EXPERIMENTAL VALIDATION OF NDA CAPABILITIES FOR MSR SAFEGUARDS: FIRST RESULTS

Matthew H. Carpenter
Los Alamos National Laboratory

Katrina Koehler
Los Alamos National Laboratory

Krystel De Castro
Los Alamos National Laboratory

David Mercer
Los Alamos National Laboratory

Sophie Weidenbenner
Los Alamos National Laboratory

Duc Vo
Los Alamos National Laboratory

Athena Sagadevan
Los Alamos National Laboratory

Daniela Henzlova
Los Alamos National Laboratory

Howard Menlove
Los Alamos National Laboratory

Mark Croce
Los Alamos National Laboratory

Michael Dion
Oak Ridge National Laboratory

Susan Smith
Oak Ridge National Laboratory

Joel Ullom
University of Colorado, Boulder

Daniel Becker
University of Colorado, Boulder

Jeffrey Sanders
Idaho National Laboratory

ABSTRACT

Current molten salt reactor (MSR) safeguards modeling efforts are limited by the lack of measurement data on realistic MSR materials as identified by advanced nuclear industry leaders and safeguards modeling experts. The challenging environment of an MSR where accessibility or direct sampling may be limited or impossible means that rapid, non-destructive characterization of fissile material and waste streams at MSRs is likely to be essential to meet regulatory requirements in a cost-effective way. An advanced systems approach is needed to meet safeguards requirements, and a main component of this approach is validated measurement performance capabilities. The goal of this project is to (1) measure gamma-ray and neutron signatures from nuclear material samples that have characteristics similar to material at an MSR facility, (2) assess limits of rapid anomaly detection and characterization of material compositions, and (3) evaluate nondestructive assay (NDA) concepts for harsh, high-radiation environments. This work will result in a comprehensive set of validated measurement capabilities for safeguards models. We present a systematic approach developed for the evaluation of NDA measurement capabilities and current results from a measurement campaign at Los Alamos National Laboratory focused on traditional and advanced gamma spectrometers. Measured materials from a spent fuel separation process at Argonne National Laboratory were chosen to evaluate limits of quantifying fissile material signatures in the presence of varying concentrations

of fission products and actinides. Both high-purity germanium and microcalorimeter detectors are capable of quantifying important nuclides, and each have unique advantages due to efficiency or energy resolution. Results suggest that direct quantification of important actinides in fuel salt may be possible by NDA for a sampling loop in an operating MSR or for salt samples.

INTRODUCTION

Current molten salt reactor (MSR) safeguards modeling efforts are limited by the lack of measurement data on realistic MSR materials as identified by advanced nuclear industry leaders and safeguards modeling experts. The challenging environment of an MSR where accessibility or direct sampling may be limited or impossible means that rapid, non-destructive characterization of fissile material and waste streams at MSRs is likely to be essential to meet regulatory requirements in a cost-effective way. An advanced systems approach is needed to meet safeguards requirements, and a main component of this approach is validated measurement performance capabilities. The goal of this project is to (1) measure gamma-ray and neutron signatures from nuclear material samples that have characteristics similar to material at an MSR facility, (2) assess limits of rapid anomaly detection and characterization of material compositions, and (3) evaluate nondestructive assay (NDA) concepts for harsh, high-radiation environments.

The experimental approach is based on a series of measurement campaigns using traditional and advanced NDA technologies to comprehensively evaluate performance with respect to material composition, spectral complexity, dose rate, cooling time, measurement environment, and other unique challenges of MSR facilities. Current MSR concepts include a wide range of possible designs including solid (salt-cooled designs) as well as liquid fuel (salt-fueled designs) with the nuclear material dissolved in molten salt for the latter [1]. Some designs may be considered a combination of the two, where certain fission products are allowed to enter the molten salt. Safeguards concepts for the salt-cooled designs will likely be more analogous to present-day reactor safeguards due to the use of fuel in solid form. The salt-fueled designs present a range of new safeguards challenges due to the liquid form of nuclear material and therefore the safeguards concepts are much less developed. As a result, salt-fueled designs are the primary focus of this NDA evaluation effort. An overview of the wide range of currently proposed MSR concepts is shown in Figure 1. It can be seen that the majority of the designs propose to use liquid fuel with several fuel variants including U and TRU fuel materials.

The NDA technologies employed in MSR safeguards will face a range of challenges in that the MSR present attributes of traditional bulk handling facilities (such as present-day reprocessing facilities), however with much smaller amounts of material added and removed. Additionally, many MSR concepts assume a closed reactor loop with the potential of on-line salt processing and refueling. From an operational standpoint, MSRs require high temperature (~700 ºC) to keep the fuel salt in liquid phase. The fuel salts represent complex mixtures of actinides and fission products and exhibit a very high radiation background. This motivates evaluation with representative measurements of two scenarios: on-line NDA measurements of an operating MSR, and laboratory measurements of small samples taken from the process where a wider range of NDA technologies are applicable.

A systematic approach was developed for the evaluation of NDA measurement capabilities and a measurement campaign at Los Alamos National Laboratory focused on gamma technology comparison was completed. Measured materials were chosen to evaluate limits of quantifying fissile material signatures in the presence of varying concentrations of fission products. Plans for neutron

measurement campaigns at LANL and joint measurements at Oak Ridge National Laboratory and Idaho National Laboratory on increasingly representative materials have also been initiated.
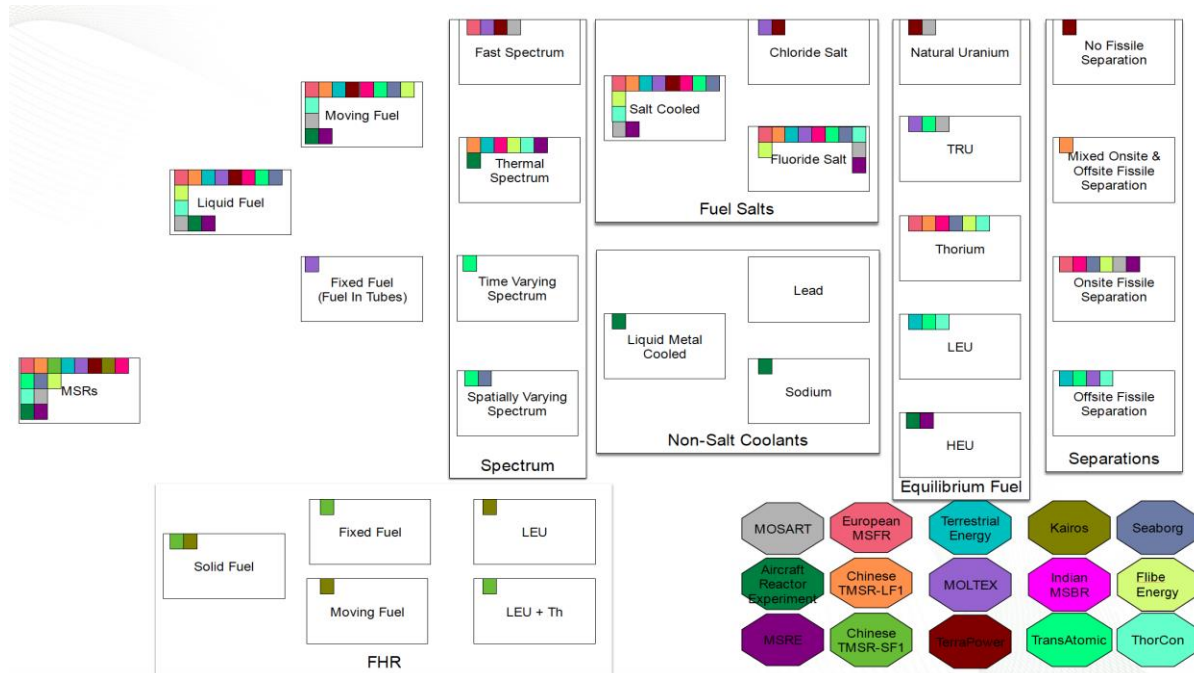

Figure 1: (From [1]) Overview of MSR technologies under development.

ANALYSIS APPROACH

While MSR safeguards requirements are still being established, it is expected that timely detection of diversion of fissile material with high detection probabilities will depend on precision measurements of material composition throughout a facility. Verification of, or detection of changes in, fissile material content is the primary objective in the MSR safeguards approach. An important goal of the project is to establish a common framework for evaluation of NDA measurement capabilities in order to facilitate comparisons between and selection of measurement technologies at key locations. Gamma-ray spectroscopy and correlated neutron counting are cornerstones of nuclear material accounting in safeguards of existing nuclear fuel cycle facilities. Experience and modeling results suggest that gamma-ray and neutron signatures will be similarly valuable for NDA of MSRs as the most direct methods of quantifying fissile material content throughout a facility.

In another Advanced Reactor Safeguards project, the Transient Simulation Framework of Reconfigurable Models (TRANSFORM) has been developed at ORNL to study the system-level dynamics of MSRs. Experimental design in this project is informed by the modeling work, and experimental results will be used to inform safeguards models. TRANSFORM provides a way to implement system-level specifications including pipe dimensions, pump characteristics and other details that are extremely useful to evaluate various operating conditions [2]. It is based on the object oriented, open-source Dymola Modelica modeling language. Currently, a variation on the Molten Salt Demonstration Reactor (MSDR) has been modeled in TRANSFORM and is being used to predict fission product movement and core reactivity. The model layout as displayed in the software is shown in Figure 2.
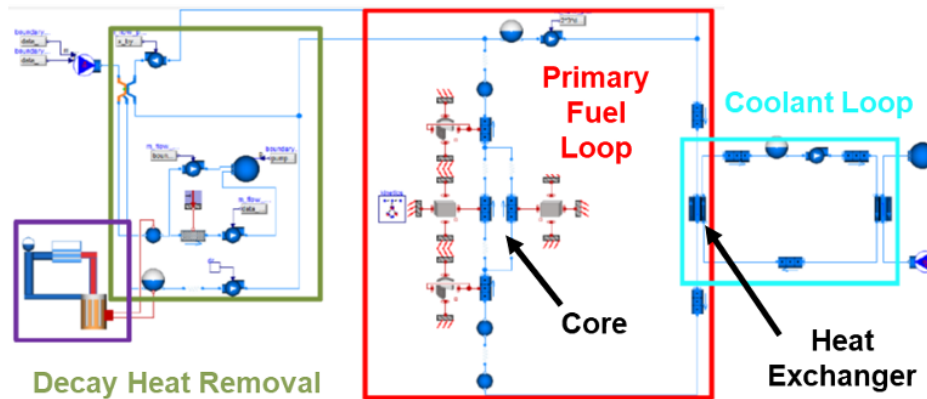
Figure 2: Layout of the MSDR in TRANSFORM being used for simulations of dynamic fission product concentrations.

Currently, an abbreviated population of fission products are handled as trace materials in the TRANSFORM model. These trace substances are carried in the bulk molten fuel-salt mixture; here it is a FLiBe salt matrix. It is possible to obtain concentrations of any fission product at any location in the model. Therefore, this is being utilized to develop an understanding of the dynamic behavior of fission product signatures. This data can be used to determine how fission product signatures vary with time and fissile content burnup. These results are used to inform experimental validation work about key measurement locations, and detector requirements.

The analysis approach begins with identifying relevant signatures. For gamma spectroscopy methods, energy peaks associated with fissile nuclides are identified and uncertainty is quantified for a given measurement time. With additional information about the process, these peaks could be compared with fission product peaks as an indicator of normal or off-normal conditions. For neutron counting methods, time-correlated signatures resulting from fission rather than (alpha, n) reactions can be used. Additionally, total neutron counting can be used in process monitoring applications to confirm presence or absence of nuclear material. The main performance metric for each measurement is the sensitivity with which it can detect changes in fissile material concentration in a given measurement time. A staged approach to measurements begins with immediately available fuel cycle materials in order to answer fundamental questions about observable signatures and progresses towards increasingly realistic MSR materials. The results are expressed in terms of the statistical uncertainty achievable for relevant signatures and intended as input values for MSR safeguards system modeling efforts.

Gamma NDA measurements include spectroscopic and non-spectroscopic technologies. Non-spectroscopic gamma detectors such as ion chambers are useful for high-dose environments but only provide a measure of total gamma-ray flux. For the complex mixtures of actinides, fission, and activation products encountered in an MSR, a non-spectroscopic gamma measurement therefore provides very limited information that generally cannot be used in quantification of specific nuclides. As a result, this evaluation of gamma NDA capabilities is focused on spectroscopic technologies. Primary metrics in gamma spectroscopy include detection efficiency, count rate capability, dynamic range, and energy resolution. Specific detector types were chosen to cover this parameter space. Sodium iodide scintillation detectors are a well-established low-resolution detector type (approximately 8% FWHM resolution at 662 keV). They have been evaluated for process monitoring using a principal component analysis approach [3]. However, their resolution is insufficient to

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distinguish nuclides in complex MSR-relevant materials. High-purity germanium (HPGe) detectors are the commercially available standard for high-resolution gamma spectroscopy, with low-energy planar detectors yielding energy resolution of approximately 550 eV FWHM at 122 keV. Microcalorimeter spectrometers are an emerging ultra-high energy resolution gamma spectroscopy technology with the first instruments now being built for use in nuclear facilities and analytical laboratories [4-5]. Energy resolution of microcalorimeter gamma spectroscopy is typically 60-100 eV FWHM at 129 keV. This work considers sodium iodide, high-purity germanium, and microcalorimeter detectors (Figure 3). Based on these results, intermediate-resolution detectors including CdZnTe will be considered in the next measurement campaigns.

To determine the sensitivity of a particular measurement method, regions with the best signatures of the various isotopes are chosen. These regions are fit with a sum of peak shapes, and the peak areas with their associated statistical uncertainty are saved. These areas and uncertainties are then scaled to correspond to a 24-hour measurement for each data type (i.e. HPGe and microcalorimeter data). Since the process is a chemical process, particular isotopes do not need to be tracked as much as elements. As a result, a strong ^{241}Am peak is all that is needed to quantify the Am content, so further quantification of the lesser constituent ^{243}Am need not be considered. For each element, the peak that provides the lowest uncertainty is used.

Neutron NDA technologies for safeguards applications focus on detection of individual and/or correlated neutron signatures from spontaneous and induced fissions within the nuclear materials. Passive neutron NDA technologies require sufficient neutron emission rate (i.e. spontaneous fission and (alpha,n) neutrons) and are typically used for assay of Pu-bearing materials [6]. Due to the low spontaneous fission yield of U-isotopes (^{233}U, ^{235}U, ^{238}U), U-bearing materials require active NDA and are typically assayed using active interrogation with an external neutron source [6].

The neutron detection technologies traditionally used in safeguards applications include primarily ^{3}He-based proportional counters and fission chambers. ^{3}He-based instruments have been a gold standard of nuclear safeguards for the past decades and provide high neutron detection efficiency to support quantitative nuclear material assay. Fission chambers are typically used in applications with high gamma dose rates, such as spent fuel measurements and are capable of operating at dose rates of up to 10,000 R/hr, however they provide very low detection efficiency on the order of 0.01%. Also, the fission chambers present shipping and regulatory control issues because of the fissile material content. A new technology developed for high gamma background applications, such as MSRs, is the High Dose Neutron Detector (HDND). The HDND technology is based on boron-lined proportional counters, which provide inherent fast signal characteristics and high gamma dose tolerance [7]. In addition, the HDND provides simultaneous monitoring of the gamma-ray emission rate. This work considers ^{3}He detectors, fission chambers, and the HDND (Figure 3).
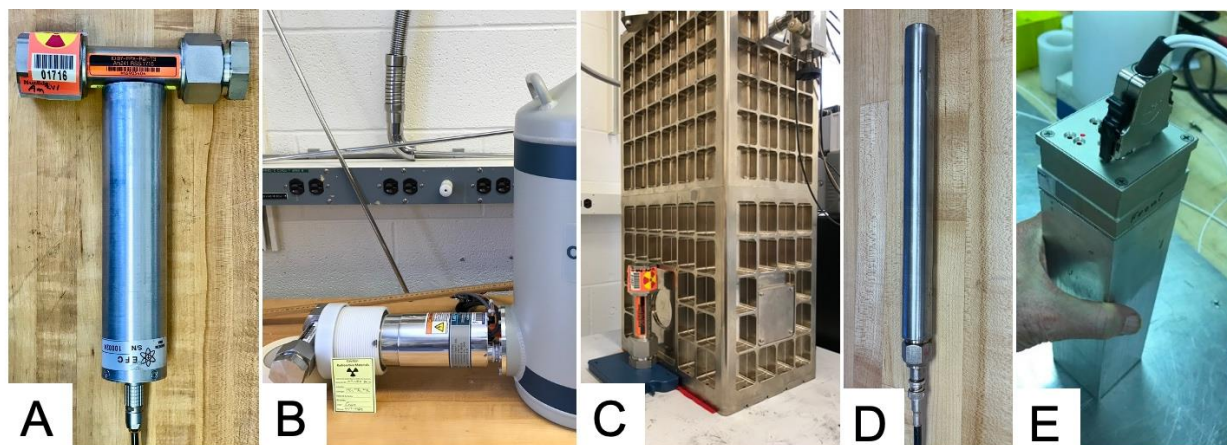
Figure 3: Primary detector types considered in this work: A) sodium iodide scintillation detector (NaI), B) planar high-purity germanium detector (HPGe), C) SOFIA microcalorimeter gamma spectrometer, D) ^{3}He neutron detector, E) mini High Dose Neutron Detector (miniHDND)

MEASUREMENT CAMPAIGN

A series of samples from an aqueous separation process of spent nuclear fuel at Argonne National Laboratory is available at Los Alamos National Laboratory. These well-characterized samples are ideal for an initial evaluation of the ability of gamma spectroscopy methods to quantify fissile material signatures in the presence of varying concentrations of fission products and as actinide components are removed from the material. Measurements of each sample were completed with a NaI(Tl) detector from EFC Company (Figure 3A, [8]), a Canberra GL1015 low-energy HPGe detector (Figure 3B, [9]), and the SOFIA microcalorimeter spectrometer (Figure 3C, [4-5]). Many of the best spectral signatures are observed in the 55-125 keV region. The NaI detector has insufficient resolution to make use of this complex spectral region for quantitative analysis, while the HPGe and microcalorimeter detectors resolve a number of important peaks. Therefore, quantitative analysis efforts focused on the two high-resolution gamma spectroscopy technologies. As components are removed from the spent fuel, especially fission products, actinide signatures become more clear. Although the age of the spent fuel used in this work leads to a moderate level of fission product activity compared with intact freshly-discharged fuel elements, in many liquid-fueled MSR designs an offgas system is designed to remove gaseous fission products. Since many fission products have Xe or Kr precursors, over 40% of fission products are expected to leave the core including a large fraction of Cs, Sr, and I which are collected in the offgas system [1]. Therefore, direct quantification of important actinides in fuel salt may be possible by NDA for a sampling loop in an operating MSR or for salt samples with a short cooling time. Further work including comparison with TRANSFORM model results is required to test this hypothesis.

Quantitative analysis of the constituents within the samples was performed on both the microcalorimeter and HPGe measurements. For each sample, key regions were fit to determine the peak areas and uncertainties, with uncertainties coming from the minimization covariance matrix. Because chemical processing in an MSR does not discriminate between the isotopes of a particular element, the area of a peak of any isotope of a particular element with the lowest uncertainty is most sensitive to possible elemental diversion. Systematic uncertainty has the potential to be significant

due to peak overlaps and complex background shapes particularly in the 100 keV region, and merits further study. The improved resolution of the microcalorimeter data, where peaks are better resolved from each other and a narrower background region is relevant, may substantially reduce this type of uncertainty contribution and therefore allow peaks in this region to be more confidently used for quantitative analysis.

An initial evaluation of miniHDND [10] was performed in preparation of future measurement campaigns at Oak Ridge National Laboratory. Basic neutron and gamma counting capability was evaluated using ^{252}Cf neutron and ^{137}Cs gamma sources in the form of high voltage plateaus. It should be emphasized that gamma sensitivity of miniHDND is tunable via gain of each signal processing amplifier. In this initial evaluation all three amplifiers were matched to provide the same gamma response. For practical applications, the gain on the gamma cell can be increased to improve gamma sensitivity, while maintaining the lower gamma sensitivity of the neutron cells and thus not affecting their performance. The ^{137}Cs high voltage plateaus indicate good gain matching of the three detector amplifiers. Note that the dose rate from these sources is very low (~60 mR/hr on contact) as this measurement was a part of an initial characterization. Further evaluation will be performed with high gamma dose rates prior to use in hot cells at ORNL. Similarly, the neutron performance was evaluated using a ^{252}Cf neutron source. Combining the gamma and neutron results, the suggested operating voltage for the instrument is ~900 V. The neutron detection efficiency for the current miniHDND configuration therefore corresponds to 0.12% for a ^{252}Cf source at the detector face.
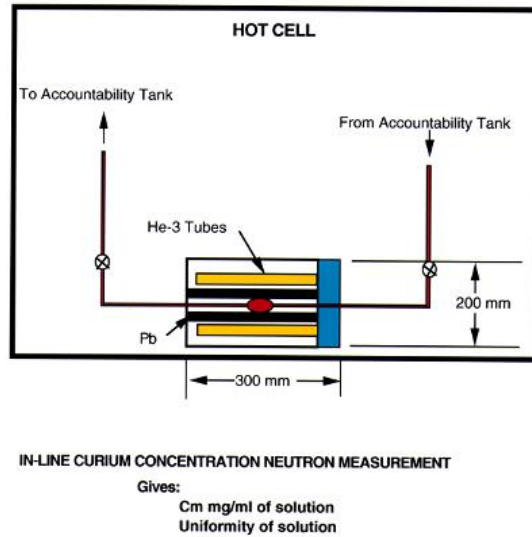

Figure 4: Illustration of on-line coincidence counting concept originally considered for reprocessing applications.

Due to the MSR facility configuration (a closed loop with potential for on-site salt processing and refueling), it can be anticipated that NDA measurements would be performed on flowing fuel salt in MSR piping, on salt samples extracted from the process or on material for refuel. The measurements of flowing fuel salt could use total neutron counting as well as total gamma counting in order to establish process monitoring capability and provide information on presence or absence of nuclear material in various streams. Higher efficiency neutron detectors could also surround a pipe to enable correlated neutron measurements for assay of flowing nuclear material. Figure 4 illustrates the concept of measuring a side stream of the reprocessing liquid that was considered in the past. Because

the MSR fuel cycle materials do not typically contain significant spontaneous fission signatures, use of ^{252}Cf source neutrons or neutron generator would have to be considered to induce fission reactions in the fuel combined with the time-correlated counting of the measuring the induced fission reactions in the uranium fissile material.

Neutron assay of samples from the process could also be used for quantitative assay, similarly to instruments used in current reprocessing and fuel fabrication facilities (AVIS [11], INVS [12]). In all such measurement scenarios, the deployed technology will have to satisfy unique MSR challenges of high radiation backgrounds, high temperatures and remote operations. The key focus of this neutron detection technology intercomparison will therefore be on a set of criteria including capability to provide reliable neutron detection in competing high gamma backgrounds, advanced process monitoring capabilities, and ability to operate in high temperature environments. Practical considerations such as robustness, maintenance requirements and compatibility with remote-handling environments will also be assessed.

Spent nuclear fuel rods available at Oak Ridge National Laboratory offer an opportunity to evaluate NDA measurement performance with extremely high activity materials [13]. The rods came from fuel assemblies irradiated at the North Anna Power Station and were shipped to ORNL in 2016. Rod average burnup is greater than 45 GWd/MTU and documented discharge dates range from 1994-2010. Some of the fuel rods are undergoing processing, which would allow comparison of measurement performance for both intact fuel rods and cut fuel elements where gaseous fission products have been released.

Figure 5: Germanium detector installed at Idaho National Laboratory's Hot Fuel Examination Facility (HFEF) used to perform post-irradiation examination on irradiated fuel rods and specimens. The detector views items inside the hot cell through a collimator which reduces gamma flux and has a BGO guard detector to reduce effects from Compton scattering.

Idaho National Laboratory is active in the development of pyroprocessing technology and has current programs exploring spent nuclear fuel separations using molten salt technology. Measurements are

planned at the Materials and Fuels Complex Analytical Laboratory (AL) to evaluate laboratory NDA of small process samples, and at the Hot Fuel Examination Facility (HFEF) to evaluate on-line measurements.

The AL routinely analyzes a wide range of samples from advanced nuclear fuel cycle development work at Idaho National Laboratory, and includes destructive and traditional nondestructive analysis capabilities. Through separate projects, a microcalorimeter gamma spectrometer is currently being built for deployment to the AL. This new instrument will be used alongside traditional NDA technologies to measure available samples from electrochemical separations relevant to on-line separations in liquid-fueled MSRs. An important outcome of this measurement campaign will be to validate the performance of advanced and traditional NDA technologies with the same relevant samples handled in a laboratory environment. In addition to the gamma-ray and neutron technologies summarized in this report, advanced CdZnTe gamma-ray systems and other spectroscopic and neutron detection technologies will be considered.

HFEF can provide measurement opportunities both in and adjacent to the hot cell where intensely radioactive spent fuel materials are available. Prior testing of the HDND at HFEF demonstrated the capabilities of this facility for evaluation of NDA technologies in an operational environment [14]. Figure 5 shows an example of a germanium detector installed at HFEF to view items inside a hot cell through a collimator. HFEF is a highly subscribed facility, especially due to COVID-related operational restrictions. Contingent on approvals, it may be possible to perform a side-by-side comparison of traditional and advanced NDA technologies at HFEF.

CONCLUSIONS

We have developed a systematic approach for the evaluation of NDA measurement capabilities and applied to a measurement campaign at Los Alamos National Laboratory focused on gamma-ray measurement technology. Measured materials from a spent fuel separation process at Argonne National Laboratory were chosen to evaluate limits of quantifying fissile material signatures in the presence of varying concentrations of fission products and actinides. Both HPGe and microcalorimetry are capable of quantifying important nuclides, and each have unique advantages due to efficiency or energy resolution. Results suggest that direct quantification of important actinides in fuel salt may be possible by NDA for a sampling loop in an operating MSR or for salt samples. Further work including measurements of additional irradiated fuel materials and comparison with TRANSFORM model results is required to test this hypothesis. Initial testing of the miniHDND in preparation for future measurement activities has been completed. Plans for neutron measurement campaigns at LANL and joint measurements at Oak Ridge National Laboratory and Idaho National Laboratory on increasingly representative materials have also been completed and the measurement campaigns are about to commence.

ACKNOWLEDGEMENTS

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