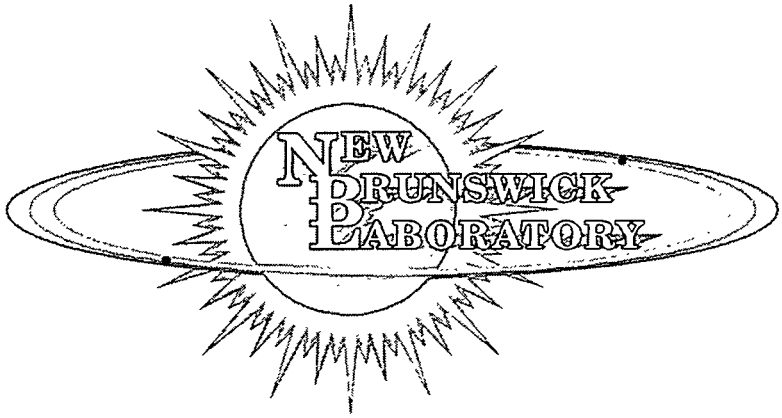
NBL-341
APRIL 1997

PROGRESS REPORT



OCTOBER 1995
THROUGH
SEPTEMBER 1996

U.S. DEPARTMENT OF ENERGY • CHICAGO OPERATIONS OFFICE • ARGONNE, ILLINOIS

U.S. DEPARTMENT OF ENERGY

CHICAGO OPERATIONS OFFICE

NEW BRUNSWICK LABORATORY
9800 SOUTH CASS AVENUE
ARGONNE, ILLINOIS 60439

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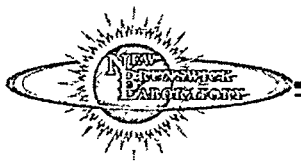

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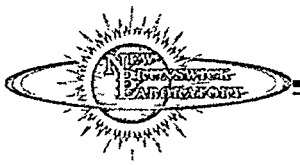

OWNERSHIP

New Brunswick Laboratory (NBL) is owned and operated by the U.S. Department of Energy (DOE). Although it is part of DOE's Chicago Operations Office system, its primary sponsor is the Office of Safeguards and Security in DOE's Office of Nonproliferation and National Security, Office of Security Affairs.

DOE MISSION

DOE is entrusted to contribute to the welfare of the nation by providing the scientific foundation, technology, policy, and institutional leadership necessary to achieve efficiency in energy use, diversity in energy sources, a more productive and competitive economy, improved environmental quality, and a secure national defense.

NBL MISSION

NBL serves as the U.S. government's central authority for nuclear material measurements and measurement evaluation. It is also the U.S. government's certifying authority for nuclear reference materials. These functions assure that the United States maintains an accurate and reliable nuclear safeguards program, particularly in the area of nuclear materials accountability. NBL's program and technical capabilities not only enhance domestic nuclear security but also support international nonproliferation efforts. Its nuclear measurements and measurement evaluation roles allow the federal government to perform independent technical audits and validate nuclear material measurements made by contractors. NBL also has the technical capability for the independent resolution of measurement and safeguards anomalies that may arise from nuclear operations and the transfer of materials between sites.

NBL HISTORY

NBL was established by the Atomic Energy Commission in New Brunswick, New Jersey, in 1949. It was initially staffed by scientists from the National Bureau of Standards who had contributed to the science of measuring nuclear materials for the Manhattan Project. At first, NBL's mission was to provide the federal government with the capability to assay uranium-containing materials for the nation's developing atomic energy program. Over the years, NBL expanded its capabilities, improving methods and procedures, developing new ones, and certifying additional reference materials for use around the world. It incorporated the capability to make plutonium measurements in 1959. During the period from 1975 through 1977, NBL was relocated from New Jersey to the current site at Argonne, Illinois.

Since its beginning, NBL has been a center of excellence in analytical chemistry and the science of measuring nuclear materials. In this role, NBL continues to make state-of-the-art measurements of elemental and isotopic compositions for a wide range of nuclear materials.

Fiscal year (FY) 1996 was a very good year for New Brunswick Laboratory (NBL), whose major sponsor is the Office of Safeguards and Security (NN-51) in the U.S. Department of Energy (DOE), Office of Nonproliferation and National Security, Office of Security Affairs. Several projects pertinent to the NBL mission were completed, and NBL's interactions with partners and customers were encouraging. Among the partners with which NBL interacted in this report period were the International Atomic Energy Agency (IAEA), NN-51, Environmental Program Group of the DOE Chicago Operations Office, International Safeguards Project Office, Waste Isolation Pilot Plant (WIPP), Ukraine Working Group, Fissile Materials Assurance Working Group, National Institute of Standards and Technology (NIST), Nuclear Regulatory Commission (NRC), Institute for Reference Materials and Measurements (IRMM) in Belgium, Brazilian/Argentine Agency for Accounting and Control of Nuclear Materials (ABACC), Lockheed Idaho Technologies Company, and other DOE facilities and laboratories.

NBL staff publications, participation in safeguards assistance and other nuclear programs, development of new reference materials, involvement in the updating and refinement of DOE documents, service in enhancing the science education of others, and other related activities enhanced NBL's status among DOE laboratories and facilities. Noteworthy are the facts that NBL's small inventory of nuclear materials is accurately accounted for, and, as in past years, its materials and human resources were used in peaceful nuclear activities worldwide.

NBL's achievements in FY 1996 are summarized below. Detailed reports are provided in the pages that follow.

MEASUREMENT DEVELOPMENT PROGRAM

It is the responsibility of the Measurement Development Program to maintain state-of-the-art assay capabilities for nuclear reference materials. Among the FY 1996 results of this program are the following:

- Development of a new mass spectrometer protocol for handling low-level uranium samples;
- Refinement of a segmented gamma scanner methodology to use for waste assay measurements;
- Validation of trace metal analyses that use inductively coupled plasma-mass spectrometry (ICP-MS);
- Investigation of the application of various performance indicator tests to the operation of its ICP-atomic emissions spectrometer;
- Assessment of NBL's waste streams; and
- Assessment of protocols necessary to reuse potassium dichromate solutions.

Critical to any analytical laboratory is its instrumentation. NBL continuously maintains and upgrades its laboratory equipment. As equipment becomes outdated or new initiatives surface that enable the staff to adapt methodologies to current guidelines (e.g., less waste), new purchases are made. Among these purchases

in FY 1996 were a direct injector nebulizer that was installed on the ICP-MS and a JCC-51 active well neutron coincidence counter.

MEASUREMENT SERVICES PROGRAM

Although the major part of the support provided by the Measurement Services Program in FY 1996 went to the SME Program and other NBL programs, services were provided to external agencies too. The program provided measurement services for uranium samples to the NRC for several of its licensees, to IRMM, and to ABACC. For example, through the NRC, NBL helped characterize the sapphire materials obtained from Kazakhstan. Other samples were characterized for use in ABACC's Sample Exchange Program as nondestructive secondary standards. In general, the types of analyses used in the Measurement Services Program included titrimetry, gamma spectrometry, and mass spectrometry.

REFERENCE MATERIALS PROGRAM

In fulfilling its mandate as the federal government's certifying agency for nuclear reference materials, NBL has continued to meet the needs of the nuclear safeguards community. NBL is pleased to report increased sales of certified reference materials and improved overall quality of the certification processes through interactions with other reference materials laboratories. In FY 1996, special nuclear materials stored at Lockheed Idaho Technologies Company in Idaho were moved to the Y-12 Plant in Oak Ridge, Tennessee, for storage. Storage of plutonium and neptunium materials assigned to the Reference Materials Program continues at Los Alamos National Laboratory.

Recently, NBL prepared and certified two new reference standards. During this report period, the performance of these standards was tested. Certified reference material (CRM) 145, the uranium (normal) assay solution standard, is one of those standards. More than 450 units of this standard were certified and are available for sale. CRM 125-A, the uranium (enriched) oxide assay and isotopic pellet standard, is also available for sale. Certified values based on International Organization for Standardization (ISO) guidelines are being determined. By next year, the modified certificates will be issued.

Other steps taken in response to customer needs include the preparation of CRM 112-A, the uranium metal assay standard, from bulk stock and its packaging into 4-gram instead of the previous 26-gram packages; the recertification and packaging of CRM 17-B, the uranium (normal) tetrafluoride standard, in 50-gram units; and the recertification of CRM 42-A, the uranium (normal) counting standard, with a uranium elemental assay of 0.5% to 4%.

The two standards (CRM 144, a plutonium triple atom spike standard, and CRM U930, an enriched uranium isotopic standard) that were reported last year as problems were removed from sale.

Another component of the Reference Materials Program is the preparation of internal quality control (QC) standards for use in various laboratory projects. The preparation and distribution of uranium and plutonium blind standards enable NBL to meet internal QC requirements. During this report period, 600 QC standards were prepared and distributed within the laboratory. The NBL Quality Control Program confirmed the validity of the CRM values.

SAFEGUARDS ASSISTANCE PROGRAM

The Safeguards Assistance Program staff provided safeguards assistance to DOE headquarters organizations, the Central Training Academy, and DOE operations offices. The staff assisted in site-specific activities both nationally (e.g., Idaho National Engineering Laboratory, Savannah River Site, Rocky Flats Environmental Technology Center, Oak Ridge National Laboratory, Los Alamos National Laboratory, and Hanford Site) and internationally (e.g., Former Soviet Union, Brazil, and Argentina). Assistance to the Carlsbad Area Office Technical Assistance Contractors was provided, with the objective of certifying the sites' abilities to ship waste to WIPP.

An example of interactions with Russia is NBL's participation in material control and accountability (MC&A) monitoring activities for the United States Transparency Agreement with Russia. Sites involved included the Ural Electrochemical Integrated Plant in Novouralsk, Russia, the Siberia Chemical Enterprise in Seversk, Russia, and the Krasnoyarsk Electrochemical Plant in Zelenogorsk, Russia.

Assistance provided to WIPP for nondestructive assay (NDA) techniques included the following: (1) help in clarifying Idaho National Engineering Laboratory's position and WIPP's position on methods for determining the total uncertainty of a system's measurement, (2) program review, (3) a special inspection to address the resolution of findings and deficiencies from a previous inspection, and (4) activities to help move facilities in the direction of Quality Assurance Program Plan (QAPP) compliance and WIPP certification.

SAFEGUARDS MEASUREMENT EVALUATION PROGRAM

The NBL Safeguards Measurement Evaluation (SME) Program is designed to monitor the quality of the destructive analyses of uranium elemental concentration and uranium and plutonium isotopic abundance measurements. This program, which focuses on nuclear facilities, provides technical support service to NN-51. When the final step of the program — the entry of all FY 1996 uranium data into the new streamlined system — was completed, the program became fully operational. This program has proven to be timely and cost-effective in terms of monitoring measurement performance at various facilities.

Another step taken toward improved efficiency was the incorporation of the Calorimetry Exchange Program into the SME Program. The first foreign participant in the SME Program (the Safeguards Analytical Laboratory in Tokai, Japan) was approved; its full participation will begin next year.

Participants in the normal uranyl nitrate portion of the program (e.g., Los Alamos National Laboratory, NBL, the Lockheed Martin Y-12 Plant, Westinghouse Savannah River Site, and Babcock and Wilcox Naval Nuclear Fuels Depot) use the information from the program to evaluate uranium assay accountability measurements on solution samples at their respective facilities. Argonne National Laboratory-West, NBL, the Savannah River Plant, and Y-12 Plant make use of enriched uranyl nitrate solutions. Ways in which these facilities benefit from the availability of samples made possible by the SME Program are described in this report. Other materials made possible by the program include uranium oxide pellets for use by NBL and four NRC licensees and plutonium in three different concentrations for use by selected facilities.

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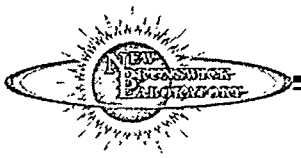

3 MEASUREMENT DEVELOPMENT

MEASUREMENT DEVELOPMENT PROGRAM

J.P. Zebrowski

The Measurement Development Program maintains state-of-the-art assay capabilities in support of the Reference Materials Program and, to a lesser extent, the Safeguards Measurement Evaluation Program and Measurement Services Program. Projects within the Measurement Development Program generally focus on increasing the efficiency and effectiveness of measurement methods, reducing waste produced by analytical methods, and upgrading and replacing obsolete analytical instrumentation.

ANALYTICAL METHODS

New Brunswick Laboratory (NBL) has finished the laboratory work necessary to qualify a new mass spectrometry protocol for handling low-level uranium samples. This protocol allowed NBL to perform confirmation measurements on environmental reference materials that had been prepared by the Institute for Reference Materials and Measurement (IRMM), Geel, Belgium. These reference materials are needed for the International Atomic Energy Agency (IAEA) environmental monitoring program.

NBL uses a segmented gamma scanner (SGS) for waste assay measurements. NBL tested the response of the SGS instrument to variations in uranium isotope ^{235}U mass amount, variations in the matrix, and changes in radial and axial positioning of the uranium within a 55-gallon drum.

Preliminary work has begun on the validation of trace metal analysis methods via inductively coupled plasma-mass spectrometry (ICP-MS). The following two methods are being pursued: a uranium matrix matching method based on an ASTM (American Society of Testing and Materials) method, and an ion-exchange separations method.

NBL investigated the application of various performance indicator tests to the operation of its Varian Liberty™ 110 inductively coupled plasma-atomic emissions spectrometer (ICP-AES). These tests measure the instrument's selectivity, sensitivity, repeatability, and reproducibility and are an important part of the instrument's quality assurance procedure.

ANALYTICAL INSTRUMENTATION

NBL has enhanced its ICP-MS through the purchase of a direct injection nebulizer (DIN). The DIN allows analysis of very small samples and results in a negligible amount of waste. Using the DIN will reduce analyte waste volumes for a daily run from ~500 to ~5-10 milliliters per day. The DIN has been purchased and installed, and the ICP-MS instrument has been optimized for use of the DIN. The funding was provided by the U.S. Department of Energy (DOE), Office of Environmental Management (EM-334), and is the result of a high-return-on-investment proposal submitted by NBL.

NBL has purchased a Canberra™ JCC-51 active well neutron coincidence counter to perform both active and passive neutron assays. The system will be used to assay uranium and plutonium in materials not readily

measurable by gamma spectroscopy or segmented gamma scanners. NBL will expect delivery of the system in FY 1997.

POLLUTION PREVENTION

NBL received funding for two projects from EM-77 (the Office of Pollution Prevention in DOE's Office of Environmental Management, Office of Site Operations) through the Chicago Operations Office, Environmental Programs Group. This funding allowed NBL to perform a pollution prevention opportunity assessment (PPOA) on the laboratory's waste streams and to test protocols and procedures necessary to reuse solutions of potassium dichromate.

UNDERGRADUATE TRAINING

NBL has again sponsored an undergraduate student in conjunction with Argonne National Laboratory's Summer 1996 Student Research Participation Program. Jennifer Walter participated in Measurement Development Program activities at NBL under the supervision of Dr. Patricia Santoliquido. This program benefited both the laboratory and the student; NBL received additional assistance on a development project, and the student obtained experience in a working analytical laboratory.

Cynthia Thompson from Valparaiso University performed her senior physics project at NBL under the supervision of Dr. David Baran. Ms. Thompson developed the InSpector™ assay system's ^{235}U enrichment procedure and performed assays of the nondestructive assay gamma standards prepared at NBL for the Y-12 Plant at Oak Ridge, Tennessee.

PUBLICATIONS AND PRESENTATIONS

A poster was presented at the DOE Pollution Prevention Conference XII, held in Chicago, Illinois. It was entitled, "Pollution Prevention in Actinide Analysis — A High Return Project."

Two posters, "Evaluation Study of Segmented Gamma Scanner Methods for Accountability Measurements" and "Development of Plutonium Isotope Dilution Mass Spectrometric Analysis," were presented at the 37th annual meeting of the Institute for Nuclear Materials Management (INMM) in Naples, Florida.

LOW-LEVEL MASS SPECTROMETRY

A.J. Traina and G.A. Sowell

New Brunswick Laboratory (NBL), in cooperation with the International Atomic Energy Agency (IAEA), accepted a task to perform the initial isotopic measurements on four solid and six liquid sample materials to be used, after certification, in a test program on laboratory capabilities in mass spectrometry. The solid materials were UO_2 powders, easily dissolved, prepared for isotopic analyses, and made to a concentration of 1 milligram (mg) per milliliter (mL) in 0.8 *M* nitric acid (HNO_3). The final sample concentration was consistent with NBL standard procedures for isotopic analysis. The six liquids were received as solutions of 45 parts per million (ppm) uranium in 1 *M* HNO_3. This concentration was lower than that typically used at NBL and required procedural modifications and testing. Because of the low concentrations of uranium in solution, extremely low contamination levels were required in the sample preparation laboratory to ensure that the laboratory that was being used for sample preparation and filament loading contributed minimally to the uranium contamination of the samples.

Laboratory C106 was selected as one of the best, if not the best, cleaner laboratories in the building. Before this work, laboratory C106 had been the subject of a contamination study associated with isotope dilution mass spectrometry. At that time, it was determined that the laboratory would sustain work at a maximum blank level of 2 nanograms (ng) of uranium per gram (g). To ensure minimum contamination, laboratory C106 was thoroughly cleaned before standards were prepared or sample materials were opened. Finally, all containers used to prepare or store standards and samples were acid washed with 3 *M* HNO_3 and then rinsed in distilled water. Aliquants of the rinse acid and distilled water were sampled for analysis by kinetic phosphorescence analysis. The assays showed zero parts per billion (ppb) of contamination per 80 g of sample material.

Before thermal ionization mass spectrometric (TIMS) analyses began, certified reference materials (CRMs) were prepared at a concentration of 45 ppm uranium in 0.8 *M* HNO_3. Three standards were selected for the study: CRMs U500, U010, and U005. The U010 and U005 standards most closely resemble the IAEA materials, which are all near normal in uranium isotopic distribution. The U500 would be used for all initial testing because of its $^{238}U/^{235}U$ ratio of approximately one (1) and the relatively high abundance of the minor isotopes. CRMs were diluted gravimetrically to minimize errors.

Approximately 53 mg of CRM U500 (U_3O_8) was weighed quantitatively and added to a 50-mL beaker. Then 10 mL of 1:1 HNO_3 was added, and the beaker, with a cover glass, was placed on a hot plate over low heat to dissolve the uranium oxide. The resulting solution was then slowly brought to dryness. After it was cooled, the material was again treated with 10 mL of 1:1 HNO_3 liquid and slowly brought to dryness without a cover glass. The uranyl nitrate solid was dissolved in about 10 mL of 0.8 N HNO_3. The solution was then quantitatively transferred to a tared, 125-mL polyethylene bottle; 0.8 N HNO_3 was added to achieve a final net weight of about 100 g. The next day, 10 mL of solution was weighed into a tared polyethylene bottle, and the weight was determined. This 10 mL of solution was again diluted to approximately 100 g in the polyethylene bottle. This second 100-mL dilution contained approximately 45 ppm of uranium per gram of solution. Cleaned glass vials were tared on a balance, and 8 g of the 45-ppm solution was added to each vial. These solutions were set aside for use in the isotopic procedure experiments. A similar procedure was used to prepare both the CRM U010 and U005 solutions in duplicate. The resulting 8-g solutions contained 47 ppm in the U500, 44 ppm in the U005, and 45 ppm in the U010.

In the TIMS analyses of samples, routine mass spectrometric procedures were followed, but at lower intensities because of the 0.09-microgram (µg) loadings on the filaments. Several variations of this

low-intensity experiment were investigated. One variation involved a regular use of the Finnigan™ software modules to run the analyses in the "AUTO" acquisition mode. This experiment failed because of a loss of signal early in the data collection. More attempts at analysis were made by using a manual step heating procedure and automatic data collection. A review of the data showed results that ranged from good to very poor. The most troublesome characteristic of the results over several turrets was their inconsistency. The major ratio, $^{238}U/^{235}U$, varied from turret to turret and from location to location. This inconsistency prevented acceptance of the method for the 45-ppm samples.

After several unsuccessful runs using turrets with various parameters, a decision was made to use the Finnigan™ "total burn" module (COL7). Again, several runs using turrets with various parameters met with limited success. Modifications were made to the module several times in an attempt to improve experimental results for the 45-ppm uranium standards. After several turrets were run, it was determined that the new COL7 module contained the same algorithmic errors as the old total burn module of software version 3.1. Because of these errors, correct abundance measurements for the four uranium isotopes in the CRMs analyzed could not be obtained. The errors could be corrected, but this would require more time than was allowed for the project. It was decided that this procedure was inadequate, and the use of total burn was terminated.

The final method used to analyze the IAEA materials was to collect the COL7 module data in an experiment with standard FIL1.5 step heating and data reduction using the module RED15. This method required the use of a correction factor, as does normal mass spectrometric analysis. The initial turrets proved to be no better than earlier attempts in terms of precision, signal stability, or run duration. Results were inconsistent from turret location to location, and the uranium minor isotopic ratios continued to give poor results. Analysis results ranged from near perfect to incomplete because of the rapid burnoff of sample material, a situation that results in no signal for data collection. Several variations of the procedure were tried to achieve consistently acceptable results for analysis of the six low-concentration samples. Finally, a set of parameters that made use of short step heating, quick lens and deflection focusing, and a strong signal intensity provided consistent results. Results were consistent from location to location and from turret to turret. The abundance measurements of the four uranium isotopes for CRMs U500, U010, and U005, as calculated on a spreadsheet, showed excellent reproducibility and accuracy.

The statistician reviewed the data sets and determined a set of confidence intervals for each of the uranium isotopic abundance measurements. The determined confidence intervals were close to those found in the NBL standard analysis method incorporating 1-µg loadings. The analyst and statistician agreed that the parameters used for the last four turrets would be used for the IAEA samples. Use of this low-level mass spectrometric method for analysis of samples containing 10-100 mg per gram provides acceptable results. These quantities may be the limit for the method. Determination of lower concentrations of uranium in solution would require further improvement of methodology, instrument modifications, and, possibly, a class 100 clean room.

EVALUATION STUDY OF SEGMENTED GAMMA SCANNER METHODS FOR ACCOUNTABILITY MEASUREMENTS

D.T. Baran, G.A. Sowell, and M.D. Soriano

INTRODUCTION

New Brunswick Laboratory (NBL) uses its segmented gamma scanner (SGS) to assay laboratory waste accumulated throughout the facility. To better understand the variability of the SGS system, NBL quantified its response. NBL tested the system by assaying 55-gallon (gal) drums with variations in ^{235}U mass amount, radial position, vertical position, and matrix material. The tests consisted of assaying four ^{235}U masses between 0.194 and 12.549 grams (g), four radial positions (r = 0, 9.31, 18.63, and 27.94 centimeters [cm]), four vertical positions (h = 2.54, 24.13, 45.72, and 67.31 cm), and three matrices (air, cadmium lined, and plastic shoe covers).

EXPERIMENTAL SET-UP

NBL used a standard JOMAR™ (Canberra) SGS to assay the 55-gal drums. The system consisted of a JOMAR™ model JSD-31 turntable and controller system, Canberra model GC1318 high-purity germanium (Ge) detector, and Canberra model 1510 Amp/ADC/HV module. The output from the Ge detector was directed to the Amp/ADC/HV module. Pulses from this module as well as output control signals from the JSD-31 controller were fed to an IBM PS/2 Model 30/286 personal computer. The entire process consisted of a double assay run-though, once with the transmission source opened for transmission measurement and once with the source closed for ^{235}U assay measurements. Each assay was broken into 18 segments. The personal computer controlled all phases of the operation from data acquisition through data analysis.

NBL developed a special source holder for the 55-gal drum, permitting radial and vertical placement of the sample. The holder consisted of four pegboard circular disks separated by three 90-cm-long metal rods and nuts. The entire apparatus slipped into the 55-gal drum.

The air matrix (empty drum) was used as a control and a test of the repeatability of the calibration constants. The plastic shoe covers were used to mimic "typical" combustible waste. The cadmium-lined matrix consisted of a small (2 × 2 × 2 inch) cube of cadmium 0.8-millimeter (mm) thick. The sources were placed inside the cube and then placed into an empty drum. This set-up tests the ability of the SGS to perform matrix corrections and attenuation calculations on particularly dense materials. Calculations show that 0.8-mm-thick cadmium has a transmission of 78% for the 185.7-keV gamma-ray.

The four ^{235}U sources, which consisted of Nuclear Regulatory Commission-quantified ash at 0.194, 1.679, 6.529, and 12.549 g each, were used to calibrate the system. The SGS system was calibrated before each type of matrix. Calibration consisted of placing each source in the geometric center of an empty drum. A least-squares linear fit to the data yielded the calibration constants.

RESULTS FROM STATISTICAL ANALYSIS

Sixteen measurements were taken on each of four sources, in which the three matrices were used and the positioning of the sources was varied both radially and vertically, for a total of 192 measurements. The NBL

statistician saw no significant effect from source placement, either radially or vertically. The results from each source were analyzed individually for matrix effects. The results are summarized in Table 1 and graphically in Figures 1-8. Figures 1-4 show the overall results for each standard and matrix. Figures 5-8 show the individual measurements for each standard and matrix.

Statistical analysis of variance (ANOVA) techniques, performed individually on the measured value for each source, revealed a statistically significant bias in the results for the metal matrix. For source A, the significance was 93%; for the other sources, the significance was greater than 99%. These results led to the conclusion, with a high degree of statistical confidence, that at levels significantly above background, the measurements made on the metal matrix were biased low. After the metal matrix data were removed from the study, there was no statistically significant difference between the plastic and air results. Finally, if the lowest ^{235}U mass source data were ignored, the overall relative difference between the assayed value and actual value would be 4.3%, with a standard deviation of 7.7%. This result suggests that the observed ^{235}U mass determination bias in the plastic matrix and air matrix results is statistically insignificant.

DISCUSSION AND CONCLUSIONS

The results suggest that NBL's SGS makes acceptable corrections to count rates for high-transmission matrices indicative of normal laboratory combustible waste. However, NBL's SGS does not make the appropriate matrix corrections or attenuation calculations for chunklike, metallic materials.

TABLE 1 Matrix Effects on SGS Analysis of ^{235}U

Standard	Matrix	Uranium (grams)	Mean Relative Difference (% for matrix)	Deviation of Relative Difference (% for matrix)
A		0.194		
	Air		-2.044	86.192
	Metal		-93.257	138.130
	Plastic		18.743	188.804
B		1.679		
	Air		22.444	15.666
	Metal		-33.157	17.411
	Plastic		12.986	21.055
C		6.529		
	Air		7.014	7.413
	Metal		-33.444	5.696
	Plastic		5.108	10.938
D		12.549		
	Air		6.907	7.496
	Metal		-30.779	7.551
	Plastic		4.010	11.014

One should not be excessively concerned with the results of the lowest ^{235}U mass source. This measurement is just above the measurable background. Nonetheless, even at a 100% to 200% statistical standard deviation, the SGS is only measuring 0.2 g of ^{235}U to a precision of 0.4 g of ^{235}U. Such small amounts of ^{235}U are not of major concern from a safeguards perspective. Controlling the background better, such as by shielding or locating the SGS system in a relatively radiation-free zone, could increase the sensitivity of the system and increase the precision and accuracy of the lower ^{235}U mass source measurements. Since gamma activity is related to the mass of ^{235}U present, measuring these lower mass amounts with any accuracy is probably not within NBL's SGS current capabilities. Consequently, without more work in improving background conditions and better defining drum matrices, NBL's SGS systems are not the system of choice for low mass (<0.5 g ^{235}U) waste assays.

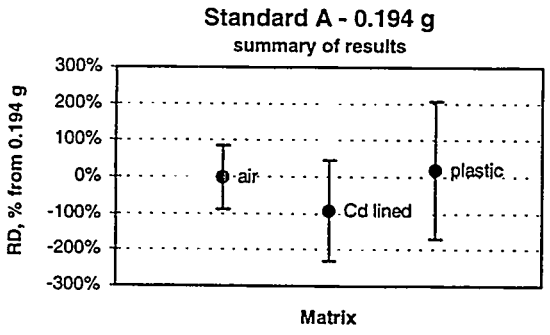

FIGURE 1 Standard A Summary

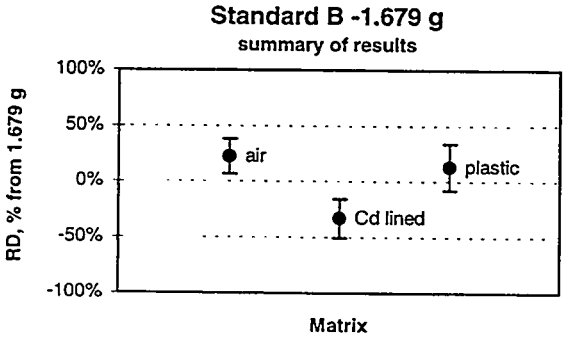

FIGURE 2 Standard B Summary

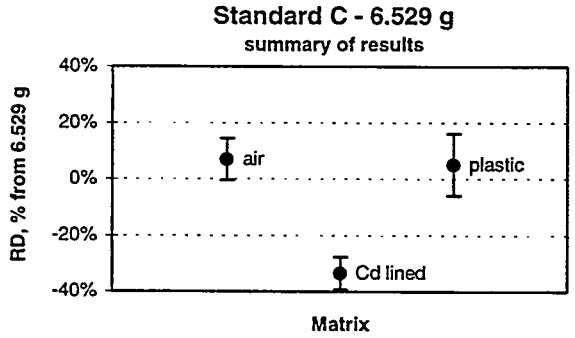

FIGURE 3 Standard C Summary

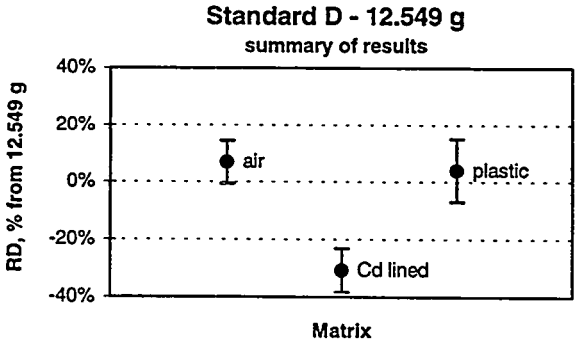

FIGURE 4 Standard D Summary

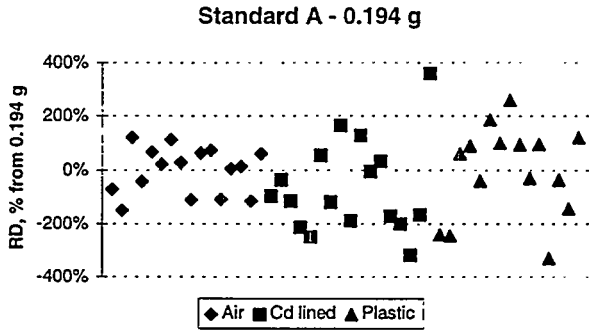

FIGURE 5 Standard A Individual Results

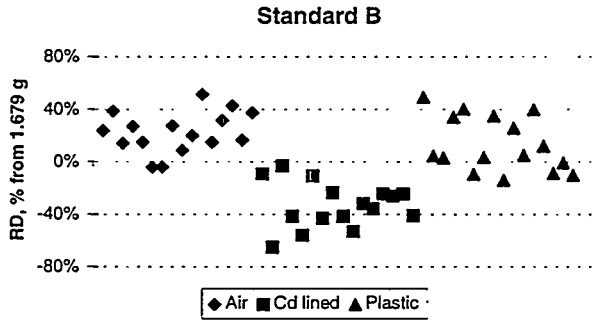

FIGURE 6 Standard B Individual Results

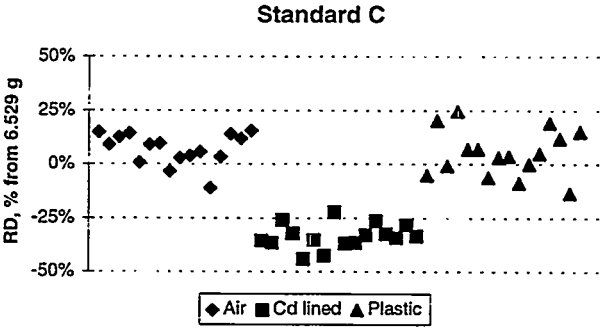

FIGURE 7 Standard C Individual Results

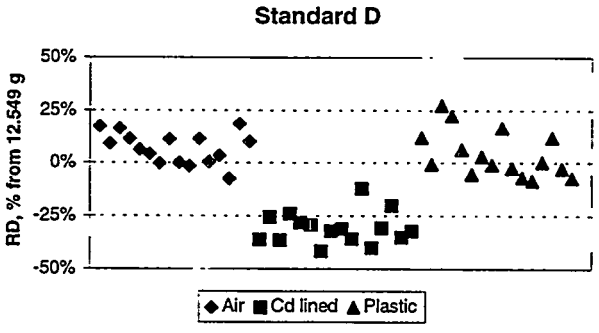

FIGURE 8 Standard D Individual Results

DIRECT INJECTION NEBULIZER

A.R. Warren and J.P. Zebrowski

New Brunswick Laboratory (NBL) purchased a direct injection nebulizer (DIN) and installed it on the inductively coupled plasma-mass spectrometer (ICP-MS) with funds from the U.S. Department of Energy's Office of Environmental Management, Office of Site Operations, Office of Pollution Prevention (EM-77). This funding was supplied as the result of a high-return-on-investment proposal submitted by NBL in fiscal year 1996. The DIN will allow ICP-MS analyses to be conducted on small sample volumes and with minimal waste generation. It is a new method of sample introduction in which sample solutions are nebulized under pressure from a small orifice located directly before the ICP's plasma. In a conventional pneumatic nebulizer, the aerosol is produced in a spray chamber and conducted to the plasma after traveling through the chamber and a conduction tube. Since the DIN allows total consumption of the sample, it produces no wastes in the nebulization step of sample introduction; in contrast, the conventional nebulizer introduces only 1-2% of the sample into the plasma, and the rest of the solution becomes waste. While a typical analysis with a spray-chamber device requires 1-10 milliliters (mL) of sample, the DIN requires less than 1 mL to do the same analysis. The optimization of instrumental parameters for the operation of the DIN is currently underway.

TRACE METAL ANALYSIS BY INDUCTIVELY COUPLED PLASMA-MASS SPECTROMETRY

A.R. Warren, P.M. Santoliquido, G.J. Orlowicz, M.D. Soriano, and J.P. Zebrowski

New Brunswick Laboratory (NBL) operates a VG/Fisons PlasmaQuad PQ2+™ inductively coupled plasma-mass spectrometer (ICP-MS). This instrument will play an important role in future analyses of impurities in uranium and plutonium materials. Work in fiscal year (FY) 1996 centered on the determination of trace levels of impurities in uranium oxide. Work included the development of methods for analysis and the optimization of the instrument itself.

Two analysis methods are currently being developed: a matrix matching method adapted from an existing American Society for Testing and Materials (ASTM) procedure, and a uranium separations method based on U/Teva resin. The ASTM-based method has the advantage of requiring very little sample treatment. The standards are matched with the same concentration of uranium as that contained by the sample, thereby compensating for the severe matrix effects. This method has inherent problems; the high concentration of uranium in the samples clogs the instrument's sampling orifices and causes degraded detection limits. The use of a direct injection nebulizer to reduce the amount of material introduced into the plasma may help alleviate the clogging problem. The alternative method being concurrently developed involves separation of the uranium from the impurities by ion-exchange chromatography before analysis of the impurities. Preliminary testing of the ASTM-based direct method was performed on synthetic samples in FY 1996. Qualification of a NBL method requires precision and bias statements, as well as limits of detection for all quantitative analytical procedures. Both the ASTM direct and the separation methods will undergo statistically based testing to provide the required information during FY 1997.

An inert sample introduction system (ISIS) has been installed in the ICP-MS to facilitate the analysis of corrosive materials. The use of hydrofluoric acid in sample digestions requires a nonglass introduction system, especially in the analysis of trace silicon, lead, and other elements found in glass and semiconductor materials. The ISIS has increased the range of elemental analyses that can be performed on the NBL ICP-MS.

POLLUTION PREVENTION OPPORTUNITY ASSESSMENT

D.E. Dallmann, J.P. Zebrowski, and M.A. Redman

The purpose of this project was to conduct a general, laboratorywide pollution prevention opportunity assessment. Because of the limited number of hours available for this project, its scope was limited to activities producing the largest volumes of radioactive, mixed, and hazardous waste.

This project was completed in several stages. The first step involved an examination of waste generation for the previous year. Waste volume, by type, was examined, as was the difficulty of disposing of each of the waste types examined: radioactive, hazardous, and mixed. Waste disposal difficulties were then examined with regard to current practices as well as those anticipated in the near future.

Individual waste streams were identified as contributors to the waste types examined. Each stream was loosely prioritized by the desirability of waste stream component reduction and by the apparent ease of performing the assessment and obtaining a feasible pollution prevention opportunity.

Radioactive waste was found to be the largest (volume) contributor to the waste generated by New Brunswick Laboratory. The total volume of radioactive waste generated in fiscal year (FY) 1996 was nearly 28 cubic meters (1,000 cubic feet). Surplus equipment made up the most significant amount of this waste. Some of the equipment had become contaminated through use, but some had become contaminated while being stored with contaminated items. It has never been difficult to find a method for disposing of radioactive waste generated by the laboratory. However, the cost involved with disposal is significant, as is the paperwork burden. Surplus equipment disposal costs for FY 1996 exceeded $25,000, and between 0.25 and 0.5 full-time equivalents (FTEs) of effort was spent completing necessary waste disposal paperwork.

Other sources of radioactive waste were also examined. A study of individual waste streams revealed that most other sources of radioactive waste were incidental and that these sources could either not be prevented or not be eliminated cost-effectively by taking advantage of pollution prevention opportunities.

The volume of hazardous waste generated at the laboratory is relatively small when compared with the volume of radioactive waste generated. In FY 1996, 30 requisitions were completed for hazardous waste disposal, 117 were prepared for radioactive waste disposal, and 27 were prepared for mixed waste. Although hazardous waste made up only a small percentage of the total waste generated by the laboratory, the majority of the hazardous waste disposal requisitions involved either expired chemicals or those for which laboratory personnel no longer had any use. By incorporating better planning and a system for getting unneeded chemicals to potential users, this type of waste should be eliminated entirely.

Mixed waste generation at the laboratory currently consists entirely of Davies and Gray titration waste. This waste stream had already been examined by NBL and Argonne National Laboratory (ANL) for several previous projects. A waste treatment option and several analytical procedures were examined. Because of the level of previous effort, Davies and Gray waste generation was outside this project's scope.

Two projects were ultimately identified as having potential for reducing the most waste with the least effort: (1) reduce the amount of surplus chemicals disposed of by the laboratory through an exchange program and (2) set up a system to avoid unnecessary contamination of surplus laboratory equipment. Ultimately, these projects were submitted to the Chicago Operations Office (CH), Environmental Programs Group, and funded by the U.S. Department of Energy's Office of Environmental Management through CH.

POTASSIUM DICHROMATE SOLUTION RECYCLING

I.W. Frank, M.D. Soriano, J.P. Zebrowski, and R.D. Oldham

INTRODUCTION

New Brunswick Laboratory (NBL) received funding from the U.S. Department of Energy's Office of Environmental Management, Office of Site Operations, Office of Pollution Prevention (EM-77), through the Chicago Operations Office, Environmental Programs Group, to explore the possibility of recovering potassium dichromate ($K_2Cr_2O_7$) solutions deemed not usable. The laboratory uses solutions of $K_2Cr_2O_7$ as the titrant in the NBL-modified Davies and Gray procedure for the determination of uranium. The titrant factor of the potassium dichromate relates weight of titrant used to weight of uranium present. Although this factor is assumed not to change over time, it is checked before sample analyses by titrating blind standards.

An individual bottle of titrant may become unusable for several reasons. At times, a solution is put aside because it has become concentrated or because it consists of too small an amount to carry out a project's titrations. At other times, the titrant is put aside because it has appeared to cause erratic results. If solutions that were discarded for benign reasons, such as factor change or amount, could be combined to form a new titrant solution and only the erratic titrant were left for disposal, the laboratory would decrease overall waste. Money could be saved; for example, solutions containing Cr^{+6} are very expensive to dispose of, since they are rated as carcinogenic by the National Institute of Occupational Safety and Health.

In this project, we first determined the criteria that the discarded titrant would have to meet to be used again. We selected the following criteria: (1) the history of the titrant must be known, (2) the titrant must never have passed through the tubing of the siphoning system formerly used in bottling titrant, and (3) the solution must contain only potassium dichromate and distilled water. By using these criteria, we hoped to separate any titrants that might have been contaminated or shown unexplained erratic results.

As a final task, we prepared a batch of recycled titrant that met the stated criteria and tested it.

ANALYSIS AND CONCLUSIONS

Potassium dichromate solutions that had been set aside for disposal but whose documented histories met the criteria described above were combined into one titrant lot and poured into four bottles. The factor for the solution in bottle 1 was determined. This titrant and another potassium dichromate solution being used routinely for current laboratory work were compared by titrating a set of standards with known values. The first set of titrations, performed immediately after the designated disposal lots were recombined, showed perfect correlation between the recombined and the current solutions. However, after four months, the current solution was unchanged, but the recombined solution seemed to have lost strength. Further testing over the next two months confirmed this change. The new factor of the recombined material was determined. It did not appear to change over the next month. Then the factor of the solution from another of the four bottles of recombined solutions was determined. Its value was the same as that from the first bottle. Apparently, the recombined solution had lost strength uniformly in both bottles and had stabilized at that value. Thus, the project generated 8 liters of usable titrant solution that previously would have required expensive disposal. In addition, the work involved in preparing a new titrant solution from potassium dichromate and water was avoided.

PERFORMANCE INDICATORS FOR QUALITY IN INDUCTIVELY COUPLED PLASMA EMISSION MEASUREMENTS

P.M. Santoliquido and J.L. Walter

A high-quality analysis consists of measurements that are selective, sensitive, repeatable, and reproducible. In an inductively coupled plasma (ICP) analysis, each of these measurement characteristics has an associated performance indicator that may be monitored by specific tests to evaluate instrument performance. Several authors (1, 2) have suggested tests that are both meaningful and easy to run. We investigated the application of these tests to the operation of New Brunswick Laboratory's (NBL's) Varian Liberty 110™ ICP instrument. We also explored the use of programs QC1, QC2, and QC3, which are provided in the instrument software.

Selectivity refers to the capability of the technique to differentiate between responses arising from different elements. Distinguishing among the emitted lights of characteristic, differing wavelengths and then resolving these lights into distinct and separate spectral peaks are fundamental components of successful multielement analysis. Resolution, which is defined as the measured peak width at half the peak maximum height, is indicative of selectivity. The narrower the peak is, the smaller the resolution value will be, and the better neighboring peaks will be resolved or separated.

Practical tests of selectivity may be made by determining the resolution of an analyte at a given wavelength and order. The program QC1 provides for the analysis of Cd at 214 nanometers (nm) in first, second, third, and fourth order. Mermet and Poussel (1) recommend using Ba at 230 nm as a test, because the majority of analytical lines used cluster around this region of the spectrum. The resolution determined for Ba at 230 nm on the NBL instrument is 6 picometers (pm).

Another practical test of resolution is to take a solution containing two elements that give adjacent peaks and see if the instrument can give a spectrum in which the two peaks are completely resolved. The NBL instrument completely resolved Mg at 202.582 nm and Zn at 202.551 nm.

Sensitivity for a given analyte may be evaluated by determining the background equivalent concentration (BEC), which is defined as the concentration of an analyte that yields a net intensity signal equal to the intensity of the background, and by determining the limit of detection. These measurements are most meaningful when made in the context of the actual analytical problem being addressed, because the matrix will significantly influence background levels. However, for purposes of general comparison, the determination of the limit of detection for nickel at 231 nm, when a nickel solution of 1 part per million (ppm) is used, provides a realistic test. This is because even with a single-element solution of nickel, significant structured background is caused by nitrous oxide band emissions. If the approach of Boumans (3) is used, the limit of detection, c_L, is related to the signal to background ratio (SBR) of the analyte line at a given concentration, c, and the relative standard deviation of the background, RSD_B, as follows:

$$c_L = [3 \times c \times RSD_B] / SBR \qquad (1)$$

The background measurement is made at 230 nm. The quantities RSD_B and SBR can also be directly compared to a ranking scale provided by Mermet and Poussel (1). We found that the NBL instrument earned an "acceptable" rating on this ranking scale.

Analytical measurements made by ICP are comparative measurements. In other words, a sample (unknown) and a standard (known) are both measured, and the unknown quantity is determined by comparing its response to that of the known. To avoid the introduction of significant error arising from differing instrumental conditions at the times of separate measurements, the plasma stability should be monitored.

The determination of precision from replicate measurements provides a measure of short-term stability, which is often referred to as repeatability. When the instrument does replicates, it simply integrates the signal again for another unit of time. There is no possibility of introducing another source of variability from driving to different wavelengths, repositioning, and relocating analyte peaks. Short-term precision on this instrument is very good and serves as a limit for the best that can be obtained for an analysis. Overall precision for an analysis involving a number of standards and samples will not be as good but, with care, may approach short-term precision magnitude.

A more severe test of precision is to repeat the analysis of a multielement solution over a longer period of time. A long-term stability study identifies instrument drift and is an indicator of reproducibility. A test devised by Mermet and Poussel (1) follows the intensity of Ar at 404.442 nm to monitor the plasma itself and two analyte lines: Ba at 455.403 nm, which is expected to be relatively insensitive to changes in power, and Zn at 206.200 nm, which is expected to be very sensitive to changes in power. The results obtained on the NBL instrument are shown in Figure 1. After the initial plasma warm-up time, the instrumental response indicated signs of drift. The intensities would be stable for approximately 30 minutes (min), drift, and stabilize again at a lower intensity.

The drift observed in this diagnostic test prompted us to check the stability of the response of the analytes of interest to us. We studied nine elements that we expect to separate from uranium as a group by an anion exchange procedure. This study was conducted by recording the mean intensities (five replicates per wavelength) for each element at 15-minute intervals, after the initial 30-min warm-up time. Graphs of the intensity normalized to the first reading were used for data analysis and are shown in Figure 2. All selected elements exhibited the same general drift trend over time, which suggests that the cause of drift was instrument related. Because the argon line, which represents plasma activity, follows the same drift pattern as the analytes, analyte distribution in the plasma and sample introduction can be ruled out as causes of the drift. Because the analyte response of all the selected elements became more stable after approximately 80-95 min, a longer plasma warm-up time for the actual analytical work is indicated.

Robustness is defined as the capability of the ICP system to accept a change in the concentrations of major elements, acids, and other elements (i.e., the matrix) without any significant variation in the line intensity of the analytes. Usually, ionic lines are more sensitive to these changes than are atomic lines. Mermet has reported (4) that the Mg(II 280 nm)/Mg(I 285 nm) line intensity ratio is an efficient indicator of sensitivity to matrix effects. A value of this ratio above 10 indicates that the ICP should not be particularly sensitive to matrix effects. On the other hand, a ratio below 4 corresponds to a high sensitivity to matrix effects. This ratio was found to be 3.95 for the NBL instrument; therefore, it will be important to consider matrix effects when developing analytical procedures.

QC2 is a program that provides for the analysis of a multielement solution containing Al, Ba, Cu, Cd, K, Mn, and Zn by the use of first-order lines. Because the selected lines provide representative coverage for the whole spectral range of the instrument, this program can be used to check the instrument's ability to accurately locate wavelengths throughout its spectral range.

QC3 provides a program for the analysis of the same elements as QC2, but the wavelengths chosen are the most intense lines and their optimal orders have been chosen. This program is useful as a general test of the overall analytical performance of the instrument as determined by checks of the factory specifications for

calibration accuracy and precision. A test solution of 50 parts per million (ppm) K and 5 ppm each of Al, Ba, Cu, Cd, Mn, and Zn is read first as a standard and immediately after that as a sample. The average value for K should be within 4% of the true value and that of the other analytes should be within 2%. The precision for five replicates of sample readings on the test solution should produce less than 3% relative standard deviation (RSD) for K and less than 1.5% RSD for the other analytes (5). The performance of the NBL instrument is acceptable in these respects.

In addition, the copper and manganese lines in the QC3 program provide a test for plasma drift caused by the buildup of material on the nebulizer and sample introduction system. This test takes the ratio of the intensity of a soft atomic line (Cu I 324.74 nm) and the intensity of a hard ionic line (Mn II 257.610 nm) to test the stability of the plasma's analytical zone. Cu/Mn ratios varying 5% or more from the initial value are an indication that the torch, nebulizer, and mixing chamber should be cleaned before proceeding with the analysis (6).

The tests described above can establish the instrumental limitations of the measurement system. With each application, the sample itself may present additional challenges, such as matrix effects and spectral interferences, which will also need to be addressed to ensure accurate analyses.

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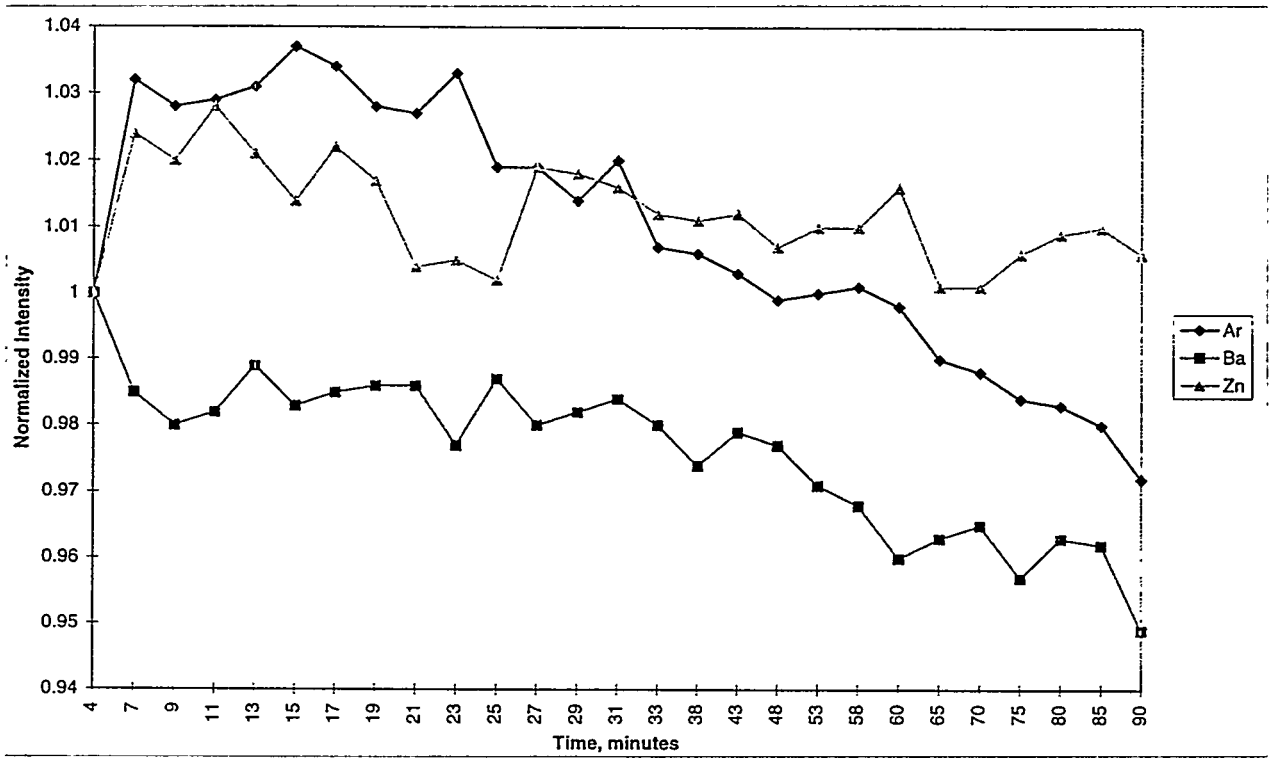

FIGURE 1 Plasma Stability Study

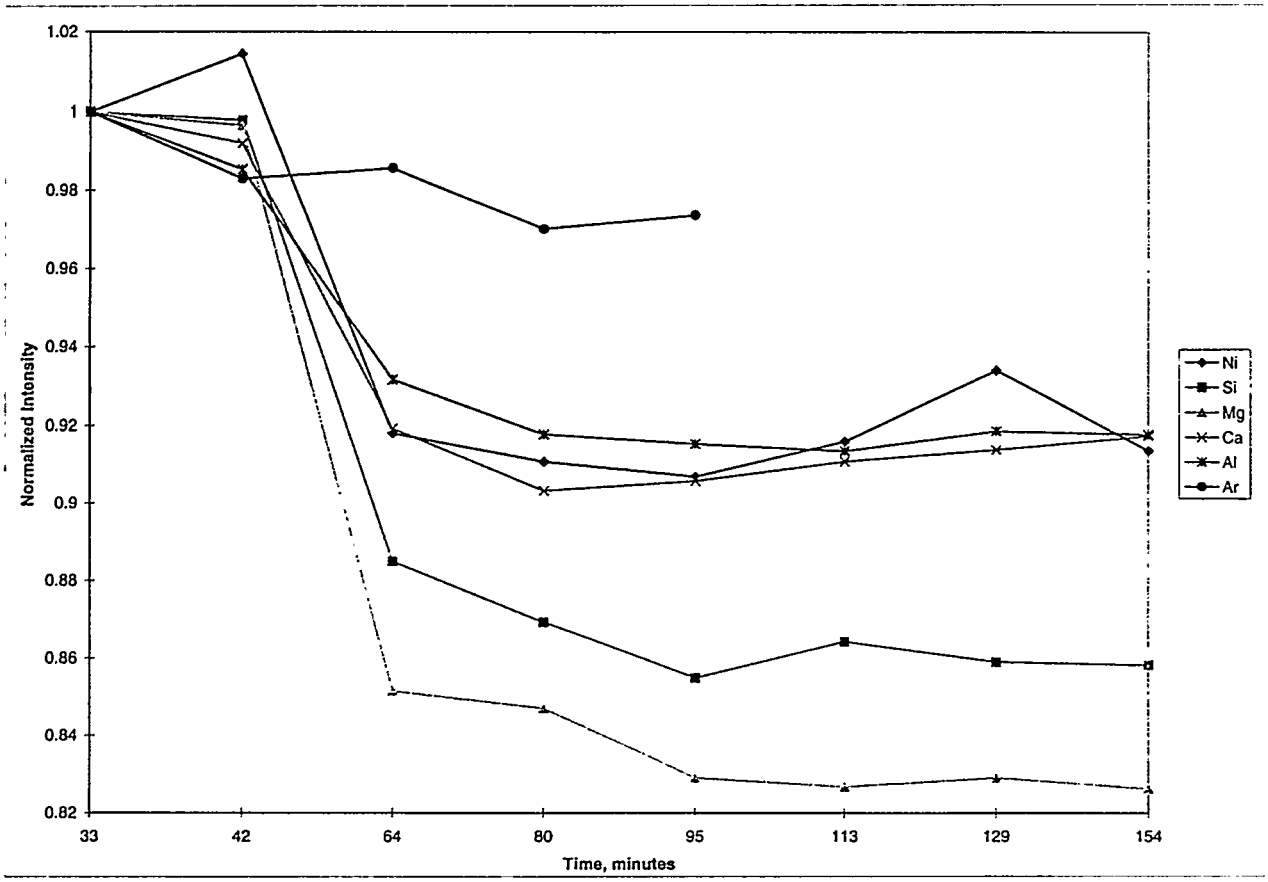

FIGURE 2 Group 1 Stability Test

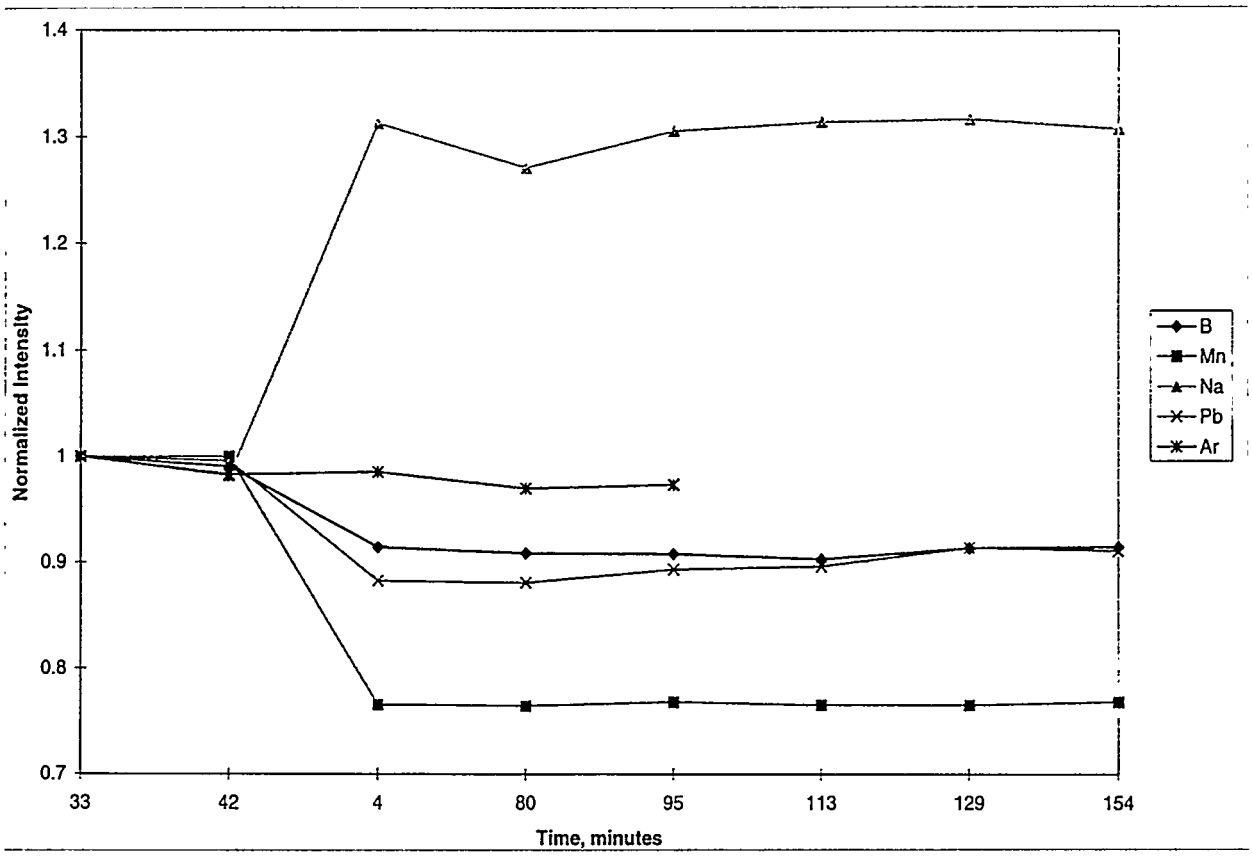

FIGURE 2 Group 1 Stability Test (Cont.)

INTERNAL NORMALIZATION TECHNIQUES FOR ISOTOPE DILUTION MASS SPECTROMETRIC ANALYSIS — APPLICATION TO PLUTONIUM

U.I. Narayanan and M.A. Legel

INTRODUCTION

Isotope dilution is a method for determining elemental and isotopic concentrations of a sample on the basis of the addition of a known quantity of element (a spike) whose isotopic composition differs from that of the sample. By measuring the change in isotopic composition of the sample-spike mixture, the elemental concentration of the sample can be calculated. Isotope dilution analysis depends on the identical elemental behavior of spike and sample throughout chemical preparation and mass spectrometric assay.

Thermal ionization mass spectrometry (TIMS) requires correction for mass discrimination due to changes in isotopic ratios with time resulting from the preferential evaporation of the lighter isotopes, a process also called fractionation.

The mass discrimination correction factors applied to the ratio measurements of a sample can be obtained by (1) measurement of a reference material standard of known isotopic composition under similar analytical conditions, (2) internal normalization by using a pair of isotopes that are inherent in the sample and preferably stable or long-lived, or (3) addition of a spike with an independent set of isotopes (a double spike). The latter two methods are considered to result in the most precise normalization. For many elements with more than four isotopes, internal normalization can be used for calculating the mass discrimination correction factor (1, 2). This internal normalization technique results in the most precise sample isotopic abundance measurements (2). In the case where isotope dilution is required to determine the concentration of a sample, the use of a double spike can correct the isotope dilution ratio for mass discrimination. Use of the double spike typically produces an order of magnitude or better external precision on an isotope dilution measurement.

Without a double spike and its capability for internal normalization, plutonium sample analysis must be corrected from external analysis of certified reference material (CRM) 128, the plutonium isotopic standard ("Plutonium-239/Plutonium-242, 1:1 Atom Ratio in Nitrate Form"). Once this "external" correction factor is determined, the value is used to correct the measured ratios in subsequent sample analyses that are run under identical sample loading and analysis conditions. A large variability in the correction factor (on the order of 0.5%) is typically observed when sample loading and analysis parameters cannot be strictly controlled. Reasons for this lack of control are numerous: (1) differences in the quantities of plutonium loaded onto the filament, especially when sample purification steps were necessary; (2) differences in chemical matrices; (3) interferences caused by sample impurities; and (4) variability in the sample loading technique and filament temperatures during both sample loading and subsequent sample analysis. The introduction of a systematic bias, with the use of an inappropriate correction factor, is directly proportional to a bias in the calculated plutonium content in moles for the sample.

The text that follows describes the exact equations and iteration techniques applied to determine plutonium concentration by isotope dilution mass spectrometry (IDMS). A triple-atom plutonium spike consisting of $^{240}Pu/^{242}Pu/^{244}Pu$ is used for the assay of plutonium samples consisting mostly of the ^{239}Pu isotope. Results of IDMS analyses that used internal versus external standards or spikes for the determination of plutonium assay are discussed in reference 3.

The equations presented in this report illustrate a method for Pu IDMS that uses four isotopes: ^{239}Pu, ^{240}Pu, ^{242}Pu, and ^{244}Pu. Several different isotopic ratios may be used for normalization and IDMS calculation if one assumes ^{239}Pu is the major isotope in the sample and ^{242}Pu and ^{244}Pu are present as minor isotopes. Equations for one system that uses $^{244}Pu/^{242}Pu$ as the normalization ratio and $^{240}Pu/^{239}Pu$ as the IDMS ratio follow. Equations for other combinations can be obtained by a simple substitution of the appropriate isotopes in the given equations.

EQUATIONS AND ITERATIONS

Mix a plutonium sample, with ^{239}Pu as the major isotope, with the plutonium triple-atom spike ($^{240}Pu/^{242}Pu/^{244}Pu$) and chemically treat the mixture to attain isotopic equilibration. In this equilibrated mixture, the abundance measurements of each isotope, expressed as a fraction of the total number of atoms present from all the isotopes, will be the sum of the total number of atoms of that isotope from the sample and the spike. The normalization atom ratio, $R^{4/2}$, can be expressed as:

$$R^{4/2} = \frac{N^4}{N^2} = \frac{F_s^4 A_s + F_u^4 A_u}{F_s^2 A_s + F_u^2 A_u} \qquad (1)$$

where

R = atom ratio,

N = number of atoms,

F = atom fraction,

s = spike,

u = unknown or sample,

A = amount in moles,

and the numerical superscripts 9, 0, 2, and 4 denote the respective plutonium isotopes 239, 240, 242, and 244.

The isotope dilution atom ratio, $R^{0/9}$, can be expressed as:

$$R^{0/9} = \frac{N^0}{N^9} = \frac{F_s^0 A_s + F_u^0 A_u}{F_s^9 A_s + F_u^9 A_u} \qquad (2)$$

Equation 2 can be rearranged and expressed in terms of A_u to give:

$$A_u = \frac{A_s (F_s^0 - R^{0/9} F_s^9)}{(R^{0/9} F_u^9 - F_u^0)} \quad (3)$$

A mass discrimination correction factor, K2, for two masses, is defined for the normalizing ratio as:

$$K2 = \frac{R^{4/2}}{{}^*R^{4/2}} \quad (3)$$

where

$^*R^{4/2}$ = observed ratio and

$R^{4/2}$ = theoretical or corrected ratio.

If one assumes that the fractionation of isotopes is proportional to the differences in isotopic masses, the mass discrimination correction function for one mass, K1, can then be expressed as a linear function of K2:

$$K1 = \left[1 - \frac{1}{2}(1 - K2)\right] \quad (5)$$

Also, the mass discrimination correction factor for one mass, K1, can be defined for the isotope dilution ratio in a manner similar to Equation 4 and rearranged in terms of $R^{0/9}$ as:

$$R^{0/9} = \frac{N^0}{N^9} = {}^*R^{0/9} K1 \quad (6)$$

The isotope dilution ratio, $R^{0/9}$, and hence the moles of sample, A_u, can be solved by using an iterative process, as described below:

- Step 1: Use the observed isotope dilution ratio, $^*R^{0/9}$, to calculate an initial value for the amount of the sample in moles, A_u, by using Equation 3.
- Step 2: Use the initial calculated A_u value to calculate the observed internal normalization ratio, $^*R^{4/2}$, for the spike, by subtracting the sample's contribution to the internal normalization ratio, by using Equation 1.

- Step 3: Use the $^{*}R^{4/2}$ value to calculate the mass discrimination correction factor observed during the analysis, K2, by using Equation 4.

- Step 4: Calculate the mass discrimination correction factor for the IDMS ratio, $K_{1,}$ by using Equation 5.

- Step 5: Calculate an initial corrected IDMS ratio, $R^{0/9}$, by using Equation 6.

Substitute the initial corrected IDMS ratio, $R^{0/9}$, back into Step 1 and repeat the sequence until K2 converges. Typically, the K2 will converge in two to three iterations. Note the values obtained for K2, K1, and $R^{0/9}$, and report A_u, the amount of the unknown sample in moles.

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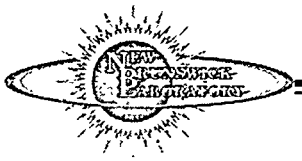


4 MEASUREMENT SERVICES

MEASUREMENT SERVICES PROGRAM

A.V. Stiffin and I.W. Frank

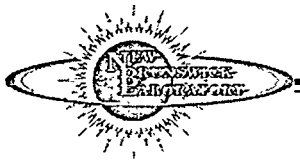
New Brunswick Laboratory (NBL), as the government's nuclear materials measurement and standards laboratory, supports nuclear safeguards programs by providing independent measurements on a variety of nuclear materials. NBL serves as a technical extension of the U.S. Department of Energy, Office of Nonproliferation and National Security, Office of Security Affairs, Office of Safeguards and Security. NBL also provides services on a contractual basis to the Nuclear Regulatory Commission (NRC). During fiscal year (FY) 1996, NBL received 77 uranium samples for analysis. A wide variety of analyses, such as titrimetry, gamma spectrometry, and mass spectrometry (MS), were performed on these samples.

NBL performed MS measurements on two sets of Institute for Reference Materials and Measurements (IRMM) environmental reference materials to verify the isotopic compositions. One set of samples consisted of four uranium oxide samples: two natural uranium oxides and two 35%-enriched uranium oxides. The samples were dissolved and analyzed by the mass spectrometrist. The second set of samples consisted of one split each from six low-level (45 micrograms of uranium per gram of solution), near-natural uranium "grandmother" reference solutions. These solutions had a nitric acid concentration of 1 *M* so it was possible to load them directly with no further sample preparation. These samples were analyzed by NBL in support of an International Atomic Energy Agency (IAEA)/IRMM program to develop reference materials for isotopic measurements on environmental samples.

NBL personnel provided measurement services assistance to the Brazilian/Argentine Agency for Accounting and Control of Nuclear Materials (ABACC) by performing uranium and isotopic assays to characterize UO_2 pellet samples to be used as nondestructive assay (NBA) secondary standards in the agency's sample exchange program.

The NRC's Office of Nuclear Materials Safety and Safeguards (NMSS) submitted six uranium samples for materials verification under the Nuclear Materials Safeguards Program. These samples included UO_2 pellets from Siemens Power Corp. Also in FY 1996, NBL provided assistance to the NRC in characterizing the SAPPHIRE materials acquired from Kazakstan. The total of 24 samples consisted of UNH crystals, UO_2 powders, and uranium metal samples.

NBL received 32 uranium samples as a result of its participation in sample exchange programs. They consisted of 16 uranyl nitrate solution samples, 4 UO_3 powder samples, 4 UF_6 solutions, and 8 UO_2 pellet samples from the NBL Safeguards Measurement Evaluation (SME) Program. NBL participated in the SME Program not only to meet the requirements for participating in a sample exchange program but also to evaluate the stability and integrity of sample materials used in the SME Program.



5 REFERENCE MATERIALS

REFERENCE MATERIALS PROGRAM

U.I. Narayanan and W.G. Mitchell

PROGRAM ADMINISTRATION

New Brunswick Laboratory (NBL), as the U.S. government's nuclear certified reference materials (CRMs) laboratory, continued providing CRMs to meet the needs of the nuclear safeguards community. These CRMs provide assurance that safeguards measurements meet essential compatibility, reliability, and traceability standards for the national measurement base. During fiscal year (FY) 1996, program activities included increasing CRM sales and improving customer service; reaching out to customers and providing customized reference materials; planning for new CRMs that meet the needs of the nuclear safeguards community; and improving overall quality of the certification processes through interactions with other reference materials laboratories.

In FY 1996, 177 units of in-stock CRMs were sold for a total of $108,472. Special orders, involving the preparation, certification, and/or verification of reference materials, resulted in additional funding of $470,500 from outside the U.S. Department of Energy (DOE), Office of Nonproliferation and National Security, Office of Security Affairs, Office of Safeguards and Security (NN-51). A brief description of these special orders is included in the text that follows. The revenue from these special orders and CRM sales for the year totaled $578,972; details on these funding sources are presented in Figure 1.

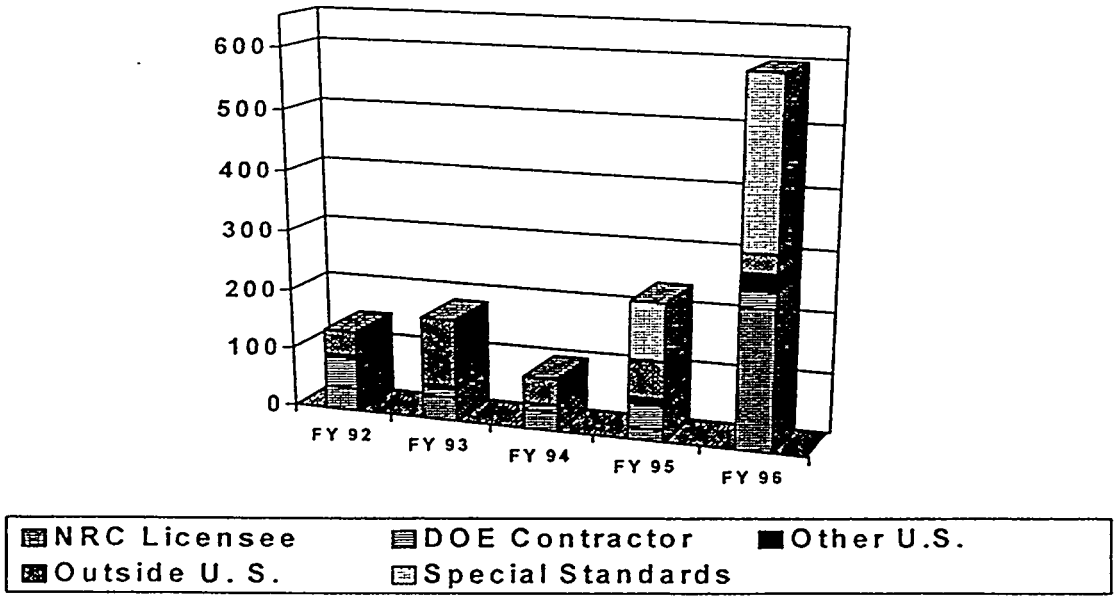


FIGURE 1 Distribution of RM Sales for FY 1992 through FY 1996

Packaging and transportation issues were addressed to resolve shipping container and certification bottlenecks. Internally, laboratory space and equipment were organized to facilitate efficient packaging operations. Sales and shipping issues are discussed in the report on Reference Materials Sales and Shipping on page 41 of this document.

The NBL Reference Materials Program's special nuclear materials (SNMs), stored at the Lockheed Idaho Technologies Company, Idaho Chemical Processing Plant, Idaho, were remeasured, repackaged, and moved to the Y-12 Plant in Oak Ridge, Tennessee, for storage. NBL continues to store the plutonium and neptunium materials assigned to its Reference Materials Program at Los Alamos National Laboratory (LANL).

During FY 1996, NBL scientists gave seven presentations on reference materials preparation, certification, availability, and uses at various professional meetings and conferences. Also, they held several different meetings with technical staff members from other laboratories or safeguards working groups (National Institute for Standards and Technology [NIST], Ukraine Working Group, Waste Isolation Pilot Plant [WIPP], LANL, Institute for Reference Materials and Measurements [IRMM], Y-12 Plant) to discuss specific technical issues related to Reference Materials Program management and projects.

NBL Reference Materials Program management staff met with, and continue to interact with, NIST staff to discuss technical program management and obtain technical references pertaining to reference materials certification protocol. NBL has adopted the International Organization for Standardization (ISO) approach to expressing measurement uncertainty for reference materials certification projects (ISO, *Guide to the Expression of Uncertainty in Measurements*, ISBN 92-67-10188-9, 1st edition, Geneva, Switzerland, 1993.)

NBL Reference Materials Program staff participated in audio/video conferences and meetings with NN-51 staff, DOE program office and operations office staff, and site (Rocky Flats Environmental Technology Center, Savannah River Site, Westinghouse Hanford Company, LANL, and Lawrence Livermore National Laboratory) representatives to discuss domestic safeguards measurements and standards needed for the stabilization and repackaging of excess nuclear materials for intermediate storage in accordance with DOE-STD-3013-96. The sites are in the process of listing the materials that will be stabilized and stored. A need for standards for measuring these materials has been identified. NBL submitted a work proposal to OSS identifying the task of fabrication of nondestructive assay (NDA) standards for this project. NBL will be involved in the planning, production, and certification of this standard by working with OSS, LANL, and other site representatives.

The preparation and characterization of plutonium NDA reference standards for waste sample measurements were discussed with WIPP staff. However, WIPP has not requested that NBL prepare these standards.

NBL has worked with the International Safeguards Project Office (ISPO)/Brookhaven National Laboratory, through NN-40, for the verification of standards for environmental reference materials that are being prepared and certified for safeguards applications at the IRMM in Belgium. NBL received six samples of uranium in nitric acid solution, referred to as "grandmother solutions," that will be used to prepare aqueous reference materials. It also received four samples of uranium oxide, referred to as "uranium base materials," that will be used to prepare soil reference materials. NBL completed verification measurements on the isotopic composition of "uranium base materials" in FY 1996.

NEW REFERENCE MATERIALS

CRM 145, the uranium (normal) assay solution standard, is now available for sale. More than 450 units of this standard (200 milligrams of uranium per unit) were certified for uranium concentration by using gravimetric

measurements. The master solution was prepared gravimetrically by dissolving a determined mass of CRM 112-A, the uranium metal assay standard, in nitric acid. Details are presented in the report on Preparation and Certification of CRM 145 Uranium (Normal) Assay Solution Standard on page 51 in this document.

Three sets of uranium concentration and isotopic standards, for a total of 113 units, were developed for and funded by the Oak Ridge Y-12 Plant Waste Management Decontamination and Decommissioning Division. The fabrication of these standards was based on needs expressed in discussions with Y-12 Plant staff, who expressed a need for traceability of the standard for use in the measurement of waste at their facility. The standards prepared are doubly encapsulated in low-atomic-weight material and can be used in site-specific matrices. The preparation and verification of the standards were presented last year in NBL Progress Report 335. A discussion of the performance of the standards is presented in the report on Preparation and Evaluation of Nondestructive Assay Standards for Uranium Gamma-Ray Spectroscopy on page 43 of this document.

CRM 125-A, the uranium (enriched) oxide assay and isotopic pellet standard, is now available for sale. Partial funding for this project was provided by the NRC. More than 3,900 units (5.4 grams of uranium per unit [g/unit]), were packaged, and a large number of these units were purchased by the NRC. Certification measurements were completed, and a certificate of analysis was issued. However, the measurement uncertainties are being reevaluated to provide certified values following the ISO guidelines. Customers are kept informed of the progress of the reevaluation. A final certificate of analysis will be issued in FY 1997.

PACKAGING OF BULK MATERIALS

To replenish the low inventory of some CRM stock, packaging of bulk materials is in progress. CRM 112-A, a primary uranium metal assay standard widely used for quality control of accountability measurements in the DOE weapons complex and worldwide, is being packaged as 4 g/unit. These packages will replace the 26-g/unit CRM 112-A currently offered for sale. Verification measurements will be completed for the certified properties of CRM 112-A; these units will be available for sale next year.

CRM 17-B, uranium (normal) tetrafluoride, is being packaged as 50 g/unit; these units will replace the 200-g/unit CRM 17-B currently offered for sale. Since this material was last certified in 1961 and the certified uranium content does not include the associated measurement uncertainties, the material will be recertified.

In FY 1995, NBL reported on the packaging of bulk material of CRM 42-A (1-4), the uranium (normal) counting standard. The uranium concentration of this set of CRMs ranges from 0.5% to 4%. Since the standard was last certified in 1957, a decision was made this year to recertify the uranium content in these standards; work is progressing to accomplish this task.

CERTIFICATION PROJECTS

In FY 1996, 600 uranium and plutonium blind standards were prepared for use at NBL and distributed among various measurement methods to meet laboratory internal measurement quality control requirements. A set of three enriched uranium gamma spectroscopy standards is being fabricated. These standards were requested and funded by the Russian/Newly Independent States Nuclear Material Security Task Force in DOE's Office of Nonproliferation and National Security, Office of Arms Control and Nonproliferation (NN-40), for use in the Former Soviet Union (FSU) states. NBL staff discussed the standards needs and specifications

with the Ukraine Working Group. This new set of standards, CRM 146, will be similar to CRM 969, the uranium isotopic standard reference materials for gamma spectrometry measurement, and can serve as a useful extension to that series. The ^{235}U enrichment levels of CRM 146 will be 20%, 54%, and 93%. The status of this project is discussed in the report on New Highly Enriched Uranium Nondestructive Assay Standards: A Beginning on page 47 of this document.

Work is in progress to certify NBL CRM 113-B, the uranium hexafluoride assay and isotopic standard. Partial funding for the project is provided by the NRC. The new standard will be a 4.5% enriched uranium isotopic and assay standard to replace out-of-stock CRM 113. The new CRM will serve the safeguards measurements needs of domestic customers at the Gaseous Diffusion Plants and also the reference materials needs for DOE activities associated with the blend-down of Russian uranium weapons materials. The status of this project is discussed in the report on Initiation of Certification of CRM 113-B, 4.5%-Enriched UF_6, on page 53 of this document.

Projects are under way to certify the following uranium isotopic solution standards: NBL CRMs U0002-A, U007, U010-A, U015-A, and U930-D. These projects are discussed in the report on Preparation and Certification of Uranium Isotopic Reference Materials on page 49 of this document. Since the bulk materials of CRMs U010-A and U930-D were expended, certification measurements were made on these materials by using existing uranium isotopic standards as bracketing standards in mass spectrometry measurements so that the CRMs could be issued early in FY 1997. However, all of these CRMs will eventually be recertified by using gravimetrically prepared uranium isotopic calibration mixes.

Planning is in progress to certify and issue the following uranium isotopic standards: NBL CRMs U010-B and U930-E. These standards, 1% and 93% ^{235}U-enriched oxides, respectively, will be packaged as 1 g/unit. There is clearly a need for these standards, and NBL currently has no stock of these isotopic reference materials for sale.

Planning is also under way to improve uranium isotopic measurements for certification. "Absolute" isotope abundance measurements will be determined by preparing synthetic gravimetric mixtures of $^{235}U/^{238}U$ from isotopically pure and well-characterized ^{235}U and ^{238}U end members. These mixtures would then be used for calibrating the mass spectrometer. Project progress will be reported in FY 1997.

In FY 1995, NBL reported problems with NBL CRM 144, the plutonium triple-atom spike standard, and CRM U930, the enriched uranium isotopic standard. These CRMs were removed from the NBL Reference Materials Catalog. Letters that discussed the problems associated with these materials and offered replacements were sent to customers. The problem areas are being addressed, and the CRMs will be available for sale next year.

REFERENCE MATERIALS SALES AND SHIPPING

M. Clapper, U.I. Narayanan, and J.B. Jeans

New Brunswick Laboratory (NBL), as the U.S. government's nuclear certified reference materials (CRMs) laboratory, provides for the sale of CRMs to meet the needs of the nuclear research and safeguards communities. To address problems in past years, NBL dealt with transportation issues, acquired certified shipping containers, and conducted an ongoing search for containers to transport specific CRMs to its foreign customers in order to improve its response to customer orders.

In fiscal year (FY) 1996, 52 different CRMs were available for sale. Purchasers of CRMs represented U.S. Department of Energy (DOE) contractors, U.S. Nuclear Regulatory Commission licensees, U.S. academic institutions, other U.S. customers, and facilities in foreign countries. A total of 64 orders (7 foreign and 57 domestic) for 216 units (10 foreign and 206 domestic), valued at $108,472, were shipped. Figure 1 depicts sales of CRM units for 1996.

Significant changes with regard to shipping NBL CRMs have occurred over the years. Domestic shipments requiring a Type A container have not posed a problem. However, Type B shipments are inherently more complicated. Domestic Type B shipments containing less than 20 curies of solid material may be shipped in a 6M container. NBL sent four 55-gallon (gal) 6M containers, whose certificates had expired, to FBF Nuclear Containers Company in Knoxville, Tennessee, for recertification in FY 1996. The containers will be used to ship the first enrichment level of the recently packaged highly enriched uranium (HEU) nondestructive assay (NDA) standards from NBL to a temporary storage facility. In addition, NBL purchased new 10- and 30-gal 6M containers for shipping smaller quantities of Type B domestic CRM orders (most typical of NBL sales). The first 30-gal 6M container was used in December 1996 for a shipment of plutonium sulfate tetrahydrate to a customer in Meriden, Connecticut.

Although the 6M containers have basically solved NBL's domestic shipping problems, shipping nuclear materials to foreign countries remains difficult. In many instances, containers certified for use within the United States are not certified for shipment of nuclear materials outside the United States, and vice versa. Two exceptions are the PAT-2 and Amersham 0666AW containers. The PAT-2 is an air-transportable plutonium container. This container will be useful for transporting all plutonium CRMs with the exception of CRM 126 (Pu metal), which is packaged in a 5- to 6-inch (in.) glass tube. (The internal C-1 capsule of the PAT-2 container is approximately 3 in. high.) NBL loaned out its entire inventory of PAT-2 containers and is now in the process of retrieving them to fill foreign plutonium orders for CRMs 136, 137, and 138.

Other viable container options were sought. DOE's Chicago Operations Office, Technical and Administrative Services Group, was consulted; it recommended the Amersham 0666AW container. However, this container has only limited availability. It was designed to fill Amersham's own shipping needs and is manufactured by a company in the United Kingdom. Amersham currently leases the container. NBL will pursue the purchase of at least one container and will also lease containers, as available, to decrease the backlog of pending foreign shipments. The Amersham container will solve many of NBL's Pu metal shipping problems (except for shipment to Canada, because Canada has not yet certified the container for Pu metal).

Certificates for the SAFPAK and SAFKEG containers, which NBL also possesses, still have not been revalidated in the United States. The U.K. certificate for the SAFKEG was expected to be issued by the end of 1996. The package will still need to be recertified by the U.S. Department of Transportation, and there is no indication as to how long this process could take. The SAFPAK certificates have not been revalidated in

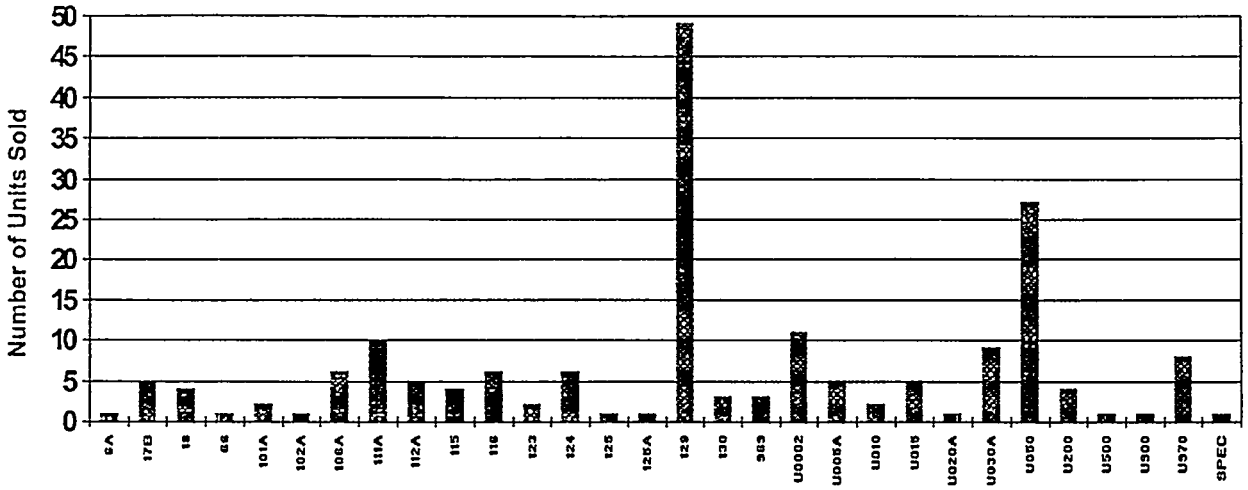


FIGURE 1 Sales Tracking of Certified Reference Materials for FY 1996

the United Kingdom. The recertification process has historically been lengthy because of the multiple reviews and various groups involved.

Annual recosting, approved by DOE-CH, allows for an annual inflation adjustment for reference material prices from the date of certification to present. The FY 1997 price list was prepared to reflect current prices. This price list, effective through September 30, 1997, appears in the FY 1997 catalog and is currently available to customers. The NBL CRM catalog is regularly updated to include several CRMs as well. The latest updated catalog was available for February 1997 distribution.

The complete array of NBL CRMs and ordering information are also now listed in the DOE Isotope Production & Distribution (IPD) catalog (distributed October 1996). This catalog is distributed internationally. NBL had not fully used this form of advertisement in the past, because NBL had been referenced only in the introduction, where it was listed as an additional resource to supply "allied products." The IPD catalog was also posted to the World Wide Web in late November 1996. Prospective customers may browse the Web site to obtain information on available CRMs and instructions on contacting the reference materials (RM) sales office to place an order. This Web site makes NBL CRM information more accessible than ever before. The Web address is http://www.ornl.gov/isotopes/catalog.htm.

NBL has designated workroom D126 as the new packaging area. The workroom is located down the hallway from the RM sales office. It will allow more efficient interaction among RM team members, result in decreased processing and handling time, and provide a larger, dedicated work area for handling the bulky containers and packaging supplies transferred from the NBL fabrication area. A facsimile ("fax") machine was ordered for the RM sales office to facilitate the receipt and handling of customer requests.

NBL continued discussions with NN-40 (Office of Arms Control and Nonproliferation in DOE's Office of Nonproliferation and National Security, which is responsible for granting final determination on contract fulfillment to foreign destinations). The intent was to isolate and alleviate bottlenecks in the process. Several suggestions were implemented. The result has been a smoother, more efficient line of communication (both offices found e-mail to be preferable), which, in several cases, has led to faster turnaround of approvals to ship foreign CRM orders. Attempts to further streamline this process will continue.

PREPARATION AND EVALUATION OF NONDESTRUCTIVE ASSAY STANDARDS FOR URANIUM GAMMA-RAY SPECTROSCOPY

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P.B. Mason, I.W. Frank, M.M. Smith, and C. Thompson

INTRODUCTION

New Brunswick Laboratory (NBL) functions as the U.S. certifying authority for nuclear reference materials and also provides site-specific safeguards assistance to U.S. Department of Energy (DOE) facilities. The waste management division of the Lockheed Martin Y-12 Plant requested that NBL prepare standards, traceable to the national measurement base, for both uranium assay and isotopic composition, for use in calibrating nondestructive assay (NDA) gamma-ray spectroscopy instrumentation. The Y-12 Plant requested three sets of standards:

1. Standards prepared from NBL certified reference material (CRM) U005A (nominally 0.5% ^{235}U) containing 10 different quantities of uranium ranging from 0.1 to approximately 7.5 grams (g),

2. Standards prepared from NBL CRM U930-D (nominally 93% ^{235}U) containing 11 different quantities of uranium ranging from 1.0 to 20 milligrams (mg), and

3. A group of standards containing 1 g U prepared from NBL CRMs with the following nominal ^{235}U enrichment levels: 0.02%, 0.5%, 1.5%, 3%, 5%, 10%, 35%, 50%, 75%, 85%, 90%, and 93%.

The NDA gamma standards prepared by NBL are fully traceable: CRMs were used as the source of uranium, isotopic verification and uranium chemical assays were performed by following strict protocols that ensured traceability, and all mass measurements were performed on balances calibrated with National Institute of Standards and Technology (NIST) traceable mass standards and for which strict quality assurance/quality control (QA/QC) procedures were followed.

The gamma standards prepared for Y-12 are relatively small, have low self-attenuation, are rugged enough to be placed in various matrices and geometries, and are reusable in new matrices/geometries. By following the "add-a-source" philosophy, Y-12 will maintain the capability to add known standards to the matrix and geometry of their choice and, in so doing, will create a set of standards that closely, if not exactly, mimics their samples in matrix and geometry. The result will be a high level of assurance in the quality of the measurements performed by an assay system calibrated by using these standards.

METHODOLOGY FOR STANDARDS PREPARATION

Containment

When methods for packaging the following nuclear material were being considered, three issues were addressed:

1. Self-absorption of the uranium gamma-ray emissions were minimized in the standards to ensure a valid calibration of the NDA instrumentation.

2. The nuclear material was doubly encapsulated to ensure containment.

3. Containment materials (primary and secondary containers, etc.) were made from low-atomic-weight materials (low Z) to minimize gamma-ray attenuation by the packaging.

Since self-absorption of gamma-ray emissions increases as uranium becomes more densely packed, a diffuse geometry for the uranium was necessary. A design was formulated that essentially fixed the uranium into the shape of a cylinder, leading to low uranium density and therefore low self-absorption. To achieve a cylindrical geometry, the uranium (added as a uranyl nitrate solution) was initially absorbed into a thin, flexible substrate. The substrate used was a filter paper composed of a borosilicate fiber woven into sheets with a polyester backing to increase its strength. The borosilicate fibers in the substrate held the uranyl nitrate within the weave. As an additional precaution, a thin layer of polyester (1 mil) with an adhesive backing was attached to the substrate. This fixed media provided added assurance that the uranium would remain within the substrate.

Aliquants of the uranium solution were dripped onto the substrate by using a squeeze bottle with a fine delivery tip in a manner that provided a relatively uniform distribution of uranium on the substrate. The substrate was allowed to air dry for a minimum of four days, after which Krylon® Crystal Clear, a spray enamel, was used to fix the uranium onto the substrate. The substrate was then folded over so that only the Mylar® backing material was exposed. It was placed between two sheets of plastic, and the plastic sheets were laminated together. The product constituted the primary container for the uranium. The plastic laminate was composed of polypropylene on the inside (the uranium-containing side) and a polyester coating on the outside. The laminated substrate was rolled up and inserted into the secondary container so that the uranium-containing paper essentially formed a cylinder around the inside of the secondary container.

The secondary containers vary in size, from 10-milliliter (mL) Teflon® Oak Ridge centrifuge tubes to 500-mL Nalgene® polyethylene bottles (leakproof). The proper dimensions of substrate that would fit into each tube size were determined. When the amount of uranium required for a standard exceeded the limit on a particular tube, the next largest-sized secondary container was used.

General Dissolution and Analysis Procedures

Thirteen different NBL CRMs were used as the source of uranium for the standards. All of the CRMs were uranium oxide (U_3O_8) powders and were certified for isotopic composition. The uranium oxide powders were dissolved in Teflon beakers in a minimum amount of Baker Ultrex II® Ultrapure nitric acid. After total dissolution, the solutions were brought to dryness and then redissolved in distilled water. Two additional drying/redissolving steps were performed to ensure removal of all excess nitric acid. Thermal Ionization mass spectrometric measurements on aliquants of the solutions were performed to verify the isotopic integrity of the solutions, thus assuring the validity of the certified values.

Chemical analyses to determine the elemental uranium concentration of the solutions used in the preparation of the standards were performed by using the NBL titrimetric method. This method has been shown to produce results with a relative deviation of within ±0.10% for a single observation, based on a 95% confidence interval. The titrant factor (in units of mg U/g titrant) was previously determined by titration of two sets of five

standards of known uranium content prepared from CRM 112-A, the uranium (normal) metal assay standard, providing traceability of the method to the national measurement base. Statistical plans of analyses were prepared before the determination of each solution concentration began.

Gamma Measurements of Standards

Measurements were performed on the Y-12 standards to measure transmission coefficients and validate the linearity of the standards. In all cases, the experimental setup included a high-purity Ge detector, a Canberra spectroscopy amplifier model 2020, an ND 581 analog-to-digital converter, and a Canberra 3105 high-voltage power supply. All electronics were connected to a μVax workstation by an ND 556 acquisition interface module. All measurements were performed in a lead-lined sample cave, and all results were background corrected.

Transmission measurements were taken through the laminated standard sheets (before insertion in the cylinder). Each individual standard was placed between the detector and a ^{169}Yb source in a geometry such that only gamma rays passing through the standard would impinge upon the detector. The ratios of the attenuated 177-kiloelectron volt (keV) and 198-keV count rate to the unattenuated 177-keV and 198-keV count rate gave the transmission of gamma rays at 177 keV and 198 keV. These transmission values were then interpolated to give the transmission at 185.7 keV. Only the 0.5% standards' transmission measurements were determined, because this set included all five different cylindrical geometries and the complete uranium mass range. The average transmission of the uranium material and the paper was calculated to be 0.9218 ±0.015. In addition, a blank standard with no uranyl nitrate solution dripped onto it was counted to calculate the transmission of the paper alone. The paper was found to have a transmission of 0.9691 ±0.0018. From the blank and standard transmissions, the average transmission through the uranium material alone was calculated to be 0.9512 ±0.015. Statistical analysis of the data suggests that the correlation between uranium area density and transmission is minimal and that a uniform correction factor of 1.026 ±0.009 can be used for all standards. This factor would be necessary to correct the calibration of a system used to assay samples not contained in polypropylene cylinders.

The linearity of the detector count rate to the mass of ^{235}U was determined for each of the three sets of standards. Linearity for each set was excellent ($R > 0.998$). Data analysis performed on the three combined sets of standards showed a slope, m, of 0.8627 ±0.0014 and an intercept, b, of 0.0 ±3.4 for the line of best fit, with a correlation coefficient, r, of 0.99985. These measurements strongly suggest the independence of the ^{235}U enrichment used in creating the standards and the independence of the gross geometry between standards (in the far-field approximation). Figure 1 shows a close-up of the mass range between 0 and 40 mg U, where most of the data reside.

CONCLUSION

NBL set out to produce fully traceable uranium gamma-ray spectroscopy standards that had low self-attenuation and were small and rugged enough to be added to representative matrices. A gamma-ray analysis of the standards demonstrates that these demands have been successfully met, and a count rate independent of enrichment and geometry has been demonstrated. Overall, the design offers great flexibility as a gamma-ray spectroscopy standard.

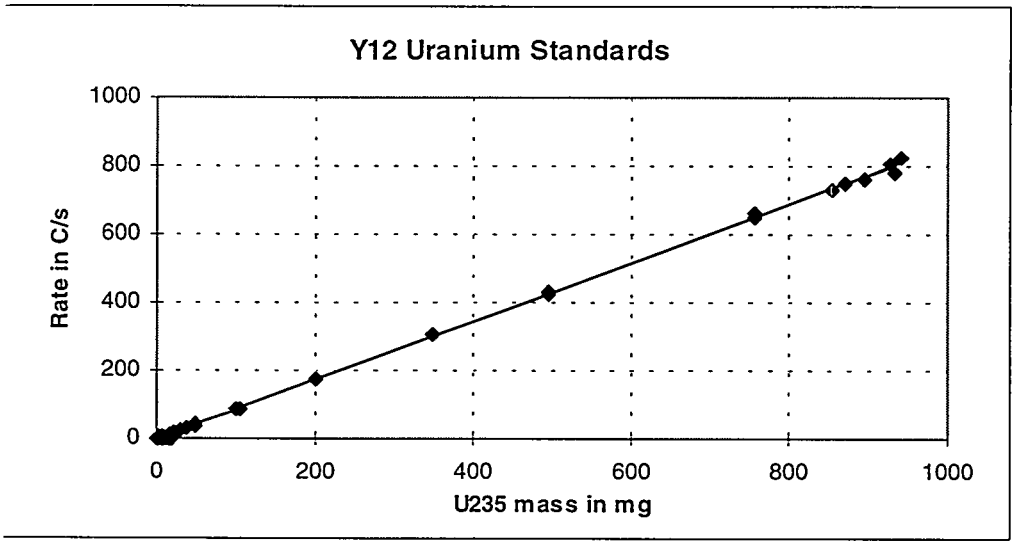


FIGURE 1 Close-Up of Mass Range between 0 and 40 mg U

NEW HIGHLY ENRICHED URANIUM NONDESTRUCTIVE ASSAY STANDARDS: A BEGINNING

D.T. Baran and U.I. Narayanan

INTRODUCTION

There is a clear and immediate need for highly enriched uranium (HEU) gamma-ray nondestructive assay (NDA) standards both internationally and domestically. New Brunswick Laboratory (NBL) has been asked by the Russian/Newly Independent States Nuclear Material Security Task Force in the U.S. Department of Energy (DOE), Office of Nonproliferation and National Security, Office of Arms Control and Nonproliferation (NN-40), to prepare a suite of more highly enriched gamma standards (i.e., spectrometry measurement standards more enriched than those presently available as certified reference material [CRM] 969, a suite of 0.3%, 0.7%, 1.9%, 2.9%, and 4.5% ^{235}U). The ^{235}U enrichments requested for use by the Former Soviet Union states of Ukraine and Kazakstan would be approximately 20%, 55%, and 93%. The proposed suite of standards, coupled with the existing NBL 969 suite, would provide facilities with NDA standards spanning the entire range of ^{235}U enrichment, from depleted to 93%. In addition, the new suite of standards would complete the required calibration range for NDA systems, resulting in strengthened and standardized material control and accountability NDA measurements throughout the DOE weapons complex and the world.

NBL proposes to assemble 20 sets of the HEU standards. Each enrichment level would be prepared in an aluminum cylinder, approximately 9 centimeters (cm) tall and 3.5 cm in radius. Each standard would have an aluminum plug, approximately 6.6 cm tall, fitting into the cylinder, compressing approximately 230 grams (g) of U_3O_8 powder to a density of 5.0 g per cubic centimeter. The aluminum bottom wall would have a thickness of 2 millimeters (mm). The known thickness of the bottom, coupled with the U_3O_8 powder compacted in the bottom, would provide an "infinitely thick" geometry. This geometry would permit NDA instruments to be calibrated by relating the 185.7 kiloelectron-volt (keV) gamma-ray count rate to ^{235}U enrichment.

PROGRESS TO DATE

NBL has secured a source of U_3O_8 powder with the appropriate enrichments from the Y-12 Plant in Oak Ridge, Tennessee. The first shipment of material, consisting of approximately 5 kilograms of 20% enriched material, arrived at NBL in early fiscal year (FY) 1997. Subsequent shipments will occur on a "just-in-time" basis to satisfy nuclear material category restraints at NBL.

NBL has secured 150 Al cans and plugs for use in the project from Frederick Manufacturing in Frederick, Maryland. These cans have undergone quality control tests by Frederick Manufacturing, showing that the bottom can thickness (the critical dimension) is 0.1999 ±0.00024 mm. This limit is well within the required tolerance of ±0.02 mm.

SAMPLING AND ANALYSIS PLAN

The preliminary plan for sampling (for any one enrichment) is to pull a 3-g sample for and from each standard unit fabricated, so a total of 20 samples is pulled during the filling procedure. These samples would then be used to evaluate material properties as described below and also to perform certification measurements.

The preliminary plan for analysis is to use these 3-g samples to determine (1) the water, volatile impurities, and uranium content of six samples by using the NBL high-precision titrimetric method; (2) the impurity content of two samples by using inductively coupled plasma (ICP)-atomic emission spectrometry (AES) and/or ICP-mass spectrometry (MS) techniques; (3) the radioactive impurities, specifically ^{232}U, ^{233}U, and ^{237}U, of one sample by using a high-resolution Ge gamma-ray spectroscopy system; and (4) the isotopic abundance measurments (certification) of 12 samples by using the thermal ionization MS technique.

The fabricated standards will be tested and evaluated to provide additional data on the performance of the standards. For each enrichment, all 20 standards will be tested for isotopic homogeneity to assure that the 185.7-keV gamma-ray area in each standard counts identically, within statistical limits, to all the other 185.7-keV gamma-ray areas of the enrichment. The data from the 185.7-keV gamma-ray areas for the three enrichment standards will be pooled to show linearity of the assembled sets. This data will then provide assurance that the area of the 185.7-keV peak is directly proportional to the count rate of the detector system, given identical detector-sample geometries. Finally, four standards per enrichment will be tested for the uniformity of U_3O_8 area density by measuring the transmission of the ^{137}Cs 661-keV gamma ray through five different points of the standard.

Fabrication and certification of the suite is expected to be completed in late FY 1997. Sixteen sets are set aside for domestic and Former Soviet Union customers, and the remaining four sets will be available for sale.

PREPARATION AND CERTIFICATION OF URANIUM ISOTOPIC REFERENCE MATERIALS

P.M. Santoliquido, A.M. Voeks, P.V. Croatto, A.J. Traina, F.E. Jones, P.B. Mason, I.W. Frank, K.D. Johnson, F.P. Orlowicz, and M.M. Smith

New Brunswick Laboratory (NBL) provides a suite of certified uranium isotopic reference materials that span enrichment levels from depleted to highly enriched. Each unit consists of a nominal 1 gram (g) of uranium in the form of a highly purified U_3O_8 powder.

In fiscal year (FY) 1995, a decision was made to replace the solid oxide form with a uranyl nitrate solution that has a nominal concentration of 1 milligram (mg) U/g solution. This change has several advantages. The original unit of 1 g U/g soliction provided much more uranium than was needed for routine isotopic analysis. This single unit would thus remain in use for an extended length of time, necessitating multiple subsampling and creating a risk of cross-contamination. The new, smaller unit provides a more appropriate quantity of uranium, resulting in less waste. Finally, the liquid form in the prepared concentration provides a ready-to-run preparation for thermal ionization mass spectrometry (MS).

Currently there are four new solution-form uranium isotopic reference materials in various stages of the preparation and certification process. Certified reference material (CRM) U930-D (nominally 93% ^{235}U) and CRM U010-A (nominally 1% ^{235}U) are near the end of the process at the statistical evaluation step, while CRM U007 (normal enrichment, nominally 0.7% ^{235}U) and CRM U0002-A (nominally 0.02% ^{235}U) are still at the beginning of the process, having only been prepared and packaged.

The preparation procedure for CRM U930-D and CRM U010-A has been reported on previously (1,2). CRM U007 was prepared from NBL CRM 112-A, the uranium metal assay standard. A piece of metal was dissolved in nitric acid and diluted to a nominal concentration of 1 mg U/g solution. CRM U0002-A was prepared from CRM U0002, one of the suite of U_3O_8 powder certified isotopic reference materials. A portion of the material was dissolved in nitric acid and diluted to a nominal concentration of 1 mg U/g solution. An automated ampulator-sealer was used to package both CRM solutions into 5-milliliter ampules with break-off tips for ease of use.

The certification analysis plan for CRM U930-D followed a statistical plan designed by the NBL Numerical Analysis Group. Two analysts performed a full isotopic analysis by thermal ionization MS by using two Finnigan™-MAT 261 instruments equipped with 13-sample turrets. In accordance with a statistically designed sampling plan, 10 ampules of CRM U930-D were selected for MS analysis. Of these, five ampules were assigned to each mass spectrometrist. Each mass spectrometrist analyzed two samples from each of the five assigned ampules and one sample from each ampule assigned to the other mass spectrometrist. The mass discrimination factor was determined by using NBL CRM U500; CRM U930 was used for quality control. Each turret included three CRM U500 aliquants, five CRM U930 aliquants, and five CRM U930-D aliquants.

The certification analysis for CRM U010-A followed a similar plan. However, in this case, the mass discrimination factor was determined by using NBL CRM U030, and CRM U010 was used for quality control. Each turret included three CRM U030 aliquants, five CRM U010 aliquants, and five CRM U010-A aliquants.

The goal of the statistical evaluation of the certification data is to provide certified values for the atomic percent and weight percent of ^{234}U, ^{235}U, ^{236}U, and ^{238}U and the uncertainties for each value. Statistical evaluations of the data for both CRM U930-D and CRM U010-A are in progress.

The certification analyses of CRM U007 and CRM U0002-A and subsequent statistical evaluations of data will be completed in FY 97.

REFERENCES

1. P.M. Santoliquido et al., "Preparation and Certification of CRM U930-D: Status Report," in NBL-335, p. 15 (1996).

2. P.M. Santoliquido et al., "Preparation and Certification of CRMU010-A: Status Report," in NBL-335, p. 16 (1996).

PREPARATION AND CERTIFICATION OF CRM 145 URANIUM (NORMAL) ASSAY SOLUTION STANDARD

P.B. Mason

INTRODUCTION

New Brunswick Laboratory (NBL) prepared and certified the uranium assay solution standard, certified reference material (CRM) 145. This standard provides a convenient and easy-to-use source of uranium for the calibration and quality control of uranium measurement instruments and systems. Additionally, the quantity and form of uranium provided with each unit of CRM 145 will reduce the wastes associated with the use of conventional uranium CRMs.

CRM 145 consists of a solution of normal uranyl nitrate in glass ampules. Each ampule contains approximately 200 milligrams (mg) of uranium in a nominal volume of 20 milliliters (mL) of 1 *M* nitric acid. The certified uranium concentration is 10.1356 ±0.0011 mg per gram of solution. The certified value is believed to lie in the uncertainty interval with a level of confidence of approximately 95%. A total of 482 ampules were manufactured, and 459 are available for sale.

PREPARATION AND CERTIFICATION

The preparation of CRM 145 was completed in fiscal year (FY) 1995, and a detailed report on its production may be found in NBL's 1995 Progress Report, NBL-335. In summary, CRM 145 was made by dissolving and diluting a quantity of CRM 112-A (uranium metal) in nitric acid and distilled, deionized water. The resulting solution was divided into aliquants and flame-sealed in glass ampules. A total of 20 units were removed for verification of the certified uranium concentration and confirmation of the relative atomic weight.

The certified uranium concentration is 10.1356 ±0.0011 mg per gram of solution. The certified value is believed to lie within the uncertainty interval with a level of confidence of approximately 95%. The certified uranium concentration value was based on the calculated preparation value and confirmed by the NBL titrimetric method. This value was calculated from the mass of high-purity CRM 112-A metal dissolved and diluted to a known mass of solution, taking into account the metal purity and buoyancy corrections. The certified relative atomic weight for CRM 145 is 238.0289. This value was based on the CRM 112-A certification measurements and verified by isotope ratio measurements determined by using thermal ionization mass spectrometry.

The uncertainty applied to the certified value was calculated according to the International Organization for Standardization (ISO) standard by using the methods described in its *Guide to the Expression of Uncertainty in Measurement* (ISBN 92-67-10188-9, 1st edition, Geneva, Switzerland, 1993). The combined standard uncertainty for the CRM 145 preparation steps was determined and represents the combined effect, at the level of one standard deviation, of uncertainty components associated with gravimetric, purity, and buoyancy correction factors. The combined standard uncertainty was then multiplied by a coverage factor of 2 to yield an approximation of the 95% confidence interval. The results are summarized in Table 1.

TABLE 1 Final Values Associated with the Issuance of CRM 145

Input Quantity (sources of uncertainty)	Method Used to Evaluate Uncertainty of Input Variable	Uncertainty of Input Quantity	Sensitivity Factor	Output Uncertainty
Mass of uranium metal	Historical performance of balance taken from Balance QA database	2.812×10^{-4} g	9.5532×10^{-5}	2.686×10^{-8}
Mass of solution	Historical performance of balance taken from Balance QA database	0.42049 g	1.0137×10^{-2}	-4.073×10^{-7}
Mass of empty bottle	Historical performance of balance taken from Balance QA database	0.10656 g	1.0138×10^{-2}	1.032×10^{-7}
Purity of CRM 112-A metal	Certificate of analysis	0.000030612	-9.686x10-7	3.103×10^{-7}
Buoyancy correction	Estimated, based on reasonable estimates of temperature, pressure, and humidity	7.487×10^{-7}	9.686×10^{-7}	7.589×10^{-9}
Combined standard uncertainty (mg U/g solution)			0.000523	
Coverage factor, k (to approximate 95% interval of confidence)			x2	
Expanded uncertainty (mg U/g solution)			0.0011	

INITIATION OF CERTIFICATION OF CERTIFIED REFERENCE MATERIAL 113-B, 4.5%-ENRICHED UF_6

M.I. Spaletto and U.I. Narayanan

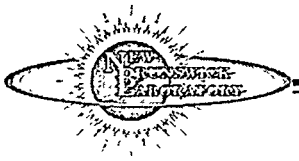
Since 1977, New Brunswick Laboratory (NBL) has had available for sale the uranium and ^{235}U standard, certified reference material (CRM) 113 ("Uranium [Enriched] Hexafluoride — UF_6 in Solid Form"). This CRM originally consisted of a low-enriched (1.7% ^{235}U) material packaged in P-10 tubes that contained approximately 10 grams (g) of UF_6 each; this is an amount suitable for performing gravimetric analysis, the method of choice for this material. Over time, this CRM was sold. Plans for replacement began in fiscal year 1996.

The primary customers identified for this CRM are the Portsmouth and Paducah Gaseous Diffusion Plants (GDPs). These two GDPs are in the process of converting from U.S. Department of Energy (DOE) contractor sites to Nuclear Regulatory Commission (NRC) licensees. Because of the change in their status, certification of this CRM is being jointly funded by DOE's Office of Nonproliferation and National Security, Office of Security Affairs, Office of Safeguards and Security (NN-51), and by the NRC.

To minimize the packaging effort, the CRM will be certified as packaged in 2-S cylinders, which hold approximately 1.8 kilograms (kg) of UF_6. The GDPs have the ability to subsample these cylinders for both gravimetric and enrichment samples. For the benefit of other national and international customers that do not have this capability, NBL will subsample the material into P-10 tubes as before, with an added charge for the repackaging.

The ^{235}U enrichment level of the new CRM was increased from 1.7% to approximately 4.5%. This higher enrichment was chosen to approximate the Russian UF_6 blend-down material receipts anticipated by Portsmouth GDP. This CRM will therefore serve the measurement needs at GDP and also assist in resolving any possible shipper-receiver differences.

A suitable base material was identified at Portsmouth GDP. NBL and U.S. Enrichment Corporation, the oversight body for the GDPs, signed a contract under which Portsmouth GDP was to supply the base material, package it into 2-S cylinders, and withdraw analytical samples for the certification. By the end of FY 1996, packaging (of 15 2-S cylinders; 1.8-kg material/cylinder) was completed, and the certification samples (60 10-g units in P-10 tubes) had been received by NBL. In FY 1997, certification measurements for both percent uranium and enrichment will be performed, a statistical evaluation will be completed, and a certificate of analysis will be issued.



6 SAFEGUARDS ASSISTANCE

SAFEGUARDS ASSISTANCE PROGRAM

M.A. Legel and W.G. Mitchell

SUMMARY

The New Brunswick Laboratory (NBL) Safeguards Assistance Program provides support for nonproliferation activities, both domestic and international. The expertise of NBL's scientific and technical staff is used to develop U.S. Department of Energy (DOE) materials control and accountability (MC&A) policies that support the safeguarding of nuclear materials at both domestic and international nuclear facilities. Site-specific assistance is provided to DOE facilities to address specific problem areas or improve safeguards for special nuclear materials. NBL is uniquely capable of providing federal employees to evaluate contractor, licensee, or foreign facility safeguards systems and performance.

During fiscal year (FY) 1996, seven NBL personnel participated as team members in seven periodic safeguards and security surveys conducted by four DOE operations offices. Site-specific assistance was provided to facilities through the Rocky Flats Field Office and Richland Operations Office. NBL staff participated in a DOE-directed Inventory Discrepancy Anomaly Resolution Team (IDART) at the Mound Facility. DOE headquarters support was provided through participation in voluntary standards organizations, the American National Standards Institute (ANSI), American Society for Testing and Materials (ASTM), International Organization for Standardization (ISO), and DOE metrology committees. In addition, NBL staff have actively participated in the meetings of the Fissile Materials Assurance Working Group in the Office of Safeguards and Security (NN-51), part of the Office of Security Affairs (NN-50) in DOE's Office of Nonproliferation and National Security. Two NBL personnel also provided Nuclear Regulatory Commission (NRC) inspection assistance and trip reports for three licensees. One NBL staff member provided support to the Carlsbad Area Office for the Waste Isolation Pilot Plant (WIPP) waste characterization activities. In addition, NBL continued to assist the Central Training Academy (CTA) in course preparation and presentation of MC&A course lectures.

PROGRAM ADMINISTRATION

The NBL Safeguards Assistance Program strives to optimize the use of NBL's technical expertise in support of DOE missions. The technical staff are experienced in various analytical laboratory techniques and procedures, instrumentation, quality assurance, and material control and accountability functions. Additionally, their participation in safeguards training courses and workshops is encouraged. A DOE CTA course consisting of CTA 139-D and CTA 140-D, Vulnerability Assessments Fundamentals and Overview, was broadcast to students locally. Ten students took the one-day overview course and three students completed the second 40-hour course. NBL personnel have taken advantage of local training workshops and training courses, such as Managing a Training Program and Table-Top Job Task Analysis, offered through the CTA. Other applicable training courses attended by NBL safeguards cadre personnel included Managing Extraordinary Customer Service; the CTA MC&A courses Waste and Residue Nondestructive Assay Measurements (MCA-248), Volume Measurement Techniques (MCA-242), and Introduction to Materials Control and Accountability (MCA-101D); and the IAEA Lessons Learned Workshop. By regularly accessing on-line data, including DOE technical standards, DOE Orders, and the DOE CTA's TRAINERNET and Web page, NBL ensures its resources remain current with DOE policy and guidance publications.

OPERATIONS OFFICE PERIODIC SURVEY ASSISTANCE

As a technical extension of OSS, NBL provided MC&A expertise to the DOE operations offices for periodic surveys of facilities. During FY 1996, seven NBL personnel joined teams to work on seven periodic safeguards and security surveys conducted by four operations offices: Albuquerque, Chicago, Idaho, and Oak Ridge. The facilities surveyed were the Idaho Chemical Processing Plant, Pantex, Los Alamos National Laboratory, Argonne National Laboratory-East, Argonne National Laboratory-West, and the K-25 and Y-12 facilities in Oak Ridge. Periodic survey assistance to operations offices was provided by Dr. Paul Croatto, Dr. Wanda Mitchell, Ms. Margaret Legel, Dr. Jeffrey Zebrowski, Dr. Usha Narayanan, Mr. Peter Mason, and Dr. Patricia Santoliquido.

SITE-SPECIFIC ASSISTANCE

At the request of a DOE operations office, NBL will assist a site by performing analyses to help the site improve its safeguards measurement posture. NBL personnel Dr. H. Rodney Martin and Dr. Wanda Mitchell worked with the Richland Operations Office and Westinghouse Hanford Company to draft performance metrics for material control and accountability. NBL personnel Dr. Wanda Mitchell and Ms. Margaret Legel assisted Rocky Flats Environmental Technology Site safeguards office personnel in their review of requirements for the site's MC&A program. An NBL team consisting of Dr. Wanda Mitchell, Ms. Margaret Legel, and Ms. Maureen Clapper reviewed the Rocky Flats MC&A plan and supporting safeguards accountability manual and provided comments and recommendations to the Rocky Flats Field Office. Assistance was provided by Dr. Usha Narayanan in the choice of reference material for a conversion of accountability methods at the Oak Ridge Y-12 Plant, as follow-up to a periodic survey. Dr. Wanda Mitchell, Mr. Robert Oldham, and Ms. M. Irene Spaletto provided assistance to the Westinghouse Hanford Company's plutonium finishing plant analytical laboratory on destructive analysis of Westinghouse Hanford's portion of samples taken by the International Atomic Energy Agency (IAEA) from materials offered for IAEA safeguards while stored at Westinghouse Hanford Company.

NBL ASSISTANCE TO DOE HEADQUARTERS

NBL personnel participated in meetings and reviewed correspondence for the OSS Fissile Material Assurance Working Group. Dr. H. Rodney Martin served as chairperson of the Technical Solutions Subgroup, Mr. Robert Oldham served as a member of the Measurement Assessment Team, and Dr. Wanda Mitchell served as a member of Facilities Transition and Operations Subgroup. Assistance was provided to NN-513 (the Field Operations Division of DOE's Office of Safeguards and Security) in the identification of safeguards requirements for nuclear materials undergoing stabilization and storage at five DOE contractor sites. NBL staff participated in a DOE-directed IDART at the Mound Facility as a result of a reported inventory difference of tritium.

NBL assisted NN-512 (the Policy, Standards and Analysis Division of DOE's Office of Safeguards and Security) by chairing a writing group for a material protection control and accountability ANSI industry standard. The language of the standard should meet the needs of various nuclear facilities, including domestic DOE facilities, NRC-licensed facilities, and international nuclear facilities. NBL also participated in the Institute of Nuclear Materials Management (INMM)/ANSI 5.2 Subcommittee on Mass Measurements Control, an ANSI/INMM N-15-5.5 Volumetric Measurement Control Committee, the ISO/Technical Committee 85/Standards Committee 5/Working Group 1, and ASTM C-26 Committee on Nuclear Fuel Cycle.

Mr. Robert Oldham attended a DOE Metrology Workshop sponsored by the DOE Technical Standards Program. He will continue to be a member of an interim steering committee tasked with promoting the use of nongovernment (voluntary) standards by DOE metrology laboratories in conformance with federal law (Public Law 104-113, Sec. 12D, March 1996), which mandates the transition from government standards and DOE Orders to recognized voluntary standards such as those approved by ANSI. A plutonium storage vault at NBL was used to test a radiofrequency (RF) tagging system under development by a commercial company, RANDTEC. Use of an RF tagging system that incorporates a tamper indication device (TID) such as a weight cell, fiber-optic seal, or movement detector could minimize time spent on vault inventory activities and reduce the frequency of required inventories.

NBL SUPPORT TO THE CENTRAL TRAINING ACADEMY

NBL personnel helped the CTA prepare and present MC&A course lectures for PHY 130, Basic Survey Course. An iteration of MCA 140, Basics of MC&A Measurements, which was scheduled to be presented at Argonne National Laboratory and use NBL for on-site demonstrations, was canceled. Personnel supporting the CTA course work are Dr. Kenneth Lewis, Mr. Robert Oldham, Mr. Michael Soriano, and Ms. Margaret Legel.

OTHER SAFEGUARDS ASSISTANCE ACTIVITIES

Two NBL personnel provided NRC inspection assistance and trip reports for three licensees in FY 1996. NRC inspection assistance was provided by Dr. David Baran and Mr. Robert Oldham. Dr. Baran evaluated two licensees, both uranium fuel fabricators, in the area of nondestructive assay (NDA) laboratory techniques and methods. The report on NBL Assistance to the NRC for NDA Techniques: A Summary on page 64 of this document provides details on these activities.

NBL provided assistance to the Carlsbad Area Office Technical Assistance Contractor in evaluating NDA techniques and methods to certify the Nevada Test Site's abilities to ship waste to WIPP. NBL participated in six on-site inspections during FY 1996. The report on NBL Assistance to WIPP for NDA Techniques: A Summary on page 62 of this document provides details on these activities.

INTERNATIONAL SAFEGUARDS ASSISTANCE

NBL personnel continued to support international safeguards projects through DOE offices NN-40 (Office of Arms Control and Nonproliferation in the Office of Nonproliferation and National Security), NN-44 (International Safeguards Division of DOE's Office of Arms Control and Nonproliferation), and NE-40 (Office of Nuclear Energy, Science and Technology) for Russia, Kazakstan, and the Argentine-Brazilian Agency for the Control of Nuclear Materials. The report on International Safeguards Assistance on page 60 of this document provides details on these activities.

INTERNATIONAL SAFEGUARDS ASSISTANCE

M.A. Legel and W.G. Mitchell

The New Brunswick Laboratory (NBL) Safeguards Assistance Program provides support for nonproliferation activities, both domestic and international. The expertise of NBL's scientific and technical staff is used to support safeguards programs in the Former Soviet Union states, Brazil, and Argentina through support from the U.S. Department of Energy (DOE), Office of Nonproliferation and National Security, Office of Arms Control and Nonproliferation (NN-40), and also from the DOE Office of Nuclear Energy, Science and Technology, Office of Facilities (NE-40).

NEWLY INDEPENDENT STATES

NBL's role in helping the newly independent states of the Former Soviet Union is to provide technical support to enhance their capabilities to control, account for, and protect nuclear material inventories against the threats of theft, diversion, and sabotage. NBL is to help develop the program plan and provide measurement standards as part of the support activities. Activities under this project support the DOE/Nuclear Regulatory Commission (NRC)/Defense Nuclear Agency (DNA)-approved program plan for technical support in the areas of physical protection and material control and accounting support to the newly independent states of the Former Soviet Union as provided for by the Nunn-Lugar Amendment. Quarterly progress reports were also provided to NN-40.

During fiscal year (FY) 1996, Dr. Wanda Mitchell served as the subject matter expert for material control and accountability (MC&A) for the Kazakstan Program, including leadership of the Aktau/Almaty MC&A team and consultations on analytical equipment for Ulba. An initial site visit to the Aktau and Almaty reactors was made in September 1995, and the trip report was completed in FY 1996. The first program plan for the Aktau and Almaty reactors was prepared, and revisions were made during the year at program planning meetings. NBL started preparations for the highly enriched uranium (HEU) gamma-spectroscopy certified reference material, CRM 146, to be used for system calibration at four Kazakstan sites as well as Ukrainian, Russian, and U.S. laboratories.

LATIN AMERICA

NBL also provides technical support for Latin American safeguards through interaction with the Argentine-Brazilian Agency for the Accounting of Nuclear Materials (ABACC). ABACC is a bilateral organization with oversight for safeguarding nuclear materials in Argentina and Brazil. NBL supports ABACC in establishing and maintaining a quality control and quality assurance program related to analytical work. NBL supports an ABACC program for the preparation of secondary isotopic standards for destructive and nondestructive analysis by characterizing samples and providing statistical evaluation of data. In FY 1996, two sets of UO_2 samples were analyzed and reported to ABACC. Reciprocal visits are also conducted between NBL and ABACC personnel. In FY 1996, Ms. M. Irene Spaletto was awarded an International Atomic Energy Agency (IAEA) Fellowship and spent two weeks at a Brazilian safeguards laboratory, Laboratoria de Salvaguardas (LASAL), in Rio de Janeiro, where she discussed uranium assay analyses and nondestructive assay standard preparation. NBL also hosted an IAEA Fellow, Ms. Sonia Gonçalves, also from LASAL, for two weeks. Monthly accounting reports were prepared and submitted to NN-44 (International Safeguards Division of DOE's Office of Arms Control and Nonproliferation) for Latin American safeguards tasks. A

representative from NBL attended the annual meetings of the permanent coordinating groups of the Argentine Ente Nacional Regulador Nuclear and ABACC held in Washington, D.C.

RUSSIAN TRANSPARENCY PROGRAM

NBL has participated in the HEU Transparency Program since its inception in 1993. Involvement in the program during FY 1996 was expanded from prior years' participation. The HEU Transparency Program is a joint program of the Russia/Newly Independent States (NIS) Nuclear Material Security Task Force in NN-40 and NE-40. The report on NBL Participation in Monitoring Activities for the U.S. Transparency Agreement with Russia on page 67 of this document provides details on these activities.

NBL ASSISTANCE TO WASTE ISOLATION PILOT PROJECT (WIPP) FOR NONDESTRUCTIVE ASSAY TECHNIQUES: A SUMMARY

D.T. Baran

New Brunswick Laboratory (NBL) provided assistance to the Carlsbad Area Office Technical Assistance Contractor (CTAC) in evaluating nondestructive assay (NDA) techniques and methods used at various U.S. Department of Energy (DOE) sites in order to certify each site's abilities to ship waste to the Waste Isolation Pilot Plant (WIPP).

NBL participated in six on-site inspections during fiscal year (FY) 1996: Idaho National Engineering Laboratory, Savannah River Site, Rocky Flats Environmental Technology Site, Oak Ridge Y-12 Plant, Los Alamos National Laboratory, and Hanford Site.

In all cases, the NDA review was only a subset of the entire review process. In general, the inspections consisted of an opening meeting, badging, safety training, personnel interviews, inspection of facilities, documentation review, and final closeout. The inspection team met with on-site management daily for a review of the day's activities. A daily inspection team meeting was also held to discuss the day's activities and plan the next day's activities. Each inspection team consisted of a team leader (CTAC), an NDA expert (NBL), one or two quality assurance experts, and one or two experts on chemical methods and the Resource Conservation and Recovery Act (RCRA). At some sites, various other inspectors, such as state or federal environmental protection agency officials, also witnessed the inspections. These inspections served to benchmark the status of each site in meeting the WIPP criteria for certifying waste for shipment. As such, the inspections were more collegial and information-based than audit- and compliance-based. It is anticipated that each of these sites will undergo an additional audit and seek certification status within the next 24 months so it can ship radioactive wastes to WIPP.

Well-defined criteria are listed for NDA systems in the WIPP waste assay criteria document. Section 9 of the third revision of the waste assay criteria document details the requirements. In general, NDA systems need to be able to quantify their precision and accuracy and the total uncertainty of the measurement. These values can be obtained from detailed studies and statistical evaluation of the system data from replicate measurements of calibration or quality control standards. In addition, the waste assay criteria require each system to have well-defined minimum detection limits and be able to distinguish between low-level waste and transuranic (TRU) waste (nominally, TRU waste exceeds 90 nanocuries per gram) with statistical certainty. The waste assay criteria also require the establishment of a quality control (QC) program for each NDA system, in which control standards can be statistically analyzed and the day-to-day performance of the system can be assured. Also required are 10% replicate analyses, and control checks are to be performed twice a shift. As part of the QC program, each system should have well-documented operating, calibration, and data review procedures.

Methods for determining the total uncertainty of a system's measurement were reviewed extensively and remain difficult to address. Each measurement system must estimate/calculate the total uncertainty of its assay value, including any statistical, and systematic contributions. Since waste drums tend to vary greatly in matrix composition, packing, and mass amounts, it is nearly impossible to estimate the total variation in a system's assay ability by using a subset of analysis data. Other challenges in meeting the waste assay criteria are maintaining thorough QC programs and training and qualification data and records. The traceability of working standards was also of concern to several sites. Many sites lacked the thorough QC programs that should address responses to out-of-control situations, scheduled statistical reviews of QC data, and

development of detailed control charts of all controlled parameters with a calculated mean and calculated 2-sigma and 3-sigma control limits. Operating procedures need to address other specific requirements of the waste assay criteria, such as 10% required replicate analyses.

In general, the site and NDA measurement systems that were reviewed were found to lack most of the required documentation and qualifications. Most NDA measurement systems have part or all of the requirements in place, but the information is not sufficient to meet the detailed WIPP waste assay criteria. These inspections served to identify the weaknesses of the NDA measurement programs and will allow the sites to better concentrate their time, effort, and money to meet the WIPP waste assay criteria.

NBL ASSISTANCE TO THE NUCLEAR REGULATORY COMMISSION (NRC) FOR NONDESTRUCTIVE ASSAY TECHNIQUES: A SUMMARY

D.T. Baran and M.A. Legel

The Nuclear Regulatory Commission, Office of Nuclear Material Safety and Safeguards (NRC-NMSS), has an established material control and accounting (MC&A) inspection program for fuel cycle licensees. New Brunswick Laboratory (NBL) has provided technical assistance to this NRC program since fiscal year (FY) 1990. During FY 1996, NBL helped the NRC evaluate two licensees in the area of nondestructive assay (NDA) laboratory techniques and methods. Both facilities are licensed by the NRC as uranium fuel fabricators. This report summarizes the inspection activities performed and the status of NDA waste measurement systems at the reviewed sites. Violations or inspector follow-up issues are the responsibility of the NRC. The use of appropriate traceable calibration standards in NDA measurement systems is of particular concern to NBL, which is the federal certifying authority for nuclear reference materials.

NBL assisted NRC inspectors in two on-site inspections during FY 1996. Dr. David T. Baran was the NDA technical expert from NBL. For both facilities, the NDA review was only a subset of the entire review process performed by the NRC inspectors. The inspections consisted of an opening meeting, badging, safety training, personnel interviews, inspection of facilities, documentation review, and a final closeout meeting. The inspection team met with on-site management daily for a review of the day's activities. A daily inspection team meeting was also held to discuss the day's activities and to plan the next day's activities. Each inspection team consisted of a team leader from the NRC, the NDA expert from NBL, and one junior inspector-in-training from the NRC. These inspections evaluated the effectiveness of nuclear material inventory control programs to verify their compliance with NRC regulations and the licensee's Fundamental Nuclear Material Control Plan (FNMCP) by specifically reviewing the facilities' NDA accountability measurement systems.

SITE 1

Site 1 employs three NDA measurement systems for accountability nuclear material measurements. The first system is a standard safeguards assay meter (SAM-II). It uses a sodium iodide detector to assay laundry before it is shipped off site. For calibration, this system uses standards similar but not identical to the laundry packages. The system calibration relates count rate to ^{235}U mass. The difference between the standards and the unknowns will slightly bias the measurements, but not by enough to be a concern. Dr. Baran found the system to have a well-written procedure, well-documented control limits and out-of-control responses, and an adequate operator training and qualification program. The working standards that are used, although not primary standards, were manufactured and measured by using in-house measurement methods that were qualified by using traceable standards.

The second system, a Canberra Q^2 waste assay system consisting of sodium iodide detectors, is used to assay waste barrels of combustible waste before incineration and polypak pails containing the ash from the incineration process. This system uses two sets of standards — one set for waste barrels and one for polypak ash — to calibrate the system, relating count rate to ^{235}U mass. The waste barrel standards are not similar in matrix, geometry, and, possibly, density to the unknowns. In addition, the Q^2 system lacks the sophistication to make matrix effects corrections on measurements. Consequently, measurements made on the barrels of combustible waste are probably biased and not of particularly good quality. However, this measurement is not the accountability measurement and serves only as a screening process for material destined for the incinerator. The polypak ash standards are identical in density, matrix, and geometry to the unknowns.

Therefore, the accountability measurement of the polypak ash is of good quality. Dr. Baran found the measurement system to have a well-written procedure, well-documented control limits and out-of-control responses, and an excellent operator training and qualification program. The working standards that are used, although not primary standards, were manufactured and measured by using in-house measurement methods that were qualified by using traceable standards.

The third system is a Canberra segmented gamma scanner (SGS) system consisting a germanium detector, transmission source, turntable, and Canberra-supplied software for matrix corrections of measurements. The SGS is used to assay waste barrels full of noncombustible waste or high-density filters and mop heads before burial. This system uses a well-defined set of calibration standards, relating the count rate to the mass of ^{235}U. In general, the standards are not similar in matrix and density to the unknowns; however, the system uses standard matrix effect corrections on the measurements, so the measurements made by the SGS are of good quality. Dr. Baran found the system to have a well-written procedure, well-documented control limits and out-of-control responses, and an excellent operator training and qualification program. The working standards that are used, although not primary standards, were manufactured and measured by using in-house measurement methods that were qualified by using traceable standards.

SITE 2

At Site 2, two NDA measurement systems are used for accountability measurements: a standard SAM-II assay counter and a germanium detector enrichment meter.

The SAM-II system consists of a single sodium iodide detector connected to the standard electronics and software package available on the purchased equipment, as developed at Los Alamos National Laboratory. The system is used to assay the quantity of ^{235}U in samples. The software package controls the selected regions of interest (ROIs) and the applied voltage, and it displays the number of counts in the ROI corresponding to the 185.7-kiloelectron-volt ^{235}U gamma-ray peak. The system is located in the incinerator area of the plant and has a restricted geometry (sample turntable and fixed detector-sample distance), ensuring some standardization. The SAM-II is qualified for the assay of high-efficiency particulate air (HEPA) filters, prefilters, and bagged trash. The SAM-II was scheduled to be replaced within 1-2 months with a new, high- purity germanium detector system operated on a personal computer system, including an on-board multichannel analyzer (MCA) and a more sophisticated data analysis software package. The new system will represent an improvement in the technology and measurement capabilities at the facility.

The germanium detector enrichment meter (E-meter) system consists of a single, high-purity germanium detector and cryostat coupled to a personal computer complete with MCA and data analysis software. The system counts standardized samples (cans containing 300 grams of uranium oxide) and relates the measured count rate to the percent of ^{235}U present in the sample. The system is located in the analytical laboratory, and it relies on standardized geometry (fixed detector-sample distance) and a well-shielded counting chamber for reproducibility of results. The system is qualified to assay ^{235}U between 2.5 and 5.0 weight percent.

The traceability of calibration standards and measurement control standards for the two NDA accountability measurement systems was reviewed. The facility's FNMCP stated that all accountability measurement systems must be calibrated by using National Institute of Standards and Technology (NIST)/NBL traceable standards. Dr. Baran found no traceability for the SAM-II system. The standards used to originally qualify the system some time between 1984 and 1986 were no longer available. No documentation on the traceability of the two uranyl nitrate standards used for quality control (QC) purposes was found. There was evidence of progress in improving this situation by using the new high-purity germanium system. Dr. Baran reviewed documentation for the creation of new sets of calibration standards for the measurement system. To obtain

well-documented, traceable working standards, records should include copies of laboratory notebooks used in preparing the samples, any outside contractor mass spectrometry results, and any reports concerning the new standards generated by the central laboratories.

Dr. Baran reviewed documentation on the traceability of the calibration standards for the E-meter system. The standards are located in a controlled area, and copies of the contractor mass spectrometry results are maintained. The documentation for a measurement control standard on file was inadequate. Dr. Baran recommended that the site provide appropriate documentation showing traceability for measurement control standard(s) as well as the qualification standards.

Dr. Baran reviewed the measurement control issues of system calibration, quality assurance (QA), and QC and found that the E-meter system had been qualified in April 1995. A review of the bimonthly QC data revealed that this system appeared to be exhibiting sufficient measurement control data. Dr. Baran recommended, as good laboratory practice, the institution of a periodic requalification. A review of the QA program showed it to be adequate; however, although data are reviewed bimonthly, it is not clear if any further statistical evaluation of the data is performed.

Dr. Baran found that the SAM-II system had been qualified for four different types of materials between 1986 and 1988. No further requalification of the system had been done, in spite of electronic component replacement. This omission was noted to the laboratory staff as a poor laboratory practice. A measurement system should be recalibrated (1) after components have been changed, (2) if there is an unresolvable out-of-control condition, (3) if statistical analysis of the QC data indicates a bias or trend, and (4) at some periodic interval. Dr. Baran recommended, as good laboratory practice, the institution of a periodic requalification. The new high-purity germanium system and new set of standards offer a good opportunity for recalibrating and instituting periodic verifications of calibration.

There were no replicate measurements on samples or standards for either the SAM-II or E-meter systems. A replicate program and the associated statistical review of the data are excellent methods to quantify a measurement system's precision and accuracy.

Dr. Baran reviewed procedures detailing the operation and qualification of the NDA accountability measurement systems. Both systems had adequate procedures documenting the system operation. Neither system had a procedure for calibration, since calibration was not routinely performed. Dr. Baran recommended that the site develop a calibration procedure that will detail the appropriate steps and appropriate calculations and mathematics needed to convert an instrument's response to known characteristic or special nuclear material mass or percentage of ^{235}U.

A review of the training and qualification records of the analysts who use the SAM-II and E-meter was performed. Records for the E-meter were complete and included a written qualification test, the acknowledgment of procedure review/revision, and records of on-the-job training. Dr. Baran did not see similar detailed records for SAM-II analysts; however, the QC coordinator did generate a report showing all qualified analysts and their respective requalification dates.

In conclusion, the NRC licensee facilities reviewed by Dr. Baran demonstrated good measurement capability in the NDA areas. Weaknesses with regard to records and documentation required to support the measurement quality were found. Further attention to detail and to the maintenance of traceability records, calibration records and data, and the statistical evaluation of QC data would enhance the MC&A portion of the NDA accountability measurement systems at these facilities.

NBL PARTICIPATION IN MONITORING ACTIVITIES FOR THE UNITED STATES TRANSPARENCY AGREEMENT WITH RUSSIA

K. Lewis and D.T. Baran

New Brunswick Laboratory (NBL) has participated in the Highly Enriched Uranium (HEU) Transparency Program since its inception in 1993. During fiscal year (FY) 1996, involvement in the program was expanded from prior years' participation, and NBL is expected to continue providing an enhanced level of support to the program. This report summarizes background program information and provides a summary of NBL experiences in monitoring activities.

BACKGROUND AND HISTORY

The Transparency Program is a joint program in the U.S. Department of Energy (DOE) sponsored by the Office of Arms Control and Nonproliferation (NN-40) in DOE's Office of Nonproliferation and National Security (NN) and by the Office of Facilities (NE-40) in DOE's Office of Nuclear Energy, Science and Technology (NE). NN-40 is responsible for transparency policy, acting as the chief spokesperson for transparency matters, and negotiating transparency agreements. NE-40 is responsible for all aspects of implementing transparency.

Transparency is defined as those agreed-upon measures that provide confidence that the arms control and nonproliferation objectives of the HEU Agreement are being met. The major nonproliferation objectives of the HEU Agreement are to provide confidence that the (1) HEU comes from nuclear weapons dismantled in Russia, (2) low enriched uranium (LEU) produced is derived from Russian HEU, and (3) LEU delivered to the United States from Russia is fabricated into fuel for commercial nuclear reactors. Transparency departs from traditional, intrusive methods of verification. Until FY 1997, the focus of the Transparency Program was on measures designed to achieve only the second and third objectives; the approach to achieving the first objective was pursued by NN through other U.S.-Russian Transparency initiatives in the Mutual Reciprocal Inspections (MRI) Agreement. In FY 1997, the focus of the HEU Transparency Program will be on all three objectives.

Table 1 shows the major program milestones that were met, allowing the program to progress to the current level of NBL staff involvement.

TABLE 1 Program Milestones

Date	Event
February 18, 1993	HEU Government-to-Government Umbrella Agreement signed; it provides for the purchase of 500 metric tons of HEU from dismantled nuclear weapons over 20 years
January 1994	HEU Purchase Contract signed at the Presidential Summit in Moscow
March 18, 1994	Protocol of HEU Transparency Arrangements signed in Washington, D.C.; it identifies U.S. and Russian facilities, establishes Transparency Review Committee (TRC) and Annex procedures, and allows deliveries of uranium and payments to begin
August 1996	Permanent Presence Office opened at UEIE

The Russian facilities subject to the HEU Transparency Agreement are:

- Ural Electrochemical Integrated Enterprise (UEIE), Novouralsk, Russia;
- Siberian Chemical Enterprise (SChE), Seversk, Russia; and
- Krasnoyarsk Electrochemical Plant (ECP), Zelenogorsk, Russia.

The U.S. facility involved in the program is the United States Enrichment Corporation/Portsmouth Gaseous Diffusion Plant (USEC/PGDP), Piketon, Ohio.

Fuel fabricators involved are:

- ABB Combustion Engineering, Hematite, Missouri;
- Framatome Cogema Fuel, Lynchburg, Virginia;
- General Electric, Wilmington, North Carolina;
- Siemens Power Corporation, Richland, Washington; and
- Westinghouse, Columbia, South Carolina.

In the United States, the Russians monitor PGDP and the fuel fabricators to assure that the LEU sent to the United States from Russia is received, sold to fuel fabricators, and made into nuclear fuel for power reactors.

In Russia, the facilities conduct the following operations. At SChE, HEU metal is received from Russian weapons dismantlement facilities; SChE also stores some weapons components and converts the HEU into chips and shavings. The HEU metal is oxidized, the oxide is purified to remove plutonium, and the oxide is packaged and shipped to UEIE and ECP. At UEIE and ECP, the oxide is fluorinated to HEU hexafluoride, and the hexafluoride is then down-blended to LEU hexafluoride, which is transferred from Russian containers to Portsmouth 30B cylinders for shipment to the United States.

At SChE, monitoring activities consist of the following:

- Visually inspect tags and seals on HEU metal chip containers.
- Observe feeding of HEU metal chips into the oxidation furnace.
- Observe the oxidation process.
- Observe HEU oxide containers removed from the process.
- Visually inspect containers and seals after packaging.
- Observe application of U.S. seals on oxide containers.
- Observe weighing/preparation of containers.

- Observe taking of oxide samples.
- Observe analysis of HEU oxide samples.

At UEIE and ECP, the following activities are conducted:

- Visually inspect containers, seals, and tags upon receipt of the oxide.
- Observe containers being weighed.
- Observe oxide containers being fed into the fluorination process.
- Observe hexafluoride withdrawal point and containers.
- Observe hexafluoride containers being weighed before and after they are filled.
- Visually inspect hexafluoride containers, seals, and tags.
- Request and observe sampling and analysis of samples (HEU, blendstock, and LEU product) from the Blend Point.
- Observe 30B cylinders being weighed before and after they are filled.
- Observe sampling and sealing sample containers.

NBL PARTICIPATION SUMMARY

The participation of NBL staff in two special monitoring visits and at the opening of the Permanent Presence Office is summarized below. NBL also hosted pretrip briefings for the entire monitoring team and involved project and headquarters personnel before two of these trips.

Special Monitoring Visit to UEIE in February 1996

Dr. Kenneth Lewis participated in a special monitoring visit to UEIE, Novouralsk, Russia, on February 12-16, 1996. At the time of this visit, Annex 3 to the protocol, Procedures of U.S. Monitoring at the UEIE, had been completed; thus, the purpose of this visit was to actually conduct monitoring activities according to the procedures.

The goals of the visit were to:

- Test daily access to the Blend Point,
- Test sampling at the Blend Point,
- Demonstrate blind sample analysis (by using UEIE standards),
- Obtain and analyze material control and accountability (MC&A) data,

- Test HEU and LEU container and process monitoring procedures,
- Test the division into three monitoring groups,
- Test the dosimeter protocol, and
- Obtain additional process information relevant to monitoring.

In addition to these major goals, other specific monitoring objectives were noted for each monitoring point on the basis of statements within Annex 3. The monitoring points from Annex 3 were the HEU Oxide Receipt/ Storage Area, HEU Oxide Fluorination Feed Area, HEU Hexafluoride Container Withdrawal Area, LEU Hexafluoride Transfer Area, Blend Point, Analytical Laboratory, and Monitors' Workrooms.

This special monitoring visit was considered to be highly successful. In prior visits to UEIE, only limited access to the processes had been obtained; in particular, there had never been such extensive access to the Blend Point. This visit essentially set the stage for achieving a successful HEU Transparency Monitoring Program.

Special Monitoring Visit to SChE in September 1996

Dr. David T. Baran participated in a special monitoring visit to the Siberian Chemical Enterprise, Seversk, Russia, on September 9-13, 1996.

SChE facilities include a uranium processing plant, centrifuge facility for enrichment, and newly installed HEU-LEU blending facility. At SChE, there were three agreed-upon monitoring points:

- Oxidation glovebox, where HEU metallic chips/shavings are converted;
- Workshop Two, where samples are taken and shipments are prepared for UEIE or ECP; and
- Analytical laboratory, where ^{235}U isotopic content is analyzed.

Opening of Permanent Presence Office, UEIE, in August-October 1996

Dr. Kenneth Lewis was appointed the head of the delegation for the opening of the Permanent Presence Office at UEIE, Novouralsk, Russia, from August 13 through October 12, 1996.

As noted above, overall processing at UEIE consists of the receipt of HEU oxide that was prepared from metal at other Russian facilities, followed by the fluorination of that oxide to HEU hexafluoride. These two steps occur in the HEU Fluorination Facility. Next the HEU hexafluoride is transferred to the Blend Point, where it is blended with a diluent hexafluoride to an LEU product. Finally, the transfer of that LEU product from UEIE vessels to the USEC/Portsmouth 30B cylinders for shipment to the USEC/Portsmouth Plant is performed in the LEU Transfer Facility. The U.S. monitors may request a blind sample analysis, in which samples are taken from the HEU hexafluoride diluent and product lines of the Blend Point and analyzed at the Analytical Centre. During such an analysis, the Analytical Laboratories in which the analyses are conducted become another monitoring point.

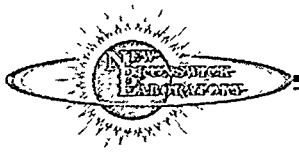
During the two-month stay at the facility, not all of the operations were active and processing material for the program. Shortly after arrival of the team, the last batch of oxide from Seversk to be converted to meet the 1996 commitment was ready to be processed. The team monitored the feeding of this material in the Fluorination Facility, after which the fluorination operation was closed down. Fluorination did not start again until two weeks before the team left. Receipt and storage of initial batches of 1997 oxide were monitored in the Fluorination Facility twice during September. On September 30, the restart of fluorination was monitored by observing three cans of newly received oxide being fed into the fluorination unit.

Most of the monitoring time was spent in the LEU Transfer Facility, where the product of the blending is transferred from the Russian containers (referred to as Technological Vessels) into the USEC/Portsmouth 30B cylinders for shipment to the United States. UEIE staff provided an operating schedule for the LEU Transfer Facility on a weekly basis so that the U.S. monitors knew when specific operations were being performed in the facility. The operation was continuous for 24 hours a day and through the weekends. Part of the monitoring consisted of gaining access to the facility and checking that the stage of operation agreed with the operating schedule provided.

Access to the Blend Point was granted early in the team's stay. During that time, the team checked U.S. seals that had been placed on the orifice plates during a change in the plates monitored by the United States in March 1996. Access was obtained again when the team requested and conducted a Blend Point sampling and blind sample analysis, which was carried out in accordance with the procedure given in the Annex of the basic agreement. The team monitored the sampling of the HEU feed and LEU diluent and product hexafluoride lines at the Blend Point and then observed the mass spectrometric analysis of these samples and blind standards in the Analytical Centre.

The set-up of the U.S. Permanent Presence Monitoring Office was successful, and monitoring activities continue with each new monitoring team. For FY 1997, special monitoring trips are scheduled biannually for the UEIE facility and bimonthly for the ECP and SChE facilities. NBL staff will provide team members for the UEIE Permanent Presence Office and for special monitoring trips to the UEIE Permanent Presence Office and SCHe.

7 SAFEGUARDS MEASUREMENT EVALUATION



SAFEGUARDS MEASUREMENT EVALUATION PROGRAM

M.I. Spaletto, M.M. Smith, M.D. Soriano, and M.E. Downey

SME PROGRAM INFORMATION

New Brunswick Laboratory (NBL) provides technical support to the U.S. Department of Energy (DOE), Office of Nonproliferation and National Security, Office of Security Affairs, Office of Safeguards and Security (NN-51), by managing a program to independently evaluate the performance of nuclear material accountability measurements in DOE nuclear facilities. This support is provided through operation of the NBL Safeguards Measurement Evaluation (SME) Program, which monitors the quality of uranium elemental concentration and uranium and plutonium isotopic abundance destructive analyses. The SME Program was implemented to replace the Safeguards Analytical Laboratory Evaluation (SALE) Program, specifically so that multiple, site-specific materials would be available.

The NBL SME Program was developed as a means to monitor and evaluate the quality and effectiveness of nuclear material accountability measurements by site, material balance area (MBA), or unit process. The program's goal is accomplished through the selection of site-specific evaluation materials, which are prepared, characterized, and distributed to participating facilities for analysis. Reported measurement data are statistically evaluated, and results are reported to the participating facility, the cognizant DOE operations office, and DOE headquarters. By using the international target values included in the NN-51 Measurement Control Guide and Measurement Improvement Plan, the SME Program reports include a determination as to whether the reporting facilities have achieved accuracy and precision within appropriate target values. Operations offices are encouraged to use these values as a guide to assessing the adequacy of site accountability measurement capabilities.

The database application for the statistical evaluation of submitted results, developed last fiscal year, is fully operational. All fiscal year (FY) 1996 uranium data have been entered into the new, streamlined system.

Five Nuclear Regulatory Commission (NRC) licensees began full participation in the program in FY 1996, according to a cooperative agreement between NN-51/NBL and the NRC. Each licensee received and analyzed two sets of samples and received statistical evaluations of the submitted data. Licensee participation is being used by the NRC to provide more timely and cost-effective monitoring of measurement performance at these facilities, when compared with on-site verification sampling by NRC inspectors.

As part of the NN-51efforts to improve efficiency, the Calorimetry Exchange Program was included in the SME Program in FY 1996; the transfer of data from the from EG&G Mound Laboratory to NBL has been completed.

With the approval of NN-51, the program has added its first foreign participant, the Safeguards Analytical Laboratory (SAL) in Tokai, Japan. SAL will participate, on a cost-recovery basis, in the analysis of highly enriched uranyl nitrate solution samples for uranium concentration by isotope dilution mass spectrometry (IDMS) and for enrichment by thermal ionization mass spectrometry. Full participation will begin in fiscal year (FY) 1997.

NORMAL URANYL NITRATE MATERIAL

The use of normal uranyl nitrate solutions enables the participation of facilities restricted from receiving shipments of enriched materials. A suite of three solutions is distributed to participants through the program. The elemental assay of each solution differs from the next by approximately 0.15% to 0.2%; the ability to differentiate these materials demonstrates good analytical capabilities.

Los Alamos National Laboratory (LANL), NBL, the Lockheed Martin Y-12 Plant, Westinghouse Savannah River Plant, and Babcock and Wilcox (B&W) Naval Nuclear Fuels are participating in the normal uranyl nitrate portion of the program to evaluate uranium assay accountability measurements on solution samples. Seventeen reports were issued to the active facilities, cognizant DOE operations offices, and NRC headquarters through September 1996. All participating facilities performed within the international target values for precision; one facility was unable to perform within the target values for bias by using its traditional analytical method. This facility is using the program to evaluate IDMS against its traditional method; in the SME Program, the traditional method has historically exhibited shifting biases from month to month. The new IDMS method has performed very well, producing results comparable to titrimetry. One new participant produced unacceptably biased titration results for its first data set; this bias was not mirrored in the laboratory's quality control standards. The laboratory was subsequently able to reduce the bias to within target values and performed acceptably the rest of the fiscal year. Another laboratory continues to produce excellent results by using a ceric titrant as a replacement for the dichromate titrant in the NBL-modified Davies and Gray titration; this accomplishment indicates that ceric titrant may be substituted for dichromate titrant at other laboratories, which will help minimize the production of mixed waste.

In FY 1997, B&W Naval Nuclear Fuels will analyze highly enriched uranyl nitrate solutions instead of the normal uranium solutions. This analysis will enable B&W Naval Nuclear Fuels to submit results on enrichment as well as uranium concentration.

ENRICHED URANYL NITRATE MATERIAL

The SME Program has available for distribution three highly enriched (approximately 90% ^{235}U) uranyl nitrate solutions: a solution of 50% enriched uranyl nitrate, and a 4% low-enriched solution for isotopic analysis. All solutions, except for the 4% enriched solution, are characterized for elemental concentration as well as isotopic distribution, enabling them to be used in IDMS.

Argonne National Laboratory-West, NBL, the Savannah River Plant, and the Y-12 Plant submitted results from isotopic analyses of the enriched uranyl nitrate solutions. Eight reports have been issued. Isotopic results from all laboratories but one were well within the international target values for bias and precision. One new participant is exhibiting a small bias that becomes increasingly more positive as enrichment decreases. In addition to enrichment measurements, one laboratory also analyzed these solutions for elemental concentration by using IDMS. This laboratory performed well within the target values, producing results very comparable to titrimetry. These results, combined with the good results obtained by another laboratory on normal uranyl nitrate solutions analyzed by IDMS, indicate that uranium IDMS, which produces less waste than titrimetry, may be developed at other facilities as an alternate method of analysis.

In addition to being used in the SME Program, the uranyl nitrate solutions, both those characterized for elemental assay and those characterized for isotopic distribution, may be used in cooperation with the Safeguards Assessment Program for performance tests during periodic material control and accountability (MC&A) inspections.

URANIUM OXIDE (UO_2) MATERIAL

With funding from the NRC, a new material was added to the SME Program: UO_2 pellets characterized for both uranium concentration and enrichment. NBL and four NRC licensees (GE Nuclear Energy Production, ABB Combustion Engineering, Siemens Power Corporation, and Westinghouse Commercial Nuclear Fuel Division) are analyzing this material. Ten reports were issued for assay, and ten were issued for enrichment; all results were well within international target values.

PLUTONIUM MATERIALS

For plutonium isotopic analysis, three solutions with differing isotopic distributions are available for use in the Program. LANL, NBL, Rocky Flats Environmental Technology Site, the Savannah River Plant, and Westinghouse Hanford Company have submitted results. Thirteen reports were issued to the facilities and cognizant DOE operations offices through September 1996. Overall performance for all facilities was within international target values for ^{239}Pu. Small abundances of ^{238}Pu proved to be the most difficult to measure; most facilities are biased high. This may indicate uranium contamination in the laboratories or incomplete separation from the interfering isobar ^{238}U, especially when accompanied by high ^{241}Pu measurements.

All plutonium participants, except for Rocky Flats, have agreed to accept samples in FY 1997 to be analyzed for plutonium concentration by IDMS. Two new solutions were prepared for this purpose in the course of a Measurement Development Program project funded by DOE's Office of Environmental Management (EM-334).

PAPERS AND MEETINGS

The first meeting of participants in the Safeguards Measurement Evaluation Program was held in FY 1996 at NBL; the minutes from the meeting were compiled and distributed. The first comparative report of facilities, covering uranium results submitted from FY 1992 through FY 1994, was prepared and distributed. Subsequently, annual meetings will be held, and annual comparative reports on program results will be issued and distributed to participants and oversight personnel.

PLANS FOR FY 1997

With the end of the Cold War, many nuclear materials are being moved into storage. Nondestructive assay (NDA) will be used to measure these materials, which avoids opening the storage containers to remove samples for destructive analysis. Use of NDA reduces overall effort and the potential for diversion, contamination, and unnecessary personnel exposure. Construction and characterization of a series of uranium NDA gamma standards are planned for next fiscal year, to be used in a "round-robin" type of sample exchange.

NBL is exploring the possibility of including other international participants in the SME Program. A proposal to include selected Argentinean and Brazilian safeguards laboratories was submitted to DOE's Office of Nonproliferation and National Security, Office of Arms Control and Nonproliferation, International Safeguards Division (NN-44).

Enhancement of the new database application for the statistical evaluation of SME Program results will continue in FY 1997. Plutonium analysis will be incorporated, and the application will be adapted to format and produce comparative annual reports. NBL is investigating an enhancement whereby participants will have access to the database to input data and query results.

In March of FY 1997, the Portsmouth and Paducah Gaseous Diffusion Plants (GDPs) will become NRC licensees. The GDPs had been participating in the SME Program as DOE contractors; the NRC will support the continued inclusion of these facilities in the SME Program.

A meeting of the Measurement Evaluation Program participants will be held in the spring of 1997. A portion of this meeting will cover the Calorimetry Exchange Program.

CALORIMETRY EXCHANGE PROGRAM TRANSFER

M.I. Spaletto and M.D. Soriano

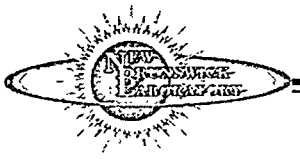
The Calorimetry Exchange Program was started in 1975 as an extension of the Rocky Flats Plutonium Metal and Isotopic Exchange Program. In 1979, Rocky Flats prepared and characterized a batch of PuO_2 powder with 6% ^{240}Pu, which is still being used as a test sample. The material was packaged into six units, each of which has a heat output of approximately 1 watt, and distributed to participants. Each participant periodically performed gamma isotopic and calorimetric measurements on the samples. The results of these measurements were compared in quarterly reports, each of which contained data covering the previous four quarters. In 1984, the program separated from the Rocky Flats Plutonium Metal and Isotopic Exchange Program, and administration of the Calorimetry Exchange Program was transferred to the EG&G Mound Facility.

Over time, changes occurred in the mission of the Mound Facility. Thus, the U.S. Department of Energy, Office of Safeguards and Security, asked New Brunswick Laboratory (NBL) to assume administration of the Calorimetry Exchange Program to increase efficiency and reduce costs to DOE. In 1996, the program was included within the Measurement Evaluation Program. Participants' data for as-yet unpublished reports, both on computer and paper, were transferred from Mound to NBL. NBL also received documentation of the characterization of the 1-watt sample and information about the administration of the program. NBL personnel underwent formal training to prepare for assumption of these new responsibilities. In June, NBL sent a letter that informed the six participants of the transfer and requested that subsequent data be sent to NBL. The participating laboratories are Los Alamos National Laboratory (LANL), Lawrence Livermore National Laboratory, two laboratories at Safe Sites of Colorado, Westinghouse Savannah River Company, and Babcock and Wilcox Hanford Company.

The last report issued by the Mound Facility covered the third quarter of 1993. Because the reports are so far behind schedule, NBL plans to issue combined fourth quarter/annual reports that will include all data not yet published for the years 1993, 1994, and 1995. Participants will be invited to an annual meeting to be held in the spring of 1997 in conjunction with the Measurement Evaluation Program meeting.

Because of changes in the DOE complex, including a large effort in the area of inventory consolidation, the Calorimetry Exchange Program participants had identified a need for new samples for the program. Hanford Company provided a suitable batch of higher-burn-up PuO_2 powder to LANL. LANL performed analyses of loss on ignition, plutonium content, plutonium isotopic distribution, and americium content on five analytical samples removed from the bulk material. LANL shipped an additional five analytical samples to NBL to perform the same analyses and supplied documentation of its results. The material, which contains approximately 12% ^{240}Pu, has been packaged into 10 units, each of which has a heat output of approximately 6 watts. When NBL analyses are completed, the data will be consolidated to provide reference values for the sample. Subsequently, the units will be distributed to participants.

In addition, an even higher wattage standard is needed to support safeguards measurements for the long-term storage of plutonium materials packaged by using the container designed from DOE-STD-3013/96. NBL and LANL will be collaborating in the production of this new standard.



8 NBL STAFF AS OF APRIL 1997

OFFICE OF THE DIRECTOR

MARGARET E. M. TOLBERT, LABORATORY DIRECTOR
Kenneth Lewis, Senior Technical Assistant
Jodi Von Rox, Administrative Assistant

ADMINISTRATIVE SUPPORT TEAM

Kathryn B. Papenfuss, Administrative Officer/Team Leader
Mary Ann Presa, Secretary
Condeocita Marcelo, Purchasing Agent
Albert Thomas, Shipper/Packager/Receiver

ES&H OFFICE TEAM

Jon Neuhoff, Physical Scientist/ES&H Team Leader
Eric Dallmann, Physical Scientist/Facility Manager
Frank Orlowicz, Physical Science Technician/DOT Certifier
Margaret Redman, Industrial Hygienist

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ROBERT D. OLDHAM, DIVISION DIRECTOR
Michelle Ficner, Secretary
Steven Goldberg, Lead Mass Spectrometrist
David Baran, Safeguards Cadre/Physicist
Paul Croatto, Safeguards Cadre/Chemist
Iris Frank, Chemist
Kimberly Johnson, Chemist
Peter Mason, Chemist
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Patricia Santoliquido, Safeguards Cadre/Chemist
Khalida Scheidelman, Chemist
Gary Sowell, Chemist
Anthony Traina, Safeguards Cadre/Mass Spectrometrist
Dennis Troutman, Electrical Technician
Arnold Warren, Chemist
Anna Voeks, Chemist

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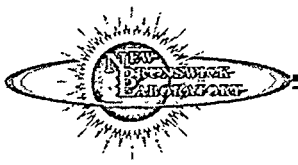
WANDA G. MITCHELL, DIVISION DIRECTOR
Margaret Legel, Safeguards Cadre/Safeguards Assistance Program Manager
Usha Narayanan, Reference Materials Program Manager
M. Irene Spaletto, Safeguards Cadre/Materials Evaluation Program Manager
Alma Stiffin, Materials Services Program Manager
Jeffrey Zebrowski, Safeguards Cadre/Acting Materials Development Manager
Maureen Clapper, Safeguards Cadre/Chemist
Mary Ellen Downey, Statistical Assistant
John Sickels, Materials Control & Accountability Representative
Marianne Smith, Statistician
Michael Soriano, Statistician

9 PUBLICATIONS AND PRESENTATONS (NBL PROJECTS)



- *Safeguards Measurement Evaluation Program Uranium Sample Exchange, FY 92 through FY 94*, Report NBL-334, New Brunswick Laboratory, Argonne, Illinois, March 1996 (M. Irene Spaletto, Marianne M. Smith, and Michael D. Soriano).

- *Minutes of the Safeguards Measurement Evaluation Program Meeting October 18-19, 1995*, NBL-336, New Brunswick Laboratory, Argonne, Illinois, July 1996.

- "Neptunium Material for the Production of a Standard," paper presented at Alternate Nuclear Material Workshop, Savannah River Site, North Carolina, 1996 (Wanda G. Mitchell and Usha I. Narayanan).

- "Technical Standard for Stabilization and Repackaging of Excess Nuclear Materials in the Complex," paper presented at Fissile Materials Assurance Working Group Meeting, U.S. Department of Energy Headquarters, 1996 (Usha I. Narayanan and Wanda G. Mitchell).

- "New Brunswick Laboratory Certified Reference Materials," exhibit and paper presented at Argonne National Laboratory's 50th Anniversary Technical Women's Symposium, Argonne, Illinois, 1996 (Usha I. Narayanan, Margaret A. Legel, Wanda G. Mitchell, and H. Rodney Martin).

- "New Brunswick Laboratory Certified Reference Materials," exhibit presented at Pittcon, Chicago, Illinois, 1996 (Usha I. Narayanan, Gary A. Sowell, Paul V. Croatto, M. Irene Spaletto, Mary Ellen Downey, and Kathryn B. Papenfuss).

- "Pollution Prevention through Reduction of CRM Unit Sample Size," paper presented at U.S. Department of Energy Pollution Prevention (DOE-P2) Conference XII, Chicago, Illinois, 1996 (Usha I. Narayanan and Colleen G. Gradle).

- "Pollution Prevention in Actinide Analysis — A High Return on Investment Project," paper presented at U.S. Department of Energy Pollution Prevention (DOE-P2) Conference XII, Chicago, Illinois, 1996 (Usha I. Narayanan and D. Eric Dallmann).

- "New and Replacement New Brunswick Laboratory Certified Reference Materials," paper presented at Institute for Nuclear Materials Management 37th Annual Meeting, Naples, Florida, 1996 (M. Irene Spaletto, Usha I. Narayanan, Patricia M. Santoliquido, and Peter B. Mason).

- "Development of Plutonium Isotope Dilution Mass Spectrometry for Routine Analysis," paper presented at Institute for Nuclear Materials Management 37th Annual Meeting, Naples, Florida, 1996, and Safeguards Measurement Evaluation Program Meeting, Argonne, Illinois, 1995 (Usha I. Narayanan, Fred E. Jones, Alma V. Stiffin, M. Irene Spaletto, Margaret A. Legel, Michael D. Soriano, and D. Eric Dallmann).

- "The Development, Availability and Uses of NBL CRMs," paper presented at Safeguards Measurement Evaluation Program Meeting, Argonne, Illinois, 1995 (Usha I. Narayanan, M. Irene Spaletto, and Wanda G. Mitchell).

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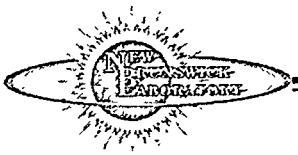

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