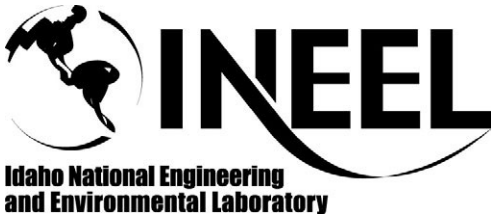


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# Multi-Detector Analysis System For Spent Nuclear Fuel Characterization

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# MULTI-DETECTOR ANALYSIS SYSTEM FOR SPENT NUCLEAR FUEL CHARACTERIZATION

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## INTRODUCTION

The Spent Nuclear Fuel (SNF) Non-Destructive Analysis (NDA) program at INEEL is developing a system to characterize SNF for fissile mass, radiation source term, and fissile isotopic content. The system is based on the integration of the Fission Assay Tomography System (FATS) and the Gamma-Neutron Analysis Technique (GNAT) developed under programs supported by the DOE Office of Non-proliferation and National Security. Both FATS and GNAT were developed as separate systems to provide information on the location of special nuclear material in weapons configuration (FATS role), and to measure isotopic ratios of fissile material to determine if the material was from a weapon (GNAT role). FATS is capable of not only determining the presence and location of fissile material but also the quantity of fissile material present to within 50%. GNAT determines the ratios of the fissile and fissionable material by coincidence methods that allow the two prompt (immediately) produced fission fragments to be identified. Therefore, from the combination of FATS and GNAT, MDAS is able to measure the fissile material, radiation source term, and fissile isotopics content.

MDAS is a new approach to the non-destructive analysis of fissile and other radioactive material. It uses the fundamental physics of fission and decay processes, very fast coincidence methods, electronics and computers available only in the past few years, and specially designed fast detectors. The time-correlated nature of signals from the array of detectors, of multiple types, is used to reduce the underlying spectroscopic backgrounds by orders of magnitude over conventional methods. MDAS uses these methods to determine the ratios of fissile isotopes, and the total quantity of fissile material in the item, using an external source of neutrons. 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 the gross counting of neutron coincidences instead of the identification of specific gamma rays selected from a spectrum of poor signal-to-noise ratio. In addition to fissile quantity, MDAS determines radioactive source term, selected nuclide inventory, and uranium enrichment. Using highly-energetic radiation, MDAS will characterizes items such as spent fuel and transuranic waste without special calibration standards or *a priori* knowledge.

The characterization data can be used independently as a qualified information for transportation, storage, or disposal of the material or to validate existing un-qualified records on the material items.

## BACKGROUND

The instrumental basis for MDAS originated from techniques used in low-energy and high-energy nuclear physics experiments performed at accelerator facilities. These techniques were first applied to measurement and verification needs of the DOE Nonproliferation and National Security Office in support of arms control and disarmament treaties.

The fundamental difference between conventional radiation detection and measurement methods and the methods used by MDAS lie in how the physical process emitting the radiation is understood and the signals generated by the detectors are collected, processed, and stored. Traditional spectroscopy and radiation detection treat radiation as a "field" or a continuous emission of energy. In reality, each time a nucleus fissions or emits radiation it is a single discrete "event" in time with individual quanta or particles of radiation. MDAS uses an event-by-event method to collect the data from the detectors, whereas traditional; spectroscopy accumulates an integrated signal. Treating radiation as a field came about because most radiation measuring equipment and detectors were "slow" with respect to the radiation emission process. In the past twenty years, radiation detectors and their support electronics have been developed that can respond in the time scales of hundreds of picoseconds ($10^{-12}$ seconds). This means that such detectors can respond to individual elements of the emitted radiation even if the emission rate is in the giga-hertz range ($10^{9}$ Hz). For comparison, this means that the decay of a source of $^{137}$Cs of one R/hr at 10 cm could be monitored on an individual atom-by-atom basis.

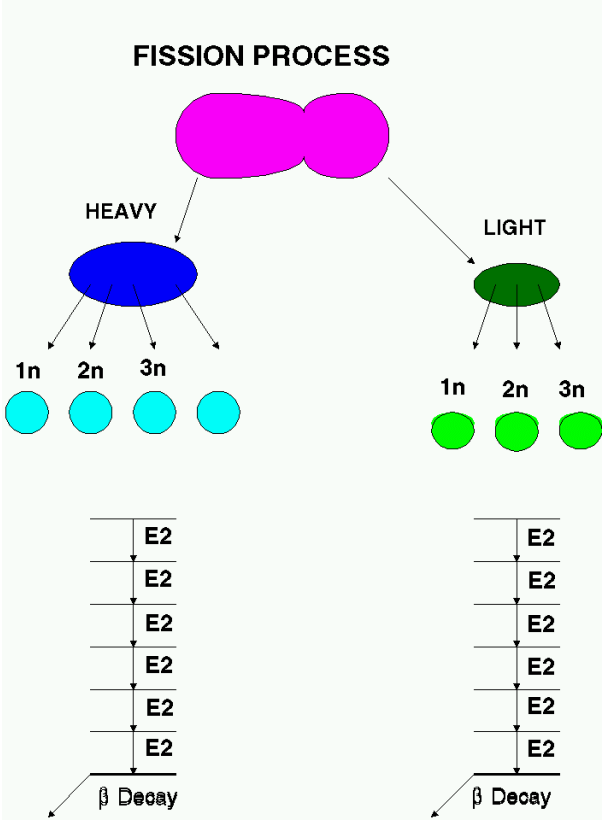
The scientific expertise that is fundamental to MDAS has been developed and validated by the experimental

nuclear physics community for many years. Innovations in computer equipment (reduced cost, reduced size, increase speed and power, networking, fiber communications, and others) allow things to be done now in real-time or within hours of a measurement that could not be done just a few years ago. The power of computer programming languages, new programming development tools, and extensive use and availability of graphical systems and interfaces reduce the need for a specialist to make NDA measurements or analyze the data. The basis and techniques of the data acquisition and analysis was developed by the physics community to satisfy the requirements of more complex and costly experiments.

In addition to new equipment, new methods for NDA use are now available. The methods used in MDAS are coincidence spectroscopy with large detector arrays[2-4], coincidence gating[5-7], list-mode data acquisition[8], fast liquid scintillators[9-12], and pulse-shape discrimination[13-15]. These combined with the new understanding of the radiation process will be addressed below.

RADIATION SOURCES

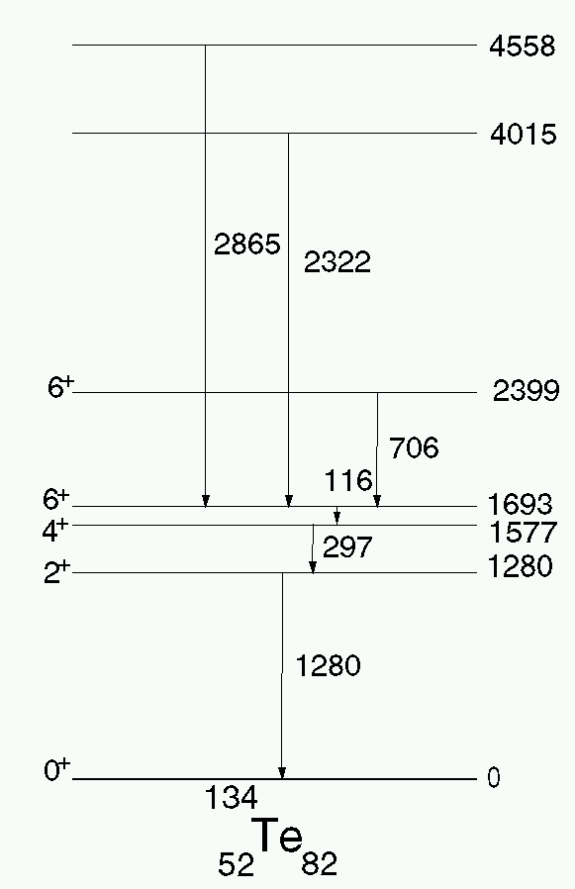
The approach used by MDAS incorporates a new understanding of the physics of radiation sources. The new understanding emerges from the correlation of the prompt radiation (gamma rays and neutrons) that occur in the fission process and in addition the multiple gamma rays that are produced by the prompt de-excitation of the isotopes produced in fission. The fission process is shown schematically in Figure 1. For the case of fission, "prompt" means a time period of approximately $10^{-22}$ seconds. The fission products are: 1) two fragments, one light mass and one heavy mass; 2) zero to 10 evaporation neutrons; and 3) several de-excitation gamma rays. The heavy-mass fragments with masses from approximately 130 mass units (A = 130) to about 145 mass units (A = 145) can have yields of 2-5% each of the total mass yield, independent of the fissioning nuclei. Except for very rare cases (approximately 1:$10^8$), fission is a binary process with the energetic (e.g., easily observable) radiation coming from the two fission fragments. The gamma rays observed as being part of the fission process are due to the de-excitation of the fragments. In previous studies, the conventional wisdom has been that the energies of gamma rays from either of the two prompt fragments lie predominately between 300 to 400 keV. This assumption was a result of the methods used in studying the fission fragments.




Until the mid- to late-1980s, fission fragments were never studied directly because of instrumental limitations that existed. Studies[16-30] undertaken by the INEEL nuclear physics staff in collaboration with partners from other DOE laboratories and universities have shown that high-energy gamma rays come from fission fragments near the doubly closed shell at Z=50 and N=82. These gamma rays can be used by a coincidence system to identify the fissioning element. Additional analysis will identify the specific isotope undergoing fission.

EXAMPLE

The even-even isotopes of $^{132,134}$Sn and $^{134,136}$Te have not only a very high yield, but also multiple gamma-ray transitions with energies of 2-4 MeV. An example is shown in Figure 2 for $^{134}$Te isotope, which, in the case of $^{240}$Pu, makes up about 5% of the fission fragment yield. We see two very high energy transitions of 2.322 and 2.865 MeV feeding the third excited state at 1.693 MeV. The vertical lines represent gamma rays that are emitted by the nucleus as it decays. Lines that are side-by-side (like the 2865, 2322, and the 706 keV gamma rays) are not in coincidence, but lines that are part of a cascade, (like

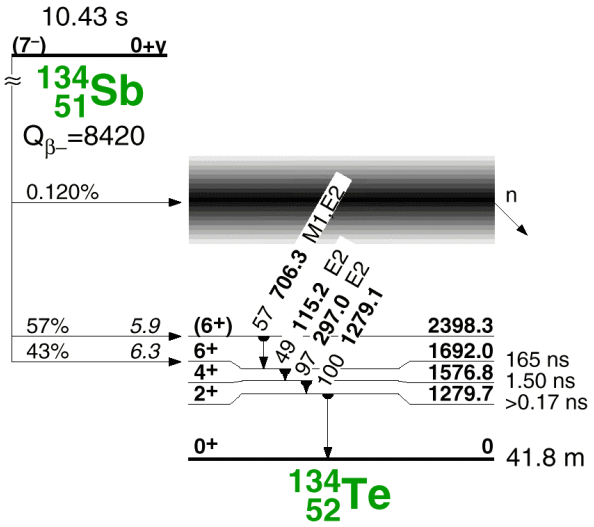



the 2865 and 1280 keV gamma rays) are in coincidence. Since, in the case of fission, the neutrons and gamma rays are all in coincidence, observing these high-energy gamma rays from one fragment makes it possible to observe the gamma rays in the partner fragment. By observing the gamma rays from both fragments, the Z (proton number) of the elements fissioning is uniquely identified. By knowing which different isotopes of the light-mass fragment are present and in what ratios, the masses (A) of the specific fission isotopes and their ratios can be determined.

Figure 3 shows the currently accepted levels in $^{134}$Te. The two high-energy gamma rays at 2.322 and 2.865 MeV have not been reported previously.

CONSERVATION RULES

The importance of proton conservation is that the two fragments contain all the protons of the original nucleus that fissioned. This results in charge (Z) conservation between the two fragments and the initial




fissile isotope.

$$Z_F = Z_H + Z_L \qquad \text{Z Conservation}$$

The two fragment isotopes and the neutrons emitted account for the mass number of the nucleus that fissioned. This is a mass number conservation that is seen in a distribution of the masses of the fragments.

$$A_F = A_H + A_L + xn \qquad \text{A Conservation}$$

Table 1 lists the light-mass fragments that are associated with the heavy-mass fragment $^{134}$Te for the fission isotopes listed in the first column. The fission half-life, in years, is included in the table as well as the specific isotopes for the cases of zero to five emitted neutrons.

The same studies[16-30] referenced previously have shown that a multiple detector array can be used to detect pairs of these fast neutrons in coincidence and determine the fissile mass present to within 50%. Proton number (Z) conservation can be seen by comparing $^{252}$Cf and $^{242}$Pu data. The equivalence of the fission of the $^{252}$Cf and the $^{242}$Pu in producing similar molybdenum fragment spectra can be accomplished by gating on the $^{134}$Te gamma rays for the $^{242}$Pu fission and on the $^{144}$Ba gamma rays from the $^{252}$Cf fission. The result is shown in Figure 4, where gamma-ray transitions of Mo isotopes are seen in both gated spectra. For all of the fissile isotopes of interest, there are several sets of fission fragment pairs that can be utilized for the purposes of identification and quantification.

| Fissioning Isotope | $T_{½}$(SF) yr | Light-mass Fragment Paired with $^{134}$Te | | | | | |
|---|---|---|---|---|---|---|---|
| | | N=0 | N=1 | N=2 | N=3 | N=4 | N=5 |
| $^{235}$U | $3.5X10^{17}$ | $^{102}$Zr | $^{101}$Zr | $^{100}$Zr | $^{99}$Zr | $^{98}$Zr | $^{97}$Zr |
| $^{238}$U | $8.19X10^{15}$ | $^{105}$Zr | $^{104}$Zr | $^{103}$Zr | $^{102}$Zr | $^{101}$Zr | $^{100}$Zr |
| $^{239}$Pu | $5.5X10^{15}$ | $^{106}$Mo | $^{105}$Mo | $^{104}$Mo | $^{103}$Mo | $^{102}$Mo | $^{101}$Mo |
| $^{240}$Pu* | $1.34X10^{11}$ | $^{106}$Mo | $^{105}$Mo | $^{104}$Mo | $^{103}$Mo | $^{102}$Mo | $^{101}$Mo |
| $^{241}$Am | $1.15X10^{14}$ | $^{108}$Tc | $^{107}$Tc | $^{106}$Tc | $^{105}$Tc | $^{104}$Tc | $^{103}$Tc |
| $^{242}$Pu* | $6.75X10^{10}$ | $^{108}$Mo | $^{107}$Mo | $^{106}$Mo | $^{105}$Mo | $^{104}$Mo | $^{103}$Mo |
| $^{252}$Cf* | 82.8 | $^{118}$Pd | $^{117}$Pd | $^{116}$Pd | $^{115}$Pd | $^{114}$Pd | $^{113}$Pd |

**Table1.** Selecting the gamma rays of a fragment identifies the fission partner in a coincidence gate. The number of protons in the two fragments must sum to the number of protons in the fissioning element. For the fission isotopes listed, the isotopes that are partners for the cases of zero to five emitted neutrons are listed for the $^{134}$Te isotope. *Spontaneous fissioning isotopes.

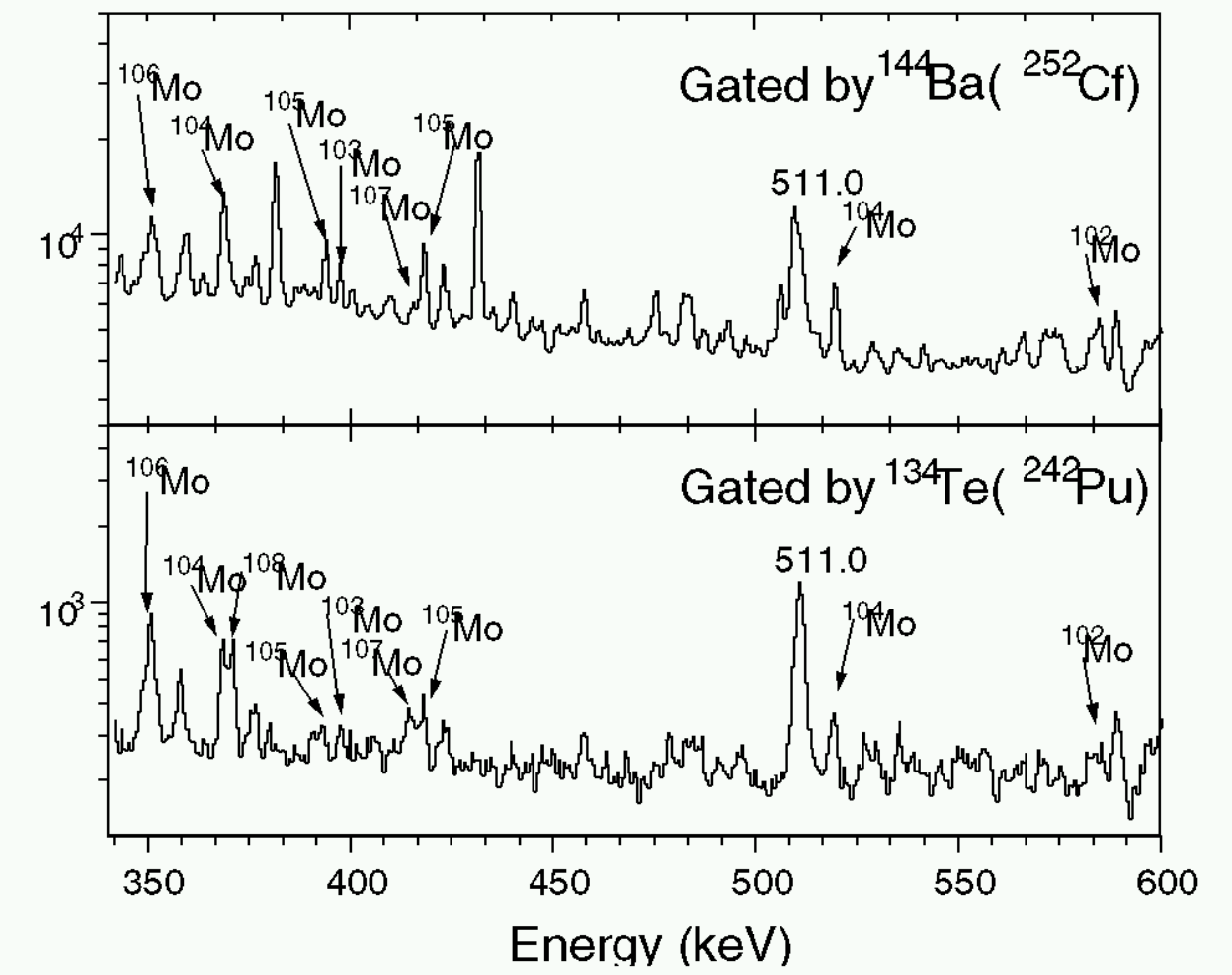



Figure 4. Two spectra from the fission of $^{252}$Cf and $^{242}$Pu gating on $^{144}$Ba lines in the californium fission and gating on $^{134}$Te lines in the plutonium fission. The partner molybdenum isotopes are labeled.

Figure 4 shows the spectra for $^{106}$Mo that corresponds to the emission of two neutrons for $^{252}$Cf and $^{242}$Pu when the complementary fragments are $^{144}$Ba and $^{134}$Te, respectively. The strong gamma ray at 350.7 keV is clearly seen in both spectra. When different isotopes of the same element are fissioning, the ratios of the relative intensities (areas under the peaks) of the gamma rays will give the ratios of the isotopes present.

MDAS DESCRIPTION

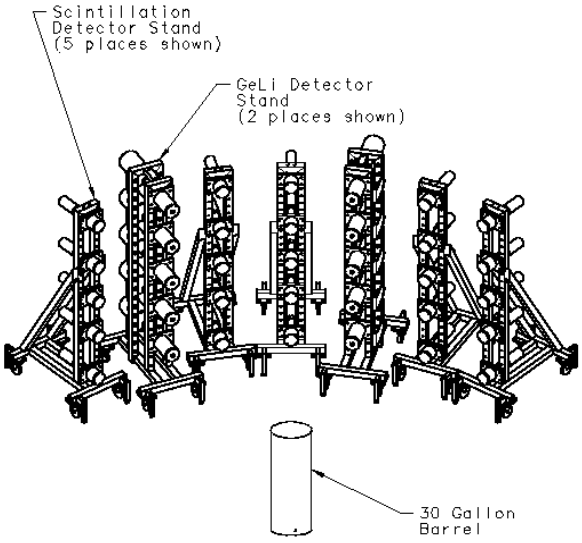



Figure 5. Drawing of one-half of the MDAS array. The array is shown to scale with respect to a cylinder representing a 30 gallon barrel.

A prototype multi-detector analysis system is being developed that will have 68 detectors; 20 high-purity germanium (HPGe) for gamma ray detection and 48 liquid scintillator detectors for neutron detection. The liquid scintillators use pulse shape discrimination[1]. This system has several advantages: fast coincidence methods, list-mode data, gamma-ray coincidence, neutron coincidence, pulse-shape discrimination, detector arrays, and data acquisition and analysis. It is important to note that MDAS integrates all the individual advantages and performs the measurement to obtain the gamma-ray and neutron data in a single operation.

Fast Coincidence Methods

Only coincidence data are recorded by the system. This means that gamma-ray energies, neutron multiplicity, and their time relations are stored by the data acquisition (DA) system when two or more detectors "see" radiation at the same time. For the system being assembled presently, this is a 50 to 100 nanosecond (ns) window. The effect of this coincidence window is to reduce the backgrounds by several orders of magnitude. Knoll[32] gives the standard formula for calculating this. The random coincidence count rate ($N_R$) for a pair of detectors is given by

$$N_R = 2\tau n_1 n_2$$

where $\tau$ is the duration of the time window, $n_1$ is the count rate in one detector, and $n_2$ is the count rate in the other detector. For example, if each of two detectors is counting at 10,000 Hz, with a 100 ns window, the random background rate is 20 counts per second. The time window that has been selected is short compared to the time windows used in the 1960s for coincidence measurements, typically hundreds of microseconds or milliseconds because of the limitations of detectors and electronics. Improvements in the last 10 years make the short time window possible, but both improved detectors and fast electronics must be used together. For example, $^3$He neutron detectors require several milliseconds to respond to the neutrons, while our liquid scintillators respond in less than a nanosecond. A pair of $^3$He detectors running at 10,000 Hz with a time window of one millisecond has a random background rate of 200,000 counts per second. Fast coincidence methods are excellent for reducing backgrounds.

List-mode Data

Another advantage is that the data are collected in "list-mode." This is a data acquisition method that does not process the data immediately, but instead stores it to computer tape and passes it to other computers for analysis. In this way, all gamma-ray energies, the number of neutrons, and the time relationships between them, are stored for every detector in the array, for each fission event, on an event-by-event basis. List mode data has two important advantages:

1. The data are stored directly from the experiment

and have the least bias from the electronics used, the DA hardware, or the analysis software used later. Data stored in this manner is easily archived for long-term record retention and can even be "re-acquired" if different analysis methods are desired. If any question as to the validity of analysis results arises, re-playing the data allows different analysis and sorting, as if the measurement were completely repeated.

2. "Gating" can be used in software that selects only coincidence relations that are important in characterizing the material. The coincidence method focuses on the difficult problems of low signal-to-noise ratios, competing channels, or high backgrounds, which degrade the signals.

The MDAS uses not only hardware gating, but also the principle of "software" gates[5-7]. This is possible because of the list-mode data storage method, and the fact that analysis is performed on a separate computer. The traditional "hardware" gates (single-channel-analyzer) cannot be changed without extensive re-calibration. A large number of these SCAs would be required for each detector, and hundreds would be required for a system like MDAS. With software gating, the selection of data of interest is done after the ADCs (analogue-to-digital converters), which allows hundreds of gates to be used on the data stream. The advantage of this method is that it not only reduces the hardware required, but further lowers the backgrounds in the analyzed data. With software gating, we only look at the particular data that is relevant to the information required.

Gamma-ray Coincidence

The method of using coincident gamma rays from the fission fragment pairs, as discussed previously in the example, is covered under the U.S. patent US5378895A. The gamma rays from the fragment pairs produced in fission are also in prompt coincidence. By identifying each fragment by its signature gamma rays, and seeing these gamma rays in coincidence, allows the pair to be identified. This identifies the fission element.

Neutron Coincidence

While gamma-ray coincidence is used to determine the fission isotopes, the fissile mass is determined using coincident pairs of fast neutrons. Only the number of pairs of neutrons observed are important, not the energy of the neutron, or the background gamma-ray field. This is a gross counting measurement that will have a small error. In addition, it ignores any possible source of neutrons other than from fission. Fission is the only non-accelerator source of coincident neutrons. In this way, neutrons from alpha reactions following the alpha decay of transuranic material are rejected by the system.

The MDAS neutron detectors are organic liquid scintillators that allow discrimination by charge collection ratios[13-15]. These detectors are extremely fast in their response (less than one nanosecond), provide good neutron-gamma-ray discrimination, and are sensitive to fast neutrons unlike $^{3}$He tubes. This is important to the coincidence-time window discussed earlier. A fast coincidence system cannot be made using $^{3}$He tubes as detectors.

Pulse-shape Discrimination

Liquid scintillators make it possible to perform pulse-shape discrimination (PSD) or charge ratio discrimination (CRD) to differentiate between neutrons and gamma rays. The particular method used for the neutron-gamma-ray discrimination is a development from our research[13-15]. We have chosen to digitize the pulse from the neutron detector in two parts and then ratio these in software. This makes the PSD very stable to environmental and other factors that can cause electronic drift and destroy the ability to discriminate between neutrons and gamma rays. Only this type of PSD will work with the short coincidence time windows. This is another important inter-relation in MDAS.

Detector Arrays

The MDAS uses arrays of detectors to achieve the high detection efficiency required for fast operation, but uses small detectors that are not overwhelmed by the high count rates that occur when measuring spent nuclear fuel. The individual detectors may be columniated and shielded to limit count rates, but the total efficiency of the system will be increased by using a large number of detectors. The traditional approach of using large single detectors will not work in a high count-rate environment. In addition, the traditional method of using multiple detectors and summing the detector outputs requires precise calibration and stability of the electronics. The MDAS approach uses software stabilization, and summing is

not an issue. The rate at which MDAS can characterize material is directly affected by the number of detectors. Increasing the number of detectors will result in shorter counting times without degrading the signal-to-noise ratio or overloading the detectors' ability to operate in an intense radiation field.

Data Acquisition & Analysis: Hardware & Software

MDAS uses high speed data acquisition methods and distributed analysis of data, both of which have been developed for high-energy nuclear physics experiments that required real-time monitoring and control. The front-end electronics are connected to the data acquisition system by dual-port memories. This eliminates the need for complex or proprietary hardware interfaces. No software execution is required in the front-end electronics, making these units as fast as is possible in hardware. New hardware does not require extensive reprogramming of the data acquisition system, thus giving MDAS a long life.

The data acquisition system is based on commercially available VME (ANSI/IEEE Std 1014-1987, ANSI/VITA-1-1994) components. VME was chosen because of the robust nature of the commercial equipment, its common use in control and acquisition systems, and the low cost of the equipment. The hardware is not as important in MDAS as the software. The data acquisition software is written in ANSI C and can be run on several VME CPUs with the proper hardware drivers. Without software processing, the front-end electronics fill a dual-port memory with a stream of integer output from the ADCs. The data acquisition system reads this memory for the data, reformats the data, stores the data to tape, and passes the data to another networked computer via a FDDI interface for immediate processing. No complex or proprietary interfaces are used by the data acquisition system. Very high data rates can be handled, with the limit being the tape system used for storage. For MDAS, this is an 8 mm tape, but an operational system would be better served by a 19 mm drive.

All processing is performed either on the networked system without interfering with the data acquisition system or as post processing at a remote site. This makes it possible both to obtain immediate results and to perform detailed analysis for regulatory reporting, data archiving, and storage for later retrieval.

Separating data processing from data acquisition means that more data can be acquired by the acquisition system as it is not hindered by the need to process the data. Also, additional analysis can be performed to correct for such items as self-attenuation of radiation in the item being measured, extended source size, failure or changes in the detectors or front-end electronics, or any unusual occurrence that arises in the measurement. Another advantage is that post analysis allows for attenuation correction of radiation from the item being measured. This reduces the dependence upon geometry and removes the need for making calibration standards for every different geometry of spent nuclear fuel or barrel of waste.

With current computer technology, processing the data from a measurement is limited only by the power of the system used for analysis. A workstation-grade machine would process the data in roughly the time needed to acquire the data for the prototype system. Processing speed can be improved in the same manner as that mentioned for the DA prototype.

CONCLUSIONS

The prototype system is the test-bed to customize the basic methods and characteristics, such that specific information can be obtained for the spent nuclear fuel and transuranic waste programs of DOE. The basic physics has been proven, and, in some special cases (arms control verification), particular systems have been developed as demonstrations. The successful prototype will also serve as the basis for an operational system, part of which involves the customization of methods and hardware for specific tasks and results.

Summarizing the important characteristics:

- Prototype uses 68 detectors
- Neutron coincidences are used to determine fissile mass
- Neutron detectors are organic liquid scintillators, very fast response
- PSD allows selection of neutron coincidence events
- Gamma-ray detectors are specially designed HPGe
- Only coincidence events are acquired as data
- Short coincidence time window reduces

random background

- Data are acquired, analyzed, and stored as list-mode format
- List-mode format is easily archived, retrieved, and re-analyzed
- Software gating selects only data of interest for analysis
- Distributed computing approach separates data acquisition and analysis

The prototype system will be used in further development tests at Argonne National Laboratory - West.

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