# Review of Uranium Enrichment Measurement for Safeguards Applications

## Sources of Special Note

Berezin, et al, present a method for using a tunable laser diode spectroscope to measure enrichment in situ:

> *The objective of this work was investigation of possibility of tunable diode laser spectroscopy (TDLS) technique application for gaseous uranium hexafluoride ($UF_6$) isotope measurement. Spectra of uranium hexafluoride gas mixture were investigated using two different Fourier Transform Spectrometers Vector 22 and Bruker 66v. Observed spectral features were identified and model spectra of different gas mixture components were developed. Optimal spectral range for measurements was determined near maximum of $UF_6$ combination band $\nu_1 + \nu_3$. Laboratory prototype of multi-channel instrument under consideration based on tunable diode lasers was built and algorithms were developed to measure gaseous $UF_6$ isotopic ratios. Diode laser used operated at the wavelengths near $\lambda = 7.68$ μm. It was placed in a liquid nitrogen cooled cryostat. Three instrument channels were used for laser frequency calibration and spectra recording. Instrument was tested in measurements of real $UF_6$ gas mixtures. Measurement accuracy was analyzed and error sources were identified. The root-mean-square random error in the $^{235}U$ isotopic content is characterized by a spread of about 0.27% for quick measurements (at times less than 1 min) and 1% for periods of more than an hour. It was estimated that the measurement accuracy could be improved by at least an order of magnitude by minimizing the error sources.[1]*

Miller gives a relatively recent (2012) summary of concepts for non-destructive cylinder assay techniques:

> *Nondestructive assay (NDA) measurements of uranium cylinders play an important role in helping the International Atomic Energy Agency (IAEA) safeguard uranium enrichment plants. Traditionally, these measurements have consisted of a scale or load cell to determine the mass of $UF_6$ in the cylinder combined with a gamma-ray measurement of the 186 keV peak from $^{235}U$ to determine enrichment. More recently, Los Alamos National Laboratory (LANL) and Pacific Northwest National Laboratory (PNNL) have developed systems that exploit the passive neutron signal from $UF_6$ to determine uranium mass and/or enrichment. These include the Uranium Cylinder Assay System (UCAS), the Passive Neutron Enrichment Meter (PNEM), and the Hybrid Enrichment Verification Array (HEVA). The purpose of this report is to provide the IAEA with new ideas on technologies that may or may not be*

*under active development but could be useful for $UF_6$ cylinder assay. To begin, we have included two feasibility studies of active interrogation techniques. There is a long history of active interrogation in the field of nuclear safeguards, especially for uranium assay. Both of the active techniques provide a direct measure of $^{235}U$ content. The first is an active neutron method based on the existing PNEM design that uses a correlated $^{252}Cf$ interrogation source. This technique shows great promise for $UF_6$ cylinder assay and is based on advanced technology that could be implemented in the field in the near term. The second active technique is nuclear resonance fluorescence (NRF). In the NRF technique, a bremsstrahlung photon beam could be used to illuminate the cylinder, and high-resolution gamma-ray detectors would detect the characteristic de-excitation photons. The results of the feasibility study show that under certain measurement geometries, NRF is impractical for $UF_6$ cylinder assay, but the 'grazing transmission' and 'secant transmission' geometries have more potential for this application and should be assessed quantitatively. The next set of techniques leverage scintillator detectors that are sensitive to both neutron and gamma radiation. The first is the BC-523A capture-gated organic liquid scintillator. The detector response from several different neutron energies has been characterized and is included in the study. The BC-523A has not yet been tested with $UF_6$ cylinders, but the application appears to be well suited for this technology. The second detector type is a relatively new inorganic scintillator called CLYC. CLYC provides a complementary detection approach to the HEVA and PNEM systems that could be used to determine uranium enrichment in $UF_6$ cylinders. In this section, the conceptual idea for an integrated CLYC-HEVA/PNEM system is explored that could yield more precision and robustness against systemic uncertainties than any one of the systems by itself. This is followed by a feasibility study on using alpha-particle-induced reaction gamma-rays as a way to estimate $^{234}U$ abundance in $UF_6$. Until now, there has been no readily available estimate of the strength of these reaction gamma-rays. Thick target yields of the chief reaction gammas are computed and show that they are too weak for practical safeguards applications. In special circumstances where long count times are permissible, the 1,275 keV $F(\alpha,x\gamma)$ is observable. Its strength could help verify an operator declaration provided other knowledge is available (especially the age). The other $F(\alpha,x\gamma)$ lines are concealed by the dominant uranium line spectrum and associated continuum. Finally, the last section provides several ideas for electromagnetic and acoustic nondestructive evaluation (NDE) techniques. These can be used to measure cylinder wall thickness, which is a source of systematic uncertainty for gamma-ray-based NDA techniques; characterize the $UF_6$ filling profile inside the cylinder, which is a source of systematic uncertainty for neutron-based NDA techniques; locate hidden objects inside the cylinder; and provide a unique identification of cylinders. Acoustic and electromagnetic NDE techniques are complementary to NDA measurements, and may improve the accuracy and continuity of*

> *knowledge of $UF_6$ measurements of interest to the IAEA. As concepts and approaches for enrichment plant safeguards continue to evolve to meet modern challenges, the conceptual ideas explored in this report, along with more traditional techniques, help define the toolkit of technologies available for $UF_6$ cylinder assay. Whether the application is an unattended cylinder verification station or an on-site inspection, the basic building blocks can be tailored to provide the best solution given competing constraints such as size and weight limitations, required precision, mechanical complexity, cost, stability, robustness, etc.*[2]

Smith and Lebrun present a design for an "online enrichment monitor" to passively and continuously measure the output of a centrifuge set:

> *The International Atomic Energy Agency is developing technology for the continuous monitoring of in process UF6 gas at centrifuge enrichment plants. These unattended detection nodes would be located on the high pressure portion of the plant's header pipes, where the gas flow from multiple cascades is consolidated. The time-dependent relative enrichment data from the online monitor will be combined with subsequent mass measurements of the corresponding feed, product and tail cylinders to produce a mass measurement of 235U in each cylinder. The On-line Enrichment Monitor (OLEM) will utilize NaI(TI) spectrometers, gas pressure gauges and possibly temperature sensors. This paper describes the conceptual design of the OLEM and the Monte Carlo modeling that has been performed to support the development process. Simulated OLEM responses are used to explore potential instrument design options (e.g. collimator configuration) and candidate data analysis methods, and to gain insight into the nature and magnitude of the various uncertainties that arise in online enrichment monitoring. Prominent among these uncertainties is the correction for the 235U signal coming from deposits on the walls of the piping; potential correction methods for this wall-deposit contribution are described. An OLEM case-study scenario is defined and the expected statistical uncertainties over a representative range of plant operating conditions (e.g. pressure, enrichment and wall deposit thickness) are calculated.*[3]

More recently, March-Leuba, et al. have performed tests on such a system:

> *As global uranium enrichment capacity under international safeguards expands, the International Atomic Energy Agency (IAEA) is challenged to develop effective safeguards approaches at gaseous centrifuge enrichment plants while working within budgetary constraints. The "Model Safeguards Approach for Gas Centrifuge Enrichment Plants" (GCEPs) developed by the IAEA Division of Concepts and Planning in June 2006, defines the three primary Safeguards objectives to be the timely detection of: 1) diversion of significant quantities of natural (NU), depleted (DU) or low-enriched uranium (LEU) from*

> *declared plant flow, 2) facility misuse to produce undeclared LEU product from undeclared feed, and 3) facility misuse to produce enrichments higher than the declared maximum, in particular, highly enriched uranium (HEU). The ability to continuously and independently (i.e. with a minimum of information from the facility operator) monitor not only the uranium mass balance but also the $^{235}U$ mass balance in the facility could help support all three verification objectives described above. Two key capabilities required to achieve an independent and accurate material balance are 1) continuous, unattended monitoring of in-process $UF_6$ and 2) monitoring of cylinders entering and leaving the facility. The continuous monitoring of in-process $UF_6$ would rely on a combination of load-cell monitoring of the cylinders at the feed and withdrawal stations, online monitoring of gas enrichment, and a high-accuracy net weight measurement of the cylinder contents. The Online Enrichment Monitor (OLEM) is the instrument that would continuously measure the time-dependent relative uranium enrichment, E(t), in weight percent $^{235}U$, of the gas filling or being withdrawn from the cylinders. The OLEM design concept combines gamma-ray spectrometry using a collimated NaI(Tl) detector with gas pressure and temperature data to calculate the enrichment of the $UF_6$ gas within the unit header pipe as a function of time. The OLEM components have been tested on ORNL $UF_6$ flow loop. Data were collected at five different enrichment levels (0.71%, 2.97%, 4.62%, 6.0%, and 93.7%) at several pressure conditions. The test data were collected in the standard OLEM N.4242 file format for each of the conditions with a 10-minute sampling period and then averaged over the span of constant pressures. Analysis of the collected data has provided enrichment constants that can be used for 1.5" stainless steel schedule 40 pipe measurement sites. The enrichment constant is consistent among all the wide range of enrichment levels and pressures used.* [4]

Close, et al. offer another cylinder measurement technique which is not sensitive to deposition on the walls of the container:

> *The pressure of uranium in gaseous UF6 inside a cylinder can be measured using X-ray fluorescence. A highly collimated source, highly collimated detector, and a very rigid geometry are required. This measurement technique is independent of the deposit that forms on the inside wall of the cylinder due to chemical reactions of UF6 gas with impurities. The technique is applicable for gaseous UF6 at low pressures where other techniques fail. The detectability limit, assuming a 25 mCi 57Co fluorescing source, is less than 1 Torr gas pressure in a 30 min count time. [5]*

Ianakiev, et al. present an active interrogation method based on the attenuation of two gamma lines in $^{235}U$:

*In this paper, we report our progress toward the development of an advanced enrichment monitoring technology for safeguarding gas centrifuge enrichment plants. We compare the $UF_6$ gas pipe attenuation and sensitivity to X-ray tube HV variations for two transmission energies: 22 keV and 25.5 keV. The first experimental enrichment results taken with a static $UF_6$ gaseous source and X-ray tube based transmission source over a wide gas pressure range are presented.[6]*

## Non-Journal Sources

Many of these sources are reports [2, 4, 7, 8] or conference proceedings [3, 9-17] from various national laboratories rather than journal publications. Alam, et al, present a novel mechanism for forensics applications[7]. The remaining three reports present either broad overviews of categories or reports on the status of testing systems for operational deployment. A conference presentation by Lewicki, et al [15], was included due to its use of spectroscopy to determine enrichment fraction (similar to my research project). Similarly, a report and a conference proceeding on an Online Enrichment Monitor (OLEM) were included as they, like my research project, report on in-situ measurements during the enrichment process[3, 4].

## Full Reference List

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