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AFCI Safeguards Enhancement Study: Technology Development Roadmap

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Abstract

The Advanced Fuel Cycle Initiative (AFCI) Safeguards Campaign aims to develop safeguards technologies and processes that will significantly reduce the risk of proliferation in the U.S. nuclear fuel cycle of tomorrow. The Safeguards Enhancement Study was chartered with identifying promising research and development (R&D) directions over timescales both near-term and long-term, and under safeguards oversight both domestic and international. This technology development roadmap documents recognized gaps and needs in the safeguarding of nuclear fuel cycles, and outlines corresponding performance targets for each of those needs. Drawing on the collective expertise of technologists and user-representatives, a list of over 30 technologies that have the potential to meet those needs was developed, along with brief summaries of each candidate technology. Each summary describes the potential impact of that technology, key research questions to be addressed, and prospective development milestones that could lead to a definitive viability or performance assessment. Important programmatic linkages between U.S. agencies and offices are also described, reflecting the emergence of several safeguards R&D programs in the U.S. and the reinvigoration of nuclear fuel cycles across the globe.

Summary

The Advanced Fuel Cycle Initiative (AFCI), a program funded by Department of Energy Office of Nuclear Energy (DOE-NE), is organized into campaigns aligned with key components of advanced nuclear fuel cycle research (*e.g.*, Fuels, Separations). The AFCI Safeguards Campaign aims to develop safeguards technologies and processes that will significantly reduce the risk of proliferation in the US nuclear fuel cycle of tomorrow. The Safeguards Enhancement Study (SES), one project within the Safeguards Campaign, is chartered with identifying promising R&D directions over time scales both near-term and long-term, and under safeguards oversight both domestic (*e.g.*, Department of Energy and Nuclear Regulatory Commission) and international (IAEA).

Reflecting the expansion of nuclear fuel cycles worldwide and a renewed interest in the safeguarding of those fuel cycles, there have been several different programmatic planning and technology assessment efforts commissioned by U.S. agencies in recent years. Perhaps the most prominent and far-reaching is the Next-Generation Safeguards Initiative (NGSI) program plan authored by the DOE Office of Nonproliferation and International Security (DOE NA-24). The NGSI program plan calls for a major US investment in policy, technology, human capital and infrastructure development considered key to the future of nuclear fuel cycle safeguards. One of the prominent supporting documents for the NGSI plan was the Advanced Safeguards Approaches (ASA-100) project commissioned by DOE NA-243, which provided a comprehensive review of current safeguards technologies, described ongoing R&D efforts, and provided recommendations regarding future investments in technology development. Meanwhile, DOE's Office of Nonproliferation Research and Engineering (DOE NA-22) is in the process of writing its long-term program plan in support of Safeguards.

This AFCI-supported document has common ground with these other roadmapping and technology assessment activities, and is complementary in many ways, but there are defining differences as well, as described below:

<u>*Scope of this roadmap:*</u> Reflecting contemporary plans and funding profiles for the AFCI program, the emphasis in this SES roadmap is on the back end of the fuel cycle as it might be developed in the United States. In the AFCI context, this means recycling approaches for both near-term (*e.g.*, mixed oxide fuel into light-water reactors) and long-term (e.g. recycled fuel into transuranic "burner" reactors). Consequently, this roadmap is focused on the safeguards needs and technologies for used fuel reprocessing, mixed oxide fuel fabrication, and the movement of used fuel and product streams from facility to facility. The safeguarding of enrichment facilities and existing commercial power reactors is not addressed because those areas are covered under other programs (*e.g.*, NA-24). Nor are "Grid-Appropriate Reactors" included explicitly in this roadmap, as the future of U.S. investment in that area is uncertain at this time.

<u>*Formulation of Gaps and Needs:*</u> The needs documented in this study, for the most part, were gleaned from recent or ongoing advanced safeguards activities outside of AFCI (*e.g.*, the NGSI roadmap and ASA-100 reports funded by NA-24) because it is the SES team's view that the high-level needs in the safeguards community are well-understood. These gaps and needs represent both domestic and international safeguards requirements because while AFCI is a US-focused program, new US nuclear fuel cycle facilities are likely to be on the IAEA Eligible Facilities list. For each need, performance targets are described. These targets, while far from definitive, attempt to set the scale for what transformational

safeguards technologies must achieve in order to significantly reduce proliferation risk. Ultimately, these performance targets will be defined by an integrated systems-level analysis but that analysis, in and of itself, is a research and development need in the safeguards community.

Roadmapping Team Composition: By design, the SES team consisted of primarily technology developers, as opposed to technology users from the IAEA or other regulatory bodies (such as the Nuclear Regulatory Commission). This means that while users (*e.g.*, former IAEA inspectors) provided insights and review, this roadmap is written primarily from the perspective of technologists interested in exploring the boundaries of signatures and science for safeguards applications. In some cases, the proposed technology solutions are inconsistent with today's inspection protocols because the hope is that technology breakthroughs will enable a completely new approach to verification activities and materials accounting.

Technology Summaries Identify Potential Path Forward and Indicate Technology Readiness: A brief (1-2 page) summary of each candidate technology is included in this report. The objectives of each summary are to highlight the potential impact on safeguards of that technology (if it were successfully developed), identify the key research questions that would need to be addressed, and outline a series of R&D milestones that would allow the concept to either be dismissed or pursued as a prototype at a representative fuel cycle facility. A coarse categorization of technical maturity, using the Department of Defense (DOD) Technology Readiness Level designations, is also included.

Each of the Candidate Technology summaries was written by recognized subject-matter experts in that field, and reviewed by at least one other recognized subject-matter expert. Over 50 different staff and reviewers from Department of Energy (DOE) National Laboratories, industry and academia contributed to this roadmap, with the goal of making it non-parochial and objective. A complete listing of the authors and reviewers for the Candidate Technology summaries is provided in Appendix A.

Taken in the context of the NA-24 and NA-22 roadmapping and technology assessment documents developed in the past year, the defining contribution of this AFCI document is the concise list of research questions and proposed milestones for each candidate technology. It is hoped that this information will be useful to programmatic planning and prioritization activities, as well as a resource to technologists working in these application areas.

The Safeguards Campaign has organized itself around three themes: 1) Advanced Instrumentation, 2) Advanced Control and Integration, and 3) Safeguards by Design. This report adopts this organization, beginning with the tables below. Those tables summarize Needs and Performance Targets, and provide overviews of each Candidate Technology in terms of potential impact and technical maturity.

Table 1. Needs and performance targets in the Safeguards Campaign areas of Advanced Instrumentation, Advanced Control and Integration, and Safeguards by Design.

Advanced Instrumentation	
Need	**Performance Targets**
Direct measurement of Pu in used fuel and actinide bearing materials	On-line measurement, Pu uncertainty <2%, no *a priori* information about fuel
Direct measurement of Pu in reprocessing streams	*Continuous*, *on-line* monitoring of Pu meeting IAEA diversion and timeliness goals • Protracted: <1 SQ per month or <1% • Abrupt: near-real-time analysis and <5%
Advanced containment and surveillance techniques	• Tag/seal technologies for near-real-time tracking of feed/product containers in fuel cycle • Radiation-based (*e.g.*, neutrons) continuity of knowledge from reactor to processing or repository
Advanced design information verification methods	Technology suite that makes design information verification integral from construction through decommissioning

Advanced Control and Integration	
Need	**Performance Targets**
Process monitoring of reprocessing streams	• *Continuous on-line* measurement of parameters (*e.g.*, radiation signatures, pH, flow) • Statistical process control methods for sensitive off-normal detection • Near-real-time visualization, analysis and reporting to facilitate decision-making by inspectors • Authenticated data transmission to enable remote, on-line facility monitoring technologies
Rapid laboratory sample analysis	Reduce sample preparation requirements and analysis time, thereby reducing manpower and cost for onsite laboratory.

Safeguards by Design	
Need	**Performance Targets**
High-fidelity integrated safeguards modeling	Safeguards by Design methodology that: • Integrates proliferation resistance and physical protection functionality and metrics • Provides discrete-event and dynamic facility simulation • Utilizes independently peer-reviewed risk/cost metrics • Includes benchmarking, blind testing using available facility data

The tabular summary of candidate technologies given below spans the continuum of technical maturity from highly-exploratory concepts to incremental improvements or adaptations of the tried-and-true. Both are important to a long-range R&D plan for nuclear fuel cycle safeguards; the former represents the building blocks that enable the creation of new and improved methods on longer time scales, while the latter can support near-term enhancements at fuel cycle facilities. The defining potential impact for each technology is important to articulate because there is no silver bullet in these technologies—each has particular strengths and may bring a different facet to meeting each need/requirement.

The Technical Readiness Level (TRL) given for each technology is based on the formulations used by DOD and NASA (see NGSI roadmap for additional information). TRL1 describes a technology in which the basic scientific principles have been observed and reported, and scientific research has begun on the topic. TRL3 describes technologies that are in the analytical and/or experimental proof-of-concept stage, while TRL6 represents a prototype demonstration in a relevant environment. TRL9 is the highest level of maturity and describes a system that has been successfully implemented in day-to-day operations.

The TRL levels in the table below are estimates based on the experience of the project team, but are not definitive. A range of TRL levels are used to indicate those technologies that have seen some level of application and deployment, but are also the subject of ongoing research.

Table 2. Candidate technologies and corresponding needs (headings, in yellow) in the areas of advanced instrumentation, advanced control and integration, and safeguards by design. The potential impact and approximate Technology Readiness Level are also provided. Candidate technologies are listed in alphabetical order under each Need.

Advanced Instrumentation		
Technology	**Potential Impact**	**Technology Readiness**
Need: Direct Pu measurement in used fuel, actinide-bearing material, reprocessing streams		
Advanced Neutron Multiplicity	Total fissile mass	5
Calorimetry	Pu mass in all matrices except used fuel	4-7
Delayed Gamma-Ray Detection	Relative ^{235}U, ^{239}Pu, ^{241}Pu, but limited penetration depth	3
Delayed Neutron Detection	Total fissile mass with emphasis on ^{235}U	4
Differential Die-Away	Total fissile mass with emphasis on ^{239}Pu	7
Electrochemical Separations	Elemental Pu, U, actinides, non-rad based	2
Hybrid K-Edge	Elemental Pu, U, actinides	4-7
Lead Slowing- Down Spectroscopy	^{235}U, ^{239}Pu, ^{241}Pu through depth of assembly	3
Nuclear Resonance Fluorescence	Isotopic masses, but with limited penetration depth	2
Passive Neutron Albedo Reactivity	Total fissile mass	3
Radiochemical Automation	Elemental Pu, U, actinides, non-rad based	2-4
Self-Interrogation Neutron Resonance Densitometry	^{235}U, ^{239}Pu, ^{241}Pu, but with limited penetration depth	3
Ultra-High Res Alpha Spec	U, Pu, actinide isotopics with improved selectivity	2
Ultra-High Resolution Neutron	Light-element indicators (*e.g.*, metallic vs oxide)	2
Ultra-High-Resolution Gamma	U, Pu concentration in outer layer of assembly	3
Need: Advanced Containment and Surveillance		
Active and Passive RFID Tags	Improved continuity of knowledge reduces NDA, DA	4-7
Need: Advanced Design Information Verification		
Radiation Imaging	Holdup monitoring and complement to laser techniques	5-6
Cross-Cutting		
Basic Nuclear Data Measurements	Improve precision of NDA measurements	4-6
Gamma Spectroscopy Algorithms	Improved Pu, U, actinide isotopic accuracy, precision	5-7
Modeling for Instrumentation	Higher fidelity and speed for evaluation, optimization	4-7

Advanced Integration and Control

Technology	Potential Impact	Technology Readiness
Need: Process monitoring of reprocessing streams		
Automated Radiochemistry	On-line monitoring to determine Pu or process changes	3-4
Electrochemical Process Monitoring	Quantitative monitoring of Pu and U mass during electrochemical reprocessing	3-4
Macroscopic Properties Monitoring	Improvement on Pu/Cm ratio over wide range of plant conditions	4-6
Modeling for Adv. Control and Integration	Higher fidelity and speed for evaluation, optimization	4-6
Multi-Isotope Process Monitor	Improvement on Pu/Cm ratio over wide range of plant conditions	2-3
Neutron Balance	Improved continuity of knowledge through fuel cycle	5-6
Statistical Process Control	On-line process control to reduce need for lab analysis	4-6
UV-Visible Spectroscopy	Oxidation states of U, Np and Pu	4-5
Need: Rapid Laboratory Sample Analysis		
Automated Radiochemistry	Reduce laboratory analysis time by automating destructive analysis and by automated sampling approaches that simplify subsequent lab analysis	3-4
Thermal Atomization Resonance Ionization Spectroscopy (TARIS)	Reduce analysis time from hours to minutes, and reduce sample prep requirements	3-4
Need: Integrated Data Authentication		
Data authentication guidelines for classes of instruments and processes	Improve continuity of knowledge, support remote facility monitoring	6-7

Safeguards by Design

Technology	Potential Impact	Technology Readiness
Need: High-Fidelity Integrated Safeguards Modeling		
Safeguards Performance Modeling	Accurate models of processes, measurements and indicators support evaluation of alternatives	4-5
Validated Systems Analysis Methods and Risk-Based Tools for Integrated Systems	Quantitative evaluation of safeguards options, using metrics reflecting proliferation resistance, physical protection and cost simultaneously?	4-5

High-Priority Near-Term Investments

As tabulated above, there are over 30 candidate technologies described in this document aiming at seven identified needs. There are typically several promising methods aligned with each need, but it is also true that there is generally no single method that can meet the need on its own. Rather, it will be a coupling or integration of multiple technologies and approaches that will best solve the problem. Using the direct measurement of elemental Pu in used fuel as an example, high-resolution x-ray spectroscopy can estimate the elemental Pu in the outer regions of the outer fuel pins in an assembly, but can say nothing about the majority (the interior) of the assembly. There are several technologies, however, that have the potential to measure fissile isotopic mass throughout the volume of an assembly (*e.g.*, lead slowing-down spectroscopy and passive neutron albedo reactivity). Together, these technologies have the potential to meet a recognized need and improve on the current approach that relies on burn-up codes and confirmatory measurements. Similar examples could be cited for other key needs.

In most cases the strengths and limitations of each technology and how they might couple for improved outcomes are not yet known. With this in mind, the SES team recommends that some near-term investment be focused on developing rigorous, quantitative frameworks for evaluating candidate technologies individually, and for evaluating the coupling of two or more methods. Due to the cost and complexity of measurement campaigns on highly-radioactive materials, limited access to representative scenarios (*e.g.*, a wide range of well-characterized PWR used fuel assemblies or real UREX processing streams on an engineering scale) and the fact that some instruments don’t yet exist, modeling and simulation becomes the key enabler to assessing and down-selecting individual technologies and combinations thereof. As much as budgets, schedule, and prototype instrument availability allow, the modeling should be supported by empirical benchmarking.

Table 3 lists the near-term investments that the SES team feels are needed to build a strong foundation for future safeguards technology development. The emphasis is on developing high-fidelity modeling that provides a realistic, and standardized environment in which to evaluate the strengths and limitations of candidate technologies.

Table 3. Overview of high-priority near-term investments in AFCI-focused safeguards technology development, as defined by the SES team.

Virtual Used Fuel Library, Source Terms and Assay Metrics
Modeling Core
Isotopic inventory and radiation emissions for a wide range of burn-ups and representative fuel types: Initially, the focus would be light-water reactor assemblies, progressing to MOX, and eventually, to fast-reactor concepts. A working group could define the tools to be used (*e.g.*, ORIGEN or CINDER for burn-up calculations, MCNPX for radiation transport), the data interfaces necessary to support instrument modeling across the community (*e.g.*, gamma-ray and neutron source terms at faces of assembly). The working group could also define a standard convention for quoting instrument performance that incorporates not only systematic effects that define accuracy, but also statistical effects that define precision.
Experimental Complement
Measurements of well-characterized LWR fuel assemblies using suite of neutron and gamma-ray spectroscopy equipment.

Virtual Reprocessing Streams, Source-Term Calculations, Dynamic Modeling
Modeling Core
Virtual Reprocessing Streams, Source-Term Calculations, Dynamic Modeling: Similar to the virtual used fuel library described above, the virtual reprocessing plant would provide the stream characteristics (*e.g.*, pH, density, elemental concentrations) and radiation source terms predicted for the reprocessing plants of tomorrow. A working group could define the appropriate modeling tools (*e.g.*, AMUSE for process stream chemistry and composition, MCNP for radiation source-term calculation) and data interfaces to support instrument evaluation. These data could then be folded into a dynamic model of a reprocessing plant that could be used to simulate normal and off-normal conditions and therefore, provide representative data feeds to statistical process control methods development. Quantitative statistical approaches such as fault tree analysis and scoring methods will be needed. Benchmarking, using available data streams from existing reprocessing plants, will be critical to building confidence in this process monitoring development framework.
Experimental Complement
Small-scale test loop(s) capable of cold and hot operations.

Programmatic Connections

In order to be successful and relevant, the AFCI Safeguards Campaign must be closely coupled to other AFCI campaigns. For example, the Safeguards Campaign should be informed and engaged in the design of reprocessing plants so that the ability to safeguard a facility is an integral part of that design process, rather than an expensive afterthought. Some of the key inter-Campaign AFCI linkages identified by the SES team are described in this report.

It is also important that the work undertaken by the AFCI program be coordinated with the larger Safeguards community, most notably the NA-24 Next-Generation Safeguards Initiative and NA-22's new Safeguards portfolio. As an immediate example, the "High-Priority Near-Term Investment" scope described above is likely well beyond the resources of the AFCI Safeguards Campaign alone, but may be within the consolidated resources of AFCI, NA-24 or NA-22. In fact, some of the scope identified above is already funded by NA-24: A spent fuel assay evaluation framework and the Coupled End To End Demonstration (CETE) project at Oak Ridge National Laboratory. Ideally, the three DOE/NNSA programs will consolidate resources and planning to ensure long-term, and complementary, R&D support for these foundational activities.

One model for how the DOE Nuclear Energy AFCI program scope would be delineated from the NA-24 programs is described here. It is based on informal discussions at AFCI Safeguards Campaign Working Group meetings, and should not be taken to represent an official viewpoint.

New U.S. nuclear fuel cycle facilities (*e.g.*, reprocessing plant or MOX fuel fabrication facility) will be subject to domestic safeguards requirements levied by the DOE and the Nuclear Regulatory Commission. It is also assumed, that they will be IAEA Eligible Facilities and subject to IAEA safeguards requirements. These facilities, therefore, represent a unique opportunity for the US to lead the development and demonstration of next-generation safeguards technology concepts, and to provide real-world examples of how the interests of individual states and the IAEA can be met simultaneously. Some specific examples of how the AFCI Safeguards Campaign could complement the internationally focused NGSI program include:

- Demonstrate how a single process monitoring infrastructure, with suitable information barriers and data authentication practices, can be utilized by both the private operator of a nuclear facility, and the regulating agency. A shared infrastructure is a key enabler for improved detection of protracted diversion scenarios, but is also a considerable stumbling block for private enterprise keen on protecting proprietary process information;
- Analyze how the accountancy requirements of DOE, NRC and IAEA interact in practice (*i.e.*, which uncertainty and timeliness requirements are the most stringent at the head-end of a reprocessing plant), and how advanced technologies might be able to help meet all requirements simultaneously;
- Demonstrate how the concept of Safeguards by Design can be put to practice on a US facility, and that safeguards requirements can be written into the facility design requirements for all three governing bodies (DOE, NRC and IAEA);
- Provide facilities and materials needed for technology development and demonstration. The Coupled-End-To-End program is an example of how US infrastructure can be utilized to support technology development aimed at global problems; the DOE complex offers many more opportunities of the sort.

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Acronyms

3DLRF	3-Dimensional Laser Range Finder
AFCI	Advanced Fuel Cycle Initiative
AMUSE	Argonne Model for Universal Solvent Extraction
ASA-100	Advanced Safeguards Approaches
BWR	Boiling Water Reactor
C/S	Containment and Surveillance
CANDU	Canadian Deuterium Uranium reactor
COTS	Commercial-Off-The-Shelf
CZT	Cadmium-Zinc-Telluride (Cd-Zn-Te)
DD	Deuterium/Deuterium
DDT	Differential Die-Away Technique
DIV	Design Information Verification
DIV	Design Information Verification
DOD	Department of Defense
DOE NA-24	DOE Office of Nonproliferation And International Security
DOE	Department of Energy
DOE-NE	Department of Energy Office of Nuclear Energy
DT	Deuterium/Tritium
EMS	Electrochemically-Modulated Separations
ENDF	Evaluated Nuclear Data File
ENSDF	Evaluated Nuclear Structure Data File
FRAM	Fixed Energy, Response Function Analysis with Multiple Efficiencies
FWHM	Full-Width at Half Maximum
GADRAS	Gamma Ray Detector Response Analysis Software
GNEP	Global Nuclear Energy Partnership
HKED	Hybrid K-Edge Densitometry
HPGe	High-Purity Germanium
IAEA	International Atomic Energy Agency
ITU	Institute of Transuranium Elements
KED	K-Edge Densitometry
KMPs	Key Measurement Points
LIDAR	Light Detection and Ranging
LSDS	Lead Slowing-Down Spectrometry
LWR	Light Water Reactor
MA-MOX	Minor Actinide Bearing Mixed Oxide
MC&A	Materials Control and Accountancy
MCNP, MCNPX	Monte Carlo N-Particle Transport Code
MGA	Multiple Group Analysis gamma-ray spectroscopy software
MIP	Multi-Isotope Process
MOX	Mixed Oxide
NDA	Non-Destructive Assay
NGSI	Next-Generation Safeguards Initiative
NRC	Nuclear Regulatory Commission

NRF	Nuclear Resonance Fluorescence
PCA	Principal Component Analysis
PNAR	Passive Neutron Albedo Reactivity
PWR	Pressurized Water Reactor
R&D	Research and Development
RADSAT	Radiation Detection Scenario Analysis Toolbox
RFID	Radio Frequency Identification
SBM	Spectroscopy Based Monitoring
SES	Safeguards Enhancement Study
SINRD	Self-Interrogation Neutron Resonance Densitometry
SNM	Special Nuclear Material
TARIS	Thermal-Atomization Resonance Ionization Spectroscopy
TES	Transition Edge Sensors
TRL	Technical Readiness Level
UREX	Uranium Extraction
UV-Vis	Ultraviolet and Visible
UWB	Ultra-Wide-Band
VR	Vulnerability Review
XRF	X-Ray Fluorescence

Contents

Figures

Tables

1.0 Introduction

This report is the primary deliverable of the Advanced Fuel Cycle Initiative (AFCI) Safeguards Enhancements Study (SES). It documents the driving gaps and needs in the domestic and international safeguards community and provides summaries of over 30 promising technologies and processes that, if successfully developed and implemented, could significantly reduce the risk of proliferation in the nuclear fuel cycles of tomorrow. Also discussed are some of the key programmatic connections to other parts of AFCI and the U.S. Department of Energy (DOE) as they are important in the formulation of AFCI R&D priorities in the context of the larger safeguards R&D community.

2.0 Approach

The roadmapping approach adopted by the SES team is summarized schematically in Figure 1, along with an example of how that process is traversed. A recognized need and the corresponding performance targets provide the starting point. In the specific example of Figure 1, the need is the direct measurement of plutonium (Pu) in used fuel (as opposed to today's confirmatory methods that rely on operator-declared information and computations) while the preliminary requirement is quantification of Pu with an uncertainty lower than is achievable with today's indirect methods—approximately two percent on Pu mass. It should be noted that, in many cases, the roadmapping need is qualitatively clear, but assigning it quantitative requirements is difficult because, for example, AFCI facility designs are still in development and systems analysis tools that illuminate how instruments and methods interact in a safeguards program are not sufficiently mature to support quantitative requirements.

Once needs and performance targets were documented, the SES team solicited input from technologists at seven DOE National Laboratories, several universities and industry to create a list of candidate technologies that may offer a solution to those needs — some near-term and some much more exploratory. In this used-fuel assay example, two of the candidate technologies were passive neutron albedo reactivity and the lead slowing-down spectrometry technique. The team identified the defining potential and impact for each technology, as compared to competing technologies and/or a baseline technology from today. In order to give an objective picture for each method, key research questions that need to be addressed in order to assess viability and compare to today's baseline were identified, and research milestones that could support such a viability assessment were articulated.

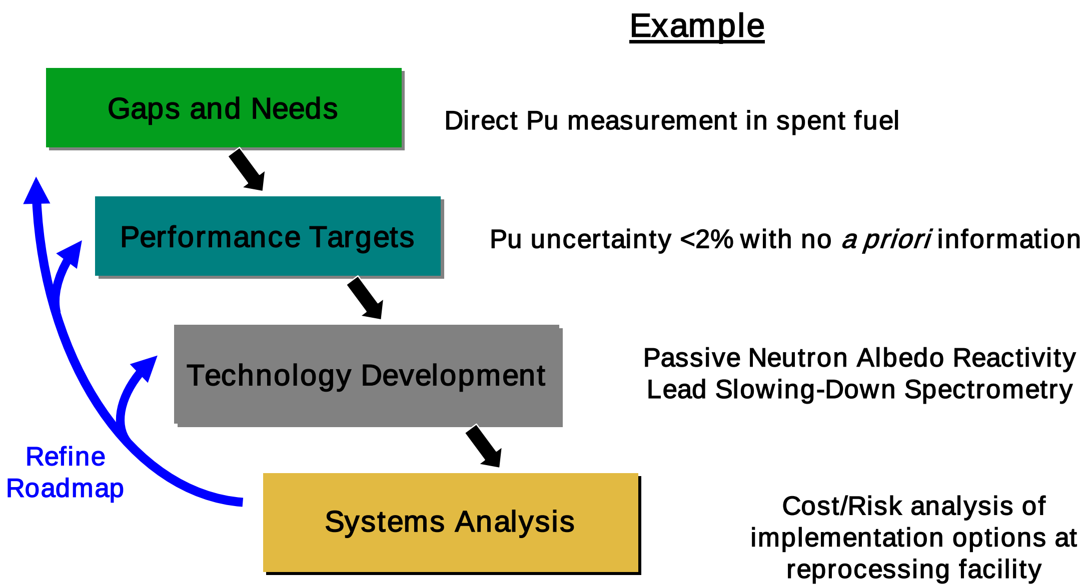

Figure 1. Overview of Safeguards Enhancement Study approach to technology roadmapping, and an example of path through that process.

The roadmap developed for AFCI Safeguards is a living document that will require updating as new information becomes available. For example, the performance targets called out in this document are preliminary and should be considered nominal starting values to be refined as technologies mature, and tools for assessing how those technologies interact (*e.g.*, Safeguards by Design) and come to fruition. The feedback loops that will make the roadmap a living document (*i.e.*, between Technology Development, Systems Analysis, Gaps and Needs) are also shown in Figure 1.

3.0 Gaps and Needs

The SES was founded on recognized gaps and needs in the international and domestic safeguards community. For example, the gaps and needs for AFCI Safeguards will often be the same as, or at least consistent with, those from recent NA-24 technology evaluation and strategy documents [Durst 2007a; Durst 2007b; Durst 2007c]. Because the emphasis of the AFCI Safeguards Campaign is the back end of the fuel cycle, particularly irradiated fuel, MOX fuel and aqueous reprocessing plants, this roadmap shares the same emphasis. Each of the identified needs is described below.

3.1 Direct Measurement of Pu in Used Fuel

The measurement of key isotopes in used nuclear fuel has been an ongoing challenge for materials control and accountability, both domestically and internationally. There are many motivations for accurate characterization of used fuel, including the following:

- Independent and timely verification of elemental plutonium content in used fuel at the head-end of reprocessing plants, as requested by the IAEA. “Independent” means that no operator-declared information is required as input to the assay process (*e.g.*, initial fuel composition, burn-up history, cooling time) and "timeliness" is related to the time it takes to convert diverted material to weapons components. The need for intact-assembly NDA measurements is likely to be even more acute for electrochemical reprocessing plants because there are apt to be no suitable measurement opportunities after the fuel enters the processing area (*i.e.*, no accountancy tank from which to draw samples that accurately reflect the total Pu mass in the fuel assembly). Note that IAEA's requirement for an *elemental* Pu total may differ from other used-fuel assay applications where, for example, fissile isotopic mass may be the primary concern.

- Shipper-Receiver accountancy as nuclear material leaves one nuclear facility (*e.g.*, cooling pond at a reactor) and arrives at another nuclear facility (*e.g.*, long-term dry storage or reprocessing plant). Such measurements help maintain continuity of knowledge on the used fuel assemblies as they move within the nuclear fuel cycle.
- Materials accountancy on used fuel that has cooled to the point that it is no longer considered "self-protecting" by regulation. As used fuel inventories stack up at U.S. nuclear power plants, for example, some of the older fuel assemblies will begin to fall under more strict accountancy measures. Methods are needed to measure those assemblies at their storage locations, and add their nuclear material content to the accountancy of the facility as required by applicable regulations.
- Burn-up credit to increase capacity of long-term storage locations. The criticality calculations that often dictate the storage capacity of used-fuel storage areas or geological repositories are typically based on conservative assumptions for burn-up, resulting in overestimation of criticality hazard and reduction in a facility's storage capacity. Nondestructive assay measurements that can accurately quantify the amount of fissile material in the used fuel, and perhaps even indicate the concentration of neutron poisons that could increase the burn-up credit, would be of great value.

State of Art

Current methods for estimating Pu mass in used-fuel assemblies are typically based on passive detection of radiation signatures from long-lived and easily measured non-fissile isotopes (*e.g.*, ^{137}Cs gamma-ray emissions or ^{244}Cm neutron emissions). These measurements are then coupled to burn-up-code calculations to create computational predictions of the isotopic inventory in each fuel assembly so as to infer Pu mass. While these passive confirmatory measurements may be appropriate in cases where the operator-declared information required for accurate burn-up calculations is trusted, as perhaps in domestic safeguards, they fall well short of the "direct and independent" goals of IAEA used-fuel verification and the desire for timeliness. Furthermore, the accuracy of the burn-up codes, for total Pu mass in typical light-water reactor fuel assemblies is generally 5-10 % due to uncertainties in irradiation conditions during the various reactor cycles, such as neutron fluence and energy spectrum in the core, axial variation along length of fuel, and uncertainties in cross-sections and isotopic production mechanisms used in the burn-up codes. Even further, the classic burn-up codes will likely be less accurate for emerging, advanced reactor types than they are for today's commercial reactors.

Based on IAEA and U.S. experience, today's indirect Pu mass measurement can attain total uncertainties of approximately 5-10 % on individual light-water reactor fuel assemblies, so long as the fuel has an ideal paper pedigree. Significantly higher values are typical for more exotic fuel types or if operator-declared information is less than ideal [Abhold 1998]. In high-volume storage or reprocessing facilities, even an optimistic uncertainty of 3% translates to "unaccounted Pu mass" of 500 kg or more per year [Menlove 2006].

The key R&D challenge in this area is identifying potential used-fuel NDA techniques, understanding their strengths and limitations, and coupling them in a way that can meet the performance objectives at key measurements points.

Performance Targets

- Pu measurement with total uncertainty <2% with no need for information from the facility
- instruments capable of integrating in typical fuel cycle operations (*e.g.*, measurements as assemblies are moved into dry storage at a reactor facility) and in various environments (*e.g.*, underwater and dry).

3.2 Direct, On-line Measurement of Pu in Reprocessing Streams

Traditional reprocessing operations generally involve an intermediate product of separated plutonium nitrate solution, with this solution being converted to plutonium oxide or a mixed uranium/plutonium oxide. Advanced fuel cycle concepts often advocate no separated plutonium, with intermediate and final product containing some level of other actinides. Just as the direct, independent and timely measurement of Pu in used fuel is critical to accurately "opening the books" at a reprocessing facility, measurements of Pu on the output streams of those plants is critical to closing the material balance.

State of Art

At present, there is very little direct, on-line measurement of Pu in the various streams of reprocessing plants. The vast majority of the Pu monitoring in the solution area of the plant is done by measurement of the level, density and temperature of the process tanks. This is supplemented by the analysis of samples taken from the tanks using a combination of chemical analysis, NDA and mass spectrometry. In the powder part of the process the nuclear material is monitored using neutron detectors arranged around the process gloveboxes that give a real time estimation of the Pu content of each box. The one-sigma uncertainty of this technique is typically greater than five percent. On-line measurements that improve the timeliness of processing solution accountancy are needed.

Performance Targets

- Protracted Diversion: Pu measurement with total uncertainty <1% or 1 SQ per month
- Abrupt Diversion: Immediate, real-time Pu measurement <5%.

3.3 Design Information Verification Techniques

Design Information Verification (DIV) is a tool used by the IAEA to verify that a facility is constructed and operated as declared by the State and operator of that facility. The State provides the IAEA with the facility's design information so that the IAEA can, as defined in the IAEA Safeguards Manual part SMR SMO 3.2, "identify the features of facilities and nuclear material relevant to the application of safeguards in sufficient detail as to facilitate verification of their design." The IAEA would like the information "as soon as the decision is taken to construct or to authorize the construction of, and to provide on an iterative basis further information on safeguards relevant features of facility design early in the stages of project definition, preliminary design, construction and commissioning." The IAEA performs DIV verification activities throughout the facility life cycle.

State of Art

The techniques for DIV have evolved over the years with more emphasis being put on DIV in the last decade and on the use of technology to assist inspectors in DIV activities. The process of DIV has two main steps: i) acquiring raw data from the plant; ii) comparing the "as-built" data and the reference model. There has been developmental work and deployment of technologies to use for DIV. The DIV activities previously relied on visual observation by inspectors to verify that the facility has not been altered or that the State and operator installed changes as they declared. In the case of gas centrifuge enrichment plants, the Low Frequency Unannounced Access inspections were DIV inspections where the inspectors verified that photographs taken by the operator and stored under seal showed the same cascade hall piping configurations at present as at the time of the original DIV photography of the cascade piping.

The IAEA has been testing the use of the 3-Dimensional Laser Range Finder (3DLRF). The 3DLRF can image certain areas within a facility and compile individual 3-D scan data to combine into a global 3-D reference model of the area. An inspector can then compare the reference 3-D model with the design drawings and photographs.

The DIV activities also can be used to detect the presence of undeclared design features and hidden facilities which could indicate undeclared nuclear activities or the diversion of nuclear material. From among several geophysical methods, the IAEA selected ground-penetrating radar as an approved technology for the detection of hidden objects and structures.

Performance Targets

- Develop technologies and processes that provide the IAEA the ability to immediately and unequivocally interpret the DIV data steams (*e.g.*, change detection algorithms for laser rangefinder images, radiation field mapping)
- incorporate radiation-based imaging systems as appropriate to improve DIV confidence.

3.4 Advanced Containment and Surveillance

Containment and surveillance (C/S) measures and monitoring complement facility safeguards approaches based on nuclear material accountancy. C/S measures and monitoring strive to permit the safeguards objectives to be achieved at acceptable costs and with minimum intrusion into routine facility operations.

State of Art

The IAEA uses the structural features of a facility, containers or equipment to establish the physical integrity of an area or items, including safeguards equipment or data, and to maintain the continuity of knowledge of the area or items by preventing undetected access to, or movement of, nuclear or other material, or interference with the items assured by seals or surveillance measures. The IAEA has several types of tags and seals. The most common seal is the metal E-cup seal attached to detect movement, tampering, and changes to nuclear material, data, facility equipment, and safeguards equipment integrity. The IAEA uses fiber optic seals to allow for *in-situ* verification of seals for certain applications. The

IAEA also uses electronic seals that can store data on opening and closing of the seal and even transmit the data remotely to the IAEA.

The IAEA uses surveillance to collect information through inspector and/or instrumental observation aimed at detecting movements of nuclear material or other items and to detect interference with containment or tampering with IAEA equipment, samples and data. The IAEA uses cameras to observe movements in many facilities. However, radiation detectors can also be used to monitor the movement of nuclear material such as the movements of used fuel into and out of a cask. Furthermore, cameras and radiation detectors can be used in concert to provide more information and to back each other up to maintain continuity of knowledge of nuclear material. Inspector observation may be used to monitor material transfers and to maintain continuity of knowledge where it is more expedient to use human surveillance for various reasons.

Between processes, it is anticipated that items will be canned and transferred through Key Measurement Points. To achieve material balance, an item must either be tagged and sealed or it must undergo NDA measurements. To save time and cost, for AFCI facilities, it is recommended that tags and seals be used to monitor containers as they pass through Key Measurement Points.

Performance Targets

Develop technologies and processes capable of near-real-time tracking of feed/product containers throughout the fuel cycle, including within individual facilities.

3.5 On-line Monitoring of Reprocessing Streams

The threat of protracted diversion of Pu from commercial reprocessing operations is a prominent concern to national and international agencies tasked with safeguarding these facilities. Ideally, process monitoring would be capable of directly measuring the flow of Pu through every stage within the reprocessing facility. However, as the previous discussion indicated, there is no current technology capable of achieving that goal.

Modern process control systems in a reprocessing plant use extensive instrumentation (*e.g.*, density gauges, flow meters, hatches opening and closing, container movements, radiation emissions) and data processing capabilities to condense information for the operating staff, to provide timely information on location and movement of materials in process for safety, quality control, and process control as well as to meet safeguards requirements. There is general agreement in the safeguards community that the proper integration of the various available parameters, such as radiation signatures, pH, and flow, coupled to state-of-the art statistical process control techniques, could significantly decrease the risk of protracted diversion scenarios enabled by subtle changes in plant chemistry or operating conditions. While these measures would not provide direct measurement of Pu concentration in the streams, they could provide sensitive anomaly detection to indicate that plant conditions are off-normal and that the risk of protracted diversion has increased.

The key R&D challenge in this area is identifying the relevant process monitoring signatures, understanding their strengths and limitations, and coupling them in a statistical process control framework (see below) that sufficiently distills the large volume of data into a form that allows the regulating body such as IAEA to make decisions regarding proliferation risk.

State of Art

Current practice is to build confidence against Pu removal from the system by monitoring flows/transfers of solutions through the main process. That is, the accountancy is based on volumetric flow that assumes constant Pu concentrations. While some solution characteristics, such as density, may be used to confirm flow estimates, that is not widespread. The operator of the reprocessing plant, with full access to all process monitoring signals and data, typically has much more robust anomaly detection capabilities than do the inspectors focused on safeguards.

Performance Targets

- User Interface: Near-real-time visualization, analysis and reporting
- Data Authentication: Develop and implement guidelines to support a new generation of remote, on-line facility monitoring technologies
- Protracted Diversion: Ability to detect Pu concentration anomalies of <1% or equating to 1 SQ per month
- Abrupt Diversion: Ability to detect Pu concentration anomalies of <5%, in real time.

3.6 Rapid laboratory sample analysis

As much of the previous discussion has indicated, the accuracy of today’s nondestructive measurements is generally not sufficient to meet materials accountability requirements. Consequently, conventional laboratory-based destructive analysis continues to be the cornerstone of materials balance estimates for nuclear fuel cycle facilities. While destructive analysis can offer excellent accuracy and precision in many cases, it may require days to weeks to get results.

Faster automated methods could support on-site laboratories and possibly move destructive analysis methods from the laboratory to on-line monitoring applications. Advances in radiochemical automation offer many opportunities to address needs in nuclear reprocessing and safeguards. Automation can reduce time, costs, and worker doses while improving safety in existing measurements, and also enable measurements not currently considered feasible because, for instance, they may be too difficult to put on-line or too costly and time-consuming for manual laboratory analysis.

State of Art

Destructive analysis is normally carried out in fixed laboratories using manual sample preparation, separation, and measurement techniques. For mass spectrometric measurement methods, the analyte must be separated from matrix components that may generate interferences.

Performance Targets

Significant reduction in time, cost and manpower associated with laboratory sample analysis in support of materials accountancy at reprocessing plants.

3.7 System Analysis Tools for Safeguards Evaluation and Optimization

Three decades have passed since the United States has designed a reprocessing plant, and existing reprocessing plants throughout the world may use dated technology. For this reason, new safeguards analysis tools are required for both the design of a future plant and the optimization of safeguards measures at existing plants — all with the goal of reducing the overall proliferation risk at the facility. The great value of systems-level analyses is that they can identify the most critical points in a materials accountancy program, and therefore, help guide prioritization of R&D agendas. For example, this roadmap describes a number of needs and associated performance targets. While those needs and targets are grounded in community experience, the way in which they interact to create the entire "Safeguards Envelope" for a facility is not yet clear. For example, if the accuracy of a process-monitoring technique improves dramatically, how does that impact the requirements for direct measurement of Pu in used fuel at the head end of the reprocessing facility? Properly designed systems analysis tools that incorporate the ideas of probabilistic risk analysis (PRA), physical protection (PP) and measurement uncertainties (both statistical and systematic) are needed to evaluate options for both new and existing facilities.

State of the Art

Accurate modeling of nuclear fuel cycle systems and the associated risk of diversion scenarios is a daunting task because it requires accurate representation of each constituent component (*e.g.*, a physical protection system or a specific measurement instrument), as well as the relationships between all components, including how uncertainty and risk propagate. While many of the pieces of a comprehensive system modeling program are available in the community, the integration into a common framework focused on safeguards metrics has not been realized (analogous to the framework utilized in PRA for nuclear reactor design).

Performance Targets

High-fidelity integrated safeguards performance modeling that

- integrates Proliferation Resistance and Physical Protection functionality and metrics
- provides discrete-even and dynamic facility simulation
- utilizes independently peer-reviewed risk/cost metrics
- includes benchmarking and blind testing using available data from actual plants.

4.0 Candidate Technologies

This section provides brief overviews of the various candidate technologies put forward by the collective experience and expertise of the study team. This list of candidate technologies spans the continuum of technical maturity from highly-exploratory concepts to incremental improvements or adaptations of the tried-and-true. Both are important to a long-range R&D plan for nuclear fuel-cycle safeguards; the former represents the building blocks that enable the creation of new and improved methods on longer time scales, while the latter can support near-term enhancements at fuel cycle facilities. These overviews are intended to highlight

- The defining technical characteristic that separates these technologies from today's baseline approaches, or other proposed approaches
- previous work in the area, and information on technical maturity
- potential impact
- relevance to Safeguards Needs outlined earlier in this report
- key research questions to be addressed
- Suggested milestones (mapped to research questions) and estimated timelines to prototype demonstrations. Note that estimated timelines assume that full funding is available to the development teams.

The candidate-technology summaries are grouped under the same major headings used for the Needs discussion, which is also the set of headings used to organize the AFCI Safeguards Campaign as a whole: Advanced Instrumentation, Advanced Control and Integration and Safeguards by Design.

4.1 Advanced Instrumentation

4.1.1 Lead Slowing-Down Spectrometry

The Lead Slowing-Down Spectrometry (LSDS) method is an active interrogation approach that taps a relatively unused dimension of nondestructive analysis: the resonance structure of fission cross-sections in special nuclear-material isotopes. The defining potential of the LSDS method is the direct measurement of fissile isotopic mass (*e.g.*, ^{239}Pu and ^{235}U) in used fuel in a nondestructive, timely fashion that requires no operator-declared information about the fuel assembly.

LSDS fuel assay begins when a pulse of interrogating high-energy neutrons is introduced into a large pile of lead. The characteristics of elastic neutron scattering in the lead pile result in "energy focusing" that produces a population of neutrons whose mean energy decreases in a predictable way, with a relatively narrow variance about the mean energy. When used fuel is loaded into the interrogation chamber located in the lead pile, the interrogating neutron population induces fission in the fissile isotopes of the fuel. Since the fissile materials have large fission cross sections that vary significantly with energy (particularly in the epithermal resonance region), the time-dependent fission neutron production rate will show a distinct signature shape for each fissile isotope (see Figure 2). This fission neutron production rate is measured by threshold fission chambers, and with appropriate analysis methods that assay signal can be used to calculate the absolute mass of fissile isotopes in the assembly. Signal extraction methods that accurately account for the non-linear effects of interrogating neutron self-shielding are a particular challenge for large matrices of used fuel, for example the typical commercial power reactor assembly.

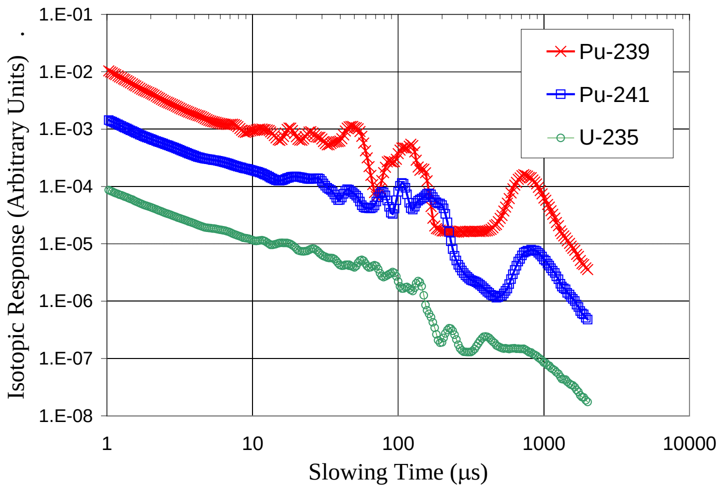

Figure 2. Examples of LSDS response functions for three different fissile isotopes.

LSDS was first explored for the assay of naval reactor fuel in the 1970s by Rensselaer Polytechnic Institute and in the same time frame for the assay of fabricated fuel pins at Karlsruhe [Krinninger 1974]. Interestingly, it has also been utilized in nuclear cross-section measurements over the last several decades. More recent work has revisited the idea both empirically [Abdurrahman 1993] and via simulation [Smith 2002; Smith 2007] for various fuel-assembly types. Recent studies have confirmed the promise of LSDS for direct measurement of ^{235}U and ^{239}Pu in intact light-water reactor used fuel assemblies [Ressler 2008].

The previous work on LSDS — mainly initial viability studies — while encouraging, were not comprehensive enough to fully characterize the strengths and shortcomings of an LSDS-based fuel assay system for several reasons. First, the studies of full-assembly fuel assay were based largely on simulation and lacked suitable empirical validation. Second, the signal extraction methods utilized for wringing isotopic information from the LSDS assay signal were relatively crude — more sophisticated methods are expected to improve the accuracy and precision of fissile isotope mass estimates by accounting for self-shielding effects. Finally, previous work made assumptions about the availability of key LSDS components (*e.g.*, high-intensity pulsed neutron sources and fission chambers lined with highly pure ^{238}U) that have not been supported by testing and evaluation.

Potential Impact
Nondestructive, timely, <u>direct</u> measurement of fissile-isotopic mass (*e.g.*, ^{239}Pu and ^{235}U) without the need for operator-declared fuel information.

Needs That Could be Addressed if Successfully Developed
Direct Pu measurement in used fuel
Direct Pu measurement in actinide-bearing materials

Research Questions
1. Can signal analysis methods quantify fissile isotopics without *a priori* fuel information, using only measured observables?
2. Can simulation and modeling methods accurately capture the degrading subtleties in realistic fuels?
3. Can ultra high-purity fission chambers be developed and fabricated at reasonable cost?

4. What is the accuracy, precision and mass-defect sensitivity for a nominal LSDS design and PWR fuels?
5. How can LSDS be integrated with other NDA techniques and/or burn-up codes to quantify total Pu?
6. What is the achievable performance for other fuel types, including MOX and advanced reactors?

Milestones	Estimated Timeline
Demonstrate assay signal analysis methods meeting performance objectives (Q1, Q4)	1 year
Full-fidelity modeling of realistic used fuel “library” and virtual assay campaign (Q2, Q4)	1 year
Empirical benchmarking, instrumentation testing at existing LSDS facilities (Q2, Q3)	2 years
Complete analysis to integrate LSDS with burn-up codes and other NDA techniques. (Q5)	2 years
Decision point: Adequate virtual performance motivates full-scale prototype?	3 years
Design and construction of prototype capable of assaying used fuel assemblies	4 years
Completion of empirical measurement campaign, prototype demonstration (Q4, Q5,Q6)	5 years

4.1.2 Nuclear Resonance Fluorescence

Nuclear resonance fluorescence (NRF) is an active interrogation approach that provides isotope-specific signatures that could be used to assay the isotopic content of a sample. NRF is not limited to fissile materials, unlike fission-inducing techniques. In addition, the photon energies involved are in the MeV range, allowing for significant penetration through material. The typical NRF signature is a set of strong, narrow (~1 eV before detector-resolution broadened) peaks. These peaks permit a very simple analysis to determine the isotopic content of the sample without operator-declared information of the sample.

NRF is stimulated by directing a beam of high-energy (~3 MeV), collimated photons at a target material. At certain resonant energies, nuclei strongly absorb and re-emit photons of the same or lower energy. The frequencies at which the NRF process occurs are unique to each isotope, and that unique fingerprint can be obtained either by measuring the energies of the scattered photons or by detecting the absorption of photons at the resonant frequency for the isotope of interest [Bertozzi 2005; Pruet 2006].

The physical process behind NRF has been understood for more than 50 years [Metzger 1959] but most work since then has utilized NRF as a tool to understand nuclear structure. Only in the last few years has NRF been applied to isotopic characterization on bulk materials, particularly under Department of Homeland Security funding for the screening of containerized cargo [Bertozzi 2005].

NRF measurements can be conducted in two basic configurations. For scattering measurements, the photons directly scattered off a sample are analyzed. For transmission measurements, the absorption by the sample material at the resonance energies is detected by analysis of the photon beam that passes through the sample. Both approaches have advantages and disadvantages for particular applications.

NRF holds promise for safeguard applications because the technique has the potential to directly determine the presence and concentration of actinide isotopes in a variety of sample matrices. However, the performance of NRF in a variety of complex samples has not been quantified. Bertozzi gives an example of a small WGPu sample encased in a steel container [Bertozzi 2005]. The capabilities and limitations of using NRF for specific AFCI applications would need further study. The simulation and prediction capabilities have not been fully tested against numerous datasets. Another issue is that while some NRF signatures of ^{232}Th, ^{235}U, ^{236}U, ^{238}U and ^{239}Pu are known [Bertozzi 2005; Warren 2007; Kniesel 2006], the NRF signatures of other, unlikely but potentially interfering, actinides are not. The next steps in assessing NRF viability for Safeguards applications include the collaboration with Domestic Nuclear Detection Office and DOE NA-22 projects for basic isotopic signature characterization, including the isotopes that are key to international safeguards applications (*e.g.*, ^{238}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu). Proof-of-principle simulations or measurements for NRF assay in realistic safeguards, such as used fuel assay or waste-product characterization can then be performed.

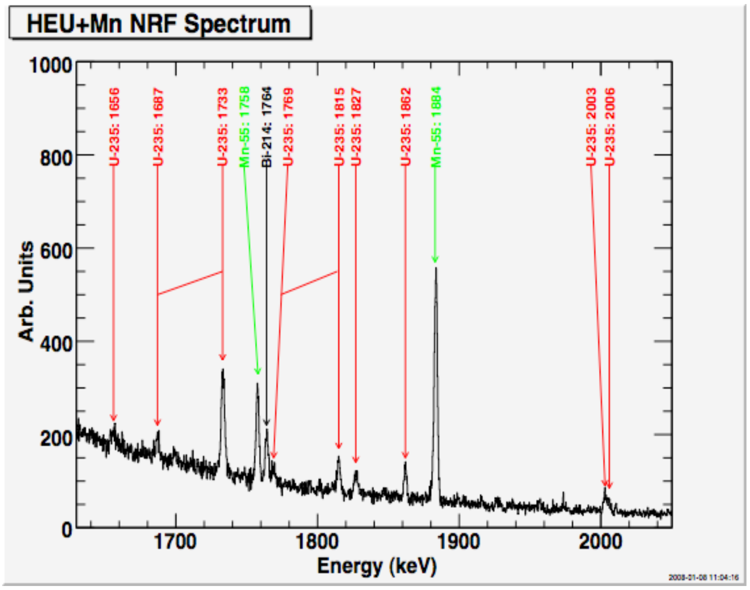

Figure 3. Measured NRF response of U-235 (red arrows) and Mn-55 (green arrows). The Bi-214 peak (black arrows) derives from natural background.

Potential Impact
Nondestructive, timely, <u>direct</u> measurement of isotopic mass (e.g. fissile and actinides) without the need for operator-declared information about the fuel assembly or fuel process.

Needs That Could be Addressed if Successfully Developed
Direct Pu measurement in used fuel
Direct Pu measurement in actinide-bearing materials
Direct measurement of Pu in reprocessing streams

Research Questions
1. What are the cross-sections and emissions energies of NRF signatures for the Pu isotopes and other actinides? Are there interferences with key signatures? Are there resonances at other energies which offer better contrast for fuel assemblies or fuel processing streams?
2. Can the measurement setup be designed so that the performance of the high purity germanium detector is not adversely affected in the radiation field of the used fuel? What are dominant backgrounds in safeguards applications?
3. Do the resonance parameters and the depth of penetration of the incident beam adequate for partial defect detection? What resonance and beam characteristics are optimal?
4. Which measurement configuration, scattering or transmission, is more likely to be successful for this application?

5. Do empirical measurements at existing NRF facilities, on unirradiated fuel samples, validate the high-fidelity modeling methods being used to extrapolate to irradiated fuel performance?
6. Do simulation and modeling methods accurately capture the variations in assay signal from used fuel of varying burn-ups?
7. What is the expected accuracy and precision for a prototype system? How is the accuracy improved with different source and detector technologies? What standards or benchmarks need to be established for high precision measurements?
8. What detector technologies can be developed to enhance the sensitivity of NRF assay techniques?
9. How does the technology work within the existing safeguards architectures? What are the footprint, cost, and safety issues for deployment? Can the system be fooled and material diverted?

Milestones	Estimated Timeline
Measure cross-sections and signature characteristics for Pu isotopes and actinides (Q1)	2 years
Demonstrate detector performance in high radiation field (Q2)	1 year
Evaluate impact of skin depth of NRF for assay (Q3)	2 years
Empirical tests of measurement configuration (scattering and transmission) for mock fuel assembly (Q4)	2 years
Empirical benchmarking at existing NRF facilities (Q5)	3 years
Full-fidelity modeling of realistic used fuel “library” and virtual assay campaign (Q6, Q7)	3 years
Decision point: Adequate virtual performance motivates full-scale prototype?	4 years
Design and construction of prototype capable of assaying used fuel assemblies (Q7,Q9)	4 years
Completion of empirical measurement campaign and prototype demonstration (Q7,Q9)	5 years

4.1.3 Advanced Neutron Multiplicity

Neutron Multiplicity Measurements has been a workhorse of material accounting for many years [Ensslin 1998]. High-efficiency thermal neutron ^{3}He well-counters have been constructed with efficiencies as high as ~67%. Primarily they have been used to quantify the mass of plutonium. Looking forward, there are two primary paths for significantly improving neutron multiplicity: (1) improving ^{3}He-based systems by using list-mode data acquisition and novel electronics, and (2) use an array of liquid scintillating detectors that are capable of detecting neutrons on a time scale around 1000 times faster than ^{3}He-based technology.

List-mode data acquisition allows ^{3}He-based systems to handle significantly higher count rates. Historically, multiplicity counting was accomplished by feeding all the signals from all the ^{3}He tubes into one electronic package. The upper count rate which could be measured was limited by that which this electronic package could handle. With list-mode, this is no longer true. The data from each ^{3}He tube can be stored independently and then combined in post-processing. As a result the count rate of the system can be increased by over an order of magnitude. In addition to list-mode, the electronic processing of the data downstream of the tubes has received little research in the safeguards arena during the past decades and is ripe for improvements.

The liquid scintillating technology advances multiplicity counting by detecting neutrons on a much faster time scale. This improves results since it reduces the background, or accidentals, significantly. With ^{3}He-based systems, the neutron thermalization time is tens of microseconds; with liquid scintillating detectors, time scales of nanoseconds are possible. A detector system using liquid scintillators has been built on the order of 1m^2 with a data acquisition system that is tuned and ready to measure neutrons and gamma-rays from nuclear material [Dougan 2007] (see Figure 4). The DAQ has a 200MHz clock that allows for nanosecond arrival timing and full digitization of the signal pulses that results in >10^5 photon rejection for MeV neutrons. Liquid scintillators are sensitive to fast neutrons (indeed it is the momentum transfer from an incoming neutron to a recoiling proton in the scintillator that allows detection) and the scintillator response time allows nanosecond timing which permits measurement of individual fission (or fission-chain) events. Liquid scintillators also preserve a measurement of the neutron momentum, which helps differentiate fission from alpha-n sources, which can aid in the determination of Pu mass and determine the value of alpha. In certain measurement context, liquid scintillators can provide a much more sensitive measurement than is possible with ^{3}He detectors, and may lead to the ability to separate different sources of fission (*e.g.*, ^{240}Pu from ^{244}Cm).

Potential Impact
Nondestructive, timely neutron measurement for detection of Pu in used fuel or actinide-bearing materials

Needs That Could be Addressed if Successfully Developed
Direct measurement of used fuel
Direct measurement of Pu in actinide-bearing materials

Research Questions
1. In the context of spent and other actinide-bearing materials, what would a practical measurement system look like given (1) the need to suppress the incident gamma flux to the detectors and (2) the need to handle the expected neutron count rate? What would the efficiency be of such a system?
2. In the context of liquid scintillators, how are the results affected by multiple scattering?
3. Is it possible to separate different sources of fission (*e.g.*, ^{240}Pu from ^{244}Cm) for practical measurements situation of interest to safeguards?
4. How might the fast time response of liquid scintillators enable new NDA techniques in both the passive and active context?

Milestones	**Estimated Timeline**
Simulate use of both detector for used fuel and other actinide-bearing materials. Make critical measurements to determine system effectiveness. (Q1)	2 year
Develop more sophisticated algorithms for photon and neutron time-correlated signals. (Q2)	1 year
Simulate use of both detector for used fuel and other actinide-bearing materials. Make critical measurements to determine system effectiveness. (Q3)	2 year
Perform relevant simulations and measurements to explore system performance of novel NDA approaches. (Q3, Q4)	3 years

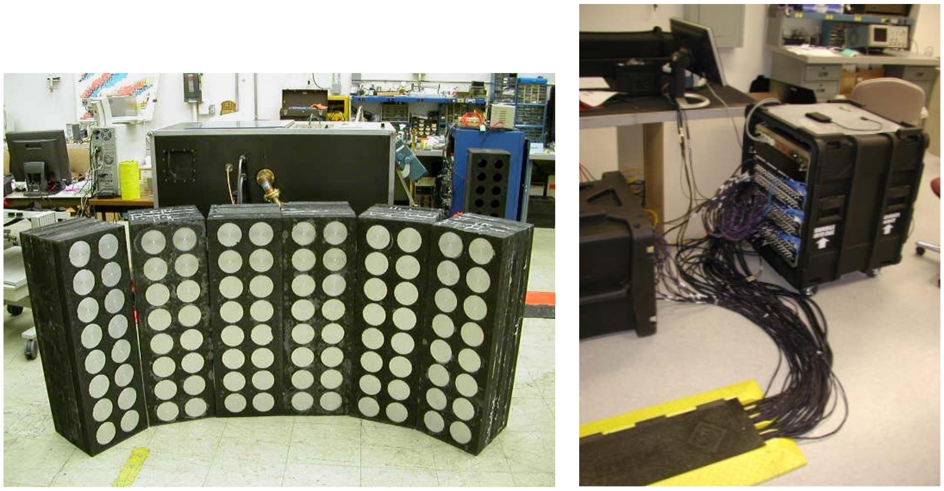

Figure 4. Liquid scintillators (left); data acquisition system (right).

4.1.4 Passive Neutron Albedo Reactivity

The Passive Neutron Albedo Reactivity (PNAR) measurement technique is a passive measurement approach that quantifies the fissile content (weighted sum of ^{235}U, ^{239}Pu, and ^{241}Pu) emphasizing the presence of ^{239}Pu and ^{241}Pu. The technique, depicted in Figure 5 below, uses neutrons that are inherently emitted by actinides in the used fuel to interrogate the fuel. Two distinct measurements of the used fuel are made. The primary difference between the two measurements is the neutron energy and fluence in the used fuel. By varying the material around the used fuel – removing the cadmium for example – high and low neutron-energy-measurement configurations can be produced. The ratios of the count rates from these two separate measurements, known as the cadmium ratios, obtained for these two measurements scale with the fissile content. The cadmium ratio can be determined with either the total or coincident neutrons [Lee 1982; Menlove 1997; Tobin 2006].

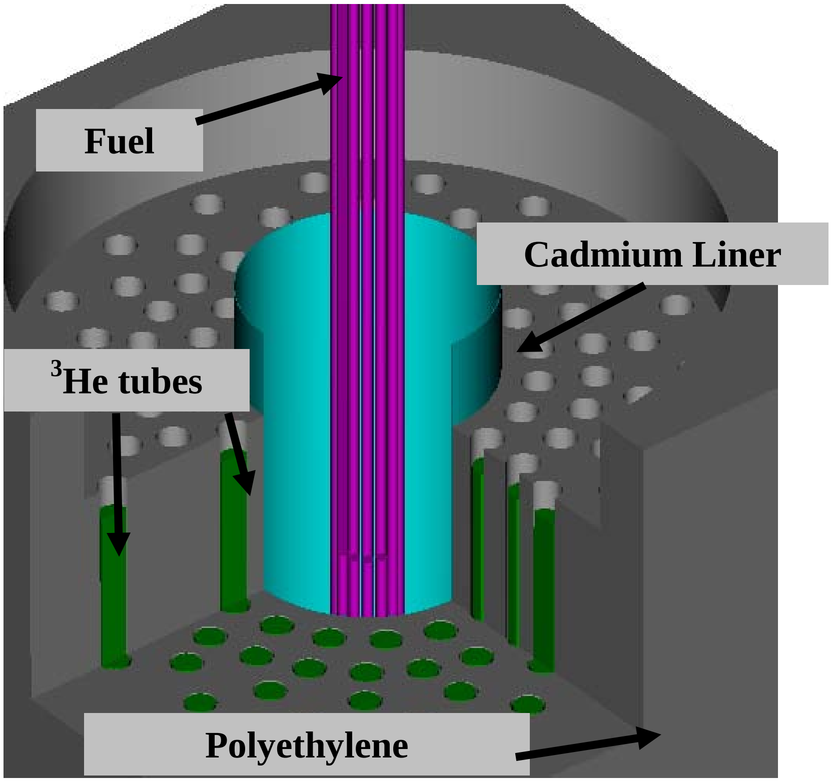

Figure 5. Schematic of PNAR instrument, including the cadmium liner that is central to discrimination of the various fissile isotopes.

As with all NDA techniques proposed for use with used fuel, PNAR will need to be integrated with other NDA techniques and/or burn-up codes to determine elemental Pu.

A PNAR instrument can be very similar to a well counter with ^{3}He tubes, polyethylene, and lead (to reduce the gamma dose to the tubes); or it could be very light (~25 kg) and relatively inexpensive if fission chambers are used with water as the moderating material. The ^{3}He based system will be able to detect coincident and total neutrons, whereas the fission chamber based system will only detect total neutrons. The cadmium ratio, determined with either singles or doubles, will vary with fissile content; it is suspected that a system based on doubles counting will be more accurate, but this needs to be verified experimentally.

The hardware used with PNAR is mature. The analysis is moderately mature; experiments using single fuel rods and a bundle of fuel rods have been performed. This technique will benefit from the development of list-mode data analysis.

Potential Impact
Nondestructive, timely, direct measurement of fissile-content (^{239}Pu + ^{241}Pu + ^{235}U)

Needs That Could be Addressed if Successfully Developed
Direct Pu measurement in used fuel
Direct Pu measurement in actinide-bearing materials
Advanced containment and surveillance technique

Research Questions
1. How to design the detector to maximize sensitivity for the three measurement situations of interest (water, borated water, air)?
2. How well will PNAR perform given the wide range (burn-up, initial enrichment, cooling time) of fuel in existence? How well will PNAR perform in various diversion scenarios?
3. How well does PNAR, in combination with burn-up codes and other NDA techniques, quantify the Pu content in used fuel?
4. How well does PNAR work with actual used fuel?
5. How well can PNAR measure various fuel types: spent MOX recycle, BWR, VVER, CANDU, etc.? How well can PNAR measure electrochemical (pyro) product? How is the accuracy of PNAR impacted by the non-isotropic nature of reactors (assemblies next to control rods, on the edge of the reactor, etc.)?

Milestones	Estimated Timeline
Complete Monte Carlo research to optimize the PNAR design (Q1)	1 year
Complete Monte Carlo research to quantify the capability of PNAR for fuel with a range of burn-up, initial enrichment, cooling time. Investigate the capability of PNAR in water, borated water, air. Investigate the capability of PNAR for several diversion scenarios. (Q2)	2 year
Complete integration effort to combine Monte Carlo results of PNAR with burn-up codes and other NDA techniques (Q3)	2 years
Decision point: Adequate virtual performance motivates full-scale prototype?	2 years
Design and construction of prototype capable of assaying used fuel assemblies	3 years
Completion of empirical measurement campaign and prototype demonstration (Q4)	4 years
Complete Monte Carlo research to quantify the ability of PNAR to measure various fuel types: spent MOX recycle, BWR, VVER, CANDU, etc. and electrochemical (pyro) product? Quantify the impacted of non-isotropic nature of reactors (assemblies next to control rods, on the edge of the reactor, etc.) on accuracy. (Q5)	4 years

4.1.5 Self-Interrogation Neutron Resonance Densitometry

Self-interrogation neutron resonance densitometry (SINRD) is a passive measurement approach that aims to quantify the mass of ^{239}Pu and ^{235}U in used fuel and other actinide bearing materials. The technique functions by measuring the absence of neutrons resulting from the fuel's absorption of these neutrons at specific resonances in the fission cross-sections of fissile isotopes. More specifically, in the case of ^{239}Pu, the ratio of the count rate recorded in a ^{239}Pu fission chamber to the count rate in a neutron flux monitor is inversely proportional to the ^{239}Pu concentration. The ^{235}U concentration can be measured in a similar manner, and simultaneously, using ^{235}U fission chambers. A preliminary design of the SINRD method is given below.

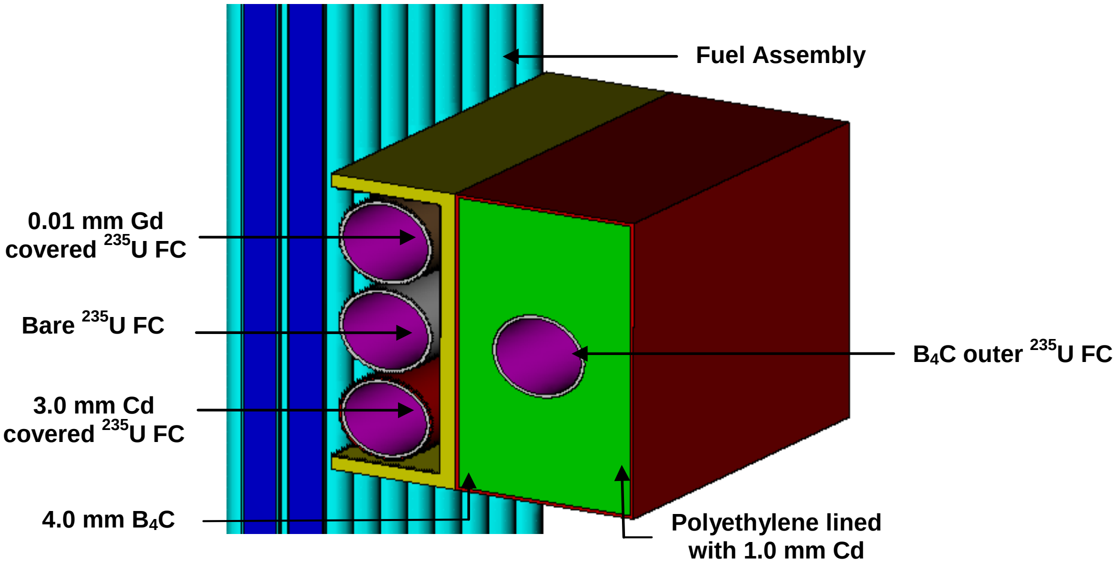

Figure 6. Schematic of SINRD instrument showing an array of fission chambers surrounded by foils with different neutron-absorbing characteristics.

The unique resonance structure in the fission cross-section of both fissile isotopes is isolated by positioning absorber filters around the fission chambers. In the example illustrated above Gd and Cd are used to alter the energy spectrum of the detected neutrons. The sensitivity of this technique is strengthened by using the same fissile materials in the sample and in the fission chamber because the attenuating effect of the resonance absorption lines in the transmitted flux is amplified by the corresponding detection resonances in the fission chamber.

As with all NDA techniques proposed for measuring used fuel, SINRD will need to be integrated with other NDA techniques and/or burn-up codes to determine elemental Pu. If SINRD were to be integrated with a burn-up code only, the ratio between the ^{239}Pu and total Pu would need to be determined. This ratio is largely a function of burn-up and, to a lesser degree, initial enrichment and cooling time. As such, signatures that measure burn-up, such as total neutron counting, can be integrated with SINRD.

A SINRD measurement system is a very light instrument that works better as the intensity of the neutron emission from the fuel increases. This is because singles neutrons are counted and fission chambers are immune to gamma ray intensity. The technique will emphasize the fuel rods that are closest to the surface of the fuel assembly and not give equal response to the interior.

The maturity of SINRD is low. Some related experiments using single fuel rods were performed [Menlove 1969]. Detector design through modeling has been the primary focus of recent research; this research indicates that less than 1% precision can be obtained in less than 100 s for ratios of the count rates in the fission chambers. These ratios scale with the mass of ^{239}Pu and ^{235}U; the ability of a SINRD system to discern the mass of each of these isotopes when they are mixed is a subject of ongoing research. Further studies with realistic isotopic concentrations in the assemblies are needed to address accuracy. The ability to detect the diversion of pins depends significantly on the location of the pins [LaFleur 2008].

Potential Impact
Nondestructive, timely, direct measurement of fissile-isotopic mass (*e.g.*, ^{239}Pu and ^{235}U) without the need for operator-declared information about the fuel assembly

Needs That Could be Addressed if Successfully Developed
Direct Pu measurement in used fuel
Direct Pu measurement in actinide-bearing materials
Advanced containment and surveillance technique

Research Questions
1. How well can the fissile content in the interior of the assembly be measured (penetrability)?
2. What is the accuracy, precision, and mass-defect sensitivity for ^{239}Pu and ^{235}U for typical PWR burn-ups?
3. How can SINRD be integrated with other NDA techniques and /or burn-up codes to quantify total Pu?
4. What is the expected performance of SINRD in water, borated water, and air?
5. What is the expected performance for other fuel types (*e.g.*, MOX recycle, BWR, VVER, CANDU)?

Milestones	Estimated Timeline
Complete Monte Carlo modeling to predict ^{239}Pu and ^{235}U accuracy, precision and mass-defect for nominal SINRD design and range of PWR burn-ups (Q1, Q2, Q4)	1 year
Complete analysis to integrate SINRD with burn-up codes and other NDA techniques (Q3)	2 years
Decision point: Adequate virtual performance motivates full-scale prototype?	2 years
Design and construction of prototype capable of assaying used fuel assemblies	3 years
Completion of empirical measurement campaign and prototype demonstration	4 years
Complete Monte Carlo modeling to predict performance for other fuel types (Q5)	4 years

4.1.6 Differential Die-away Technique

The differential die-away technique (DDT) is an active measurement approach that quantifies the fissile content (weighted sum of ^{235}U, ^{239}Pu, and ^{241}Pu) emphasizing the presence of ^{239}Pu and ^{241}Pu. A DDT measurement begins with the burst of 14 MeV neutrons produced by a neutron generator. The primary detector only counts neutrons above 1 eV. Data is only collected after the neutrons from the neutron generator have moderated to below 1 eV in energy. Hence, the data is collected after the neutrons from the generator are thermal. The detected neutrons are therefore from prompt fissions induced by the thermal neutrons originating from the neutron generator burst. Since the average energy of induced fission neutrons is greater than 1 MeV, these are the only neutrons counted by the detectors. A 100-Hz repetition rate is representative for a DDT system [Caldwell 1983; Jordan 2007b; Jordan 2007a; Hollas 1997].

The signal from ^{239}Pu and ^{241}Pu is stronger than that from ^{235}U. This is due to the combination of (1) the larger thermal cross section of ^{239}Pu and ^{241}Pu as compared to that of ^{235}U (thermal cross sections of ^{235}U, ^{239}Pu, and ^{241}Pu are 586 barns, 748 barns, and 1013 barns, respectively) and (2) the larger induced fission multiplicity of ^{239}Pu and ^{241}Pu as compared to that of ^{235}U (induced thermal fission multiplicities are 2.9, 2.8, and 2.4 for ^{239}Pu, ^{241}Pu, and ^{235}U, respectively). If commercial-grade Pu, with five times as much ^{239}Pu as ^{241}Pu, is assumed, the effective Pu thermal cross section is 792 b. Hence, 1.62 times more prompt neutrons are emitted from ^{239}Pu and ^{241}Pu than from ^{235}U per induced thermal fission. As with all NDA techniques proposed to date for measuring Pu in used fuel, DDT will need to be integrated with other NDA techniques and/or burn-up and neutron transport codes to determine elemental Pu. The greater sensitivity to fissile-Pu content may be important in this integration effort.

Since the DDT involves interrogating the fuel with thermalized neutrons, self-shielding is a concern. However, it is expected that neutron multiplication will propagate neutrons to the center of the fuel assembly. Hence, the presence of neutron multiplication will effectively compensate for self-shielding because the neutrons that are lost to the self-absorption process will create additional neutrons from the fission reaction.

The use of DDT techniques in air alone has been demonstrated successfully. The use of DDT techniques in air, water and borated water needs to be investigated. For all three situations, the strength of the neutron generator needed is above what is commercially available; hence research in neutron generator development is needed. Furthermore, the presence of water in and around the fissile material is also unique for DDT; the self-shielding effect of this needs investigation.

Potential Impact
Nondestructive, rapid, direct measurement of fissile-content (^{239}Pu + ^{241}Pu + ^{235}U) emphasizing the ^{239}Pu and ^{241}Pu fissile contents

Needs That Could be Addressed if Successfully Developed
Pu measurement in used fuel
Pu measurement in actinide-bearing materials

Research Questions
1. How to design the detector to maximize sensitivity for the three-measurement configurations of interest (water, borated water, air)? What is the extent of self-shielding in each of the three measurement situations? How strong of a neutron generator is needed?
2. How well will DDT perform given the wide range (burn-up, initial enrichment, cooling time) of fuel in existence? How well will DDT perform in various diversion scenarios?
3. How well does DDT, in combination with burn-up codes and other NDA techniques, quantify the Pu content in used fuel?
4. How well does DDT work with actual used fuel? What is the design of the neutron generator that can produce the necessary neutron intensity?
5. How well can DDT measure various fuel types: spent MOX recycle, BWR, VVER, CANDU, etc.? How well can a delayed neutron measurement system quantify Pu in an electrochemical (pyro) product? How is the accuracy of DDT impacted by the non-isotropic nature of reactors (assemblies next to control rods, on the edge of the reactor, etc.)?

Milestones	Estimated Timeline
Complete Monte Carlo research to optimize the DDT design, quantify self-shielding and determine the neutron generator strength (Q1)	1 year
Complete Monte Carlo research to quantify the capability of DDT for fuel with a range of burn-up, initial enrichment, cooling time. Investigate the capability of PNAR in water, borated water, air. Investigate the capability of DDT for several diversion scenarios. (Q2)	2 year
Complete integration effort to combine Monte Carlo results of DDT with burn-up codes and other NDA techniques (Q3)	2 years
Decision point: Adequate virtual performance motivates full-scale prototype?	2 years
Design and construction of prototype capable of assaying used fuel assemblies. Included in this design is a neutron generator capable of delivering the necessary neutron intensity.	3 years
Completion of empirical measurement campaign and prototype demonstration. (Q4)	4 years
Complete Monte Carlo research to quantify the ability of DDT to measure various fuel types: spent MOX recycle, BWR, VVER, CANDU, etc. and electrochemical (pyro) product? Quantify the impacted of non-isotropic nature of reactors (assemblies next to control rods, on the edge of the reactor, etc.) on accuracy. (Q5)	4 years

4.1.7 Delayed Neutron Detection

The delayed neutron measurement technique is an active measurement approach that quantifies the fissile content (weighted sum of ^{235}U, ^{239}Pu, and ^{241}Pu) emphasizing the presence of ^{235}U. The technique functions by measuring delayed neutrons emitted by fission fragments in the seconds following an active-interrogation induced fission. There are many isotopes produced when ^{235}U, ^{239}Pu, or ^{241}Pu fission. The time dependence of the delayed neutrons emitted from these isotopes is often described by six time groups. The half-lives of these groups vary from ~1/5th of a second to ~1 minute [Rinard 2001].

A potential implementation scenario starts when a neutron generator produces a burst of neutrons near the fuel; ideally, the generator will surround the fuel. After all the neutrons from the interrogating burst are gone (~1 ms), the total neutrons emitted from the fuel (combination of Cm and delayed neutrons) are measured for ~19 ms. The pattern of neutron bursts followed by total neutron counting is repeated to obtain the desired precision. The delayed neutrons need to be significant in quantity relative to the curium background.

When ^{235}U fissions, ~1.6% of the neutrons emitted are delayed. When ^{239}Pu fissions, ~0.6% of the neutrons emitted are delayed. When ^{241}Pu fissions, ~1.6% of the neutrons emitted are delayed. Assuming commercial-grade Pu, with five times as much ^{239}Pu as ^{241}Pu, ~0.77% of the neutrons emitted from Pu would be delayed neutrons. Hence, on a per-fission basis, 2.1 times more delayed neutrons are emitted from ^{235}U than from Pu. As with all NDA techniques proposed to date, a delayed neutron system will need to be integrated with other NDA techniques and/or burn-up codes to determine elemental Pu. The fact that the delayed neutron signal from ^{235}U is stronger than from fissile Pu may prove useful in integrating among techniques to determine elemental Pu since most other neutron based systems emphasize Pu fissile.

It is expected that a neutron generator with an intensity of ~10^{12} n/s will be needed to interrogate a commercial grade used fuel assembly. An intensity of ~10^{13} n/s is expected to be possible with present deuterium/tritium (DT)-based technology (14 MeV), and ~10^{11} n/s is possible with a deuterium/deuterium- (DD) based system. The DD-based system is preferable due to the lower energy spectrum (2.2 MeV) and because it is tritium free. It is desirable to reduce the neutron energy to below 1 MeV before it interrogates the fuel to avoid inducing fissions in ^{238}U. Research into improving neutron generators needs to be done. One factor that may make the neutron generator design easier is that the target of the neutron generator can be made very large. The target would ideally wrap around the fuel assembly.

Delayed neutron analysis is mature. The neutron generators needed are thought to be possible with present technology but have not yet been produced and need to be customized to the used-fuel measurements context. The underwater used fuel application of the high-yield neutron sources needs to be developed.

Potential Impact
Nondestructive, timely, measurement of fissile-content (^{239}Pu + ^{241}Pu + ^{235}U) emphasizing ^{235}U content

Needs That Could be Addressed if Successfully Developed
Pu measurement in used fuel
Pu measurement in actinide-bearing materials

Research Questions
1. How to design the detector to maximize sensitivity for the three measurement situations of interest (water, borated water, air)? How strong a neutron generator is needed?
2. How well will delayed neutron detection perform given the wide range (burn-up, initial enrichment, cooling time) of fuel in existence? How well will delayed neutron detection perform in various diversion scenarios?
3. How well does delayed neutron detection, in combination with burn-up codes and other NDA techniques, quantify the Pu content in used fuel?
4. How well does delayed neutron detection work with actual used fuel? What is the design of the neutron generator that can produce the necessary neutron intensity?
5. How well can delayed neutron detection measure various fuel types: spent MOX recycle, BWR, VVER, CANDU, etc.? How well can a delayed neutron measurement system quantify Pu in an electrochemical (pyro) product? How is the accuracy of delayed neutron detection impacted by the non-isotropic nature of reactors (assemblies next to control rods, on the edge of the reactor, etc.)?

Milestones	**Estimated Timeline**
Complete Monte Carlo research to optimize the delayed neutron detection design and determine the neutron generator strength (Q1)	1 year
Complete Monte Carlo research to quantify the capability of delayed neutron detection for fuel with a range of burn-up, initial enrichment, cooling time. Investigate the capability of PNAR in water, borated water, air. Investigate the capability of delayed neutron detection for several diversion scenarios. (Q2)	2 year
Complete integration effort to combine Monte Carlo results of delayed neutron detection with burn-up codes and other NDA techniques (Q3)	2 years
Decision point: Adequate virtual performance motivates full-scale prototype?	2 years

Design and construction of prototype capable of assaying used fuel assemblies. Included in this design is a neutron generator capable of delivering the necessary neutron intensity.	3 years
Completion of empirical measurement campaign and prototype demonstration. (Q4)	4 years
Complete Monte Carlo research to quantify the ability of delayed neutron detection to measure various fuel types: spent MOX recycle, BWR, VVER, CANDU, etc. and electrochemical (pyro) product? Quantify the impacted of non-isotropic nature of reactors (assemblies next to control rods, on the edge of the reactor, etc.) on accuracy. (Q5)	4 years

4.1.8 Delayed γ-Ray Detection

The measurement of delayed gamma-rays is an active measurement technique that quantifies total fissile content and may be utilized to provide crude indications of relative fissionable isotope abundance. (Because the gamma rays are emitted following beta-decay of fission products, their yields are only weakly sensitive to the fissionable isotope as explained in the last paragraph). The technique focuses on monitoring delayed gamma rays emitted following beta-decay of excited fission fragments following neutron or photon interrogation. On average, ~six to eight delayed gamma rays are emitted per fission event, thus providing up to three orders of magnitude more signal than delayed neutrons. Furthermore, the decay of delayed gamma-ray emission is considerably slower than delayed neutrons, with timescales for emission ranging from several seconds up to years (*e.g.,* long-lived fission products). By utilizing the relatively large delayed gamma-ray yield for fissionable material identification and quantification, several advantages may be possible, including increased sensitivity, increased accuracy, and decreased inspection time. While all of these may not be available simultaneously, application specificity could govern which of these advantages should be sought.

Any active inspection technique that utilizes delayed gamma-rays must have the ability to discriminate between delayed gamma-rays emitted from fission reactions and those emitted from alternative reactions within non-fissionable materials. However, because the total delayed gamma-ray emission per fission reaction carries an average of ~7 MeV, there is a substantial gamma-ray yield above ~2.5 MeV that occurs at long times following the fission reaction. Initial measurements show sensitivity at the 95% confidence level for ~560 mg, with a mass determination accuracy of 1% for ~200 g of ^{238}U. These sensitivity and accuracy measurements were performed with a 600 s data collection period, with approximately 18 times less bremsstrahlung intensity than was utilized for similar delayed neutron measurements [Hunt 2008]. For safeguards, optimizations will need to be implemented and the effects of other fissionable material (*e.g.,* Np, Am, Cm) need to be investigated.

Prior research utilizing neutron- and photo-induced fission has shown that detecting specific gamma-rays that originate exclusively from fission fragments allows for the detection of nuclear materials [Gehrke 1994; Hollas 1987; Jones 2005; Jones 1996; Twomey 2007; 2003; Norman 2004; Slaughter 2007]. This detection results because the majority of the initial fission fragments are in excited states and their energy stability is attained through emission of these delayed gamma rays, which have discrete energy levels. Unfortunately, isotopic identification is not necessarily obtained because the fragments produced during the fission process remain fairly constant from one isotope to another. However, the mass distributions of the fragments vary significantly from isotope to isotope, particularly for those fragments that make up the light fragment peak. Thus, time-dependent measurements and/or ratios of specific de-excitation gamma-ray production can provide signatures of specific fissionable isotopes. Which particular gamma-ray lines/ratios show the greatest variation from one isotope to another and the ability to measure these variations needs to be investigated. As with all spectroscopy measurement techniques, discriminating signals from potentially large gamma-ray backgrounds can be problematic.

Potential Impact
Nondestructive, timely, measurement of fissile-content

Needs That Could be Addressed if Successfully Developed
Fissile material content measurement in used fuel
Fissile material content measurement in actinide-bearing materials

Research Questions
1. How to design the detector to maximize sensitivity for measurement situations of interest? How strong a neutron generator/photon source is needed? Which delayed gamma-ray lines/ratios produce the most useful signatures?
2. How well will delayed gamma-ray detection perform given the wide range (burn-up, initial enrichment, cooling time) of fuel in existence? How well will delayed gamma-ray detection perform in various diversion scenarios?
3. How well does delayed gamma-ray detection, in combination with burn-up codes and other NDA techniques, quantify the content in used fuel?
4. How well does delayed gamma-ray detection work with actual used fuel? What is the design of the neutron generator/photon source that can produce the necessary neutron/photon intensity?
5. How well can delayed gamma-ray detection measure various fuel types: spent MOX recycle, BWR, VVER, CANDU, etc.? How well can a delayed gamma-ray measurement system quantify fissile content in an electrochemical (pyro) product? How is the accuracy of delayed gamma-ray detection impacted by the non-isotropic nature of reactors (assemblies next to control rods, on the edge of the reactor, etc.)?
6. To what extent is MCNPX or the recently upgraded COG capable of successfully predicting the intensity of individual β-delayed fission product gamma-rays?

Milestones	**Estimated Timeline**
Complete simulations/numerical calculations to optimize the delayed gamma-ray detection design, determine the neutron generator/photon source strength, and identify delayed gamma-ray lines/ratios of interest (Q1)	1 year
Complete simulations/numerical calculations to quantify the capability of delayed gamma-ray detection for fuel with a range of burn-up, initial enrichment, cooling time. Investigate the capability of delayed gamma-ray detection for several diversion scenarios. (Q2)	2 years
Complete integration effort to combine calculation results of delayed gamma-ray detection with burn-up codes and other NDA techniques (Q3)	2 years
Decision point: Adequate virtual performance motivates full-scale prototype?	2 years
Design and construction of prototype capable of assaying used fuel assemblies. Included in this design is a neutron generator/photon source capable of delivering the necessary neutron/photon intensity.	3 years
Completion of empirical measurement campaign and prototype demonstration. (Q4)	4 years

Complete Monte Carlo research to quantify the ability of delayed gamma detection to measure various fuel types: spent MOX recycle, BWR, VVER, CANDU, etc. and electrochemical (pyro) product? Quantify the impact of non-isotropic nature of reactors (assemblies next to control rods, on the edge of the reactor, etc.) on accuracy (Q5, Q6)	4 years

4.1.9 Ultra High-Resolution Alpha Spectroscopy

The ultimate target for Ultra High-Resolution Alpha Spectroscopy is to eliminate the need for the present sample/remote laboratory/mass spectrometer system for accurately determining the mass and isotopics of Pu. The goal is to replace the status quo with faster, on-line equipment that doesn't generate any waste. The ultra high-resolution alpha spectroscopy technology also goes by the name of microcalorimeter (μcal). Most of the hardware is the same in a photon-based and an alpha-based μcal system but the detector surface does need to be altered and the analysis must be customized.

A wide range of actinides of interest to safeguards, in particular all the major Pu isotopes (except ^{241}Pu), decay by alpha decay. The energy of the emitted alpha particle is unique to the nuclei of origin and, as such, can be used to quantify the presence of each alpha emitting isotope. Virtually all the alpha particles emitted have energies in the range of 4 to 6 MeV. Because alpha particles are heavy and charged, they deposit their energy in a very thin layer. For this reason the aqueous samples of interest are deposited on a substrate that is positioned very close to the μcal detector with a vacuum gap between the detector and the sample.

Present alpha spectroscopy systems have a FWHM resolution of ~10 keV as compared to the expected ~1 keV FWHM resolution of a μcal alpha spectroscopy system. This order of magnitude improvement in resolution has the potential to significantly change the way in which an alpha spectroscopy system functions when measuring actinide solutions. For example, it is likely possible to measure all the actinides from one sample – eliminating the need to chemically separate the approximately five actinides of interest. Furthermore, since separation is no longer necessary, the introduction of tracer elements is eliminated. Hence, any material extracted from the material stream can return to the material stream completely unaltered, eliminating the need for waste treatment.

Microcalorimetry has been an active area of research for over two decades. The application of μcal to alpha spectrometry has received relatively little attention as compared to photon detection [Horansky 2008; Coron 1985; Giuliani 2000]. Preliminary experimental work was done and the great promise of great resolution was confirmed; for example, the alpha particles emitted from ^{239}Pu and ^{240}Pu were observed to be distinct from one another for the first time ever.

Potential Impact
Determination of Pu isotopics, and other alpha-emitting actinides, from DA samples in less than 24 hours. Elimination of DA waste stream.

Needs That Could be Addressed if Successfully Developed
Rapid laboratory sample analysis
Direct measure of Pu in reprocessing streams
Direct Pu measurement in actinide-bearing materials

Research Questions
1. For a variety of reprocessing streams, including a mixture of present streams (PUREX) and future stream (UREX$^+$), what does the expected alpha spectrum look like given the expected resolution of a μcal system as well as the demonstrated resolution of a μcal system?
2. Given the present sample-taking methodology, what steps need to be taken in sample preparation? What is the chemical methodology for this process?
3. What is the optimum means for introducing an on-line μcal system at a reprocessing facility? How would an industrial-scale automated alpha spectroscopy system function from sample collection, sample preparation, and measurement, to waste return?
4. How can the measurement speed be improved?
5. How well will a μcal alpha spectroscopy system perform with actual reprocessing streams?

Milestones	Estimated Timeline
Complete modeling of several reprocessing streams, including a mixture of present streams (PUREX) and future stream (UREX$^+$), to produce the expected alpha spectrum for both the expected resolution of a μcal system as well as the demonstrated resolution of a μcal system (Q1)	1 year
Determine sample taking methodology: list the steps needed in sample preparation. Establish the necessary chemical methodology. (Q2)	3 year
Decision point: Adequate virtual performance motivates full-scale prototype?	3 years
Determine the optimum means for introducing an on-line μcal system at a reprocessing facility. Provide a conceptual design of an industrial-scale automated alpha spectroscopy system that contains sample collection, sample preparation, measurement and waste return. (Q3)	3 years
Complete array design to enable faster measurement (Q4)	4 years
Complete measurements of reprocessing stream material with a μcal alpha spectroscopy system (Q5)	5 years

4.1.10 Ultra-High Resolution Neutron

In the arena of long-range enabling R&D, high-resolution fast-neutron spectrometers are an enabling technology for non-destructive analysis (NDA) of nuclear materials, especially of fissile materials with characteristic neutron emissions [Niedermayr 2004]. They also allow the detection of light elements in actinide matrices, such as oxygen in Pu [Friedrich 2006]. Neutron spectrometers allow analyses though shielding impenetrable to gamma-rays, for example for on-line monitoring of processing plants.

Superconducting fast-neutron spectrometers (“Ultra-N-Spec”) offer an order of magnitude higher energy resolution than conventional semiconductor or gas-based detectors [Niedermayr 2004], and significantly higher efficiency than time-of-flight spectrometers [Friedrich 2006]. In addition, they have a simple response function and allow straightforward discrimination against gamma or neutron-scattering events [Niedermayr 2007].

Superconducting fast-neutron detectors consist of a neutron absorber crystal and a thin film sensor operated at the transition between its superconducting and its normal state (transition edge sensor, TES).

Neutron capture in an (n,α) reaction deposits a total energy $E_{neutron} + Q_{(n,\alpha)}$, and the resulting temperature increase is measured as a change in sensor resistance. Operation at very low temperatures reduces thermal noise and allows an energy-dependent energy resolution below 10 keV for fast neutrons in the MeV range.

Prototype fast-neutron spectrometers have been built based on ~mm^3 TiB_2 or ~cm^3 enriched ^{6}LiF neutron absorbers coupled to superconducting Mo/Cu sensors. They are cooled to their operating temperatures around 0.1 K in an adiabatic demagnetization refrigerator (Figure 7, left). The TiB_2-based prototypes have achieved an energy resolution of 5.5 keV at a total energy of 2.9 MeV [Niedermayr 2004]. The larger ^{6}LiF-based devices have achieved and energy resolution below 50 keV for MeV neutrons [Friedrich 2006]. They have efficiency above 1% for 1 MeV neutrons, and their response above $Q_{Li\text{-}6} = 4.78$ MeV is mostly set by the (n,α) absorption cross section in ^{6}Li (Figure 7, center and right) [Niedermayr 2007].

Potential Impact
Nondestructive, timely neutron measurement for on-line process monitoring or detection of light elements in actinide matrices (such as oxygen), precision measurements of yield and spectra of (α,n) reactions

Needs That Could be Addressed if Successfully Developed
Process monitoring of reprocessing streams
Rapid laboratory sample analysis

Research Questions
1. What specific safeguards needs could be addressed using neutron spectroscopy?
2. Can pixellated systems achieve sufficient efficiency and gamma discrimination at reasonable cost?
3. Can the instrument perform at count rates sufficient for safeguards applications?

Milestones	Estimated Timeline
Working group to identify whether neutron spectroscopy is key enabler (Q1)	0.5 year
Development of high-efficiency prototype neutron spectrometer with liquid-cryogen-free refrigerator (Q2)	2 years
Complete proof-of-principle measurements for specific safeguards applications (Q3)	3 years
Demonstration of capability as on-line process monitor (Q3)	4 years

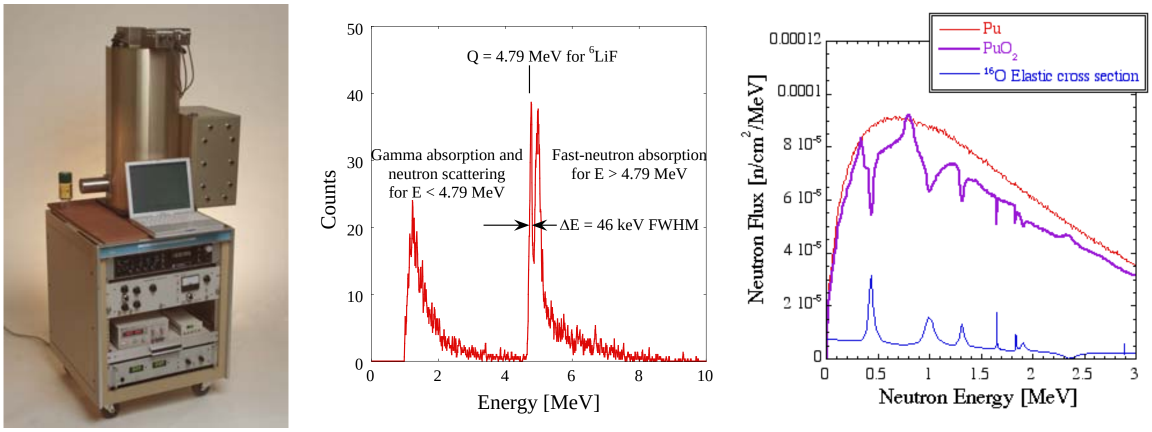

Figure 7. (Left): Cryogenic neutron spectrometer. The neutron detector is held in the cryostat on top of the rack at ~0.1K at the end of the cold finger within ~2 cm of the radioactive neutron source at room temperature; (Center): MCNP simulation of the neutron spectrum for Pu and PuOx; (Right): Fast-neutron spectrum of a Cf-252 source, showing 46 keV energy resolution at MeV energies and good discrimination between fast-neutron absorption and neutron scattering events.

4.1.11 Ultra-High Resolution Gamma

Precise accounting of nuclear material is essential to detect and deter the diversion of fissile material, and gamma-spectroscopy is routinely used for qualitative and quantitative non-destructive analysis (NDA) in this safeguards context.

Ultra-high energy resolution increases the precision of NDA in nuclear TRU mixtures whenever HPGe spectra are affected by line-overlap, *e.g.*, for Pu and U analysis at ~100 keV [Terracol 2004; Drury 2005; Friedrich 2007; Hoover 2007; Horansky 2007]. This can replace DA with NDA in certain applications. Alternatively, ultra-high energy resolution reduces the time required to characterize a sample with a certain precision. It also increases the sensitivity for detecting weak emission lines above a constant background, as required for direct detection of Pu above the Compton background of fission products in used fuel.

Superconducting gamma-spectrometers (“UltraSpec,” “Microcalorimeter”) operated at temperatures around 0.1 K offer an order of magnitude higher energy resolution than conventional high-purity germanium (HPGe) detectors at ~100 keV [Drury 2005; Doriese 2007]. This can increase the precision of NDA accordingly. The detectors work by measuring the increase in temperature upon absorption of a gamma-ray with a sensor film operated at the transition between its superconducting and its normal state, and are therefore also referred to as transition edge sensors (TESs) or microcalorimeters (µCal). A bulk ~mm^3-sized absorber is attached to the sensor, and several such pixels are typically operated in parallel for increased efficiency [Doriese 2007].

Current ultra-high resolution gamma-spectrometers are based on superconducting tin absorbers and Mo/Cu bilayer or multilayer TES detectors. For user-friendly operation at the required operating temperatures, refrigerators have been built that attain ~0.1 K at the push of a button without the use of cryogenic liquids (Figure 8, left). The detectors have achieved an energy resolution between 30 and 90 eV

FWHM for Gamma-energies at 100 keV (Figure 8, center) [Friedrich 2007; Doriese 2007]. Specialized digital signal processors allow each 2 mm × 2 mm pixel to be operated at rates above 100 counts/s (Figure 8, right) [Dreyer 2008]. 112-pixel detector arrays are currently under development for count rates at 10,000 counts/s.

Potential Impact
Nondestructive, timely, precise low-energy gamma ray measurement of actinides without the need for chemical separation

Needs That Could be Addressed if Successfully Developed
Direct Pu measurement in used fuel
Direct Pu measurement in actinide-bearing materials
Rapid laboratory sample analysis

Research Questions
1. For which gamma-spec scenarios does <100 eV resolution substantially improve performance?
2. Can pixellated systems achieve sufficient efficiency and gamma discrimination at reasonable cost?
3. Can the instrument perform at count rates sufficient for safeguards applications?
4. Can current analysis codes be adapted for ultra-high-res response functions?

Milestones	Estimated Timeline
Working group to identify where ultra-high-res gamma spectroscopy is key enabler. (Q1)	0.5 year
Development of high-efficiency prototype neutron spectrometer with liquid-cryogen-free refrigerator (Q2)	2 years
Complete proof-of-principle measurements for specific safeguards applications. (Q3)	3 years
Complete proof-of-principle quantitative analysis using modified isotopic codes. (Q3)	3 years
Demonstration of capability as on-line process monitor (Q3)	4 years

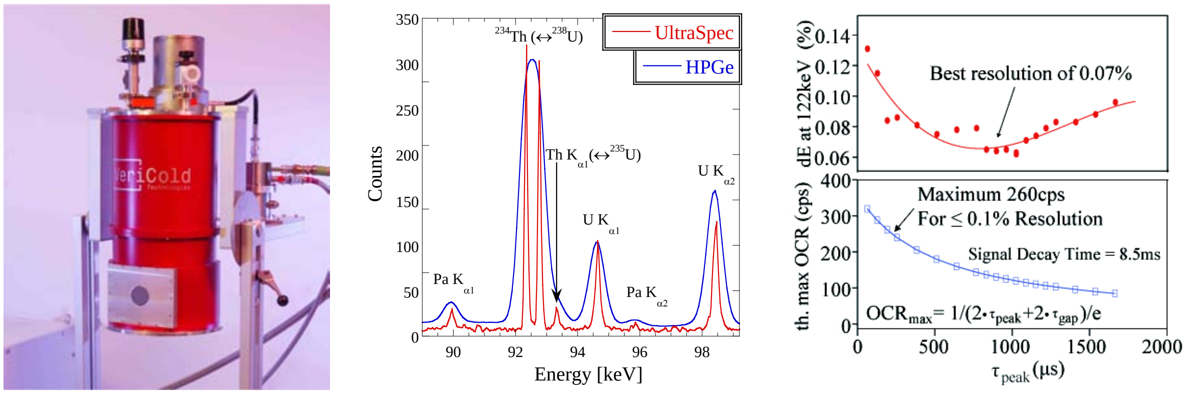

Figure 8. (Left): Cryostat for liquid-cryogen-free UltraSpec detector operation at 0.1 K; (Center): Gamma spectrum of LEU, once taken with UltraSpec and once with a conventional HPGe detector; (Right): Specialized DSP readouts allow >100 counts/s despite slow signals with several ~ms decay times.

4.1.12 Electrochemically Modulated Separations

Electrochemically-modulated separations (EMS) have been utilized for the separation and trace analysis of uranium [Pretty 1998] and plutonium [Clark 2006]. EMS benefits from two-stages of selectivity: redox state adjustment and redox-specific complexation at the modified electrode. For U and Pu, a simple anodized glassy carbon electrode is highly specific for uranium or plutonium accumulation under reducing and oxidizing conditions, respectively, effecting a separation using only an applied potential. Several EMS applications are envisioned for materials accountability in reprocessing: 1) isolation, purification, and release into a clean, less-hazardous solvent stream for destructive analysis (DA); 2) purification and concentration on the working electrode surface for nondestructive analysis (NDA, *e.g.*, gamma or neutron spectroscopy); or 3) isolation and removal of nanogram-size solids for DA.

Both DA and NDA are desired for monitoring aqueous reprocessing streams and for safeguarding acute or protracted material diversions. EMS will allow rapid separations of a slip-stream sample for either NDA or DA (Figure 9). Currently, DA begins with grab sampling prior to laboratory-based separations and mass spectrometric analysis. On-line separations using EMS with addition of quantitative spikes would result in significant reduction in analytical cost and hazards. Direct NDA of the process stream is currently limited by activation and fission product interferences that would be eliminated by EMS.

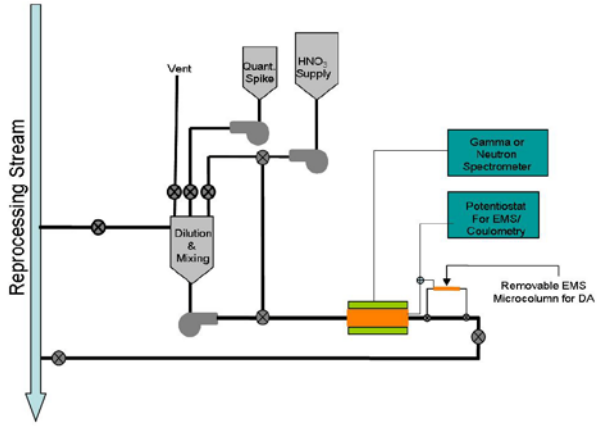

Figure 9. Conceptual diagram of the EMS system for NDA monitoring and DA sampling of the reprocessing stream, providing on-line separation and spiking for quantification.

Prior work has demonstrated that Pu can effectively be isolated from a 10^7 excess of uranium. Figure 10 illustrates how effectively Pu can be isolated from other components likely to be present in the reprocessing stream. The next steps in implementing EMS for monitoring reprocessing streams are the design, testing, and demonstration of a scaled-up separation cell with dissolved nuclear fuels.

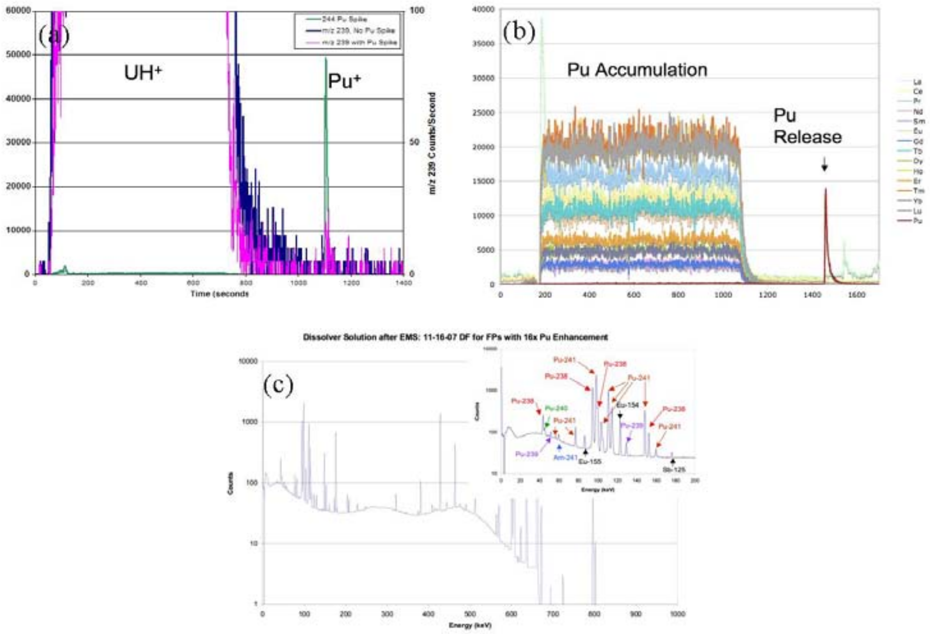

Figure 10. Temporal ICP-MS signal shows the accumulation of Pu as (a) uranium (UH+ monitored) and (b) rare earth elements pass through prior to the release of plutonium. (c) Resulting gamma SYNTH spectrum is also shown, demonstrating the promise of isotope dilution gamma spectrometry for Pu detection.

Potential Impact
Rapid, on-line, direct quantitative NDA of Pu in aqueous reprocessing streams
Rapid isolation and solid sampling (≤ 1 ng) for lab-based Pu and U concentration analysis

Needs That Could be Addressed if Successfully Developed
Direct measurement of Pu in reprocessing streams
Rapid laboratory analysis

Research Questions
1. Can electrochemically modulated separations processes be scaled up to dimensions needed to monitor fuel reprocessing?
2. Is the anodized glassy carbon electrode surface robust in the high radiation environment? Can it be regenerated on-line?

Milestones	Estimated Timeline
Scale up and laboratory testing of laboratory based EMS system (Q1)	1 year
Evaluation of measurement uncertainties and limits of detection (Q2)	2 years
Design and automation of EMS system (Q3)	3 years
Testing of production prototype	4 years

4.1.13 X-Ray Transmission/Fluorescence Techniques

Mass Spectrometry remains the standard measurement for input accountancy in reprocessing plants. Coulometry remains the standard for plutonium accountancy. Davies-Gray Titration will likely remain the standard for accountancy for pure uranium measurements. However, these are time-consuming measurement techniques that require considerable sample preparation, and therefore contribute to waste-handling concerns for laboratories. Hybrid K-Edge Densitometry (HKED) is the most familiar label for a class of nondestructive X-ray transmission and fluorescence techniques that have been developed to meet the same analytical-lab objectives as those methods described above. HKED can provide accuracies that rival the current laboratory techniques, but is performed directly on samples drawn from process equipment, in a very timely manner (count times of a few tens of minutes) with no sample preparation and subsequent waste issues and the samples can be returned directly to the processes. As the name implies, these transmission/fluorescence techniques are a “hybrid” of two long used techniques, K-edge densitometry and X-ray fluorescence, to significantly enhance their capabilities. Each of these techniques is described below.

The K-Edge Densitometry (KED) method uses a highly-collimated X-ray beam passing through a solution sample of well defined path length. Its transmission is measured as a function of energy in critical energy regions. The abrupt change of the transmitted X-ray intensity at the the K-level absorption edge is a measure of the concentration of elements with nearby K-edge energies (see Figure 11). A pure K-Edge instrument requires a series of carefully characterized solutions of uranium and/or plutonium for establishing a calibration curve. Interferences arise from elements having their K absorption edge in the same energy region as the element under investigation. Because U and Pu have similar K-edge energies, their relative concentrations are difficult to distinguish using K-edge measurements alone. It is for these mixed U/Pu samples, which are the core of many accountancy measurements in a reprocessing plant, that the K-Edge technique needs to be augmented with X-ray fluorescence. In X-ray fluorescence (XRF), an X-ray beam of larger divergence stimulates the emission of characteristic X-rays from uranium and plutonium. The intensities of the induced X-rays may be used for the determination of the U/Pu ratio in a sample or, after appropriate calibration, for the absolute determination of the respective amounts of an element. In its simplest explanation, the technique derives the quantitative ratios of elements based on the relative count rate recorded in specific peak regions of interest [Camp 1981].

The combination of K-Edge Densitometry with X-ray fluorescence resulted in the so-called Hybrid K-Edge (HKED). This instrument uses a single X-ray source for both parts of the analysis, the K-edge absorption and the fluorescence excitation. It has proven to be an extremely useful analytical tool in the verification of reprocessing input solutions, in large part because it allows the simultaneous and quantitative determination of uranium and plutonium. This can even be done directly from samples of highly-radioactive input solutions. Accuracies of a few tenths of a percent have routinely been achieved, rivaling the more traditional techniques but with more timely results and reduced analytical wastes. This technique was used for in-line aqueous uranium-plutonium flow measurements of reprocessed nuclear fuel at the Institute of TransUranium Elements (ITU) at Karlsruhe [1981] (Figure 12).

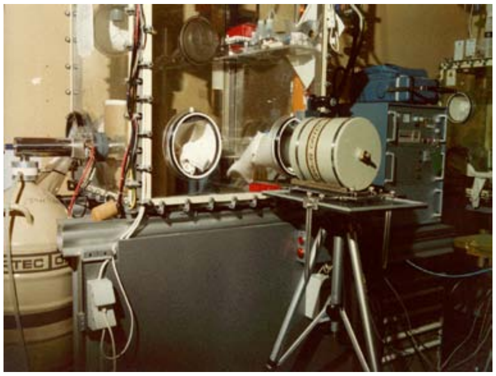

Figure 11. X-ray fluorescence analysis at Institute of TransUranium Elements [Camp 1981].

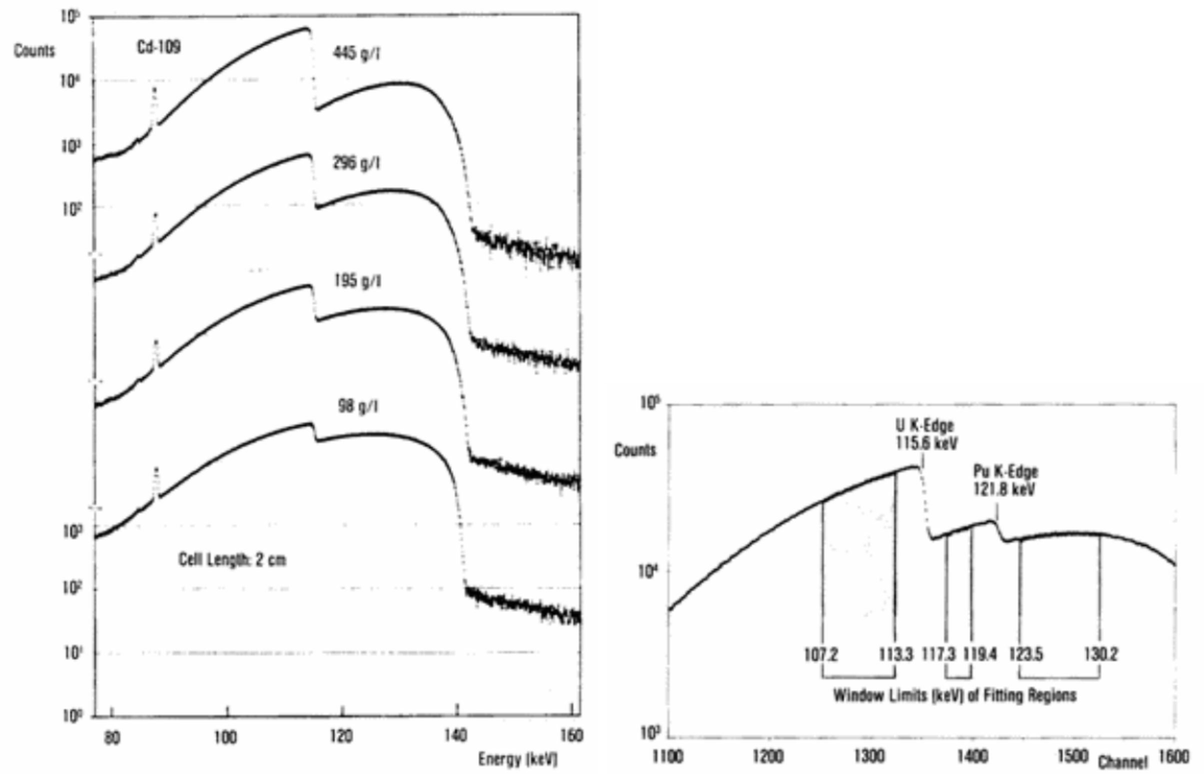

Figure 12. Example spectra from HKED, showing the variation in intensity and spectral shape with uranium and plutonium concentration.

More recent work has expanded on the idea of HKED, and has indicated that the utility of the method might be further expanded in reprocessing plant monitoring. For example, it may be possible to provide concentration ratios for other actinides such as Am and Cm. There has also been preliminary work to extend the absorption-edge densitometry concept beyond the K-edge to the L-edge [Wang 2006]. In this work, a Compton-suppressed spectrometer and list-mode data acquisition have been used to wring additional sensitivity from the method, and it has shown promise for determining the ratios of minor isotopes as well.

Potential Impact
Rapid, waste-minimizing, accurate measurement of elemental U, Pu and minor actinide concentrations in samples drawn from reprocessing streams

Needs That Could be Addressed if Successfully Developed
Rapid laboratory sample analysis
Direct measurement of Pu in reprocessing streams

Research Questions
1. Can the technology be extended to the measurement of other minor actinides (*e.g.*, Am, Np) and what is the corresponding accuracy and precision in samples representative of AFCI-defined separation processes?
2. How does the accuracy and precision of these methods change for on-line implementations?
3. Can the incorporation of additional absorption edges significantly improve performance over commercially available systems for either laboratory or on-line implementations?
4. Can instrumentation variants (*e.g.*, Compton-suppression or coincidence techniques) improve performance over commercial systems?

Milestones	Estimated Timeline
Develop test bed to support head-to-head evaluations of different techniques and instrumentation combinations. (Q1, Q2)	1 year
Identify the most promising alternative signatures perform comparative studies. (Q1, Q2)	2 years
Built advanced prototypes (*e.g.*, Compton-suppressed or coincident), evaluate. (Q4)	2 years
Deploy on-line (and laboratory) prototypes for evaluation at reprocessing plants. (Q3)	3 years

4.1.14 Advanced Tags and Seals

Tags and seals will be necessary for containment and surveillance of items, in particular between Materials Balance Areas within a facility and during shipments. An effective tag or seal could provide the necessary confidence in continuity of knowledge, reducing the need for NDA measurements. Advanced tags and seals require innovations to meet more stringent and expanded performance requirements. The need for active seals and tags to enable near real-time accountancy with longer periods between physical access requires innovation in several technical areas. The active seal should function reliably in proximity to neutron and gamma sources over its expected lifetime. Power use should be minimized so battery life is sufficient for the inventory period, and robust authentication, confidentiality and tamper-resistance and protection are needed. An active tag seal network should be simple to install, configure, and maintain, and adaptable to many material monitoring applications. Data security and data management of the active tag is a critical requirement, necessary to support the anticipated growth in joint use safeguards monitoring equipment.

The incorporation of more difficult to duplicate physical authentication measures into the active seal is desirable, to deter and detect seal substitution. Passive tags and seals should also incorporate more complex physical authentication independently of the seal integrity indicator, to deter and foil sophisticated tampering. Both active and passive seals should maintain integrity and function over a wide range of temperatures and environmental conditions.

Radio Frequency Identification (RFID) tags can help overcome the line-of-sight limitations of optical signal tags, but they face serious challenges at their current state of technology. Most of the commercial-off-the-shelf (COTS) RFID systems operate in very narrow frequency bands that make them vulnerable to detection, jamming and tampering, as well as having difficulty around metal containers. Furthermore, the commercial passive RFID tags have short range, and finally the active tags that provide long ranges have limited lifetimes. In addition, such tags are known to perform poorly around metallic environments. Authentication of COTS systems may require additional system modifications to generate a trusted RFID system. One promising avenue of exploration is ultra-wide-band (UWB) RFID technology for tags and seals. Some of the improvements to the current COTS systems include: The ability to work around metals and cluttered environments, accurate geo-positioning with fine spatial resolution (*i.e.*, cm accuracy), and low probability of intercept and detection [Nekoogar 2008].

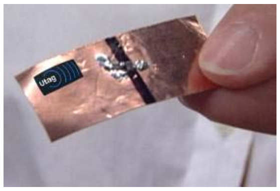

Figure 13. UWB passive tags are small, have an unrestricted lifetime, and can operate in harsh propagation environments and have geo-location capability.

A suite of technologies under development may be integrated to provide system solutions for active inventory control and monitoring, with robust authentication and seal integrity functions. Key features for next-generation devices include anti-tamper enclosures, integral radiation shielding, and authenticated data transmission [Dowla 2005; Nekoogar 2008; Schoeneman 2005].

Potential Impact
Improved continuity of knowledge reduces need for DA, NDA

Needs That Could be Addressed if Successfully Developed
Active and Passive RFID tags

Research Questions
1. Can anti-tamper, size, battery, environmental requirements be met in cost-effective tag?
2. Multi-attribute tag and seal technology: can multiple signatures be combined effectively?

Research Milestones	**Estimated Schedule**
Design, build, test prototype tags with integral radiation shielding, anti-tamper enclosures (Q1)	1 year
Design, build, test prototype multi-attribute tag and seals (Q2)	2 years
Complete proof-of-principle sensor-tag integration	2 years

4.1.15 Radiation Imaging for Design Information Verification

The 3-D laser (lidar) scanning technology is now being utilized for Design Information Verification (DIV) applications. That technology has the ability to assess whether the physical features of a facility (*e.g.*, equipment and piping) are unchanged from previous verification visits, but laser scanning does not necessarily provide indication of whether the material flows in a facility (*e.g.*, streams moving through a complex network of piping) are off-normal in some way. Radiation imaging, most particularly gamma-ray imaging, has the potential to help meet that need.

Some gamma-ray imaging techniques are capable of 3-D source intensity mapping for an imaging system located at a fixed position, a feature that has proven to be useful in homeland-security applications, for example [Ziock 2004; Vetter 2002]. Visual and/or range data from visual cameras and laser range scanners can then be merged into the 3-D gamma-ray imaging process for superior contrast, shorter image reconstruction processing times, and direct identification of physical objects containing radioactivity. The integrated gamma-ray and range mapping can then be either inspected by an automated program or user, or compared with previously stored maps to identify changes [Mihailescu 2006; Burks 2006]. Figure 14 provides examples of how a Compton-scatter imager was utilized in proof-of-principle measurements. The use of high-resolution spectroscopic gamma-ray imagers, such as the one used in that figure, could open the door for the tracking of average isotopic concentration (*e.g.*, ^{152}Eu) over the acquisition time, a potentially useful parameter for detecting anomalies in how the plant is being used over extended time periods.

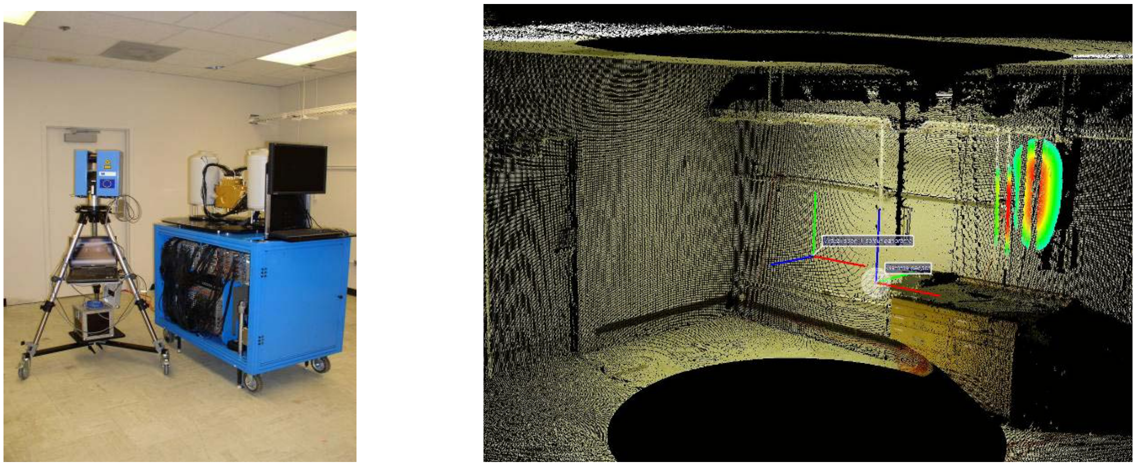

Figure 14. (Left): A 3-D Design Information Verification (DIV) laser range scanner and a Compact Compton Imager (CCI) gamma-ray camera on a mobile cart used to acquire the 3-D integrated room model, with accompanying source-intensity map (right).

A combined use of a spectroscopic gamma-ray imager with a large field of view and a lidar scanner also has the potential to aid materials accountability by enabling more accurate real-time holdup and material accumulation measurements in bulk facilities across the nuclear fuel cycle (enrichment, fuel fabrication, and reprocessing) for international safeguards. The 3D-GIVS may be utilized for measurements in inaccessible parts of facilities, glove boxes, and along ducts and pipes. Furthermore, errors that commonly arise from underestimating holdup due to self-attenuation can be partly averted because the system would provide information on the density, distribution and shape of the material, thereby permitting more accurate self-absorption corrections.

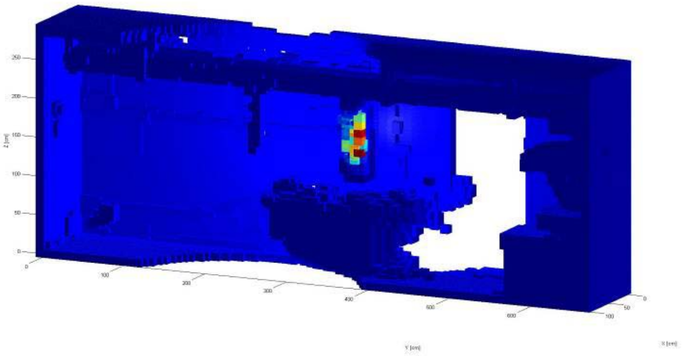

Figure 15. Example of a 3-D gamma-ray image of a 1-m long ^{152}Eu gamma-ray source, integrated with a voxellized LIDAR scan of the room.

Potential Impact
Augment today's 3-D laser range-finding information about a plant's physical features with additional information about the plants operation and processing streams.

Needs That Could be Addressed if Successfully Developed
Advanced design Information verification
Process monitoring

Research Questions
1. How do the high background fields in reprocessing plants impact the spatial resolution and activity-quantification capabilities of 3-D gamma-ray imaging?
2. What spatial and energy resolution, and collection efficiency, are required to meet DIV objectives and timelines?
3. Can a nominal imager design, combined with automated anomaly detection methods, provide sufficient sensitivity to detect off-normal conditions expected in diversion scenarios?

Research Milestones	**Estimated Schedule**
Simulate integrated imaging performance, assuming a range of imager characteristics (spatial and energy resolution, and collection efficiency) in representative scenarios with intense and varying backgrounds. (Q1, Q2, Q3)	1 year
If viability assessments show promise, deploy prototype imager/LIDAR system and perform mock DIV inspections. (Q2, Q3)	2 years

4.1.16 Basic Nuclear Data Measurements

Safeguards projects encounter nuclear data issues when (1) new Non-Destructive Assay (NDA) technologies or schemes are modeled using sophisticated simulation codes such as MCNP(X) or GEANT or (2) when assessments of various architectures are performed for specific inspection regimes and fuel cycles. These simulations are used to lay the groundwork for proposing and planning new projects and also to optimize the design or analysis of different configurations. Many calculations can be performed quickly, whilst individual experiments involving SNM require extensive authorization and are costly. Simulations of fielded experimental interrogation configurations can be used to interpret the measured data. And very importantly, simulations can extensively explore "what if?" questions.

Simulation capabilities are built upon quality-controlled nuclear cross section and decay databases, in the ENDF nuclear data library. These evaluated databases incorporate the detailed information available from experiments and from nuclear models, and allow transport simulations to model the underlying physical phenomena. For nuclear reactor problems, the ENDF database has been shown to be accurate through comparison with many benchmark-quality measurements. The resources used include the ENSDF nuclear structure library and NSR references library, and these resources are typically accessed through a variety of web-based dissemination projects.

While the infrastructure for benchmarking, collaborating on compiling and evaluating new information for database efforts, and disseminating these results are supported by the DOE Office of Nuclear Physics in the Office of Science, the impetus for making the relevant measurements, benchmarks, or relevant nuclear reaction model calculations are expected to be supported by those programs interested in those areas.

A specific question regarding the development of advanced safeguards is whether the current fundamental nuclear data are sufficient to allow for the proper development of appropriate NDA techniques to measure the nuclear materials that would be processed in an advanced fuel cycle. The ability to efficiently design and develop advanced NDA techniques for safeguarding nuclear fuel will be dependent upon the accuracy with which the radiation characteristics of this fuel can be simulated based on fundamental physics data. The modeling capabilities that are needed for this development include depletion analysis methods to confirm and refine the declared isotopic composition of the incoming used fuel [Santi 2007b; Santi 2007a] as well as Monte Carlo methods to simulate the radioactive emissions from the material and their transport from source to measurement/detector location. In addition, the gamma-rays emitted from actinides, fission products, and activation products must be accurate for predictions based on computer simulations to be reliable. The uncertainties associated with the gamma-ray energies should also be well known. Because the expectations for advanced fuel cycles include the ability to reprocess used fuel from different types of reactors, the characteristic properties of the nuclear material that are reprocessed are expected to vary, depending on the properties of the used fuel that is being reprocessed, and the process itself, *e.g.*, aqueous, electro-chemical. Thus, the models that are used in this effort cannot be easily tuned specifically for a given reactor type, discharge burn-up, cooling-time, etc., but rather must be capable of accurately modeling the properties of used fuel taking into account a variety of characteristics as described above. The accuracy and precision of these models will directly depend on the quality of the fundamental nuclear data upon which the models are based.

An initial evaluation has been performed to determine the current status of the fundamental nuclear data that are needed to accurately model the various properties of the nuclear materials that are expected to be processed within AFCI [Santi 2007b; Santi 2007a]. The data that have been reviewed in this evaluation include prompt neutron multiplicity distributions for nuclides that undergo either spontaneous or neutron-induced fission, half-lives, gamma-ray branching ratios and energies, and (α,n) cross sections. The first part of the evaluation was performed to determine if any major gaps exist in the current nuclear data. The second part of the evaluation performed a rudimentary sensitivity analysis to determine how the uncertainties associated with the fundamental nuclear data affect uncertainties associated with the modeling of radiation from potential AFCI materials. Because a number of different reprocessing technologies are currently under consideration, the sensitivity analysis was limited to materials associated with a specific reprocessing process.

While this initial evaluation was able to identify weaknesses in the fundamental nuclear data which appear to have an affect on the ability to model the radiation signatures from materials, a key question remains as to the relative impact the uncertainties associated with the fundamental nuclear data will have on the ability to design and implement advanced safeguards instrumentation. In order to perform such a study to determine the cost/benefit ratio of performing measurements to improve the quality of the fundamental nuclear data, measurement scenarios will have to be defined which specify the type of material to be measured, the type of measurement to be performed, and the target accuracy and precision for a given measurements technique and account for the characteristics of the environment within which the measurements will be made. Another area that was not sufficiently explored in the initial evaluation is whether the appropriate nuclear data exists to develop new detection technologies such as nuclear resonance fluorescence, and lead-slowing-down spectroscopy.

Potential Impact
Improved precision and accuracy in modeling the properties of nuclear materials and characteristics of radiation signatures and environments for NDA detector development and analysis, and lower costs of viability studies

Needs That Could be Addressed if Successfully Developed
Nuclear Data Measurements

Research Questions
1. Based on input from advanced safeguards technology developers, what are the nuclear data needs?
2. What are the cross sections for photon excitation of 238Pu, 240Pu, 241Pu and 242Pu for nuclear resonance fluorescence?
3. Can a methodology be developed to determine the impact of reducing specific nuclear data uncertainties or gaps on the ability to safeguard nuclear material within a facility?
4. Based on a cost/benefit analysis, what are the top priorities in terms of the basic physics measurements that are needed for creating advanced safeguards?
5. In what facilities can basic physics measurements be performed and on what time scale?
6. Are the gamma-ray energies emitted from actinides, activation products and fission products accurate, and are their uncertainties well known?

Milestones	**Estimated Timeline**
Determine nuclear data needs based on input from safeguards technology developers. (Q1)	1 year
Measure the cross sections for photon excitation of ^{238}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu for nuclear resonance fluorescence. (Q2)	2 years
Develop methodology for determining impact of reducing specific nuclear data uncertainties based on potential improvement in safeguarding nuclear material. (Q3)	2 years
Create prioritized list of nuclear data needs and associated measurements. (Q4)	3 years
Create library of gamma rays for actinides, activation products and fission products, with uncertainties.	3 years
Identify facilities and timeline for performing high priority basic physics measurements. (Q5)	4 years

4.1.17 Calorimetry

Calorimetric assay is the determination of the mass of radioactive material through the combined measurements of its thermal power by calorimetry, and if necessary, its isotopic composition using either gamma spectroscopy or mass spectroscopy. Calorimetric assay is presently the most precise and accurate Nondestructive Assay (NDA) technique for assaying plutonium-bearing items. The nominal precision that can be achieved with this technique is ~0.5% for low-power items (≤ 0.2W) and ~0.1% for high-power items (≥ 1W). Because calorimetry measures the thermal power from the entire item, the technique does not have any uncertainty due to sampling errors as occurs with analysis techniques. Since the heat-measurement result from calorimetry is essentially independent of material and matrix type, it can be used on any material form or item matrix, assuming no additional sources of heat besides the radioactive decay of the material are present within the material (*e.g.*, chemical reactions, phase transitions, etc.). The thermal power measurement can be made traceable to a U.S. or other National Measurement Systems through the use of electrical heat standards to either directly calibrate the calorimeter or to calibrate secondary ^{238}Pu heat standards. As a result of the high precision and accuracy associated with calorimetric assay, the technique can be used to prepare secondary standards for other NDA systems that are based on neutron and gamma-ray detection [Rakel 1987; ASTM 2003; Hyman 1999; Lestone 2000].

One of the most successful radiometric calorimeter designs fit the general category of heat-flow calorimeters. A heat-flow calorimeter consists of a sample chamber insulated from a constant temperature environment by a thermal resistance and a means to measure the temperature difference across the thermal gradient produced by the thermal resistance. The temperature difference across the thermal resistance is related to the heat generated from within the sample chamber. When an item is placed in the calorimeter the temperature gradient across the thermal resistance is disturbed and the gradient changes with time until it converges to a constant value and equilibrium is achieved. Because it is necessary for the calorimeter to reach thermal equilibrium in order to determine the thermal power of the item, calorimetry measurement times are typically longer than other NDA techniques and can range from one to several hours. Various factors can affect the measurement time associated with a calorimeter including the type of material being measured, thermal instability of the sample due to endothermic or exothermic reactions occurring within the sample, the packaging of the material, the total mass of the item, the initial temperature of the item relative to the final temperature equilibrium temperature of the item/calorimeter, the type and construction of the calorimeter being used, the use of an equilibrium prediction algorithm, and the required assay accuracy.

Heat-flow calorimeters have been used to measure thermal powers from 0.5 mW (0.2 g low-burn-up plutonium equivalent) to 1000 W for items ranging in size from less than 2.54 cm to 60 cm in diameter and up to 100 cm in length. Examples of the current state of the art in radiometric calorimetry are solid-state heat-flow calorimeters that utilize thermopile components as heat sensors [Bracken 1997; Bracken 1998; Bracken 2000]. Solid-state heat sensors, which passively produce an electrical signal proportional to measured heat flow, allow heat-flow calorimeters to be portable, insensitive to mechanical strains, scalable to any size or shape, and intrinsically low-noise.

Calorimetric assay can be applied to a number of different radionuclides: plutonium, HEU, ^{233}U, ^{237}Np, ^{242,244,245}Cm, ^{250,252}Cf, ^{241,242m,243}Am, tritium, and fission products [Bracken 2007]. For the types of nuclear materials that are being considered as part of the Advanced Fuel Cycle Initiative (AFCI) which will keep various minor actinides co-mingled with plutonium, calorimetric assay is a viable option for accurately measuring the mass of the plutonium within these materials even in cases where the plutonium isotopes only contribute a smaller fraction of the total thermal power of an item because of the high accuracy and precision associated with the calorimetry measurement of an item. This assumes that the weight fractions of the relevant heat-producing actinides in the material can be determined using either a Destructive Analysis or NDA measurement technique.

Potential Impact
Pu mass determination in all matrices of safeguards interest except used fuel

Needs That Could be Addressed if Successfully Developed
Direct Pu measurement in actinide-bearing materials

Research Questions
1. What are the practical limits for using calorimetric assay within a reprocessing facility?
2. What are the types of calorimeters that are most applicable or adaptable to meeting the measurement and facility needs identified in Q1?
3. How can the required measurement time for the calorimeter(s) identified in Q2 be reduced while maintaining appropriate levels of accuracy and precision?
4. What accuracy and precision can be achieved when performing calorimetric assay on solutions?

Milestones	Estimated Timeline
Complete studies using facility modeling techniques to determine the effectiveness and impacts of using calorimetric assay to measure materials at various points within a reprocessing facility. (Q1)	1 year
Complete research to determine the necessary design requirements for a calorimeter and identify candidate calorimeter designs which can currently meet or be adapted to meet these design requirements. (Q2)	1 year
Complete research using measurements and calculations to determine methods for minimizing calorimetry measurement times. (Q3)	1 year
Complete research to determine the ability of performing calorimetric assay on solutions. (Q4)	2 years

4.1.18 Enhanced Gamma-Ray Spectroscopy Algorithms for Isotopic Analysis

Advanced Fuel Cycle Initiative (AFCI) product materials present new challenges for the NDA technique of isotopic analysis using gamma-ray spectroscopy. A key distinguishing feature between past materials-analysis experience and AFCI materials is the likelihood of increased concentration of minor actinides promoted by higher burn-up and used-fuel recycling. Many of these minor actinides have relatively intense gamma-ray emissions and can therefore complicate the determination of the plutonium and uranium isotopic in two primary ways:

1. Minor-actinide emissions can directly interfere with the plutonium and uranium gamma rays and make it more difficult to cleanly resolve peaks and obtain accurate isotopic analysis, even when using high-resolution spectrometers.
2. Although absolute mass of minor actinides may be low, the activity and gamma-ray yields can be high, resulting in a significant increase in overall count rate, and therefore underlying Compton

continua, underneath the plutonium and uranium peaks of interest. This degrades the signal-to-noise ratio, thereby degrading the precision of isotopic analysis. Longer measurement times may be required to reach desired isotopic uncertainties.

To better understand the challenges the new AFCI materials might present, the Safeguards Campaign undertook measurements of minor actinide-bearing mixed-oxide (MA-MOX) pellets using a planar high-purity germanium (HPGe) detector and a coaxial HPGe detector [Santi 2007a]. Figure 16 shows an example spectrum from the MA-MOX pellets (^{237}Np/Pu ratio of 0.1 and a ^{241}Am/Pu ratio of 0.15) as measured using the planar detector. The isotopic analysis software FRAM [Sampson 1989] was used to analyze the collected data to indicate how the current generation of isotopic analysis codes might perform for materials containing relatively high levels of minor actinides.

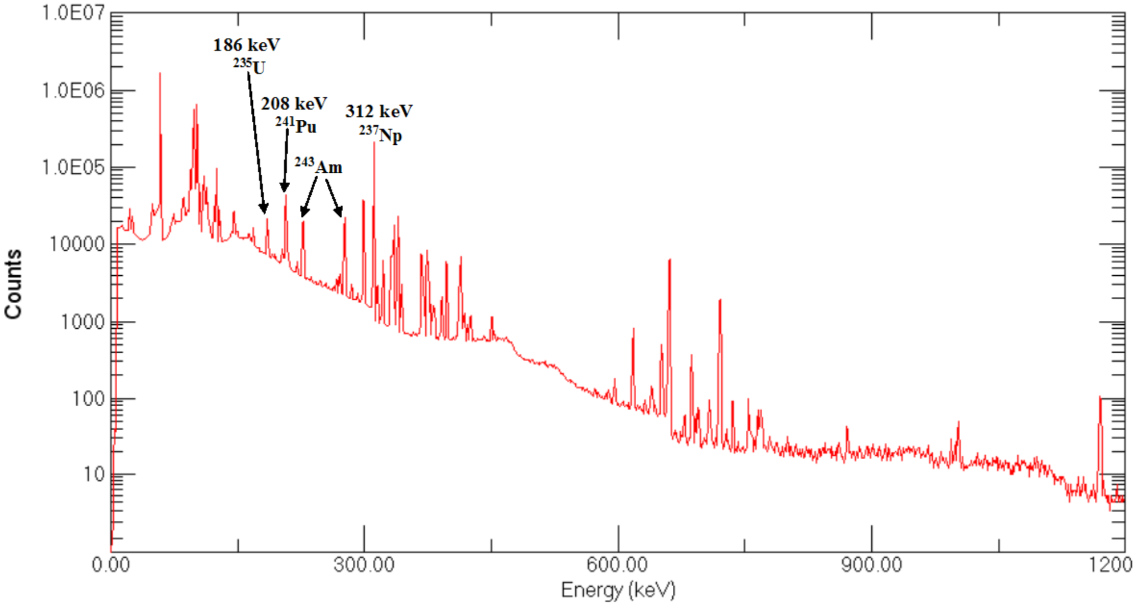

Figure 16. Spectrum measured using a planar detector of the MA-MOX pellets.

Observations from this initial evaluation of MA-MOX fuel pellets indicate the current status of gamma-ray spectroscopy algorithms for safeguards, and where additional research is needed.

1. For this specific sample, FRAM was able to analyze the MA-MOX spectra using the gamma rays in the 120-500 keV energy region, an energy region key to Pu and U isotopic analysis. There were no significant direct interferences, but the background continuum was elevated by higher-energy emissions from the minor actinides.
2. The ^{240}Pu effective precision was about 10% for a 2-hour acquisition time. This is about a factor of ten worse than that of the normal plutonium or MOX.
3. The x-ray region (~90 keV) often offers the best precision for elemental composition, but due to interferences with minor-actinide lines, that region was not accurately analyzed by FRAM.
4. The higher-energy (200-1,010 keV) region analysis was complicated due to the higher-energy emissions from ^{241}Am at 662 keV and 722 keV, and therefore the isotopic results based on this region were poor.

The MA-MOX measurements highlight the need to assess how today's isotopic analysis algorithms, for example FRAM and MGA [1990] might be improved to accurately assay the types of materials likely to be produced in the proliferation-resistant advanced fuel cycles. For example, the fact that minor-actinide emission may completely obscure regions used heavily by today's algorithms, but leave other regions accessible, means that algorithms could be designed to be more adaptive. Furthermore, it is desirable to quantify MAs such as Np and Am.

The continued development of gamma-spectroscopy algorithms is important to a comprehensive safeguards program because spectroscopy-based isotopics are often a key complement to other methods as a means to achieving the overall goal of fissile mass accountability values. For example, gamma-spectroscopy-based Pu isotopics can be coupled to singles and/or coincidence neutron rates to determine the absolute mass of Pu in a sample.

Potential Impact
Improved ability to directly measure U, Pu isotopics and elemental composition for actinide-bearing material flow in advanced fuel cycles

Needs That Could be Addressed if Successfully Developed
Direct Pu measurement in actinide-bearing materials
Direct measurement of Pu in reprocessing streams
Nuclear data measurements
Rapid laboratory sample analysis

Research Questions
1. What are the most problematic actinides in terms of U, Pu masking and what analysis regions are most impacted by their presence?
2. For a range of prospective advanced nuclear fuel cycle material types (*e.g.*, MA-MOX), with a range of actinide concentrations, what are the expected limits of accuracy and precision for isotopics and elemental composition, when assuming HPGe spectrometers?
3. Can algorithms be enhanced to recognize the presence of major interferences and adapt the methods accordingly?
4. How would algorithms need to be adapted to accommodate ultra-high-resolution spectrometers, and do they enable improved performance?

Milestones	**Estimated Timeline**
Identify the minor actinide isotopes and concentrations likely to arise in prospective advanced fuel cycle material flows. (Q1)	1 year
Using simulated assay of representative materials matrices as a basis, perform a study of algorithm performance to identify gaps and research needs. (Q2)	2 year
Develop enhanced analysis methods as necessary, and reevaluate the limits of performance. (Q3)	4 years
Explore the impact of ultra-high-resolution spectrometers, and accompanying isotopic analysis algorithms, through simulation and measurement. (Q4)	4 years

4.1.19 Detection Instrumentation Modeling Methods

Modeling and simulation is used in virtually all stages of the technology development lifecycle, from the early exploratory R&D to the deployment stage. The modeling tools for radiation detection scenarios, being quite mature and widely used, are particularly well-integrated into that lifecycle. For example, modeling can be used to assess the viability of an on-line gamma-spectroscopy instrument aimed at extracting a weak signature from a strong background. Once candidate systems are identified, modeling is used for design optimization (*e.g.*, designing collimation around a gamma-ray spectrometer to minimize background effects). The performance of conceptual and deployed systems can then be predicted by putting them in virtual environments similar to those expected in a fuel reprocessing plant or a used-fuel storage pool, for example.

Radiation transport modeling methods used in the radiation detection community can be broadly categorized as stochastic (Monte Carlo) or deterministic. Familiar examples of the former used frequently by the radiation detection community are MCNP [MCNP—a General Monte Carlo N-Particle Transport Code 2003], COG [Buck 2002], and GEANT4 [Agostinelli 2003]. Monte Carlo codes allow tremendous geometric flexibility, have well-tested cross-section libraries, and simulate the physics of the problem on a particle-by-particle basis. However, since individual particles must be tracked through each interaction, Monte Carlo codes can be very slow for large problems with significant scattering or attenuation where few of the emitted particles ever reach a detector region. Additionally Monte Carlo solutions are not global; the characterizations of the radiation field are only obtained in regions that are selected prior to the calculations.

Examples of three-dimensional deterministic transport codes include TORT [Rhoades 1997], PARTISN [Alcouffe 2005], and RADSAT [Smith 2008, in press]. These solve the linear Boltzmann transport equation numerically by discretizing in space, angle, and energy. Deterministic codes offer several advantages that motivate efforts to apply them in complex radiation detection problems. First, the solution is global–at the completion of a calculation, the angular flux is known in every element of the computational mesh so that a detector could be “walked” through the problem during post-processing. Second, there is no statistical noise in the solution; the solution represents “infinite real measurement time,” which is particularly attractive for simulation of long-dwell and/or high dose-rate detection scenarios. The third, and perhaps greatest advantage, is that deterministic methods can be extremely fast compared to Monte Carlo methods for scattering- and attenuation-dominated problems such as those with large-volume or highly shielded sources. These speed advantages are particularly notable for code packages that utilize one-dimensional transport, such as GADRAS [Mitchell 2008].

In addition to the radiation transport algorithms, there are complementary modeling tools that support the definition of source terms and material composition needed to build realistic models of the materials being interrogated. For example, SOURCES-4C [Wilson 2002] can calculate the neutron spectra produced in a wide variety of reactions (*e.g.*, alpha, n reactions from uranium and plutonium-bearing matrices and spontaneous fission), and codes like ORIGEN and CINDER calculate isotopic production and depletion. For gamma-ray spectroscopy applications, automated means for defining gamma-ray source terms are needed. Codes such as RadSrc (Rad Source) [Hiller 2007] and GADRAS are capable of such calculations, but may need to be refined or expanded to meet the specific needs of new radiation transport packages.

While many of the tools in the radiation detection and nuclear engineering community are relatively mature in their specific application areas, additional investment is needed to enhance and refine them for the specific needs of safeguards applications. Some potential examples of those enhancements include

- Development of improved variance reduction methods for the calculation of neutron coincidence system responses. Deterministic methods to calculate neutron coincidence distributions also warrant further investment because they have the potential to estimate neutron coincidence system responses very rapidly [Mattingly 2008].
- Development of hybrid deterministic/stochastic and fully deterministic differential/integral transport solvers. Deterministic methods to solve the differential transport equation in three spatial dimensions generally perform poorly in low-scattering regions due to ray effects. In contrast, neither deterministic methods to solve the integral transport equation nor stochastic methods suffer from performance degradation due to ray effects. Hybrid codes that adapt their solution method to the density of each spatial region promise substantial performance enhancements of current codes.
- Materials library for materials important to safeguards applications such as oxide fuel variants, shielding concretes, detector-component materials. This library could be used to automatically generate the material input information for transport codes (*e.g.*, materials cards for MCNPX).
- Improvements in burn-up/depletion modeling, utilizing the latest supporting empirical data and providing fidelity (*e.g.*, radial variation in fuel pins) important to nondestructive assay system modeling.
- Testing and evaluation of deterministic transport methods for safeguards scenarios that are particularly time consuming using Monte Carlo methods (*e.g.*, the highly-scattering matrix of a lead-slowing-down spectrometer used in used fuel assay.

Potential Impact
More rapid and accurate instrumentation modeling methods will enable more confident evaluations of concept viability and predicted system performance. All phases of instrument design and evaluation will benefit, including Safeguards Performance Modeling efforts that rely on accurate estimates of statistical and systematic uncertainties of instrument performance.

Needs That Could be Addressed if Successfully Developed
Detection instrumentation modeling and simulation is a cross-cutting activity that will provide benefits in Advanced Instrumentation, Advanced Control and Integration and in Safeguards by Design.

Research Questions
1. Based on a survey of instrument developers, modelers and systems-level analysts, what are the needs and gaps?
2. What are the highest-priority needs to address, and what is the estimated impact of modeling improvements in that area?

Milestones	**Estimated Timeline**
Form working group and perform survey of instrument modeling needs. (Q1)	3 months
Prioritize needs and develop execution roadmap with budget estimates. (Q2)	6 months
Complete tasking as funding levels allow	ongoing

4.2 Advanced Control and Integration

4.2.1 Statistical Process Monitoring

Statistical process control and monitoring are large and relatively mature topics outside of nuclear safeguards [Choi 2008; Crosier 1988; Chen 2002]. During FY08, progress was made on evaluating a small subset of options to monitor various data streams at reprocessing facilities, such as tank level, volume, density, and temperature taken in near real time from in-tank dip-tube pressure sensors. In this discussion, we assume a context in which

- Many data streams will be monitored on different time scales from many different facility processing and storage areas;
- Periodic decisions are required regarding whether the facility is operating as declared, and if not, identify the most likely misuse scenario(s);
- In the international context, the possibility of data falsification must be considered among possible misuse scenarios [Burr 1994].

In this context, several research areas have emerged as a result of the FY08 efforts, including

1. the need for data-driven pattern recognition (operating as declared or misuse A or misuse B or period-driven (at the end of each day or balance period, make a judgment)
2. the need to contrast data-driven decision making from time- or period-driven decision making
3. methods to accommodate within-data-type temporal correlations together with between-data-type correlations
4. data fusion at feature, score, or decision level to reach overall decision [Dass 2007]
5. issues involving multiple testing and associated false alarm rates [Burr 2003]. Allowing a high “false discovery” rate might be acceptable, provided there is efficient, timely anomaly resolution, such as J. Howell’s model-based diagnostic methods [Howell 1997].
6. Regardless of whether anomaly resolution is used, there will always be a need to assess false negative rates for a given low overall false-positive rate (low false-positive rate after anomaly resolution if anomaly resolution is invoked as part of the procedure) and misuse data will usually require realistic simulation, so quantifying and dealing with computer model uncertainty will be a major issue [Higdon 2008].

Regarding 1), there are many established pattern-recognition methods, some of which have been applied in safeguards’ contexts. Other methods continue to be developed.

Area 2) involves the unexplored link between process monitoring and nuclear material accounting, which typically involve decision making at pre-established times, such as interim inventory periods or periodic clean outs (material balance periods).

Challenge 3) involves both between- and within-variable patterns, thus complicating the types of pattern recognition tools required.

Regarding 4), a relevant empirical study of data fusion for biometric devices involves the same generic questions: to what extent should data from the individual data streams be preprocessed prior to fusing with other data sources. An extreme form of preprocessing, for example, is to convert the data from a given data stream into a vote regarding whether the facility is operating as declared.

Several ongoing efforts fall under 5), but more needs to be done. J. Howell's concept [Howell 1997] is perhaps the most tested and mature, involving using model-based reasoning to resolve anomalies with the ultimate goal of still correctly labeling each time period as "operating as declared" or not.

Finally for 6), the required quality of simulated misuse scenarios superimposed onto real or simulated normal facility data is a difficult question [Durst 2007a]. One current thought is a relatively simple perturbation study that varies model assumptions and parameters that are used to generate data from each misuse scenario.

Potential Impact
Large volumes of process monitoring data must be efficiently and effectively evaluated to segregate facility operations into "normal," "normal but atypical," "misuse," or "unknown."

Needs That Could be Addressed if Successfully Developed
Process monitoring of reprocessing streams
Data collection, integration, semi-automated evaluation

Research Questions
1. How to handle data streams having different time scales? Consider allowing the most-frequently updated data sources to "trigger" evaluation of other data sources, and consider other options.
2. Combine data streams at feature, score, or decision level to reach overall decision regarding "operating as declared" or not.
3. Compare "data-driven" to "time-driven" decision rules. A "time-driven" rule makes a decision at the end of pre-chosen periods (such as material balance periods). A "data-driven" rule monitors the data assuming "normal plant operation" until the data suggests either a restart of the evaluation period or that "abnormal plant operation" occurred.
4. What are best options to accommodate within-data type temporal correlations and between-data type correlations?
5. How accurate should simulated misuse scenarios be? Consider computer model uncertainty.

Milestones	**Estimated Timeline**
Empirical study via simulation to evaluate options for data streams having different time scales (Q1)	1 year
Empirical study via simulation to evaluate options for how to combine ("fuse") data streams (Q2)	1 year
Empirical study via simulation, preferably using real data and real data that include the effects of simulated misuse (Q3 and Q4)	2 years
Decision point: Are the empirical studies using simulation adequate to recommend the chosen method(s) in Q1–Q4? This involves Q5. It is anticipated that related efforts in computer model uncertainty evaluation can be modified to help with the decision point question.	3 years

4.2.2 Multi-Isotope Process (MIP) Monitor

Modern industrial reprocessing techniques, including the PUREX and UREX+ family of separations technologies, are based on solvent extraction between organic and aqueous phases. In these bi-phase systems, product (actinide) and contaminant (fission and activation products) elements are preferentially driven (thermodynamically) to opposite phases, with small amounts of each remaining in the other phase [Benedict 1981]. The distribution of each element, between the organic and aqueous phases, is determined by major process variables such as acid concentration, organic ligand concentration, reduction potential, and temperature. Hence, for consistent performance of the separation process the distribution of each element between the organic and aqueous phases should be relatively constant. During “normal” operations the pattern of elements distributing into the product and waste streams at each segment of the facility should be reproducible, resulting in a statistically significant signature of the nominal process conditions. Under “abnormal” conditions, such as those expected under some protracted diversion scenarios, patterns of elements within the various streams would be expected to change measurably. The MIP monitoring approach utilizes changes in the concentrations of gamma-emitting contaminants as evidence of changes to the process chemistry [Smith 2007; Schwantes 2008; Orton 2008]. In-process surveillance by the MIP monitor is accomplished by coupling the gamma spectrometry of the streams with Principal Component Analysis (PCA). PCA is a chemometrics tool that finds combinations of variables (principal components or PCs) that best describe the variance between differing datasets [Malinowski 2008]. The MIP technique is then capable of evaluating the patterns of the gamma-emitting contaminants for statistically relevant signs of potential changes to the process chemistry [Smith 2007; Schwantes 2008; Orton 2008].

Initial modeling and experimental efforts have suggested that the MIP concept may be viable for monitoring process conditions. The modeling efforts include the use of ORIGEN-ARP [Croff 1983] to provide a list of isotopes contained in used fuel. The simulated used fuel composition is then used in the AMUSE (Argonne Model for Universal Solvent Extraction) [Regalbuto 2004; Hensley 1995] code to calculate the expected distribution of the elements, reported as the relative molar concentrations, into the organic and aqueous phases. This information is then fed into the Synth [Hensley 1995] or SuperSynth code to simulate the gamma spectra that one would obtain from the feed, aqueous and organic solutions. Experimentally, the MIP concept is currently being tested on two different dissolved spent-fuel assemblies that have undergone solvent extraction at different acid concentrations.

Examples of recent modeling efforts and how they support the evaluation of the MIP concept are shown in the figures below. Results of the burn-up and decay of used fuel from a moderately burned pressurized water reactor (PWR) assembly were obtained from ORIGEN. Those results were then fed to AMUSE to calculate the elemental distribution into the aqueous phase after solvent extraction as a function of acid strength. Resulting gamma spectra were then generated for the aqueous, feed and raffinate streams. The gamma spectra simulations (Figure 17) were based on a small Cd-Zn-Te (CZT) detector that will be used in the laboratory demonstration. The solvent extraction process was simulated for various acid concentrations and the spectral data from these simulations were analyzed using PCA to identify trends in the spectral patterns. In addition to various acid strengths, various burn-ups were also simulated. The resulting PCA plots are presented in Figure 18.

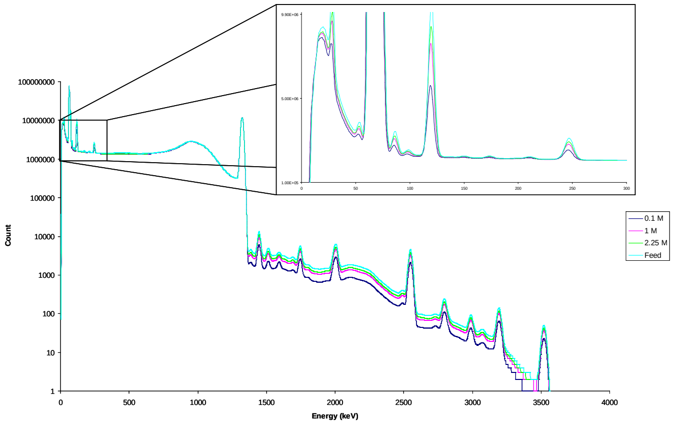

Figure 17. Simulated spectra from a CZT detector of the feed and raffinate of the first stage of the PUREX separation of a 28 MWd/kgU PWR rod. One gram of U was modeled and the uranyl nitrate concentration in the feed for all separations was 1.3 M.

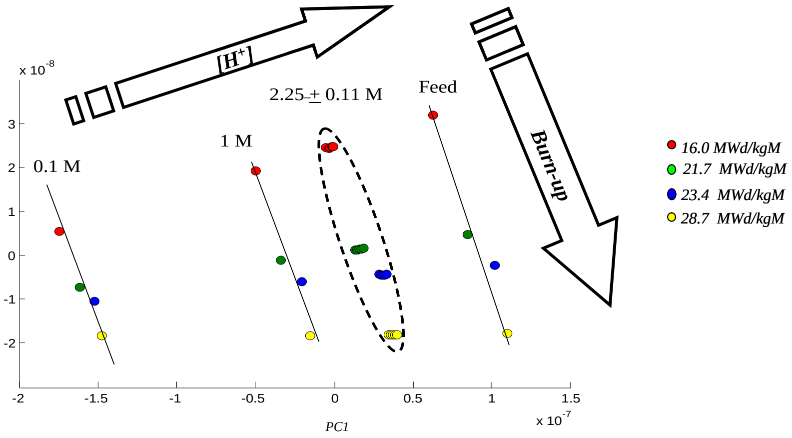

Figure 18. Case Study: MIP Monitor utility for distinguishing burn-up of dissolved fuel and changes in acid concentration. 10% variance in acid strength used to represent "industrial variations" of 1st stage of PUREX/UREX flowsheet ([H+] = 2.25 M). CZT detector; 3600s count time.

Figure 17 shows the Synth simulated spectra that are to be expected from a CZT detector when surveying the gamma emissions from dissolved used fuel and the aqueous solutions after solvent extraction processing. The figure clearly shows how the spectra visibly change as a function of [H+] in the aqueous feed stream during solvent extraction. When spectral changes as a function of fuel burn-up are modeled and considered using PCA, one can extract information about burn-up as well as acid strength. Figure 18 illustrates how information on burn-up and acid strength can be extracted from the MIP Monitor in PCA space. To do this effectively, preprocessing of the spectral data was necessary to enhance differences as a function of the two variables of interest: burn-up and [H+]. Figure 18 also illustrates how PCA might be attenuated to identify changes in process variables of interest within a process. It is envisioned that a statistical alarm limit could be set around the PCA points, shown in Figure 18, to signal when process conditions have deviated from nominal conditions.

Only preliminary results from experimental studies are currently available. However, these results do provide some credence to the modeling results, suggesting the MIP Monitor will be sensitive to changes in acid concentration, as well as burn-up and/or cooling time.

Potential Impact
Cost-effective, nondestructive, near-real-time, autonomous monitor of process conditions at a reprocessing facility

Needs That Could be Addressed if Successfully Developed
Process monitoring of reprocessing streams

Research Questions
1. Do experiments confirm simulations of the expected sensitivity of MIP to process conditions?
2. What are the experimentally determined sensitivities of MIP to changes in major process conditions?
3. Can PCA be "tuned" to extract useful information (burn-up, acid strength, etc) from measured gamma patterns?
4. Are there design/operability considerations that need to be addressed for deployment in a high-background facility?

Milestones	**Estimated Timeline**
Demonstrate MIP Monitoring technology meeting performance objectives. (Q1)	1 year
Sensitivity testing from simplified lab experiments benchmarked against model predictions. (Q2)	1 year
Comprehensive study of MIP Monitor sensitivity to all major process variables. (Q2&3)	2 years
Design and construction of prototype for high background facility operation. (Q4)	3 years
Deployment of prototype to a pilot-scale facility. (Q4)	3.5 years

4.2.3 Macroscopic Properties Monitoring

Increasing safeguards effectiveness in used-fuel reprocessing plants is a great challenge to the national and international communities. The International Atomic Energy Agency (IAEA) has established international safeguards standards for fissionable materials at the reprocessing plants to ensure that significant quantities of weapons-grade nuclear material are not diverted over a specified time frame. Because proliferant diversions are possible via deliberate modification of the flowsheet chemistry, it is necessary to confirm proper operational performance to verify facilities operate under adequate safeguard-declared conditions. In any reprocessing facility, variability in process is expected under normal operating conditions. Diversions of fissile materials would cause off-normal variations detectible in real-time with this monitoring platform. On-line real-time monitoring of the flowsheet radiochemical streams provides a unique capability to rapidly identify unwanted/suspect deviations from normal operation conditions.

This multiparametric monitoring includes measurements of physicochemical parameters of the radiochemical streams of variable compositions and identification of unique signatures of the separations process that is operating at normal conditions. Coupling the real-time measurements with the ability to search chemical libraries of physicochemical, spectroscopic, and chemical properties provides a path for detecting modified or unwanted chemical agents in the various process streams.

The mobile chemical behavior of Pu, which depends on the presence of acid, nitrate, solution ionic strength, temperature, etc., allows prediction of its concentration in the nuclear fuel cycle process streams based on the measurements of the macroscopic properties. Existing Pu signature libraries will be expanded as applicable to the reprocessing conditions and needs. For example, solutions containing high concentrations of U, complexing agents, transition metals, and fission products will be measured. Statistical analysis and modeling will allow calculation of Pu concentration based on these measurements. Chemometric modeling will be used to process the data. This new methodology could be universally applied to aqueous and organic solutions and could be easily integrated with other on-line measurements such as spectroscopic and radiological.

A demonstration of this methodology has been accomplished by the coupling of physical property measurements with spectroscopic measurements for the predictive measurement and analysis of chemical components within Hanford tank waste streams. The on-line monitoring of waste streams was successfully demonstrated by combining spectroscopic measurements with physiochemical measurements (conductivity, density, temperature) in real-time quantitative determination of chemical components in the waste [Bryan 2005]. This new on-line monitoring system, which features Raman spectroscopy combined with a Coriolis meter and conductivity probe, has been recently developed to provide immediate chemical data and flow parameters of high-level radioactive waste streams [Bryan 2007]. This process monitoring system uses two models to predict the concentration of components of solutions of high-brine/high-alkalinity wastes during retrieval from Hanford waste storage tanks. The spectroscopic data were fit using a Classical Least Squares model (SaltPred®) to predict the concentrations of nitrate, nitrite, chromate, aluminate, phosphate, sulfate, carbonate, and hydroxide salts. The physiochemical data (conductivity, density, and temperature) were fit using a Partial Least Squares model (NaPred®) to predict the sodium concentration for the retrieved waste streams. Figure 19 shows the measured sodium values versus fitted predictions for several hundred different waste stream simulants.

It is feasible to extend the on-line methodology to fuel reprocessing streams for the predictions of U and Pu. Equations have been derived using density, conductivity and temperature measurements for the determination of Pu, U, and nitric acid concentration within fuel reprocessing solutions [Clark 2002; Sakurai 1996; Gildseth 1972; Kumar 1998; Charrin 2000; Yamamoto 1988]. The derived equations correlating Pu concentration to density have included the effects of temperature, nitric acid, and in some cases, uranium [Gildseth 1972]. More than 700 composition and density data taken from technical literature were fit to the derived equation, yielding, for the Pu(IV)/U(VI)/$HNO_{3(aq)}$ system, an equation that is valid for all combinations of Pu(IV), U(VI), and HNO_3 in water with a 0.75% relative uncertainty.

Other process monitoring technologies can be combined with physical parameter measurements to reinforce the ability to predict fuel reprocessing solution compositions. Two complimentary systems are the Multi-Isotope Process (MIP) monitor and the spectroscopy-based monitoring (SBM) system–funded under the Global Nuclear Energy partnership (GNEP) Safeguards and Separations Campaigns respectively. The MIP Monitor is designed to take advantage of the gamma signal of process streams to identify spectral patterns that shift with changes in process chemistry. Spectroscopic-based monitoring (SBM) using UV-vis-NIR and Raman spectrometers continuously monitor chemical compositions of the process streams including actinide metal ions (U, Pu, Np), selected fission products, and major reagent chemicals.

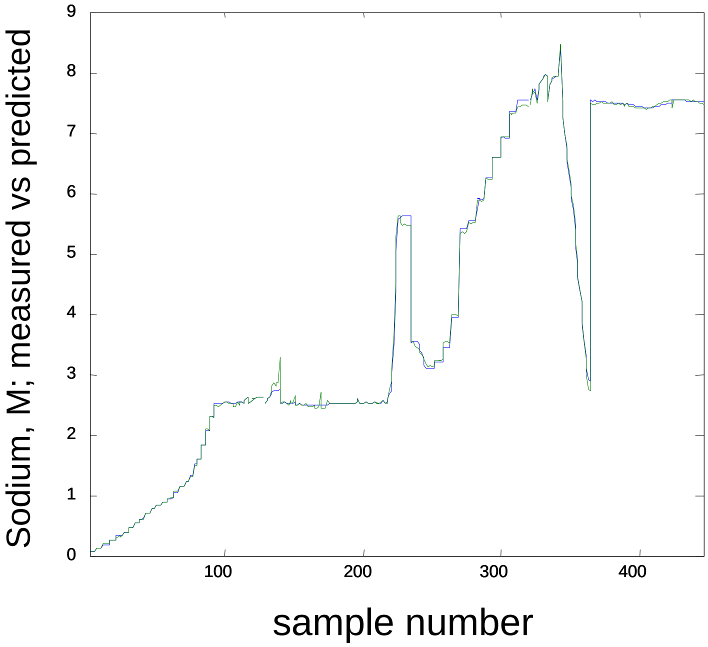

Figure 19. Partial Least Squares (PLS) model used to predict sodium concentration from conductivity, density and temperature data from multiple Hanford tank waste simulants. Data (blue line) and fitted (green line) values displayed; this data represents several hundred different waste simulant compositions.

Potential Impact
Nondestructive, timely, direct measurement of Pu and U content within reprocessing streams without the need for operator-declared information about the fuel assembly

Needs That Could be Addressed if Successfully Developed
Direct measurement of Pu in reprocessing streams
Process monitoring of reprocessing streams

Research Questions
1. Can Pu be determined within simulant reprocessing solutions?
2. Can modeling of simulants accurately capture the subtleties and differences in the Pu and U assay from actual used fuel of various origin and burn-up?
3. If previous inquiry justifies a prototype facility that can assay irradiated fuel assemblies, what is the appropriate design and where should it be sited?
4. Could the accuracy and precision achieved with prototype-system assays of a wide range of fuels significantly enhance materials accountability in advanced fuel cycles?

Milestones	**Estimated Timeline**
Deploy testbed to support simulant measurements. (Q1)	1 year
Demonstrate Pu and U measurements on simulant feeds under processing conditions. (Q1)	1 year

Measure and model of actual used fuel. (Q2)	2 years
Decision point: Adequate performance of Pu and U measurements in actual fuel motivates full-scale prototype?	2 years
Design of prototype monitoring system capable of assaying reprocessing fuel solutions (Q3)	3.5 years
Complete empirical measurement campaign and prototype process monitoring system demonstration. (Q4)	4 years

4.2.4 Neutron Balance

A neutron balance is a proposed new approach to safeguarding among the front-end facilities in a used-fuel processing cycle. The methodology applies to UREX+, PUREX, and electrochemical (pyro) processing. A neutron balance can involve integrating the safeguards among the following:

- used-fuel storage in reactor pool
- interim dry storage facility for used fuel assemblies
- wet storage at the reprocessing facility
- head-end hot cell for shearing assemblies.

It can also be used to give confidence that the material that left one facility arrived at another. The method is based on the measurement of the total neutron emission rate. The usefulness of this measurement is built on two facts: 1) The spontaneous fission neutron emission is almost entirely from ^{244}Cm (> 96%), hence the change in the neutron emission with time varies as the half-life of this isotope. 2) Pu and Cm are intimately connected while the Pu is in used fuel assemblies; meaning if Pu is removed, it is likely that ^{244}Cm is removed as well [Menlove 2006].

The temporal scope of the “neutron accountancy” might span from discharge until the fuel is dissolved in the input accountability tank. It is also possible to use the balance concept between point-to-point transfers. The remainder of this discussion will focus on using the neutron balance for the broader scope of providing an integrated safeguards approach among the locations listed above. Such a system could operate in an unattended mode; the measurements could be integrated into regular fuel movements. The balance is based on the conservation of neutrons; since ^{244}Cm is the source of the vast majority of the neutrons, this balance could be called a ^{244}Cm balance. The term neutron balance is more appropriate since it includes the small contribution of Pu spontaneous fission and neutrons generated from multiplication.

There are three primary reasons that the underlying assumptions of the neutron balance concept might be violated: 1) Significant neutron emission arises from isotopes other than ^{244}Cm, 2) multiplication varies slightly in time due to the contributions of ^{241}Pu, and 3) multiplication varies due to the medium in which the assemblies are measured. Each of these effects is expected to be small and could be accounted for as described below.

Regarding 1), since ^{240}Pu dominates the non-^{244}Cm spontaneous fission source, inclusion of this one isotope would maintain the known time dependence of neutron emission. The contribution of ^{240}Pu to the total neutron emission could be estimated from the singles neutron count rate. If the total ^{240}Pu can be estimated to an accuracy of ~10%, the uncertainty in the total neutron source variation with time would be impacted by ~0.5%. Regarding 2), the multiplication in the assembly is dominated by ^{235}U and ^{239}Pu. The impact of ^{241}Pu, the only multiplying isotope that changes significantly over the time scales of interest, can be estimated and corrected for by the singles count rate as discussed above for ^{240}Pu. Uncertainty in the ^{241}Pu content will result in a few percent of uncertainty in the overall neutron emission. By measuring in air the multiplication the significance of this uncertainty can be reduced if desirable. Regarding 3), this uncertainty can be eliminated by always measuring in the same medium (water, borated poly, air). If this is not possible, corrections for the change in medium should be possible within a few percent uncertainty. Note that the ^{240}Pu and ^{241}Pu corrections discussed above would not require unique burn-up code calculations. For a given assembly type, a general correction for each of these isotopes as a function of the singles rate would be accurate enough.

In aggregate, it is anticipated that the total neutron emission can be estimated to 5% or better. It is important to note that over the time scale involved in shipping an assembly between any two locations, the neutron count rate does not change at all. For these cases, the singles count rate would likely have an uncertainty of less than 2%; that for counting statistics would be much less than 1%, however the uncertainty will be determined by systematic uncertainties and calibration.

In the context of dry storage, assemblies would be measured before loading into the cask as well as when they are removed. It is not thought possible to include a measurement of an entire dry storage cask in the balance.

The neutron balance will likely provide assurance of the absence of diversion over the many years (reactor pool, dry storage, etc.) that plutonium exists in assemblies – from the discharge of the used fuel until the head-end shearing cell where item accountancy changes to bulk accountancy. Since one meter of a used fuel assembly emits ~100 million neutrons per second, a 0.01% efficient detector would achieve 1% counting uncertainty in 1 second.

One of the strengths of this technique is that no mass is being quantified. Hence, the uncertainty in going from a count rate to mass is not present. The initial count rate is taken as the starting point so that the only parameter needed to apply the neutron balance idea is the half-life of the Cm isotopes responsible for the majority of emissions. Furthermore, a neutron balance could take advantage of C/S measurements used to track fuel assemblies through the head end of a reprocessing plant and other key measurement points used to support traditional Shipper Receiver Difference measurements. High-energy neutrons are ideal for such a balance due to their penetrability through relevant materials: the fuel assemblies, storage containers, and shielding material.

Potential Impact
Self-consistent monitoring of used fuel over many years - from the fuel leaving the reactor until it enters the input accountability tank

Needs That Could be Addressed if Successfully Developed
Physics-based continuity of knowledge for used fuel

Research Questions
1. How much does the neutron intensity change throughout the fuel cycle? In particular how do the neutron emitting sources in used fuel change over the first couple of years after discharge as a

	function of initial enrichment and burn-up?
2.	How well does the neutron balance concept work for a real facility?

Milestones	Estimated Timeline
Complete quantification of the neutron intensity for the measurement locations of application to the neutrons balance. In particular quantify the neutron emitting sources in used fuel over the first couple of years after discharge as a function of initial enrichment and burn-up. (Q1)	2 year
Complete installation of a prototype neutron balance system in a representative fuel handling facility. (Q2)	4 years

4.2.5 Automated Radiochemistry

Destructive analysis is a fundamental and often critical component of radiochemical measurements. Conventional laboratory-based destructive analysis is a cornerstone of materials balance estimates for nuclear fuel cycle facilities, and offers excellent precision in many cases, but can require days to weeks to get results. Faster automated methods are needed to support on-site laboratories, and to move destructive analysis methods from the laboratory to on-line monitoring applications. Automation may also support IAEA laboratories involved in safeguards.

Destructive analysis is normally carried out in fixed laboratories using manual sample preparation, separation, and measurement techniques. For mass-spectrometric measurement methods, the analyte must be separated from matrix components that may generate isobaric or molecular interferences. For radiometric methods, the analyte should be radiochemically purified prior to measurement to address matrix effects, spectral interferences, and prepare thin sources. Although gamma-emitting radionuclides may be determined by non destructive analysis in some measurement scenarios, separations may still be required in many cases if the gamma emissions are weak and/or the sample matrix is radiologically complex.

Advances in radiochemical automation offer many opportunities to address needs in nuclear reprocessing and safeguards. Automation can reduce time, costs, and worker doses while improving safety in existing measurements, and also enable measurements not currently considered feasible (*e.g.,* too difficult to put on-line or too costly and time-consuming for manual laboratory analysis). Hence, it would be shortsighted to consider automation only in the context of existing measurements, where it may reduce human radiation exposure and manual labor. New automated radiochemical measurement capabilities may enable new measurements–and measurements at new locations (*e.g.*, on-line) – to support safeguards objectives. The ability to automatically separate radionuclides of interest from the background and from each other may allow radiation detection techniques currently used just for nondestructive analysis to be more informative or accurate.

Automation is emerging as an increasingly important area of radioanalytical chemistry, and developments in this area can be used as a basis to develop automated analysis methods to support safeguards. Currently radiochemical automation is either practiced or under development in the following areas:

- IAEA laboratories have automated aspects of the analysis of used fuel using robotic arm technology in radiological glove boxes [Beugelsdijk 1998; Brandalise 1994; Zahradnik 1996; Ziegler 1999].

- Heavy-element researchers automate the rapid separation and measurement of short-lived heavy isotopes generated from target samples [Gaggeler 1997; Zvara 1999; Schaedel 2007]. This method has been instrumental in the discovery of several new elements and isotopes [Wilk 2008] [Oganessian 2006].
- Automated methods have been developed to support the analysis of samples related to environmental management of U.S. DOE sites, linking automated radiochemical separations to radiometric or mass-spectrometric detection [Grate 1998; Grate 2003; Grate 2008a].
- Automated process monitoring instrumentation has been developed for nuclear waste processing. An instrument for determination of ^{99}Tc in Hanford nuclear waste can perform sample preparation, separation and detection all in less than 15 minutes for on-line analysis [Grate 1998; Grate 2003].
- Radiochemical sensors have been developed for detection and determination of radionuclides in water and process samples [Grate 2008b; Hofstetter 1999].
- Automation methods are under development to support analyses of environmental samples related to nuclear nonproliferation [Auer 2004].
- Medical isotope production requires separation of short-lived daughter products from their longer-lived parent isotopes. Interest in new isotopes and the desire to perform this separation at-site are leading to increasing interest in automated radiochemical separation units.
- Requirements for the laboratory analysis of large numbers of potentially contaminated samples have motivated research into the automation of radiochemical analysis. These samples may be from contaminated environmental sites or bioassay samples from an accidental or intentional nuclear event [Lariviere 2008b; Lariviere 2008a].

Automation has been enabled, in part, by the confluence of developments in robotics, digital fluidic instrumentation, and modern radiochemical separation materials. Some of these automation approaches have been reviewed, and comprehensive reviews are published or are in the press [Grate 2003; Grate 2008a].

Development of new semi-automated or automated methods to support safeguards can address measurement applications at IAEA laboratories, at on-site laboratories (at reprocessing plants), at the interface between process sampling and on-site laboratories, and at on-line process locations. The last category can support detection of diversion, process control, and/or provide additional information with safeguards value.

As an example, one of the most pressing challenges to adequately safeguarding a nuclear fuel reprocessing plant is the need for on-line assay of the process solutions, especially the plutonium-bearing solution from the used fuel dissolver [Durst 2007a]. Currently there is no on-line (destructive analysis) monitoring technology to determine the chemical composition of reprocessing solutions, particularly for specific actinides of interest. The best currently available method to analyze the composition of these reprocessing streams is to grab samples and perform conventional laboratory analysis.

The safeguarding of reprocessing methods and facilities has been a first priority for analysis by the ASA-100 Project Team (commissioned by the NA-243 Office of NNSA) [Durst 2007a]. A key conclusion of this team, as stated in their 2007 report, is that on-line monitoring is one of the overarching needs:

> "Develop on-line assay techniques to measure the plutonium, uranium, and actinide content of aqueous process solutions… [because] the concentration of nuclear material in the main process streams and inventory vessels is of fundamental safeguards importance."

Furthermore, the report notes that even with on-site laboratories, "it is still difficult to obtain sample results in a timely manner."

On-line process monitoring could provide near real-time destructive assay of key strategic fluid streams within reprocessing plants, and provide the following advantages if deployed in key measurement points: (1) immediate results, (2) more frequent results, (3) amenable to remote monitoring by immediately relaying results elsewhere, (4) security (monitors could be placed in secured containers), and (5) may reduce the frequency and cost of grab sampling.

Automated process monitoring instrumentation, mentioned above for the determination of total ^{99}Tc in Hanford nuclear waste, can automatically perform sample preparation, separation and detection for the destructive analysis of nuclear processing solutions [Grate 2003; Grate 2008a]. Techniques such as this could be developed for plutonium in acid dissolver solution, for example.

Potential Impact
Timely destructive assay, including on-line process monitoring at key measurement points, as well as streamlined automated sampling and laboratory analysis, will support and improve nuclear safeguards

Needs That Could be Addressed if Successfully Developed
Direct measurement of Pu in reprocessing streams
Process monitoring of reprocessing streams
Rapid laboratory sample analysis

Research Questions
1. What are key "enabling" signatures and associated measurement points for reprocessing facilities and MOX fuel fabrication plants (regardless of whether the final measurement is made on-line or on a sample transported to a laboratory)?
2. What are key needs and opportunities for on-line destructive analysis (DA) measurements? Are there measurements not currently considered feasible which could be enabled by automation?
3. What are the shortcomings of key on-line nondestructive analysis (NDA) techniques in current use? How can DA improve upon these measurements for on-line applications?
4. What are the current DA techniques being utilized at on-site laboratories or IAEA laboratories? Are there presently bottlenecks, inefficiencies, facility or labor issues in these laboratories that could be reduced or eliminated via automation? Could the implementation of automation systems substantially increase throughput of DA samples?

5. What are most promising sample prep/separation/detection combinations for addressing on-line application needs?
    a. What sample preparation chemistries and sample preparation system architectures can be used to automate the initial DA steps for the determination of specific radionuclides in specific sample or process matrixes?
    b. What separation chemistries and automated separation system architectures can isolate the analytes from the prepared matrix and deliver them to detectors to support the determination of specific radionuclides from specific sample or process matrixes?
    c. What detection approaches, in combination with automated sample preparation and separation, provide the required measurement results?
    d. Conceptualize and develop overall architectures to combine sample preparation, radiochemical separation, and on-line detection in integrated analyzer instruments for laboratory and on-line applications, especially where the architecture can be adapted to implement various sample preparation and separation chemistries for each of various analytes of interest.
6. What are the most promising automated methods for sampling, sample preparation, and separation to address DA needs at IAEA and on-site laboratories?
7. Based on benchtop experiments, what levels of analytical performance can be achieved for automated radiochemical methods in various applications, *i.e.*, time required to obtain a measurement result, analytical capacity and throughput, accuracy, and precision?
8. How will this projected performance improve the overall safeguards stance of a facility in terms of typical nonproliferation metrics (*e.g.*, Safeguards by Design metrics)?

Milestones	**Estimated Timeline**
Compile key analytes and measurements points for reprocessing facilities and MOX fuel fabrication plants that are *currently known from prior studies*. (Q1-4)	1 year
Complete a comprehensive look at key analytes, signatures, and measurement points – as well as current methods, shortcomings, and gaps– for all phases of the nuclear fuel cycle, as they relate to on-line measurements, on-site laboratories, and IAEA laboratories. (Q1-4)	2 years
Determine the shortcomings of current NDA techniques that may be addressed by the use of DA techniques, especially where automation enables the DA techniques to be practical. (Q3)	2 years
Analyze current DA techniques, from sampling to laboratory analysis, to determine bottlenecks, inefficiencies, and analytical gaps that may be addressed through automated radiochemistry. (Q4)	2 years
Select and begin applied research and development projects to develop automated DA methodology for *known needs* for on-line analyzers, (Q5) including a. on-line determination of Pu in acid dissolver solution at reprocessing plants b. rapid automated sample preparation system to increase sample throughput in the destructive assay of MOX fuel samples	3 years
Select applied research and development projects to develop automated DA methodology for *additional needs* identified in the comprehensive analysis of analytes, signatures, measurement points, shortcomings and gaps. (Q5)	4 years

For known analytes, such as plutonium isotopes, select applied research and development projects to develop or adapt radiation detection techniques for use as the detection component of on-line DA monitors, where the synergy of the on-line analyzer, which will reduce backgrounds and simplify the matrix, and the detector enable new measurement approaches to meet safeguards objectives. (Q3,5)	4 years
Select applied research and development projects to develop automated DA methodology to address shortcomings of on-line NDA techniques. (Q3,5)	2 years
Select applied research and development projects to develop automated DA methodology to support at-site sampling and analysis needs and IAEA laboratory, focusing on *known needs,* including automated sampling and separation interfaces between on-line sampling points and at site laboratories that reduce the needs for hot cells, simplify or eliminate laboratory sample preparation. (Q6)	3 years
Select applied research and development projects to develop automated DA methodology to support at-site sampling and analysis needs and IAEA laboratory, focusing on for *additional needs* identified in the comprehensive analysis of analytes, signatures, measurement points, shortcomings and gaps. (Q6)	4 years
Determine the experimental analytical performance of promising approaches, based on initial projects. (Q7)	4 years
Determine the experimental analytical performance of promising approaches, based on additional projects. (Q7)	5 years
Assess the impact of DA methods on safeguards, based on performance projected from analytical measurements. (Q8)	5 years
Identify approaches for the development of engineered prototypes. (Q8)	5 years

4.2.6 UV-Visible Spectroscopy

Ultraviolet and Visible (UV-Vis) spectroscopy is a valuable tool for monitoring the concentrations of metals in a process feed. A beam is passed through the solution of interest, and molecules in the solution that interact with the beam absorb energy at discrete wavelengths. Due to molecular vibrational-rotational effects, wavelengths of light on either side of the central absorption wavelength are also absorbed. In particular, this technique has been very valuable for studying the different oxidation states of actinides. This, in turn makes it an ideal tool for nondestructive analyses of actinide-bearing solutions.

In the context of nuclear safeguards, UV-Vis spectroscopy is viewed as potentially valuable in monitoring the status and operations in a complex aqueous chemical processing plant, such as a fuel-reprocessing facility. Two types of applications are envisioned: actinide concentrations in reprocessing streams, and monitoring of important stream parameters. One of the needs stated in this roadmap is for the “direct measurement of Pu in reprocessing streams” with the specific requirement of continuous, on-line monitoring of Pu meeting IAEA diversion and timeliness goals:

- protracted: <1 SQ per month or <1%
- abrupt: near-real-time analysis and <5%.

Recent experiments have shown that continuous, on-line UV-Vis monitoring is valuable in tracking the concentration of uranium in an effluent stream from a UREX flow sheet. The figure below shows the UV-Vis spectra (wavelength vs. absorbance) that is characteristic of U(VI) plotted against time. The major absorbance band between 400 and 410 nm is stable up to about 11:45, when a feed flow rate was changed. The resulting change in the uranium concentration in the product stream is clearly reflected in the spectra shown. While these results support the feasibility of the technique, questions remain as to its value in a complex, actinide-rich solution.

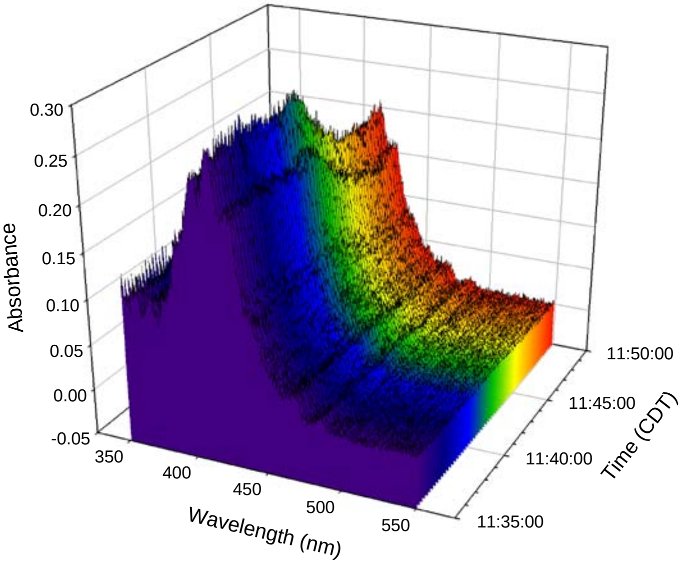

Figure 20. UV-Vis spectra as a function of time in a UREX process stream.

Potential Impact
Nondestructive, timely, continuous measurement of actinide concentrations and oxidation states

Needs That Could be Addressed if Successfully Developed
Direct Pu measurement in reprocessing streams
Process monitoring of reprocessing streams

Research Questions
1. Can specific signals be obtained from spectra of highly complex streams? For example, can Pu and Np be characterized and quantified in the presence of a relatively large concentration of U? This will determine which streams can be monitored to give useful information.
2. Can the spectrometer system be sensitive enough to detect and characterize small concentrations of actinides?
3. Can the uncertainty of the measurement be kept low enough to result so that small changes can be detected?
4. Can the fiber optic interface needed for continuous, real-time operation be designed to work reliably in complex streams, in a hot cell environment and in the presence of high radiation fields?

Milestones	Estimated Timeline
Obtain data for U (VI) at concentration ranges relevant for UREX+ and PUREX in both aqueous and organic phases. (Q2)	1st year
Run process scenarios to increase or decrease the concentration of U (VI) in process streams to determine the sensitivity of the system to relevant changes in stream compositions. (Q3)	1st year
Obtain data for other actinides in the presence of U at concentration ranges relevant for UREX+ and PUREX in both aqueous and organic phases. (Q1, Q2)	2nd year
Run process scenarios to increase or decrease the concentration of other actinides in process streams to determine the sensitivity of the system to relevant changes in stream compositions. (Q3)	3rd year
Develop a simulant formula that includes all of the metals, anions and organic compounds that are UV-Vis active and would be present in process streams. (Q1)	2nd year
Obtain data for all actinides in the simulant at concentration ranges relevant for UREX+ and PUREX in both aqueous and organic phases. (Q1)	4th year
In proximity to unshielded used fuel, obtain data for all actinides in the simulant at concentration ranges relevant for UREX+ and PUREX in both aqueous and organic phases. This will need to be done several times to establish any degradation over time. (Q4)	5th year

4.2.7 Actinide Sensors for Electrochemical Process Monitoring

The majority of process-monitoring technologies discussed in this report are targeted at aqueous reprocessing, as it is the most common fuel reprocessing approach globally, and has been the baseline assumption in the AFCI program. However, the U.S. has a considerable history in electrochemical (also known as “pyroprocessing”) separations, and that technology is a prominent part of the long-range AFCI plan in which fast burner reactors are used to close the fuel cycle. The safeguards monitoring of the electrochemical process, however, is complicated by the fact that the initial fuel separation takes place in a molten salt bath (*i.e.*, an extremely harsh environment) and the dynamics of the separation process are complex. Techniques for monitoring the concentration of uranium and plutonium are needed to demonstrate that electrochemical separations processes can be accurately monitored, and reliable materials accountancy can be realized.

Solid-state-ionic sensors, also known as ionic conducting ceramics, have been used as sensors in molten media for over 40 years [Barmat 1960]. The examples for their commercial applications are oxygen sensors, which can provide *in-situ* determination of oxygen activity in liquid metals and alloys [Gileadi 1993], and Na sensors, which can provide *in-situ* measurement of Na activity in molten aluminum and aluminum silicon alloys [Dunn 1980; Dexpert-Ghys 1982]. The advantages of these types of devices are simplicity, reliability, and high *in-situ* sensitivity.

The ionic conducting ceramic sensor technology may be a useful tool for on-line monitoring of actinide concentrations in molten salt electrolyte for electrochemical processing. Some ceramics (solid electrolytes) show appropriate ionic conductivity at the operating temperatures of electrochemical processing. Such solid electrolytes are generally stable compounds which can withstand the harsh environment of molten chloride salts and hot cell operations. Therefore, solid electrolyte based sensors are well-suited to high-temperature molten-salt applications

Figure 21 and Equation [1] given below show a schematic of how the ionic sensor can be operated for on-line monitor Pu^{3+} concentration (activity) in the molten-salt (LiCl-KCl based) electrolyte.

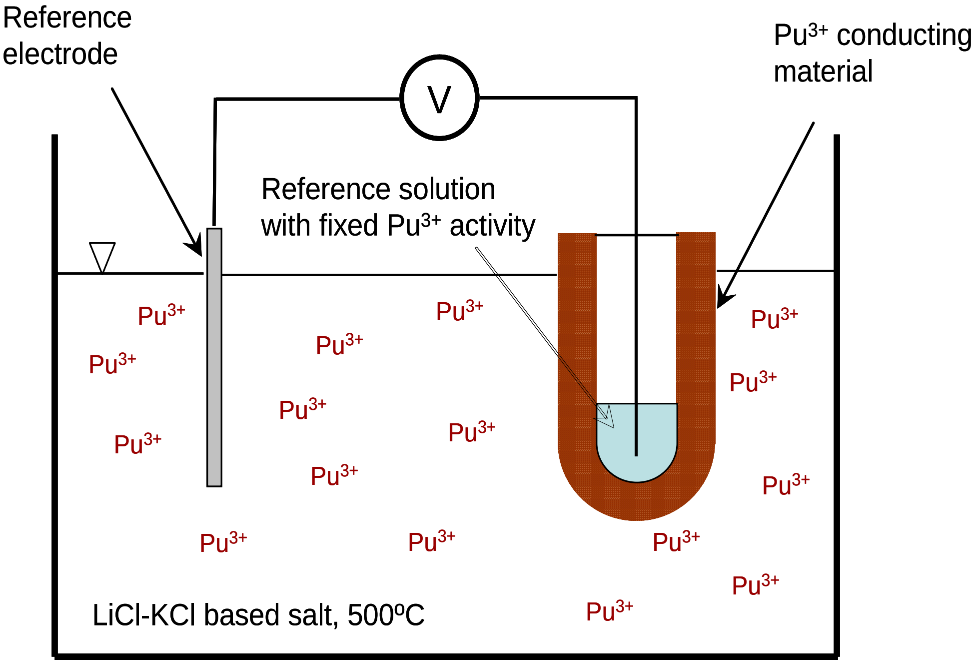

Figure 21. Schematic diagram of the ionic sensor for determining Pu^{3+} activity.

$$V = \Delta E = \frac{RT}{nF} \ln a_{Pu(reference)} - \frac{RT}{nF} \ln a_{Pu^{3+}} = A - B \ln a_{Pu^{3+}} \qquad [1]$$

Here, ΔE is a measured potential difference, R is the universal gas constant, n is number of electrons, T is the absolute temperature, and *F* is the Faraday constant. Equation [1] is called the Nernst Equation, which indicates the measured voltage difference [Zhang 1996]. ΔE, is a function of the activity difference of Pu^{3+} in the molten salt to be measured and Pu^{3+} in the reference solution. Since the activity of Pu^{3+} in the reference solution is known and constant in the electrorefiner operating conditions, the measured ΔE is a logarithmic function of the Pu^{3+} activity in the electrolyte. Any activity (concentration) change in the electrolyte will result in a simultaneous change in ΔE, which is directly measurable.

However, the most challenging task in applying the solid-state-ionic sensor technique to a fission-products-laden molten salt is to identify and synthesize a solid electrolyte (ionic conducting ceramic material) that functions as an actinide sensor. The actinide sensor materials must exhibit high melting temperature, high ionic conductivity, high sensitivity, and high selectivity. No appropriate sensor material is commercially available, so R&D is needed to achieve that goal. Once candidate sensor types are identified, the sensitivity of the instruments to uranium and plutonium concentration must be quantified, along with the expected mass uncertainty expected for production-scale electrochemical systems.

Potential Impact
Real-time, on-line measurement of U, Pu and other actinide concentrations in electrochemical processing

Needs That Could be Addressed if Successfully Developed
Direct Pu measurement in reprocessing solutions
On-line monitoring of actinide concentrations in reprocessing solutions

Research Questions/Challenges
1. Can suitable sensor materials, specific to each element of interest, be successfully developed?
2. Can these sensors be successfully fabricated?
3. What is the estimated accuracy in laboratory-scale test systems?
4. Can adequate sensitivity and selectivity be achieved at production scale, with molten salts laden with fission products?

Milestones	Estimated Timeline
Develop appropriate technique for synthesizing actinide sensor materials. (Q1)	1 year
Characterize the actinide sensor materials by XRD m SEM, or other techniques. (Q1)	2 years
Fabricate sensor devices with surrogate material and test them in molten salts for sensitivity and selectivity. (Q2 and Q3)	2 years
Fabricate sensor devices and test them in molten salts to determine the accuracy of measurements. (Q3)	3 years
Demonstrate the actinide-sensor applications at engineering scale, using fission-products-laden molten salt. (Q4)	4 years

4.2.8 Rapid Mass Spectrometry

Thermal-atomization resonance ionization spectroscopy (TARIS) is an advanced mass spectrometry technique that could speed up the measurement time of Pu or other elements in mixed reprocessing samples. TARIS is a combination of three established processes [Cipiti 2007]. First, a 10µL liquid processing sample is atomized on a hot tantalum plate in a vacuum chamber (thermal atomization). Next, three laser wavelengths are used to selectively excite and ionize a particular element group (resonance ionization). Finally the ionized element group is accelerated into a mass spectrometer for measurement.

The promise of TARIS is that the resonance ionization step selectively “pulls” out the element of interest, so no prior chemical separation is required. It may be possible to take measurements of mixed reprocessing samples with a sampling time on the order of minutes, a drastic improvement over current methods which can take about 8 hours. However, the uncertainty of the measurement has yet to prove that TARIS will be comparable to existing techniques.

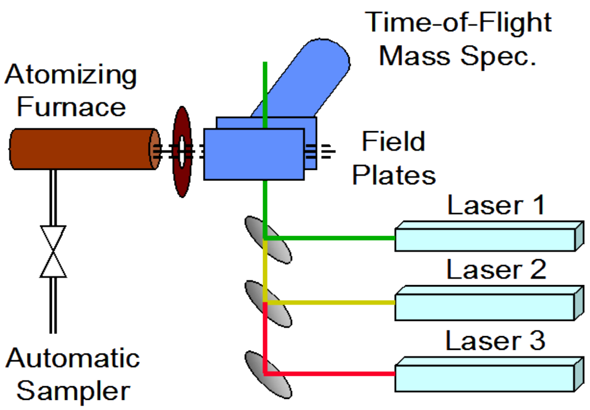

Figure 22. TARIS concept.

Accelerated measurement times will save a considerable number of man-hours in the analytical laboratory. This technique could also allow for many more sampling points to be taken which would be a requirement for achieving true near-real-time accountability of elemental quantities. A faster sampling rate also makes it possible to take multiple samples of a solution for improved statistical precision. All of these benefits point toward improved monitoring of continuous processing, which will be the paradigm of reprocessing plants in the future.

The key challenge of TARIS is to demonstrate a precise measurement of one element in a mixed actinide sample. R&D must determine the overall precision, selectivity, the effect of interferences, and overall measurement time. In addition, TARIS must be robust for this application.

Potential Impact
Can decrease the analytical measurement time from 8 hours to 20 minutes—may offer new safeguards paradigms by eliminating the need for plant flushout.

Needs That Could be Addressed if Successfully Developed
Direct measurement of Pu in reprocessing streams
Rapid laboratory sample analysis

Research Questions
1. Can we achieve the desired level of selectivity in mixed actinide samples?
2. What is the overall level of precision/uncertainty?
3. Is it worth it to build a multi-elemental unit with tunable lasers?
4. How much will these units cost?

Research Milestones	**Estimated Schedule**
Demonstrate measurement and determine uncertainty of Pu in mixed actinide samples.	1 year
Develop Pu-TARIS prototype.	1 year
Demonstrate on actual reprocessing solutions.	2 year

Determine robustness of operation, unit cost, waste recycle.	2 years
Design tunable laser system for multi-elemental capability.	3 years
Demonstrate tunable laser system on actual reprocessing solutions.	4 years

4.2.9 Data Authentication Methods

To permit valid conclusions to be drawn from safeguards data, it is essential that this data is known to be authentic. That is, it must be known that the data originated from the intended source, that the data was not changed in transit, and that it is not a repeat or delayed copy of previous data. To provide this assurance, a combination of equipment authentication procedures, protection of the equipment from tampering, and cryptographic authentication of the data is employed [Kryukov 2007]. All systems that will be used for international safeguards by the IAEA must pass a vulnerability assessment (VA) or a vulnerability review (VR) before the systems are fully authorized for safeguards use [Tolk 2006]. The putative adversary for these analyses is assumed to have full knowledge of all aspects of the system and is also assumed to have unlimited access to the facility. The adversary is further assumed to be highly motivated to defeat the system and to have national resources available for mounting the attack.

Safeguards systems and equipment that will be proposed for use by the IAEA or another international safeguards organization should be designed with authentication included, instead of attempting to add security later. Failure to integrate authentication measures early in the design results in systems that are expensive or perhaps impossible to deploy with adequate security to pass the VA or VR. It is important that the hardware and software design be transparent. For software, this includes source and binary code, development tools, etc. For hardware, schematics, IC specifications, etc.

The Joint DOE-DoD Authentication Task Force established a community consensus on the basic principles and guidelines for authentication of data, software, and hardware [2001], and work continues to develop supporting technology components for integration into future systems, such as secure unique identifiers and tamper-indicating electronic seals [Schoeneman 2005]. Hardware and software authentication is the process of gaining assurance that a system performs its purpose (and only its intended purpose) robustly and precisely as intended over the long lifespan of the instrument. Simple systems are easier to authenticate than complex systems; thus, where possible, extra functionality should be limited. A number of commercial and DOE-developed tools (such as ROSE) can aid domain experts in the authentication of software in both binary and source form [White 2006; White 2008].

A data-authentication priority for AFCI is the identification of areas where authentication is likely to be the most important, and the integration of data authentication into the design stages of that instrument or process. Data authentication techniques are being examined for incorporation into the Safeguards Performance Model.

Potential Impact
Incorporation of data authentication into the design early will provide long-term savings in preventing later plant retrofits, and could enable the sharing of data transmission infrastructure between private operators and regulatory inspectors.

Needs That Could be Addressed if Successfully Developed
Integration of data authentication methods

Research Questions
1. Joint-use equipment data authentication and sharing systems?
2. Universal data security module for remote and unattended equipment operation?
3. Intrinsic code verification for larger software and memory systems?
4. Full revalidation methods for monitoring systems that have been in the physical custody of others?

Research Milestones	Estimated Schedule
Define standard architecture and protocol for authentication and sharing of monitoring data. (Q1)	1 year
Complete prototype system for data authentication and confidentiality via adaptable sensor interface. (Q2)	1.5 years
Demonstrate ability to verify larger software modules and larger memory areas. (Q3)	2 years
Demonstrate suite of revalidation tools for hardware, software authentication after custody exchange. (Q4)	3 years

4.2.10 Modeling and Simulation for Advanced Control and Integration

Traditional materials accounting technology has been used for nuclear materials control and accountancy (MC&A) to detect the diversion of material produced in nuclear chemical process facilities using primarily sample collection and off-line analysis. These methods have not been shown to have the adequate resolution or timeliness required for in-process inventory measurements in large-scale process plants. One way to improve upon current methods in such facilities is to utilize process data in concert with modeling and simulation methods to develop near-real-time estimates for all fissionable material flows.

Nuclear separation process chemistry is well-understood and can be simulated using steady-state computer codes like AMUSE (Argonne Model for Universal Solvent Extraction) when appropriate input parameters are known. However, we cannot currently model or predict overall plant performance or the response of the system to typical operational changes such as feed variations or operator actions. Advanced computational methods will allow management and interpretation of the large dataset from the envisioned sensor array in real time. This on-line process data coupled with a detailed model of the whole plant can enable real-time materials accountancy and plant status. These sensor data can also be used to detect process anomalies and to determine if the anomaly has any safeguards significance.

Mitigation of any anomalous condition in the plant would require the following steps:

1. Detect and identify the condition and display details; this can be done by routine monitoring of simple physical and radiological sensors.
2. Determine if the condition is of safeguards significance; this would require a detailed plant model to predict the detailed effects of the anomaly.
3. Identify corrective actions; this action would be interactive between the operator and the observer, making use of the plant model.

4. Monitor the response of the system to the corrective action; the response would be predicted by the plant model and require only routine process monitoring to confirm.

In the first stage of development we will identify the current state-of-the-art in process monitoring technology and develop software to identify anomalies. In the second stage of development we will develop the software and interfaces to identify possible causes, mitigations and ramifications for the observed anomaly.

Potential Impact
Continuous, nondestructive and remote, monitoring of an entire process leading to real-time inventory

Needs That Could be Addressed if Successfully Developed
Process monitoring of reprocessing streams
Data collection, integration, semi-automated evaluation

Research Questions
1. What sensors are available and which will provide the most valuable information?
2. How can the data be evaluated in real time, to determine abnormalities in the process; how can uncertainties and drift be handled as to minimize false alarms?
3. How can a “normal status” be defined, and used for comparison?
4. Given a specific abnormal data point, how can we use AMUSE (or similar process codes) to determine a) What physical or chemical condition is reflected in the abnormality? b) How can this abnormality be confirmed? c) What can be done to recover? d) What is the effect of the abnormality on the overall plant status? e) What are the safeguards ramifications of this status? and f) whether any materials could have been diverted?
5. How can this approach be designed so that it is useful to both the operators and the observers?

Milestones	**Estimated Timeline**
Prepare a white paper on available, demonstrated and promising sensor technologies. (Q1)	1 year
Collect several data streams from a complex aqueous process, including known process upsets and anomalies. (Q2, Q3)	1 year
Using this data set, evaluate and develop software for recognizing the upsets and anomalies. (Q2, Q3)	2 year
Develop software that will take a recognized anomaly and use an established process code to calculate the complete status of the process. (Q4)	2 year
Develop software that will take that process status and calculate possible causes and remedies. (Q4)	3 year
Develop software that will take that process status and calculate the process status at all points during the upset, and determine if any materials could have been diverted. (Q4)	4 year
Design the user interface so that both the operator and the observer can use the same data to maximum value. (Q5)	5 year

4.3 Safeguards by Design

4.3.1 Safeguards Performance Modeling

Safeguards performance modeling is important for objectively evaluating the impact of new instruments or safeguards protocols in a facility for nuclear material accounting. When appropriate metrics are implemented, this performance modeling can also be used as a basis for a cost/benefit analysis, thereby supporting the prioritization of advanced safeguards R&D.

Among the examples of performance modeling is a tool intended to assist in the design of the safeguards system for a reprocessing plant [Cipiti 2007]. The UREX+1a process is emulated using discrete-event modeling techniques. An example of the front end of the plant is shown in Figure 23. The model tracks material flow throughout the plant, including cold chemicals, bulk flow rates, elemental quantities, and solids. The processing equipment volumes are included along with the expected separation efficiencies throughout the facility.

The model also includes measurement blocks that can simulate any type of accountability measurement throughout the plant. The user can specify information such as sampling rate, random and systematic error, drift, and calibration to represent different instrumentation. The measurement blocks are used to test the accountability system as well as to virtually test new instrumentation that may advance the state of the art. A diversion block can be used to determine the system response to a misuse scenario. Other performance modeling efforts have focused on how safeguards might be considered in the design and construction phases of fuel cycle facilities [DeMuth 2007].

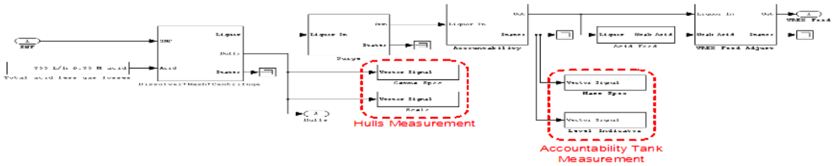

Figure 23. Schematic of discrete-event modeling as applied to Safeguards performance modeling of the front end in a reprocessing plant.

Prominent R&D questions for Safeguards performance modeling include

- What are the proper risk/cost metrics to consider?
- What data sets are available to test and validate the models?

- What advanced detection algorithms can be used to enhance performance from multiple sensors and monitoring systems?
- How robust are the systems to manipulation by an insider adversary?

The modeling effort will only be as useful as the accuracy of the assumptions built into the models. However, a high level of detail is not required to use the models to prioritize which new measurement technologies described here should make the most impact. Statistical techniques for accurate estimation of the value of information provided by measurements will be required.

Potential Impact
Decision-making tool for advanced instrumentation; virtual testing for meeting regulatory requirements; potential expansion into integrated safeguards, security, and safety model

Needs That Could be Addressed if Successfully Developed
High-fidelity integrated safeguards modeling

Research Questions
1. Can we rely on these types of models and assumptions to design new plants?
2. Can the model be expanded to include item accounting, security, and data authentication?
3. Can the model be benchmarked?

Research Milestones	**Estimated Schedule**
Combine measurement model with cost/benefit model.	0.5 year
Expand the model to include used fuel receipt and product conversion.	1 year
Include sigma MUF analysis and evaluate various statistical tests.	1 year
Incorporate Data Validation & Security to expand the model to one integrated system.	2 years
Benchmark the model.	3 years
Develop full accountancy plant design.	3 years

4.3.2 Validated Systems Analysis and Risk-based Tools for Integrated Systems

The need for integration within and among safeguards and security areas, as well as safety and operations, is an issue that is being addressed in all of the current efforts for developing future nuclear facilities (Gen IV, AFCI, IAEA). In the past, individual assessments of these areas have been performed through the use of detailed analysis techniques that have not been integrated in the design, evaluation, and operation of a facility [Garcia 2001; 2006]. This often has led to inefficient and costly design and operational requirements. The major benefit of integrating these areas is more cost-effective and efficient design and operation of nuclear facilities. Effective integration of these areas, however, will require the development and implementation of a framework, methods and validated systems analysis and risk-based tools for modeling and evaluating integrated facility design and operation [Darby 2007]. Commercially available software should be utilized for components of the analysis where appropriate.

Ongoing efforts are generally taking two approaches: one to extend and combine established, existing methods and tools to achieve a variety of “integration” within and among safeguards and security areas, as well as safety and operations, in some cases; another to develop simplified metrics that can be used to rate alternative design configurations [Martins 2007]. These scoring metrics and weighting functions are developed through expert elicitation [Butler 2005]. These ongoing efforts have many significant similarities and overlap, although they may focus on different integration issues. To support the design, licensing, and operation of future nuclear facilities, one key issue is to define the level of integration that is desired and to address the question of the extent of integration among safeguards, security, safety and operations that is achievable. Within this context, future efforts should look to coordinating and leveraging ongoing efforts, systematically addressing commonalities and differences, and developing strategies for an overall framework, then envisioning advanced analysis techniques for implementation. Long-term, the goal is a framework, methods and tools that can be applied for modeling and evaluating integrated systems from the beginning of facility design and throughout facility operation.

Potential Impact
Improvements in cost-effectiveness during the design, construction, and operation phases of the plant

Needs That Could be Addressed if Successfully Developed
High-fidelity integrated safeguards modeling

Research Questions
1. What are principles, objectives, and performance metrics for the design and evaluation of safeguards systems? For example, for a physical security system, the design principles are detect, delay and respond.
2. What performance characteristics and data are required for design and evaluation of a safeguards system?
3. What are the key relationships among safeguards, security, safety systems, and operations for a recycle facility, and how can they be modeled in an integrated facility design?

Research Milestones	**Estimated Schedule**
Develop principles and performance metrics for safeguards systems.	1 year
Implement and demonstrate first level integration of safeguards and security systems elements in the safeguards performance model.	1 year
Identify key relationships among systems for an integrated design.	1 year
Develop and demonstrate process and tools for design and evaluation of safeguards systems; identify modeling and analysis tools to support process.	2 years
Develop data base of safeguards performance data to support systems analysis .	3 years
Develop and demonstrate framework for design and evaluation of conceptual recycle facility that integrates safeguards and security systems; extend framework to address safety and operational elements; identify modeling and analysis tools to support framework.	5 years

5.0 Key Programmatic Connections

In order to be successful and relevant, the AFCI Safeguards Campaign must be closely coupled to other AFCI campaigns and the safeguards community as a whole. For example, the Safeguards Campaign should be informed and engaged in the design of reprocessing plants so that the ability to safeguard a facility is an integral part of that design process, rather than an expensive afterthought. Some of the key linkages identified by the SES team are described below.

5.1 AFCI Campaigns

Facility Design: All facilities designed under the AFCI program are expected to incorporate the "Safeguards by Design" concept—the idea that the best possible safeguards must be an important part of the early baseline design exercises. In addition, any designs produced by subcontractors or industrial partners must be held to the same metric. Fuel cycle facilities (*e.g.*, U purification plant, U enrichment plant, fuel fabrication plant, reprocessing plant, waste operations, repository, and transportation infrastructure) must contain safeguards hardware and procedures as an integral part of their design. The safeguards campaign should provide facility engineers with guidelines, requirements, and footprints to be used in early designs.

Systems Analysis: Safeguards must be one of the many factors included in the system analysis of any fuel cycle or part of a fuel cycle. Any change in hardware, procedures, or capacity could have a profound effect on the ability to safeguard a facility, or require changes in specific safeguards issues. The Safeguards and System Analysis Campaigns must work together to incorporate accurate and useful safeguards data and requirements into the global systems model.

Reactor Design: The Reactor Campaign currently includes a task to collect and evaluate key nuclear data (e.g. fission and absorption cross-sections of fissile and actinide isotopes). While much of this work can be directly utilized by the Safeguards Campaign, there may also be Safeguards-specific data needs that could be communicated to the Reactor nuclear data effort.

Separations: Close coupling between the Safeguards and Separations Campaigns is important so that candidate separations schemes that will be amenable to safeguards and safeguards implementation planning can be integrated into the design of separations processes. For example, if the Separations campaign should decide that only electrochemical processes will be pursued in AFCI, the Safeguards Campaign may need to re-focus a considerable part of a portfolio that is currently heavy on investment and experience in safeguarding aqueous processes.

Cross-Cut Campaigns: The Safeguards Campaign must communicate its computational needs to the Modeling and Simulations Cross-cut team, and be able to harvest tools developed in the Modeling and Simulations program. The complexity of the envisioned facilities leads to multiple possible safeguards issues; modeling and simulation can provide a tool for fully exploring these issues. Good communication with the regulatory cross-cutting activities in AFCI is also important, as decisions made in that activity could drive the performance requirements for safeguards systems.

5.2 Other Programs

DOE NA-24: The Next-Generation Safeguards Initiative (NGSI), a program to be executed by NA-24, is likely to be the largest safeguards technology development program in the United States in the near future. It will be imperative that the AFCI program continue its close integration with NA-24 and the emerging NGSI, so that R&D investments are made in a complementary fashion.

IAEA: Due to the multinational and cooperative nature of the program, transparency in all facilities and in all countries is key to the acceptance of the AFCI program. Because AFCI has unique goals, and the facility designs are quite different than previous designs, new approaches to safeguards must be developed and utilized in collaboration with the Agency. As an example of a relevant International Atomic Energy Agency (IAEA) activity, a multi-laboratory team is investigating the role and potential value of process monitoring data for safeguards monitoring and detection activities. Specific areas of interest include potential diversion path scenarios, and data analysis techniques to enhance detection capabilities. This effort will be symbiotic with AFCI work in facility modeling.

DOE NA-22: This office has a long history of developing technologies that are relevant to safeguards, but that focus has recently been sharpened with the creation of a new NA-22 program area called Safeguards R&D and Alternate Radiological Source Development. Although funding in the program is relatively limited in the initial year, it will be important that the AFCI Safeguards Campaign is informed and engaged in the new NA-22 program to ensure complementary goals and project undertakings. Examples of existing NA-22 projects with immediate relevance to AFCI Safeguards are the construction of a small-scale centrifugal contactor to foster development and testing of process monitoring technology, the liquid-scintillator advanced multiplicity counter, a used-fuel pin counter and microcalorimeter detectors.

University Collaborations: There are a number of programs, either already created or being considered, that are providing support for safeguards-relevant R&D at universities in the United States. The sustainability of a pipeline of creative ideas and technical staff in safeguards will rely on these programs. The AFCI program will strive to strengthen the ties between the more basic R&D being performed at universities and the specific needs and applications being pursued by the AFCI Safeguards Campaign.

6.0 Reference

1. Abdurrahman, NM, et al. 1993. "Spent-Fuel Assay Performance and Monte Carlo Analysis of the Rensselaer Slowing-Down-Time Spectrometer." Nuclear Science and Engineering 115:279-96.

2. Abhold, ME, ML Collins, and GW Eccleston. 1998, Verification Methods for Spent Fuel in Sealed Multi-Element Casks. Report No. LA-UR-98-129, Los Alamos National Laboratory, Los Alamos, NM.

3. Agostinelli, S, et al. 2003. "Geant4 - a Simulation Toolkit." Nuclear Instruments & Methods in Physics Research, Section A (Accelerators, Spectrometers, Detectors and Associated Equipment) 506:250-303.

4. Alcouffe, RE, et al. 2005. PARTISN: A Time-Dependent, Parallel Neutral Particle Transport Code System. Los Alamos National Laboratory, LA-UR-05-3925

5. ASTM. 2003. Standard Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting. ASTM Standard C 1207,

6. Auer, M, et al. 2004. "Intercomparison Experiments of Systems for the Measurement of Xenon Radionuclides in the Atmosphere." Applied Radiation and Isotopes 60:863-77.

7. Barmat, M. 1960. Thermoelectric Materials and Devices. Reinhold, New York.

8. Benedict, M, TM Pigford, and HW Levi. 1981. Nuclear Chemical Engineering. 2nd ed., McGraw-Hill, New York.

9. Bertozzi, W, and RJ Ledoux. 2005. "Nuclear Resonance Fluorescence Imaging in Non-Intrusive Cargo Inspection." Nuclear Instruments & Methods in Physics Research, Section B (Beam Interactions with Materials and Atoms) 241:820-5.

10. Beugelsdijk, TJ, and RM Hollen. 1998. "Robotics and Automation in Radiochemical Analysis." in Handbook of Radioactivity Analysis, ed. MF L'Annunziata, pp. 693-718. Academic Press, San Diego.

11. Bracken, DS, R Biddle, and R Cech. 1997. "Design and Performance of a Vacuum-Bottle Solid-State Calorimeter." in Proceedings of the 38th Annual Meeting of the Institute of Nuclear Materials Management, Phoenix.

12. Bracken, DS, R Biddle, and CR Rudy. 1998. "Performance Evaluation of a Commercially Available Heat Flow Calorimeter and Applicability Assessment for Safeguarding Special Nuclear Materials." in Proceedings of the 39th Annual Meeting of the Institute of Nuclear Materials Management, Naples.

13. Bracken, DS, and P Hypes. 2000. "Solid-State Calorimeter." in Proceedings of the 41st Annual Meeting of the Institute of Nuclear Materials Management, New Orleans.

14. Bracken, DS, and CR Rudy. 2007, Principles and Applications of Calorimetric Assay. Report No. LA-UR-07-5226, Los Alamos National Laboratory, NM, Albuquerque.

15. Brandalise, B, et al. 1994. "Robotized Equipment for the on-Site Analysis of Fissile Material." in International Nuclear Safeguards 1994 Vision for the Future, Vol 1, pp. 755-60. International Atomic Energy Agency, Vienna.

16. Bryan, S, D Levitskaia, and SI Sinkov. 2005, Process Monitor Development Project: Acceptance Test Report. Report No. PNNL-Report-47187, Pacific Northwest National Laboratory, Richland, WA.

17. Bryan, SA, and TG Levitskaia. 2007. "Monitoring and Control of UREX Radiochemical Processes." pp. 176-79. American Nuclear Society, La Grange Park, IL 60526, United States, Boise, ID, United States.

18. Buck, R, E Lent, and S Hadjimarkos. 2002. COG: A Multiparticle Monte Carlo Transport Code: Users Manual. UCRL-TM-202590.

19. Burks, M, et al. 2006. "Imaging Performance of the Si/Ge Hybrid Compton Imager." pp. 6-10. IEEE, Fajardo, Puerto Rico.

20. Burr, T, J Jones, and L Wangen. 1994. "Multivariate Diagnostics and Anomaly Detection for Nuclear Safeguards." in Proc. 35th Annual Meeting Inst. Nuclear Materials Management, pp. 9 p.

21. Burr, TL, et al. 2003. "Solution Monitoring: Quantitative and Qualitative Benefits to Nuclear Safeguards." Journal of Nuclear Science and Technology 40:256-63.

22. Butler, J, et al. 2005. "The Adoption of Multi-Attribute Utility Theory for the Evaluation of Plutonium Disposition Options in the United States and Russia." Interfaces 35.

23. Caldwell, JT, et al. 1983. "Experimental Evaluation of the Differential Die-Away Pulsed-Neutron Technique for the Fissile Assay of Hot Irradiated Fuel Waste." pp. 296-9. Battelle Press, Richland, WA, USA.

24. Camp, DC, WD Ruhter, and KW MacMurdo. 1981. "Determination of Actinide Process- and Product Stream Concentrations Off-Line or at-Line by Energy Dispersive X-Ray Fluorescence Analysis." pp. 155-60. ESARDA, Karlsruhe, West Germany.

25. Charrin, N, P Moisy, and P Blanc. 2000. "Contribution of the Concept of Simple Solutions to Calculation of Density of Ternary and Quaternary Solutions." Radiochimica Acta 88:25-31.

26. Chen, Y, et al. 2002. "Multi-Dimensional Regression Analysis of Time-Series Data Streams." pp. 323-34. Morgan Kaufmann Publishers, Hong Kong, China.

27. Choi, H, H Ombao, and B Ray. 2008. "Sequential Change-Point Detection Methods for Nonstationary Time Series." Technometrics 50:40-52.

28. Cipiti, BB, PE Rexroth, and NL Ricker. 2007, Safeguards Performance Modeling of a UREX+1a Reprocessing Plant. Report No. SAND2007-6586, Sandia National Laboratories, Albuquerque, NM.

29. Clark, J, W. J., et al. 2006. "Electrochemically Modulated Separation, Concentration, and Detection of Plutonium Using an Anodized Glassy Carbon Electrode and Inductively Coupled Plasma Mass Spectrometry." Analytical Chemistry 78:8535-42.

30. Clark, SB, and C Delegard. 2002. "Plutonium in Concentrated Solutions." in Advances in Plutonium Chemistry 1967-2000, ed. DC Hoffman. The American Nuclear Society, La Grange Park, Illinois.

31. Coron, N, et al. 1985. "A Composite Bolometer as a Charged-Particle Spectrometer." Nature 314:75-6.

32. Croff, AG. 1983. "Origen2: A Versatile Computer Code for Calculating the Nuclide Compositions and Characteristics of Nuclear Materials." Nucl. Technol. ; Vol/Issue: 62:3:Pages: 335-52.

33. Crosier, RB. 1988. "Multivariate Generalizations of Cumulative Sum Quality-Control Schemes." Technometrics 30:291-303.

34. Darby, JL, et al. 2007, Framework for Integrating Safety Operations, Security, and Safeguards in the Design and Operation of Nuclear Facilities. Report No. SAND 2007-6429, Sandia National Laboratories, Albuquerque, NM.

35. Dass, SC, and AK Jain. 2007. "Fingerprint-Based Recognition." Technometrics 49:262-76.

36. DeMuth, S. 2007. "Advanced Safeguards for the Global Nuclear Energy Partnership (Gnep)." in European Nuclear Conference, Brussels.

37. Dexpert-Ghys, J, M Faucher, and P Caro. 1982. "Lanthanum Β-Alumina Phase Doped with Europium: Optical Investigation by Dye Laser Site-Selective Excitation." Journal of Solid State Chemistry 41:27-38.

38. Doriese, WB, et al. 2007. "14-Pixel, Multiplexed Array of Gamma-Ray Microcalorimeters with 47 Ev Energy Resolution at 103 Kev." Applied Physics Letters 90:193508-1.

39. Dougan, A, et al. 2007. "New and Nondestructive Neutron and Gamma-Ray Technologies Applied to Safeguards." in JAEA-IAEA New and Novel Technologies Meeting. UCRL-PROC-237318.

40. Dowla, F, et al. 2005. "Method for Remote Powering and Detecting Multiple Uwb Passive Tags in an Rfid System." USA IL-11592.

41. Dreyer, JG, et al. 2008. "Development of Digital Signal Readout System for Large Tes Arrays." Journal of Low Temperature Physics:958-63.

42. Drury, OB, SF Terracol, and S Friedrich. 2005. "Quantifying the Benefits of Ultrahigh Energy Resolution for Gamma-Ray Spectrometry." pp. 1468-79. Wiley-VCH, Warsaw, Poland.

43. Dunn, B, and GC Farrington. 1980. "Fast Divalent Ion Conduction in Ba++, Cd++ and Sr++ Beta" Aluminas." Materials Research Bulletin 15:1773-7.

44. Durst, C, et al. 2007a, Advanced Safeguards Approaches for New Reprocessing Facilities. Report No. PNNL-16674, Pacific Northest National Laboratory, Richland, WA.

45. Durst, C, et al. 2007b, Advanced Safeguards Approaches for New Fast Reactors. Report No. PNNL-17168, Pacific Northwest National Laboratory, Richland, WA.

46. Durst, PC, et al. 2007c, Advanced Safeguards Approaches for New TRU Fuel Fabrication Facilities. Report No. PNNL-17151, Pacific Northwest National Laboratory, Richland, WA.

47. Ensslin, N, et al. 1998. "Application Guide to Neutron Multiplicity Counting."

48. Friedrich, S. 2007. "Nuclear Diagnostics with Cryogenic Spectrometers." Nuclear Inst. and Methods in Physics Research, A 579:157-60.

49. Friedrich, S, et al. 2006. "Neutron Absorption Spectroscopy for Identification of Light Elements in Actinides." Nuclear Instruments & Methods in Physics Research, Section A (Accelerators, Spectrometers, Detectors and Associated Equipment) 559:745-7.

50. Gaggeler, HW. 1997. "Fast Chemical Separation Procedures for Transactinides." in Robert A. Welch Foundation Conferences on Chemical Research XXXI The Transactinide Elements, pp. 43-63, Bern, Switzerland.

51. Garcia, ML. 2001. The Design and Evaluation of Physical Protection Systems. Butterworth-Heinemann, Boston.

52. Gehrke, RJ, et al. 1994, Application of PINS and GNAT to the Assay of 55 Gal Containers of Nuclear Waste. Report No. EGG-NRE-11236, Idaho National Energy Laboratory, Idaho Falls, ID.

53. Gildseth, W, A Habenschuss, and FH Spedding. 1972. "Precision Measurements of Densities and Thermal Dilation of Water between 5° and 80°C." Journal of Chemical Engineering Data 17:402-09.

54. Gileadi, E. 1993. Electrode Kinetics for Chemists, Chemical Engineers, and Materials Scientists. New York, NY.

55. Giuliani, A. 2000. "Particle and Radiation Detection with Low-Temperature Devices." Vol 280, pp. 501-8. Elsevier, Helsinki, Finland.

56. Grate, JW, and OB Egorov. 2003. "Automated Radiochemical Separation, Analysis, and Sensing." in Handbook of Radioactivity Analysis, ed. MF L'Annunziata, pp. 1129-64. Academic Press, San Diego.

57. Grate, JW, and OB Egorov. 1998. "Automating Analytical Separations in Radiochemistry." Analytical Chemistry 70:779A-88A.

58. Grate, JW, OB Egorov, and MJ O'Hara. 2008a. "Automation of Extraction Chromatographic and Ion Exchange Separation Methods for Radiochemical Analysis and Monitoring." in Ion Exchange Solvent Extraction, ed. B Moyer, Vol 19. Taylor and Frances Group, Accepted for publication.

59. Grate, JW, et al. 2008b. "Radionuclide Sensors for Environmental Monitoring: From Flow Injection Solid-Phase Absorptiometry to Equilibrium-Based Preconcentrating Minicolumn Sensors with Radiometric Detection." Chemical Reviews 108:543-62.

60. Gunnink, R. 1990, MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances, Volume I, Methods and Algorithms. Report No. LA-11720-MS, Lawrence Livermore National Laboratory, Oakland, CA.

61. Hensley, WK, et al. 1995. "Synth - a Spectrum Synthesizer." pp. 229-37.

62. Higdon, D, et al. 2008. "Computer Model Calibration Using High-Dimensional Output." Journal of the American Statistical Association 103:570-83.

63. Hiller, L, et al. 2007. Radsrc/Monte Carlo Code Interface Manual. UCRL-TR-229498.

64. Hofstetter, KJ, PR Cable, and DM Beals. 1999. "Field Analyses of Tritium at Environmental Levels." Vol 422, pp. 761-6. Elsevier, Ann Arbor, MI, USA.

65. Hollas, CL, et al. 1997, Matrix Effects Corrections in Ddt Assay of 239pu with the Cten Instrument. Report No. LA-UR-97-3408, Los Alamos National Laboratory, Salt Lake City, UT.

66. Hollas, CL, DA Close, and CE Moss. 1987. "Analysis of Fissionable Material Using Delayed Gamma Rays from Photofission." Vol B24-B25, pp. 503-5, Denton, TX, USA.

67. Hoover, AS, et al. 2007. "Monte Carlo Studies of High Resolution Microcalorimeter Detectors." pp. 5 pp. IEEE, San Diego, CA, USA.

68. Horansky, RD, et al. 2007. "Superconducting Absorbers for Use in Ultra-High Resolution Gamma-Ray Spectrometers Based on Low Temperature Microcalorimeter Arrays." Nuclear Inst. and Methods in Physics Research, A 579:169-72.

69. Horansky, RD, et al. 2008. "Superconducting Calorimetric Apha Particle Sensors for Nuclear Nonproliferation Applications." Applied Physics Letters 93:123504.

70. Howell, J, and S Scothern. 1997, A Prototype Diagnostic Aid for a Tank Monitoring System. Report No. SRDP-R244, UK Safeguards R&D, Glasgow.

71. Hunt, AW, and ET Reedy, Idaho State University. 2008. "Conversations." ed. DL Chichester, Idaho Falls.

72. Hyman, P, V Gupta, and D Sullivan. 1999. "Nuclear Materials Safeguards Matrix-Specific Qualification & Continuous Bias Correction Programs." New Brunswick Laboratory, Argonne, IL.

73. INL. 2003. "Better Than Dogs." in *Need to Know*, INL National Security Newsletter, Vol 3. Idaho National Laboratory, Idaho Falls.

74. JDDATF. 2001, Guidelines for Authenticating Monitoring Systems. Report, Joint DOE-DoD Authentication Task Force.

75. Jones, JL. 1996. "Detection of Pulsed, Bremsstrahlung-Induced, Prompt Neutron Capture Gamma-Rays with a Hpge Detector." in SPIE 1996, Vol 2867, pp. 202. SPIE, Crete.

76. Jones, JL, et al. 2005, Pulsed Photonuclear Assessment (Ppa) Technique: C.Y.04 Year-End Progress Report. Report No. INL 05-02583, Idaho National Laboratory, Idaho Falls, ID.

77. Jordan, KA, and T Gozani. 2007a. "Pulsed Neutron Differential Die Away Analysis for Detection of Nuclear Materials." Nuclear Inst. and Methods in Physics Research, B 261:365-8.

78. Jordan, KA, J Vujic, and T Gozani. 2007b. "Remote Thermal Neutron Die-Away Measurements to Improve Differential Die-Away Analysis." Nuclear Inst. and Methods in Physics Research, A 579:407-9.

79. Kniesel, G, and U Bardey. 2006. "An Analysis of the Correctness and Completeness of Aspect Weaving." pp. 10 pp. IEEE Computer Society, Los Alamitos, CA, USA.

80. Krinninger, H, E Ruppert, and L Hintze. 1974. "Neutron Self-Shielding Factors for Tantalum in the Energy Range of 45 Ev to 50 Kev." Nuclear Instruments and Methods 117:45-60.

81. Kryukov, V, et al. 2007. "Trusted Processor: A Result of the Evolution of Information Barrier Technologies." in Institute of Nuclear Materials Management Annual Meeting. INMM, Tuscon.

82. Kumar, S, and SB Koganti. 1998. "Prediction of Densities of Mixed Organic Solutions Containing Uo2(No3)2 and Nitric Acid." Journal of Nuclear Science and Technology 35:309-12.

83. LaFleur, AM, et al. 2008. "Nondestructive Measurements of Fissile Material Using Self-Indication Neutron Resonance Absorption Densitometry (Sinrad)." in 8th International Conference on Facility Operations – Safeguards Interface, Portland.

84. Lariviere, D, et al. 2008a. "Automated Flow Injection System Using Extraction Chromatography for the Determination of Plutonium in Urine by Inductively Coupled Plasma Mass Spectrometry." Journal of Analytical Atomic Spectrometry 23:352-60.

85. Lariviere, D, et al. 2008b. "Rapid and Automated Analytical Technologies for Radiological/Nuclear Emergency Preparedness." in Nuclear Chemistry: New Research, in Press, ed. AN Koskinen. Nova Publishers, Hauppauge NY.

86. Lee, DM, and LO Lindquist. 1982, Self-Interrogation of Spent Fuel. Report No. LA-9494-MS, Los Alamos National Laboratory Los Alamos, NM.

87. Lestone, JT, TH Prettyman, and JD Chavez. 2000. "Performance of the Skid-Mounted Tomographic Gamma Scanner for Assays of Plutonium Residues at Rfets." in Procedings of Institute of Nuclear Material Management 41st Annual Meeting, New Orleans.

88. Malinowski, E. 2008. Factor Analysis in Chemistry. John Wiley & Sons, New York.

89. Martins, JL, et al. 2007. "Scores - a Tool for Performance-Based Safeguards Inspections." in European Safeguards Research and Development Association 29th Conference.

90. Mattingly, J, and ES Varley. 2008. "Synthesis of the Feynman-Y Neutron Multiplicity Metric Using Deterministic Transport,." in American Nuclear Society Annual Meeting, Anaheim.

91. MCNP—a General Monte Carlo N-Particle Transport Code. 2003. Los Alamos National Laboratory, LA-UR-03-1987

92. Menlove, HO. 2006, Advanced Safeguards Integration for the UREX+ Head-End Fuel Cycle - the Use of a Neutron Balance for Safeguards in the UREX+ Fuel. Report No. LA-UR-06-6113, Los Alamos National Laboratory, Albuquerque, NM.

93. Menlove, HO, and DH Beddingfield. 1997, Passive Neutron Reactivity Measurement Technique. Report No. LA-UR-97-2651, Los Alamos National Laboratory, Los Alamos, NM.

94. Menlove, HO, et al. 1969. "A Resonance Self-Indication Technique for Isotopic Assay of Fissile Materials." Nuclear Applications 6:401 et seq.

95. Metzger, FR. 1959. "Resonance Fluorescence in Nuclei." Progress in Nuclear Physics 7:54-88.

96. Mihailescu, L, et al. 2006. "Combined Measurements with Three-Dimensional Design Information Verification System and Gamma Ray Imaging - a Collaborative Effort between Oak Ridge

National Laboratory, Lawrence Livermore National Laboratory, and the Joint Research Center at Ispra." pp. Size: PDF-file: 9 pages; size: 1.1 Mbytes, United States.

97. Mitchell, DJ, and J Mattingly. 2008. "Rapid Computation of Gamma-Ray Spectra for One-Dimensional Source Models." in American Nuclear Society Annual Meeting, Anaheim.

98. Nekoogar, F, F Dowla, and Springer. 2008, Ultra-Wideband Radio Frequency Identification Systems. Report, Lawrence Livermore National Laboratory, Berkeley, CA.

99. Niedermayr, T, et al. 2007. "Unfolding of Cryogenic Neutron Spectra." Nuclear Inst. and Methods in Physics Research, A 579:165-8.

100. Niedermayr, T, et al. 2004. "Microcalorimeter Design for Fast-Neutron Spectroscopy." Vol 520, pp. 70-2. Elsevier, Genoa, Italy.

101. Norman, EB, et al. 2004. "Signatures of Fissile Materials: High-Energy Γ Rays Following Fission." Nuclear Instruments & Methods in Physics Research, Section A (Accelerators, Spectrometers, Detectors and Associated Equipment) 521:608-10.

102. Oganessian, YT, et al. 2006. "Synthesis of the Isotopes of Elements 118 and 116 in the 249cf and 245cm+48ca Fusion Reactions." Phys. Rev. C 74.

103. Orton, C, et al. 2008. "Advanced Safeguards Technology Demonstration at Pacific Northwest National Laboratory." in Proceedings of the 49th Annual INMM Conference, Nashville.

104. Pretty, JR, DC Duckworth, and GJ VanBerkel. 1998. "Electrochemical Sample Pretreatment Coupled on-Line with Icp-Ms: Analysis of Uranium Using and Anodically Conditioned Glassy Carbon Working Electrode." Analytical Chemistry 70:1141.

105. PRPP. 2006, Evaluation Methodology for Proliferation Resistance and Physical Protection of Generation IV Nuclear Energy Systems, Revision 5. Report No. GIV/PRPPWG/2006/005, Proliferation Resistance and Physical Protection.

106. Pruet, J, et al. 2006. "Detecting Clandestine Material with Nuclear Resonance Fluorescence." Journal of Applied Physics 99:123102-1.

107. Rakel, DA. 1987. "In-Field Calibration of Neutron Correlation Counters Via Calorimetry and High Count Rate Gamma Ray Isotopic Abundance Measurements." pp. 239-49. IAEA, Vienna, Austria.

108. Regalbuto, MC, et al. 2004. "Solvent Extraction Process Development for Partitioning and Transmutation of Spent Fuel." in *Actinide and Fission Product Partitioning and Transmutation*, Eighth Information Exchange Meeting, pp. 373-85. Nuclear Energy Agency Organization for Economic Co-operation and Development, Las Vegas.

109. Ressler, JJ, LE Smith, and KK Anderson. 2008. "Lead Slowing Down Spectroscopy for Direct Pu Mass Measurements." in American Nuclear Society, Institute of Nuclear Materials Management Facility Protection Conference, Portland, OR.

110. Rhoades, WA, and DB Simpson. 1997. The TORT Three-Dimensional Discrete Ordinates Neutron/Photon Transport Code. Oak Ridge National Laboratory, ORNL/TM-13221

111. Rinard, PM. 2001, Application Guide to Shufflers. Report No. LA-13819-MS, Los Alamos National Laboratory, Los Alamos, NM.

112. Sakurai, S, and S Tachimori. 1996. "Density Equation of Aqueous Solution Containing Plutonium (IV), Uranium (Vi) and Nitric Acid." Journal of Nuclear Science and Technology 33:187-89.

113. Sampson, TE, GW Nelson, and TA Kelley. 1989, Fram: A Versatile Code for Analyzing the Isotopic Composition of Plutonium from Gamma-Ray Pulse Height Spectra. Report No. LA-11720-MS, Los Alamos National Laboratory, Albuquerque, NM.

114. Santi, P, M Fensin, and D Vo. 2007a, Status of Nuclear Data for Advanced Safeguards. Report No. LA-UR-07-6496, Los Alamos National Laboratory, Albuquerque, NM.

115. Santi, P, et al. 2007b. "The Role of Nuclear Data in Advanced Safeguards." in Advanced nuclear fuel cycles and systems (GLOBAL 2007), Boise.

116. Schaedel, M. 2007. "Superheavy Element Chemistry-Achievements and Perspectives." Journal of Nuclear and Radiochemical Sciences 8:47-53.

117. Schoeneman, JL, SAND2005-0598A, INMM 2005 Annual Conference. 2005. "Secure Sensor Platform (Ssp) for Materials' Sealing and Monitoring Applications." in Institute of Nuclear Materials Management Annual Meeting. INMM, Phoenix.

118. Schwantes, JM, et al. 2008. "Multi-Isotope Process (MIP) Monitor: A near-Real-Time Monitor for Reprocessing Facilities." in Transactions of the American Nuclear Society Annual Meeting, Anaheim.

119. Slaughter, DR, et al. 2007. "The Nuclear Car Wash: A System to Detect Nuclear Weapons in Commercial Cargo Shipments." Nuclear Inst. and Methods in Physics Research, A 579:349-52.

120. Smith, L, et al. 2008, in press. "Deterministic Transport Methods for the Simulation of Gamma-Ray Spectroscopy Scenarios." in IEEE Transactions on Nuclear Science.

121. Smith, L, et al. 2007. "Next-Generation Online Mc&a Technologies for Reprocessing Plants." in Global 2007 Advanced Nuclear Fuel Cycles and Systems Conference, Boise, ID.

122. Smith, LE, and NM Abdurrahman. 2002. "Neutron Spectrometry for the Assay of High Fissile Content Spent Fuel." Nuclear Technology 140:328-49.

123. Terracol, SF, et al. 2004. "Ultra-High Resolution Gamma-Ray Spectrometer Development for Nuclear Attribution and Non-Proliferation Applications." Vol Vol. 2, pp. 1006-13. IEEE, Rome, Italy.

124. Tobin, SJ, et al. 2006. "Non-Proliferation Technology Development Study for UREX." in IAEA Symposium on International Safeguards: Addressing Verification Challenges, Vienna, Austria.

125. Tolk, K, et al. 2006. "Design of Safeguards Systems for Authentication." in Symposium on International Safeguards - Addressing Verification Challenges, Vienna.

126. Twomey, TR, AJ Caffrey, and DL Chichester. 2007. "Nondestructive Identification of Chemical Warfare Agents and Explosives by Neutron Generator-Driven Pgnaa." United States.

127. Vetter, K. 2002. "Gamma-Ray Tracking: New Opportunities for Nuclear Physics." Nuclear Physics News 12.

128. Wang, TF, WD Ruhter, and GP Russ. 2006. "Safeguards and Environmental Measurements Using Compton Suppressed Ge Detectors." in IAEA Symposium on International Safeguards, Vienna.

129. Warren, GA, et al. 2007. "Nuclear Resonance Fluorescence of 235u above 3 Mev." pp. 2047-9. IEEE, Honolulu, HI, USA.

130. White, G. 2006. "Strengthening Software Authentication with the Rose Software Suite." in Institute of Nuclear Materials Management 47th Annual Meeting, Nashville.

131. White, G. 2008. "Tools for Authentication." in Institute of Nuclear Materials Management, Nashville.

132. Wilk, PA, et al. 2008. "Separation of Group Five Elements by Reversed-Phase Chromatography." J. Radioanal. Nucl. Chem. 275.

133. Wilson, WB, et al. 2002. SOURCES-4c: A Code for Calculating (Alpha, N), Spontaneous Fission, and Delayed Neutron Sources and Spectra. Los Alamos National Laboratory, LA-UR-02-1839

134. Yamamoto, M. 1988. "Determination of Nitric Acid Concentration in 30 Vol Tbp-N-Dodecane by Measuring Dielectric Properties." Journal of Nuclear Science and Technology 25:540-47.

135. Zahradnik, P, and H Swietly. 1996. "The Robotized Chemical Treatment of Diluted Spent Fuel Samples Prior to Isotope Dilution Analysis." Journal of Radioanalytical and Nuclear Chemistry 204:145.

136. Zhang, L, et al. 1996. "Electrochemical Sensor for Measuring Magnesium Content in Molten Aluminium." Journal of Applied Electrochemistry 26:269-75.

137. Ziegler, T, and K Mayer. 1999. "Development of an Optimized Method for Faster and More Reliable Automated U/Pu/Np Separations." Radiochimica Acta 86:123-28.

138. Ziock, KP, et al. 2004. "Large Area Imaging Detector for Long-Range, Passive Detection of Fissile Material." IEEE Transactions on Nuclear Science 51:2238-44.

139. Zvara, I. 1999. "Studies of the Chemistry of Transactinides by Fast Separation Techniques." in NATO Science Series, Vol 53, pp. 123-34. NATO.

Appendix A: Authors and Reviewers of Technology Summaries

Technology	Lead Lab	Primary Author(s)	Reviewer(s)
Advanced Instrumentation			
Lead Slowing Down Spectroscopy	PNNL	Eric Smith (PNNL)	Victor Gavron (LANL),Yaron Daron (RPI)
Nuclear Resonance Flourescence	PNNL	Glen Warren (PNNL)	Steve Korbly (Passport Systems), Micah Johnson (LLNL)
Advanced Neutron Multiplicity	LANL	Steve Tobin (LANL)	Les Nakae (LLNL)
Passive Neutron Albedo Reactivity	LANL	Steve Tobin (LANL)	Samer Kahook (SRNL)
Self-Interrogation Neutron Resonance Densitometry	LANL	Howard Menlove (LANL)	Eric Smith (PNNL)
Differential Die-Away	LANL	Steve Tobin (LANL)	Samer Kahook (SRNL)
Delayed Neutron Detection	LANL	Steve Tobin (LANL)	Dennis Slaughter (LLNL)
Delayed Gamma Detection	INL	David Chichester and Matt Kinlaw (INL)	Dennis Slaughter (LLNL), Steve Tobin (LANL)
Ultra-High-Res alpha	LANL	Mike Rabin (LANL)	Owen Drury (LLNL)
Ultra-High Resolution neutron	LLNL	Stephan Friedrich (LLNL)	Mike Rabin (LANL)
Ultra-High Res gamma	LLNL	Stephan Friedrich (LLNL)	Mike Rabin (LANL)
Electrochemically-Modulated Separations	PNNL	Douglas Duckworth and Scott Lehn (PNNL)	Shelly Li (INL)
Hybrid K-Edge	ORNL	Mike Ehinger (ORNL)	Tzu-Fang Wang (LLNL), Mike Collins (LANL)
Active and Passive RFID Tags	SNL	Keith Tolk (SNL)	Faranak Nekoogar (LLNL), Jennifer Tanner (PNNL), Chris Pickett (ORNL)
Automated 3-D Feature Recognition	ORNL	Mike Ehinger (ORNL)	Ana Caiado (ORNL), Lucian Mihaelescu (LBL)
Radiation Imaging	PNNL	Eric Smith (PNNL)	Ehinger (ORNL), Burke (LLNL)
Basic Nuclear Data Measurements	LANL	Peter Santi (LANL)	Winifred Parker (LLNL), Michael Todosow (BNL), Dennis McNabb (LLNL)
Calorimetry	LANL	Peter Santi (LANL)	Lee Refalo (SRNL)
Gamma-Ray Spectroscopy Algorithms	LANL	Duc Vo (LANL) and Tzu-	Eric Smith (PNNL), Tzu-Fang Wang (LLNL)

		Fang Wang (LLNL)	
Modeling for Instrumentation	PNNL	Eric Smith (PNNL)	John Mattingly (SNL)
Advanced Control and Integration			
Statistical Process Control	LANL	Tom Burr (LANL)	Jon Schwantes (PNNL)
Multi-Isotope Process Monitor	PNNL	Jon Schwantes (PNNL)	John Crebs (ANL)
Neutron Balance	LANL	Steve Tobin (LANL)	Ben Cipiti (SNL), Chuck Georgen (SRNL), Ehinger (ORNL)
Physical Properties Monitoring	PNNL	Sam Bryan (PNNL)	John Crebs (ANL)
Automated Radiochemistry	PNNL	Jay Grate (PNNL)	Tim DeVol (Clemson)
UV-Visible Spectroscopy	ANL	Alan Bakel (ANL)	Jamie Warburton (UNLV), Keith Bradley (LLNL)
Actinide Sensors for Electrochemical Processing	INL	Shelly Li (INL)	Doug Duckworth (PNNL)
Thermal Atomization Resonance Ionization Spectroscopy (TARIS)	SNL	Ben Cipiti (SNL)	Don Craczyk (ANL), Matt Douglas (PNNL)
Data authentication guidelines instruments and processes	SNL	Peter Merkle (SNL)	Dick Kouzes (PNNL)
Modeling and Simulation for Adv. Control and Integration	ANL	Alan Bakel (ANL)	Jim Sprinkle (LANL, NA-24)
Safeguards by Design			
Safeguards Performance Modeling	SNL	Ben Cipiti (SNL)	Ehinger (ORNL), Tom Burr (LANL)
Validated Systems Analysis Methods	SNL	Felicia Duran (SNL)	Peter Hester (ODU), Trond Bjornard (INL)

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