



NONDESTRUCTIVE ASSAY TECHNIQUES APPLIED TO NUCLEAR MATERIALS

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INTRODUCTION

Los Alamos National Laboratory was created at the birth of atomic energy and has played an important part in its application and control ever since. Nuclear material safeguards was born with the initial production of nuclear materials in the early 1940s. The Los Alamos Safeguards Program has worked for 30 years to improve the worldwide control and accounting of nuclear materials, and it is vigorously pursuing a program of application and research to meet the problems of today and the future.

The international community needs to develop methods to assure that plutonium and highly-enriched uranium will not be diverted from reactor use to nuclear weapons production. The International Atomic Energy Agency (IAEA), which was created by the international community in 1957, is tasked with the responsibility of safeguarding these materials. The treaty on Nonproliferation of Nuclear Weapons specifies that every "non-nuclear-weapon state" (all countries except China, France, the United Kingdom, the United States, and Russia) is obliged to conclude an agreement with the IAEA for applications of safeguards to all its peaceful nuclear activities. The purpose of these safeguards is to verify that the country of concern is not diverting nuclear materials from peaceful uses to nuclear weapons.

The concept of safeguards is significantly different in nuclear weapons states. The concern is not diversion of nuclear materials from peaceful research and energy production to nuclear weapons production; rather, it is maintenance of detailed knowledge of the location and quantities of nuclear materials, through adequate measurements and bookkeeping, as well as physical protection of the materials. These aspects, known commonly as Materials Protection, Control and Accounting, are essential to preventing illicit diversion of nuclear materials to individuals and organizations involved in clandestine operations (e.g., terrorists and rogue countries). These methods are also applied in non-nuclear-weapons states to prevent unauthorized access and removal of nuclear materials. There are three aspects that need to be in place in order to maintain a valid safeguards system:

1. Physical protection — guarding the access to nuclear materials using physical protection and surveillance.
2. Accounting systems — computer based accounting systems that provide the current location of nuclear materials, quantities, and the uncertainty in the assayed values.
3. Measurement systems — detectors, data acquisition systems and data analysis methods that provide accurate assays of nuclear material quantities for the accounting system.

This paper will discuss nondestructive assay (NDA) techniques as they apply to the third aspect of safeguard systems, measurement systems. NDA is defined as “the quantitative or qualitative determination of the kind and/or amount of nuclear material in an item *without alteration or invasion of the item.*” This is contrasted with destructive analysis, which is the process of taking small samples from the item in question, analyzing those samples by chemical analysis, destroying the original nature of the samples in the process (hence the term “destructive”), and applying the results to the entire item.

Measurement of nuclear materials before packaging may be performed either destructively or nondestructively. However, after the materials are packaged it is not economically feasible to open the containers for sampling for destructive analysis. NDA is then the method of choice for analyzing the entire item rapidly, inexpensively, and accurately. NDA measurements are the only reliable techniques available for items that are physically and chemically heterogeneous.

Over the past 30 years, numerous techniques that use the atomic and nuclear properties of actinides have been developed for reliable, rapid, accurate, and tamper-proof NDA of nuclear materials. We devote the rest of this article to NDA methods and their application to nuclear materials. More details and references can be found at <http://www.nis5.lanl.gov>.

We distinguish between two types of NDA measurements: the first involving the detection of spontaneously emitted radiation, produced by the natural radioactive decay processes; the second involving the detection of induced radiation produced by irradiating the sample with an external radiation source. Most of the uranium and plutonium isotopes spontaneously emit radiation that can be detected away from the sample. For example, ^{240}Pu undergoes spontaneous fission emitting two to three neutrons per fission event. Uranium-235 emits a 186-keV gamma ray following its decay by alpha emission. In general, neutrons provide better assay of bulk nuclear materials because they are much less attenuated compared to gamma rays. Consequently, quantitative uranium assay usually involves inducing fissions in ^{235}U using an external neutron source. However, gamma rays are used to determine the enrichment of uranium samples using the 186-keV gamma-ray radiation emitted by ^{235}U . They are also used for waste assay measurements where the material density is low enough to allow most of the produced gamma rays to escape the sample.

NEUTRON MEASUREMENTS

Fission is not the only nuclear process that emits neutrons: (α, n) reactions with light elements (such as oxygen, chlorine, fluorine, and others) cause the neutron emission

rate to be dependent on the chemical composition of the material. The effect of these neutrons on the neutron assay process can be isolated by coincidence counting.

A schematic passive neutron counter is depicted in Figure 1. The plutonium sample is placed in the center of a polyethylene barrel. Helium-3 filled proportional counter tubes are positioned in the polyethylene, parallel to the central axis of the barrel. The neutrons emitted from the sample are thermalized in the polyethylene and are subsequently captured by the ^3He in the proportional counters. The capture process produces a proton and a triton, which in turn produce a pulse in the counter. The train of pulses produced by the counter is analyzed by a shift register. This circuit specifically provides the intelligence to distinguish (on average) between correlated neutrons emitted by the fission process, and random neutrons emitted by (α,n)

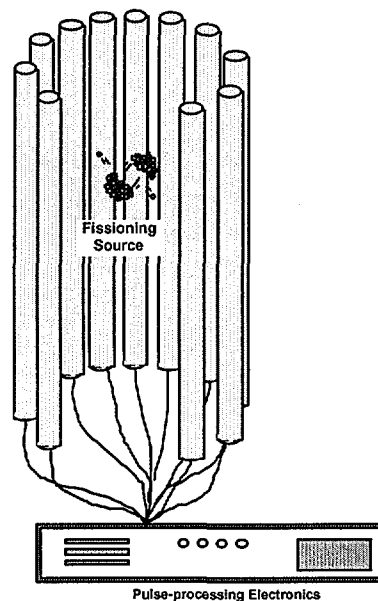


Figure 1: Schematic of passive neutron coincidence counting. A fission source emits neutrons that are moderated in a polyethylene barrel (not shown) and subsequently captured in ^3He proportional counter tubes. The pulses produced by the tubes are analyzed by a shift register which determines the number of time-correlated neutron pairs.

reactions. This is done by analyzing the time correlations between the pulses. The (α,n) neutrons are uncorrelated in time, whereas fission neutrons are emitted two to three at a time, and therefore, a significant fraction of these neutrons come in pairs (and sometimes in higher multiples). By analyzing the number of coincident pulses (within a specified time window, typically around $60\ \mu\text{s}$) compared to the number of single pulses, one can determine the ^{240}Pu content of the material, as well as the contribution of (α,n) reactions. This process is known as coincidence counting. The error in the mass of a plutonium sample using coincidence counting can be as low as 1%.

When analyzing large items, additional bias is caused by multiplication. The effect of multiplication can be measured from triple-correlated neutron pulses triples. A full measurement of singles (uncorrelated pulses), doubles (two-fold coincidence events), and triples provides a complete determination of the (α, n) reaction rate, the ^{240}Pu spontaneous fission rate, and the multiplication of the sample. This process is known as multiplicity counting.

State-of-the-art neutron detectors for safeguards applications contain a large number of ^3He proportional tubes in order to attain neutron detection efficiencies approaching 80%. Such high efficiencies are necessary in order to detect triples with good statistics. The tubes are strategically placed in the polyethylene moderator to maximize efficiency while also minimizing the effects of sample position and constituents on an assay. Other materials used in detector construction are chosen to minimize the escape of neutrons and to ensure the safety of personnel when the radioactive materials are being measured.

Neutron measurements of ^{235}U are performed by inducing fissions with an external neutron source because spontaneous fission rates are orders of magnitude lower than from plutonium isotopes. Typically, AmLi neutron sources, with an average neutron energy of ~ 0.5 MeV, are used in the Active Well Coincidence Counter.

A more sensitive measurement technique that is applicable to large volumes of material containing uranium involves the use of a shuffler. The shuffler measures delayed neutrons following fissions induced by an intense (typically 10^9 neutrons/s) ^{252}Cf source. The sample is placed in a large cavity surrounded by ^3He proportional counters. The ^{252}Cf is moved mechanically (or shuffled) into the cavity, irradiates the sample for several seconds, and is then removed. The proportional counters then count the delayed neutrons for a similar period of time. This process is repeated over and over until adequate counting statistics are obtained.

CALORIMETRY MEASUREMENTS

Heat production is characteristic of all radioactive decays. Plutonium produces a significant amount of heat from this effect (~ 2.5 watts/kg for weapons-grade material). The heat output cannot be masked by radiation shielding, is not affected by the physical or chemical composition of the sample or other materials (matrix), and is therefore used as one of the most accurate NDA methods for plutonium. Traditional calorimeters that have been used to assay plutonium involve a heat bath kept at a constant temperature and two insulated cylinders immersed in the bath. The sample is introduced into one of the cylinders, and the other remains vacant and serves as a reference arm. Both cylinders are wrapped with resistive wire, and each forms two sides of a Wheatstone bridge. The cylinder with the sample heats up and the resistivity increases, debalancing the bridge. The equilibrium deviation is an accurate measure of the temperature, and hence of the sample heat output (power). A typical measurement takes four to eight hours, in order to obtain an accuracy on the order of 0.1%. Uranium-235 has much lower heat production compared to ^{239}Pu , making it much more difficult to measure. Research is being performed to increase the sensitivity (currently about 0.001K) to measure the very small temperature changes that would enable use of calorimeters for assay of ^{235}U in bulk samples.

GAMMA-RAY MEASUREMENTS — ISOTOPIC DISTRIBUTION

Both neutron counting and heat measurement assay techniques require accurate knowledge of the isotopic composition of the sample in order to determine the mass of plutonium or uranium. Neutron coincidence counting measures mainly the ^{240}Pu component in plutonium, and calorimetry is sensitive to small quantities of isotopes other than ^{239}Pu that produce more heat because of their shorter lifetime. The isotopic distribution of actinide materials can be measured nondestructively by careful analysis of the item's gamma-ray spectrum measured with a high-purity germanium detector. The technique quantifies the ratio of all gamma-emitting isotopes in the item to a common isotope. It uses the known intrinsic intensities of the gamma rays from the actinide isotopes to internally self-calibrate each measurement, so that measurements are made without prior knowledge of the measured item and without any calibration of the measurement system. In this fashion, the method can measure items of arbitrary physical and chemical composition, items of arbitrary size and geometry, and items inside containers of unknown size and composition. Typically, measured samples range in size from a few tens of milligrams to many tens of kilograms in measurement times that range from ten minutes to one hour.

FUEL MEASUREMENT TECHNIQUES

There are several methods that can determine the burnup of reactor fuel and its plutonium content. The Fork Detector measures the intensity of gamma rays and neutrons from spent nuclear fuel. The neutron rate determines burnup, and the gamma rate combined with the burnup determines the cooling time. The actual fissile content of the fuel is inferred by using reactor burnup codes. The Research Reactor Fuel Counter (RRFC) is a more recent development and directly measures the fissile content by counting neutron coincidences induced by an AmLi neutron source. It is typically used for materials test reactor fuel because of its high enrichment. Other fuels that have a high-curium content would overwhelm the active interrogation and bias the result. The Passive RRFC can be used for these fuels to determine the total plutonium and curium concentrations. It can also be used for fuels that have a low initial enrichment. Other methods involve interrogation with large external neutron sources, such as a shuffler, that use a ^{252}Cf source, a high-fluence d-t neutron source currently being developed at Los Alamos, or a synchronous programmable neutron generator.

ADVANCED NDA TECHNIQUES

The initial thrust of NDA development for safeguards emphasized measurements of pure materials that were used for weapons building or reactor fuel elements that contained PuO_2 . In recent years, more emphasis is being placed on safeguards issues associated with the disposition of salts and other residues containing plutonium in significant quantities. These residues often contain large quantities of americium isotopes, alpha-particle emitters, that cause a large neutron background (the alpha particle interacts with the light element in the residue producing neutrons). Two techniques that are currently available to measure these residues are Tomographic Gamma Scanning (TGS) and Add-a-Source.

TGS involves the simultaneous measurement of gamma rays from an external source penetrating a barrel, and gamma rays emitted by plutonium in a container. This is analogous to medical imaging the internal organs of patients by medical facilities, albeit with lower resolution. The barrel is rotated, translated, and elevated during the measurements, providing multiple projections of the drum that are reconstructed into a tomographic image. The image is used to correct the assay for the source(s) position and for the attenuation of emitted gamma rays by the materials in the drum.

Add-a-Source is a method used with large volume items in neutron coincidence counters to correct for the effect of different materials in the items. A ^{252}Cf neutron source is positioned at three locations in the counter, adjacent to the drum being sampled. A correction factor is derived from the known source intensity measured without the drum and compared to the measurements with the drum. This correction factor is then applied to the drum assay results obtained from coincidence counting. This method is useful when dealing with large volumes of light-element materials, as long as the background neutron rate is not too high.

IMPLEMENTATION OF NDA TECHNIQUES

The NDA techniques we have developed are used around the world in nuclear materials control and accounting. Neutron and gamma-ray (isotopic) measurements are the fundamental building blocks of IAEA safeguards. These measurements are used to assay ^{239}Pu and ^{235}U in Japanese facilities at Rokkasho and elsewhere; to assay fuel rods in Aqtau, Kazakhstan; to measure the vast quantities of nuclear materials in Russia; to quantify nuclear materials in the U.S. nuclear complex; by EURATOM throughout Europe; and in many other places around the world. Calorimetry is used as the gold standard for NDA measurements in the United States, and is being considered for use elsewhere.

The techniques that we have developed are also used for waste stream assay. The Rocky Flats facility uses our TGS (that was commercialized) and our "SuperHENC (trailer-mounted, super-high-efficiency neutron counter) to determine the plutonium content of barrels or crates of waste being shipped to the Waste Isolation Pilot Plant in Carlsbad, New Mexico. The TGS typically measures 208 L waste drums with an average thickness below 30 g/cm^2 , providing a better than 10% accuracy. Japanese facilities use waste-drum assay systems (incorporating neutron measurements) to determine fissile material content in their waste streams.

In many facilities, the NDA is performed remotely, and the information is combined with other remote monitoring information such as video frames (typically taken infrequently, but speeded up when triggered by a change), door switch signals, external radiation monitors, etc. The information is collected by an elaborate code system (Multi-Instrument Collect), combined with operator declarations, and scanned for anomalies. When the code points to an anomaly, it can be investigated by an inspector, who looks at the detailed record (video, radiation, NDA, and operator declarations) and makes a

determination. This saves the inspector the necessity to be present on site and sort through a vast mass of information from all the different sensors.

SUMMARY

Nondestructive assay is a suite of techniques that has matured and become precise, easily implementable, and remotely usable. These techniques provide elaborate safeguards of nuclear material by providing the necessary information for materials accounting. NDA techniques are ubiquitous, reliable, essentially tamper proof, and simple to use. They make the world a safer place to live in, and they make nuclear energy viable.