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Fast Enrichment Screening for Safeguards Applications

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Abstract. Methods for rapid non-destructive uranium enrichment classification of large containers are of importance to safeguards and counter-terrorism agencies. There is often a need to quickly categorize and segregate suspect containers as 'depleted' or 'enriched' on a 'Go/No Go' basis. Recent improvements in gamma spectroscopy technologies have provided the capability to perform rapid field analysis using portable and hand-held devices such as battery-operated medium and high resolution detectors (including lanthanum halide and high purity germanium). Furthermore a new generation of portal monitors are currently under development with advanced spectroscopic capabilities. Instruments and technologies that were previously the domain of complex lab systems are now widely available as touch-screen "off-the-shelf" units. Despite such advances, the task of enrichment analysis remains a complex exercise. This is particularly so when surveying large items such as drums and crates containing debris of unknown density and composition contaminated with uranium. The challenge is equally applicable to safeguards inspectors evaluating large items and for interdiction of illicit special nuclear materials in mass transport e.g. shipping containers at ports and borders. The variable shielding, container size, lack of matrix knowledge, wall thickness and self-shielding compound this problem. Performing an accurate assessment within the short count time window demanded of the field operative, leads to the need for a reliable method that can adapt to such conditions and is robust to a wide dynamic range of counting statistics. Several methods are evaluated with reference to the performance metrics defined in applicable standards (such as ANSI N42.43-2006). The primary issue is to minimize the bias that can result from attenuation effects, particularly as the gamma emissions from U235 are low energy and therefore highly susceptible to absorption in large containers with metal scrap. Use of other radiometric signatures such as bremsstrahlung radiation and neutron emissions are considered in addition to photopeak ratio analysis. Benchmark comparisons are performed against well-established enrichment analysis methods such as PC-FRAM, MGAU, and the Enrichment Meter. The suitability/limitations of the more traditional methods are discussed.

1. Introduction

A need exists for non-destructive assay equipment and techniques that can perform rapid uranium enrichment classification. In particular the ability to perform real-time 'Go/No Go' segregation of large objects and containers as 'depleted' or 'enriched' provides a major transformational impact for safeguards inspectors. Materials being released from nuclear facilities must be categorized for domestic and international materials control and accountability. At ports and borders, the same technology can be used to screen large containers suspected to contain illicit shipments of enriched uranium with a sufficiently high throughput and accuracy to minimize impact to flow of commerce.

2. Requirements

The specific requirements include:

- Capability to perform non-destructive/non-intrusive scanning of a whole container at a time. Container sizes will vary from small suitcase size packages through to full size intermodal shipping containers.

- One of the greatest technical challenges involves compensating for the presence of matrix materials inside the container including mixtures of organics and metals with varying density.
- Survey times must be configurable to the specific application. For example in a high throughput customs/border application the survey must complete in less than five minutes. In the safeguards arena more time is available but generally in order to be effective operationally, the survey must be completed in one hour or less for the entire container in order to achieve a reasonable throughput. The performance goal is very much driven by the need to minimize false alarms (i.e. mis-categorization).
- Reliability and ruggedness of the equipment must also be taken into account when designing a system for the end user. Hard working long life components should be sought. Electronics should feature minimal gain drift or automatically compensate for this. Tough field conditions must also be taken into account, operating in four seasons in extreme real world environments from the desert to the arctic.
- Ease of operation and maintenance is another factor. Self- or auto-calibration should be included in the design and where possible off-the shelf components used for detectors and electronics. Ready access for spare parts and servicing is critical for the long term viability of such a system. The system software and user interface must be designed for rapid operation in the field.

3. Traditional Methods for Enrichment Measurement

There are three widely used methods for determining enrichment based on gamma spectroscopy:

3.1. Infinite Thickness Method

The Infinite Thickness Method (or Enrichment Meter Technique) uses either two or three region-of-interests to measure the 185.7 keV peak and its associated background area based on a calibration using reference samples under a well-defined geometry. The method is best suited for bulk samples which easily meet the infinite thickness requirement and is applicable to items containing depleted uranium (< 0.72% U235 fraction) up to highly enriched uranium (HEU) with greater than 90% U235 enrichment. In general, three conditions must be met:

- The sample must have a uniform distribution: both isotopically and by matrix distribution.
- The daughter activities must be in secular equilibrium with the parent uranium activities.
- The material must be infinitely thick with respect to the 185.7 keV gamma peak.

The method becomes less accurate when non-representative references are available or the material type, packaging and gamma ray background is varying from sample to sample. Also, a large gamma ray background can reduce the accuracy of the results.

The enrichment meter relies upon the sample container wall being well known. A correction factor is required if the wall thickness is variable (usually achieved by an ultrasonic measurement).

3.2. Peak Ratio or Intrinsic Calibration

Measurements based on the intrinsic calibration method avoid the need for calibration with physical standards. Isotopic ratios are determined from measured gamma spectrum using corresponding gamma and x-rays from the decay of all isotopes, taking into account physical phenomena such as the energy dependence of detector efficiency, self-absorption in the sample and attenuation in the container and filters. For uranium spectra, the x-ray analysis method [1] uses analysis of the XKα region (89-99 keV) where fairly abundant but strongly overlapping gamma and x-ray signatures from the U235 and U238 daughter nuclides Th231 and Th234 occur. This approach requires secular

equilibrium between U238 and its daughter nuclides, which is reached about 80 days after chemical separation: the method is, therefore, not suited to freshly separated uranium materials.

The x-ray region is highly susceptible to interference from other x-rays that may be present in the sample (such as elevated Th, Np237, and Ac227). For this reason, the intrinsic calibration method has also been further developed to include a full-spectrum analysis mode that analyzes peaks across the full energy of the spectrum [2]. This technique is used only with high-resolution (HPGe) systems. For wall thicknesses of greater than 10 mm of steel, the low energy (89-100 keV) region is heavily attenuated, so the higher energy region (121 – 1001 keV) is favoured. With this method it should be noted that it is not necessary to know the cylinder wall thickness.

Whereas MGAU exclusively uses the low energy region, PC-FRAM is supplied with several 'parameter files' that allow an user to select an appropriate analysis route. Furthermore, non-equilibrium conditions between U238 and Pa234m can be corrected for with PC-FRAM. However, in all situations, the user must pay careful attention to the possibility of interfering lines and non-physical response functions that can result from energy calibration drift, poor counting statistics or other sources of bias. In particular the presence of elevated levels of thorium can cause problems for the PC-FRAM analysis in the 121 – 1001 keV region.

Other commercial software packages such as ISOTOPIC [3] software follow similar principles to the PC-FRAM method using peaks across the full energy region of the spectrum.

3.3. Peak Fitting Method

An improvement to the traditional enrichment-meter is made by fitting computed response profiles to the observed data of NaI spectra in the 130 to 290 keV region. A computer analysis code called NaIGEM [4] (NaI Gamma Enrichment Measurements) is used to perform automatic correction for changes in gain and detector resolution and account for Compton continuum and interference peaks. Additionally, the calibration of the system requires only a single reference sample.

Designed specifically for the IAEA verification of UF_6 cylinders, a fully integrated electrically cooled portable HPGe system is available from AMETEK Ortec known as the Portable UF_6 Cylinder Verification System [5]. This package includes a tungsten collimator and the system is operated on the device using a touch screen LCD display with on board software that is an adapted version of the IAEA NaIGEM enrichment meter. The 50mm diameter x 30 mm depth HPGe detector is cooled with a Stirling Cooler that can operate on battery or AC power.

4. PERFORMANCE SPECIFICATION

In order to define specific performance requirements for rapid enrichment screening, it is useful to examine relevant existing standards relevant to safeguards.

The US Department of Homeland Security Domestic Nuclear Detection Office (DNDO) has adopted the ANSI/IEEE American Standards Committee on Radiation Instrumentation N42 standards against which to test and evaluate radiation detection instruments. The two most relevant standards relating to technical performance criteria are: ANSI N42.34-2006 and ANSI N42.43-2006.

American National Standard Performance Criteria for Mobile and Transportable Radiation Monitors Used for Homeland Security (ANSI N42.43-2006) specifies the operational and performance requirements for transportable and/or mobile radiation monitors used in homeland security. Performance requirements and tests are given in relation to radionuclide identification. The standard broadly covers devices that do not have permanent mounting platforms including those mounted to vehicles, trailers, watercraft and cranes and those used while being carried by a person such as a backpack.

American National Standard Performance Criteria for Hand-Held Instruments for the Detection and Identification of Radionuclides (ANSI N42.34-2006) defines U235 and U238 as nuclides of interest but does not call out the need for the instrument to be able to determine enrichment of any uranium identified.

ISO 22188:2004 specifies methods and means of monitoring for inadvertent movement and illicit trafficking of radioactive material. It provides guidelines on the use of both stationary and portable (e.g. hand-held) instruments to monitor for radiation signatures from radioactive material. Emphasis is placed on the operational aspects, i.e. requirements derived for monitoring of traffic and commodities mainly at border-crossing facilities including maritime ports, airports, and similar locations where goods or individuals are being checked. This standard does not address the issue of detection of radioactive materials at recycling facilities, although it is recognized that trans-boundary movement of metals for recycling occurs, and that monitoring of scrap metals may be done at the borders of a state.

The IAEA has developed a series of guidelines and prescriptive specifications relating to performance of instrumentation used for detecting radioactive materials crossing international borders [6] and an extensive study has been performed on existing equipment and techniques [7]. These IAEA standards are broadly in line with the ANSI N42 requirements.

Another relevant international project is the Illicit Trafficking Radiation Assessment Program (ITRAP). This falls under the organization of the European Union in collaboration of the IAEA, World Customs Organization (WCO), INTERPOL and the US DNDO. ITRAP provides funding and laboratories for collaborative studies and testing of equipment for border monitoring. The aim of ITRAP is to develop consensus standards, establishing technical requirements and address practical issues that relate to border radiation detection equipment [8].

5. FAST SCREENING TECHNIQUES

The quickest and simplest means of discriminating HEU from DU is comparison of the strongest photopeaks i.e. 185.7 keV for U235 and the 1001 keV Pa234m daughter of U238 (assuming the U238 is in secular equilibrium with its daughters). This comparison can be a useful indicator: a strong 185.7 keV line without 1001 keV line present will be a good indication that the material is highly enriched. Conversely with depleted uranium it is very likely that only the Pa234m daughter peaks will be visible with the U235 low energy peaks dropping below the Compton continuum as combined result of their weak emission on strong absorption in surrounding shielding materials.

However, caution should be applied when attempting to get a meaningful measure of enrichment by a ratio of the two lines. The significant difference in attenuation can lead to large errors in enrichment unless the shielding, matrix and source materials are well characterized. Therefore a more suitable photopeak for enrichment determination is the 258 keV (Pa234m) peak. This peak is emitted at one tenth of the production rate of the profligate 1001 keV line so this requires a more substantial quantity of uranium in order to be detected under the reference scenarios. Under the reference performance evaluation conditions defined above, this line is detectable for 200g HEU in 600 seconds. For a 60 second count, 750g HEU is required. With depleted, natural, and low enriched (5-10%) uranium, this peak is detectable with their respective reference quantities of uranium in 60 seconds.

For the 258 keV peak, the weak branching ratio is not the only issue. This low energy line will not penetrate more than 3 cm of steel. However, it should be noted that for a large ISO shipping container (heterogeneously filled with a distributed steel matrix), the “visible” outer 3 cm would represent around 3,000 - 4,000 kg of steel. In other words, this is the portion of the box from which the emitted 258 keV photons are detected. Assuming such a container has a net weight in the range 10,000 – 20,000 kg, a gamma based enrichment survey would cover 15-40% of the box contents. Furthermore if we make an argument that the source material is most likely in the form of multiple sources randomly located rather than localized in a single “worst case” point, there is only a small probability that all the uranium present would escape the surveyed region.

It is therefore possible to develop a simple threshold criterion for various survey scenarios by comparison of the 258 keV peak with U235 peaks. The strong peak at 205 keV is most suitable as it lies closest in energy. A simple peak area comparison (205/258 keV) can be developed as follows:

- Mock up the survey scenario with various enrichment sources (e.g. from DU to 10% LEU enrichment and HEU if available).
- Take measurements on these sources with an HPGe detector at various positions with a suitable collimator.
- Repeat the surveys with varying thicknesses of steel plate in front of the detector to simulate shielding (1mm, 3mm, 5mm).
- Analyze the data with a suitable enrichment analysis software e.g. PC-FRAM. Note that the uranium parameter files bundled with PC-FRAM v4.3 and later take care of most of the known issues (interference from thorium etc.) over the range 121 keV to 1001 keV.
- For a representative sample of survey scenarios at different positions and compare the ratio of net peak areas at 205 keV and 258 keV to the known enrichment and correlate to FRAM's measured enrichment.
- For the DU, Nat U, LEU and HEU boundaries:
    - Determine spectral quality indicators that allow correction for shielding and the presence of interfering photopeaks (e.g. thorium).
    - Determine the threshold peak ratios that define each boundary.

Another means of discrimination of HEU from DU is by evaluation of the bremsstrahlung radiation spectrum. The bremsstrahlung spectrum above 100 keV is produced from the high energy beta particles emitted from Pa234m decay. This produces a gamma-ray continuum with a mean energy of 400 keV [9]. This continuum lies underneath the gammas usually used for uranium identification. Such radiation is useful for identifying HEU, particularly in shielded scenarios. HEU normally has very weak photon production in the 1001 keV region, but the bremsstrahlung radiation from Pa234m decay (U238 daughter) produces a strong signal with sufficient energy to penetrate the shielding. By measuring a region of the bremsstrahlung spectrum (e.g. 400 - 600keV) and comparing to the major photopeaks at 185.7-, 258- and 1001 keV it is possible to classify the uranium enrichment and make an evaluation of the nature of the shielding involved.

Finally some consideration should be given to the sample's neutron emission. U235 does not produce spontaneous fission neutrons and U238 produces only a very weak signal from this mechanism. However neutron emission by (alpha, n) does occur in uranium compounds and is often easily detected. U234 is the dominant source of this emission and the U234 fraction follows strongly the U235 enrichment - in fact there is little variation in U234/U235 ratio for DU, NatU and LEU. Of course in the case of pure metal, the neutron emission is negligible regardless of enrichment, but for uranium oxide and uranium hexafluoride the neutron emissivity will be a strong indicator of enrichment. A simple measure of enrichment can therefore be provided by comparing the total neutron count rate with the 1001 keV photon. This method is potentially one of the least susceptible to bias due to attenuation effects as both the emitted fast neutrons and high energy photons can penetrate through thick layers of metal debris. Care should be taken to verify that no other source of neutron emission could be present in the sample (for example Pu240, Cf252 and Cm244).

6. CONCLUSIONS

The measurement of uranium enrichment can be a relatively straight-forward approach as long as the measurement is made in a clean environment under well-defined conditions and parameters. Unfortunately, real-world applications do not meet these criteria. Whether measuring waste or cargo

the measurement is complicated by high backgrounds, interfering signals (NORM, medical isotopes, etc), unknown shielding characteristics, and stand-off distance.

While the 1001 keV peak of U238 can be detected in most real-world applications, further information as to the enrichment of the uranium is problematic since the presence of the 185.7 keV peak of U235 is susceptible to high backgrounds, shielding, and stand-off. Other enrichment techniques using x-rays or low-energy gamma peak ratios are also hampered. For measurements of waste containers or inspections of UF_6 cylinders and facilities, the problems encountered with possible interferences is overcome with longer counting times and some knowledge as to the attenuating characteristics of the item. However, in security environments (ports of entry) the need is for quick analysis with very limited information on the item being measured.

With a combination of techniques using improved handheld technology, it is possible to rapidly screen enrichment in order to classify containers as containing 'depleted' or 'enriched' uranium on a 'Go/No Go' basis. The safeguards inspector should understand both the capabilities and the limitations of this method and be aware of the possibility of sources of bias and interference including matrix attenuation, self shielding and the effects of masking radiation.

REFERENCES

1. A.N. Berlizov and V. V. Tryshyn, "Study of the MGAU Applicability to Accurate Isotopic Characterization of Uranium Samples," IAEA-SM-367/14/05/P.
2. D. T. Vo and T. Sampson, "Uranium Isotopic Analysis with the FRAM Isotopic Analysis Code," LA-13580, May 1999.
3. R.C. Hagenauer et al, "The Evaluation of Program Isotopic to Nondestructively Quantify Uranium and Plutonium Samples at the JRC in Ispra", ESARDA, May 2003.
4. R. Gunnink, "A Guide for Using NaIGEM, PC Version 1.51b for DOS and Windows," December 2002.
5. Ametek Ortec, Portable UF_6 Cylinder Verification System, MICRO-UF6-PKG-1.
6. IAEA-TECDOC-1312, Detection of radioactive materials at borders, September 2002 and K.E. Duftschmid, IAEA Technical/Functional Specifications for Border Radiation Monitoring Equipment).
7. IAEA-TECDOC-1596-CD, Improvement of Technical Measures to Detect and Respond to Illicit Trafficking of Nuclear and Radioactive Materials, Results of a Coordinated Research Project 2003–2006, July 2008.
8. P. Beck, ITRAP, "Illicit Trafficking Radiation Detection Assessment Program," Final Report, February, 2001.
9. P. Ragan, S. Abousahl, Illicit Trafficking: Monte Carlo Modelling of Shielded Uranium Source Gamma Spectra from the NaI Detector, Institute for Transuranium Elements, Joint Research Centre, European Commission, Karlsruhe, Germany.