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Author(s): J. L. Parker
T. E. Sampson

Submitted to: IAEA Symposium on International Safeguards
Vienna, Austria
March 14-18, 1994
(EXTENDED SYNOPSIS)

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IMPROVED IMPLEMENTATIONS OF TWO GAMMA-RAY MEASUREMENT METHODS*

J. L. Parker and T. E. Sampson

Los Alamos National Laboratory, Safeguards Assay Group N-1, MS E540
Los Alamos, New Mexico 87545 USA

Most of the basic gamma-ray measurement methods useful in international safeguards were discovered long ago, often independently by different investigators in different countries. As detectors and associated electronic equipment have improved and as a better understanding of the measurement physics has evolved, the implementation and usefulness of the methods have also improved. Specific recommendations for areas of developmental effort have been made at an IAEA Advisory Group Meeting.¹ We herein report results of improved and more useful implementations of two measurement methods. Neither of them is entirely new, and neither has seen the use they perhaps deserve. It is hoped that these methods, in improved form, may contribute more significantly to international safeguards.

The first method is that of quantitative assay for plutonium or uranium (or both) in small samples of reasonably homogeneous materials. "Small" for purposes of such gamma-ray assay might be defined as volumes ≤ 15 ml, in cylindrical vials with diameters ≤ 2 cm, and depths ≤ 5 cm. For finding the plutonium content of small samples, current procedures usually use high-resolution gamma-ray spectrometry to find the effective ^{240}Pu fraction followed by neutron coincidence counting to find the total plutonium content of the sample. By optimizing and improving methods developed mostly for larger containers of waste or scrap,² it is possible, in many cases, to use a higher efficiency germanium detector and get a reasonably accurate value of the total plutonium content from the same spectrum used to find the plutonium isotopic distribution. Past experience, and current preliminary results indicate that bias levels of 1% or less are often possible. Precisions of $\sim 1\%$ for 1 gram of ^{239}Pu in 1000 s should be possible; greater masses give better precision and lower masses give worse precision in the same time. Clearly, however, precisions will depend on detector efficiency; sample-to-detector distance; sample size, shape and packaging as well as plutonium mass and counting time. For best accuracy at least one standard is desirable, but if a standard containing plutonium or uranium is not available, a gamma-ray source, such as ^{133}Ba , can be used with little loss of accuracy. For best accuracy the corrections for gamma-ray self-attenuation should be based on a measured transmission through the sample, but for small samples of known and regular geometry gamma-ray ratios can be used to get quite good corrections. Accuracy and precision for ^{235}U measurements are not quite as good as for ^{239}Pu because of the lower energy (185.7 keV compared to ~ 400 keV) upon which the quantitative assay is based.

The second method is that of uranium isotopic distribution measurements by intrinsic self-calibration methods similar to those used for measuring plutonium isotopic distributions. Uranium enrichment measurements by the standard "Enrichment-Meter" methods are very accurate for infinitely thick samples of uranium-bearing materials, but the method fails completely for noninfinitely thick samples of unknown composition and packaging. The first known application of the intrinsic self-calibration idea to uranium measurement goes back to 1976.³ Others have worked on similar or modified schemes since. At Los Alamos National Laboratory, we have improved the analysis of the spectra of arbitrary uranium-bearing materials by using a better relative efficiency model and by adding the ability to use all the available spectral information from ~ 50 keV to ~ 1 MeV (including the x-rays from uranium decay.) Over a wide range of enrichments, the $^{235}\text{U}/^{238}\text{U}$ ratio can be measured with accuracies of 2% or better. For the higher enrichments, the ratio of ^{234}U to the other isotopes is also measurable. In the low-energy uranium K x-ray region, the

*Work supported by US Department of Energy, Office of Arms Control and Non-proliferation and DOE/DOS, Program of Technical Support to IAEA Safeguards.

thorium x-rays emitted in the uranium decay are essentially ^{235}U signatures in almost all uranium isotopic mixtures likely to be encountered. They are used as such in our analysis methods. With the improved implementation, it is still necessary to assume equilibrium of the thorium daughters of ^{235}U and ^{238}U with their parents to obtain accurate measurements. Such is usually the case for shut-down uranium facilities where this scheme should work well for measuring the enrichments of holdup deposits.

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