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National Measurement Laboratory Office of Measurements for Nuclear Technology Annual Report 1980

John D. Hoffman

Director
National Measurement Laboratory

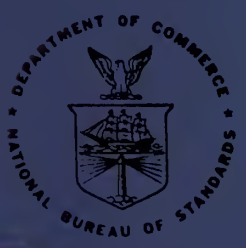
H. Thomas Yolken

Chief, Office of Measurements
for Nuclear Technology

National Measurement Laboratory
U.S. Department of Commerce
National Bureau of Standards
Washington, DC 20234

January 1981

Annual Report, Fiscal Year 1980
(October 1, 1979 - September 30, 1980)



U.S. DEPARTMENT OF COMMERCE
NATIONAL BUREAU OF STANDARDS

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NATIONAL MEASUREMENT LABORATORY OFFICE OF MEASUREMENTS FOR NUCLEAR TECHNOLOGY ANNUAL REPORT 1980

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U.S. DEPARTMENT OF COMMERCE, Malcolm Baldrige, *Secretary*
NATIONAL BUREAU OF STANDARDS, Ernest Ambler, *Director*

Preface

This annual report is a summary of the National Bureau of Standards (NBS) Measurements for Nuclear Technology (MNT) Program for Fiscal Year 1980. The MNT activities at NBS are divided into two programs: Nuclear Safeguards and Nuclear Waste Management. The Nuclear Safeguards Program was the first to be initiated near the end of Fiscal Year 1977, while the Nuclear Waste Management Program was initiated near the end of Fiscal Year 1980. However, NBS did make significant contributions to the nuclear safeguards field in years past. For example, in the 1950's, C. J. Rodden and other NBS scientists founded a laboratory for the measurement of nuclear fuels at NBS. This laboratory was later transferred to the U.S. Atomic Energy Commission and became known as the New Brunswick Laboratory. In addition, work by W. Shields and co-workers in the 1960's established NBS as a world leader in the development of ultra-high accuracy mass spectrometry for chemical and isotopic assay of nuclear fuels. NBS Standard Reference Materials for the chemical and isotopic assay of uranium and plutonium provide the foundation for nuclear safeguards measurements throughout the world.

In a somewhat different area of safeguards technology, J. Schleter and co-workers at NBS provided an entirely new approach to the problem by applying systems analysis techniques. In the early 1970's, they developed and implemented a technique that is known as diversion path analysis. Many other organizations are now applying systems analysis techniques to safeguards problems.

In the mid 1970's, interest in standardization of measurements for nuclear safeguards was furthered by the recognition of a need for an NBS program in this area from many organizations. This recognition and support resulted in the establishment of the Measurements for Nuclear Safeguards Program at NBS.

The Department of Energy (DOE) in late August 1979 invited the National Bureau of Standards to consider the establishment of an appropriate technical assistance program to meet the measurement standards needs of the waste disposal problem.

Because of the urgency of the request and the complexity of the problem, NBS assembled a study group consisting of 13 experts in a variety of scientific and engineering disciplines. The group was asked by NBS management to study the problem in a three-month period (Sept. 15 - Dec. 15, 1979), to explore critical questions, and to define an appropriate NBS technical program in a final report to be completed and reviewed by March 31, 1980. The report was completed on March 31, 1980, and submitted to DOE. The goal of the NBS program stated in the report is to provide a measurement base (including reference methods, Standard Reference Materials, Standard Reference Data, measurement assurance procedures, and scientific laws and concepts of waste behavior) to allow other organizations to (a) measure short-term behavior, (b) predict long-term behavior, (c) choose acceptable disposal systems, and (d) provide quality control of processing and assembly in nuclear waste management. The NBS Nuclear Waste Management Program was initiated in September 1980 and is completely supported by DOE funds.

The MNT Program is one of several matrix managed efforts at NBS. The programs provide a focus for the activity, both inside and outside NBS. The programs provide funds and directions for needed technical work that is done within NBS Centers. People remain attached to Centers but are funded for specific activities in a matrix management approach. A program provides central organization for outside inquiries and for distribution of results. Often programmatic focus can provide improved assistance to outside agencies with measurement problems. The MNT Program is operating in this manner.

We hope that this annual report will provide the technical community with a view of the Program's direction. We encourage members of the community to make recommendations to us concerning the program's future direction.

B. Stephen Carpenter
William P. Reed
H. Thomas Yolken
Office of Measurements for
Nuclear Technology

TABLE OF CONTENTS

1. Introduction

The Office of Measurements for Nuclear Technology (OMNT) was established to provide national measurement standards needed to safeguard nuclear materials. In addition, during the past year, a task force of NBS scientists examined the need for measurement standards in nuclear waste management, resulting in the formation of a program in nuclear waste management. Thus, the OMNT now consists of two separate activities, Nuclear Safeguards and Nuclear Waste.

Nuclear Safeguards

Introduction

Rapid and sensitive methods to detect the diversion of nuclear materials are an essential objective of the measurements for nuclear technology program. This objective can be achieved with the aid of accurate and precise methods of measurement of nuclear materials at all phases of manufacture, use, and transfer to maintain material accounting. Effective and universal measurement standards are required to attain the necessary accuracy of measurement.

The NBS program for nuclear safeguards serves three sets of clients: the Nuclear Regulatory Commission (NRC) and the U.S. commercial nuclear industry; the Department of Energy (DOE) (as developer of nuclear safeguards technology and as regulator of nuclear facilities operated for DOE) and U.S. Government-owned DOE nuclear facilities; and the International Atomic Energy Agency (IAEA) and countries that have nuclear facilities under the jurisdiction of the Non-Proliferation Treaty. The FY 1981 NBS nuclear safeguards program is funded by DOE and the Department of State (DOS).

Goals and Objectives

The goals and objectives of the NBS program are derived from the perceived needs of the sponsoring agencies - to reduce the risk to the public from the illegal diversion of nuclear materials by providing a common and accurate standardization basis for measurement of nuclear fuels. This goal is to be achieved for all of the measurements that need to be made throughout the fuel cycle on a wide variety of material forms. In order to meet this goal, our objectives are to provide measurement standards and services for the four major measurement areas for nuclear safeguards accountability. These are listed below.

Tasks

To achieve these goals and objectives, three task areas are utilized that combine similar technological resources of NBS. These three task areas are

(a) Bulk and Nondestructive Measurements for Nuclear Safeguards
(b) Chemical Measurements for Nuclear Safeguards
(c) Systems Studies and Statistical Analysis

Nuclear Waste Management

Introduction

In August 1979, DOE invited NBS to consider establishment of a technical program that would contribute to the measurement standards foundation required for disposal of nuclear waste. A group of NBS scientists was asked by the

management of NBS to examine the needs for measurement standards in nuclear waste management and, if desirable, to recommend a technical program. The NBS study (funded by DOE) was completed, approved by the NBS Executive Board, and submitted to DOE on March 31, 1980. DOE staff have reviewed the NBS proposal and have provided a total of $1,850,000 to NBS to initiate a portion of the program in FY 1980 (last quarter) and FY 1981. DOE intends to increase the funding level in future years to allow additional activities to be included in the program. NBS and DOE will most likely consummate a Memorandum of Understanding in the nuclear waste management area.

Goals and Objectives

The summary of the NBS report on nuclear waste management follows . . . The National Bureau of Standards proposes to support the national program in nuclear waste disposal by: assisting in the development of measurement techniques and standards to assess stability, providing knowledge of factors controlling stability of the waste package, determination and evaluation of data critically needed for evaluation of alternative waste forms and packages, and development of measurement standards and procedures needed for quality control. This is an appropriate role for NBS for several reasons. First, accuracy is required in difficult measurements that are essential to the solution of a national problem. Second, competence in similar measurements on other materials already exists at NBS. Third, credibility of results is extremely important; the traditional NBS measurement and standards role of third party objectivity with no promotional or regulatory interest is appropriate. Fourth, certain voluntary consensus standards are needed, as well as regulatory standards; the existing extensive work of NBS in support of the voluntary consensus standards system makes a good base for similar standards work in support of nuclear waste disposal.

The strategy of the NBS program recognizes the need to disseminate existing NBS capability, mainly in the area of short-term performance, and to concurrently develop the new scientific basis needed for long-term prediction, and to provide the additional basis needed for short-term performance capability. For example, work is needed to improve the accuracy of existing leach tests and develop consensus on standard tests to determine short-term performance. This work is critically needed but must be supplemented by work on the mechanisms controlling release so that long-term predictions can be made. NBS will accordingly pursue parallel and interactive efforts centered respectively on reference leach tests involving chemical analysis and on the kinetics of long-term release. The NBS strategy also recognizes the importance of collaboration with work already in progress in a number of government, industry, and university laboratories. NBS will make optimum use of its existing experimental facilities in carrying out this work. NBS will concentrate on model systems using the same chemical compounds and structures, but employing isotopes with no or relatively low levels of radioactivity. Work on actual, high activity waste forms will be carried out in a cooperative manner at existing hot cells in other laboratories.

With the foregoing considerations in mind, the overall goal of the NBS program can be stated as follows: To provide a measurement base (including methods, data, traceability to basic standards, and assurance procedures) and scientific laws and concepts of waste behavior to support: (a) measurement of short-term behavior, (b) prediction of long-term behavior, (c) choice of acceptable disposal systems, and (d) quality control of processing and assembly in nuclear waste management.

The NBS program is developed in terms of work needed on each component of the multi-barrier system and the need to monitor the system. The following technical themes run through much of the work:

(a) the interactions between components of the system
(b) quantitative analysis of elements and phases
(c) stability of phases over relatively short times
(d) leach and corrosion rates over relatively short times
(e) mechanisms and quantitative basis of phase changes, mechanical deterioration, leaching, corrosion, absorption, and transport
(f) predictive scientific laws and concepts to allow choice of disposal systems
(g) test methods and procedures for quality control

Tasks

To achieve these goals and objectives, the NBS program on nuclear waste has developed, for FY 1981, three tasks which combine similar technology resources of NBS. These three task areas are

(a) Leachability Mechanisms for Waste Forms
(b) Nuclear Waste Form Reference Materials and Data
(c) Containers, Overpack, Corrosion and Backfill

2. Personnel

The Office of Measurements for Nuclear Technology (OMNT) is in the NBS National Measurement Laboratory (see organizational chart in Appendix). The OMNT consists of two major programs: Measurements for Nuclear Safeguards and Nuclear Waste Management. Each program is run by the Team Management concept, which allows members from different centers or programs within NBS to help the programs run effectively. At the present time, the OMNT interacts with four centers in NML and two centers and the Continuous Process Technology Program in NEL. The Management Team concept has been demonstrated to be a fruitful approach to matrix management.

An FY 1980 organizational chart is shown in the Appendix, Section F. Tables I and II provide a summary of the NBS-OMNT personnel.

Table I

Summary of NBS Personnel Involved in the OMNT Safeguards Program

Technical or Area	Principal Investigators
A. Bulk Measurements	
Density, Flow, Volume, Mass, Process Measurements	G. Baumgarten, D. Cooper, A. Gaigalas, J. Houser, E. Johnsen, F. Jones, B. Robertson, J. Whetstone, T. Yeh
B. Destructive Chemical and Isotopic Assay	
Chemical Assay, Mass Spectrometry, Alpha Spectrometry, Moisture Determination	J. Fassett, E. Garner, J. Gramlich, F. Jones, W. Kelly, H. Kingston, L. Lucas, L. Machlan, J. Moody, L. Moore
C. Non-Destructive Assay	
Calorimetry, Gamma Techniques, Neutron Techniques	D. Bakshi, J. Behrens, C. Bowman, A. Carlson, S. Carpenter, T. Cheng, J. Colwell, D. Ditmars, R. Fleming, M. Ganoczy, D. Garrett, R. Johnson, L. Lucas, M. Meier, T. Mitlehner, R. Schrack
D. Statistics	
Measurement Assurance Programs, Reference Materials, Calibrations, In-Plant Error Analysis	H. Ku, J. Lechner, W. Liggett, C. Spiegelman
E. Safeguards Systems Studies	S. Bologa, M.D.K. Maltese, J. Schleter
F. User's Guide, Safeguards Accountability, UF_6 Mass MAP	E. Johnsen

TABLE II

Summary of NBS Personnel Involved in the OMNT Waste Program

Technical or Area	Principal Investigators
A. Waste Forms	
1) Leachability Mechanisms	
Concepts - Theory	R. Mountain, R. Munro
Radiation Effects	T. Cheng
Transport, Thermochemistry, Microstructures	R. Roth, A. Franklin
Mechanical Properties	S. Wiederhorn
2) Research and Reference Materials, Data Measurement Methods	
Phase Diagrams of Relevant Ceramics	R. Roth
Leachability Research Materials (Glass)	W. Haller, D. Cronin
Leachability Reference Test Methods	H. Kingston
B. Containers, Overpacks, Sleeves	
Corrosion Mechanisms Data and Methods Feasibility of Long-Term Deep Test	J. Kruger
C. Backfill	
Mechanisms Research Materials Data and Test Methods	F. Yokel, R. Chung
D. Mathematical Support	
Statistical Engineering	H. Ku
Submodeling	J. Fong

3. Measurements for Nuclear Safeguards Program - A Technical Overview

A description of the technical activities in each of the Program's four areas is given in this section. In addition, accomplishments during FY 1980, the third full year of the Program, are reviewed.

A. Non-Destructive Assay

The NBS effort on standards for non-destructive assay (NDA) of nuclear materials is motivated by the need for reliable, accurate, timely and economic methods for the measurement of special nuclear materials. A wide variety of activities are being carried out in NBS laboratories to fulfill this objective. The activities may be grouped into three major categories: reference methods, standard reference materials, and data development and calibration.

(1) Reference Methods

Reference methods are methods having demonstrated precision and accuracy. Due to their complexity, these techniques are generally suited only for a specially equipped laboratory. These reference measurement methods are used to characterize reference materials or as references for developing and calibrating simpler techniques that are inherently less accurate.

(a) Resonance Neutron Radiography

Advances in data gathering and in analysis have been made during the past year. After development of a high resolution one-dimensional positron sensitive neutron detector and the successful demonstration of its ability to analyze fresh fuel pellets, it was obvious that the speed of data gathering could be greatly improved by the development of a two-dimensional detector. In collaboration with Oak Ridge National Laboratory, a two-dimensional positron sensitive proportional counter was developed. Initial tests on the device have shown high resolution ($\sim$ 1.0 mm) and further development on the device is underway to improve the speed of data taking. A different type of two-dimensional detector, shown in Figure 1, has been developed in cooperation with Surface Science Corporation. A small detector about 1" in diameter having a resolution of about 0.1 mm has been developed that utilizes a multi-channel plate electron amplifier. Plans for testing the device are now underway.

Computer calculations have shown that inhomogeneities in sample thickness, that would cause large errors in the assay of constituents, can be compensated for and the error reduced by a factor of about 20 by shape analysis of the resonance line in the absorption spectrum. The dependence of this effect on sample thickness is shown in Figure 2. With this philosophy in mind, two samples of spent fuel have been measured treating the whole 1-cm diameter slice of fuel as one analysis area. Figure 3 shows the transmission spectra obtained for the two samples. Analysis of the data is now underway, but a casual examination shows quite obvious differences in isotopic composition between the two samples. Table 1 lists some of the more obvious examples. The two samples are from the same fuel rod. One sample is taken from the end of the rod where the neutron flux is relatively low, and one cut is taken from the center of the rod where the neutron flux is greater. It is clear that the transuranic isotope buildup is greater in the center cut, and conversely the depletion of the ^{235}U is less on the end cut. The development of this technique should be a valuable tool in spent fuel analysis for the safeguards program to determine reactor history and fuel rod composition.

This work is being performed in the Center for Radiation Research under the sponsorship of NRC.

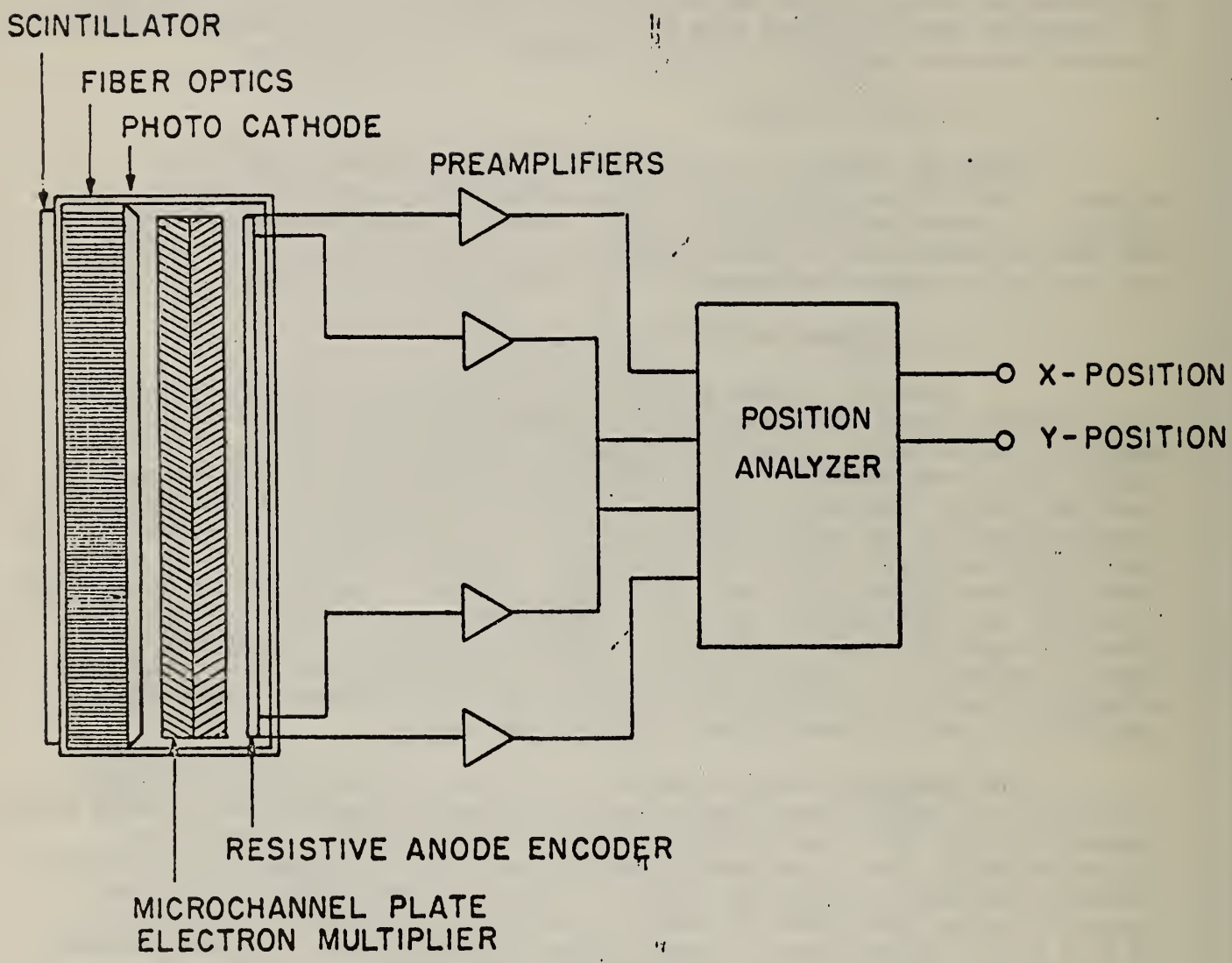


Figure 1. Schematic of two-dimensional neutron detector. The scintillator material is 1.0 mm thick enriched Li glass. The system has a dead time of 8 microseconds.

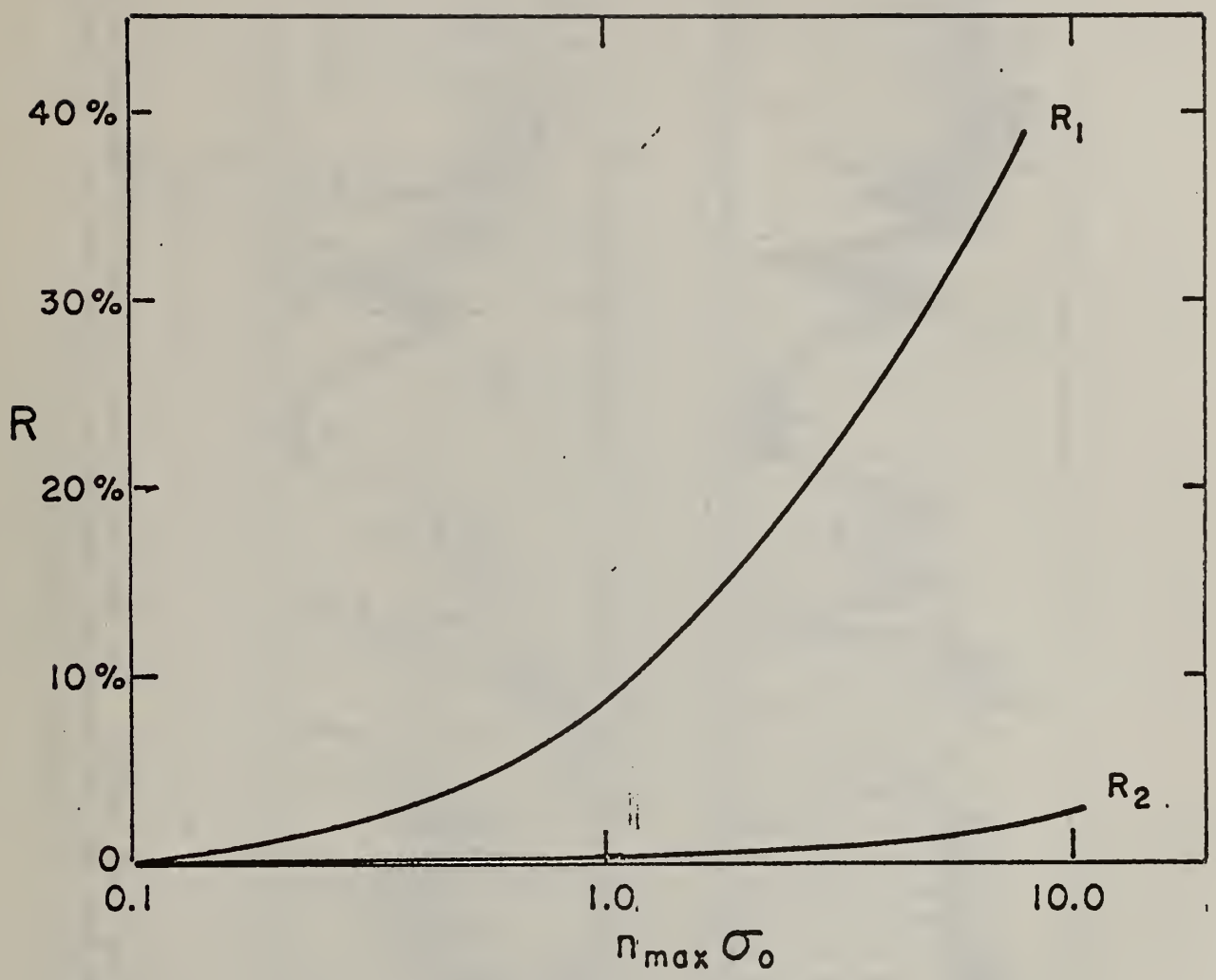


Figure 2. Error induced in sample analysis by non-homogeneity of sample. The graph shows the error induced by a sample whose density varies by a factor of ten. The maximum density is n_{max} atoms per barn, and the peak cross section is σ_0 barns/atom. The curve labelled R_1 shows the error obtained by conventional analysis. The curve labelled R_2 shows the error obtained when a two-component analysis is used.

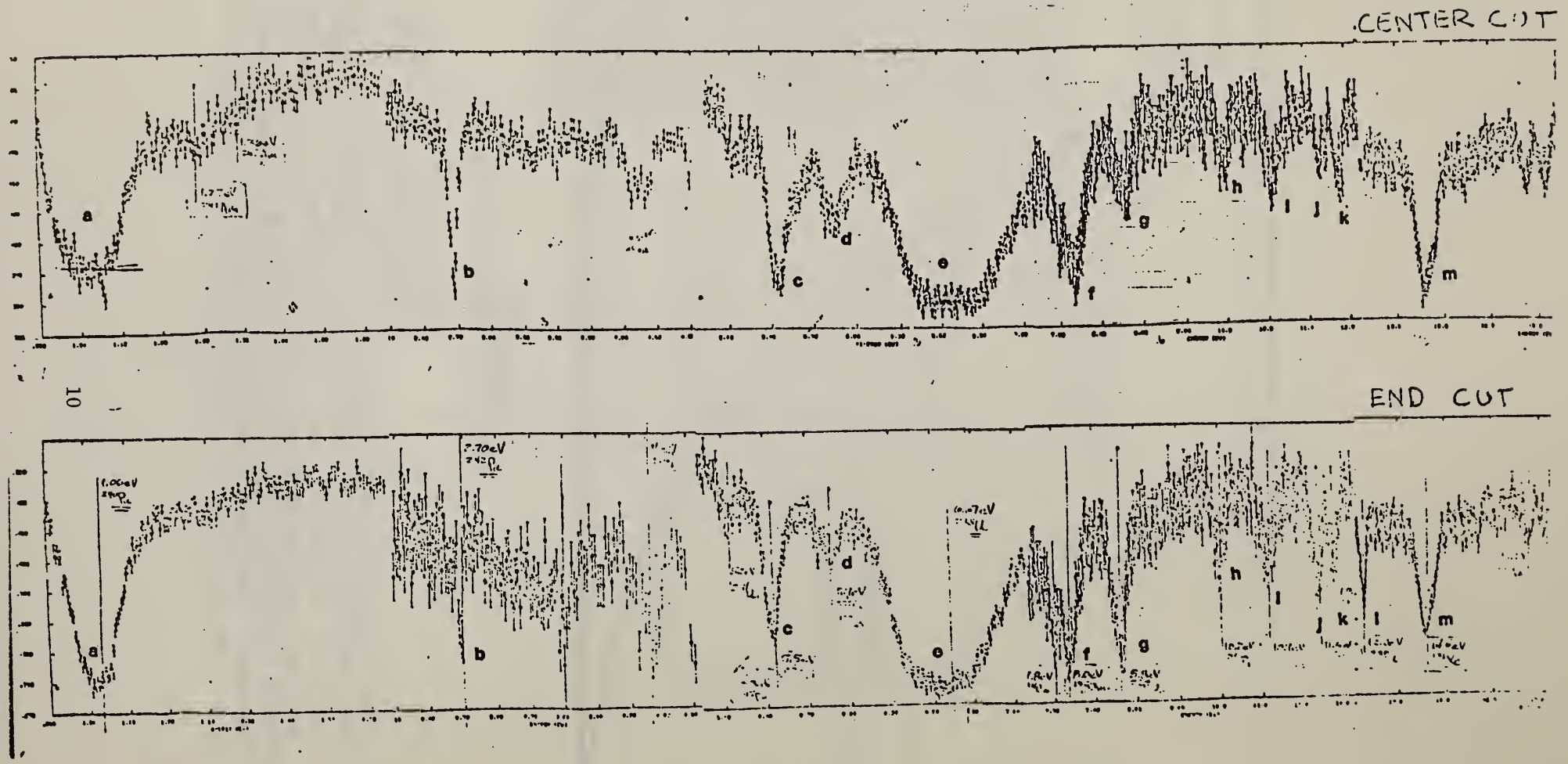


Figure 3. Neutron transmission of spent fuel samples. The upper curve shows the neutron transmission for a sample cut from the center of a spent fuel rod, the lower curve from the end of the same rod. The abscissa is neutron energy from 1.0 eV to 20.0 eV. The resonances are identified in Table 1.

Table I

Resonance in Fig. 3	Neutron energy (ev)	Atom	Remarks
a	1.06	^{240}Pu	Less in end cut
b	2.70	^{242}Pu	Much less in end cut
c	5.5	^{236}U	Less in end cut
d	5.9	^{241}Pu	Less in end cut
e	6.67	^{238}U	Same in both
f	8.0	^{152}Sm	Less in end cut
g	8.8	^{235}U	More in end cut
h	10.2	^{240}Pu	Less in end cut
i	10.9	^{239}Pu	Less in end cut
j	11.6	^{235}U	More in end cut
k	11.9	^{239}Pu	Less in end cut
l	12.3	^{235}U	More in end cut
m	14.4	^{131}Xe	Less in end cut
n	19.3	^{235}U	More in end cut

(b) Van-de-Graaff Techniques

A program to develop Van-de-Graaff based measurement systems for safeguards standards has been completed in collaboration with the Los Alamos Scientific Laboratory (LASL). Dr. Michael Meier of the Neutron Physics Group has been at LASL since December 1978. Dr. Meier has been working on two major programs during the past two years at LASL. The first of these areas was to develop a technique such that a User's Guide for the Van-de-Graaff based Small Sample Assay Station (SSAS) with the associated support hardware and software could be produced. This Guide is to acquaint potential users of the Van-de-Graaff based SSAS with the associated support hardware and software. The general operation of the SSAS is described in Ref. 1. In brief, the proton beam is pulsed with a repetition rate of 10 s^{-1}. The 100-millisecond period is divided into four intervals:

1. 0-35 ms--beam on target for neutron production, rate 10^{10}-10^{11} n/s and SSAS preamp biased off;
2. 35-50 ms--beam off, SSAS preamp biased on and gain stabilizing;
3. 50-90 ms--beam off, SAS preamp gain stabilized and scaler electronics counting delayed neutrons; and
4. 90-100 ms--beam off, SSAS preamp biased off and scaler electronics gated off.

The interrogating neutron flux is monitored by a cadmium-covered ^{3}He proportional counter (^{3}He) mounted on top of the SSAS, a cylindrical fission chamber (FC) that surrounds the sample position, a modified long counter (MLC), and the proton current incident on the neutron producing target. The first three of these signals are supplied to integral discriminators with biases set between the low pulse-height tail and the thermal peak (^{3}He counters) or fission fragment peak (fission chamber). The target current is supplied to a commercial current integrator, and the output pulses, proportional to total charge, are scaled in a scaler board [2] along with the discriminated monitors' pulse trains.

The SSAS is a very efficient detector based on 39 moderated ^{3}He proportional counters. During the time that its preamp is biased on and stable, logic pulses corresponding to ^{3}He(n,p)T events are supplied to another channel of the scaler. Samples are assayed by measuring the ratio of SSAS events in the delayed neutron counting period to the interrogating neutron flux. A rabbit system is used to transport the samples from the Van-de-Graff console to the irradiation position in the SSAS. An associated clock is started when the sample is transmitted and an "enable acquisition" dc level (EAL) shifts from zero to five volts after a preset interval in which the irradiated sample comes to equilibrium.

The Van-de-Graaff LSI-11-related hardware is comprised of the LSI-11 chassis, the interfaces and scaler board, a CRT terminal, and a LA180 printer. There are two types of software control for this system, the automatic program control and the semi-manual control by the console keyboard. The second area was the development of procedures for measuring the hydrogen content of unfired fuel rods using a ^{252}Cf hydrogen monitor and the issuing of a User's Guide for the monitor.

A ^{252}Cf hydrogen monitor has been used to determine the hydrogen contents of 144 unfired ("green") fuel rods being circulated in the New Brunswick Laboratory fuel measurement evaluation program. The monitor was calibrated with

standards fabricated by LASL analytical chemistry group CMB-1. Measurements were made relative to these standards and overall accuracies of ±3% were achieved.

A major problem in the fast neutron assay of High Temperature Gas-cooled Reactor (HTGR) unfired ("green") rods is the presence of hydrogen (H), which moderates the average energy of the interrogating flux and thereby changes the response of the system. Preliminary work has shown that this effect enhances the response by about 25% for the 150-mg H content of the rods and 10% for the 450-mg H content of the polyethylene encapsulation provided for the HTGR fuel measurement evaluation program, sponsored by New Brunswick Laboratory (NBL). To make this correction with confidence, accurate measurement of the hydrogen content must be made. To this end, the hydrogen monitor [3] has been calibrated and used to measure H content of these rods.

The rods are right circular cylinders 1.4 cm in diameter and 5.0 cm high composed of TRISO-coated UC_2 and ThO beads embedded in a carbon matrix. For the intercomparison each rod is encapsulated in a sealed polyethylene vial.

The hydrogen monitor has been calibrated for measurements in the vicinity of 600-mg hydrogen. With the current calibration and standards, the estimate accuracy is ±4.5 mg, which is based on the difference in assay results for the same unknown for two different standards and measurement precisions. Before proceeding with work that improves the precision of the monitor, more work will need to be done to remove a ±3-mg discrepancy. Likely candidates for investigation are the exact form of the response, which is likely to be more complex than the simple linear response, and use of calibration standards, which more closely match the rods.

The software and procedures to be used for calibration and running of the ^{252}Cf hydrogen monitor have been documented in a User's Guide. Computer software was developed for use with the DEC LSI-11 using the Version 3 operating system.

This work is being performed in the Center for Radiation Research under the sponsorship of DOE.

References

[1] Adams, E.L., Bourret, S., and Meier, M.M., "Small Sample Assay Station Users' Guide", to be published as LASL (LAMS) Information Report.

[2] Bourret, S., et al., "Model 516 LSI-11 Six-Channel Scaler", to be published as part of Ref. 1.

[3] Meier, M.M., and Adams, E.L., "HTGR 'Green' Rod Intercomparison - I. Hydrogen Assay", to be published as part of Ref. 1.

(c) Neutron Tomography

Another major NDA effort is to develop the methodology for high-accuracy measurements and establish reference standards using resonance neutron tomography with reactor-produced neutrons.

The efforts devoted during fiscal year 1980 for the resonance neutron tomography portion of the NRC supported nuclear safeguards program were (1) furthering and continuing development of software programming and the library data set; (2) the finalizing of hardware designs and subsequently the letting out of construction bids; and (3) the use of three low enrichment U-235 fuel pellets to experimentally verify the "resonance neutron self-indication effects" with the NBS Reactor.

The need for continuing development of software programming and the library data set was evident as Eq. 3 in the OMNT Annual Report for 1979 indicated.

$$S_k \propto \int \phi_o(E) e^{-\sum_{i=1}^{4} \sigma_{T_i}(E) X_i} \sigma_{F_k}(E) dE$$

$$\simeq S_k^o - \sum_{i=1}^{4} K_{ik} X_i + \sum_{i=1}^{4} L_{ik} X_i^2 + \sum_{i \neq j} M_{ijk} X_i X_j$$

where $S_k^o \equiv \int \phi_o(E) \sigma_{F_k}(E) dE$; $K_{ik} \equiv \int \phi_o(E) \sigma_{T_i}(E) \sigma_{F_k}(E) dE$;

$L_{ik} \equiv 1/2 \int \phi_o(E) \sigma_{T_i}^2(E) \sigma_{F_k}(E) dE$; and $M_{ijk} \equiv 1/2 \int \phi_o(E) \sigma_{T_i}(E) \sigma_{T_j}(E) \sigma_{F_k}(E) dE$.

S_k^o is proportional to the detector response with no sample.

To ensure that all X_i's would have unique values that are orthogonal to one another, i.e., X_i cannot be a function of $X_{i \neq j}$ or their combinations. This requires that the cross sections as functions of neutron energy (mainly resonance neutrons) have to be clearly and distinctively described for each and every fissile isotope involved. Our present library data set utilizes 200 cross section data points for each fissile isotope. Their effects will be determined by comparing theoretically calculated K_{ik} and L_{ik} values with experimentally measured ones which is by fitting the experimentally generated curves of fissile sample thickness vs. fissile detector reaction rate for each different type of fissile material.

In order to solve the multiple non-linear equation set of the above kind, a derivative method was used. An example involving composites of three types of isotopes is illustrated here.

$$S_k(X_1, X_2, X_3) \simeq S_k(X_1) + S_k(X_2) + S_k(X_3) - 2S_k^o + \sum_{i \neq j} M_{ijk} X_i X_j \qquad (1)$$

where X_1, X_2, X_3 are the thicknesses of three different types of fissile isotopes, and M_{ijk} is defined as before

$$M_{ijk} \equiv 1/2 \int \phi_o(E) \sigma_{T_i}(E) \sigma_{T_j}(E) \sigma_{F_k}(E) dE$$

If we introduce a filter of known thickness and type dX_1 in the neutron beam, then

$$S_k(X_1 + dX_1, X_2, X_3) \simeq S_k(X_1 + dX_1) + S_k(X_2) + S_k(X_3) - 2S_k^o$$

$$+ 2(M_{12k} X_2 + M_{13k} X_3)(X_1 + dX_1) + \sum_{i \neq j \neq 1} M_{ijk} X_i X_j \qquad (2)$$

(2) - (1)

$$dS_k(dX_1) \equiv S_k(X_1 + dX_1, X_2, X_3) - S_k(X_1, X_2, X_3)$$

$$\simeq S_k(X_1 + dX_1) - S_k(X_1) + 2(M_{12k} X_2 + M_{13k} X_3) dX_1 \qquad (3)$$

$$dS_k(dX_1) \simeq C_{k1} + 2L_{1k}X_1dX_1 + 2(M_{12k}X_2 + M_{13k}X_3)dX_1 \quad (4)$$

where $C_{ki} \equiv -K_{ik}dX_i + L_{ik}(dX_i)^2$.

The non-linear equation set (1) is now linearized

$$\frac{dS_k(dX_1)}{2dX_1} \simeq \frac{C_{k1}}{2dX_1} + L_{1k}X_1 + M_{12k}X_2 + M_{13k}X_3 \quad (5)$$

The software program for solving Eq. (5) has been incorporated into the VAX-11/780 computer.

The detail design of the reactor-based resonance neutron safeguard experimental system was completed last April and has successfully gained the approval of NBS Reactor Hazards Evaluation Committee. The NBS Shops Division is responsible for the construction of the system and the letting of bids to private contractors. The scheduled completion date is February 1981. The collimator is made up of nesting tubes of various diameters so the beam size can be changed from 1/4" (0.64 cm) to about 2 inches (5.08 cm) to facilitate faster count rates with compromised resolution for larger containers and to accommodate neutron radiography use. The retrieved collimator tubes will be stored in an auxiliary shield.

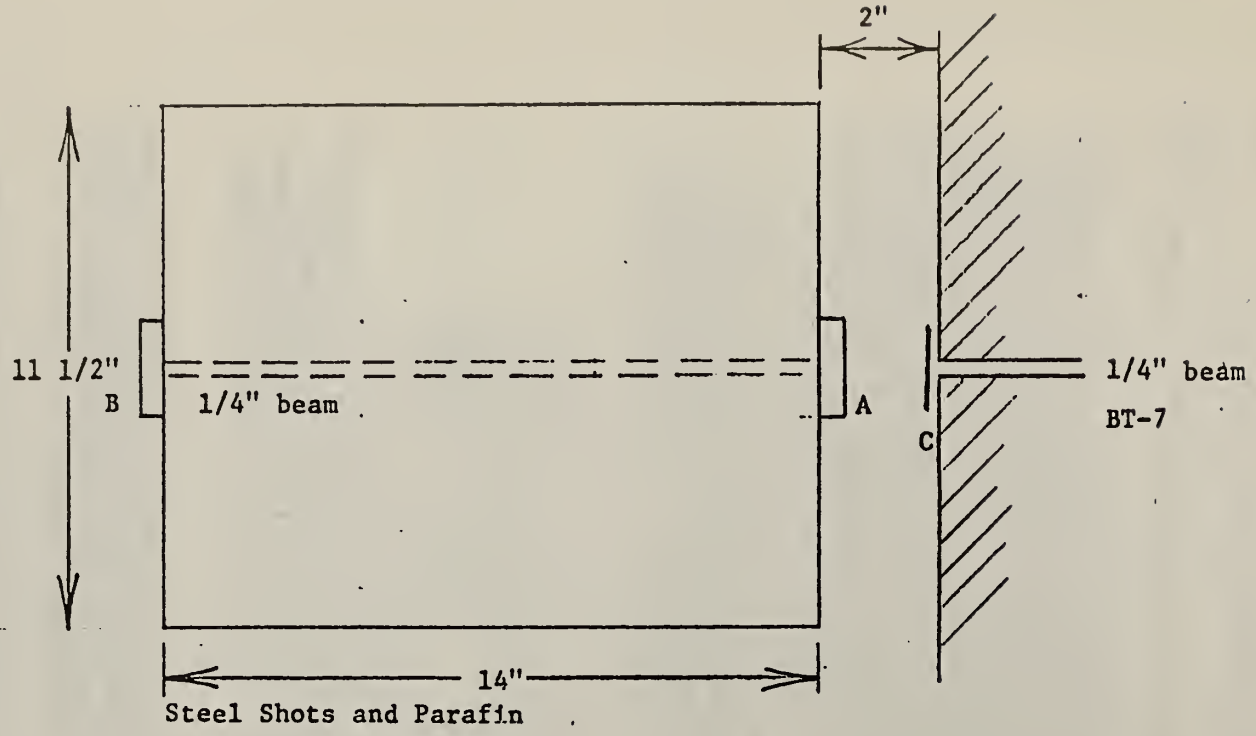
In order to verify the resonance neutron self-indication effect with the NBS Reactor facility, an experiment was conducted to quantitatively determine the U-235 and U-238 contents in three nuclear fuel pellets supplied by General Electric Wilmington Manufacturing Department. The known information is as follows:

Isotopic U-235 enrichment %	% U	Density	Length	Diameter
3.9572	88.15	95.9243	0.432"	0.410"
2.5174	88.168	96.56	0.4395"	0.410"
1.194	88.14	95.52	0.434"	0.410"

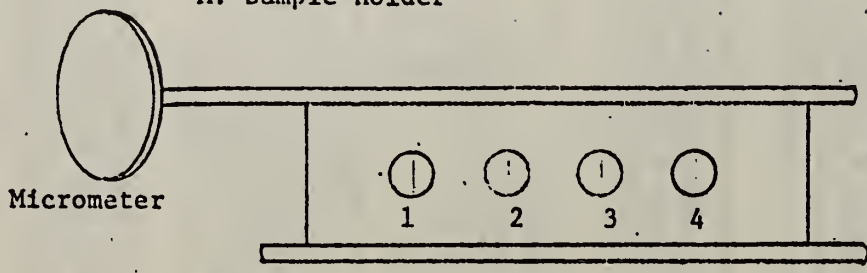
A horizontal neutron beam of 1/4" (0.64 cm) diameter was extracted from the NBSR core on port BT-7. The thermal neutron portion of the beam was removed by using a 0.04" (0.102 cm) thick Cd sheet as filter. The filtered beam was then transmitted through the fuel pellet and detected by NBS double fission ionization chamber detectors [4].

The experimental setup and fuel pellet holder are shown in Figure 4. The holder was made with a lucite block of approximately 15 cm long, 5 cm wide and 1 cm thick. Four holes were drilled to accommodate the three fuel pellets and a blank reference hole. The electronics block diagram is shown in Figure 5.

The three fuel pellets, along with the blank hole, were scanned by the neutron beam and detected with U-235, Pu-239 double fission chamber detectors and then with U-235, U-233 detectors. The reason for using Pu-239 and U-233 detectors was that the fission cross section for U-238 was very small.



A: Sample Holder



Lucite block 15 cm x 5 cm x 1.07 cm diameter of the four holes 1.04 cm top and bottom of lucite are covered by .025' Al sheets. The position of the lucite will be controlled by a micrometer. Holes #2, #3, and #4 will contain the pellets.

B: Fission Detectors
NBS double chamber fission detectors
U-233, U-235 and Pu-239 3 mg/cm^2 thick

C: Cd Sheet .102 cm thick

Figure 4. Neutron Telescope and Fuel Pellet Holder Assembly

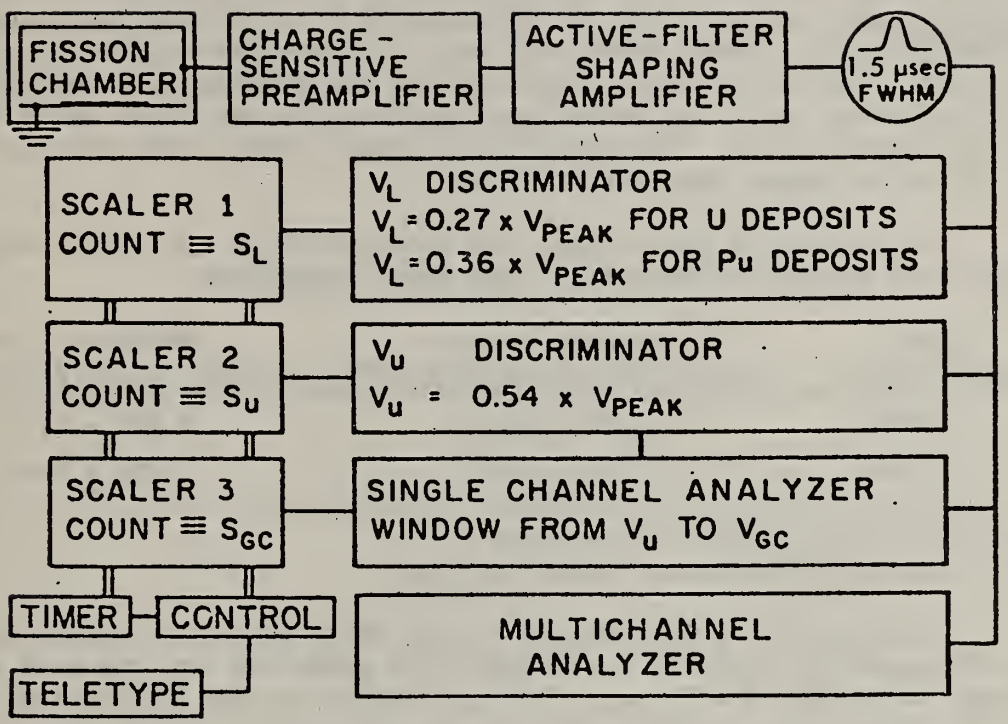


Figure 5. Each side of the dual fission chamber is monitored independently by a triple-scaler pulse-processing system as shown here. The integral discriminator counts, S_L and S_U, are the primary (redundant) counting data. The relative positions of the discriminator levels V_L and V_U to the peak of the pulse-height distribution, V_{PEAK} are established with the aid of the multi-channel analyzer. The SCA count S_{GC} provides a sensitive gain check; $V_{GC} = 1.37\ V_{PEAK}$.

A count rate of $\gtrsim$ 1000 cps was obtained using very thin fission foil in all detectors, thus no corrections were made for detector dead time, self-absorptions, and background noise. A total count of $\gtrsim$ 50,000 was accumulated for each data point.

The equation for U-235, U-238 mixed composition sample says

$$S_k \simeq S_k^o - K_{ik}X_i + \sum_{i=1}^{2} L_{ik}X_i^2 + \sum_{i=j} M_{ijk}X_iX_j \qquad (6)$$

where i can be U-235 or U-238 and K can be U-233, U-235 or Pu-239.

If the neutron flux $\phi_o(E)$ is known, one can then calculate the coefficients S_k^o, K_{ik}'s, L_{ik}'s and M_{ijk}'s, and thus evaluate U-235 and U-238 contents in all three pellets using the measured S_k values from either the U-235, Pu-239 detector set or U-235, U-233 detector set. Another approach is using two fuel pellets as known standards, and if we further neglect the contributions from the second order terms, then one can experimentally determine the various K_{ik}'s and thus the first order approximation for the U-235 and U-238 contents in the third pellet. The dropping of second order terms will introduce an error $\sim$ 15% for a sample thickness of 1 mm.

The following table lists the measured U-235 and U-238 contents in grams in the third pellet as compared to the known quantities.

	Known quantities*	Measured with U-235, Pu-239 detectors**	Measured with U-233, U-235 detectors**
U-235	0.2174	0.185 $\pm$ 0.03	0.193 $\pm$ 0.03
U-238	5.2764	5.581 $\pm$ 0.05	5.476 $\pm$ 0.49

*Corrected for 1/4" neutron beam size.
**Errors were estimated second order effects.

The measured U-235 content in the third pellet is about 11% of the known U-235 quantity when the U-233 and U-235 detectors set are used and about 15% with the U-235, Pu-239 set. The accuracies for the measured U-238 content in the third pellet are 4% and 6%, respectively, for the two sets of detectors. The fact that all neutrons with energies below 0.5 eV were filtered out in the beam caused the Pu-239 detector to perform less satisfactorily relative to U-233 and U-235 detectors, since 60% of the resonance strength for Pu-239 lies in the 0.3 eV region. Nevertheless, the results indicate a reasonably accurate estimation of U-235 and U-238 contents can be obtained in a very short time ($\lesssim$ 1 min.) with the self-indication method with counting statistics of 0.1%.

One also sees the need for incorporating the second order terms. Presently we are preparing a set of standard Pu-239, U-235, and U-238 discs of various thicknesses from NBS Standard Reference Materials which will be used in an upcoming experiment to measure $\phi_o(E)$ as well as these first order and second order coefficients. This will afford a much more stringent test of our methodology and reduce the measurement uncertainty to less than 1%.

In summary, it would require approximately one day to inspect a nuclear waste container approximately the size of a gallon paint can. This conclusion assumes that it would require 110,000 transmission measurements with a resolution of 4 mm and 3% counting statistics. The assumption is made that the container will be scanned at 64 points in a plane orthogonal to its axis. The container will be rotated in two-degree increments following each orthogonal 64-point scan. Following each complete 180° rotation cycle, the container will be stepped axially and the process repeated for a total of 20.

The amount of time needed for the entire interrogation, together with resolution and counting statistics, can be greatly improved by incorporating a linear series of detectors and a reactor power of 20 MW.

This program is being carried out in the Center for Materials Science under the sponsorship of NRC.

References

[4] Grundl, J.A., Gilliam, D.M., Dudey, N.D., and Dopek, R.J., Nucl. Tech. 25, 237 (1975).

(d) Calorimetry

The overall goal of the calorimetric activity is to provide measurement assurance and traceability to National Standards for calorimetric measurements made in the industrial assay of plutonium-bearing solids. This activity aims at the establishment of an automated calorimetric measuring system which will be used to periodically check the certified power of encapsulated plutonium heat sources submitted by major industrial producers.

Major steps were taken in FY 1980 to realize the NBS goal of providing traceability to National Standards for calorimetric measurements made in the industrial assay of plutonium-bearing solids. These included (1) further testing of a Mound-designed heat-flow calorimeter, together with an automated data acquisition system (this included power measurements on two Mound-fabricated, encapsulated plutonium heat standards); (2) design and start of construction of an NBS heat-flow calorimeter; and (3) preparation of an NBS "General Reference Material" Report of Investigation to accompany Mound-calibrated plutonium heat standards.

(1) The twin-bridge, heat-flow calorimeter obtained previously on loan from the Mound facility was tested in measuring the total decay power of two calibrated plutonium heat sources (nominal powers 0.25 W and 4.0 W, respectively). The sources were loaded automatically, and calorimeter data were recorded with an automated data acquisition system built around a Hewlett-Packard* 9835 desktop computer. The NBS power data for both sources had a range of 20 ppm and a standard deviation of about 10 ppm. They agreed with the Mound predicted powers for these sources within the combined measurement uncertainties of NBS and Mound.

(2) Design specifications for an NBS twin-bridge, heat-flow calorimeter were developed, using NBS experience with the Mound calorimeter as a guide. While the gross physical features and operating principles of the NBS calorimeter follow the Mound design, some changes in construction materials, such as use of stainless steel for all submarine parts and use of O-rings instead of gasket seals, were used to improve the reliability. In addition, the calorimeter was designed to by symmetrical, so that either of its two sensing chambers could be used as a reference chamber. The main calorimeter body has been completed, and we are presently constructing the thermal sensing elements, including heaters and resistance thermometers.

(3) In order to establish traceability to national standards for calorimetric heat-flow measurements in assaying radioactive materials, a service similar to the one offered by NBS to the radiopharmaceutical industry has been proposed. This is essentially a Measurement Assurance Program (MAP) in which traceability to national standards is established through a programmed exchange of measurement artifacts between major suppliers (in this instance, of calibrated radioactive heat sources) and NBS. As a first step in this direction, power measurements have been carried out on three Mound-calibrated heat standards in an NBS ice calorimeter, and it has been determined that the NBS and Mound

*Certain commercial equipment, instruments, or materials are identified in this paper in order to adequately specify the experimental procedure. In no case does such identification imply recommendation or endorsement by the National Bureau of Standards, nor does it imply that the material or equipment identified is necessarily the best available for the purpose.

measured powers are in agreement within their combined uncertainties for all three standards. Cooperative efforts between Mound and NBS scientists, together with the Office of Standard Reference Materials, have produced a "General Reference Materials" Certificate (GM-10) which documents these NBS/Mound intercomparisons. Mound will be authorized to distribute this certificate (together with its own calibration certificate and data), along with each newly-calibrated source it provides. A copy of the NBS certificate for GM-10 appears as Figure 6. As soon as the NBS heat-flow calorimeter is operational, a formal MAP between Mound and NBS covering calibrated plutonium heat standards will be started.

This work is being carried out in the Center for Thermodynamics and Molecular Science under the sponsorship of NRC and DOE.

(2) Standard Reference Materials for NDA

(a) U-235 SRMs

A major portion of the effort in this Sub-Task has continued to be the development of the low-enriched U_3O_8 NDA Gamma-Ray Spectrometry NBS SRMs. This effort has been a collaborative one with NBL, LASL, NBS and the Euratom Laboratories of JRC-Ispra, CBNM and KfK. During November 1979, a meeting was held at CBNM in Geel, Belgium, to review the results from each organization on the pre-experiment study, as well as to finalize the procedures and measurements needed for the certification of the SRMs.

The final design of the aluminum containers for the material was approved (Figure 7), and the production of the 900 cans needed was done by the NBS Shops. Upon completion, these containers were then shipped to CBNM for critical dimension measurements and filling with low-enriched U_3O_8. The filling of containers with the ^{235}U isotopic standards began with the 0.318% material, and continued in the order of increasing enrichment (0.7128%, 1.943% and 2.950%). The 4.5% enriched material, which is to be included within the set of Reference Materials, has not arrived at CBNM.

Prior to the canning of the material, 13 (1-gram) samples were taken from each isotopic enrichment batch for mass spectrometry analysis, and one (20-gram) sample was taken for the $^{232}U/U$ determination by gamma-ray spectrometry. In addition, each 20-gram sample will be sent to NBL for uranium assay. A set of ultrasonic devices will be placed in the top of each can after filling, and then a representative number of each batch will be sent to NBS for critical evaluation by gamma-ray spectrometry.

Effort also has been devoted to investigating the contribution of ^{238}U decay daughter products (^{234}Pa and ^{234m}Pa) to the 185 keV gamma-ray line observed from low-enriched ^{235}U samples. This study has continued through the use of uranium oxide samples of known ^{235}U isotopic abundance. Gamma-ray spectroscopy has been performed on 11.5-gram samples whose ^{235}U isotopic abundances ranged from 0.0004% to 0.7198%. This isotopic range was chosen because the contribution from ^{234}Pa and ^{234m}Pa is most significant.

In addition to the gamma-ray spectroscopy, these samples are to undergo chemical separation, since secular equilibrium exists. The uranium is separated from its decay daughter products, thorium and protactinium. After separation, the two fractions (uranium and thorium-protactinium) are then measured by gamma-ray spectrometry. By monitoring the uranium fraction over

Figure 6

Special Reference Material Report

GM 10

Encapsulated Plutonium Heat Source

Introduction

The enclosed encapsulated plutonium heat source provides a convenient, measured standard for calorimeter calibration [1] in the calorimetric assay of plutonium-bearing solids. This report summarizes the NBS measurements made on this reference material. The reference is produced and certified by the Mound Facility*, Monsanto Research Corporation, Miamisburg, Ohio.

Description of Source

This heat source, fabricated and calibrated at the Mound Facility, consists of a doubly encapsulated sample of plutonium-oxide. The sealing of both inner and outer capsules has been carried out using documented and reproducible welding procedures. Nondestructive testing (radiography and He leak tests) has been conducted to ensure the integrity of each capsule. Since the active materials are largely alpha-particle emitters, helium gas will accumulate within the inner capsule. The capsules have a conservative design life of five years. The lack of long-term compatibility and creep data for the materials of construction precludes the assignment of longer working lifetimes to the sources at this time. Therefore, it is strongly recommended that this source be returned three years from the date of receipt to the Mound Facility for reevaluation and recertification. This source should be handled with the normal precautions for alpha-particle emitters and stored in the container provided when not in use.

Certification

This source has been calibrated at the Mound Facility using Mound heat-flow calorimeters and electrical standards traceable to the National Bureau of Standards. A separate certificate issued by the Mound Facility accompanies this reference material. Selected Mound plutonium heat sources similar to the one accompanying the Mound certificate have been independently measured in a precision Bunsen ice calorimeter at NBS. The calibration results accompanying that certificate are in the form of computer printout and microfiche giving the decay power calculated from these measurements, day-by-day, for a three-year period after the calibration. Also included are the physical parameters of the source including materials of construction and measured radiation dose rates.

*Mound Facility is operated by Monsanto Research Corporation for the U.S. Department of Energy under Contract No. DE-AC04-76-DP00053.

Figure 6 (continued)

NBS Measurements on Mound-Encapsulated Sources

Each measurement in a precision Bunsen ice calorimeter reflects the average (unweighted) of at least 10 independent determinations of heat flux (power) for a single source. Three such measurements were carried out on Mound sources of nominal power, 0.23W, 1.5W, and 1.0W, and are summarized below. The first two are documented extensively in [2].

	Mound Source Designation		
	0.23WB	1.5WB	1.0WK
Mound Predicted Power[a] [W]	0.22544	1.44942	0.97027
NBS-Measured Power[a] [W]	0.22517	1.44936	0.96996
Number of NBS Measurements	16	19	11
s_m[b] [W]	0.00005	0.00004	0.00034
Estimated NBS Overall Uncertainty[c] [W]	0.0003	0.0007	0.0014

[a] Individual data for each source calculated for a single time mid-way through the measurements series for that source.

[b] s_m = Computed standard deviation of the mean.

[c] Sum of maximum conceivable systematic errors and 99% confidence limits for the mean.

At NBS measurements were carried out by D. Ditmars of the Chemical Thermodynamics Division and at Mound Facility by K. C. Jordan, Senior Research Specialist.

Packaging and reshipment of this source to the Mound Facility for testing and recalibration must follow all applicable Department of Transportation, U.S. Department of Energy and/or U.S. Nuclear Regulatory Commission regulations. All shipments should be addressed to:

C. L. Fellers, Group Leader
Safeguards Research and Development
Monsanto Research Corporation
Mound Facility
P.O. Box 32
Miamisburg, Ohio 45342

[1] ANSI Standard N15.22-1975, "Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control", available from American National Standards Institute, 1430 Broadway, New York, New York 10018.

[2] D. Ditmars, Intl. J. Appl. Radiat. Isotopes 27, 469 (1976).

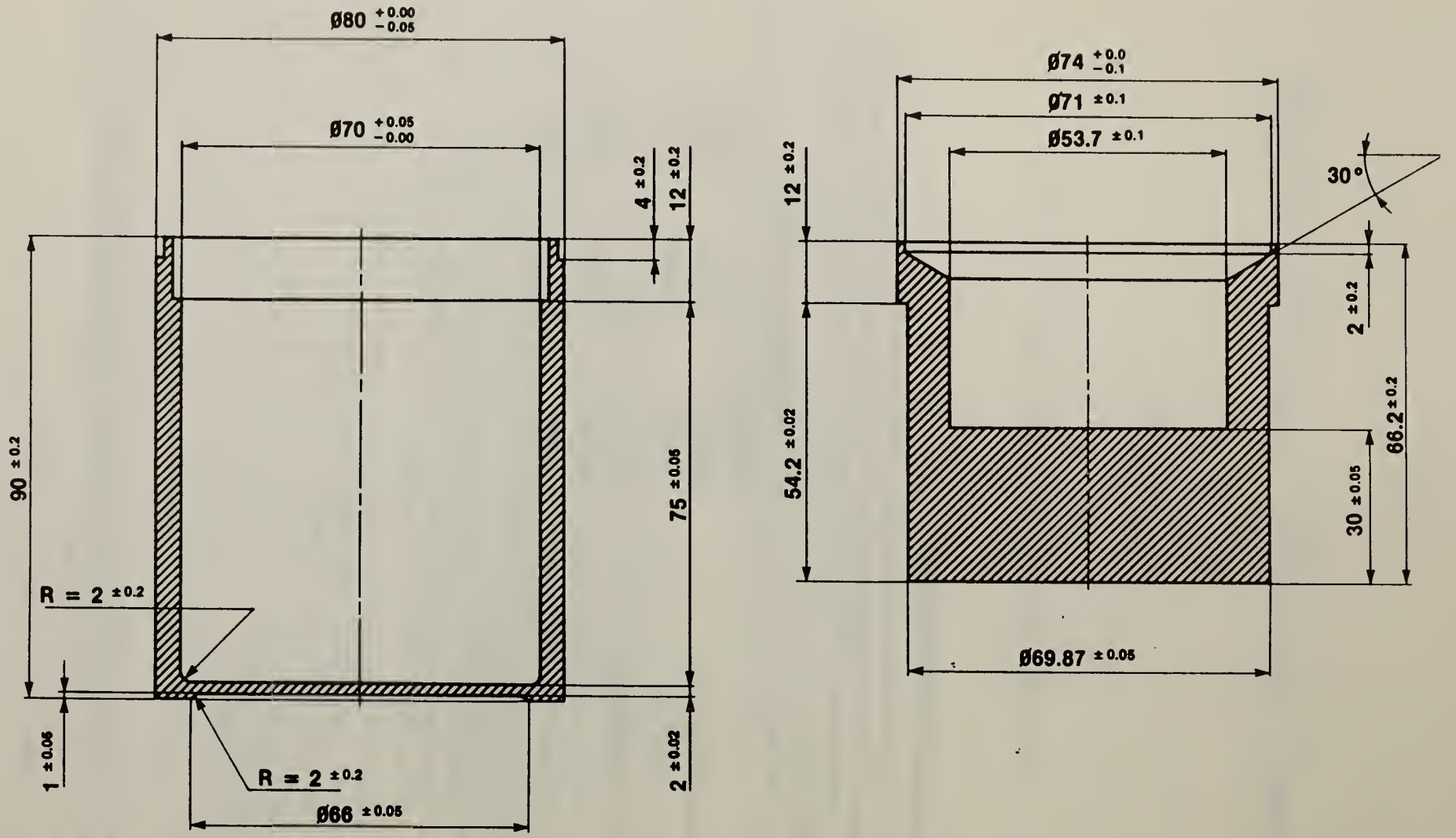


Figure 7. New Aluminum Container Design Drawings

time, an indication of the 185 keV gamma-ray line buildup reflects the protactinium interference due to ^{238}U decay, while the thorium-protactinium fraction provides more quantitative information based on the protactinium decay.

This work is being carried out in the Center for Analytical Chemistry under the sponsorship of NRC and DOE, with statistical help from the Center for Applied Mathematics.

A commercially available, pure-germanium detector has been ordered, evaluated, and accepted. This detector is presently being calibrated and further tested for stability in preparation for its use in the certification of the NBS Plutonium Isotopic Reference Materials, particularly the NDA standards.

Much discussion and information collection has been carried out, both individually and at group meetings, to determine more accurately the needs of the national (and, to a lesser extent, international) safeguards community for plutonium Standard Reference Materials (SRMs). A meeting of representatives from all of the major U.S. facilities involved was held at NBS on October 15 and 16, 1980. At that meeting the needs and position of the U.S. safeguards community were defined in preparation for a later meeting to discuss joint international efforts to certify these SRMs on an international basis.

This work is being carried out in the Center for Radiation Research under the sponsorship of DOE, with statistical help from the Center for Applied Mathematics.

(3) Data Development and Calibration

(a) Radioactivity Program

The half-life of ^{241}Pu has been followed by measuring the ingrowth of ^{241}Am in a sample of ^{241}Pu purified in March 1977. $4\pi\alpha$(LS)-γ-coincidence and -anticoincidence counting methods have been used. The results to date remain consistent with a half-life of 14.35 $\pm$ 0.15 y. It is planned to measure the sample periodically for several more years.

The half-life of ^{240}Pu is being determined from measurements of the total alpha-particle-emission rate of a well-characterized sample of $^{240}PuO_2$. This $^{240}PuO_2$, prepared at the Los Alamos Scientific Laboratory, has been provided to several national laboratories for measurement of the ^{240}Pu half-life as part of their participation in the DOE Half-Life Evaluation Committee. The results of the measurements are to be published jointly, as was the case with the previous ^{239}Pu half-life study.

This work is being carried out in the Center for Radiation Research under the sponsorship of DOE, with statistical help from the Center for Applied Mathematics.

(b) Fission Track Monitor Evaluation

One of the activities completed within NBS during the last fiscal year was the field testing and evaluation of a fission track reactor power monitor for the International Atomic Energy Agency (IAEA). The purpose of this task was to evaluate the capability of the track-etch monitor for providing an independent record of the nuclear reactor's power history and to provide information concerning the neutron energy spectrum.

The track-etch monitor evaluated was an externally-powered device designed to record the neutron flux from operating reactors and similar sources for extended periods of time. A strip of 35 mm wide polymer tape is drawn past a group of fissionable deposits which have been exposed to the local neutron flux. A fraction of the fission fragments generated penetrate the tape producing radiation-damaged tracks that are subsequently etched and counted to permit a reconstruction of the temporal flux history to which the monitor was exposed. Each monitor is equipped with sufficient tape to cover a year of reactor operation time. The monitor is designed to operate at a neutron flux of 10^6 $ncm^{-2}s^{-1}$.

In one of the actual field tests, a monitor was installed above the shroud at the Maine Yankee Power Reactor in Wiscassett, Maine, for ten months. During this period of time, the 855 Megawatt reactor underwent four unscheduled shut-downs and over eight changes in reactor power-levels. The 14.5 meter tape was removed from the monitor, chemically etched to reveal the fission tracks, which were measured. The investigation of the tape indicated that the monitor was able to record the changes in reactor power but had suffered several mechanical malfunctions, and furthermore was location-dependent for optimum neutron flux measurement. A second monitor was placed in the Wisconsin Public Service Corporation's Kewaunee Nuclear Power Plant for a shorter period of time, and similar information was obtained.

The final conclusion reported to the IAEA was that the monitor would technically perform the functions that were required of it; however, it would not be completely reliable due to frequent mechanical failure. The IAEA was also advised that the only means to achieve the unattended monitoring of power reactors was to completely redesign the motor-gear train assembly of the monitor.

This work is being performed in the Center for Analytical Chemistry under the sponsorship of the International Safeguards Project Office (ISPO) at the Brookhaven National Laboratory.

B. Destructive Analytical Chemistry

The goal of the Destructive Analytical Chemistry Task is to provide standardization for wet chemical and mass spectrometric methodology used for the assay of nuclear materials. The research of this Task is divided into three major components:

- Research to produce and certify Standard Reference Materials (SRMs)
- Support for Measurement Assurance Programs (MAPs)
- Chemical and isotopic measurements to support NDA

When requested, and as needed, assistance is provided to other domestic laboratories in government and industry. Assistance is also provided to other countries and to international and/or multinational organizations in the accurate measurement of certified reference materials.

(1) Standard Reference Materials

The research, development and production necessary to certify and distribute an NBS SRM generally requires the cooperative effort at NBS of Program Offices, Centers and Divisions. In addition, there is usually a need for support of materials fabricators and of other academic, government or

industrial laboratories. This approach is beneficial, cost effective and conserves the limited resources available for standardization by utilizing the best scientific talent available.

(a) ^{244}Pu Spike SRM

Approximately 900 bottles containing 1 mg of ^{244}Pu spike have been prepared at Los Alamos Scientific Laboratory (LASL). Measurements have been made at NBS using isotope dilution mass spectrometry (IDMS) to determine the isotopic composition and elemental concentration of this material. These measurements and those made at New Brunswick Laboratory (NBL) by coulometry and at LASL by coulometry and IDMS will be used to certify the ^{244}Pu as an NBS Standard Reference Material. All measurements have been completed.

NBS analyzed seven ^{244}Pu samples. Each was spiked with one of three SRM 949e (plutonium assay standard, 97.456% ^{239}Pu) solutions prepared for this spike calibration and equilibrated by reduction and oxidation of the mixed solutions. This work was done by LASL and NBS personnel at LASL. Prior to mass spectrometric analysis at NBS, the samples were purified by ion exchange chromatography to remove daughter products, primarily ^{238}U and ^{241}Am, which represent potential mass spectrometric isobaric interferences.

A pulse-counting detector was installed in a standard NBS-design thermal ionization mass spectrometer, replacing the conventional Faraday cage detector, in order to make high sensitivity measurements requiring only nanogram quantities of plutonium per determination. Thus, the radiation hazard and instrument contamination was reduced by greater than a factor of 1,000 relative to the conventional triple-filament mass spectrometric procedure which requires 4 μg of sample per determination.

The pulse-counting measurement system consists of a 17-stage electron multiplier of Swiss manufacture with the "Rajchman" structure and Cu-Be conversion dynode, a high-speed non-overloading discriminator-amplifier, and a high-speed commercial counter. The voltages to the dynodes of the multiplier are fed off a high-impedance divider string external to the vacuum and supplied by a commercial power supply. The theoretical deadtime of the system as calibrated by a double-pulse generator with nanosecond pulse capabilities was verified by an empirical calibration utilizing the uranium isotopic SRMs.

The mass spectrometric loading technique utilized was an electrodeposition process in which the plutonium is plated directly onto the sample filament. Approximately 0.1 to 0.2 ng of Pu was plated onto the filament in a reproducible fashion as evidenced by the mass spectrometric behavior observed throughout the analytical program. This amount was determined by high-efficiency α-counting measurements made on representative filaments.

The correction factor for isotopic fractionation was determined from the analysis of plutonium isotopic SRMs 947 and 948. Multiple determinations were made throughout the spike calibration program to guarantee control of the measurement process and provide an indication of measurement precision. The correction factor for fractionation determined from the ^{240}Pu/^{239}Pu certified/experimental values for the standards was applied to all experimental ratios. The compositions of the SRM 949e spike material and ^{244}Pu material were determined by duplicate mass spectrometric analyses of three samples of each material. Duplicate determinations of the seven spiked ^{244}Pu samples were also made. A total of 54 mass spectrometric analyses were made using an estimated 5-10 ng of Pu. Conventional procedures would have required 200 μg.

The grand average of all calculated concentrations (1.9948 ± 0.0012 μM/mL) indicates a precision in ratio measurement of ∿ 0.1%, which is consistent with precision of standards measurements and experimental ratios. The dominant error in this measurement procedure is due to the extrapolation of the correction factor determined on the $^{240}Pu/^{239}Pu$ ratio of the standards to the $^{244}Pu/^{239}Pu$ ratio of the unknowns. This factor increases the confidence limit for the result to 0.16%. The results of the collaborating laboratories are summarized in the following table.

Comparison of Results
^{244}Pu Concentration

Lab	Technique	Samples	Determinations	Value	Measured S.D.
NBS	IDMS (^{239}Pu Spike)	7	16	1.9947	±0.0012
NBL	Coulometry	9	9	1.9941	±0.0004
LASL	Coulometry	5	5	1.9944	±0.0015
	IDMS (^{242}Pu Spike)	1	6	1.9925	±0.0014

This work is being performed in the Center for Analytical Chemistry under the sponsorship of DOE.

(b) ^{233}U Spike SRM

Approximately 400 ampoules of a ^{233}U spike, SRM 995, have been prepared containing 5 mg U in 10 mL of HNO_3 (1 + 19). Before ampouling, the uranium was purified by anion exchange chromatography to remove ^{229}Th and other daughter products.

The concentration of uranium in the ampoules of the SRM were determined by thermal ionization isotope dilution mass spectrometry following a sampling plan supplied by the NBS Center for Applied Mathematics: eight ampoules were chosen for use in the assay of the SRM. Separate aliquots from each of the ampoules were spiked with known amounts of ^{235}U (SRM 993) and uranium metal (SRM 960, assay standard). In addition, eight aliquots of SRM 993 were spiked with SRM 960 as a cross check on the assay of these materials. Thus, all three SRMs have been assayed against each other.

The statistical design of the experiment allowed for the assessment of the following parameters: ampoule-to-ampoule variations among the ^{233}U samples, sample-to-sample variations among the ^{235}U and natural uranium metal spikes, inconsistency in the concentration previously assigned to the ^{235}U spike SRM, and finally the measurement error in the concentration of the ^{233}U spike SRM. No ampoule-to-ampoule or sample-to-sample variations were detected from the statistical evaluation of the data. The concentration of ^{233}U spike (SRM 995) was found to be 489.93 μg U/g with an uncertainty at the 99% confidence interval of ±0.037%. This uncertainty can be divided into a ±0.011% random error component and a ±0.026% possible systematic error. The isotopic composition of SRM 995 was also determined mass spectrometrically using both Faraday cage and ion counting detection systems. The ^{233}U content of the SRM was found to be 99.9245 ± 0.0006 atom percent.

This work is being performed in the Center for Analytical Chemistry under the sponsorship of NRC, with statistical help from the Center for Applied Mathematics.

(2) Methodology for Chemical Assay of Nuclear Materials

High-precision and high-accuracy chemical assay techniques are essential if there is to be adequate safeguarding of the nuclear materials in the fuel cycle. These techniques, in conjunction with well characterized SRMs, help provide a sound base for materials measurement and accountability. Chemical assay procedures are also essential to the preparation of high-accuracy primary reference materials of known isotopic composition for mass spectrometer calibration.

(a) Moisture in PuO_2

The experimental phase of this Sub-Task was completed on schedule in this fiscal year, culminating the work on moisture in plutonium dioxide (PuO_2).

An investigation was made of vessels, lids and septa for relatively short-term storage of methanol containing water extracted from metal oxides or other materials; storage prior to titration of the methanol-water mixture is convenient. The two systems of most promise are the Reacti-Vial System* consisting of borosilicate glass vials (5 mL capacity) or flasks (10 or 25 mL capacity) and Teflon vial closures with Mininert* valves; and Vacutainers* (10 mL capacity), partially evacuated test tubes with rubber stoppers. The Reacti-Vial System is satisfactory, but care must be taken to leak-test the valve. The most satisfactory of the Vacutainers investigated is the Becton-Dickenson* No. 6440, with uncoated inner surface and glycerine-lubricated pink rubber stopper. The Vacutainer does contribute water vapor from the head space; the contribution of water from the pink stopper is much less than that from the red stopper in more recent use.

The paper, "Telescopic Viewer in Syringe Calibration", appeared in the February 1980 issue of Analytical Chemistry. A manuscript, "Calculation of Solvent-Water Mixture Volumes", has been accepted for publication in Analytical Chemistry. The manuscript presents a procedure which enables calculation of the ratio of the volume of a solvent-water mixture to the sum of the volumes the components would occupy separately; expressions have been derived for the general case and have been applied to the determination of water by Karl Fischer titration of water extracted from various substances into methanol which is then introduced into a titration vessel volumetrically.

A manuscript, "Buoyancy Correction and Weighing on Analytical Balances", by R.M. Schoonover and F.E. Jones, has been submitted to Analytical Chemistry. This manuscript treats, in detail, buoyancy corrections to be applied to weighing on analytical balances. These corrections are applicable to the several weighings involved in the Karl Fischer titration determination of moisture in PuO_2.

A manuscript reviewing the moisture in PuO_2 problem and discussing moisture determination methods is in preparation.

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*Certain commercial equipment, instruments, or materials are identified in this paper in order to adequately specify the experimental procedure. In no case does such identification imply recommendation or endorsement by the National Bureau of Standards, nor does it imply that the material or equipment identified is necessarily the best available for the purpose.

A visit was made to the Monsanto Mound Laboratories at Miamisburg, Ohio, to discuss the possibility of making moisture measurements (by Karl Fischer titration) on PuO_2 at Mound. The suggestion was very well received by Mound personnel, and there appeared to be no insurmountable technical problems. Contacts were then made to acquire PuO_2 on which to make the measurements. It was decided that the better course of action would be to make the measurements at a facility which had proximate access to an excellent source of supply of several different batches of PuO_2. Consequently, a visit was made to the Hanford Engineering Development Laboratories (HEDL) operated for DOE by the Westinghouse Hanford Company at Richland, Washington, to arrange to acquire several samples of PuO_2 from the Rockwell Hanford Operations, and to make the measurements at HEDL. In preparation for the measurements, Karl Fischer titration apparatus was modified for use in a glovebox and other accessory equipment was acquired.

Measurements of the moisture content of PuO_2 were made by Karl Fischer titration on three 15-gram samples from three different suppliers: Rockwell Hanford Operations (RHO) (sample designated Pu 113); the Savannah River Plant (Pu 124); and Los Alamos Scientific Laboratories (Pu 135). The moisture content of 200-milligram samples of the same oxide was determined using a Model 902H Moisture Evolution Analyzer operated at 400 °C. The Karl Fischer work in the glovebox and the Moisture Evolution Analyzer work were done by a HEDL technician. Each of the 15-gram samples was milled for 15 minutes in 25 milliliters of methyl alcohol in a Spex Mixer/Mill vial containing three tungsten carbide pellets supplied by RHO. As a blank, 25 milliliters of methyl alcohol was similarly milled. A methyl alcohol-water mixture prepared at NBS and injected into Vacutainers was used to standardize the titrator. The Karl Fischer (KF) titration measurements were carried out very successfully; the mean values of the moisture content and the corresponding relative standard deviation of the mean are shown in the table below. The values determined using the Moisture Evolution Analyzer (single observations) are also shown in the table. Since the MEA result for Pu 113 was much lower than the Karl Fischer result, HEDL subsequently made MEA measurements on two samples taken from different places in the can; the results were 0.0735% and 0.0750%. Since the KF and MEA results on Pu 124 and Pu 135 were in satisfactory agreement, it is apparent that the milling and extraction in methyl alcohol removed more moisture from Pu 113 than did the oven at 400 °C in the MEA. The results of the KF measurements will be presented at the January 1981 meeting of ASTM Committee C26 at Ft. Myers, Florida, with the ultimate intent of having the KF procedure included in an existing ASTM Standard.

Measurements of the Moisture Content of PuO_2

	KF Titration		Moisture Evolution Analyzer
	Mean Value of Moisture Content (%)	Standard Deviation of Mean	Mean Value of Moisture Content (%)
Pu 113	0.3008	0.0043	0.0857
Pu 124	0.7490	0.0078	0.704
Pu 135	0.4702	0.030	0.500

This work is being performed in the Center for Mechanical Engineering and Process Technology under the sponsorship of DOE.

(b) Resin Beads

High sensitivity isotopic measurements of uranium and plutonium by thermal ionization mass spectrometry using an anion resin bead as the emitter has been investigated. The use of the resin bead has been studied for its promising advantages in facilitating shipping, minimizing sample size, and sample handling in the isotopic analysis of uranium (U) and plutonium (Pu) for required nuclear safeguard measurements. Some of the specific advantages of the resin bead technique result from (1) the selective absorption by the beads of U and Pu from the fission and actinide produces in dissolver solutions, (2) the ability to load and analyze beads directly in the mass spectrometer, and (3) high ion yield from the bead matrix which markedly reduces the requires sample size and thus the potential radiation hazard.

The NBS effort in the U and Pu isotopic analysis program has been directed at the critical evaluation of the resin bead technique with respect to achievable precision and accuracy. Factors which affect this are ionization efficiency, ion beam stability, fractionation control, and isobaric interferences. The parameters associated with the loading of U on resin beads have been investigated for the purpose of producing reference bead materials. The loading procedure has a demonstrated affect on loading efficiency (amount of U per bead divided by the total bead capacity) and ionization efficiency, parameters which critically affect mass spectrometric reproducibility and precision. These investigations have required the development of individual bead assay procedures, batch assay procedures, and extensive mass spectrometric investigations.

As a part of the NBS effort, a round robin has been organized to introduce laboratories which have the capability of making high sensitivity measurements to the resin bead technique and to receive, in turn, an indication of measurement state-of-the-art as related to U and Pu resin bead analysis. The response to requested participation in the round robin was excellent, and 12 laboratories agreed to participate. The laboratories have been sent the first set of samples and partial results have been received. These first samples contained U at the 30 ng and 300 ng levels. The second samples will contain U at the 2 ng level and the third set of samples will contain Pu at the 2 ng level. A fourth set of samples containing both U and Pu at the ng level is planned.

The resin bead program has been operated in close contact with Oak Ridge National Laboratory (ORNL), which is the principle promoter and active user of the technique. A set of resin beads containing both U and Pu were prepared at ORNL and independently analyzed by both ORNL and NBS. These beads contained material representative of 30,000 megawatt-days burn-up from a LWR and had been spiked with ^{242}Pu and ^{233}U for isotope dilution analysis. A set of the beads has been forwarded to the IAEA in Zeibersdorf, Austria, for calibration and standardization of their instrumentation.

Analysis of this mixed uranium/plutonium bead material has demonstrated that to reproduce isotopic fractionation, the variability of which limits measurement precision and accuracy, requires careful control of the time and temperature program of the mass spectrometric procedure. Although this statement is generally true for all thermal ionization measurements, the fact that two elements, both at the nanogram level and both with five isotopes, are measured from a single sample makes control of isotopic fractionation more critical and more difficult. The mass spectrometric procedure used must also allow correction for the mutual ^{238}Pu and ^{238}U isobaric interferences. Initial

attempts to burn-off Pu quickly prior to the U measurements required too hot a temperature which resulted in severe uranium isotopic fractionation. Subsequently, a procedure was followed that utilized a lower temperature and slower rate of Pu burn-offs, and, although necessitating a correction of a few percent for the ^{238}Pu in the ^{238}U signal, resulted in much less severe and much more reproducible uranium isotopic fractionation. The measurement precisions for the $^{242}Pu/^{239}Pu$ and the $^{233}U/^{238}U$ ratios of this material were 0.15% and 0.10%, respectively (1σ).

This work is being performed in the Center for Analytical Chemistry under the sponsorship of DOE and ISPO.

(3) Investigations of Sample-Mounting Techniques for Uranium Isotopic Analysis by Thermal Ionization Mass Spectrometry

The accuracy and precision of mass spectrometric isotope ratio measurements is limited by the ability to control isotopic fractionation during the ionization process. This mass-dependent fractionation appears to be dependent on the molecular species being volatilized from the sample filaments. In the case of uranium, the per mass fractionation correction (ratio of true ratio to measured ratio) can be altered by as much as 0.1% depending on the conditions under which the uranium nitrate solution is dried on the sample filaments prior to insertion into the mass spectrometer.

The effects of static and dynamic air flow environments have been investigated, along with different drying temperatures which produce different uranium oxides on the filaments. Microscopic examination of the filaments during sample drying has revealed that the filament temperature at which conversion to the various oxides occurs is not reproducible and that in some cases the oxide form changes during the few minutes between completion of the sample mounting and insertion into the mass spectrometer. These observations have resulted in the development of a drying technique which keeps the drying temperature low to prevent oxide formation. This technique has been successfully applied to the assay of the ^{233}U spike SRM and has demonstrated the capability of accuracies and precisions of better than 0.05%.

Additional studies directed toward the identification and control of the chemical species formed on the mass spectrometer filaments is continuing with the eventual goal of achieving routine precisions of 0.01% for uranium isotopic measurements.

This work is being performed in the Center for Analytical Chemistry under the sponsorship of NRC and DOE.

C. Bulk Measurements

The Bulk Measurements Task occupies a position of strategic importance in both the NBS Nuclear Safeguards Program and in nuclear safeguards in general. The activities of the Task involve the determination of the quantity of nuclear material in specific areas and processes in a nuclear fuel cycle plant and between specific areas and processes. Measurements of mass, volume, density, flow, pressure and temperature contribute to the determination of the quantity of nuclear material; the measurements of these physical variables interact with each other and with the activities of the other three Tasks in the NBS Nuclear Safeguards Program.

(1) Flow

A system has been designed and constructed for checking the calibration of the NBS laser Doppler velocimeter (LDV) system. The LDV system measures fluid velocity inside our 5 cm diameter by 60 cm long glass test section. The calibration check is necessary because when the laser beam passes through the curved glass, the beam is bent in a way that is difficult to describe theoretically and with sufficient accuracy. This is true even though the round glass tube is totally immersed in water contained within flat glass windows. The calibration system uses a wire traveling longitudinally through the pipe at a known constant velocity. The wire can be moved to any location in the pipe cross section by two stepping motors interfaced to the same computer that operates the LDV system.

Figure 8 shows the measured fringe spacing as a function of the wire position across the beam waist. The spacing is constant to within a fraction of a percent for both the green and blue beams. Figure 9 shows that the measured fringe spacing at various locations in the pipe volume is constant to within a fraction of a percent. As demonstrated by the data, the calibration of the LDV system has been completed successfully.

Work on the correlation flow meter has progressed from the preliminary version used to measure the velocity of bubbles in water flowing in a pipe to a more sensitive and noise-free system which is under construction at present. The sensing electrode consists of a wire offset from the ground plane and facing an electrically active plate located across a 3/4" glass tube. The wire and the active plate form a capacitor which, together with a balancing capacitor, are connected to the input of a tuned amplifier. The capacitive bridge was balanced to about 50 μV out of 10 V at a frequency of 1 MHz. The output of the tuned amplifier was fed into a phase sensitive detector (PSD) operating at 1 MHz. Due to the sensitivity of the above detector it was found that the output of the PSD changed over a period of minutes. This variation was traced to changes in the properties of the flowing tap water, for example, its bubble content. To eliminate the slow variation, a design of a computer based feedback circuit was initiated. The feedback circuit samples the integrated output of the PSD and adjusts the values of two photoresistors located in the bridge circuit in order to minimize the PSD signal.

This work is being performed by the Continuous Process Technology Program under the sponsorship of DOE.

(2) UF_6 Mass Measurement Implementation Program

The objective of this Sub-Task is the improvement of in-facility weighing of large UF_6 cylinders. The effort involves the implementation of a Measurement Assurance Program (MAP) for the measurement of the mass of UF_6 cylinders, filled with UF_6 or empty. The MAP provides continually verifiable evidence of performance of the mass measurement process, giving timely and quantitative uncertainty statements for the mass measurements.

A draft standard operating procedure was sent to participating organizations for comment.

The 30B Replica Mass Standard (RMS), with mass determined by NBS, was first shipped to Oak Ridge to be used in calibrating the Oak Ridge In-House Standards. The RMS clyinder was then shipped from Oak Ridge to the General

Electric Co. (G.E.) Facility at Wilmington, NC, where it was used to recalibrate the G.E. In-House Standards, and then on to the Combustion Engineering Co. facility at Hematite, MO, where they were used to calibrate In-House Standards.

This work is being performed in the Continuous Process Technology Center and the Center for Applied Mathematics, under the sponsorship of NRC.

Figure 8. TEST BEAM WAIST LOCATION

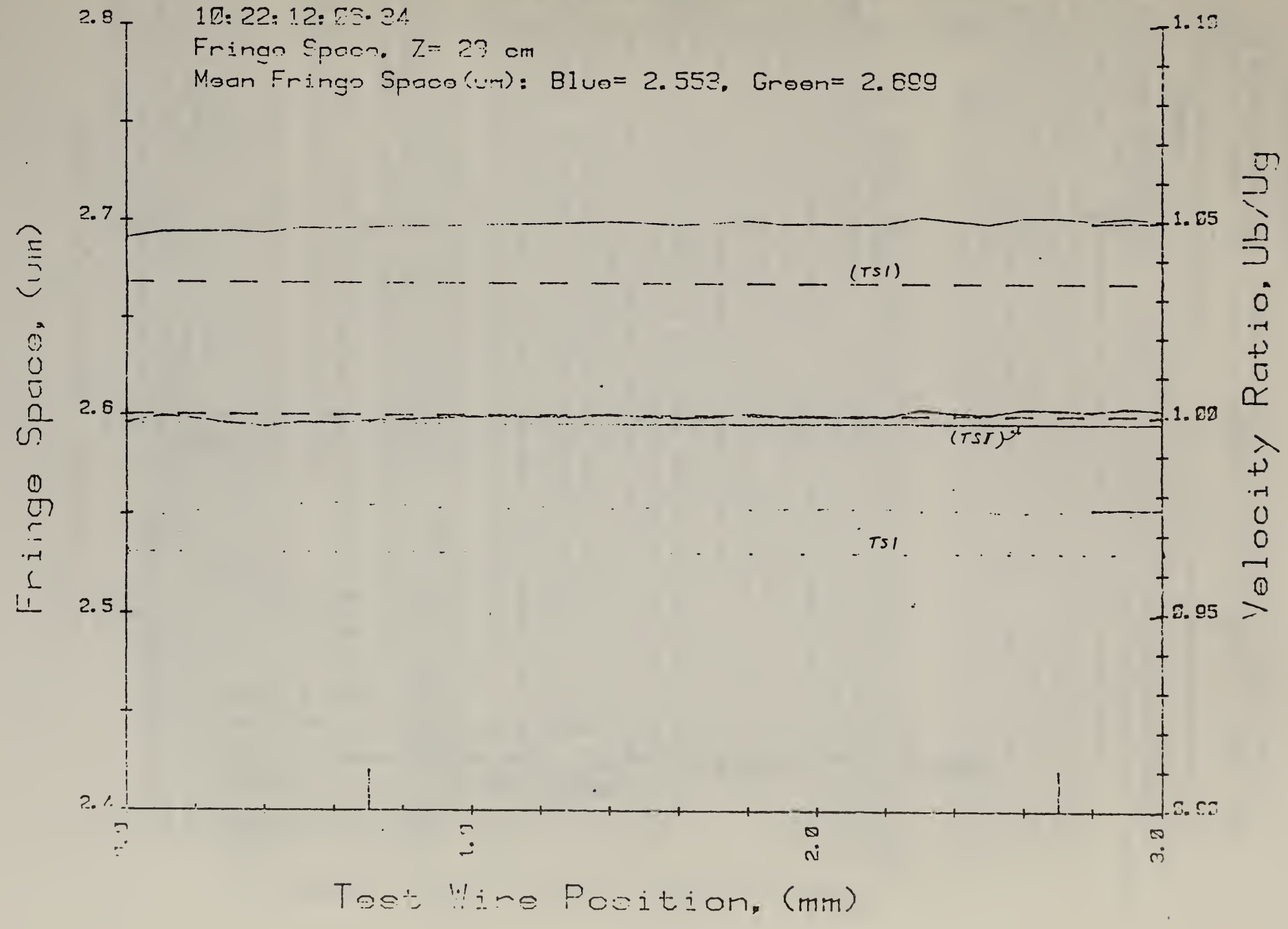
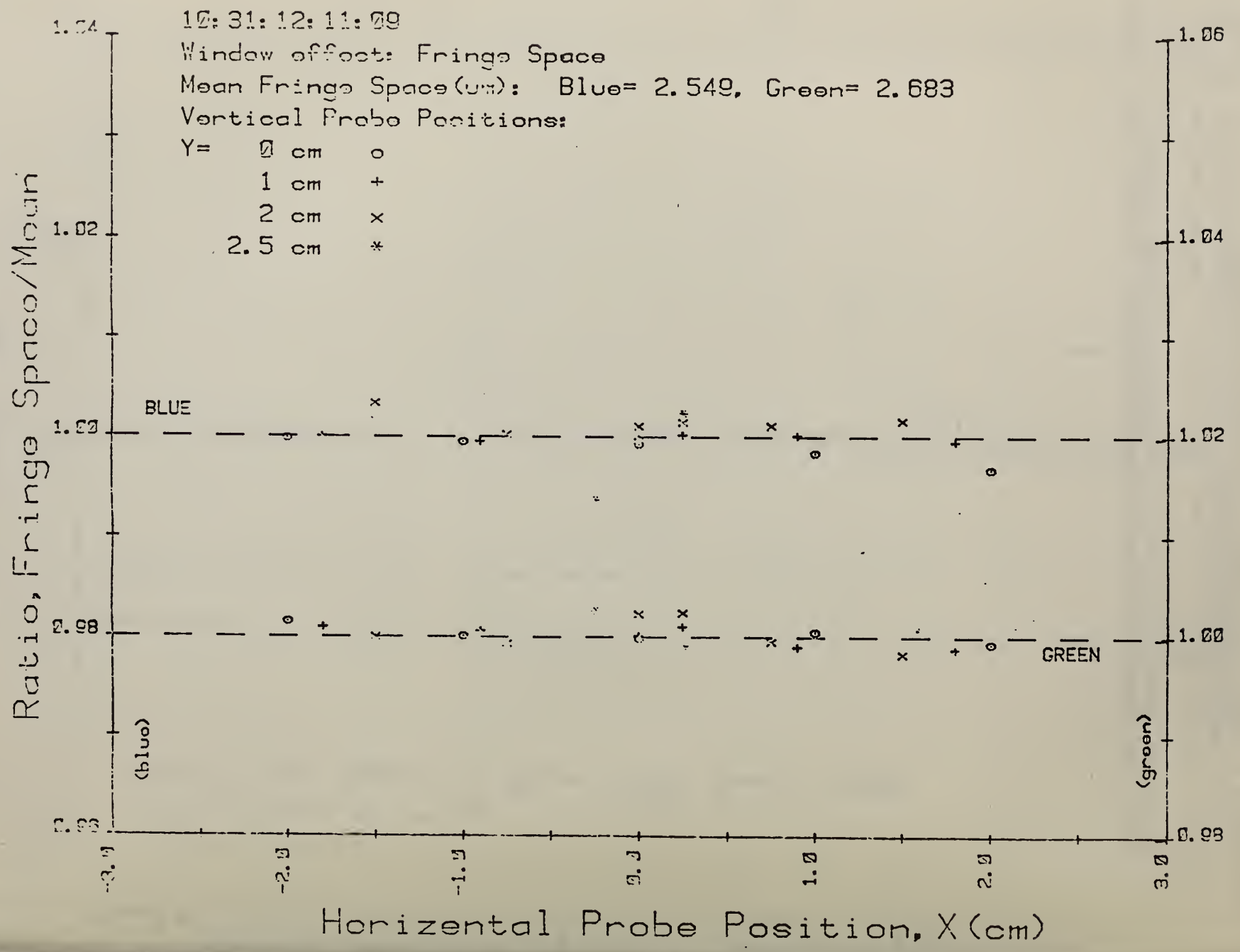


Figure 9. LDV CALIBRATION



D. Applied Mathematics

The applied mathematics effort in support of safeguards consists not only of independent efforts, but also of collaborative efforts with other scientists at NBS.

(1) Some Effective Statistical Approaches for Presenting Interlaboratory NDA Enrichment Measurements. In order to realistically evaluate measurements on prototype and primary reference materials measured by several different nondestructive assay (NDA) labs, it is necessary to distinguish real differences between laboratory findings and artificial differences such as those caused by artifacts of individual peak area calculations. This report provides guidance for locating and assessing the magnitude of these peak area artifacts. In particular, it provides a variety of algorithms for linear peak area computations which make the standards appear either most or least homogeneous, under a variety of statistical criteria. The criteria include the usual F statistic as well as the standardized range.

(2) Robust Inference for Items Measured a Few Times. Estimation methods that de-emphasize the measurements that appear to be most in error offer the possibility of improved performance. This possibility, which occurs when the measurement error is non-Gaussian, is explored for some measurement designs not covered by previous work on robust methods. In these designs, each item in a collection of somewhat dissimilar items is to be measured two or three times, and the total for the entire collection is to be estimated. The methods are based on the assumption that the measurement errors for different items are from the same population. A method is proposed for each of three cases: three replicate measurements on each item, two replicate measurements by one method and one measurement by another method on each item, and two replicate measurements on each item. All the methods use the differences between the measurements on the same item to obtain a scale estimate by which the sizes of the measurement errors are judged. How the methods adjust the measurements differs from case to case. When there are three measurements per item, the most dissimilar of the three is downweighted. When there are only two measurements per item, the measurements for the entire collection are used to judge which measurements to downweight. The methods include estimates of the uncertainty due to measurement error, which are also obtained from the differences between the measurements on the same item.

(3) Inventory Difference Calculations: Problems and Possibilities (preliminary abstract of report in preparation). The origin, development, and termination of the "error study" task funded by the Nuclear Regulatory Commission at the National Bureau of Standards is presented in this report.

Other reports which grew (at least partially) out of this study are listed and related to the intent of this study.

A general approach to the calculation of Inventory Difference (ID) and its Limit of Error (LEID) was devised and refined through visits to a selected fuel-fabrication facility. This approach is discussed, with emphasis on the importance of having measurements "under control" and on the possibility of automating the computations. The concepts of "measurement process" and "Measurement Assurance Program" are also discussed and their importance shown. Possible problem areas, both those observed at the studied facility and others, are given, along with possible directions for resolution.

Finally, new approaches to the calculation of ID and LEID being developed elsewhere are described and their relation to the approach being considered herein elucidated.

All of the statistical work is being done in the Center for Applied Mathematics with the support of NRC and DOE.

E. Safeguards Systems Studies

The objective of Safeguards Systems Studies is to provide technical support to the safeguards program of the DOE Office of Safeguards and Security (DOE/SS) and the U.S. Initiative Program for international safeguards. Systems analysis studies are performed leading to a more effective utilization of data and information from all sources for achieving safeguards goals. Program efforts contribute sound technical input for design, development and operation of DOE/SS and domestic nuclear plant programs that will be effective in protecting against covert diversion of special nuclear materials (SNM) by persons authorized access to the material or international programs aimed at detecting diversion by the State. Task activities addressed completion of the Concepts for Safeguards Information Systems study, modification of Diversion Path Analysis Computer Program 2, and adaptation of Diversion Path Analysis concepts to evaluation of international safeguards as applied by the International Atomic Energy Agency.

(1) Concepts for Safeguards Information Systems

Concepts for Safeguards Information Systems (CSIS) are based on the theory that meaningful safeguards decisions can only be formulated through effective use of information from all pertinent sources, both within and, when appropriate, from outside of DOE. Examination of DOE facilities, operations offices and headquarters safeguards activities, coupled with the information flowing in the present system, served as the basis of the safeguards information system framework defined in CSIS. CSIS is generic in nature and, in this context, the framework is defined by a heirarchy of decision makers whose safeguards responsibilities and tasks are specified. Based on these specifications, the information needs of each decision maker were developed and aligned with the information generated, records maintained and interactions between decision makers throughout the heirarchy. The application of CSIS will provide a check of completeness of information flow and content of flow between decision makers and serve primarily as a guide for designing, upgrading and implementing a site-specific information system for safeguards.

Development of the concepts and framework for the generic safeguards information system were completed and the draft document describing CSIS was submitted to DOE/SS.

(2) Diversion Path Analysis

Diversion Path Analysis (DPA) is a safeguards evaluation tool that can be used to determine the vulnerability of domestic material control and material accounting (MC&MA) subsystems of safeguards to the threat of theft of SNM by a knowledgeable insider. DPA specifically addresses diversion of SNM from its authorized location within a domestic plant by a person who has access to the process area and/or the material. It is used to evaluate the ability of the MC&MA subsystems to detect the loss of a fraction of the amount of SNM needed to construct a clandestine nuclear explosive. Using the methodology, facility personnel systematically determin (1) how, from the diverter's viewpoint, to covertly acquire SNM and conceal the theft from the MC&MA subsystems; (2) how soon the MC&MA subsystems would indicate the theft

and (3) what modifications to the plant's safeguards system would be necessary to eliminate, or reduce the severity of, the identified vulnerabilities.

DPAs completed during the last year by contractor personnel at Idaho and Savannah River were assessed by NBS at the request of the Directors of Safeguards and Security for the respective DOE Operations Offices. The Savannah River personnel developed a Diversion Vulnerability Index while performing the analyses at that site. This Index represents a crude cost-benefit measure and assisted contractor management in selecting modifications for implementation. Because of its potential usefulness at other sites, DPA Computer Program 2, which assists in assembling modification data, was revised to include computation of the Index.

(3) International Safeguards

In recent years, the International Atomic Energy Agency (IAEA) Board of Governors has requested the IAEA Secretariat to increase the extent to which the effectiveness of IAEA safeguards is gauged by objective analysis. A method has been proposed, which is an adaptation of Diversion Path Analysis, to increase both the uniformity and objectivity of effectiveness evaluation analyses. Development of the method represents a cooperative effort being performed under the U.S. Initiative Program. The method is intended to direct, with minor variations, three distinct modes of application. First, it can be used to guide generation of the model safeguards approach and associated design assessment of a generic facility type. Second, it can be used to develop the facility safeguards approach, for a specific facility that resembles the generic facility, and derive the implementation assessment for that approach. Third, it can be employed to carry out the performance evaluation of an IAEA safeguards inspection of the facility based on the facility safeguards approach developed during the second mode of application.

A draft set of instructions and guidelines was developed for conducting the first mode of application. This draft is currently undergoing revision based on experience gained in developing case studies for two types of generic facilities. The implementation experience has indicated, however, that the guidance provided for this phase of the analysis is both practical and credible.

F. Users Guide, Safeguards Accountability Instrumentation and Techniques

The primary objective of the Users Guide is to provide specific recommendations on how facilities should be designed to accommodate the necessary measurements to meet the SNM accountability goals of the safeguards program. The Guide will discuss how the measurement instruments and accountability systems should be installed and will outline the supporting services required. The Guide will also discuss facility design considerations so that the instruments can be efficiently calibrated, operated and repaired, either manually or remotely.

The preliminary draft of the Guide, which was completed during the previous fiscal year, was revised to reflect certain directives from the NRC. Discussions were held with the persons at Mound Laboratories who are preparing a Handbook of Nuclear Safeguards Measurement Methods. The purpose of these discussions was to assure that the two documents would be complementary.

The first rough draft of the Guide was completed and copies sent to contributors for comment and/or correction. In its draft form, the document had a length of approximately 18,000 words.

After the cut-off date for comments, May 30, 1980, the Guide was rewritten into its final form. It is now undergoing editorial review.

4. Measurements and Standards for Nuclear Waste Management - A Technical Overview

The Program of Measurements and Standards for Nuclear Waste Management was established late in fiscal year 1980, after an extensive study of the materials and measurements problems involved in nuclear waste management. As a result, most of the efforts expended have focused on program startup. However, technical progress has been made in specific areas.

The technical plans for the Nuclear Waste Program are divided into the following areas: (1) Waste Forms Leachability Mechanisms; (2) Waste Forms Research, Reference Materials Data and Measurement Methods; and (3) Containers, Backfill and Math Support.

A. Waste Forms - Leachability Mechanisms

This task will review existing efforts by others, as well as carry out experiments in selected areas at NBS to improve and understand the mechanisms involved in leaching of radioactive waste species from an immobilizing matrix. In order to accomplish this, the role or effect of radiation, mechanical properties, microstructure, microchemistry, thermodynamics, transport mechanisms, thermochemistry, and various other disruptive processes will be investigated.

Planned activities for fiscal year 1981 include the completion of a set of Monte Carlo calculations to analyze the energy deposition processes in the waste matrix, evaluation of existing models of mass transport in solids for their applicability to the nuclear waste problem, and institution of experimental studies concerning the structural stability and transport properties of crystalline material containing large ratios.

B. Waste Forms - Research, Reference Materials Data and Measurement Methods

This task includes the development and dissemination of generic research materials with well-characterized elemental composition to aid in the development and promulgation of leach tests. Materials (glass) with varying leach rates containing non-radioactive cesium, strontium, etc., will be provided.

Current activities include advising the Materials Characterization Center (MCC) on the MCC-1 leach test and round robin, and providing a borosilicate glass for use in that round robin. Development of a set of generic reference leach tests in cooperation with Battelle's MCC is a long range goal. During FY 1981 NBS will develop the capability to perform several of the proposed reference leach tests and, if required, suggest changes to the procedures to improve precision.

In addition to the above work, a literature survey is planned to compile, evaluate, and publish a compendium of standard reference data on phase equilibria of materials used for nuclear waste encapsulation. This will be done through the existing mechanisms used to publish "Phase Diagrams for Ceramists", co-sponsored by NBS and the American Ceramic Society. The compendium will include critically evaluated phase diagrams which are one of the components needed for waste form processing and for characterizing the compatibility and interaction of the waste form with the geologic environment.

During fiscal year 1981, a comprehensive literature survey of all existing published (archival) literature, including the first five volumes of "Phase Diagrams for Ceramists", will be undertaken.

C. Containers, Backfill and Mathematical Support

This task is focused on the container and backfill material and includes also the statistical and quality control effort for the whole program.

In the area of containers, existing efforts to understand corrosion mechanisms will be reviewed, and specific research will be carried out in selected areas to improve the understanding of degradation mechanisms of candidate materials, particularly as they apply to long-term prediction and accelerated testing.

During fiscal year 1981, existing data and literature will be reviewed on possible canister degradation mechanisms and test methods, and state-of-the-art information will be assembled for publication.

In order to provide well-characterized materials for use in NBS tests and materials research by other laboratories, a complete stock of analyzed research alloys will be developed during the coming year.

Also planned is a cooperative effort with the MCC in surveying and evaluating existing data on underground corrosion of potential canister materials. A bibliography of existing data and literature on corrosion of alloys underground should be completed.

Work on the backfill effort for the Nuclear Waste Program includes a review of existing efforts elsewhere, as well as performing selected research at NBS to improve the understanding of the basic functional mechanisms (excluding sorption coefficient) related to emplacement of backfill materials. The information obtained will be disseminated via publications and will provide support to other NBS efforts. In addition, a workshop on backfills is being planned for September 1981.

In order to promote compatibility of testing, the development and dissemination of research materials is planned. Possible candidate materials include common clays such as illite, kaolinite, and montmorillinite, as well as zeolites and sand-bentonite mixtures. These materials will be evaluated and characterized for homogeneity, elemental composition, structure, physical size, loss of weight on drying, and stability in packaging.

Extensive use of statistical services available at NBS is planned for the effective use of the interlaboratory testing required in the performance of these tasks.

5. Appendix

A. NBS Seminars on Measurements for Nuclear Technology

NBS Nuclear Safeguards Seminar, "Safeguards Non-Destructive Assay Program", by Walter G. Martin, U.S. Nuclear Regulatory Commission, February 21, 1980.

NBS Nuclear Safeguards Seminar, "Measurements for Nuclear Materials Safeguards - Their Development, Calibration, and Evaluation", by Carleton Bingham, Director, New Brunswick Laboratory, March 6, 1980.

NBS Nuclear Safeguards Seminar, "Overview of International Nuclear Safeguards", by William M. Murphey, Arms Control and Disarmament Agency, April 25, 1980.

B. Committee Participation and Consulting Activities

(1) Committee Participation

James W. Behrens - Member, DOE Cross Section Evaluation Working Group (CSEWG), Standards Subcommittee; Member, Safeguards Committee, Institute for Nuclear Materials Management

Charles D. Bowman - Member, Department of Energy (DOE) Nuclear Data Committee

Stephen M. Baloga - Member, Technical Program Committee for American Nuclear Society Topical Conference, "Measurement Technology for Safeguards and Materials Control", Kiawah Island, SC, November 26-30, 1979

B. Stephen Carpenter - Board of Directors, American Nuclear Society; Member, Isotope and Radiations Division, Technical Committee for Nuclear Safeguards; Member, Institute of Nuclear Materials Management Subcommittee INMM-9.3 on Non-destructive Assay of Standards Committee N15 on Methods of Nuclear Material Control; Member, Department of Energy, Office of Safeguards and Security, Ad Hoc Task Group on Measurements and Standards

Randall S. Caswell - Member, International Society of Radiology, International Commission on Radiation Units and Measurement

Allan D. Carlson - Member, DOE Cross Section Evaluation Working Group (CSEWG), Standards Subcommittee

David A. Ditmars - Member, INMM 8.4, "Calibration Techniques in the Calorimetric Assay of Pu-Bearing Solids"; Member, NBS Pu Steering Committee

James A. Grundl - Member, ASTM E10.05, Committee on Neutron Dosimetry

J.M. Robin Hutchinson - Chairman, International Committee of Radionuclide Metrology (ICRM), Subcommittee on Low-Level Techniques Group; Member, American National Standards Institute (ANSI) Committee on Nuclear Instruments and Detectors; Secretary, ANSI Subcommittee N42.2 on Procedural Standards for Calibration of Detectors for Radioactive Measurements

Harry H. Ku - serves on the Steering Committee of the Safeguards Analytical Laboratory Evaluation Program

James A. Lechner - Member of American Society for Testing and Materials (ASTM) Committee C26 on Nuclear Fuel Cycle; Member of ASTM Committee N15; advising the Institute for Nuclear Materials Management Committee 9.4 on revised standard for measurement control of NDA processes

Lawrence A. Machlan - Member, Plutonium Steering Committee at NBS; Member, ASTM C26 (Nuclear Fuel Cycle)

William P. Reed - Member, ASTM C26, "Nuclear Fuel Cycle"; Member, ANSI N15.9, "Nondestructive Assay"

Paul E. Pontius - Member, INMM8.1 Subcommittee on Mass Calibration Techniques for Nuclear Material Control; Member, INMM8.2 Subcommittee on Volume Calibration for Nuclear Material Safeguards

James R. Whetstone - Member, INMM8.2 Subcommittee on Volume Calibration for Nuclear Material Safeguards

(2) Consulting Activities

Stephen M. Baloga and John C. Schleter - Consultants to DOE Savannah River Operations Office during assessment of Diversion Path Analyses for Savannah River Plant facilities; Consultants to DOE Idaho Operations Office during assessment of Diversion Path Analyses for Idaho National Engineering Laboratory facilities

Stephen M. Baloga - Consultant to DOE Savannah River Operations Office on statistical sampling plans and nuclear material measurements for inventory verification

Charles D. Bowman - Performed a neutron dosimetry review for the Fusion Materials Irradiation Test Facility (FMIT), Hanford Engineering Development Laboratory; performed linac neutron measurements for Lawrence Livermore National Laboratory, Physics Division; performed gamma ray laser research for Los Alamos Scientific Laboratory, Physics Division

Ronald G. Johnson - Performed photonuclear research for Lawrence Livermore National Laboratory

Paul E. Pontius - Consultant to LASL on Bulk Measurements

Roald A. Schrack - Performed measurements for international safeguards on special nuclear material for International Safeguards Project Office

C. Publications (from October 1, 1979 to September 30, 1980)

Baloga, S. M., and Hakkila, E. A., "Measurement Trends for Future Safeguards Systems", LA-UR-80-1817, Los Alamos Scientific Laboratory, June 1980; also, Nuclear Materials Management, Proceedings Issue, Vol. IX (in press).

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Canada, T. R., and Carpenter, B. S., Editors, Proceedings of Measurement Technology for Safeguards and Materials Control, NBS Special Publication 582 (1980).

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D. Talks (from Oct. 1, 1979 to Sept. 30, 1980)

Baloga, S. M., "Statistical Problems Associated with the Verification of Nuclear Material Inventories at DOE Facilities", NBS Statistical Engineering Laboratory Seminar, March 11, 1980.

Baloga, S. M., "Measurement Trends for Future Safeguards Systems", INMM 21st Annual Meeting, Palm Beach, FL, June 30-July 2, 1980.

Baumgarten, G. P., Brame, V., Cooper, D. G., and Robertson, B., "Automated Tank Calibrator", American Nuclear Society Topical Meeting, Measurement Technology for Safeguards and Material Control, November 26-30, 1979, Kiawah Island, SC.

Behrens, J. W., "Non-Destructive Assay of Fresh Nuclear Fuel Using Resonance Neutron Radiography", Rennselaer Polytechnic Institute, Troy, NY, March 3, 1980.

Bowman, C. D., "Molecular Binding Effects on Neutron Nuclear Cross Section", Univ. of Missouri, Columbia, MO, Oct. 31, 1979; Univ. of Missouri, Rolla, MO, Nov. 1, 1979; Washington University, Nov. 2, 1979.

Bowman, C. D., "Resonance Neutron Radiography", Atomic Energy Research Establishment, Harwell, England, March 14, 1980.

Bowman, C. D., "White Source Use in a Neutron Standards Laboratory", IAEA Consultants Meeting on Neutron Source Properties, Kossuth Lajon University, Debrecen, Hungary, March 17, 1980.

Bowman, C. D., "Accelerator Source for Neutron Nuclear Data Measurements", as chairman of the Accelerator Sources Working Group, IAEA Consultants Meeting on Neutron Source Properties, March 17-21, 1980.

Bowman, C. D., "Neutron Standards Measurements and Related Physics Research", American Physical Society Meeting, Washington, DC, April 30, 1980.

Bowman, C. D., "Neutron Source for 10-50 MeV Cross Sections", as chairman of the Neutron Sources Working Group of the Conference on 10-50 MeV Cross Sections, Brookhaven National Laboratory, May 19-21, 1980.

Bowman, C. D., "Neutron Powder Diffraction Measurements Using the NBS 100-MeV Electron Linac", Lawrence Livermore National Laboratory, Livermore, CA, Sept. 15, 1980.

Bowman, C. D., "Efficient Neutron Production Using 12 MeV Electrons", Sixth Conf. on the Application of Accelerators in Res. & Industry, Nov. 3, 1980, Denton, TX

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Fassett, J. D., Kelly, W. R., Machlan, L. A., and Moore, L. J., "Mass Spectrometric Isotope Metrology of Uranium on Resin Beads", 23rd Conf. on Analyt. Chem. in Energy Tech., Gatlinburg, TN, Oct. 10, 1979.

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Garrett, D., and Cheng, T., "Nuclear Safeguard Interrogation of Waste Containers with Resonant Neutrons", talk presented at the 40th ASNT National Fall Conference on Quality Testing Show, 1980, Houston, TX.

Gramlich, J. W., "Sample Mounting Techniques for High Accuracy Uranium Isotopic Analysis", 28th Annual Conference on Mass Spectrometry and Allied Topics, New York, NY, May 27, 1980.

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Reed, W. P., "The Role of Standard Reference Materials in Achieving Measurement Traceability", American Nuclear Society Topical Meeting, Measurement Technology for Safeguards and Material Control, November 26-30, 1979, Kiawah Island, SC.

Schleter, J. C., "Diversion Path Analysis", DOE/NRC R&D Exchange Seminar, June 13, 1980.

Schrack, R. A., "Quantitative Isotope-Selective Assay Using Resonance Neutron Radiography", University of Glasgow, Glasgow, Scotland, March 25, 1980.

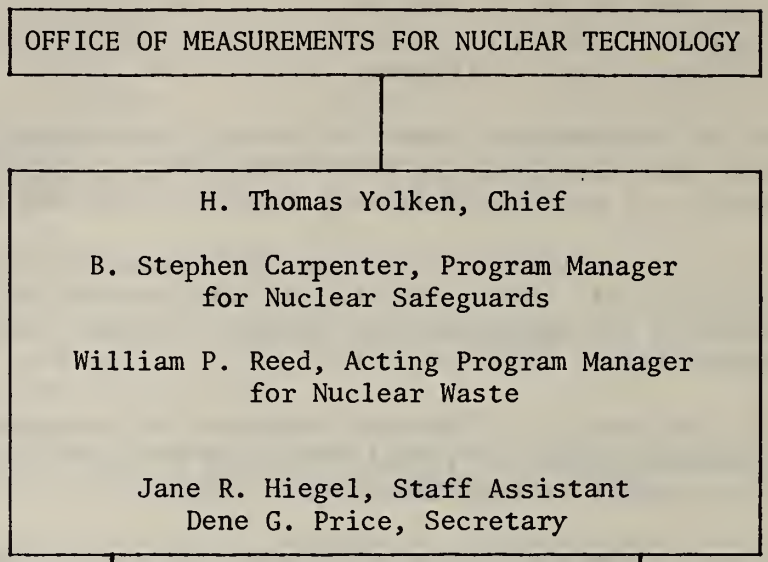
Schrack, R. A., Behrens, J. W., Johnson, R. G., and Bowman, C. D., "Resonance Neutron Radiography Using an Electron Linac", Sixth Conference on the Application of Accelerators in Research and Industry, Nov. 3, 1980, Denton, TX.

Yolken, H. T., "National and International System of Measurement Standards for Nuclear Safeguards", U.S. DOE/IAEA International Training Course on Nuclear Materials Accountability and Control for Safeguards Purposes, Santa Fe, NM, May 27-June 6, 1980.

E. Sponsored Symposia and Workshops

Measurement Technology for Safeguards and Materials Control (Kiawah Island, SC), November 1979

F. Office of Measurements for Nuclear Technology



Safeguards Management Team*

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Nuclear Waste Management Team*

Thomas E. Gills (OSRM)
Wolfgang K. Haller (CMS)
Howard S. Kingston (CAC)

*Bench Scientists devoting part of their time helping to manage the program.

G. Functional Statements

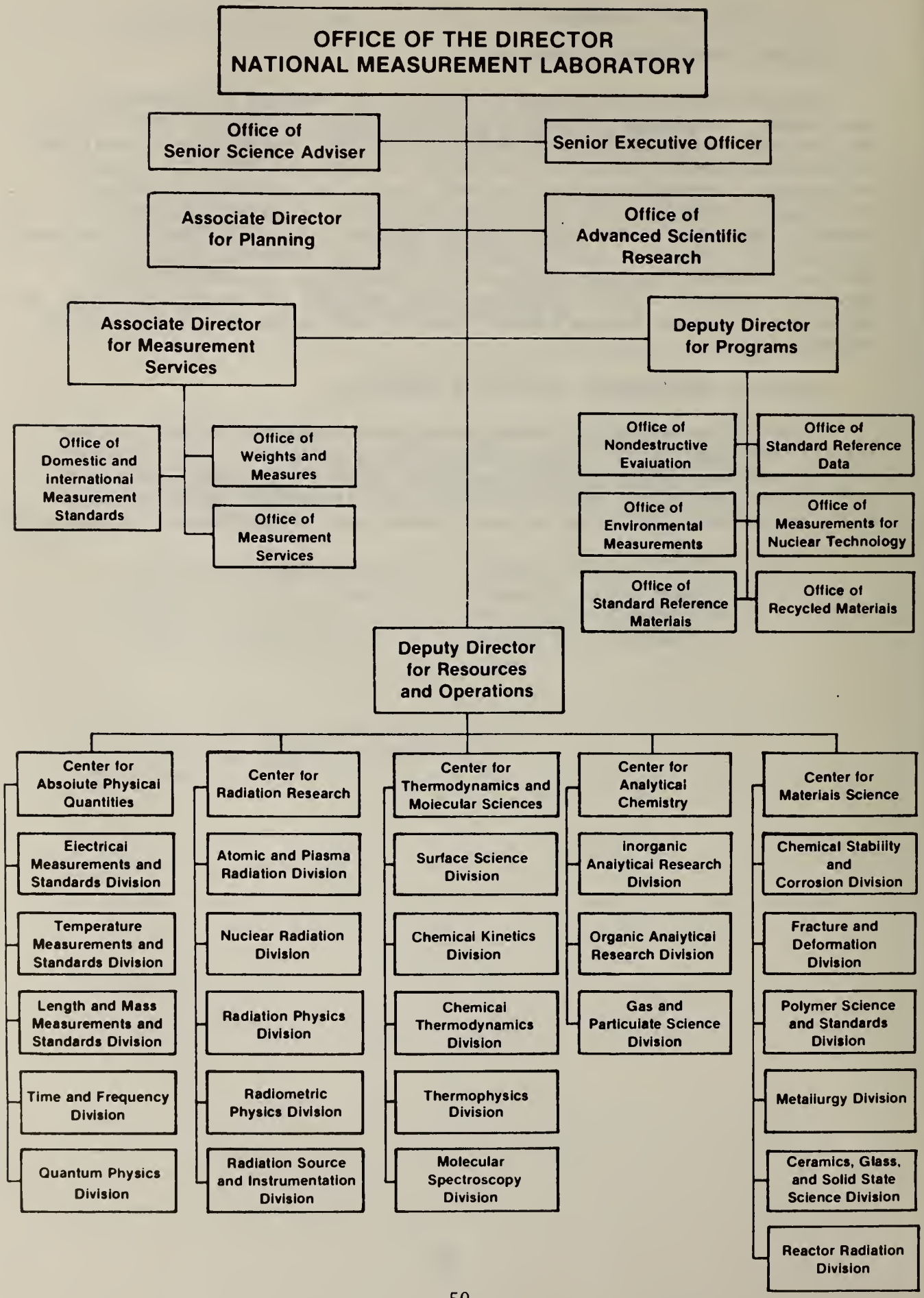
NATIONAL MEASUREMENT LABORATORY

Provides the national system of physical and chemical and materials measurement; coordinates the system with measurement systems of other nations, and furnishes essential services leading to accurate and uniform physical and chemical measurement throughout the Nation's scientific community, industry, and commerce; conducts materials research leading to improved methods of measurement, standards, and data on the properties of materials needed by industry, commerce, educational institutions, and government; provides advisory and research services to other government agencies; conducts physical and chemical research; develops, produces, and distributes standard reference materials; provides standard reference data; provides calibration services; and collaborates with the Bureau's major organizational units in carrying out its responsibilities.

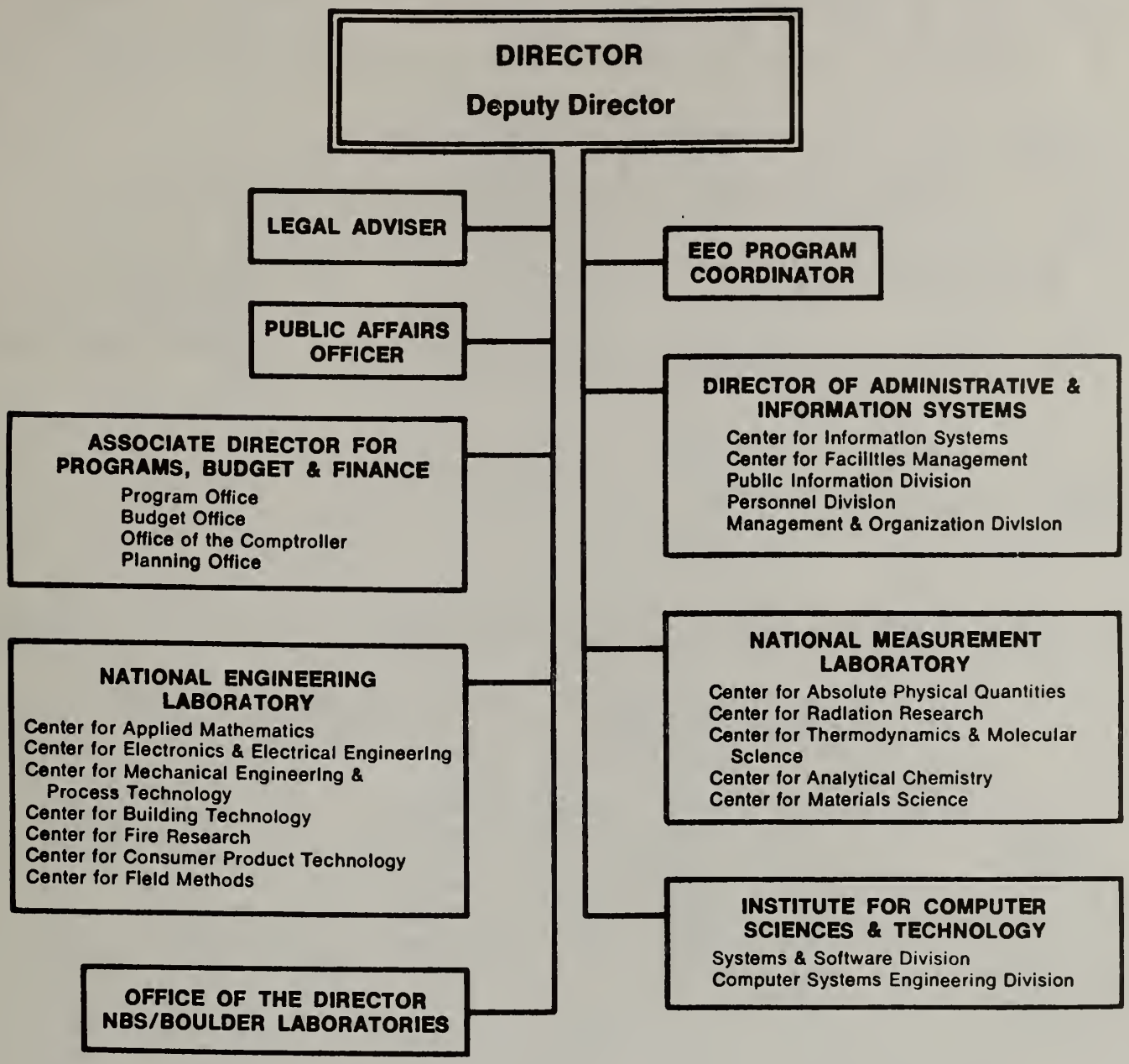
OFFICE OF MEASUREMENTS FOR NUCLEAR TECHNOLOGY

Provides the Nation with needed measurement standards and services for nuclear technology in the areas of nuclear waste management and safeguarding nuclear materials; plans, directs, and coordinates the Program's laboratory work that is carried out within NBS units; and disseminates these standards and services domestically and to other nations and to international organizations.

H. NML Organizational Chart



I. NBS Organizational Chart



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10. SUPPLEMENTARY NOTES

[ ] Document describes a computer program; SF-185, FIPS Software Summary, is attached.

11. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)

This annual report is a summary of the National Bureau of Standards (NBS) Measurements for Nuclear Technology (MNT) Program for Fiscal Year 1980. The MNT activities at NBS are divided into two programs: Nuclear Safeguards and Nuclear Waste Management.

The MNT Program is one of several matrix managed efforts at NBS. The programs provide a focus for the activity, both inside and outside NBS. The programs provide funds and directions for needed technical work that is done within NBS Centers. People remain attached to Centers but are funded for specific activities in a matrix management approach. A program provides central organization for outside inquiries and for distribution of results. Often programmatic focus can provide improved assistance to outside agencies with measurement problems. The MNT Program is operating in this manner.

12. KEY WORDS (Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)

accountability guide; bulk measurements; isotopic assay; leachability studies; nondestructive assay; nuclear safeguards; nuclear waste management; Standard Reference Materials; statistical evaluation

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