# A new instrument for rapid $UF_6$ enrichment level determinations in the field

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## Abstract

We have developed a new portable instrument for the determination of $UF_6$ levels of enrichment in the field for nuclear safeguards applications at gas centrifuge enrichment plants (GCEP). The method employs a Single-Use Destructive Assay (SUDA) sampler for collecting $UF_6$ gas, which is absorbed as uranyl fluoride in a solid zeolite matrix, and then directly analyzed for isotope ratios. For this purpose, a small amount of the matrix is heated in a high-temperature micro-crucible, which produces an atomic beam of uranium. This atomic beam is interrogated by a narrow linewidth diode laser and the isotopic absorption spectrum is measured. Our instrument then determines the enrichment level by comparing the relative contributions of $^{235}U$ and $^{238}U$ to the absorption signal. The analysis takes less than 10 min and there is no need for sample preparation or chemical separations. This technology is promising for bias defect enrichment determination of uranium samples collected at GCEPs.

## Introduction

Currently, mass spectrometry remains the gold standard for high-precision, high-sensitivity analysis of nuclear material samples from GCEPs. However, equipment size, complexity, and requirements for sample preparation (dissolution, separation) and utility (voltage/power, high vacuum, compressed gases, strong acids) make mass spectrometry unsuitable for deployment in the field, which is highly desirable for nuclear safeguards applications. Despite efforts to miniaturize instrumentation, a truly portable mass spectrometer with appropriate actinide mass range, high sensitivity, simple operation, and reasonable use requirements remains unrealized. A portable instrument capable of conducting isotopic analyses of uranium and, in some cases, plutonium is needed, especially when a quick analysis turnaround of bulk samples is needed. We have designed, built, and deployed a Fieldable Atomic Beam Isotopic Analyzer (FABIA) to overcome the issues of current analytic technology to meet these needs (Fig. 1). FABIA provides the isotopic determination of uranium, plutonium, and other actinides in collected samples with high sensitivity, resolution, and speed [1–4]. The analytical method is based on conducting laser absorption spectroscopy through an atomic beam of the sample under analysis. The absorption spectrum of the sample reveals its isotopic composition. This instrument analyzes uranium samples in the field in less than 10 minutes. There is no need for sample preparation, no chemical waste is generated, and the support infrastructure for the instrument is minimal, requiring only 800W of electrical power. Unlike mass spectrometry, FABIA's analytical technique is optical in nature and, thus, immune to isobaric interferences—that is, interferences from isotopes of different elements that have the same mass, such as $^{238}$U and $^{238}$Pu. Because the FABIA instrument weighs fewer than 20 kilograms and measures 30 × 30 × 35 cm, it can easily be sited at plants for ease of analysis. Specifically for Safeguards applications at GCEPs, the method employs a Single-Use Destructive Assay (SUDA) sampler for collecting $UF_6$ gas [5,6], which is absorbed as uranyl fluoride in a solid zeolite matrix, and then directly analyzed spectroscopically for bias defect enrichment determination of uranium by our instrument.

*Figure 1. Fieldable Atomic Beam Isotopic Analyzer (FABIA).*

## Experimental

The method consists of generating an atomic beam of uranium and measuring the absorption of light from an intersecting laser beam. The atomic beam is generated by a resistively heated tantalum foil micro-crucible, capable of reaching temperatures as high as 2600°C with minimal electrical power requirements, and without the need for active cooling. A few milligrams of $UF_6$ gas is collected by the SUDA sampler, which consists of a 10 × 3 mm cylindrical aluminum foam, loaded with zeolite (Fig. 2). The SUDA sampler collects $UF_6$ gas in the zeolite matrix, which hydrolyzes the gas into uranyl fluoride ($UO_2F_2$). The aluminum foam is then loaded into the instrument for analysis.

*Figure 2. SUDA sampler consisting of a cylindrical aluminum foam loaded with zeolite and mounted on a standard KF-16 fitting.*

Fig. 3 depicts the experimental setup. The SUDA sampler is placed at the bottom of the micro-crucible. The crucible is resistively heated inside a vacuum chamber to 2000 ºC. The chemical decomposition of uranyl fluoride at high temperatures is very well understood [7]. In addition to sublimation, the following chemical reactions occur when solid uranyl fluoride is heated:

$$^{5}/_{2}UO_2F_2(s) \rightarrow UF_5(g) + {}^{1}/_{2}O_2(g) + {}^{1}/_{2}U_3O_8(s)$$

From this equation, we observe that 60% of the uranium atoms in the uranyl sample are contained in the oxide. As part of our efforts to develop a fieldable system for the determination of uranium enrichment by laser absorption spectroscopy, we have developed chemical reduction methods for the production of neutral uranium atomic beams from oxides using lutetium as reducing agent [3]. Thus, after the initial decomposition of uranyl fluoride into the oxide, atomic beams of uranium can be generated using lutetium according to the following reaction:

$$^{3}/_{8}U_3O_8(s) + 2Lu(s) \rightarrow Lu_2O_3(s) + {}^{9}/_{8}U(g)$$

The uranium vapor escaping the cylindrical crucible forms a collimated atomic beam, which is intersected at right angles by the beam from a tunable external-cavity diode laser. The laser wavelength is centered at the 861.031 nm transition of uranium. A silicon photodiode detects the transmitted light absorbed by the atomic beam, showing absorption bands corresponding to each isotope type present in the sample.

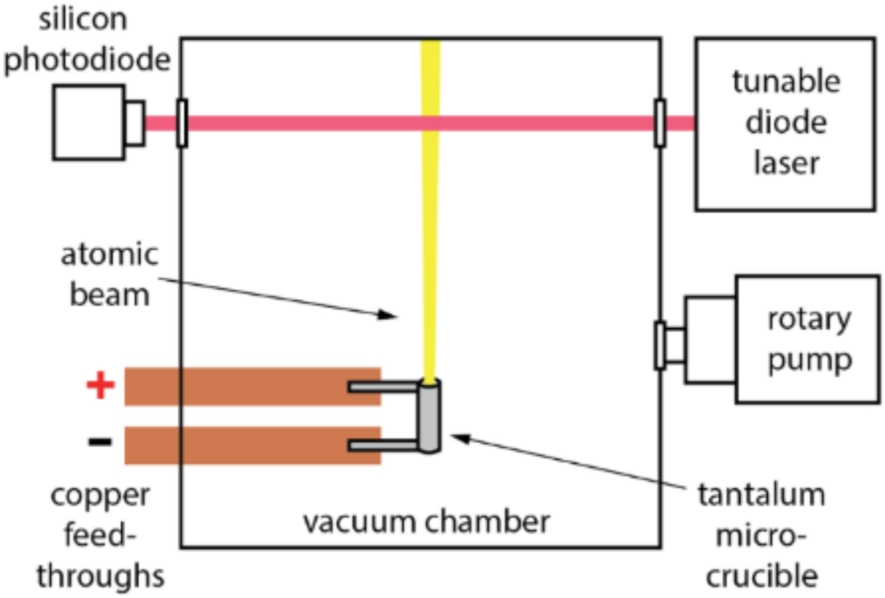

*Figure 3. Schematic diagram of the experimental method for isotopic analysis in atomic beams.*

The laser beam has a diameter of a few millimeters, and its power is relatively low—5 mW—to avoid saturation of the atomic transition. The narrow spectral linewidth of the laser (< 300 kHz), combined with a low-divergence atomic beam that reduces transverse Doppler broadening, allows us to resolve the isotope shift easily. During the sample analysis, the laser center frequency is rapidly tuned through a range of a few gigahertz at a scan rate of 10 Hz. The absorption signal captured by the photodiode is synchronized with the scan rate of the laser and digitized by a data acquisition module controlled by a custom LabVIEW software program. Fig. 4 shows FABIA's user interface running on a built-in Apple iPad.

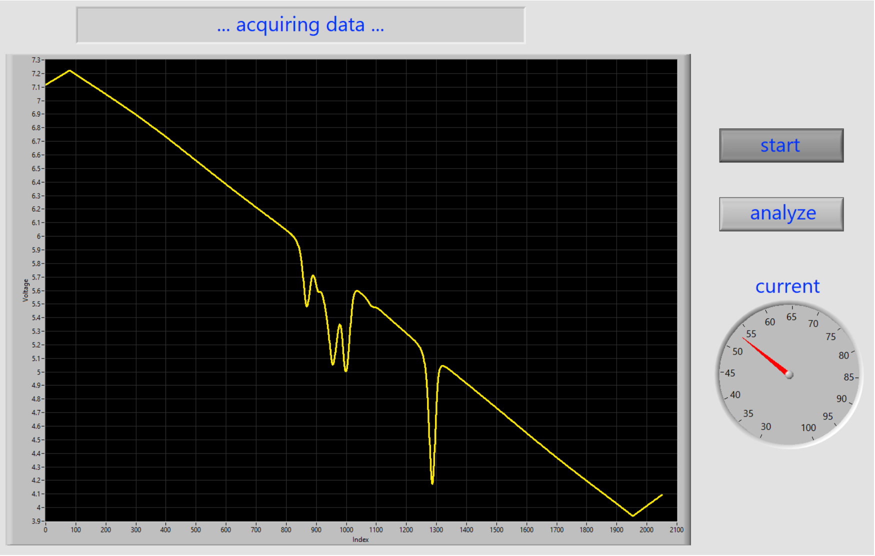

*Figure 4. FABIA's user interface. Shown is a raw spectrum of 63% enriched uranium.*

## Results

We have analyzed SUDA samples containing $UF_6$ at 4.7%, 9.2%, and 92.2% levels of enrichment (expressed as $^{235}U/(^{235}U + ^{238}U)$ atom percentage). The equivalent uranium mass in each SUDA sample was 7 to 8 mg. Fig. 5 shows typical spectra obtained for these samples. The multiplet to the left originates from the $^{235}U$ isotope, and consists of a superposition of 21 hyperfine transitions due to its non-zero nuclear spin. The single peak on the right originates from the $^{238}U$ isotope. By measuring the relative areas of these two features, the isotope ratio can be determined. A comparison of the measured values to those obtained by thermal ionization mass spectrometry (TIMS) reveals a measurement accuracy of 0.5%, and a precision of 1%, in the worst cases.

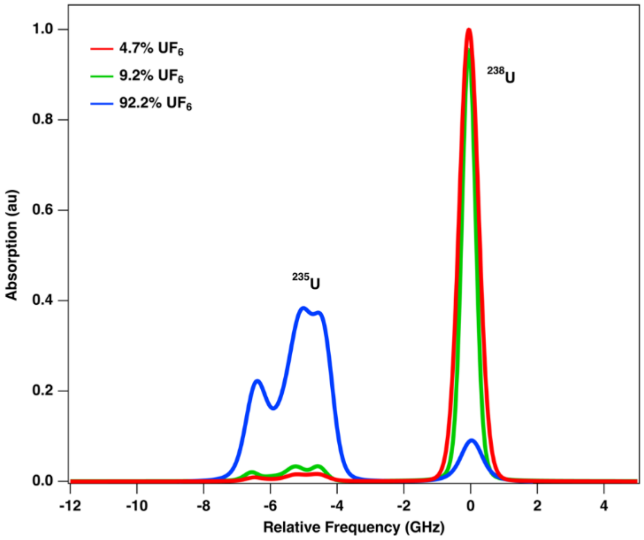

*Figure 5. Typical absorption spectra of SUDA samples*

In addition, we deployed FABIA to a $UF_6$ laboratory at Pacific Northwest National Laboratory (Richland, WA, USA) for a field test. We successfully demonstrated the analysis of sampled $UF_6$ gas of various levels of enrichment using SUDA samplers (Fig. 6).

## Discussion

The main advantage of FABIA over competing technologies is its small size and, consequently, its portability. At the heart of our invention lies the high-temperature micro-crucible [1]. This 10-mm-tall crucible, made from tantalum foil, can be heated to temperatures up to 2600°C and allows the vaporization of most refractory materials in the form of an atomic beam. This in turns allows the atoms to interact with a laser individually, without the deleterious effects of Doppler broadening, allowing users to identify the isotopic composition of the sample. Mass spectrometry also deals with individual atoms (strictly speaking, ions) generated by a variety of means, but these ions then need to be exposed to a large magnetic field over a long distance to measure their mass-to charge ratio to resolve the isotopic composition of the sample. This translates into a very bulky instrument.

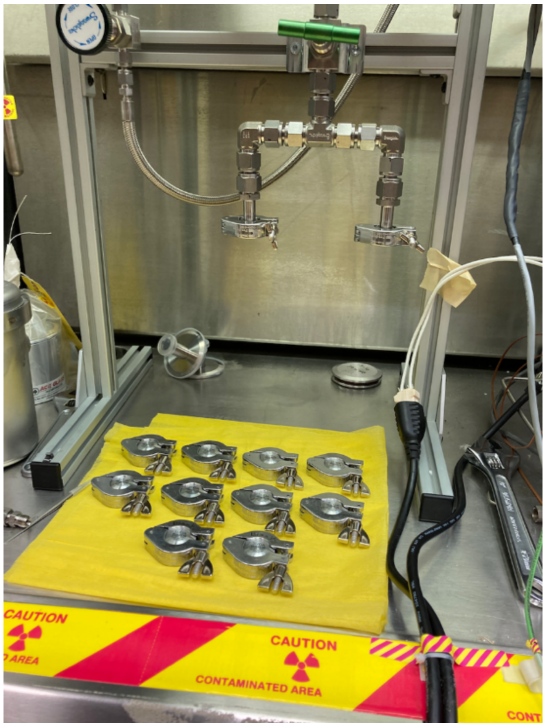

*Figure 62. A set of SUDA samplers used to collect UF$_6$.*

Another advantage of FABIA is that it identifies the isotopes based on their optical properties—that is, the frequency of light that is absorbed over a very narrow band—on the order of a few picometers. Because atoms of a given element absorb light over a very narrow band, no interference from other isotopes is present in the sample. Thus, there is no need to conduct elemental separations before analysis. Conversely, mass spectrometry measures the charge-to-mass ratio of the atoms present in the sample. There are innumerable cases of nuclear samples that contain isotopes of different elements that have the same mass-to-charge ratio, the so-called isobars: $^{238}$Pu and $^{238}$U, $^{241}$Pu and $^{241}$Am, H$^{238}$U and $^{239}$Pu, etc. The mass-to-charge ratio of these pairs is practically the same and interfere with each other. Thus, every sample analyzed by mass spectrometry must undergo a time-consuming process that consists of dissolution by strong acids followed by elemental separation by ion chromatography. This process takes days to complete, cannot be conducted in the field, and increases the cost per sample.

In addition, the analysis time for FABIA is less than 10 minutes, whereas most other techniques, including sample preparation, can take several days to completion. In the case of analyses for Safeguards, this advantage has the potential to greatly improve in the ability of inspectors to immediately take appropriate actions when an out-of-compliance sample is found, as opposed to waiting weeks to obtain results at a fixed laboratory. FABIA has been developed to be fully automated and does not require extensive training for operation. The user interface, developed in LabVIEW, is highly intuitive, and the data analysis happens in real-time.

## Conclusion

We designed, built, and field-tested the Fieldable Atomic Beam Isotopic Analyzer (FABIA), a portable instrument for conducting isotopic analysis of nuclear materials in the field. FABIA can analyze samples at GCEPs, which reveals immediately whether uranium has been enriched beyond reactor-grade levels. Total analysis time, including chamber evacuation, sample heating and data collection, takes less than 10 min.

## Acknowledgement

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