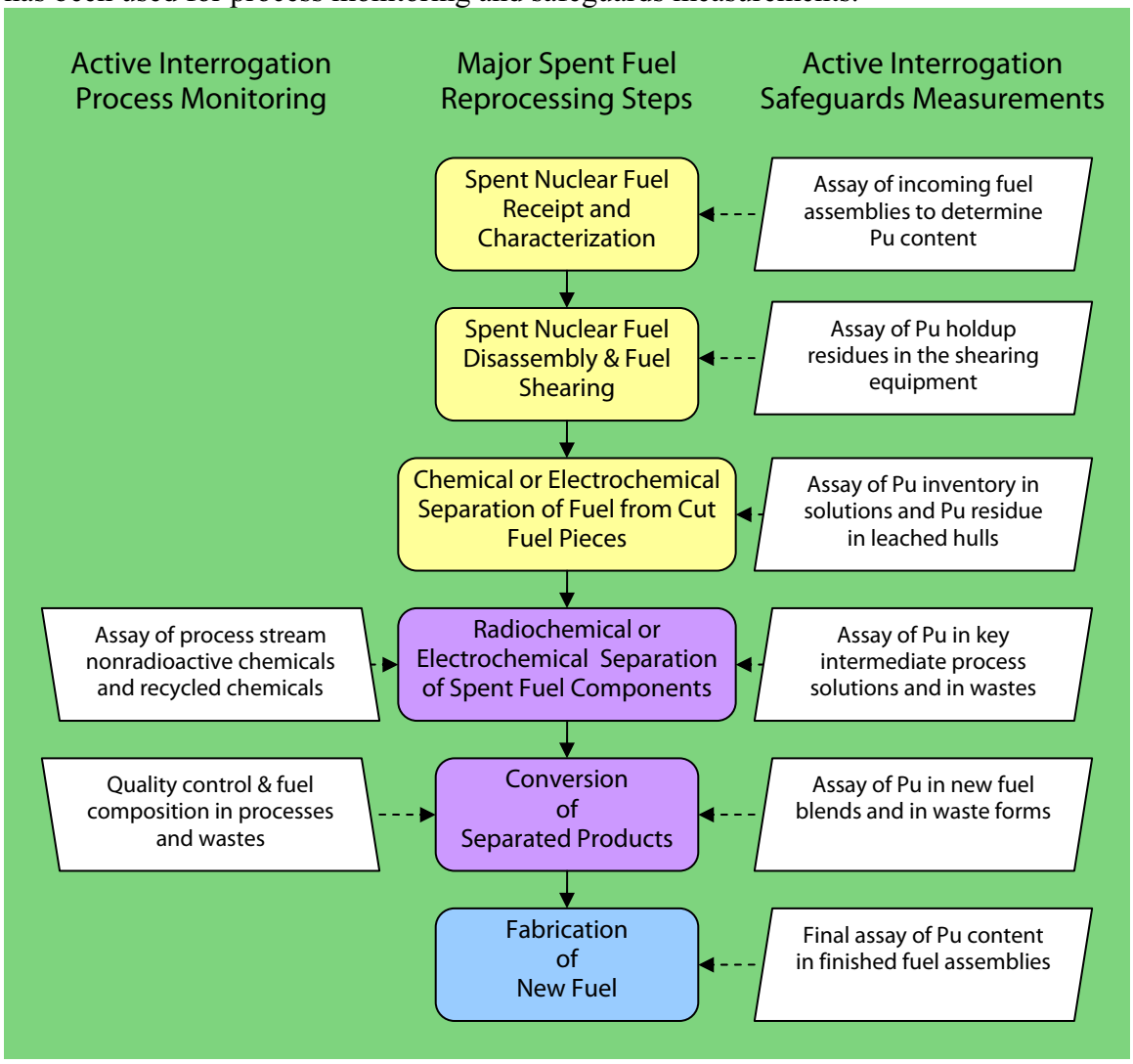
The GNEP program primarily involves the development of several large-scale research and demonstration facilities related to advanced nuclear fuel cycles. These facilities include an advanced reactor, a fuel treatment facility, and a fuel cycle research and development facility. Critical to the success 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 of the GNEP fuel reprocessing/recycling facilities. Challenges in this area will be more difficult than previously encountered in fuel reprocessing facilities around the world because the advanced fuels to be used in the GNEP fuel cycle 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 in the GNEP fuel cycle 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 interfering sources 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 GNEP 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 in the GNEP facilities. 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 it’s safeguard requirements can be ascertained.

Today’s industrial nuclear fuel reprocessing facilities at Sellafield, UK, Cadarache, France, and Rokkasho, Japan are large and complicated installations; the next generation GNEP 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 the future GNEP 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 preformed 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.

Figure 1 Major steps in the fuel recycling process and areas where active interrogation has been used for process monitoring and safeguards measurements.



Active Interrogation in Nuclear Process Monitoring and Safeguards

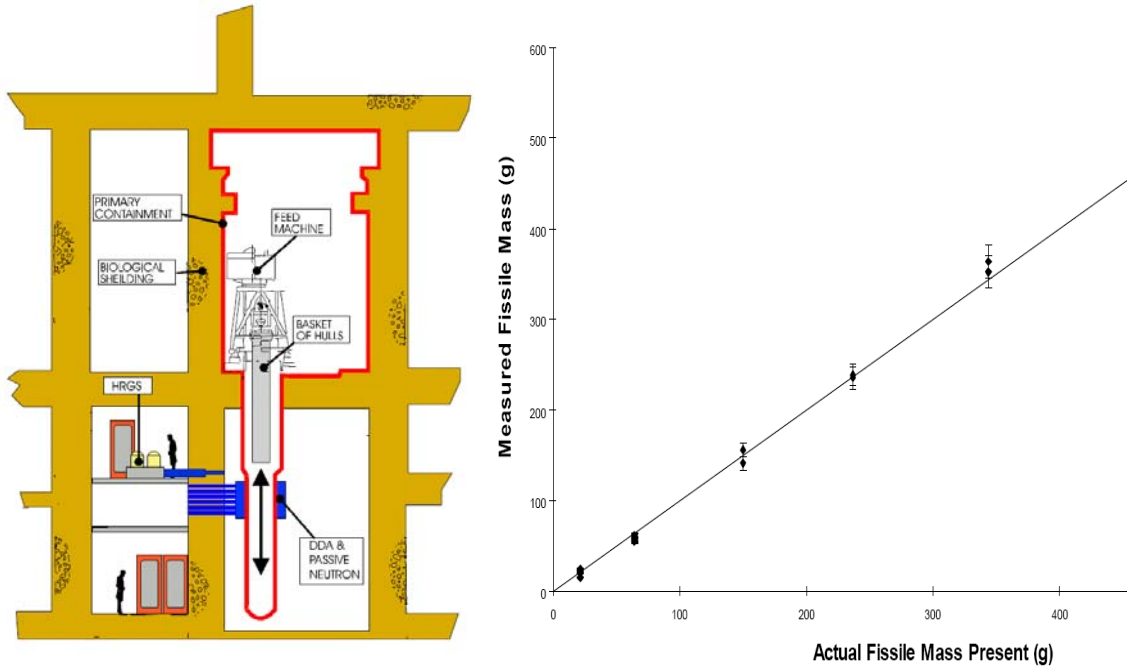
Active interrogation as a tool for nuclear safeguards and process monitoring in the nuclear fuel cycle has been the subject of many research projects and has been explored in nearly every aspect of the reprocessing cycle. A simple flow sheet showing the major steps involved in spent nuclear fuel reprocessing 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 the next generation GNEP 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.[6 - 10] 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 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.[11 - 14] Results at the THORP plant in the UK (Figure 2) have demonstrated this technique to be capable of achieving good measurement precisions over a range residual fissile material quantities in leached hull. Also evident in this figure is that a large, well-shielded facility is needed to safely handle and process these materials.

Figure 2 Scale drawing of a BNFL Instruments Ltd. leached hull analyzer installed at the THORP reprocessing plant in Sellafield, UK (left), and a plot showing the instrument's measurement linearity and precision for quantifying residual fissile content remaining in the hulls (right).[14]



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.[15 - 17] 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 Safeguards Scoping Experiments at INL

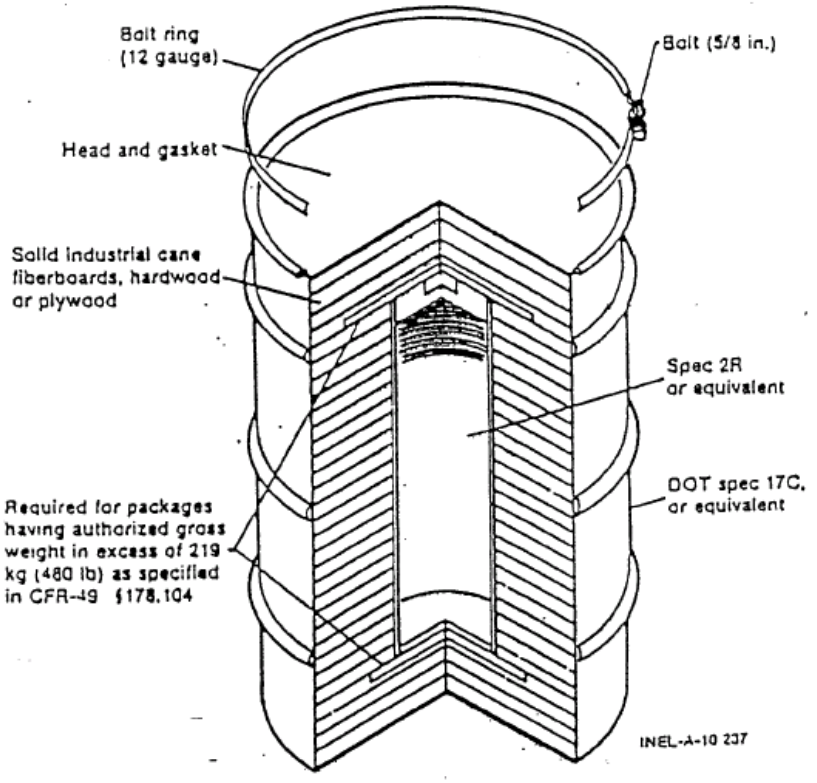
Idaho National Laboratory is ideally suited for investigating and demonstrating advanced safeguards instrumentation using active interrogation techniques. In addition to existing national and homeland security active interrogation programs mentioned in the introduction, and their related test, evaluation and support facilities, the Laboratory also hosts a complete set of fuel reprocessing and fabrication facilities within the Materials and Fuels Complex (MFC) (Figure 3). Included in this Complex are hot cells at the **Hot Fuel Examination Facility (HFEF)** suited for *spent nuclear fuel receipt and characterization* and *spent nuclear fuel disassembly and fuel shearing*, hot cells in the **Fuel Conditioning Facility (FCF)** suited for *chemical or electrochemical separations of fuel from cut fuel pieces, radiochemical or electrochemical separation of spent fuel components*, and *conversion of separated products,* and glove boxes at the **Fuel Manufacturing Facility (FMF)** suited for *fabrication of new fuel*. Additionally, the Complex also supports the **Special Nuclear Material (SNM) Storage Area** and the **Transient Reactor Test Facility (TREAT)**, places where general purpose safeguards experiments may be performed using uranium and plutonium.

Figure 3 The INL Materials and Fuels Complex (MFC)

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.[1] 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 (see Figure 4).[2]

Figure 4 The DOT 6M drum used for these experiments and a photo of the HEU fuel rodlets.[18]



HEU fuel rodlets prior to loading into the DOT 6M drum.

The detectors used in these experiments included a 0.5 m^2 liquid scintillator array and a 2 m^2 array of helium-3 filled proportional counters (Figure 5). 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, as seen in the pictures. The active interrogation source for these experiments was a Thermo Electron MP320 electronic neutron generator (ENG) using deuterium which produced a monoenergetic neutron spectrum of 2.5 MeV neutrons with an intensity of roughly 1 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. For tests with the liquid

[1] 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.
[2] These fuel rodlets were from INL's zero power physics reactor (ZPPR).

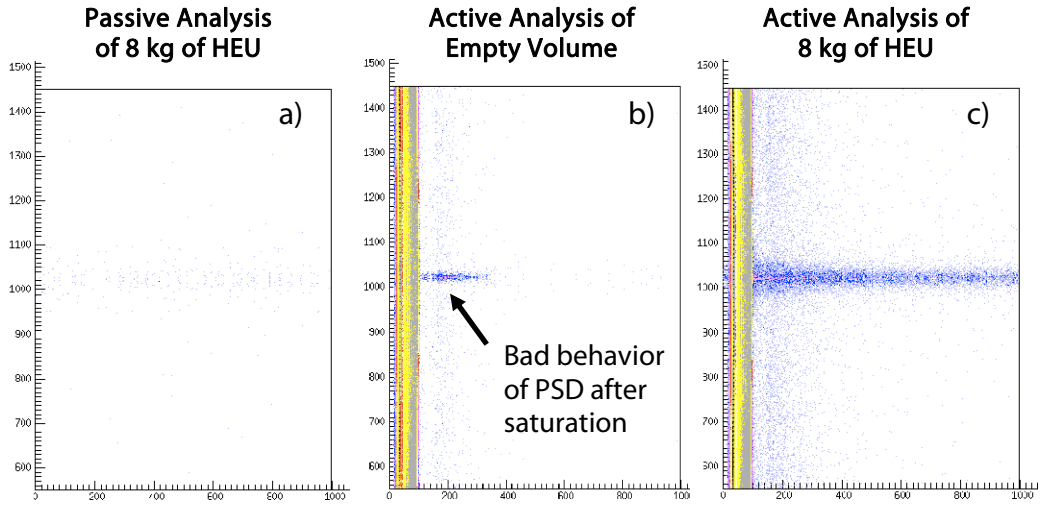
scintillator array the generator was pulsed at a rate of 10,000 Hz with a pulse width of 10 microseconds; with the He-3 array the generator was pulsed at a rate of 300 Hz with a pulse width of 166 microseconds.

Figure 5 The liquid scintillator array (top picture) and helium-3 array (bottom picture) used for interrogation of the 6M drums. The neutron generator can be seen in the bottom picture.

Data was collected and pre-processed for these experiments using ORNL's Nuclear Material Identification System (NMIS) electronics hardware, evaluating single counts in each detector, time-correlated pairs of counts between multiple detectors, and neutron multiplicity.[19] Measurement were taken for durations of 15 to 30 minutes. From the liquid scintillator array pair-counting data plots are shown in Figure 6. In these plots the abscissa values correspond to the time the first neutron event is detected, following a neutron generator pulse, in any particular detector; the time scale for these measurements is 0.1 microseconds. The ordinate values in these plots correspond to the relative times or arrival of the first and second neutron pulses in a pair, in units of 0.1 microseconds, where the time value of 1028 corresponds to a zero difference (i.e., two pulses are counted in separate detectors simultaneously). Pulses (pairs of counts) are symmetrically distributed above and below the 1028 line.

While the neutron generator is on multiple pairs of pulses are continuously recorded at all time separations. After the generator pulse is over this source of randomly correlated neutrons no longer exists and the only correlated source of neutrons remaining in the experiment are a) those originating from fission caused as the neutron population in the drum thermalizes and produces follow-on fission in the HEU (the die-away region) and b) from the decay of beta-delayed fission products that emit neutrons (the delayed neutron region), which are not time correlated. As seen in the figure, the passive signature from the drum is very weak. The active background signature shows a processing artifact but otherwise, after the end of the neutron pulse, the correlated pair spectrum is essentially empty. The spectral signature from the 8 kg HEU drum, in contrast, shows a very strongly correlated paired neutron signature out until 100 microseconds and the start of the next pulse. The measurement of pairs of neutron events following irradiation is a very powerful technique for detecting SNM, is resilient to random background signatures, and can be insensitive to nearby photon radiation.

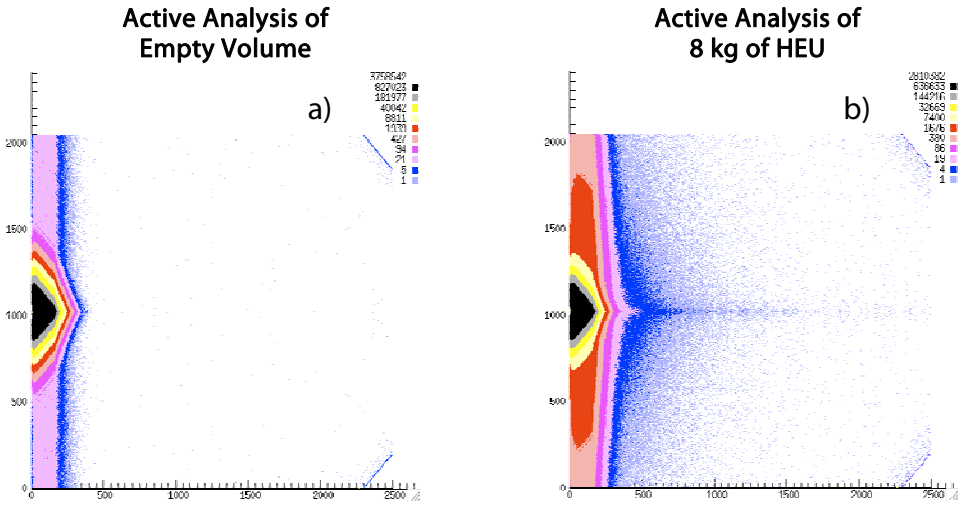
Figure 6 NMIS data with the liquid scintillator array of a) the 6M drum loaded with 8 kg of HEU without the external neutron source, b) the array response with the ENG without HEU, and c) the array response with 8 kg of HEU and the ENG operating.[1]



[1] Plots courtesy of P. Hausladen, ORNL.

Similar to the liquid scintillator data, paired neutron counting plots are also shown in Figure 7 for data collected with the He-3 detector array. Rather then detecting fast neutrons directly as with the liquid scintillators, these detectors rely on first moderating the fast neutrons in the polyethylene shroud that surrounds the He-3 tubes. As a result of this process the time correlations between neutron events in these detectors are broadened versus those seen in the liquid scintillators; this can be seen in Figure 7b where the correlated neutron pairs following the end of the neutron pulse are spread much wider in time. Despite this broadening though the existence of a time correlated neutron source is still clearly evident using the He-3 detector arrays as well.

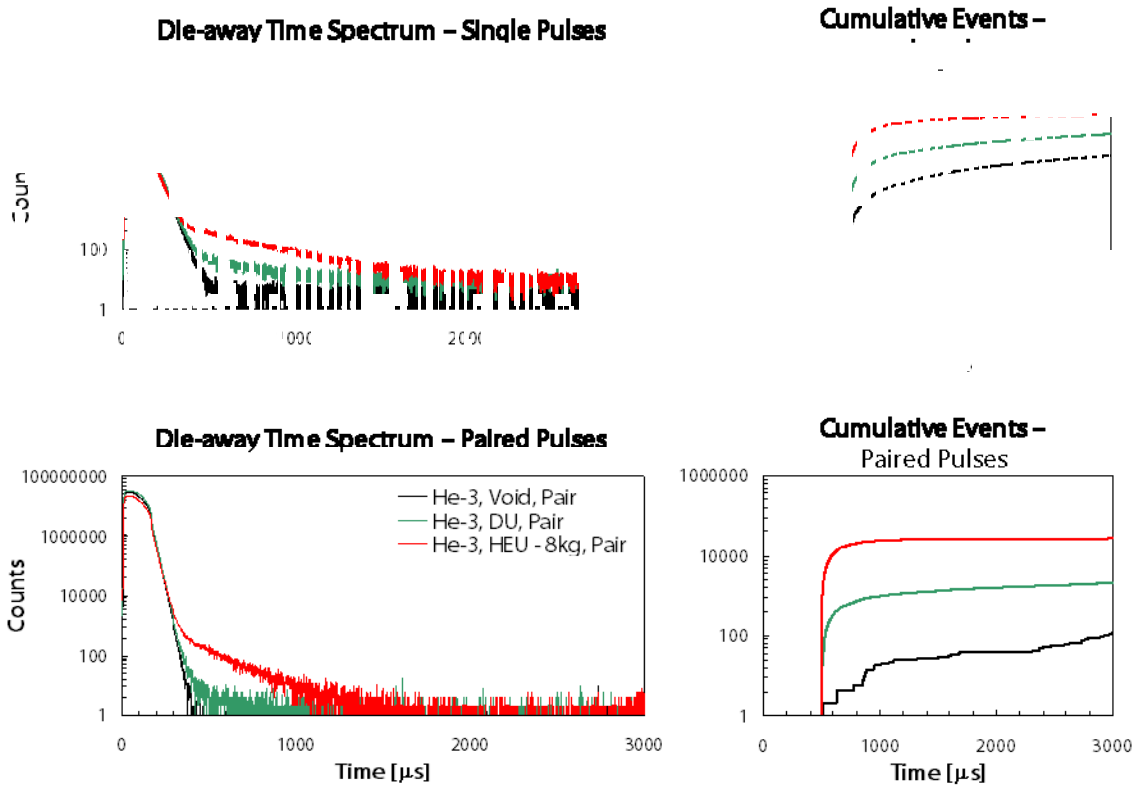
Figure 7 NMIS data with the He-3 detector array of a) the array response with the ENG without HEU and b) the array response with 8 kg of HEU and the ENG operating.[1]



From the helium-3 data further analysis has also been performed to assess the relative sensitivity of this technique for detecting and eventually quantifying the HEU in the drums; data from this analysis is presented graphically in Figure 8. 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. Measurement times for collection of this data were approximately 40 minutes. 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 beta delayed neutron emitters fission products and the detectors inherent background count rate (the delayed neutron region).

[1] Plots courtesy of P. Hausladen, ORNL.

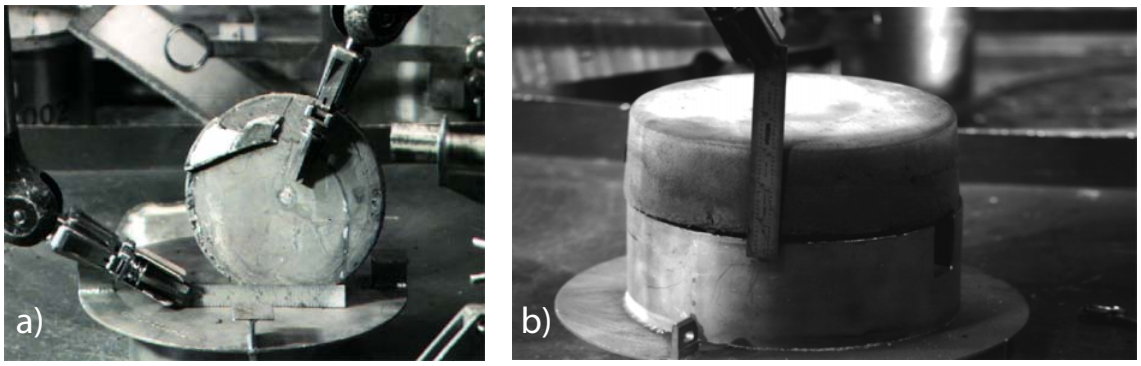
Figure 8 Die-away time spectra and cumulative event spectra (beginning 350 microseconds after the start of the neutron pulses) from the He-3 array for both single count events and paired counting events (events separated by less than 650 microseconds) measuring the active background, the 6M Drum signature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



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 8, a further improvement in signal-to-noise is seen in the die-away of this data versus the single pulse counts. Simply 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 probably also serve to improve the detection efficiency of these techniques.

Extending these observations to the GNEP safeguards context it is clear that active interrogation could be used to perform SNM material inspections of bulk materials including spent fuel hull claddings, solid waste forms, or low-activity waste drums. Images of two bulk material forms, a metals waste form and a uranium metal ingot, from spent fuel reprocessing activities at INL that might also be analyzed using active interrogation are shown in Figure 9. Extending these preliminary results further, and as shown in previously published literature, the technique also has applicability in process monitoring to detect and quantify SNM content in future nuclear reprocessing facility process streams.

Figure 9 Photo a) shows a metal waste form from INL's electrochemical reprocessing (pyroprocessing) facility created from melting down spent fuel hull claddings from the Experimental Breeder Reactor – II (EBR-II) fast reactor, photo b) shows a uranium ingot made from melting and casting reprocessed EBR-II fuel after undergoing electrochemical reprocessing.



Future Active Interrogation Safeguards Work at INL

Future work at INL will proceed towards applying active interrogation technology to find measurement solutions for GNEP safeguards challenges. In particular, work will be performed to identify unique measurement problems anticipated in the GNEP facilities where active interrogation can be used. These investigations will be carried out in consultation with experts in aqueous and electrochemical reprocessing in order to identify unique situations associated with the GNEP fuel cycle (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. The applicability of using active interrogation techniques to monitor both radioactive and non-radioactive process streams in advanced GNEP facilities will also be investigated. INL has a long established partnership of collaborating with other Department of Energy laboratories and universities in the field of active interrogation. These relationships will be leveraged in support of this work including an already existing partnership with local colleagues associated with Idaho State University.

Acknowledgement

The experiments described in this report 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. Special thanks are extended to Paul Hausladen for preparing the time correlation plots and to Paul and Jim Johnson for assembling together the raw data used to analyze the helium-3 detector results. The work in this report was sponsored by the Department of Energy GNEP Safeguards Campaign and the Department of Homeland Security's Domestic Nuclear Detection Office (DNDO).

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