#### INEEL ENVIRONMENTAL MANAGEMENT NONDESTRUCTIVE ASSAY ACTIVITIES

Y. D. Harker, J. D. Cole, M. W. Drigert, R. J. Gehrke, J. K. Hartwell, J. L. Jones, J. W. Mandler, C. A. Mcgrath, C. V. McIsaac, G. K. Becker, J. C. Determan, G. A. Cordes, and M. J. Connolly

## Idaho National Engineering and Environmental Laboratory P. O. Box 1625 Idaho Falls, ID 83415

#### ABSTRACT

Successful management of the U. S. Department of Energy (DOE) environmental management wastes at the Idaho National Engineering and Environmental Laboratory (INEEL) has required the development of advanced nuclear based nondestructive assay (NDA) technologies and methodologies. These activities have been directed towards addressing current and future NDA needs associated with (a) spent nuclear fuel, (b) contact-handled transuranic (TRU) waste, (c) remote-handled transuranic waste, and (d) environmental monitoring and remediation programs.

The types of activities undertaken include:

- 1. Fission physics research to develop NDA technology based on correlated gamma and neutron data for assay of high activity wastes.
- 2. Radiation transport analysis to support development of quantitative prompt gamma neutron activation analysis and gamma ray transport analysis for simple gamma assay systems.
- 3. Development of gamma and neutron based down-hole probes for subsurface characterization.
- 4. Development of accelerator based, high-energy x-ray fluorescence to support subsurface contaminant modeling.
- 5. NDA applied to radionuclide characterization of TRU waste.
- 6. Validation of NDA characterization data for waste containers.

This paper presents an overview and discusses the application and benefits of these activities.

## **INTRODUCTION**

The Idaho National Engineering and Environmental Laboratory (INEEL) is both a customer and developer of nondestructive assay (NDA) systems. The programs in which the INEEL is involved and for which NDA plays a major role are: the Spent Nuclear Fuel program, the Contact Handled Transuranic (TRU) Waste program, Remote Handled TRU Waste program, and the Environmental Monitoring and Remediation programs. In order to meet present and future needs in NDA, the INEEL is involved in the following research and development efforts.

Fission Physics Research is directed toward advancing a revolutionary technique that can assay fissile nuclides (e.g., U-235, Pu-239, etc.) directly without apriori knowledge of fissile material constituents.

This concept is based on the use of multi-detector arrays consisting of both neutron detectors and highpurity germanium (HPGe) gamma-ray spectrometer detectors operating in fast coincidence. At INEEL, this concept is being developed in a two pronged approach; i.e., (a) develop a prototype detector system for assaying high-activity samples of spent nuclear fuel or remote handled TRU waste and (b) develop the database needed for analyzing the prompt fission fragment coincidence data. The prototype system is called the multi-detector analysis system (MDAS), and its development is sponsored by National Spent Nuclear Fuel program and the Mixed Waste Focus Area/RH-TRU program. The fission database generation effort is sponsored by the INEEL Verification and Validation program.

The Gamma Radiation Transport Analysis development effort is directed toward enlarging the role that simple gamma-ray spectrometer systems can play in assaying certain types containerized transuranic waste. In this effort, it was demonstrated that with analysis techniques developed at INEEL, assay results consistent with regulatory and shipping requirements can be obtained using simple passive gamma spectrometer systems. Therefore, such gamma-ray spectrometers can be utilized to provide absolute assay data with less capital cost and often less operating cost.

The Gamma and Neutron Based Down-Hole Probe development program began because the Environmental Monitoring and Remediation programs are in need of better methods to characterize subsurface concentrations of nuclear waste and hazardous materials. INEEL is developing two probes which can be inserted in test holes or monitor wells to determine subsurface concentrations of hazardous materials such as chlorinated solvents, and subsurface concentrations of fissile materials such as plutonium, uranium. One probe will detect hazardous material via prompt gamma neutron activation analysis and the other probe will identify fissile nuclides by the prompt fission neutron detection.

The Accelerator Based, High-Energy X-ray Fluorescence techniques are being developed to support subsurface contaminant modeling. A proof-of-concept assessment has been made of an electron accelerator-based system to interrogate and detect uranium, transuranics, and heavy metals in soil samples representative of the Radioactive Waste Management Complex (RWMC) Subsurface Disposal Area (SDA). Software to monitor the operation of NDA equipment is also being developed and has been demonstrated in this research.

NDA Applied to Radionuclide Characterization of TRU Waste was initiated to reduce ambiguities in radioassays where standard analytical techniques are employed.

The validation of characterization data for waste containers is being approached through, first, automated data validation systems using both expert system automated data validation techniques and pattern recognition approaches and, second, the development of waste matrix surrogates. Data validation can be the most costly and time consuming component in certifying waste for shipment and disposal. In one program, the INEEL has developed and is now implementing an automated data review and validation system, which is designed to reduce the time required for persons to review and validate NDA data. Based on a predefined set of rules, the automated system reviews the data and provides the data reviewer with a list of anomalies or concerns, which require the attention by the data

reviewer. By reducing the number of items that a data reviewer has to actually review, the automated system saves time and money. In another program, the INEEL is working to specify, design, and fabricate waste matrix surrogates. The surrogates are used to support nondestructive waste assay system development, end-user acceptance testing, NTWP Performance Demonstration Program (NDA PDP) in addition to NDA characterization compliance demonstration activities supporting technical audits.

#### FISSION PHYSICS RESEARCH

It has been long been recognized that simultaneous detection and identification of prompt fission fragments and associated prompt neutrons can, by charge and nucleon conservation, determine, which isotope had undergone fission. As an example, identifying Te-134, Mo-105 and a single neutron in coincidence would indicate that a Pu-240 nucleus had undergone spontaneous fission. In early systems designed to assay fissile materials, such a principle could not be adequately utilized because such systems lacked the time response, energy resolution and/or sensitivity to detect the prompt fission fragments and neutrons in very narrow time coincidences. With the advent of large detector arrays utilizing multiple high-purity germanium (HPGe) gamma-ray detectors with very fast coincidence technology, the ability to gather correlated time and radiation type data was made possible. These first detector arrays (e.g., Gamma sphere, Euroball, and Eurogam) were expensive and were primarily used to study the physics of fission. Some of the INEEL physicists were able to participate in these fission physics experiments and have utilized that knowledge to develop spin-off systems that support different application programs at INEEL (1).

In the late 1990's, INEEL undertook the task of developing a revolutionary prototype multi-detector array, similar to the physics detector arrays, which could be used to assay the fissile content of high activity samples; e.g., spent nuclear fuel and/or remote handled transuranic waste. The prototype system, called Multi-Detector Analysis System (MDAS), is currently undergoing testing to determine its performance characteristics and to demonstrate its ability to assay fissile materials under high activity environments. It is an integration of the basic understanding of physics and the new results obtained in recent fission studies with large arrays of detectors (2). The system has 68 detectors, 20 HPGe and 48 xylene detectors. The xylene detectors are liquid scintillators that can distinguish between gamma-ray events and neutron events through the use of pulse-shape discrimination techniques.

The time-correlated nature of the signals from the array of detectors, of multiple types, is used to reduce the underlying spectroscopic backgrounds by orders of magnitude over conventional methods. This is done by not recording the data in the time intervals between correlated signals. By means of analysis software, MDAS compensates for extended source size, radiation attenuation, high backgrounds and non-fissile radioactive material present. The quantification of fissile material is determined from counting of neutron coincidences. Fissile material composition; i.e., the relative abundance of different fissile nuclides, is determined from time-correlated fission fragment decay gamma rays.

The MDAS prototype configuration, initially assembled at the TREAT facility of Argonne National Laboratory (ANL) in Idaho, was designed to assay spent nuclear fuel rods. A slightly different

configuration has been designed for assaying RH-TRU waste. In order to provide an enhanced capability to do concept testing, without getting bogged down in operational details, the MDAS detectors have been reconfigured at an experimental station on a neutron beam line at the Oak Ridge Electron Linear Accelerator (ORELA). The ORELA phase of this project is to provide an integrated test of the MDAS using an intense and well-characterized neutron source. Later his year, the ORELA/MDAS setup will be used to demonstrate performance in assaying high-activity samples. Later in the program, MDAS detector array will be reconfigured back at INEEL where it will be combined with its own dedicated neutron source.

To support the analysis of data taken by MDAS technology, a completely new database of correlated prompt fission fragment decay data and neutron multiplicity data had to be established. Such a database has been developed for spontaneous fission nuclides; e.g., Pu-240 and Cf-252. However, no such data exists for neutron-induced fission. Beginning in 1998, the INEEL established, with a strong collaboration from the ANL Physics Division, a smaller detector array at the ANL Intense Pulsed Neutron Source (IPNS). Under the INEEL Fission Physics research program, the IPNS detector stand has been used to take correlated fission fragment gamma-ray decay data and correlated neutron data form neutron-induced fission of U-235, Pu-239 and U-233. The analysis of these data is a lengthy process and many collaborators are working on the task. To date, preliminary fission fragment decay-gamma correlation matrices, which can be used by MDAS type assay systems, have been produced for neutron-induced fission of U-235 and Pu-239.

In addition to providing data for MDAS type assays, the IPNS experiment is providing new data into the aspects of physics of fission, data heretofore unavailable. As a result, new nuclear models are being proposed by the nuclear theorists to explain the previously unobserved phenomena found in the new fission data.

This past year a second fission physics experiment has been installed at ORELA. This second experiment will complement the IPNS experiment by providing correlated fission fragment data as a function of the interrogating neutron energy, whereas the IPNS experiment is limited to thermal neutron energy.

## GAMMA RADIATION TRANSPORT ANALYSIS

The INEEL transuranic waste characterization program currently uses a Canberra Q-2 gamma assay system to determine isotopic ratios only. This method is strictly a passive gamma spectrometer system. The INEEL NDA research program has been investigating how such a simple passive gamma spectrometry system, and others like it, can be used to perform absolute assays, which would meet the requirements for shipping and storage of transuranic waste. Under this research, an analysis method has been developed to use a simple passive gamma system as a stand-alone assay device. In contrast to most common commercial assay techniques, which typically use auxiliary transmission sources, this analysis method relies on gamma signatures from the primary detector assay spectra. The spectral data are combined with physical dimensions of a drum, the net weight of the matrix in the drum, the fill height of the matrix and general chemical information about waste matrix to calculate an attenuated efficiency

curve in real-time. The primary assumptions are: (a) the drum, either by rotation or by physical composition, has azimuthal symmetry about the centerline of the drum, (b) the matrix can be represented by a right circular cylinder of uniform density, and (c) the radionuclide composition can be represented by cylindrical shells within the matrix cylinder. These simplifying assumptions greatly reduce the complexity of the problem and allow for the solution of an attenuated efficiency curve for each drum assayed.

The analytical efficiency method, developed under the INEEL NDA research, was applied to the analysis of gamma-ray spectra from the INEEL Stored Waste Examination Pilot Plant (SWEPP) Gamma Ray Spectrometer (SGRS). The analyzed data were from assays ranging from surrogate drum tests to real waste drum assays. Based on the data analyzed so far, the analysis method has produced absolute values for plutonium mass, which are, on the average, approximately 15% biased low from the known values. In addition, the drum-to-drum variation of the assay results about the known values are comparable to those observed for the SWEPP Passive Active Neutron (PAN) assay system.

Currently, this analysis method is being incorporated into the next version of the SWEPP Gamma Ray Analysis Package, used as the production analysis code for the SGRS. This will give the SGRS the capability to perform absolute assays as well as provide isotopic mass ratios. Adding the absolute assay capability to the SGRS improves the overall throughput capacity of SWEPP when characterizing drums for shipment to WIPP and it also provides a much needed backup to the INEEL PAN assay system.

# GAMMA- AND NEUTRON-BASED DOWN-HOLE PROBES FOR SUBSURFACE CHARACTERIZATION

The INEEL is currently developing advanced nuclear-based NDA instruments and methods for use in subsurface characterization. In particular, gamma- and neutron-based down-hole probes are being developed together with pertinent nuclear data required in the analysis and interpretation of the data obtained by these instruments. In addition, a facility is being developed where this technology can be tested and the instruments calibrated.

Two different down-hole probes are being developed. Both utilize neutron generators and radiation detectors. The neutron-gamma (NG) probe will measure, in the passive mode, the gamma-emitting radioactive constituents of the subsurface material. In the active mode, it will measure elemental composition, using gamma rays from neutron capture and neutron inelastic scattering. The NG probe is a major improvement over the active gamma down-hole probes currently used (which use isotopic neutron sources) in that it can measure more elements (e.g., carbon and oxygen). This is very important to the Waste Area Group (WAG) 7 effort, which needs to know the carbon-to-chlorine ratio in order to determine how much carbon tetrachloride is present in the waste buried at the Radioactive Waste Management Complex (RWMC). In addition, measuring these additional elements can provide information for use in determining the category of waste being interrogated.

The prompt fission neutron (PFN) probe will measure, in the passive mode, the neutron flux emitted by the subsurface material. In the active mode, it will measure fissile material to concentrations as low as about 10 nCi/g, neutron die-away, and the macroscopic neutron removal cross section. These measurements are of special interest to the WAG 7 effort. The PFN probe will have a lower limit of detection for plutonium that is much lower than that attainable via passive gamma-ray spectrometry. In addition, in most cases (i.e., the absence of appreciable amounts of other high neutron capture cross section materials), it can measure the chlorine concentration without suffering from the saturation effects that prevent the neutron-gamma technique from accurately measuring chlorine concentrations above about 15%.

Pertinent nuclear structure data needed in the analysis and interpretation of down-hole gamma-ray spectrometric data are being measured. High-quality gamma-ray spectra from actinides (including aged material in which sufficient amounts of progeny are present to be detectable) are being measured. They will be used to correctly identify the radioactive components in down-hole gamma logs and in other waste at the RWMC. Gamma-ray production rates are being measured for neutron inelastic scattering as a function of neutron energy. These measurements, being performed at the Los Alamos Neutron Science Center (LANSCE), are providing data for use in modeling efforts to calculate the gamma-ray spectrum from a complex sample. The production rates for alpha-induced reactions on light elements are being measured. These data are being used to develop enhanced methods for assay of waste containing fissile material and to identify the chemical form of actinides in waste. Modeling is being investigated as a means to calibrate the NG instrument. Since most INEEL down-hole probe applications involve inhomogeneous and usually unknown matrices, traditional calibration methods using a series of homogeneous, known standards are not practical. Calibration using modeling techniques is, therefore, being investigated. It is expected that these modeling techniques will enable the NG probe to provide accurate, quantitative results.

The Subsurface Neutron Interrogation Facility (SNIF) is being developed for testing and calibration of nuclear-based down-hole NDA instruments. This facility, located at INEEL Test Reactor Area (TRA), consists of surrogates with center holes in a gravel matrix. Two sizes of surrogates (2 ft and 4 ft diameter, each up to 17 ft in length) will be available. The surrogates will consist of cylindrical containers into which a matrix material is placed (together with a non-homogeneous array of other constituents, if desired). The smaller surrogate will be used for testing down-hole probes, performing sensitivity and parametric studies, and obtaining benchmark data to validate models. In general, the larger surrogate will be used for calibration and benchmark purposes.

As stated above, the down-hole probes are being designed to enhance the down-hole logging capabilities for the WAG 7 and other subsurface characterization efforts, and the SNIF is being designed to support the development and testing of down-hole NDA instruments. Benefits to the INEEL have already been realized from these research studies. For example, the data on the gamma rays emitted by alpha-induced reactions on light elements have enabled us to identify the chemical form of americium in INEEL Subsurface Disposal Area Pits 4, 9, and 10 and to identify and correct erroneous conclusions initially made from interpretation of down-hole logging results at RWMC. The gamma-ray spectral data obtained from actinides has enabled us solve assay problems that recently

arose at SWEPP and to correct errors in recently published data concerning the gamma-ray spectrum from Am-241. These data have also assisted us in interpreting logging data from the above-mentioned Pits 4, 9, and 10, enabling us to identify the unexpected presence of excess Np-237 (i.e., much more than can be attributed to the decay of Pu-241 or Am-241) and Th-228 not coming from the decay of Th-232.

## ACCELERATOR-BASED, HIGH-ENERGY X-RAY FLUORESCENCE TO SUPPORT SUBSURFACE CONTAMINANT MODELING

## Unconventional Atomic X-ray Fluorescence (UAXRF)

A proof-of-concept assessment has been made of an electron accelerator-based system to interrogate and detect uranium, transuranics, and heavy metals in soil samples representative of the Radioactive Waste Management Complex (RWMC) Subsurface Disposal Area (SDA). This technology assessment is based heavily on the "unconventional" Atomic X-ray Fluorescence (UAXRF) research being performed at the Idaho State University's Idaho Accelerator Center (IAC).

In this interrogation process, an accelerator accelerates electrons up to several MeV's in kinetic energy. These electrons interact with a high-Z material to produce high-energy, continuous-energy, bremsstrahlung (photons) radiation. Based on the selected electron beam energy, these photons can be highly penetrating. These penetrating and scattered photons will probe atomic energy levels (up to 100's keV) in the XRF process occurring during the accelerator pulse. The unconventional nature of the XRF comes from the use of these high-energy, highly penetrating bremsstrahlung photons, the high intensity level of XRF stimulation, and the associated specialized detection systems.

The testing configuration consists of a soil sample surrounded by several detectors: a thin window (~1%) high-purity germanium, a larger volume (~10%) high-purity germanium (HPGe), and a large 5x5 sodium iodide (NaI). The ~1% HPGe detector is sensitive to XRF emissions. On the other side of a 2-meter thick concrete wall is the electron accelerator. The electron accelerator is a variable-energy unit capable of producing electrons with energies up to 12-MeV. A 15-cm diameter hole connects the detector side from the accelerator side. The electron accelerator is centered with the hole and then laser-aligned with the soil sample located in the detector room.

Preliminary tests have used in homogenous test samples: a gold/tungsten composite foil with sand and thorium with lead. Results using a 6-MeV electron accelerator have shown thorium characterization through about 1-cm of lead shielding. The tests also show that it is possible to detect uranium and heavy metals (high Z down to Z ~30) in the soil column at concentrations below 100 ppm. Lower concentration levels can be detected for materials with higher Z. UAXRF results with gold/tungsten composite foils have shown foil detection through up to 10 cm of sand. Additional experimental and numerical optimizations are still required. In addition, by using the results from various fixed-position, detectors surrounding the soil sample, a 3-D image can be formed by rotating the soil sample column. Linear resolutions of less than 3 mm can be expected.

#### Pattern Recognition for NDA Operation Data Sets

NDA technologies provide information to characterize materials and container contents in a nonintrusive manner. The resulting characterization information then constitutes an historic, archival record that is used to support evaluations of waste processing and disposal techniques and strategies to be used with the waste configurations. It is of essence that the data collected and the decisions based on that data be verifiable into the future for environmental regulators and stakeholders and the general public.

Therefore, the INEEL has been researching intelligent information processing techniques that will monitor the performance of NDA technologies as they are in operation during the characterization process. The computer software uses the patterns of historic operation data to evaluate the ongoing equipment operation. Pattern recognition techniques provide real time capability to monitor the performance of NDA systems and interpret the resulting performance data by acting across the time stamped vector of system parameters. The entire variable vector thus is considered a pattern and is analyzed in real time to extract pattern changes that can be interpreted as data characteristics or operating system anomalies.

The initial application of these multivariate pattern recognition techniques was to monitor the operation of the variable energy electron accelerator during the research on UAXRF methods in soil characterization. This research and the results to date are discussed above.

An important part of this application, as in all similar applications, was simply to determine the important operational parameters that must be monitored in order to analyze equipment health and performance. Once these parameters were determined, the accelerator was instrumented appropriately to collect the parameter data. Consistent procedures were established and followed for accelerator operation, and the operation data was automatically collected and analyzed in near real time during the NDA characterization process. Data and user interfaces were accomplished with the commercial LabView software package.

Through this approach, the quality level of the archival characterization data can have a firm basis in the assurance that the nondestructive assay technology equipment was performing according to established specifications. This research is now past the proof-of-concept stage and the results will be used in future INEEL NDA research, development, and operations projects.

#### NDA APPLIED TO RADIONUCLIDE CHARACTERIZATION OF TRU WASTE

The  $\gamma$ -ray spectra of most actinide elements and their progeny are complex, especially in the energy region below about 300 keV. There are few  $\gamma$ -rays above 300 keV with high-emission probabilities. This means that in the energy range up to the 300 keV peak, interferences between actinides are quite common. Correction for these peak interferences is even more difficult when the relative abundance of each actinide can vary widely, as much as several orders of magnitude. Further, since the actinides in TRU waste have been decaying for many years, progeny are often prominent that are not normally observed in actinide material that has been freshly separated within the last year. In some cases, these

progeny may even dominate the spectrum. An example is the 440-keV  $\gamma$  ray of Bi-213 that is emitted by aged U-233 but not by freshly separated U-233 (3). As a result, this peak can be very useful in radioanalysis of aged U-233. In other cases where the alpha activity is very high, as with gram quantities of Am-241, alpha-induced reactions with low-Z elements, especially fluorine, can produce promptly emitted  $\gamma$ -rays that further add to the complexity of the  $\gamma$ -ray spectrum (3-6). Therefore, it is prudent to identify each and every peak in a spectrum of actinide waste to avoid large errors due to unidentified interferences in the reported results.

Because progeny gamma-ray peaks following long-half-life daughter and parent activities are often detected during Ge gamma-ray spectral analysis of TRU waste, the decay equations can often be used to determine the time since the parent actinide was pure. This fact was used to determine the potential presence of Np-237 in excess of that occurring from the decay of the parent, Am-241. This was done by measuring the intensity ratio of the 312-keV  $\gamma$ -ray peak of Pa-233 to the 322-keV  $\gamma$ -ray peak of Am-241. Under special circumstances this ratio can even be used to estimate the age of the waste (7).

Also observable in Ge gamma-ray spectra of TRU waste is the presence of prompt reaction gamma-ray peaks resulting from nuclear reactions initiated by the alpha particles commonly emitted by actinides, with the low-Z elements present in the waste. In particular, the elements Be, B, C, N, O, and F have low energy reaction thresholds. When the neutron/gamma emission ratio is known, these reaction gamma rays may be used to determine the fraction of the total neutrons emitted from the waste that are due to neutrons produced from ( $\alpha$ , n) reactions. To do this, corrections for gamma-ray attenuation in the waste must be made. An initial attempt to analyze TRU waste for fissile material by this method resulted in assays that were within 30% for about one third of a set of INEEL TRU waste drums of various matrixes (3).

The above methods were developed as a result of questions by the waste assay group after employing analytical techniques that left ambiguities in the radioassays. These more robust methods of radioanalysis were developed as a consequence of the Ge spectrometry studies of the actinide elements funded by the INEEL Verification and Validation Research Program. This project involved the acquisition, analysis, and presentation in color graphics of the gamma-ray spectrum, the decay chain,  $\gamma$ -ray energies and emission probabilities, and the decay scheme of individual, pure actinides. The decay data is evaluated data downloaded from the Brookhaven National Laboratory (BNL) Evaluated Nuclear Structure Data File (ENSDF). The Gamma-ray Spectrometrists in today's environment through PC computers and the World Wide Web (see Web Site at http://id.inel.gov/gamma). It has been initially funded to expand the R. L. Heath original Ge Gamma-ray Spectrum Catalogue into the actinide region using present-day HPGe detectors.

Since all of the acquired actinide spectra for the Ge Gamma-ray Spectrum Catalogue were accumulated with Ge spectrometers equipped with the INEEL-developed dual-energy pulser, in principle the spectra of individual radionuclides can be combined. By combining specific fractions of the individual actinide spectra one could, therefore, form composite spectra simulating the actual waste. Such a process can

be a very useful tool to evaluate potential peak interferences during spectral analysis and to the quality assurance of radioanalytical results.

#### VALIDATION OF NDA CHARACTERIZATION DATA FOR WASTE CONTAINERS

There are three separate programs at the INEEL that are addressing the validation of NDA characterization data for waste containers. The first uses expert system technology, the second uses pattern recognition technology, and the third involves the specification, design and construction of waste matrix surrogates.

#### **Expert System Technology**

Three projects based on expert system technology are currently ongoing. These are: (a) the data review expert system for the Passive/Active Neutron and gamma assay system at the SWEPP facility at INEEL, (b) the data review system for the High Efficiency Neutron Counter (HENC) and FRAM systems at the Radioassay Nondestructive Test (RANT) facility at the Los Alamos National Laboratory (LANL), and (c) the development of a rule editor that extends the capabilities of the data review expert system. This overview will briefly discuss the data review expert system and the rule editor tool and propose potential applications of these tools both for waste NDA and more generally.

The data review expert system addresses the need to know and demonstrate the quality of NDA waste characterization data generated for use in the National Transuranic Waste Program (NTWP). It is a requirement that each NDA measurement receive an independent technical review by a qualified expert. With hundreds of raw and reduced data acquired on each waste drum and hundreds of thousands of such drums in the national inventory, this requirement mandates a large and costly data review/validation effort. For this reason, researchers at the INEEL have developed a generic expert system framework for automated review/validation of waste NDA data. The expert system is designed around an objectoriented and rule-based framework that can be easily customized for application at sites with different NDA system configurations and waste inventory properties. The system is designed in a two-level approach. At the top level, the logic of the data review process is encoded as a set of rules, and it does not change between implementations at different sites. The logic is written in terms of generic definitions of data items and their attributes, with logic functions to test the validity of a data item. At the lower level, the expert system is customized for application at sites with different NDA system configurations and waste inventory properties. Here the data items and their attributes are given definitions that are specific to the implementation site. Now that the data review expert system is developed, each implementation of the system needs only define the specific data items to be reviewed. The data review expert system itself is already written at the top level, and operates on the specific data items at the lower level.

The rule editor tool is designed to complement and extend the capabilities of the data review expert system. It will greatly aid both the development of new systems and the maintenance of existing ones. The rule editor is a user interface that presents an existing set of rules through a graphical user interface and allows the user to modify the rule definitions. New rules can also be specified through this user

interface. The system works by storing the fundamental rule data in a database, independent of any programming language syntax. This means the information is presented to the user in a way that is independent of the CLIPS expert system programming language in which the rules will ultimately be expressed. This makes rule editing and specification much easier for the user. A program then reformats this information, adds CLIPS syntactic elements, and incorporates the new expert system code in the generic portions that are never modified. While the rule editor could be used by both software specialists and others, further INEEL work is required (and is proposed) to allow non-software specialists (e.g., data review personnel) to modify and specify rules. This work would involve developing an advanced debugging capability within the data review expert system, such that the operation of new or modified rules could be verified and problems quickly isolated.

Due to its design, the data review expert system is applicable wherever large amounts of data have been mandated for extensive review. It is currently being implemented for review of waste NDA data generated at facilities at both the INEEL and LANL. The data review expert system is currently planned for implementation with all of the NDA systems at LANL's RANT facility, and it is being considered for wider use within the DOE complex. ANTEC, a private company, is interested in applying this system at the Rocky Flats Environmental Test Site in Colorado. In addition, data off of other waste characterization instruments, such as headspace gas analysis, could also be reviewed by this system. Looking further, this system could potentially be applied to other applications such as materials control, materials accountability, and spent fuel characterization.

#### Pattern Recognition for NDA Measurement Data Sets

Pattern recognition for NDA measurement sets is applicable to individual safeguards waste NDA systems as well as interconnected systems consisting of multiple measurement stations. A project at INEEL is considering pattern recognition software for state-of-the-art NDA measurement techniques in order to exploit the measurement data sets for information not otherwise noted or utilized in conventional data analysis techniques. Pattern recognition techniques are able to take advantage of additional information contained either directly in the data or in conjunction with other information related to the measurement. Conventional approaches to these NDA process categories are generally based on first principle techniques, which for the most part are time and resource intensive and in many cases subject to unknown error components. Often conventional techniques are incapable of recognizing the presence of a bias sources subsequently not addressed or corrected for in the NDA assay quantification.

The two systems from which data have been acquired for this project are the Combined Thermal Epithermal Neutron and Crated Waste Assay Monitor systems, both developed at the Los Alamos National Laboratory NIS-6 Group. Pattern recognition techniques will be employed for the purpose of identifying data features and relationships useful to address one or both of these NDA processes. The intelligent data processing approach is planned to identify and extract unknown data features and relationships not previously used in data analysis due to constraints inherent in conventional approaches. Features and relationships of interest are those which can be used to improve NDA system design, data acquisition strategies, data reduction methods, data interpretation, operational status and data validation.

Once identified, such features will be evaluated to identify both their physical meaning and the manner in which they can be utilized in NDA measurement improvement.

#### Specification, Design and Fabrication of Waste Matrix Surrogates

Waste matrix surrogates representative of DOE complex waste forms have been and continue to be specified, designed and fabricated. Both drum and box type waste matrix surrogates have been fabricated and deployed. The surrogates are used to support NDA system development, end-user acceptance testing, NTWP Performance Demonstration Program (NDA PDP) in addition to NDA characterization compliance demonstration activities supporting technical audits. The box type waste surrogates are designed such that the differing matrix types, e.g., metals, combustibles, plastics, can be combined in multiple combinations in a single measurement configuration box. This affords sites the ability to demonstrate that characterization of wastes of differing mixtures can be properly accomplished thereby improving overall waste packaging and disposition efficiency. To date a total of 35 surrogate matrix drums have been fabricated and deployed in the drum NDA PDP program, 14 different drum surrogates produced for use at the INEEL SWEPP characterization facility and 10 box type surrogates produced for the box NDA PDP and box assay system development. Five more different types of box waste matrix surrogates are being designed and fabricated in FY01 in this program.

Waste matrix surrogates for four different waste type categories are also being designed and fabricated in support of the TRU and Mixed Waste Focus Area (TMFA) Inner Layer Confinement Reduction project. TRU waste is typically packaged within one or more polyethylene (PE) or polyvinyl chloride (PVC) bags with one or more of these multi-layer bags of waste placed in one container (drum or box). These plastic bags are termed inner confinement layers. Flammable gas build-up occurs within these packages (or inner confinement layers) as the result of hydrogen generation caused by radiolysis of hydrogenous materials by the alpha emitters in the waste. The Nuclear Regulatory Commission has imposed a flammable (i.e. hydrogen, methane, etc.) gas concentration limit on this contact-handled transuranic waste transported using the Transuranic Package Transporter, Model II (TRUPACT-II). However, if the inner confinement layers could be breached the hydrogen would have an escape path to the TRUPACT cavity where it may be captured on hydrogen getters.

Tests are planned to evaluate a process by which the inner confinement layers may be breached. Four simulated waste types with "worst case" packaging configurations will be tested. Twelve drums will tested for each waste type, for a minimum of 48 separate tests. Each drum will contain various packaging configurations with differing surrogate waste materials, void spaces within the packages, and different types of waste containers within the bags. It is expected that if inner confinement layers can be breached for these "worst case" configurations, layers could be breached for other configurations. The project will determine the number of bags breached by type of packaging configuration (e.g., number of layers of confinement, type of plastic, type of waste, void volume in the inner bags and void volume in the liner bags, etc.). Also, the limitations associated with the process will be identified.

UltraTech International, Inc. is the inventor of the process. The tests will take place at their facility in Jacksonville, Florida. Several hundred waste packages in varying configurations are being designed and fabricated at the INEEL North Holmes Lab Complex to support the project.

#### CONCLUSIONS

This paper was written to provide an overview of the NDA technologies being developed and supported at the INEEL in response to the Department of Energy need to successfully manage the environmental wastes at the INEEL. The NDA technologies that were presented are directed towards addressing current and future needs associated with spent nuclear fuel, contact-handled transuranic waste, remote-handled transuranic waste, and environmental monitoring and remediation programs. Some of the technologies are already deployed, and some are still under research. They are all intended to provide a diversity of solutions to the problem of addressing both radioactive and nonradioactive waste. The focus is on nondestructive assay because this type of technology assays unknown wastes without disturbing the waste container or excavating the subsurface soil. Companion software tools are under development to monitor the performance of the NDA equipment and interpret the analysis data.

As work on this portfolio proceeds the INEEL is identifying the strengths and weaknesses of each NDA technology being developed. The INEEL feels strongly that successfully managing their environmental wastes is a big challenge, and that the NDA technologies under development will provide cost effective approaches to meeting the challenge at INEEL and then to transfer the technologies to other waste sites around the DOE complex.

#### REFERENCES

- 1. M. W. Drigert, J. D. Cole, and E. L. Reber, "Fission Assay Tomography," Nucl. Instr. and Meth., vol. A364, 1995, p. 394.
- J. H. Hamilton, A. V. Ramayya, S. J. Zhu, G. M. Ter-Akopian, Y. T. Oganessian, J. D. Cole, J. O. Rassmussen, and M. A. Stoyer, "New Insights from Studies of Spontaneous Fission with Large Detector Arrays," Prog. Part. Nucl. Phys., 1995, vol. 35, p. 635-704.
- R. J. Gehrke, J. K. Hartwell, C. A. McGrath, E. W. Killian, Y. D. Harker, Twedell, D. M. May, L. V. East, and C. V. McIsaac, "Peeling the Onion? The Gamma-Ray Spectrometry of Transuranic Waste", INEEL/EXT-2000-0002, Proc. of the 7<sup>th</sup> NDA Waste Characterization Conference, 2000, vol. 1, p. 203.
- C. E. Moss and J. T. Caldwell, "Assay of TRU Wastes Containing (α, n) Sources," Proceedings of the 27<sup>th</sup> Annual Meeting of the Institute of Nuclear Materials Management, New Orleans, LA, 1986 June 22-25, p 427.

- S. Croft, "The Thick Target (α, n) Yield Of Fluorine Compounds for a Broad Range of Actinides," EUR 17665 EU, Proceedings of the 19<sup>th</sup> Annual Symposium on Safeguards and Nuclear Material Management, held in Montpellier, France, 1997 May 13-15, p. 397.
- 6. A. V. Shumakov, G. N. Vlaskin, V. F. Kositsyn, and M. A. Naumov, "Estimated Detection Limits of B, C, N, O, and F In  $\alpha$ -Emitters by Using  $\gamma$ -Radiation from ( $\alpha$ , x $\gamma$ ) Reactions," Radiochemistry, 1994, vol. 36, no. 1, p. 62.
- 7. R. J. Gehrke, L. V. East, and Y. D. Harker, "Information in Spectra from Sources Containing 'Aged' <sup>241</sup>Am as from TRU Waste," Waste Management, 2000, vol. 20, p. 555.