BNL-66293

Confirmatory Measurements for Uranium in Nuclear Weapons by High-Resolution Gamma-Ray Spectrometry (HRGS)

J. R. Lemley, L. Forman, W. R. Kane, P. Vanier, Brookhaven National Laboratory
Upton, NY 11973 USA 516/344-2916 FAX: 516/344-7533

W. Kiehl, J. Wright, Pantex Plant
Amarillo, TX 79120 USA 806/466-4292 FAX: 806/477-6411

Abstract

High-resolution gamma-ray spectrometry (HRGS) measurement techniques have been developed to confirm HEU in the presence of plutonium in US nuclear weapons and have now been tested with all types of weapons in the enduring stockpile. A combination of high-resolution spectral features assures that both uranium and plutonium components are present. The most useful spectral features for uranium are the ratio of the 186- and 1001-keV peaks associated with the decay of ^{235}U and ^{238}U; the profiles of the 186-, 1001- and 2614-keV gamma-ray peaks versus position along the axis of a device; and the peak area of the 2614-keV gamma ray from decay of ^{232}U (a contaminant from processing reactor-irradiated uranium in gaseous diffusion cascades). Using design information, quantitative acceptance-rejection criteria can be established.

Battery-powered HRGS equipment meets rigorous safety standards for use with nuclear weapons containing conventional high explosives. Portable HRGS equipment can be operated either in process buildings or in the field bunkers used for storage of nuclear weapons at Pantex. HRGS techniques disrupt normal operations minimally. HRGS provides additional assurance, beyond low-resolution measurements, regarding the presence of all SNM in nuclear weapons being transferred to or from the Department of Energy's (DOE) custody.

Introduction

In May 1995, the DOE Office of Safeguards and Security (OSS, NN-51) clarified policy regarding confirmatory-measurement requirements for items, including weapons, containing both uranium and plutonium. Consistent with requirements in DOE 5633.3B and succeeding Orders, measurements of all special nuclear material (SNM) types (e.g., plutonium and highly enriched uranium (HEU)) are required for items upon receipt and prior to weapon disassembly where the SNM types are in chemically and physically discrete forms. Work by Brookhaven National Laboratory at the Pantex Plant has demonstrated that confirmatory techniques based on HRGS can provide high assurance regarding the presence and distribution of both weapon-grade plutonium and HEU in weapons.

Characteristic Spectral Features of Nuclear Weapons

For the purpose of confirming the presence of SNM, a nuclear weapon can be viewed as an assembly of radioactive materials that emit and absorb gamma rays surrounded by materials that attenuate gamma rays. Uranium and plutonium materials themselves both emit gamma rays and also attenuate gamma rays through self-absorption. Radioactive gamma-absorbing materials may

surround other radioactive materials of interest. The radioactive materials useful in confirmatory-measurements are uranium isotopes, plutonium isotopes, decay products, and impurities. Attenuation is energy dependent and occurs through self-absorption in the source materials themselves and in intervening materials between the source and the detector. The amounts and configurations of the source and absorbing materials account for the spectral features that are useful for confirmatory measurements. Removal, substitution, or altering the configuration of the emitting and absorbing materials would cause detectable changes in the high-resolution spectra.

HRGS measurements on complete weapons were made with collimated high-purity germanium (HPGe) detectors so that spectral and spatial signature elements could be clearly resolved and the data compared with design information. Assurance regarding the presence of HEU can be derived from a combination of the following HRGS features:

- The 186-keV/1001-keV intensity ratio;
- Axial scanning – HRGS measurements at multiple positions along the axis of a weapon using a collimated detector so that spatial profiles of gamma-ray intensities at various energies can be compared;
- The intensity of the 2614-keV gamma ray associated with the decay of ^{232}U;
- Weapon-specific signatures (e.g., spectral features or position dependence of gamma-ray intensities associated with a particular weapon type).

The 186-keV/1001-keV intensity ratio depends on source strength (related to quantity, configuration, and enrichment of uranium) and on the energy-dependent differential attenuation of these two gamma rays in materials intervening between source and detector. Axial scanning is carried out by measuring spectra at various positions along the axis of a weapon, as shown schematically in Figure 1. A confirmatory procedure based on scanning sometimes can be simplified to require measurements at only two positions.

Some spectral signatures provide most effective confirmation when measured at an optimum location. For a particular signature the optimum location can be determined from design information or can be established empirically through systematic measurements on the specific weapon type.

The 2614-keV gamma ray associated with decay of ^{232}U is very penetrating because the attenuation of gamma rays in uranium is near minimum around this energy. ^{232}U (half-life 72 a) is produced when uranium is irradiated in a nuclear reactor; possible production paths have been summarized by Peurrung.[1] If the irradiated uranium is subsequently fed into a uranium enrichment plant, the ^{232}U contamination moves toward the top of the cascade. In a gaseous diffusion enrichment cascade, ^{232}U contamination continues to appear, preferentially in enriched product, even when irradiated uranium is no longer being fed into the plant. Once present in HEU, ^{232}U and its decay products can provide spectral signatures for HEU.

Scanning Along Axis of Device

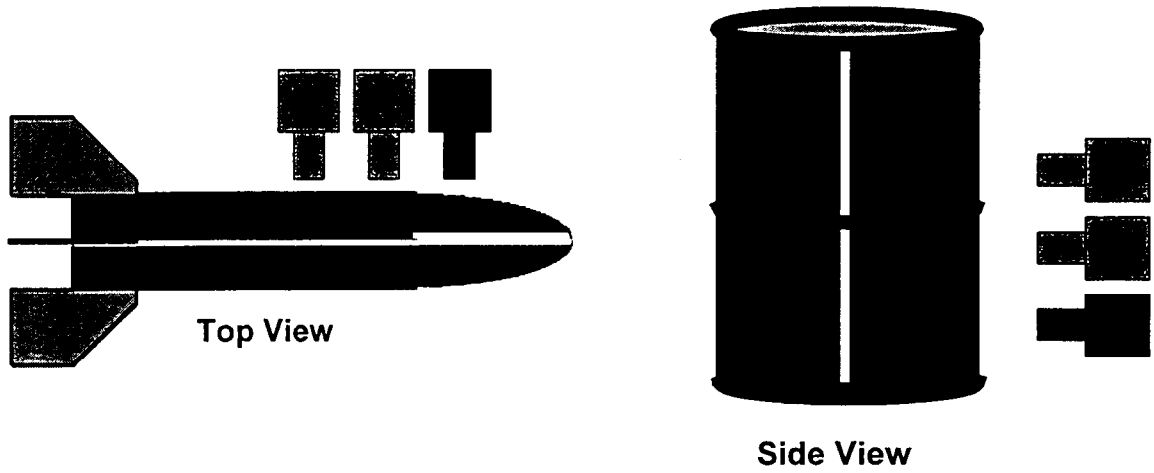
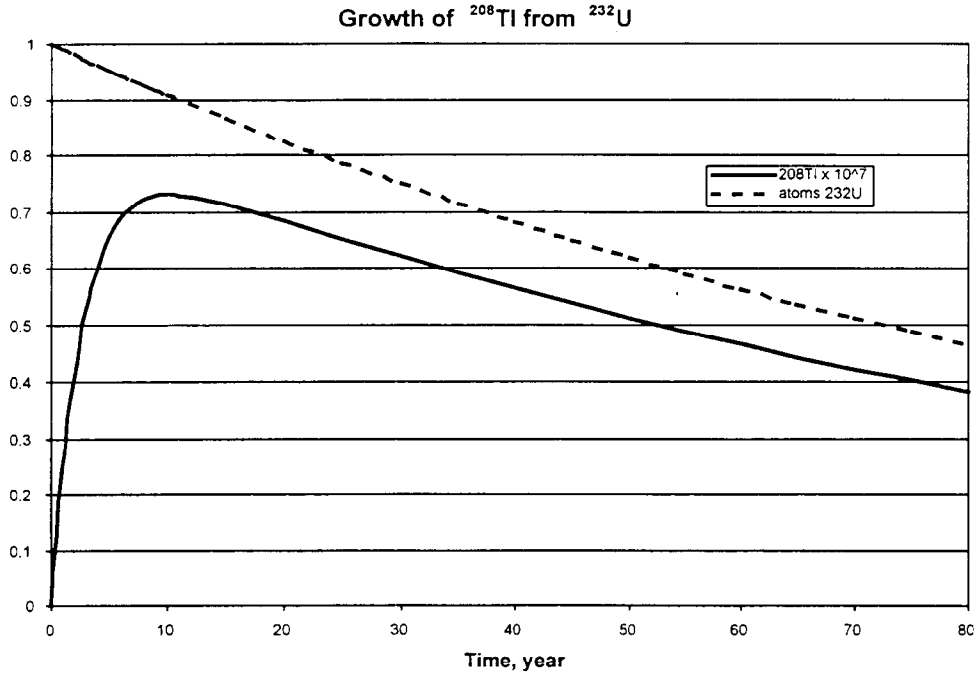


Figure 1. Detector positioned for scanning along the axis of a device. Left, device with axis horizontal; right, device with its axis vertical.

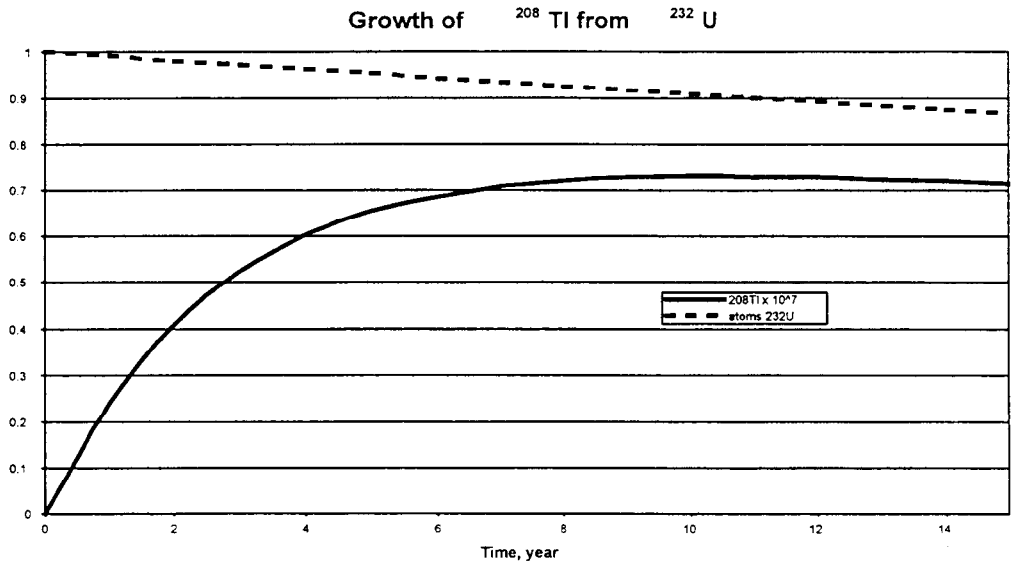
Since ^{232}U is an isotope of uranium, it cannot be removed from HEU in any practical way. However, the decay products of ^{232}U, including ^{208}Tl, whose decay results in the 2614-keV gamma ray, are not present in uranium hexafluoride withdrawn from an enrichment plant. Other processing of uranium for the manufacture of nuclear weapons may alter the concentration of uranium decay products.

^{232}U has a 72-year half-life. ^{208}Tl occurs at the end of the ^{232}U decay chain and beta decays to ^{208}Pb, which is stable. The longest-lived nuclide in the ^{232}U decay chain is ^{228}Th with a 1.9-year half-life; half-lives of the other nuclides in the chain are much shorter. The half-lives of ^{232}U and ^{228}Th account for the change with time of the 2614-keV radiation associated with HEU.

Assuming no ^{232}U decay products are present following some uranium processing operation, such as withdrawal of HEU from an enrichment plant, the concentration of ^{208}Tl relative to the parent ^{232}U changes as shown in Figure 2. The intensity of the 2614-keV radiation is proportional to the number of atoms of ^{208}Tl and reaches a maximum after several times the half-life of the longest-lived nuclide in the chain, ^{228}Th. Figure 2b. shows that the 2614-keV intensity is maximum after about 10 years but reaches 90% of this maximum in about 5 years. Eventually ^{208}Tl reaches secular equilibrium with its parent ^{232}U where the ratio of the number of daughter atoms to parent atoms becomes constant, and the 2614-keV intensity changes with the 72-year half-life of the ^{232}U parent. The 2614-keV intensity remains greater than 50% of its maximum from about 2 years to more than 80 years.



Part a)



Part b)

Figure 2. Growth of ^{208}Tl from decay of ^{232}U. Decay of ^{208}Tl produces the 2614-keV gamma ray.

The naturally occurring radioactive isotope ^{232}Th also results in the 2614-keV gamma. The decay chains of ^{232}U and ^{232}Th converge at ^{228}Th. After ^{228}Th, the decay chains of both nuclides are identical and result in penetrating gammas at 238, 510.7, 583, 727, 860, 1620 and 2614 keV. The

chain ends with beta decay of ^{208}Tl to ^{208}Pb and the production of gammas at 583 and 2614 keV with fixed intensity ratio.

The ways in which the levels of ^{228}Th are populated make it possible to readily distinguish ^{232}U from ^{232}Th. ^{232}U alpha decays directly to ^{228}Th, but only levels less than or equal to 874.6 keV above the ground state of ^{228}Th are populated. In comparison, ^{232}Th first alpha decays to ^{228}Ra which then beta decays to ^{228}Ac. Beta decay of ^{228}Ac to ^{228}Th feeds higher energy levels in ^{228}Th than does alpha decay of ^{232}U, some 32% of the beta decay going to the 968.8-keV level in ^{228}Th. Radiative decay of this level produces gamma rays at 969 keV and 911 keV with relative intensities of 37 and 62% respectively.

Detection of gamma rays at 911 and 969 keV in the correct intensity ratio, along with a peak at 2614-keV, would indicate that the 2614-keV peak was due, at least in part, to decay of ^{232}Th rather than ^{232}U. BNL's HRGS measurements on weapons at Pantex, were carefully checked for peaks at 911 and 969 keV to determine whether any part of an observed 2614-keV peak might be due to natural thorium background. After correction for background, a 2614-keV peak with an axial intensity profile that is consistent with component dimensions and other design information can provide strong assurance regarding the presence of HEU in a weapon.

Quantitative acceptance/rejection criteria based on a combination of spectral features of the kinds listed above have been established for each type of weapon. The quantitative acceptance limits for various tests are derived from and must be consistent with theoretical calculations using design information, systematic trends supported by design information, and statistical variation observed in multiple measurements. The combination of HRGS spectral features provides high assurance regarding the presence of HEU in weapons. HRGS methods can also raise the level of assurance regarding the presence and configuration of weapon-grade plutonium although confirmatory measurements based on low-resolution methods are considered adequate for confirming the presence of plutonium. Of course, other elements of the safeguards programs implemented by DOE and the Department of Defense provide further assurance and defense in depth for the special nuclear materials used in nuclear weapons.

Practicality of HRGS Confirmatory Measurements on Weapons

Although HPGe detectors must be operated at cryogenic temperatures, HRGS equipment is sufficiently portable, can be battery powered, and is sensitive enough that data with satisfactory statistical accuracy can be obtained within acceptable counting times. HRGS measurements are passive and do not depend on active interrogation with penetrating radiation. HRGS equipment satisfies the rigorous safety regulations required for use near nuclear weapons containing chemical high explosive. Because of these characteristics, HRGS measurements can be made in both the manufacturing and the storage areas of an assembly/disassembly plant. HRGS confirmatory-measurement methodology could be implemented with reasonable cost and manageable impact on operations. Technically trained staff can carry out HRGS confirmatory-measurement procedures under the supervision of staff trained in nuclear measurements.

References

1. Anthony J. Peurrung, "Predicting ^{232}U Content in Uranium," Pacific Northwest National Laboratory, Report PNNL-12075, December 1998.

Acknowledgment

This work was performed under the auspices of the U.S. Department of Energy, Contract No. DE-AC02-98CH10886.