

# NUCLEAR MATERIALS MANAGEMENT



JOURNAL OF THE INSTITUTE OF NUCLEAR MATERIALS MANAGEMENT

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# EDITORIAL

**DR. WILLIAM A. HIGINBOTHAM**  
Brookhaven National Laboratory  
Upton, New York



Before I started in the safeguards field, I was in nuclear instrumentation. Boron tri-fluoride filled proportional counters were developed in the 1930's, and geiger tubes in the 1920's. But sodium-iodide scintillation counters, which could be used to measure the energies of gamma-rays were not developed until after the war and silicon and germanium detectors came late in the 1950's. Silicon detectors soon replaced the very complicated and difficult to use gridded ionization chambers for alpha particle spectrometry and the germanium detectors opened a new world for high resolution gamma-ray spectrometry. In every case, there was a good idea followed by a great many practical problems before these detectors became available at reasonable cost and operationally reliable.

In order to use radiation detectors, one needs very high quality amplifiers and fancy electronic circuits to sort the signal pulses according to size and sometimes also by shape. There were a few very clever individuals in several different countries who collaborated in the development and improvement of the detectors and the circuits needed to use them. Vacuum tubes were replaced by transistors and transistors by integrated circuits on chips. Analogue circuits were replaced by digital circuits and a pocket computer today is better than a computer my group built in the 1950's which filled a large room and consumed an enormous amount of power.

In 1944 I invented the first electronic counting circuits which would operate reliably to over 200,000 hertz (or on random pulses spaced by 5 microseconds). In the fall of 1945 I built the first multichannel pulse height analyzer, a rack full of tubes and power supplies. Today, electronic counters are 1,000 times faster and portable 8,000 channel analyzers operate reliably for days on a few batteries.

This fall I attended the annual Institute of Electrical and Electronic Engineers Nuclear Science Symposium. I saw a number of friends who have been active in the instrumentation field for thirty years or so. About one quarter of the papers were presented by scientists and engineers from other countries, as has long been the case. Applications of the instruments were broader than ever before: medicine, space, high energy physics, nuclear energy, environment, safeguards, radiation protection, etc.

It is an exciting time. The discovery and development of nuclear energy stimulated these developments in the early days and nuclear energy and safeguards have benefited from them, as have medicine, many industries, education, and the ability of individuals, organizations, and governments to plan and to operate. The tools for research, data storage and analysis have improved by several orders of magnitude during the last thirty years. But these are tools. Ingenuity and wisdom are still essential for their constructive use.

Safeguards people have been involved in the electronics revolution and we are taking advantage of it. But wisdom of a very special sort is essential if safeguards is to fulfill its promise to society. Safeguards is an international undertaking on behalf of society as a whole. We have the tools, and most of those involved in safeguards at the political and the technical levels have the good will. But I know that some of us view the purposes and the means to achieve them rather differently, in different parts of the world and even within its several parts. It is important, very important, to take advantage of the latest developments in electronics and statistical analysis. But it's even more important that all of us involved in safeguards agree on what it is we are trying to do and on how to do it together.

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## CHAIRMAN'S COLUMN

**YVONNE M. FERRIS**  
 Rockwell International  
 Golden, Colorado



Have you ever wondered why your summer Journal sometimes arrives at Christmas and contains September meeting announcements? I'm exaggerating, of course, but a late journal can be annoying. One reason for its tardiness is lack of technical articles. The INMM does not always have a pool of articles to draw from and, therefore, must wait on those in the approval chain. Having been a member of this organization for over 12 years, I am quite familiar with the many talents of our more than 700 members. We are privileged to have among us some of the most knowledgeable nuclear scientists, engineers and accountants in the world. Lack of papers, therefore, can not possibly be caused by lack of subject matter or talent. The historical dedication and support of the membership also precludes lack of interest in the Journal. Some other reason or reasons, therefore, must be the cause of the occasional shortage of papers. I believe it is scientific involvement. The Institute is filled with such busy people doing so many things to improve safeguarding of nuclear material that setting aside enough time to prepare a paper for publication oftentimes seems impossible. I would encourage each of you, however, not to give in to the temptation of putting off or putting aside publishing your work. We all know the obvious reasons for publishing: 1. if you don't publish your work someone else might publish it instead, 2. if work is not published others can not benefit from knowledge of it. The latter can not be stressed too much. The INMM is the only professional society dedicated to the principle of safeguarding nuclear material. This includes material control and accounting, physical protection, transportation and waste management. Chances are, if you publish what you are doing, a large proportion of our membership will not only be interested in it but also can benefit from it. Remember, this is our Journal and only through our support and personal involvement can all of us benefit from it.

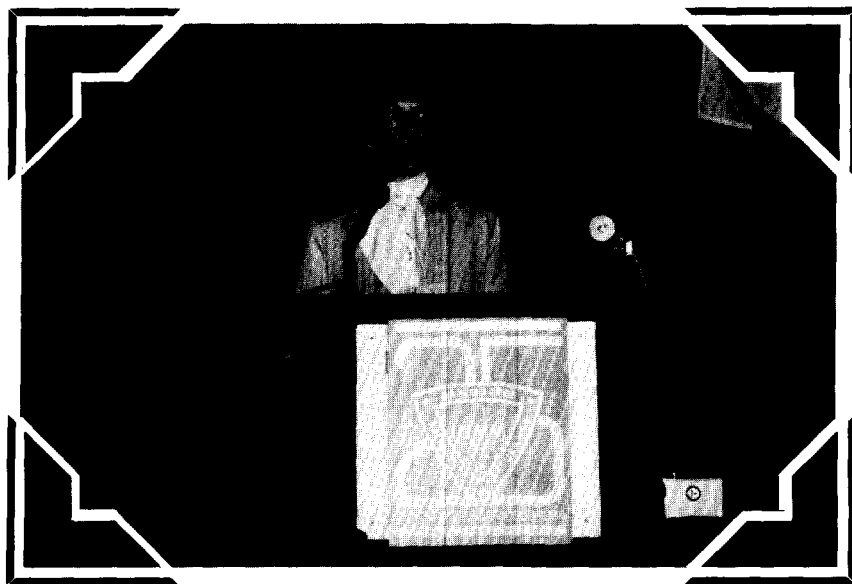
## INSTITUTE ADOPTS DYNAMIC NEW LOGO



At the last Executive Committee Meeting, INMM Directors enthusiastically endorsed a new logo for the Institute. The new logo incorporates the Institute's increased emphasis upon waste management and transportation in addition to safeguards. The professionally designed logo shows the dynamic image of the Institute in the nuclear community.

# INMM 1984 ANNUAL MEETING HIGHLIGHTS

Retiring Chairman John Jaech welcomed members and guests of the Institute at the 25th Annual Meeting in Columbus, Ohio. The Institute's 1985 Annual Meeting will be held July 21-24 at the Albuquerque Regent Hotel in Albuquerque, New Mexico, USA.



Newly elected Chairman Yvonne Ferris presented her goals for the Institute during her two-year term as Chairman.



During a break in the Annual Meeting, Local Arrangements Chairman Harley Toy met with Bob Keepin and Tom Shea.



Chairman John Jaech congratulated the first group of Institute Fellows (L to R: William Higinbotham, G. Robert Keepin, Ralph Lumb and Sam C.T. MacDowell). Fellows not present include James E. Lovett and Richard A. Schneider.



INMM Secretary Vince DeVito (Goodyear Atomic) receives the 1984 Distinguished Service Award from Chairman John Jaech.



Dr. Carl Bennett (Battelle Northwest) receives the 1984 Distinguished Service Award from INMM Chairman John Jaech.

## INMM 1984-85

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**VICE CHAIRMAN** Charles M. Vaughan

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**TREASURER** Robert U. Curl

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Thomas E. Shea

James P. Shipley

Robert J. Sorenson

Dennis W. Wilson

### 1984-85 COMMITTEE CHAIRMEN

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Annual Meeting Exhibits	James C. Hamilton
Annual Meeting Program	John F. Lemming
Annual Meeting Contributed Papers	Charles M. Pietri
Annual Meeting Invited Papers	Robert Brooksbank
Annual Meeting Local Arrangements	Denis L. Mangan
Annual Meeting Registration	Gary Carnival
Awards	Ralph E. Caudle
Bylaws & Constitution	Roy G. Cardwell
Certification	Barbara M. Wilt
Education	Harley L. Toy
Examining	James E. Lovett
Headquarters & Journal	John W. Messervey
Journal Technical Editor	William A. Higinbotham
Long Range Planning	Sam C. T. McDowell
Membership	Roy G. Cardwell
Material Control & Accounting TWG	Darryl B. Smith
N-14 Standards	John W. Arendt
N-15 Standards	George A. Huff
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Safeguards	Leon D. Chapman
Training Coordinator	Dean D. Scott
Transportation TWG	James R. Clark
Waste Management TWG	E. R. Johnson

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Pacific Northwest	Herb E. Smith
Southeast	Wendell L. Belew
Japan	Ryohei Kiyose
Vienna	Marco Ferraris

### INMM STAFF

Executive Director	John E. Messervey
Administrator	Beth Perry
Accounting Services	Carol Vraney

## INMM CALENDAR OF EVENTS

### FEBRUARY 12-15, 1985

Material Control & Accounting TWG  
Process Control Monitoring  
Opryland Hotel  
Nashville, TN

Chairmen

Robert Dube

Donald Emon

Darryl B. Smith

### FEBRUARY 28-MARCH 1, 1985

Executive Committee Meeting  
Albuquerque Regent Hotel  
Albuquerque, NM

Chairman

Yvonne M. Ferris

### APRIL 30-MAY 2, 1985

Waste Management TWG Seminar  
Hyatt Regency Washington on Capitol Hill  
Washington, DC

Chairmen

E. R. Johnson

John McBride

### JULY 22-25, 1985

26th Annual Meeting  
Albuquerque Regent Hotel  
Albuquerque, NM

Chairman

Charles M. Vaughn

### SEPTEMBER 16-18, 1985

Transportation TWG Seminar  
Hyatt Regency Washington on Capitol Hill  
Washington, DC

Chairmen

John W. Arendt

James R. Clark

### TO BE ANNOUNCED

Information Display and Control Workshop

Chairman

J. D. Williams

### TO BE ANNOUNCED

Error Propagation Seminar

Chairman

Darryl B. Smith

### TO BE ANNOUNCED

Shortcourse on Safeguards (Certification)

Chairman

Barbara M. Wilt

# JAPAN CHAPTER REPORT

The Executive Committee of the Japan Chapter of INMM met in Tokyo on August 10, 1985 to confirm the following results of the election of officers and committee members for FY 1984-85 and FY 1985-86:

Chairman	Ryohei Kiyose (Univ. of Tokyo)
Vice Chairman	Mitsuho Hirata (JAERI)
Secretary	Yohko Iwamatsu (NMCC)
Treasurer	Reinosuke Hara (Sieko)
Members at Large	Tohru Haginoya Kazuhisa Mori Hideo Kuroi Masumichi Koizumi

Executive Committee meetings were also held on June 7, 1984 and November 10, 1983.

Members of the Japan Chapter as of August, 1984 are 90 in number, showing a steady increase over the previous years (82 in FY 1982-83). Members are from the following organizations:

Nuclear Energy Organizations	37
(Nuclear Material Control Center, JAERI, PNC, JAIF)	
Government Offices (Sci. Tech. Agency)	2
Universities	6
Industries, Electric Power	9
Others	36
	<u>90</u>

The 5th Annual Meeting was held in Tokyo on Tuesday, June 26, 1984. 125 persons participated in the meeting, including W. Alston of IAEA (Vienna Chapter), 49 Japan Chapter members and 75 non-members. The program of the meeting was as follows:

**Opening Remarks** H. Kuroi (Program Chairman)

**Chapter Chairman's Speech** Y. Kawashima (Chairman)

**Invited Special Session,** Chairman: M. Kuramoto (NMCC)

- (1) Recent International Situation of Safeguards  
T. Maruyama (STA)
- (2) Standing Advisory Group for Safeguards Implementation  
T. Haginoya (MMC)
- (3) Problems and Pleasures of an IAEA Inspector in Japan  
W. Alston (IAEA)

**Session 1** (All invited), Chairman: M. Koizumi (PNC)

- (1) Spent Fuel Verification by Ultraviolet Cherenkov Emission  
A. Nakaoka (EPCRI) et al.
- (2) ANS/IAEA Topical Conference on Safeguards Technology—  
The Process/Safeguards Interface  
S. Takahashi (PNC) & M. Kikuchi (NMCC)

**Session 2 Material Balance—I,** Chairman: K. Ikawa (JAERI)

3 Technical papers were presented by Y. Hisamatsu (CRC), H. Sano (JNF), and M. Omae (PNC)

**Session 3 C/S and PP,** Chairman: M. Kajiyoshi (NMCC)

2 technical papers were presented by M. Masuda (JNFS) and H. Kuroda (PNC)

**Session 4 Inspection,** Chairman: K. Tsutsumi (PNC)

2 technical papers were presented by K. Ikawa (JAERI) and H. Mizuno (PNC)

**Session 5 Measurement & Analysis,** Chairman: H. Kawamoto (PNC)  
3 technical papers were presented by M. Akiba (PNC), K. Ochiai (PNC), and C. Konagai (Tosiba)

**Session 6 Material Balance—II** Chairman: T. Osabe (JNF)

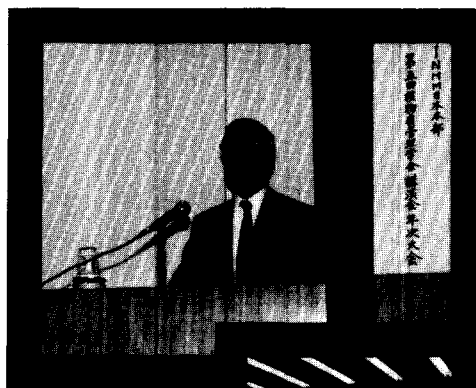
3 technical papers were presented by H. Nishimura (JAERI), and H. Ihara (JAERI), and H. Oake (MMC)

**General Business Meeting** Chairman: R. Kiyose (Vice Chairman)

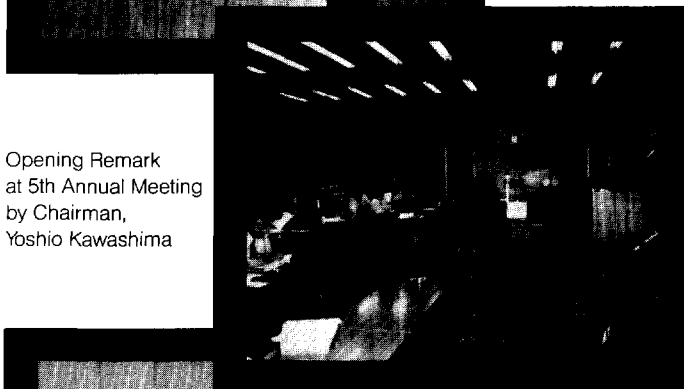
- (1) Business Report by M. Hirata (Secretary)
- (2) Audit Report by R. Hara (Treasurer)

**Closing Remarks** R. Kiyose (Vice Chairman)

The program committee for the 5th Annual Meeting was composed of H. Kuroi (Chairman, JAERI), K. Ikawa (JAERI), H. Kawamoto (STA), M. Kajiyoshi (NMCC), T. Osabe (JNF), K. Tsutsumi (PNC), S. Yamagami (MMC), and H. Wakabayashi (U. of Tokyo).



Chairman:  
Yoshio Kawashima



Opening Remark  
at 5th Annual Meeting  
by Chairman,  
Yoshio Kawashima



Program Chairman:  
Hideo Kuroi



Vice Chairman:  
Ryohei Kiyose



# AWARDS COMMITTEE REPORT

## RALPH E. CAUDLE, CHAIRMAN

The 25th Annual Meeting Banquet provided the opportunity for Chairman John Jaech to recognize this year's awards recipients. Carl Bennett and Vince DeVito were named to receive the 1984 MERITORIOUS SERVICE AWARDS and Bernard Gessiness was named to receive the 1984 DISTINGUISHED SERVICE AWARD.

Carl Bennett, of Battelle Memorial Institute and a senior member of the INMM, is currently involved in technical studies aimed at improved nuclear materials management on an international scale. He has made major and lasting contributions to the theory and practice of safeguards in his more than four decades of work in the field. His professional career has also gained him prominence in the field of statistics, particularly in the development and application of statistical methods to the physical and life sciences. He has also attained wide recognition in behavioral and social sciences for his work related to models and indices for crime statistics. Of special interest to the Institute are Carl's contributions in the fields of domestic and international nuclear materials safeguards. These contributions which are well known to fellow workers include international advances in inspection methodology of and evaluative methods for safeguards effectiveness.

Vince DeVito's contributions to both the nuclear industry and the INMM for more than thirty years have been many. In the early 1950's, he began his nuclear career with Goodyear in nuclear materials control. From this beginning he was made head of that department, thence to Manager of Safeguards and Security, and in 1982 was made Plant Manager of the Portsmouth gaseous diffusion facility. He has made many contributions to the business and technology of separation enrichment and is recognized both nationally and internationally for his work. Vince has been an active member of the Institute since 1960 and served as its Secretary since 1972. Much of the growth and stability of INMM is directly attributable to his hard work during a time of transition from a fledgling nuclear group to a full grown technical society; and it has been through his many voluntary hours that INMM has maintained the necessary management organization to operate it successfully.

Bernie Gessiness has been in nuclear materials management since 1952 when he came to NLO, Inc. Since 1959, he has been Supervisor of Nuclear Materials Control having previously held the position of Chief of Analytical Development. As supervisor, he has for many years administered the ADE/ERDA/DOE materials management

## 1985 AWARD NOMINATIONS (S)

I nominate: \_\_\_\_\_

of: \_\_\_\_\_  
Company Name/Address

for the:  Distinguished Service Award  
 Meritorious Service Award

Justification: (Qualifications/Contributions)

\_\_\_\_\_  
\_\_\_\_\_  
\_\_\_\_\_  
\_\_\_\_\_  
\_\_\_\_\_  
\_\_\_\_\_  
\_\_\_\_\_

Signature

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I nominate: \_\_\_\_\_

of: \_\_\_\_\_  
Company Name/Address

for the:  Distinguished Service Award  
 Meritorious Service Award

Justification: (Qualifications/Contributions)

\_\_\_\_\_  
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\_\_\_\_\_  
\_\_\_\_\_  
\_\_\_\_\_

Signature

Return to: INMM  
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programs at NLO maintaining liaison between the company and other contractor and licensee facilities. He is both a past Chairman and Vice-Chairman of INMM and has been continuously active in the organization since its early beginnings. He was one of the first four Certified Nuclear Materials Managers of the INMM. Bernie is an internationally recognized authority on inventory control of strategic materials of high intrinsic value. He wrote and published the first nuclear materials control and accountability plan for a DOE contractor that has since been used as a model for other sites.

It is now time for us to begin the nomination process for the 1985 awards. Your help is needed. Please use the enclosed award nomination form to recommend those candidates you consider worthy of recognition.

#### Chronology of Awards

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##### Distinguished Service Awards

1979—W.A. Higinbotham  
1980—Louis Doherty  
1981—Roger M. Smith  
1982—G. Robert Keepin  
1983—International Atomic Energy Agency,  
Department of Safeguards  
1984—Bernard Gessiness

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##### Meritorious Service Awards

1979—None  
1980—Douglas E. George  
1981—None  
1982—Ronald D. Smith and John H. Ellis  
1983—Edward Owings and Duane A. Dunn  
1984—Carl A. Bennett and Vincent J. DeVito

---

##### Student Awards

1979—Mark H. Killinger  
1980—Mohammad Sharafi, M.I.T.  
1981—Houng Y. Soo, University of Washington  
1982—Paul E. Benneche, University of Virginia  
1983—Terry L. Zimmerman, Idaho State University  
1984—None

---

##### One-Time Awards

1978—Industry Award, presented to Tri-State Motor Transit, Inc.  
1982—In Appreciation Award,  
presented to E.R. Johnson and Associates

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# A REMINISCENCE, 1944

## Chapter 4\*

### R.D. Smith

In 1944 when I arrived in Oak Ridge, I was assigned to Barrack M. It was neither better nor worse than many other barracks I'd been assigned to since I had entered the Army. On further consideration, let me say, it may have been the best. The quality of a barrack is the quality of the men who occupy it.

I remember one very strange barrack in which I was an inmate at Camp Santa Anita in California. Almost all the other guys were Texans, and it seemed as though every one of them had a guitar. The song of the day was "Tumble-Weed." I've never heard such a cacophony...no two guys in the same key...nobody who really knew the song. (But there it is again. Everyone was singing or trying to.)

Barrack M was different. There everything was the game of bridge in every spare minute! Now I had learned the Culbertson system, and later Goren, and I knew the more common conventions; but I wasn't close to being in the same league with those guys. When I did play with them, I quickly became just a rack for the fourth hand.

One group of four...Schwartz (Columbia), Sumner (Cornell), Yarazunis (RPI), and Symmes (Tufts)...were honing and fine-tuning Goren. As I remember, they had it down to one-half points. No bid meant what it sounded like it meant until the final blow was struck. And, believe me, the pair who struck that blow almost always made it.

The reason I said "group of four," above, is that "table of four" would have been inaccurate. We had no tables. We played on our bunks.

There is something I would like to add at this point to explain why Barrack M was so different. It is a quote from the SED (Special Engineer Detachment) yearbook published in late 1945.

"The average Army General Classification Test Score for the detachment is 133, undoubtedly one of the highest averages for any single unit in the Army."

Well, I happen to know my AGCT score; and I sure dragged the average down. The AGCT is (or was) a classical IQ test. It was such a pleasure to be surrounded by near-geniuses. An average guy like me almost never won a game of any kind but the winners were almost always gracious. Yes, there were a few haughty ones; but there always are whether they have brains or not. And our outfit had brains!

We were very like a small college. We had a football team, a baseball team, and a basketball team, though I don't remember the latter clearly. But I remember the football and baseball teams who toured around the local small colleges and Army camps and almost always won.

The football schedule and results were as follows:

Opponent	SED	Opp.
Appalachian State Teachers College	27	6
Tennessee Polytechnic Institute	12	6
Fifth Division Field Artillery†	53	0
Milligan College	13	6
Tenth Infantry†	6	33

†Both from Camp Campbell, Kentucky

\*Note: Volume XIII, Number 1/Spring, 1984 incorrectly titled Chapter 3 as Chapter 2.

For example, and I hope I don't embarrass the gentleman, there was Robroy (Bob) Millican—a huge man—who had played on the line with Texas A & M. He stood an inch or two over six feet and weighed something like 240 to 260. There may have been a little fat on him, but it was the hardest fat I've ever felt. Those poor small colleges never knew what hit them when confronted by a line that was the likes of Bob.

As you know by now I have a terrible time keeping things in chronological order. Nine months before I was sent to Oak Ridge, I was sent to Lehigh University (Bethlehem, Pennsylvania) in something called the Army Specialized Training Program (ASTP). There was a whole battalion of us at Lehigh, divided as usual, into companies. Bob Millican was in my company.

There were a bunch of bored professors teaching a bunch of bored guys courses they mostly had had before. I suppose the review was good for us, but it seemed interminable. Naturally we had compulsory "calisjumpics" and other exercises. Our company elected touch football as our exercise, so we divided up into teams. Our team drew the team Bob was on as our perpetual opponent, so we developed a strategy. I am glad there was only one of him on that team.

But what a strategy! I was to oppose Millican on the line. I was to give up over 100 pounds and maybe six inches in height. We were counting on his good nature. We all knew he was good natured. If that tank of a man had come straight ahead I would have been crippled for life, at least. The strategy worked; when he paused to pick me up and set me aside, we tended to gain yardage. Once in a while our team even won. He was and is a prized friend.

After the war had ended—I've forgotten whether we were still in service or not—Bob bought a car. The assembly lines for civilians had begun to run again! You can imagine the pent-up demand. What a terrific car! I guess we were out of service by that time and working in Oak Ridge as civilians—lots more profitable than being G.I.'s. Well, that was a grand car. It had everything from soup to nuts both inside and out except it had wooden bumpers. Wooden bumpers! They were massive and undoubtedly of the best hardwood, but they sure were funny looking. I don't know to this day what the problem was. Maybe the steel people couldn't retool fast enough. Maybe the demand was too great. At any rate, wooden bumpers! Bob and his wife took us for a number of enjoyable rides in that car.

Back at the second laboratory where I worked, there was always a furor; we were trying to do a thing that had never been done before. I've never seen so much concentrated nitric acid—whole sinks full. What came to us were the target items—experimental ones—from the alpha buildings. The alpha buildings were the first stage in the Calutron isotope separation process. There were and are five of them; they are used for other things now. Apparently some of the scientists were trying to correct their aim in those magnetic fields. That beam of ions did not behave as it should all the time. So, we were to prepare analytical samples from various areas of the targets by very carefully washing or burning them. I say washing or burning.

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The principal part of the target was usually a copper thing we called a "can." It didn't look at all like a can but who cares. The rest of the stuff was graphite. Intricate pieces that had to be scraped or burned or both.

Well, working on the cans was simple if one liked nitric acid. Since I was the only man in the laboratory, women were assigned this nasty job. Each girl was provided with a chisel, elbow-length rubber gloves, and many instructions on where the nearest bottle of base was to neutralize that acid. Believe me, the girls listened. Inevitably, a girl would chisel her glove once in a while and run for the sodium bicarbonate solution.

Of course we had hoods, and pretty good ones. I suppose there were efficient fans up on the roof. Now everyone knows that when one immerses copper or uranium in concentrated nitric acid, a by-product of the dissolution is nitrogen peroxide—a nasty, acrid, brown gas. What many don't know is that it entrains uranium pretty well and, I believe, copper also. So from time to time we'd wash down the insides of the hoods. One night the acid had been drained from the sinks. I called on an agile, small woman and told her to wash down the hoods, to put the wash in five-gallon bottles, and to label them carefully.

She had to be small and agile to get up in those hoods. She knew all the preparations—a towel or something over her hair, the rubber gloves to her elbows—it was altogether a nasty job. But I only watched for a minute or so because I had so many other things to do. Hours later I came back. Here was a row of five-gallon bottles each neatly labeled "Hood Worsch." Well, she labeled the words just like she spoke them.

I also had a pair of twins working for me. They couldn't have been more than eighteen, probably less. During the war we weren't fussy—if they wanted to work, we took them. They were cute. One of them did all the talking for both. The other never said a thing. They were good workers and quick to learn. Today I would call them nice little girls. Then, that would have been an insult—they were war-workers.

One day after a three-day break, the silent one came in wearing a long-sleeved sweater. It was definitely not sweater weather, so I asked the talker why the sweater. She told me her sister had gone to Chattanooga during the break with a sailor boyfriend and he had persuaded her to get tattooed! The talker even got "little silence" to show me the tattoo. My word! It covered the whole outside of her left upper arm. There was a scroll on which it said "Love" and the sailor's first name. There were also cupids and the usual hearts and flowers. It was schlock, but this was a master-piece of schlock!

The talker explained that their mother did not know about the tattoo and that "little silence" vowed never to let her see it. I can imagine how that worked out.

I sure hope the sailor came back and married "little silence." They would have had great one-sided conversations. You see, the sailor's first name was somewhat rare. If she married someone else—say a "John" or a "Richard"—the poor groom would have topped himself to death within a year confronted with that tattoo.

There was one girl in the laboratory who was twenty and looked thirty—not that she looked bad, just older than her years. She elected to take a solitary and dirty job in a room of the laboratory in which no one else ordinarily worked. She was the one with the chisel who scraped the pieces of graphite. I had noticed that she always put her large purse near her on the laboratory bench where she was working. Who's to argue with small eccentricities when impeccable work is being done?

Then one day I was walking through that room and there she was zonked out on the floor. I sure didn't know what was going on, but I made the right phone calls—guards, ambulance, etc. They took her away.

At age twenty-two I was a little shy about these things, but nevertheless I went to Mary who was about fifteen years my senior. I asked her what had happened and was it female trouble? Mary said simply, "She ran out." As she explained, it all came together—the big purse—the solitary job. The poor girl had probably been an alcoholic from the age of ten. She drank what we called "sploe", the Tennessee sour mash "moonshine" whiskey, raw and unadulterated. I had noticed a sort of burn on her upper lip, but it meant nothing to me so I dismissed it. Some misfortune had prevented her from replacing her bottle.

There was nothing to do but to explain it to my supervisor and together we went to his supervisor. That factotum must have done some pretty fancy explaining too, but we needed that girl. If her normal state was drunk, so be it. She never acted drunk. She was lucid and efficient.

One day I was arriving at work in the North parking lot and found myself walking maybe twenty yards behind her. She hollered something at the guard like "What do you say Jack?" He said something like, "You okay today, Miss?" She said, "Yes" and walked in.

I think the whole exchange meant, "Do you have your bottle, and is it full?" There was no way for me to verify that; it's only my suspicion. But I think there would have been an exchange of bottles at the guard shack had she said "No."

Do I mean anyone could work at Y-12? Heavens, no! There are a few who were faulted but too good to lose. There were a few who were faulted whom we couldn't lose fast enough! But, that's for another episode.

NEXT: Gunk and more gunk.

# N14 ACTIVITIES REPORT

John W. Arendt recently assumed the chairmanship of the N14 Committee with Meriam Pellettieri serving as vice chairperson and Marilyn M. Warrant as secretary. The N14 management committee includes:

Chairman/John W. Arendt	Members/Richard T. Haelsig
Vice Chairman/Meriam Pellettieri	James W. Lee
Secretary/Marilyn M. Warrant	Edmund C. Tarnuzzer

A management committee meeting was held in Knoxville, TN on November 7, 1984. The N14 Committee membership including continued interest to serve is being updated and a full committee meeting is being planned for the second or third quarter of 1985.

Preliminary planning has been started to conduct a seminar on packaging and transportation of radioactive materials to be held in Washington, DC in September, 1985.

An update on the status of current standards is:

Standard No. and Title	Status
<b>ANSI N14.2</b> , "Tiedowns for Transport of Fissile and Radio-active Material Containers Greater than One-Ton Truck Transport"	Negative ballots are being resolved
<b>ANSI N14.6</b> , "Special Lifting Devices for Shipping Containers Weighing 10,000 Pounds (4500 Kg) or More for Nuclear Materials"	Negative ballots are being resolved

**ANSI N14.9.2**, "Packaging of Nuclear Power Plant Radio-active Wastes for Transportation" (after this standard is approved and issued, N14.9.1 will be withdrawn) Ready for balloting by N14

**ANSI N14.10**, "Guide for Liability and Property Insurance in Shipping Nuclear Materials" Approved by N14 conditioned on addressing comments

**ANSI N14.23**, "Design Basis for Resistance to Shock and Vibration of Radioactive Material Packages Greater than One-Ton in Truck Transport" Negative ballots are being resolved

**ANSI N14.24**, "Barge Transport of Radioactive Materials" Negative ballot is being resolved

**ANSI N14.27**, "Carrier and Shipper Responsibilities and Emergency Response Procedures for High-way Transportation Accidents Involving Truckload Quantities of Radioactive Materials" Approved by N14 conditioned on addressing comments

**ANSI N679 (renumbered to N-29)**, "Guide for Writing Operating Manuals for Radioactive Materials Packaging" Ready for balloting by N14

ANSI 14.3, N14.5, N14.7, N14.9.1, N14.10.1, and N679 (will be renumbered N-29) are being reviewed to determine whether the standards should be revised, reaffirmed, or withdrawn.

A complete status report of all standards and/or working groups will be included in the next activities report.

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# BOOK REVIEW

**LESLIE G. FISHBONE**  
Brookhaven National Laboratory

SOCIAL AND ECONOMIC ASPECTS OF RADIOACTIVE WASTE DISPOSAL: CONSIDERATIONS FOR INSTITUTIONAL MANAGEMENT  
Panel on Social and Economic Aspects of Radioactive Waste Management, National Research Council  
National Academy Press, Washington, D.C. 1984 xii, 175 pp.

"The socioeconomic and institutional issues associated with high-level radioactive waste management are complex and challenging." Having alerted the reader that this will be a difficult task, the National Research Council's Board on Radioactive Waste Management launches into an analysis of these issues. The degree of difficulty quickly becomes evident. To consider how much this study does to unravel the complexity, or to meet the challenge, some background information is first required.

In 1980, the Department of Energy (DOE) requested the National Research Council to conduct a study of the socioeconomic aspects of nuclear waste repository siting. The council responded by impaneling, under the aegis of its Board of Radioactive Waste Management, a study group to address these social and economic issues. This group, hereafter referred to as the panel, was composed of sixteen members, largely social scientists with university affiliations. The panel met nine times between 1980 and 1983, solicited views and reactions from interested individuals and organizations, was briefed by various DOE staff and contractors and discussed its approaches with representatives of environmental organizations. Drawing on these resources, as well as experience of individual members, the panel produced the study which is the subject of this review.

In defining the scope of its study, the panel chose to restrict itself to high-level radioactive waste management, concentrating on the disposal of spent fuel in a mined geologic repository. (The decision not to consider low level radioactive waste management was, I think, a bit unfortunate in light of present need.) The panel extended its mandate, however, to include a consideration of issues related to the transportation of radioactive waste or spent fuel, as well as repository siting, construction and operation. Within these confines the study undertook to do four things: (1) Identify major socioeconomic considerations in the location, construction and operation of a generic repository. (2) Assess what is known about these considerations, and the extent of the associated data base. (3) Determine the applicability of what is known to siting a repository. (4) Suggest a strategy for incorporating socioeconomic factors into the repository siting process.

One must begin such a study by defining "socioeconomic" in an operational sense. The panel chose a broad definition, in which socioeconomic apparently refers to "virtually any non-technical effect." This definition is contrasted to the narrow view that the term should be limited to quantifiable changes in local demographic, economic or residential patterns. Such a broad definition is certainly useful in a scoping study, to make certain that things aren't overlooked; but there is also the danger, I think, of becoming overwhelmed. Having adopted the definition, almost at the outset the panel seems to have concluded that the existing body of social science knowledge is inadequate for the task of siting a radioactive

waste repository. We are told that more research is needed; doubtless this is true. One wishes, however, that the panel had devoted more attention to assessing the state of knowledge according to a restricted set of quantifiable factors. Beginning with a fairly narrow definition, where analyses might be more intensive, might provide some guidance on how to address additional issues which may be perceived as the definition is broadened. To simply list a broad spectrum of potential concerns, unranked in terms of perceived impact, in areas where there is little precedent, and then say that social science knowledge is at present inadequate, is not much help to the reader uninitiated in unraveling complexity. In fact one could be left with *some sense of frustration—as I was—in that accumulating the requisite social science database may seem an even more formidable task than generating the technical information required, or in that general socioeconomic considerations may be perceived as so intractable that they will be ignored, and a series of ad hoc approaches developed.* I doubt if the panel wished to evoke either of these extreme perceptions.

The panel considered socioeconomic factors relating to four separate areas: general public concern over nuclear power, transportation of radioactive waste or spent fuel, repository siting and institutional issues. Each area is addressed in a separate chapter of the report. The format varies from chapter to chapter and does not bear any particular relation to the four charges to the panel, as listed above. Each chapter more or less stands alone, with the overall effect that the report seems a bit disjointed. This may be inevitable, at least to some extent, in a report where different sections are prepared by different people; presumably this is the case here.

The chapter on public concerns begins by citing evidence that public concern over nuclear power has increased in recent years. It considers the correlation of demographic factors with public concern and concludes that women are more concerned than men; possible reasons are discussed but the panel found that, thus far no authoritative explanation of this "gender gap" has been offered. Other demographic correlations are then found to be less well understood. Possible causes for public concern are next addressed. The panel finds some evidence to support the roles of four factors: fear, the nature of the hazard presented by nuclear power, conflict of value systems, and institutional credibility and distrust, in generating public concern. Interestingly, the panel finds that the trend of research results does not support the position that public concern is the result of public ignorance.

The chapter closes with a critical perspective on methodological and data-base limitations which almost sounds like a disclaimer, and which, as the panel points out, might lead the reader to question the applicability of social science to the problem at hand. (See above). It is argued that the reverse is true, although I wish this had been made more clear. It seems that the major difficulties involve questions of interpreting the impact of actions taken or positions held by various factional groups, of unbiased polling of the general public, and the fact that the data themselves may be in a state

of flux. In particular, regarding opinion polls, the panel feels that "...extending the conclusions of this body of research to policy applications in repository siting is of doubtful merit." (It would be interesting to learn if the panel would apply the same caveat in other areas, such as waste shipping, emergency planning, etc.). Evidently the panel believes that such studies may be most useful as qualitative indicators of those factors which merit careful attention in repository siting.

I think there may be something of an inconsistency in the panel's finding on the role of "public ignorance" in generating public concern, with respect to their other findings. In discussing the possible role of fear in causing public concern, the perceived similarity between *nuclear weapon explosions and reactor accidents is cited as a contributing factor*. Surely, this is a manifestation of "public ignorance." When considering the effect of the unique risk presented by nuclear power in generating public concern, it is argued that the public views risks associated with nuclear power and risks associated with other activities in a nonequivalent manner. Would this be the case if the public were more aware (i.e. less ignorant) of those equivalencies which do exist? The panel in fact cites one nonequivalent study in which the most knowledgeable people were the most strongly supportive of nuclear power. This is the sort of thing which the panel feels merits further study. The panel identifies a need for improved public understanding in any event.

This brings us to a final comment on this chapter: It is almost entirely concerned with overall perceptions of "nuclear power," and thereby suffers some of the shortcomings of the broad approach as mentioned previously. Nuclear waste management issues are a sub-set of the issues involved with nuclear power. Indeed, a large fraction of low level radwaste is generated by operations which have nothing to do with nuclear power. Since certain of the concerns about nuclear power, such as reactor accidents, may have very little to do with siting a repository, public concern over nuclear power may or may not mirror specific concerns with repository siting. I suspect that some people who strongly object to the spread of power reactors might be much less resistant to the development and siting of a repository to dispose of spent fuel or high-level waste. It would have been helpful to have had some insight into the extent to which waste management concerns could be decoupled from overall perceptions of nuclear power. Perhaps a review of attitudes toward low level waste management, where needs are more pressing, where sites have been established, where more sites are being considered, and where regional political alliances are currently being formed, would have been useful, particularly in view of the regional approach discussed by the panel later on.

Issues related to transportation are integral to the waste management network. In addressing these, the panel considered a reference case of 113 reactors operating in the period 1995-2000, and analyzed various options for repository siting and transportation mode (truck vs. rail). Shipping patterns for different assumptions about repository location were developed for the panel by Oak Ridge National Laboratory, using existing computer models.

The panel considered the socioeconomic effects associated with the various options. While no firm recommendations were offered, I think the reader would find these considerations quite instructive. The main tradeoff seems to be on the question of a single site, which would create a transportation "funnel" and a greater regional inequity, vs. a regional approach where several repositories are constructed in different parts of the country. The panel considers a perception of regional equity as an important and perhaps necessary condition for achieving social consensus on a waste management program. At the same time the panel acknowledges that disputes could occur more frequently in the search for multiple sites. This seems to me to be a balanced presentation which is very much to the point, and defines the issue in a manner which will be useful in further considerations.

The panel identified several obstacles to a system relying predominantly on rail transport, as the DOE presently intends, and recommends that these receive further review. Overall, with regard to the waste management network, the panel emphasized that many of the socioeconomic interrelationships are not well understood, and stressed the need for increased effort in this area, as compared with the level of effort on repository technical design. The argument is compelling, and I would only remark that the two should go hand in hand, since repository design will certainly influence the spectrum of siting options available.

In considering the socioeconomic issues involved with the siting of the actual repository itself, the panel restricts itself to the immediate vicinity of the repository. A generic repository site is described, which envisions a seven-year construction period, involving up to 4200 workers. Subsequently, about 1000 workers are required for operations. Socioeconomic characterization assumptions for three regional sites are presented for reference, drawn from DOE analyses.

The panel distinguishes two types socioeconomic effect: "conventional effects" which may result from the siting of any large-scale industrial operation, and "special effects," which are associated with the radiological characteristics of the repository. I found this approach appealing. At this point the panel simply lists the types of conventional effects which could occur during repository siting construction and operation. These include things such as changes in property value, noise, social pathologies, location transfer costs and institutional adaptations. The panel seems to take a "wait and see" position at present and does not attempt a rank ordering of these effects, either in terms of their probability of occurrence, or on whether the individual changes would be beneficial or detrimental. The panel devotes one and one half pages to comment on special socioeconomic effects associated with the radiological nature of the facility. Here the panel seems to feel that at present not much can be said, but warns that these effects could well exceed the conventional effects associated with the repository, and be difficult to deal with. The discussion closes with a recommendation for a general program to deal with socioeconomic effects in repository siting, including a prompt evaluation of effects which appear as the

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siting actually takes place, and development of effective means of mitigating or compensating for these effects, by marshalling both financial and technical resources. One can hardly argue with the spirit of these recommendations; some however might wonder what level of resources the panel would contemplate.

The report closes with a discussion of institutional means for addressing socioeconomic effects in repository siting. Most of the discussion centers on the Nuclear Waste Management Policy Act (NWPA). The panel argues that the time schedule imposed by the order may be too restrictive for DOE to adequately provide for local concerns or participatory opportunities. The panel also concludes that there is an institutional gap in the Act, in that the Act relies on the participation of state agencies but does not specifically provide a means whereby concerns of the affected population would be taken up by these state agencies. Since the Act is in fact in place, the panel argues for an increased role for informal processes in allowing local public participation and in resolution of disputes.

One panel member in a supplementary comment goes farther and advocates an increased normal role for public participation, both in rule-making and in adversarial procedures. Neither view offers comment on the level of public participation envisioned. The discussion ends with a comment on institutional requirements to address socioeconomic issues in transporting radioactive wastes, and in particular argues for the elimination of inconsistencies in existing NRC-DOT regulations. (I think that there has been some clarification of this matter in recent court actions, which probably occurred after this report was completed.) Finally, the report contains a useful appendix describing the results of Oak Ridge's transport model calculations.

To return to the original question I believe this study is helpful in at least two ways. First, while one might wish for a more incisive treatment in certain areas, one must also accept the state of the art. Those findings which seem relatively clear at this point deserve, I think, to be taken seriously in developing waste management strategies. In other areas, this report is probably most useful in providing a critical perspective on the role of social science in coping with these issues, which will probably vary from reader to reader, and may in itself be a socioeconomic issue.

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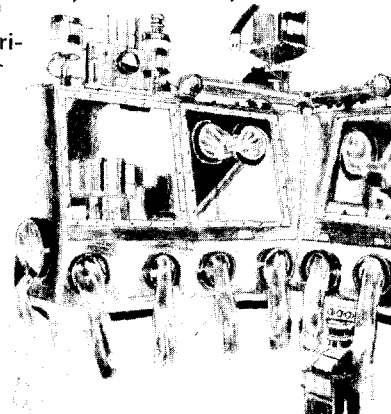
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# NDA TECHNOLOGY FOR SAFEGUARDING URANIUM HEXAFLUORIDE

**N.S. BEYER, D.R. TERREY**

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## Abstract

The measurement of uranium hexafluoride ( $UF_6$ ) is an important safeguards activity which is conducted on a regular basis during inspections of facilities handling  $UF_6$ . For the past six to eight years, most of the measurements of  $UF_6$  for safeguards purposes have been made upon solid  $UF_6$  contained in large storage cylinders. This technique combines the weighing of cylinders with a determination of the U-235 enrichment accomplished by gamma ray spectroscopic measurement of the material. To enhance the reliability of cylinder measurements, new weighing instrumentation is now being used, consisting of a small, portable load-cell apparatus which can be used in conjunction with existing cylinder handling equipment. With it, an independent confirmation can be made of weight of  $UF_6$  in cylinders. Recently, the " $UF_6$  Gas Phase Enrichment Monitor" has become available. This instrument can be used to measure the uranium-235 enrichment of a sample of gaseous  $UF_6$ ; the enrichment is determined directly by simultaneous measurement of the U-235 and total uranium concentrations. Calibration using standards of known enrichment is easy and reliable and the accuracy and precision is better than those of measurements on solid  $UF_6$ . It has a direct application at enrichment facilities but it could also be used to measure samples of  $UF_6$  gas collected from storage cylinders.

## NDA Measurement of $UF_6$ Contained in Storage Cylinders

Until recently, most of the safeguards type measurements of  $UF_6$  have been made upon solid  $UF_6$  contained in large 30A or 30B type storage cylinders. The technique, which is fast, relatively simple and non-destructive is well documented in the literature [1,2,3,4]. However, for the sake of completeness, a short summary of the method is presented here.

Gamma ray spectrometric measurements are made of the 186 keV gamma rays characteristic of

and emitted by the U-235 contained in the  $UF_6$ . Since the quantities of solid  $UF_6$  contained in the cylinders exhibit "infinite thickness" to 186 keV radiation the "enrichment meter" method can be applied to determine U-235 enrichment.

High resolution gamma ray measurement instrumentation (i.e. multi-channel analyzer and Ge detector) is used in order to avoid problems of interference from radiation from non-volatile daughters of U that may be plated on the inner surface of the cylinder. The detector should be mounted horizontally on its dewar so that measurements can be made at elevations as low as 30 cm above the ground. A conventional multi-channel analyzer can be used for data acquisition and reduction. A portable ultrasonic thickness gauge, which may be obtained commercially, should be used to measure the wall thickness to an accuracy of  $\pm 0.1$ mm so that corrections can be made for the exponential absorption of gamma rays by the heavy steel walls of the storage cylinders. The detector should be firmly fixed in a lead collimator with a wall thickness of 1 to 2 cm. The measurement point can be either on the side or end of the cylinder but should be 6 cm below the full height as determined by tapping the cylinder with a metal object and noting the change in sound. Measurement times in the range 5-10 min are usually required to obtain acceptable statistical uncertainties for low enriched U. The cylinder wall thickness should be measured at a point within the area covered by the gamma ray measurement.

A commonly used calibration technique consists of declaring one or more cylinders as standards and using as a calibration constant the net 186 keV counts per unit enrichment (%) obtained for the standard. Sometimes two "standard cylinders" are used (e.g. a natural and a 3% enriched). Given the choice of enrichments to use for standards, those with higher enrichments should be used when using only one standard. The simple peak stripping method using a straight line interpolation to obtain the background under the 186 keV peak is quite satisfactory if consistently applied. For the case of using only one standard, the equation for the enrichment of a cylinder with unknown enrichment

U-5/U and a wall thickness X is:

$$U_5/U = (U_5/U)_s \cdot C/C_s \cdot e^{\mu(X-X_s)}$$

where:  $U_5/U$ ,  $(U_5/U)_s$  are the enrichments of the unknown and the standard; respectively  $C$ ,  $C_s$  are the net counts in the 186 keV peak for the unknown and standard, respectively;  $X$ ,  $X_s$  are the cylinder wall thickness of the unknown and the standard, respectively, and  $\mu$  is the linear attenuation coefficient of the cylinder wall material. When two standards are used, correction is made to zero wall thickness.

It is important to note that  $\mu$  may differ appreciably from its theoretical value because of the effects of the poor geometry used for this measurement; therefore, for best accuracy,  $\mu$  should be measured using flat plates of the same material as the cylinders in a geometry closely simulating that of the field measurement.

This "enrichment meter" type measurement of  $UF_6$  storage cylinders has been routinely applied during the past six to eight years during physical inventory verifications conducted for international safeguards purposes at bulk handling facilities. It is combined with the weighing of cylinders so that the amount of U-235 can be determined. Inspection personnel have applied the technique with varying degrees of success and generally the measurement precision has varied from  $\pm 3.0\%$  to  $\pm 6.0\%$  (1 sigma) relative. Some of the main sources of error which have been encountered include: uncertainties in the ultrasonic measurement of wall thickness, variations in background radiation encountered at the cylinder storage locations and difficulties in reproducing geometry and the possibility of cavitation of the solid  $UF_6$  at the points viewed by the detector. Equipment instability and malfunction, caused some problems in the past but is much less common now.

Accuracy of the results is of course dependent upon the reliability of the values of enrichment associated with the operator's cylinders which were used as standards. Until recently it has been impossible to take a sample from these cylinders for mass spectrometric analysis at Headquarters. Sometimes the analyser - detector system is calibrated at Headquarters prior to the inspection by using Headquarters standards. These may consist of containers of  $UO_2$  or  $UF_4$  and are considerably different from  $UF_6$  contained in a typical type 30A or 30B cylinder. Corrections for the difference must therefore be applied to establish a proper calibration. At other times, it has been possible to calibrate the equipment against large storage hoppers of  $UO_2$  available at the facility. The hoppers can usually be sampled and samples sent to Headquarters for a mass

spectrometric analysis to establish the desired independent calibration.

Another aspect of the technique which can affect the accuracy and independence of the results, is the difficulty of confirming the calibration of the operator's weighing system which is used to weigh the cylinders. The gross weight of a cylinder filled with  $UF_6$  approaches 3000 kg and it is difficult for the inspector to provide standard weights covering this weight range. Only relatively small standard weights are usually available to the inspector and thus he can confirm only a small weight range of the operator's weighing system.

#### Load-Cell Based System for Weighing $UF_6$ Cylinders

A system which permits safeguards inspectors to make independent weight-verifying measurements on  $UF_6$  cylinders to a high degree of accuracy using inexpensive portable equipment, has been developed for the IAEA by the U.S. National Bureau of Standards, through the U.S. Support Programme to IAEA safeguards. This system is based on a small load cell and has been tested in various locations in Europe, Asia, and North America. The system was designed to meet a number of requirements. In addition to having an accuracy of about  $\pm 1$  kg, the system had to be easily transportable, relatively inexpensive, easy and quick to use, and it had to comply with operator's safety requirements. All of these objectives have been achieved, and this Load Cell Based System (LCBS) is now in routine inspection use, and has already been applied to the weighing of other large items, such as fuel assemblies.

A sketch of the system is shown in Figure 1. The heart of the system is the load cell, which weighs less than 4kg and is about 14cm long and 8cm in diameter. A strain gauge is incorporated in the load cell, whose electrical resistance is changed by the application of an external load. This change in resistance is sensed and read out on a digital display unit. The load cell plus read-out combination must be calibrated to obtain the best accuracy and the IAEA is in the process of setting up a suitable calibration facility at the Seibersdorf laboratory. Included in the system illustrated are two other mechanical components, the so-called "flexures", mounted above and below the load cell. The purpose of these is to compensate for any possible asymmetries in the suspension system which could produce off-axial forces in the load cell. Such forces could give erroneous readings, when one is trying to achieve the highest levels of accuracy (1 part in  $10^4$ ). For routine inspection use, the flexures will be omitted from the system, since comparative tests have shown that without them, the error level in weighing cylinders is of the order of 1kg, which is quite satisfactory for verification purposes. Commensurate advantages are simplification of the assembly and reduction in head-room re-

quired between the lifting device and the cylinder. Maintenance of this error level without flexures depends to some extent on good design of the other mechanical components in the system, which are usually specific to the facility where the load cell is being used. Usually, as shown, there are connecting blocks, an upper to mate with the facility's lifting crane, and a lower, to mate with the facility's hardware, which is attached to the cylinder for lifting. The O-ring used in the illustrated case was needed to make connection between the facility's lifting hook and the LCBS upper block. The whole system described so far is designed for 30A and 30B UF<sub>6</sub> product cylinders, which, filled, generally weigh about 2 1/2 tons. The capacity of the LCBS could be extended to include the larger feed cylinders which may weigh up to 16 tons, but larger components would be needed, and the system would not be so readily transportable. However, a twin load - cell system has been developed and successfully tested for this application (7). All the components used in the system are commercially available items, with the exception of the connecting blocks, which are specially designed. A transducer-simulator is provided which contains a set of calibrated resistors and acts as a standardizer, used in order to assure that the characteristics of the electronic display unit have not changed. To permit greater freedom, a portable battery pack has been prepared, for application in those areas where no AC power is present. All components together weigh less than 20kg. Omission of the flexures saves about 4kg.

There are some corrections which need to be made to the raw data which are read out from the display. One is the application of the calibration curve for the particular load cell which is being used. Another correction to be made is a temperature correction. A typical calibration function would be:

$$L = \frac{R_t - 0.17}{0.99994}$$

where  $R_t$  is the temperature-corrected display reading and  $L$  is the load on the cell. This shows the almost complete correspondence of the display to a direct reading of the weight in kilograms. When calibrations are made, a temperature calibration is also included for each cell, or, properly, for each strain-gauge/bridge combination, since some cells have two bridges in one unit. Finally, the local value for  $g$ , the gravitational acceleration must be allowed for, since the value, in general, will be different from that at the site of the calibration. The sum of these corrections is usually about 1-2 kg.

In trials at one commercial plant, the measurements were repeated in various ways. First, the set of cylinders to be weighed was

measured with the normal apparatus, but using a hydraulic lift cylinder between the facility lifting hook and the LCBS. The purpose of this was to cushion the load cell from any possible shock during the lifting process. However, this cylinder was quite cumbersome to use, and there was a desire on the part of Agency personnel to eliminate it if at all possible. We learned that results were not significantly improved by the presence of the cylinder, and so it was decided not to use it in the future. A repeat test some months later at a different plant showed that the system had not been damaged by removal of this element. The standard deviation, on cylinders weighing almost 3000 kg, was  $\pm 0.5$ kg, and the average bias (the difference between the corrected readings from the LCBS and the facility declared values) was - 0.14 kg. Measurements were repeated with the flexures removed from the system, which resulted in a somewhat larger bias and standard deviation in the values measured ( $\pm 0.63$  kg and - 0.78 kg).

At the second plant, (with fewer measurements) no significant difference was found between the cases with and without flexures. Finally, measurements were repeated with the system suspended from a fork-lift truck, to see if it is feasible to use the LCBS in an area where there is no easily available overhead crane. The results were less good here, for the few cylinders attempted by this method, with an average bias of -1.5kg. For most cases, however, this accuracy would still be acceptable. At other facilities where the LCBS has been tested the biases were larger, between 1.25 and 1.5kg. In one case, the cylinders were stored outdoors and the measurements were made in a bright desert sun. The fear here, is that the temperature corrections may not have been adequate, because the measuring thermometer could give the temperature only of the surface of the load cell, which may not have been the same as the temperature of the strain gauge within. Shielding the equipment from direct sunlight may alleviate this problem.

#### The Gas-phase UF<sub>6</sub> Enrichment Monitor

This instrument has been developed for the IAEA under the Australian Support Programme and from the inception of the development work, computer control of the measurement process was envisaged. The control programme has undergone continual improvement during the development and testing phases, with input from both safeguards development staff and safeguards inspectors who have been given opportunities to try out the instrument.

The response of inspectors to this development has been very favourable. The programme runs on an HP-85 desk-top computer equipped with a 16K memory module and a ROM drawer containing the following ROMs: INPUT/OUTPUT, PLOTTER/PRINTER, MATRIX, ADVANCED PROGRAM. General purpose and RS-232 interfaces are also needed. The programme itself tests for the presence of the correct ROMs and an error message is given if they are not present. A

schematic diagram of the instrument is shown in Figure 2 and a photograph in Figure 3. The general purpose interface is used to sense the status of the vacuum gauge and liquid nitrogen trap. The RS-232 interface is the link with the multi-channel analyser.

The principle of operation of the instrument is extremely simple and can be understood by reference to Figure 2. A small sample of the quantity of  $UF_6$  whose enrichment is required, is taken from a storage cylinder or from any other appropriate sampling position. It has been assumed that samples are taken under vacuum conditions by liquid nitrogen cryopumping. The minimum sample size required to achieve the design accuracy of 1% for a  $UF_6$  sample, 3% enriched in U-235, is 3g. A larger sample is acceptable but does not improve the accuracy.

The  $UF_6$  sample bottle is connected to the enrichment monitor and the  $UF_6$  is allowed to evaporate into the previously evacuated volume of the measurement chamber. The total uranium content of the  $UF_6$  gas within the chamber is determined by measuring the attenuation of the 59.5 keV gamma rays from an Am-241 source placed beneath the chamber. The gamma rays must traverse the volume of the chamber to reach a 5" x 1" NaI detector placed directly above the chamber. For gamma-ray energies of about 60 keV, the absorption process of gamma rays in matter is almost entirely by the photo-electric process. Furthermore, the absorption cross-section has an effective  $Z^4$  dependence. Thus the uranium content of a  $UF_6$  sample is more than three orders of magnitude more effective in the absorption process than the total fluorine content. In addition, even large quantities of an HF impurity in the  $UF_6$  sample have no effect on the measurement process.

The principal gamma ray from the decay of U-235 at 185.7 keV has a 54% branch probability. It is sufficiently separated in energy from other interfering gamma ray lines (such as those from the decay of U-238 and its daughter products) that a measurement of its intensity with a NaI detector provides an accurate estimate of the total U-235 content of the  $UF_6$  sample. Thus from a comparison of the two count rates in the same NaI detector, the enrichment of the  $UF_6$  sample can be determined. Of course the system must be calibrated with standards previously measured by mass spectrometric methods.

With time there is a slow build-up of a uranium deposit (principally  $UF_4$ ) on the wall of the measurement chamber and thus it is necessary to subtract this background for each measurement. The magnitude of the background to be subtracted is determined by repeating the NaI count with the measurement chamber evacuated. This second measurement also serves to provide the unattenuated gamma ray count rate in the NaI detector from the Am-241 source.

The complete instrument comprises:

- A vacuum system for evacuating the counting chamber, admitting the sample, and recovering the  $UF_6$  after measurement.
- A 5" diameter x 1" thick sodium iodide detector and photomultiplier, with the necessary amplifier and HV supply, connected to a multi-channel analyser.
- The necessary pressure and vacuum gauges to control the sample handling.
- An interfaced HP-85 computer which guides the operator through the entire operating sequence via an interactive routine and performs the necessary data reduction.

The measuring sequence is as follows:

- Evacuate the measuring chamber
- Attach the  $UF_6$  sample cylinder to the admittance valve
- Evaporate the  $UF_6$  sample into the measurement chamber
- Perform the measurement (1000 sec counting time)
- Re-condense the  $UF_6$  into the sample cylinder by cooling it with liquid nitrogen
- Evacuate the system in preparation for the next sample.

Provision is made in the interactive HP-85 programme for:

- checking the performance of the instrument by measuring a standard,
- regular checking of background level,
- informing the operator of the correct valve operating sequence and proper pressure values through the measurement cycle,
- automatic monitoring of the liquid nitrogen level,
- help to the operator in case of any malfunction during operation,
- calculation of the measured sample enrichment and its uncertainty,
- Calculation of the difference between measured and declared enrichment and an indication of whether the difference is significant.

The results obtained with the system have been very satisfactory. Of the 62 measurements made at one location, 34 were within one SD of the plant value, 18 were between one and two SD, 10 were between two and three SD. For 40 measurements on product samples the mean RSD was  $\pm 0.94\%$ ; for 12 measurements on a single (very dirty) feed sample the mean RSD was  $\pm 2.23\%$ ; for 9 measurements on tail samples (counting time 1000 sec) the mean RSD was  $\pm 2.65\%$ ; and for two measurements on one tail sample (counting time 4000 sec) the mean RSD was  $\pm 1.28\%$ .

With a second instrument at a different location, the mean standard deviation for thirteen 400 second counts was  $\pm 2.36\%$ , relative, in

measuring product material of about 3.3% enrichment. This instrument has been installed in a facility for an extended field test under plant conditions, for a period of six months.

To measure a single sample using the instrument with 1000 second counting time takes about 30 minutes. Taking account of start-up, standardization and background checks, about 10 samples per day could be measured. The HP-85 computer programme is designed to lead the operator through the correct operating sequence, and requires a correct response from the operator before proceeding at each stage; if an incorrect response is given, the computer will not progress to the next stage.

#### Conclusion

With the developments described in this paper, the NDA method based on gamma spectrometric enrichment measurements on UF<sub>6</sub> product cylinders can now be used by safeguards inspectors with a greater degree of confidence. The load-cell based weighing system provides a transportable, non-intrusive means for independent verification of cylinder weights. About 15 minutes set-up time is required, after which about 10-15 weighings per hour may be performed, without removing the cylinders from their storage location. Error level is of the order of 1 kg or better.

The gas-phase enrichment monitor allows the determination of the enrichment of a sample of UF<sub>6</sub> with an error level of  $\pm 1\%$ ; after initial set-up and standardization, which takes about one hour, the sample handling and measurement time is about 30 minutes per sample.

Thus with the availability of these instruments it has now become possible to establish with a high degree of reliability and independence the amount of U-235 contained in UF<sub>6</sub> storage cylinders. Some residual UF<sub>6</sub> gas usually exists above the solid material and it is relatively simple to take a sample of this gas. Some facility operators collect samples of gas in this way and use them for verifying mass spectrometrically the enrichment of their UF<sub>6</sub>. Therefore the cylinders chosen as "standards" for the gamma spectrometric NDA method could be sampled and an enrichment analysis immediately carried out by using the Gas-Phase Enrichment Monitor. When this procedure combined with the use of the load cell for independent weighing has been approved for routine use and clearance has been obtained from the operators of the facilities, the degree of reliability of cylinder NDA measurements will be considerably improved.

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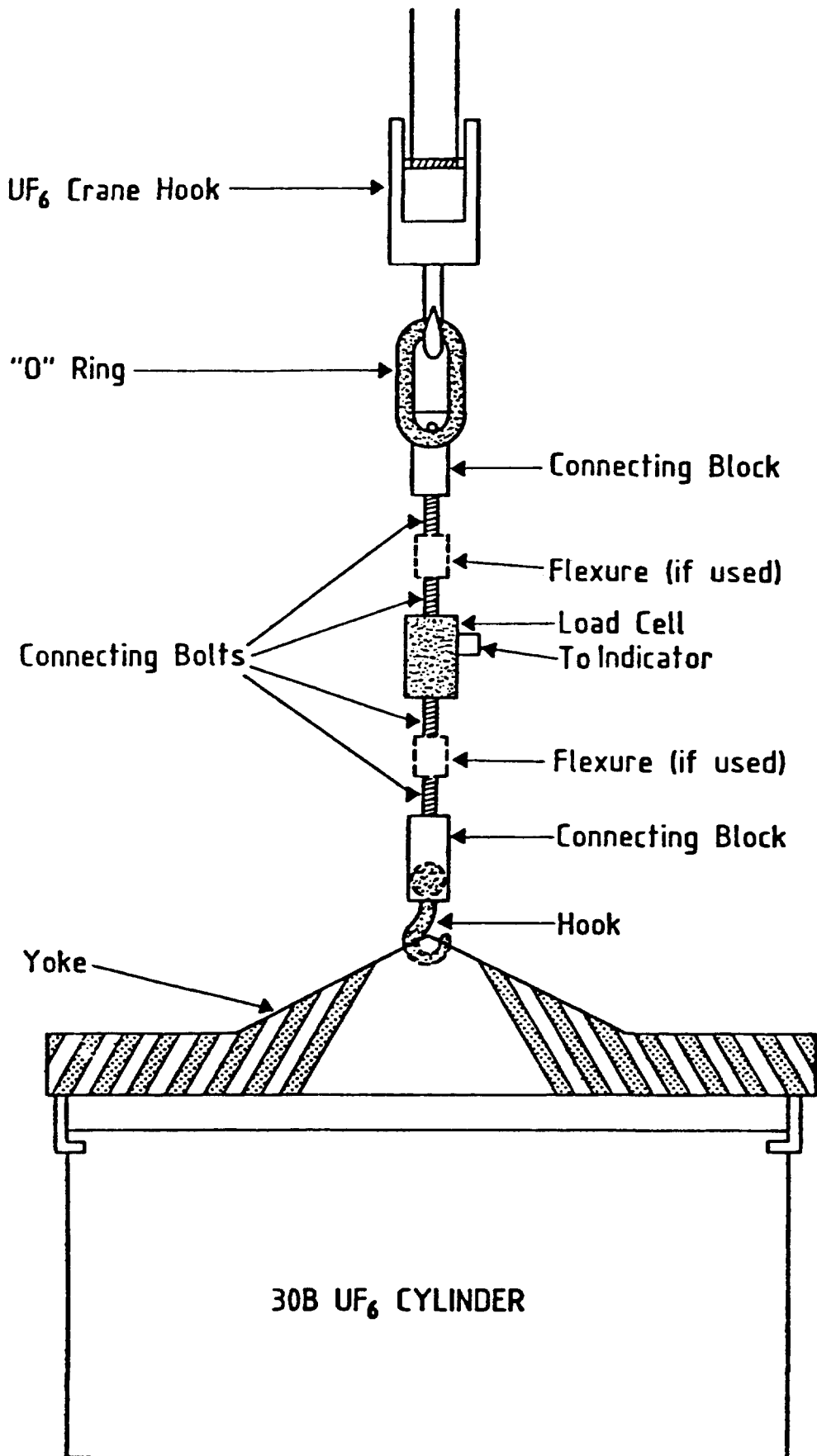


Fig. 1 - Side View of Assembled LCBS

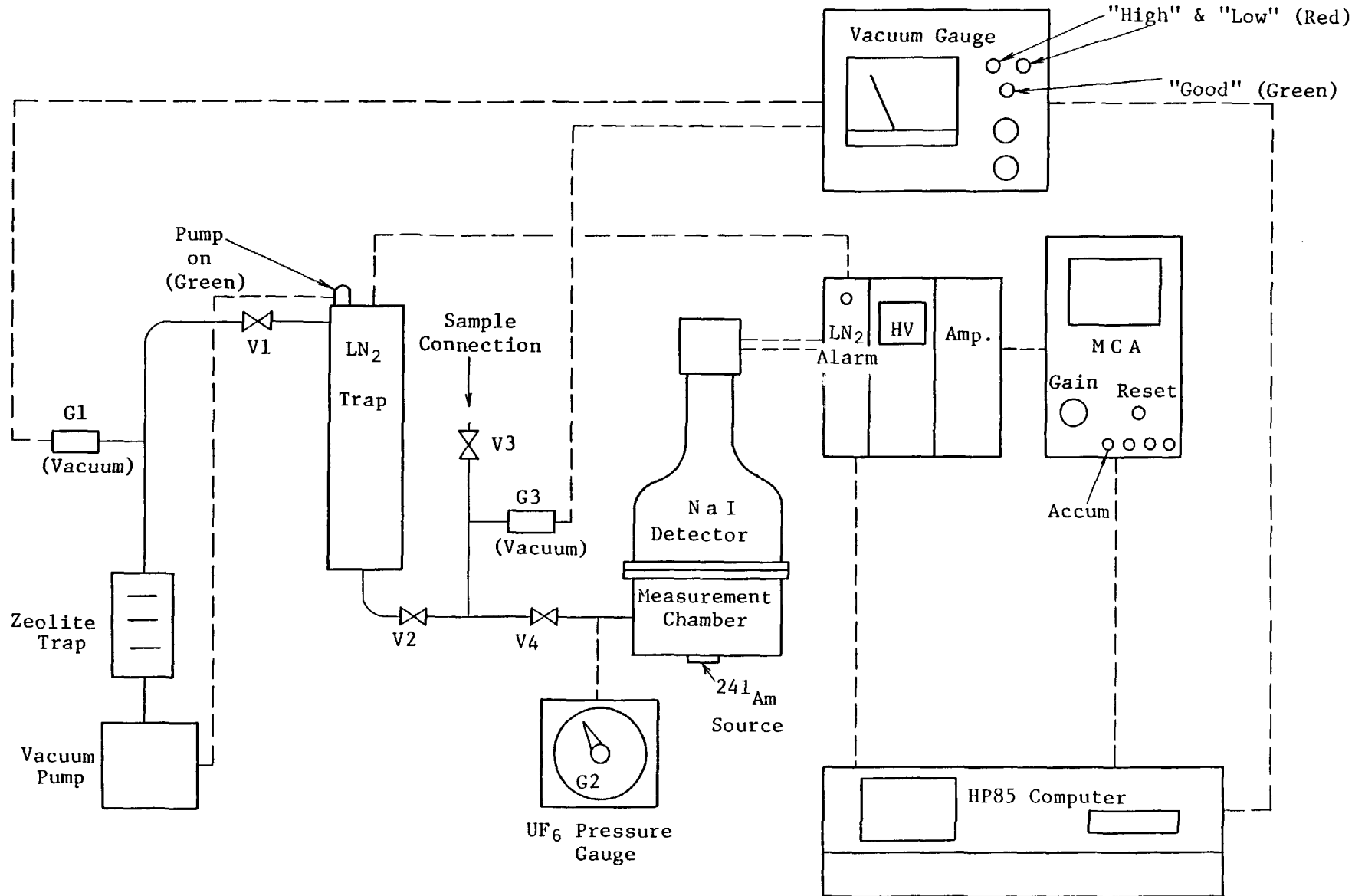


Fig. 2 - Gas-Phase  $UF_6$  Enrichment Monitor



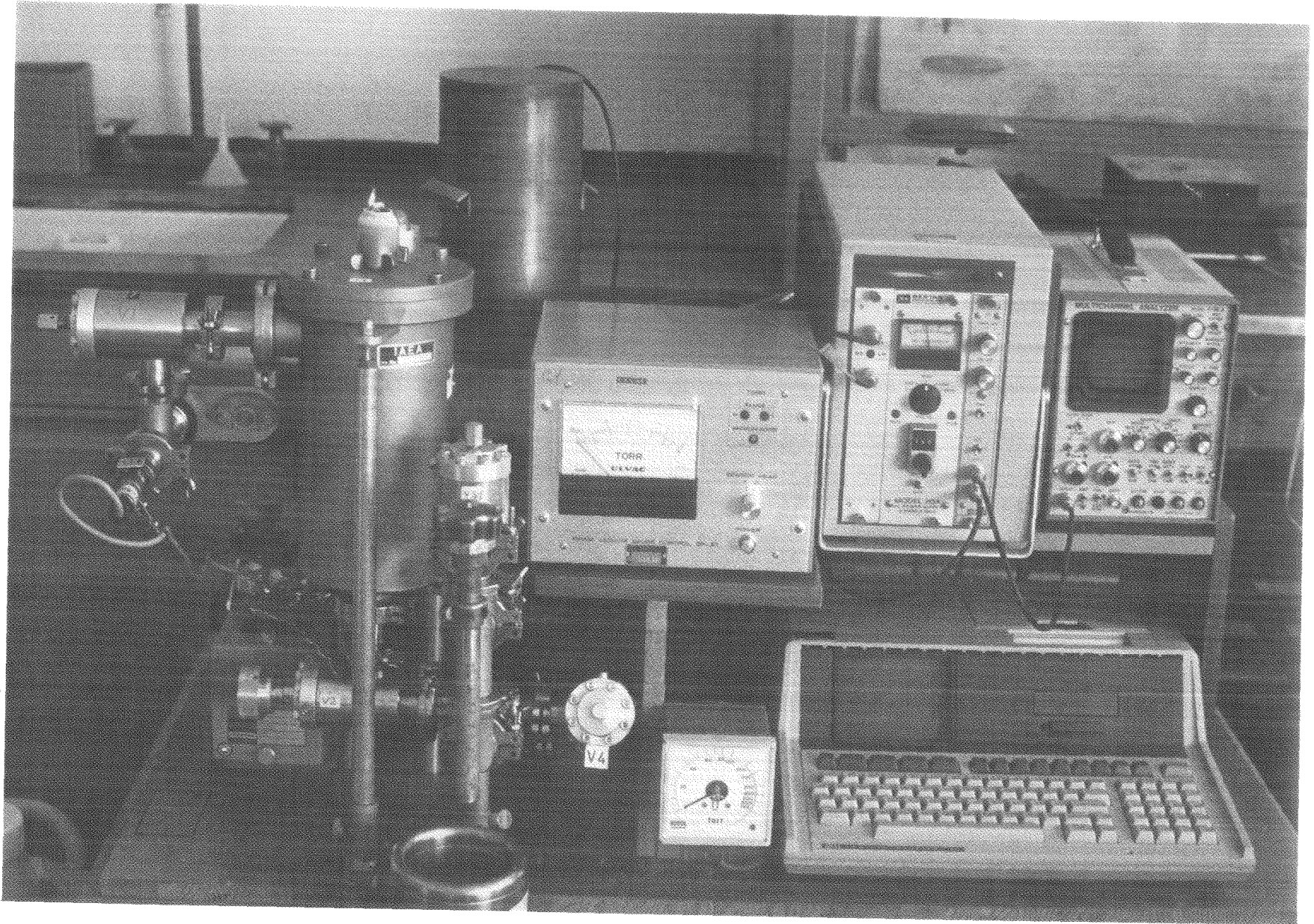


Fig. 3 — Photograph of Gas Phase UF<sub>6</sub> Enrichment Monitor



# CONSISTENCY CHECK APPROACH TO QUANTITATIVE VERIFICATION

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## ABSTRACT

The mean for some given quality characteristic of a population of  $N$  items is to be estimated by selecting  $n$  items at random and measuring these items with measurement device #1, a nondestructive assay (NDA) device. This NDA device is then calibrated by randomly selecting a subset  $k < n$  of the  $n$  items and measuring them by measurement device #2, a destructive assay method. The problem is to estimate the population mean using the  $(n+k)$  measurement results and to find the variance of the estimate.

As a specific example, the population may be  $N$  sintered fuel pellets of nominally the same enrichment. The quality characteristic may be the enrichment (% U-235). Measurement device #1 may be a SAM-2, and measurement device #2 a mass spectrometer.

## Problem Statement

This study was prompted by a question posed in connection with the inspection of a nuclear facility, and the title of the paper is inspired by this question. The question dealt with the consistency of NDA results within a given population of items to be inspected, and the inferences that can be made based on such consistency. The problem was then broadened to include the aspects dealt with in this paper.

## Mathematical Model

Let

$y_i$  = measured response for item  $i$  with measurement device #1

$x_i$  = measurement response for item  $i$  with measurement device #2

$\mu_i$  = true response for item  $i$

$$x_i = \mu_i + \delta + \epsilon_i \quad (1)$$

$$y_i = \beta \mu_i + \eta_i \quad (2)$$

$\delta$  = systematic error with measurement device #2

$\epsilon_i$  = random error with measurement device #2

$\eta_i$  = random error with measurement device #1

$\beta$  = unknown calibration parameter

It may be bothersome to some readers to include a systematic error for the destructive method but not for the nondestructive device. The  $\delta$  quantity relates the DA measurement to the true quantity, while any systematic error in the NDA is accounted for by the  $\beta$  parameter, the calibration constant. Of course, errors in estimating  $\beta$  will result in a systematic error in the NDA measurement after the calibration is completed, but a priori in the model, a separate identification of a systematic error in  $y_i$  is unnecessary.

Assume that

$\delta$  is  $N(0, \sigma_\delta^2)$

$\epsilon_i$  is NID  $(0, \sigma_\epsilon^2)$

$\eta_i$  is NID  $(0, \sigma_\eta^2)$

$\mu$  = population mean =  $\frac{1}{N} \sum_{i=1}^N \mu_i$

$\sigma_\mu^2$  = population variance  
 $= \frac{1}{N} \sum_{i=1}^N (\mu_i - \mu)^2 / (N-1)$

Thus,  $E(\mu_i) = \mu$

$\text{var } \mu_i = (N-1) \sigma_\mu^2 / N$

$\text{cov}(\mu_i, \mu_j) = -\sigma_\mu^2 / N$

The above definition of  $\mu$  and  $\sigma_\mu^2$  is made to treat the case when the population of items to be characterized is of finite size. For  $N$  large relative to  $n$ , the model is altered by assuming that  $\mu_i$  is a random variable with mean  $\mu$  and variance  $\sigma_\mu^2$ .

## Parameter Estimation

For simplicity in notation and exposition, it is assumed without loss of generality that the items measured by both devices are numbered 1 to k and those measured only by device #1 are numbered (k+1) to n. Then, the pairs of responses ( $x_i, y_i$ ) for  $i=1, 2, \dots, k$  are used to estimate the parameter  $\beta$ . In this approach,  $\beta$  is estimated by

$$\tilde{\beta} = \bar{y}/\bar{x} = \frac{\sum_{i=1}^k y_i}{\sum_{i=1}^k x_i} \quad (3)$$

There are alternate estimates of  $\beta$  that could be used. In practice, one would want to examine  $y_i/x_i$  for each item to check for outliers. However, for the small value of  $\sigma_\mu$  assumed in this model, and with errors in both variables,  $\tilde{\beta}$  is a reasonable estimator to use.

In finding the weighted estimate of  $\mu$ , first consider the k observations ( $x_i, y_i$ ). In assigning the weight c to each  $x_i$  and (1-c) to each  $y_i$ , the estimate of  $\mu$  based on only these first k observations would be

$$\begin{aligned} \dot{\mu} &= c \frac{\sum_{i=1}^k x_i}{k} + (1-c) \frac{\sum_{i=1}^k y_i}{\tilde{\beta} k} \\ &= c \frac{\sum_{i=1}^k x_i}{k} + (1-c) \frac{\sum_{i=1}^k x_i}{\tilde{\beta} k} \\ &= \frac{\sum_{i=1}^k x_i}{k} \end{aligned} \quad (4)$$

and it is noted that the first k observations with device #1 are used only to estimate  $\beta$  (i.e., for calibration purposes); the estimate of  $\mu$  based on only these k observations depends entirely on the observations made with device #2.

For all n data points;  $x_i$  for  $i=1, 2, \dots, k$  and  $y_i$  for  $i = (k+1), (k+2), \dots, n$ , the weighted estimate of  $\mu$  is

$$\tilde{\mu} = a \frac{\sum_{i=1}^k x_i}{k} + (1-a) \frac{\sum_{i=k+1}^n y_i}{\tilde{\beta}(n-k)} \quad (5)$$

The problem is to choose the parameter a to minimize the variance of  $\tilde{\mu}$ , and to find the variance for that value of a.

### Variance of $\tilde{\mu}$ as Function of a

Equation (5) is rewritten in terms  $\mu_i, \delta, \epsilon_i$ , and  $\eta_i$  using equations (1)-(3). The expression for  $\tilde{\mu}$  reduces to

$$\tilde{\mu} = \frac{a}{k} \left( \sum_{i=1}^k \mu_i + k \delta + \sum_{i=1}^k \epsilon_i \right) \quad (6)$$

$$+ (1-a) \left( \frac{\sum_{i=1}^k \mu_i + k \delta + \sum_{i=1}^k \epsilon_i}{\beta \sum_{i=k+1}^n \mu_i + \sum_{i=k+1}^n \eta_i} \right)$$

$$(n-k) \left( \beta \sum_{i=1}^k \mu_i + \sum_{i=1}^k \eta_i \right)$$

The variance of  $\tilde{\mu}$  is found by Taylor's series expansion. The partial derivatives of  $\tilde{\mu}$  are found with respect to  $\mu_i, \delta, \epsilon_i$ , and  $\eta_i$ . These simplify as follows:

$$\frac{\partial \tilde{\mu}}{\partial \mu_i} = a/k; \quad i=1, 2, \dots, k \quad (7)$$

$$\frac{\partial \tilde{\mu}}{\partial \mu_i} = (1-a)/(n-k); \quad i = (k+1), (k+2), \dots, n \quad (8)$$

$$\frac{\partial \tilde{\mu}}{\partial \delta} = 1 \quad (9)$$

$$\frac{\partial \tilde{\mu}}{\partial \epsilon_i} = 1/k; \quad i=1, 2, \dots, k \quad (10)$$

$$\frac{\partial \tilde{\mu}}{\partial \eta_i} = -\frac{(1-a)}{k\beta}; \quad i=1, 2, \dots, k \quad (11)$$

$$\frac{\partial \tilde{\mu}}{\partial \eta_i} = \frac{(1-a)}{(n-k)\beta}; \quad i=(k+1), (k+2), \dots, n \quad (12)$$

Then by Taylor's series approximation:

$$\begin{aligned} \text{var } \tilde{\mu} &\approx \frac{(N-1) \sigma_\mu^2 \left[ \frac{a^2}{k} + \frac{(1-a)^2}{n-k} \right] - 2\sigma_\mu^2}{N} \\ &\bullet \left[ \frac{k(k-1)a^2}{2k^2} + \frac{(n-k)(n-k-1)(1-a)^2}{2(n-k)^2} + \frac{k(n-k)a(1-a)}{k(n-k)} \right] \\ &+ \sigma_\delta^2 + \frac{\sigma_\epsilon^2}{k} + \sigma_\eta^2 \left[ \frac{(1-a)^2}{k\beta^2} + \frac{(1-a)^2}{(n-k)\beta^2} \right] \end{aligned} \quad (13)$$

The coefficient on  $\sigma_\mu^2$  reduces to

$$\frac{(a^2 n + k - 2ak) - 1/N}{k(n-k)} \quad (14)$$

Therefore,

$$\begin{aligned} \text{var } \tilde{\mu} &\approx \left[ \frac{(a^2 n + k - 2ak) - 1/N}{k(n-k)} \right] \sigma_\mu^2 \\ &+ \sigma_\delta^2 + \frac{\sigma_\epsilon^2}{k} + \frac{n(1-a)^2 \sigma_\eta^2}{k(n-k)\beta^2} \end{aligned} \quad (15)$$

### Optimum Value for a

To find that value of a which minimizes var  $\tilde{\mu}$

$$\frac{\partial \text{var } \tilde{\mu}}{\partial a} = \frac{2(an-k) \sigma_\mu^2}{k(n-k)} - \frac{2n(1-a) \sigma_\eta^2}{k(n-k)\beta^2} = 0$$

one DA measurement. Let the constraint be  $E = 100$ , so that from (23),

$$10k+n = 100$$

The optimum value for  $k$  is found from (24), where, from (25)

$$\begin{aligned} D &= [2025(0.2025+1) + 8100]10^{-8} \\ &= 10535.06 \times 10^{-8} \end{aligned}$$

Ignoring the  $10^{-8}$  factor that appears in both the numerator and denominator,

$$\begin{aligned} k &= \frac{100}{10} \frac{(105350.6 - 4050 \sqrt{105350.6})}{(105350.6 - (40.50)^2)} \\ &= 8.9 \end{aligned}$$

Choose  $k=9$  and  $n=100-90 = 10$

10 measurements would be performed by NDA, and 9 of the 10 by DA. From (21),

$$a = 0.9832$$

and from (22),

$$\begin{aligned} \text{var } \tilde{\mu} &= 10^{-8} [225 + 225 + 8100 (0.1092)] \\ &= 1335 \times 10^{-8} \\ \sigma_{\tilde{\mu}} &= 0.0037\% \text{ U-235} \end{aligned}$$

Continuing with this example, suppose the measurement error standard deviation for the NDA were smaller, say  $\sigma_{\eta} = 180$  rather than 400. Then  $r=1$  rather than 0.45.

$$\begin{aligned} D &= [2025(2) + 8100] \times 10^{-8} \\ &= 12150 \times 10^{-8} \\ k &= \frac{100}{10} \frac{(121500 - 90 \sqrt{121500})}{(121500 - 8100)} = 7.9 \end{aligned}$$

Choose  $k=8$  and  $n=100-80 = 20$

20 measurements would be performed by NDA and 8 of the 20 by DA. From (21),

$$a = 0.70$$

and from (22),

$$\begin{aligned} \text{var } \tilde{\mu} &\approx 10^{-8} [225 + 253 + 8100 (0.0875)] \\ &= 1187 \times 10^{-8} \\ \sigma_{\tilde{\mu}} &= 0.0034\% \text{ U-235} \end{aligned}$$

As the examples indicate, and as is intuitively clear, NDA measurements play a major role whenever the process variance,  $\sigma_{\mu}^2$ , is large in a relative sense, for then it is important to measure many items by some means. For small  $\sigma_{\mu}^2$ , on the

other hand, it is not necessary to measure many items, but those that are measured should be measured by a precise method, i.e., by DA. Other generalizing statements may also be made, e.g., the cost parameter,  $c_0$  is important, as is the ratio of measurement error standard deviations,  $\sigma_{\eta}/\sigma_{\epsilon}$ .

which yields the solution

$$a = \frac{k\beta^2 \sigma_\mu^2 + n\sigma_\eta^2}{n(\beta^2 \sigma_\mu^2 + \sigma_\eta^2)} \quad (16)$$

Note that as  $\sigma_\eta^2 \rightarrow 0$ , i.e., as the measurement error for device #1 (NDA) becomes small relative to the sampling error, then  $a \rightarrow k/n$ , and all  $n$  observations are equally weighted regardless of which measurement device is used. At the other extreme, as  $\sigma_\eta^2$  gets large in a relative sense, (or as  $\sigma_\mu^2$  gets small)  $a \rightarrow 1$ , and only the items measured with device #2 (DA) carry any weight. Keep in mind that this result is from the perspective of estimating  $\mu$ ; as a check for consistency on an attributes basis, the device #1 results would still have value.

#### Minimum Variance of $\tilde{\mu}$

Upon inserting (16) into (15), the variance of  $\tilde{\mu}$  corresponding to the optimum choice of  $a$  is

$$\text{var } \tilde{\mu} \cong \sigma_\delta^2 + \sigma_\epsilon^2/k + A\sigma_\mu^2 + B\sigma_\eta^2 \quad (17)$$

where

$$A = \frac{(k\beta^4 \sigma_\mu^4 + 2k\beta^2 \sigma_\mu^2 \sigma_\eta^2 + n\sigma_\eta^4) - 1/N}{kn(\beta^2 \sigma_\mu^2 + \sigma_\eta^2)^2} \quad (18)$$

$$\text{and } B = \frac{(n-k)\beta^2 \sigma_\mu^4}{kn(\beta^2 \sigma_\mu^2 + \sigma_\eta^2)^2} \quad (19)$$

Note that  $A$ , the coefficient on  $\sigma_\mu^2$ , reduces to  $(1/n-1/N)$  when  $\sigma_\eta^2 = 0$  and hence  $a = k/n$ . In this event, the sampling error is zero when  $n=N$ , i.e., when all items in the population are measured.

The results simplify somewhat if we define the ratio

$$r = \beta\sigma_\mu / \sigma_\eta \quad (20)$$

Then, the optimum choice for  $a$ , from (16) and (20) is

$$a = \frac{kr^2 + n}{nr^2 + n} \quad (21)$$

and the variance of  $\tilde{\mu}$  corresponding to this choice of  $a$  is, from (17)-(20),

$$\text{var } \tilde{\mu} \cong \sigma_\delta^2 + \sigma_\epsilon^2/k + \sigma_\mu^2 \left[ \frac{kr^4 + (k+n)r^2 + n}{kn(r^2+1)^2} - 1/N \right] \quad (22)$$

#### Example

Suppose  $k = 3$   
 $n = 20$

$$\sigma_\mu = 0.0090\% \text{ U-235}$$

$$\sigma_\delta = 0.0015\% \text{ U-235}$$

$$\sigma_\epsilon = 0.0045\% \text{ U-235}$$

$$\beta = 20,000$$

$$\sigma_\eta = 400$$

assume  $N$  is large.

Then, from (20),  $r = 180/400 = 0.45$

From (21),  $a = 0.8569$

From (22),

$$\text{var } \tilde{\mu} \cong 10^{-8} [225+675+8100(0.2856)]$$

$$\cong 3213 \times 10^{-8}$$

$$\sigma_{\tilde{\mu}} = 0.0057\% \text{ U-235}$$

#### Optimum Allocation of Measurements

For given total measurement effort, we find  $k$  and  $n$  to minimize the variance of  $\tilde{\mu}$ .

Let  $c_0$  = number of measurements that can be performed with measurement device #1 (NDA) for same cost as one measurement with device #2 (DA).

$E$  = Total fixed effort: equivalent number of measurements that can be performed by NDA.

The constraining relationship is

$$E = c_0 k + n$$

For given  $E$  and  $c_0$ , the values of  $k$  and  $n$  that minimize the  $\text{var } \tilde{\mu}$  given by (22) are found by replacing  $n$  in (22) by  $(E-c_0k)$ , partially differentiating  $\text{var } \tilde{\mu}$  with respect to  $k$ , equating to zero, and solving for  $k$ . The solution is

$$k = \frac{E}{c_0} \frac{(c_0 D - r \sigma_\mu \sqrt{c_0 D})}{(c_0 D - r^2 \sigma_\mu^2)} \quad (24)$$

$$\text{where } D = \sigma_\epsilon^2(r^2+1) + \sigma_\mu^2 \quad (25)$$

If, in a given application,  $k$  is found to equal or exceed  $E/c_0$ , the optimum solution is  $k=E/c_0=n$ , i.e., all measurements would be performed by measurement device #2, destructive analysis.

#### Example

The previous example is continued with the additional information that  $c_0=10$ , i.e., 10 NDA measurements can be performed for the same cost as

# RECOMMENDED DETAILED GUIDELINES FOR STATES' SYSTEMS OF ACCOUNTING FOR AND CONTROL OF NUCLEAR MATERIALS

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## ABSTRACT

In 1980 the IAEA published IAEA/SG/INF/2 "Guidelines for States' Systems of Accounting for and Control of Nuclear Material", to assist Member States in establishing, maintaining and reviewing their SSACs. These guidelines were quite general and did not address the details of the specific elements of SSACs. It was brought to the attention of the Agency that more detailed guidance was needed in the implementation of certain elements of SSACs. To provide the additional detail it was decided to prepare detailed guidelines for SSACs at various types of facilities which Member States could use, if they wished, to establish and maintain their SSACs. This paper briefly describes the background and discusses the basis on which the Agency decided that such detailed guidance would be useful. The paper then describes the approach taken to produce recommended detailed SSAC guidelines in terms of seven principal elements of an SSAC at the facility level and the detailed guidelines under development for each element. The paper addresses progress to date by presenting a discussion of the principal elements and their detailed guidelines. The paper concludes by discussing briefly future plans and possible areas for future efforts.

## INTRODUCTION

For some years in the past, Agency experience indicated a need for guidance to Member States in establishing and maintaining their State Systems of Accounting for and Control of Nuclear Material (SSACs). In 1980 the Agency published IAEA/SG/INF/2 "Guidelines for States' Systems of Accounting for and Control of Nuclear Material." This document presented general guidelines for the requirements and functions of SSACs, including the elements of the system and the performance expected at both the State Authority and the facility levels.

While the INF/2 document was considered a practical help to Member States, the guidelines were quite general and did not address the details of the specific elements of SSACs. Agency experience still indicated that there was a need for improvement in several elements of SSAC implementation. This situation was addressed in the Safeguards Implementation Reports for both 1981 and 1982. Agency inspectors often found a lack of information regarding measurement precision and accuracy, either because the information was not available or was of doubtful quality. They found that records were not kept in a uniform logical manner so as to allow retrieval of data. Often it was found that it was almost impossible to trace reports to substantiating records and sometimes the records did not substantiate the reports. They also found that often no physical inventory was actually taken or if taken it was inaccurate, incomplete or both so that the value of the associated material balance was questionable. It was apparent that there was further need for additional detailed guidance for certain elements of SSACs.

To develop this additional guidance, tasks were initiated under the U.S. Support Programme in Vienna and at the Brookhaven National Laboratory in the United States. It should be clearly understood that the purpose of these tasks is to provide technical details of an effective nuclear materials accounting and control system which Member States may use, if they wish, to establish and maintain their SSACs. If a system is developed using the detailed guidelines it would be expected that it would be compatible with any requirements for such systems included in subsidiary arrangements or facility attachments. However, there is no intention that the reports produced would add to, subtract from, or amend in any way the rights and obligations defined for the IAEA and the States in documents INFCIRC/66/Rev. 2, INFCIRC/153 (corrected) or any specific

safeguards agreement concluded with the IAEA pursuant to these documents.

#### SEVEN PRINCIPAL ELEMENTS OF AN SSAC

To develop a structure for presenting these additional guidelines, the requirements and functional elements presented in INF/2 and the requirements for safeguards systems of INFCIRC/153 were considered. Table 1 shows the 12 sections in INF/2 which identify the areas in which a State Authority should establish requirements. Table 2 shows the sections in INF/2 which identify the functional elements that should be established by the State Authority. Table 3 shows the measures called for by INFCIRC/153 in an SSAC. Some of these measures would be functions of the State Authority, some would be functions of a facility operator, while some could be functions of both. The mix of functions would, of course, be expected to vary from State to State.

If one were to attempt to structure detailed SSAC guidelines using either the State Authority level requirement areas identified in Table 1 or the facility level functional areas identified in Table 2 there would be considerable duplication among the guidelines, because the same subjects are addressed in several of the sections. There are interrelationships among the measures of INFCIRC/153, the State Authority requirements areas and the facility level functional areas. For example, in Table 2, measurements are called for in Subsections 3.3.3, 3.3.4, 3.3.5, 3.3.6 and 3.3.7. These subsections, in turn, refer in Table 1, to Subsections 2.4.3, 2.4.5, 2.4.6 and 2.4.9, which also call for measurements of nuclear material. Further INFCIRC/153 paragraph 32(a) calls for a measurement system for nuclear material. Similarly other elements of the system are called for in various sections and in INFCIRC/153.

Table 1

IAEA/SG/INF/2  
Areas for State Authority Requirements

- 2.4.1 Starting point, termination and exemption of accounting and control
- 2.4.2 Categorization of nuclear material
- 2.4.3 Material balance areas
- 2.4.4 Records and reports system
- 2.4.5 Measurement system
- 2.4.6 Nuclear material flow
- 2.4.7 Physical inventory taking
- 2.4.8 Shipper/receiver differences
- 2.4.9 Material balance closing
- 2.4.10 Measurement control
- 2.4.11 Application of containment and surveillance measures
- 2.4.12 International transfers of nuclear material

Table 2

IAEA/SG/INF/2  
Functional Areas at the Facility Level

- 3.3.1 Starting point, termination and exemption of accounting and control
- 3.3.2 Categorization of nuclear material
- 3.3.3 Material balance areas and key measurement points
- 3.3.4 Facility Accounting and control system
- 3.3.5 Flow measurements
- 3.3.6 Physical inventory taking
- 3.3.7 Measurement uncertainty in the material balance
- 3.3.8 Containment and surveillance
- 3.3.9 Miscellaneous controls

Table 3

INFCIRC/153 (Corrected) Paragraph 32  
SSAC Structure

- (a) A measurement system for nuclear material;
- (b) The evaluation of precision and accuracy of measurements and the estimation of measurement uncertainty;
- (c) Procedures for identifying, reviewing and evaluating shipper/receiver differences;
- (d) Procedures for taking a physical inventory;
- (e) Procedures for evaluation of unmeasured inventory and unmeasured losses;
- (f) A system of records and reports showing material balance area nuclear material inventory and inventory changes;
- (g) Procedures to assure that the accounting procedures and arrangements are being operated correctly; and
- (h) Procedures for submission of required reports to the Agency.

Seven categories of information are addressed throughout the different subsections. These seven categories can be defined and described so that they include all of the facility level functional elements of Section 3.3, the State Authority requirement areas of Section 2.4, and the measures called for by INFCIRC/153. By considering these seven categories as principal elements of an SSAC, guidelines can then be prepared for these principal elements that would cover all of the subject matter for the facility level functional elements and the relationship of those functional elements to the State Authority requirements and the measures called for in INFCIRC/153. Table 4 lists these seven principal elements of an SSAC. To assure coverage of the various requirements and functions that have been identified for SSACs the scope of the guidelines for each of these seven principal elements was developed as follows:

Table 4

Seven Principal Elements of an SSAC

1. Nuclear Material Measurements
2. Measurement Quality
3. Records and Reports
4. Physical Inventory Taking
5. Material Balance Closing
6. Containment and Surveillance
7. Organization and Management

(1) Nuclear Material Measurements

Guidelines at the facility level would address selection of material balance areas and selection of key measurement points for material flows and for inventory. Procedures for determining quantities transferred into or out of MBAs would be addressed, including measurement of nuclear material received, produced, shipped, lost or otherwise removed from inventory as well as for quantities on inventory.

At the State Authority level, guidelines would address the standards and criteria the Authority should consider in establishing requirements for facility measurement systems.

(2) Measurement Quality

Guidelines at the facility level would address the procedures for determining and controlling the precision and accuracy of the nuclear material measurements. Data collection and treatment procedures for determining the uncertainty of measurements also would be addressed.

At the State Authority level, guidelines would address the standards and criteria the Authority should consider in establishing requirements for measurement uncertainties and for measurement control.

(3) Records and Reports

Guidelines at the facility level would address the procedures and data that would need to be maintained to accurately record and report inventories and inventory changes, including adjustments for physical inventories, biases, shipper-receiver differences, mistakes, etc. Included would be discussion of both accounting and operating data that would be needed to obtain accurate statements of quantity, form and disposition of nuclear materials. Data and procedures needed for striking periodic material balances would be covered. Guidelines for batch identification and batch data handling also would be included. Data flow and control to assure accurate and timely reports as required by the State Authority and the Agency also would be covered.

At the State Authority level, guidelines would address the records and report system that would need to be maintained by the Authority in accordance with Agency agreement requirements, as well as criteria and standards the Authority should consider for facility records and reports systems.

(4) Physical Inventory Taking

Guidelines at the facility level would address the procedures that the facility operator would need to establish and follow to assure timely and accurate physical inventories. Such procedures would include pre-inventory procedures such as scheduling, preparing inventory work plans, assigning personnel, preparing working papers and inventory lists, etc. Also addressed would be the inventory taking procedures, including listing and checking procedures, item identity and check-off procedures as well as general provisions and criteria, such as the basis for accepting prior measurements, the extent of process clean-out required, and criteria for inventory stratification and categorization.

At the State Authority level guidelines would address the criteria, standards and schedules the Authority should consider in establishing requirements for facility physical inventory taking to assure compliance with Agency agreement requirements and to assure accurate inventories.

(5) Material Balance Closing

While one might consider Material Balance Closing a part of PIT it is, in fact, the PIT that is a part of closing a material balance. Guidelines at the facility level would address the data elements needed to close a material balance. The results of the PIT are, of course, a major element. There are other, equally necessary, data elements which would be addressed in the guidelines. These include book inventory data, accounting data and operating data on which to base statistical evaluations of the material balance, i.e. the MUF and  $\sigma$ MUF.

At the State Authority level, guidelines would address the criteria and rationale that the Authority should consider in establishing the requirements for a Material Balance Closing, including criteria for MUF and  $\sigma$ MUF.

(6) Containment and Surveillance

Guidelines at the facility level would address techniques of containment and surveillance that a facility operator might use to supplement and simplify accounting and control measures.

At the State Authority level guidelines would address those measures that the

Authority might employ to provide assurance of the integrity of the facility system. Measures that the Authority may require of the facility operator or permit the facility to use instead of some accounting measures also would be addressed.

#### (7) Organization and Management

Guidelines at the facility level would address the organization of the accounting and control function in terms of assignment of responsibilities to persons and units of the facility to assure controls and internal checks for the system. Also included would be guidelines for the establishment of the starting point for accounting and control of the material, criteria for categorization of the material and procedures for the termination of and exemption from controls, including those for waste and discards. The starting point, termination, etc. would, of course, be in accordance with State Authority established criteria which would be addressed in guidelines at the Authority level. Guidelines for system management at both the facility and State Authority level would address the procedures that could be used to assure proper operation of the system. This would include audits, and other controls that might be used to provide confirmatory information or to indicate anomalies in the system.

#### GUIDELINE DEVELOPMENT

With these elements defined we then went to Agency staff, in particular Operations people, to identify those elements for which they thought Member States needed more guidance. We also sought to establish priorities with regard to facility types and to find out the extent of detail and depth of coverage that was believed to be needed.

As a result of these many conversations, priority has been given to the development of guidelines at the facility level for the first five principal elements, i.e. Measurements, Measurement Quality, Records and Reports, PIT, and Material Balance Closing. The other two elements, Containment and Surveillance and Organization and Management may be covered at a later time but are not now included in the task schedule. After numerous iterations, including various outlines and drafts reviewed by Agency staff, we concluded that the guidelines should be presented in a separate document for each facility type. Each such report would address the principal elements of the SSAC as applicable to the specific facility type with examples to show the application to that facility type. Also, it was concluded that the first facility type to be covered would be low-enriched uranium conversion and fuel fabrication facilities.

Again after many iterations, including review of outlines and drafts by Agency staff and by consultants, two reports have been published in the series. One is STR-150 entitled "Detailed Description of an SSAC at the Facility Level for a Low Enriched Uranium Conversion and Fuel Fabrication Facility." Table 5 shows an abbreviated Table of Contents for STR-150. In an effort to make the report easier to understand and use, more than 60 tables and figures are included. The second report is STR-159 entitled "Detailed Description of an SSAC at the Facility Level for Research Reactors" with contents similar to STR-150.

Many have commented that the 200 plus page document of STR-150 is too long. However, many who expect to use the report as they visit facilities in Member States have commented that this is the kind of detail that is needed. We expect that the LEU facility report will be one of the longest. The MOX facility report will probably be as long. Others such as the report for research reactors will be shorter.

Table 5

#### STR-150 Table of Contents

##### Part 1. Introduction

- 1.1 General
- 1.2 Principal Elements of an SSAC
- 1.3 Purpose and Scope of this Document
- 1.4 The Model Plants

##### Part 2. Nuclear Material Measurements

- 2.1 Introduction
- 2.2 Material Balance Areas
- 2.3 Key Measurement Points
- 2.4 Materials and Material Types
- 2.5 Measurement Methods
- 2.6 Documentation
- 2.7 Example

##### Part 3. Measurement Quality

- 3.1 Introduction
- 3.2 System Qualification
- 3.3 Standards and Calibration
- 3.4 Sampling
- 3.5 Control Programme
- 3.6 Administration
- 3.7 Example



Part 4. Records and Reports

- 4.1 Introduction
- 4.2 Accounting Records
- 4.3 Operating Records
- 4.4 Accounting Reports
- 4.5 Data Handling
- 4.6 Examples of Records and Reports

Part 5. Physical Inventory Taking

- 5.1 Introduction
- 5.2 Inventory Measurements
- 5.3 Inventory Organization and Planning
- 5.4 Conduct of Inventory
- 5.5 Post-Inventory Activities
- 5.6 Example

Part 6. Material Balance Closing

- 6.1 Introduction
- 6.2 The Material Balance Equation
- 6.3 Evaluation of MUF
- 6.4 Other Parameters Affecting MUF
- 6.5 Example of Material Balance Closing and MUF Evaluation

Appendix A. The Reference Facility

As we approached completion of the first report we again went to Agency staff to decide which facility type to address next. We had established some priorities in our earlier discussions but we wanted to check again. The results of these conversations is the list of facility types shown in Table 6, in the order of their priority for report preparation. Tentative schedules call for all or most of these to be completed by June of 1986.

Table 6

SSAC Guideline Reports Planned

Research Reactors  
Critical Facilities  
Power Reactors  
Research Facilities  
State Level Systems  
Reprocessing Facilities  
MOX Facilities  
Enrichment Facilities

# THE SAFEGUARDS ADVANTAGES OF NAIVE BIAS CORRECTION

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## Abstract

In a measurement control situation where periodic measurements of known quantities (standards) are interspersed among production measurements of unknowns, a simple method of handling biases is to base the correction to an unknown solely on the observed error of the closest (in time) standard measurement. The statistical properties of this "naive" bias correction method are derived, and its utility for safeguards objectives is discussed. It is pointed out that no statistical evaluation of the data is needed to make such a correction, that the statistical properties of the corrected estimates are independent of the nature of systematic errors or biases, and that in most cases the correction results in considerably improved estimates of the total amounts of material in strata containing many items. By contrast, this correction method should generally not be applied if one is interested only in the contents of individual items. Because facility operational and administrative needs do not necessarily coincide with material accountancy goals, it is argued that it is reasonable that measurement and measurement control data should be treated differently for the different purposes. Thus, the bias correction should not be applied to book values, but accumulated by strata and used for the correction of inventory differences.

## 1. Introduction

Current policy dictates that the significance of an inventory difference (I.D.) is to be evaluated in relation to its limit of error (LEID). LEIDs which can be calculated on the basis of error propagation or similar techniques are preferable to those which are historically derived because (1) they encourage good measurement performance, (2) they provide insight that can be used to improve I.D. behavior, and (3) they can be adapted to changing throughput or inventory conditions.

The problem of realistically modelling the factors which contribute to I.D. behavior mathematically is, however, a difficult one. The difficulties fall into two categories: first, there are effects for which measurement control

data is unavailable, and, second, where such data are available, large statistical uncertainties may attend estimates of key parameters derived from them. While not attempting to minimize the former category of problems, this paper will concentrate on the latter.

Considerable space in this journal and elsewhere (1-12) has been devoted to the problem of handling long-term errors, especially by Jaech, who uses a model involving random errors, long-term systematic errors, and biases. Within the context of the model, and the scope of the questions asked, the issue has been thoroughly studied. This paper will make somewhat different assumptions and will allow a different type of solution.

An additional point of view may be justified on the following grounds:

- Many errors show a complex time-dependence of means (systematic effects) which is not easily accommodated by the pure systematic/pure random error model. If one makes bias corrections on the basis of an inappropriate model, the results are unpredictable.
- If bias corrections are not made, there remains the task of characterizing statistically the complex systematic effects for the purposes of error propagation. In theory, analysis-of-variance techniques are applicable, but in practice a long period of stable data is necessary before such methods can be used with confidence and accuracy. It is often debatable whether the effects should be characterized in statistical terms at all.
- When the pure random/pure systematic model is applicable, it is very difficult to make good estimates of long-term systematic error variances. One estimate that has been suggested (Ref. 1 p. 89, for example) is the squared difference between a mean value and an actual value of a series of measurement-control runs of a standard:  $\sigma^2 = (\bar{x} - \mu)^2$ .

Jaech<sup>(2)</sup> points out that this is about the best that can be done, but calculates that generally one has about a 68% chance of being off by at least a factor of 2, and a 15 - 20% chance of being off by a factor of 16. If these types of estimates are used to compute the dominant terms in the LEID, the LE-LEID comparison will have questionable utility. There is also the question of the significance of long term (multi period) I.D.'s, which must take into account correlations between long term systematic errors; these are even harder to estimate.

The method proposed here addresses all the above problems for material strata for which representative measurement control data exist by making continual bias corrections. The corrections should in many circumstances make for better I.D.s and more accurately estimated L.E.I.D.s; however, the corrections may vary well, depending on circumstances, worsen estimates of the contents of individual items. It is therefore recommended that the corrections be accumulated for the purposes of safeguards only.

Interestingly, when conditions are stable, and the pure random/pure systematic error model is valid, the method described here produces the same error correction term on a stratum basis as some of those previously suggested.<sup>1,2</sup>

## 2. The Naive Bias Correction Method; Statistical Properties

In the situation that we will describe, a sequence of measurements are made on (for example) the product items of a facility over a period of time. At fixed intervals, the same measurement process produces a value for a known standard. The measurement process does not "know" which are the standards, so that systematic errors affecting the item measurements also affect the standard measurements. The time interval between standards measurements will be called T. The bias correction method calls for a bias correction term to be calculated for each standard measurement, applicable to all items measured in the time interval of length T centered at the point at which the standard was measured; thus if the difference between the measured and known value of the standard is  $\delta$ , the bias correction is the amount of nuclear material corresponding to a systematic error of  $\delta$  applied throughout the interval. The total bias correction is the (algebraic) sum of the corrections for all intervals (all standards measurements). A bias-corrected inventory difference can be computed from the bias-corrected strata values.

The error structure that will be assumed here is not completely general, but complex enough to illustrate the kinds of effects that will clearly occur if this type of correction is calculated. Three types of errors are assumed: random, short-term systematic (which is really

very-short-term systematic), and long-term systematic. Random errors are independent from measurement to measurement. Long-term systematic errors are constant during the intervals of time of length T; otherwise their behavior is arbitrary, and need not even be characterized statistically. The concept thus incorporates biases and systematic effects whose correlation periods are long compared with the sampling interval. In order to illustrate the consequence of systematic effects of duration short compared with T, the interval is divided into J periods, each of which has a systematic error associated with it. This is the short-term systematic error. An expression for the variance of the corrected estimate of the total amount of material in all items of a stratum is derived. The result is, of course, that the long-term systematic effects tend to be reduced in both the corrected estimate and the variance of the corrected estimate, while the components due to short-term and random errors are enhanced.

The mathematical model is

$$X_{ijk} = \mu_{ijk} + \epsilon_{ijk} + \zeta_{ij} + \eta_i$$

$$(i = 1 \dots I, j = 1 \dots J, k = 1 \dots K)$$

$$Y_i = \mu_0 + \epsilon_{i**} + \zeta_{iJ/2} + \eta_i$$

$$(i = 1 \dots I)$$

$$\mu_1 = \mu_0 + \epsilon_0$$

$$C_i = M(Y_i - \mu_1)$$

$$X = \sum_{ijk} X_{ijk} - \sum_i C_i$$

where

i is an index denoting the sequence of standards measurements or time intervals; there are I total intervals.

j is an index indicating time periods within the time interval; each interval is subdivided into J periods.

k denotes the sequence of item measurements within each period; K items are measured in each period.

$X_{ijk}$  and  $\mu_{ijk}$  are the measured and actual values associated with measurement i, j, k.

$\epsilon_{ijk}$  is the random error (the associated standard deviation will be  $\sigma_r$ )

$\zeta_{ij}$  is the short-term systematic error (with standard deviation  $\sigma_s$ )

$\eta_j$  is the long-term systematic error (with standard deviation  $\sigma_{sys}$ )

$Y_i$  is the observed value of the standard at measurement i

$\mu_0$  and  $\mu_1$  are the actual and "known" values for the standard;  $\epsilon_0$  is the associated error

$\epsilon_{i**}$  is the random error associated with the standard measurement

$\zeta_{iJ/2}$  is the short-term systematic error of the standard measurement; it is identical to  $\zeta_{ij}$  where  $j$  is at the midpoint of the time interval.

$C_i$  is the bias correction for interval  $i$ .

$X$  is the bias-corrected estimate of the total amount of material  $\sum_{ijk} \mu_{ijk}$ .

$M = JK$  is the number of items measured in an interval

This situation is illustrated in Figure 1.

The correction factor  $C_i$  is given by:

$$C_i = JK (\epsilon_{i**} + \zeta_{iJ/2} + \eta_i - \epsilon_0)$$

Therefore the bias-corrected estimate for the  $i^{\text{th}}$  interval is

$$\begin{aligned} (\sum_{jk} X_{ijk}) - C_i &= \sum_{jk} (\mu_{ijk} + \epsilon_{ijk}) + K \sum_j \epsilon_{ij} + M \eta_i \\ &\quad - JK(\epsilon_{i**} + \zeta_{iJ/2} + \eta_i - \epsilon_0) \\ &= \sum_{jk} (\mu_{ijk} + \epsilon_{ijk}) - JK\epsilon_{i**} + JK\epsilon_0 \\ &\quad + K (\sum \zeta_{ij} - (J-1)\zeta_{iJ/2}) \\ &\quad \quad \quad J \neq J/2 \end{aligned}$$

The variance associated with the error  $(X - \sum_{ijk} \mu_{ijk})$  is therefore

$$\begin{aligned} N\sigma_r^2 + (JK)^2 I\sigma_r^2 + N^2\sigma_o^2 + IK^2\{ (J-1)\sigma_s^2 + (J-1)^2\sigma_s^2 \} \\ = \sigma_r^2 (N + M^2 I) + N^2\sigma_o^2 + IJK^2(J-1)\sigma_s^2 \\ = (M^2 I + N) \sigma_r^2 + N^2\sigma_o^2 + ((J-1)/J)(M^2 I)\sigma_s^2 \end{aligned}$$

This variance, of course, contains no systematic errors except for that associated with the uncertainty in the reference standard; all others have been corrected for. On the other hand, the random error, which usually enters with a factor of  $N$ , is now increased by a term  $M^2 I \sigma_r^2 = (N^2/I)\sigma_r^2$ . The question of whether or not the bias-corrected variance will be larger or smaller than the uncorrected version essentially rests on whether this term is larger or smaller than the contribution of the longer-term systematic errors. To a crude approximation, contributions from these

errors take the form  $N^2\sigma_{sys}^2$ , so that if  $(\sigma_r^2/I\sigma_{sys}^2)$  is less than one (which will usually be the case for  $I$  greater than, say, 10) the bias-corrected variance will be smaller.

The term involving short-term systematic error behaves like a random error when  $J$  is large compared to 1. When  $J = 1$ , it disappears, having become a long-term error. When  $J = 2$  or 3, there is very little difference between the corrected and uncorrected contribution.

Having argued that the corrected estimate is generally better than the uncorrected one for estimating the contents of large numbers of items ( $N$  and  $I$  large), and also that the short-term errors involved in its variance are easier to characterize and estimate, it should also be noted that the original error variance for an individual item is

$$\sigma_r^2 + \sigma_s^2 + \sigma_{sys}^2$$

whereas the corrected value will usually have an error

$$2\sigma_r^2 + 2\sigma_s^2 + \sigma_o^2.$$

Therefore, unless the systematic error is known to be considerably larger than the random and short-term errors, the correction may very well result in a poorer estimate of the contents of individual items.

Finally, it is worth noting that Jaech<sup>1,2</sup>, for example, uses a bias correction which is calculated on the basis of the average factor  $(\sum_i Y_i)/I - \mu_1$  which is applied to all items. This is made under the assumption  $\zeta_{ij} = 0$  and  $\eta_i = \eta$  (no short-term systematic error and constant long-term error). Under these assumptions, both methods yield the same total correction factor  $\sum_i C_i = (N/I)(\sum_i \epsilon_{i**}) + N\eta - N\epsilon_0$ .

### 3. An Example

An example will be given to illustrate and amplify the preceding theory. It will deal with a hypothetical set of drums of uranyl nitrate solution produced over a period of three months; the U-235 content is determined by net weight, a concentration determination, and mass spec. value for isotopics. We are concerned with the errors in the isotopics. Relevant values are:

Net weight per item: 60 kg.

Number of items.: 100 in first 45 days, 200 in second 45 days

Number of standards determinations: 40 (approximately one every 2 days)

Concentration: 50 grams U per kg. of solution

A hypothetical set of measurement control results are presented in Figure 2. A simple T-test would show that the mean value is insignificantly different from zero, indicating that a bias correction should not be made, even though clear systematic effects are present; perhaps by chance, or perhaps because of some common cause, the systematic error has shifted at about the same time as the output changed. To ignore the systematic effects would be incorrect. Another way to proceed would be to go ahead and make the bias correction on the entire three months of data regardless of the insignificant result: all results would therefore be shifted upward by .0033% to compensate for the negative mean. This would of course also be incorrect, because the higher production in the second half of the time period means that in fact the biases shown will actually over estimate the total U-235 content. The same effect would also occur if the concentration increased or the weight per item increased over the time period.

The naive bias correction would be computed as follows. There would be 5 items corresponding to each of the 20 standards determinations in the first half of the period. The observed discrepancy for the first measurement is about -0.00045; this corresponds to

$$(5) (-0.00045) (60) (50) = -6.8 \text{ gr. U-235}$$

A similar calculation would be made for each of the 40 points, and the results summed. In this case the total correction is + .11 kg. U-235. This number would then be subtracted from the observed total value of U-235 in all items, along with corrections for the other measurement techniques. To determine the error variance associated with the bias-corrected isotopic measurement process, one must estimate the random error. This could be done by a series of determinations on a standard performed at one point in time in the usual manner, or one could, for example, use the standard deviation of the first twenty points of Figure 2, under the assumption that conditions were stable in that interval. The latter procedure has the advantage that it would include the short-term systematic errors. This procedure yields a value of about .04%, or  $(0.0004)(60)(50) = 1.2$  grams U-235. The total variance for the two time periods must (and can) be computed separately since M is different in the two periods;

$$\begin{aligned} V &= (M_1^2 I_1) \sigma_r^2 + (M_2^2 I_2) \sigma_r^2 \\ &= (5)^2 (20) (1.2)^2 + (10)^2 (20) (1.2)^2 \\ &= 3600 \text{ gr}^2 \end{aligned}$$

This procedure is fairly simple and rests on a reasonably good data-base. If, on the other hand, no bias correction were to be made, one is left with the problem of characterizing the complex long-term systematic errors represented by

the data shown in Figure 2 for the purposes of error-propagation.

#### 4. Concluding Remarks:

The objectives of measurement systems from the point of view of the facility operator and from the point of view of safeguards overlap, but do not coincide. By and large, the requirements of process or quality control are to obtain the best values for individual items. (In addition, the values a facility enters on its books may be subject to administrative or contractual constraints). The safeguards authority, on the other hand, has no interest whatsoever in the contents of individual items, except insofar as they can be used in the overall material balance. Given the choice between a random error and a somewhat smaller systematic error, the operator will choose the systematic, and the safeguarder will choose the random.

The operator desires a measurement control system which will help insure that every measurement is being made with reasonable accuracy; the safeguards wants a system which functions superbly on the average.

Given the differing objectives, there is no reason why the operator and safeguarder should use measurement and measurement control data in the same way; seldom is a particular statistical procedure optimal for two different purposes. The bias correction method suggested here is intended to fulfill safeguards functions: to provide more accurate estimates of material balance quantities which have accurately computable error variances. It is not suggested that it be used to correct book values. It would be useful, however, to carry computed values of the correction factors in the computer files so that bias-corrected versions of material balance quantities could be calculated.

The advantages of this type of bias correction procedure are the following:

1. It eliminates systematic effects of duration long compared with the time between standards measurements, thereby considerably reducing error variances for most material balance quantities.
2. Variances of the bias-corrected quantities can be computed on the basis of estimates of random or very-short-term systematic errors, which are much easier to make than characterizations of longer-term systematic effects. Except for the error due to standard uncertainty, these variances can be added together from one material balance period to another.
3. The bias correction computation can be carried out trivially by computer. No data need be analyzed, and no tests of

significance performed to carry out the correction.

4. The procedure automatically takes into account changes in material characteristics or plant throughput with time.

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Figure 1

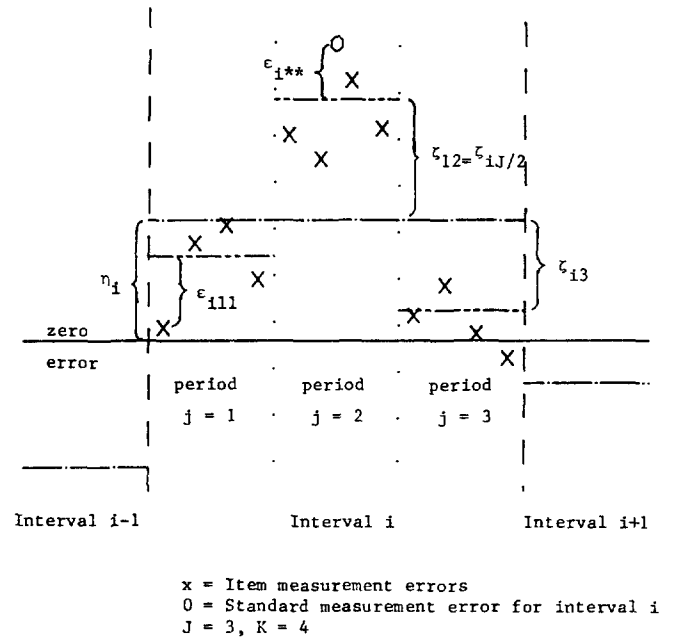
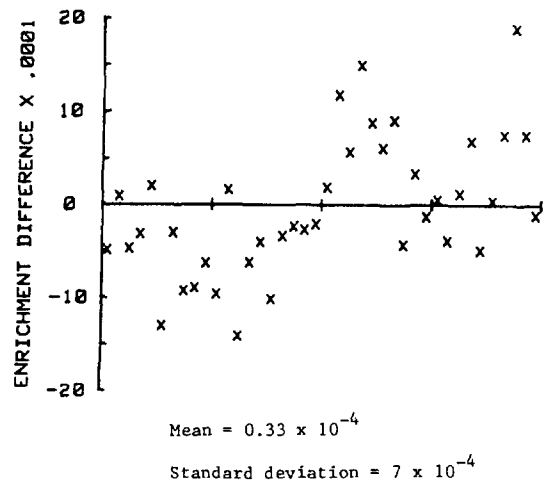


Figure 2



# INTERNAL STANDARDS APPLIED TO ISOTOPIC RATIO MEASUREMENTS OF URANIUM AND PLUTONIUM\*

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## ABSTRACT

Application of internal standards to measurement of isotopic ratios in analyses of uranium and plutonium has yielded improvement in precision and accuracy of about a factor of 5 for single filament analyses of nanogram-sized samples. These results emphasize the desirability of producing the necessary enriched isotopes in sufficient quantity and purity to foster the widespread application of the technique. It will have especially significant impact on nuclear safeguards programs.

## INTRODUCTION

Internal standards (also known as double spikes) have been in use for some years to improve isotopic ratio measurements in thermal ionization mass spectrometry. First suggested by Dietz, et al.,<sup>1</sup> the technique involves spiking the sample with a mixture of two isotopes whose ratio is known to a very high degree of accuracy. By comparing the measured value of this ratio to the known, a bias correction can be calculated that is applicable to the specific conditions of the individual analysis. The principal variation in bias, and one that is impossible to control with a high degree of certainty, is sample-filament chemistry. For application of an average bias correction, the traditional way of correcting for what is simplistically termed "mass discrimination," the same segment of the isotopic fractionation curve must be used in the collection of data for all samples. In practice, it is difficult to do this, and it is this problem that limits precision in pulse counting systems to about  $\pm 0.5\%$ .

Because the use of an internal standard allows one to correct for the individual sample-filament chemistry of a given analysis, great improvement in accuracy and precision is realized with its application. Dodson has investigated the theoretical basis of the technique.<sup>2,3</sup> Among the elements to have been analyzed in this way are U,<sup>4,5</sup> Pb, Ca, Ba, and Mo;<sup>6</sup> the internal standard technique has been applied to strontium for some time in

geological applications; in this case, the natural  $^{86}\text{Sr}/^{88}\text{Sr}$  ratio, neither isotope of which has a radiogenic precursor, serves as a built-in internal standard in strontium-rubidium age-dating measurements. Internal standards for a given element are applicable to any sample of the same element irrespective of its source. Techniques to reduce the effects of fractionation, such as overcoating the sample with a barrier layer,<sup>7,8</sup> are also applicable: use of internal standards will improve precision and accuracy beyond what is otherwise available regardless of other mass spectrometric details.

One important application of this technique will be in the area of safeguards. By combining it with resin-bead sampling,<sup>9</sup> it will be possible to approach precisions and accuracies of  $\pm 0.1\%$  for 1-3 ng samples of uranium and plutonium. Analyzing such small quantities is very important for assaying dissolver solutions of spent fuel.

This paper reports a larger body of results for uranium than have heretofore been available and presents convincing evidence for the power of the technique; the first results for use of a plutonium internal standard are also described.

## EXPERIMENTAL

The mass spectrometer used is a two-stage,  $90^\circ$  magnetic sector instrument with an abundance sensitivity of over  $10^6$ .<sup>10</sup> It is equipped with a pulse counting detection system for analysis of small samples. The resin bead sample loading technique was used to provide 1-3 ng of uranium or plutonium.<sup>9</sup>

It should be stressed that the applicability of internal standards is independent of instrumental and sample-loading details. We obtained improvement in precision and accuracy of about a factor of 5 and would expect similar improvements for any mass spectrometer-sample loading system; Callis has reported precisions of better than 0.01% using a commercially available mass spectrometer with a triple filament source.<sup>11</sup> His work required much larger samples than that reported here; our work is directed toward assaying highly radioactive solutions and analysis of small samples where this level of quality has only been

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approached in the past by analyzing much larger samples. We are presently engaged in a collaborative project with the International Atomic Energy Agency, the Center of Nuclear Studies at Saclay, France, and the French reprocessing plant at LaHague. The purpose of this study is to evaluate internal standards for uranium and plutonium under field conditions; samples from LaHague will be prepared from scratch in duplicate, and the results obtained through use of the internal standard will be compared to those obtained through conventional techniques.

An iterative procedure was used to obtain the required bias correction for each run of each sample; a general solution was derived that is applicable to any spike for any element, and the necessary calculations incorporated into existing computer programs.<sup>12</sup> Thus, since an analysis is ordinarily comprised of 10 runs, 10 individual bias corrections were applied to each sample, one for each run.

Two uranium double spikes were synthesized from enriched  $^{233}\text{U}$  and  $^{236}\text{U}$ . Two spikes were necessary because insufficient high purity ( $> 99\%$ )  $^{236}\text{U}$  is available for isotope dilution work. The compositions of the two spikes are summarized in Table 1. Note that the low purity spike is nearly 5%  $^{235}\text{U}$ , which necessitates such a large correction to the 235 mass position that it is not possible to refine  $^{235}\text{U}/^{238}\text{U}$  ratio measurements using it. Hence, the low purity spike was used in isotope dilution analyses, where the high  $^{235}\text{U}$  content did not affect the results, and the high purity to improve  $^{235}\text{U}/^{238}\text{U}$  ratio measurements. For these latter, a sample-spike ratio of about 10/1 was used to reduce the correction to the 235 mass position.

These spikes were isotopically calibrated by measuring the abundances of all isotopes in the pure double spikes. NBS-500 is a certified uranium isotopic standard with a  $^{235}\text{U}/^{238}\text{U}$  ratio of 0.9997. This material was then used as an internal standard to determine very accurately the  $^{233}\text{U}/^{236}\text{U}$  ratio in the double spikes. This known ratio was then used to establish the  $^{235}\text{U}/^{238}\text{U}$  ratio in the spikes. The concentration of the low purity standard was determined by using it to assay a gravimetrically known uranium sample.

The plutonium internal standard was synthesized by mixing high purity  $^{242}\text{Pu}$  and  $^{244}\text{Pu}$ ; the details are given in Table 2. The  $^{239}\text{Pu}/^{240}\text{Pu}$  ratio of a certified isotopic standard, NBS-947, was used as an internal standard in the manner already described for uranium to establish the isotopic composition of the mixture. NBS-949, certified as 99.99% Pu, was used to determine the concentration.

Like high purity  $^{236}\text{U}$ ,  $^{244}\text{Pu}$  is in extremely short supply, precluding its routine use in isotope dilution analyses. Only the minimum spike necessary was added to the individual sample solutions for these two internal standards. Adding the spike to the sample on the filament failed to yield good results, probably due to failure to achieve equilibrium between the two.

## RESULTS AND DISCUSSION

The effect of the high purity internal standard was evaluated by processing data from replicate analyses of two uranium samples in two different ways: by application of our previously used average bias correction per mass and by executing the internal calibration calculations. The results are shown in Table 3. Significant improvement in precision was noted in each case. The  $^{235}\text{U}/^{238}\text{U}$  ratios reported cannot be directly compared to their accepted values because each reflects a contribution from the spike. The average bias correction (0.29% per mass) was determined by calibrating with NBS-500 and refining with routine checks against NBS-010 ( $^{235}\text{U}/^{238}\text{U} = 0.010140$ ). Note the deviation from the average value of the two bias corrections determined using the internal standard calculations. These reflect the averages of the six independent analyses that were made and exemplify the problem. The values for a typical individual analysis ranged from 0.033 to 0.335% per mass for NBS-010. These are "best case" analyses: pure starting materials, reproducible sample loading, etc.; conditions would be substantially worse for typical pay samples.

Using an internal standard, we usually observe the bias correction to vary  $\pm 10$ -20% as filament conditions change within a single analysis. It thus appears that the application of an average bias correction is at best a crude approximation and that the problem addressed by internal standards is indeed the limiting factor in the precision of isotopic ratio measurements.

As an extreme test of the double spike technique, a sample spiked with the low purity internal standard was run to exhaustion, data being accumulated for about five hours. The  $^{238}\text{U}/^{233}\text{U}$  ratio using an average bias correction was  $0.6370 \pm 0.0067$ , a relative standard deviation of over  $\pm 1\%$ . Using the internal standard technique, the result was  $0.6343 \pm 0.0004$ , a relative standard deviation of  $\pm 0.07\%$ . Thus, with data taken under conditions far more adverse than would be expected in a normal analysis, excellent precision was still obtained.

To evaluate the accuracy of the technique, a suite of NBS isotopic standards was analyzed. The  $^{235}\text{U}/^{238}\text{U}$  ratios covered the range of 0.004919 for NBS-005 to 17.349 for NBS 930. Results are given in Table 4. Each standard was analyzed at least six times; no analyses were rejected. Accuracy is indicated by listing the measured  $^{235}\text{U}/^{238}\text{U}$  ratio divided by the theoretical value.

The only instance of the measured ratio deviating from the certified value by more than 0.1% is for NBS-750, where it is 0.17% high. This deviation is at present unexplained; further analyses of NBS-750 produced the same result.

The low purity uranium standard was checked against two of the monthly samples distributed by the SALE program. Six analyses of sample H-66 (consensus value 87.78% U) gave  $87.76 \pm 0.04\%$  U.



Fourteen analyses of the 300 series sample (consensus value 88.08% U) gave  $88.06 \pm 0.07\%$  U. The higher standard deviation for the latter may reflect inhomogeneities reported for this sample. In both cases, precisions were better than 0.1%.

The plutonium internal standard was found to give results comparable to the uranium internal standards: precisions and accuracies (where known) of 0.1% or better.

#### CONCLUSIONS

From the results reported above, it is clear that internal standards provide a powerful technique for improving both precision and accuracy of isotopic ratio measurements. Our routine results are improved by at least a factor of five over those previously obtained.

One caveat: it is absolutely essential to know the isotopic composition of the internal standard to a high degree of accuracy. Errors here will not be reflected by loss of precision but by poorer accuracy: the corrections needed to be applied to isotopes common to both sample and spike will be in error, and this will be directly reflected in the results.

Although the technique is applicable to any element with enough reasonably stable isotopes, one obvious use is in safeguards programs, both domestic and international. Accurate assay of uranium and plutonium is crucial in this context, and the internal standard offers a means of substantial improvement in the measurements with essentially no alteration of current analytical techniques required. Sample preparation and spiking are identical to present practice; only mass spectral data treatment is different.

We are presently making a "double double" spike, one containing both uranium and plutonium, each as an internal standard. It will be applied to analysis of spent fuel dissolver solutions and is included in the evaluation presently underway at LaHague mentioned earlier. Such an approach would be valid for any situation where the uranium/plutonium ratio does not vary over too wide a range. Unfortunately, widespread application of this technique will not be possible until sufficient quantities of high purity  $^{236}\text{U}$  and  $^{244}\text{Pu}$  become available.

To generate  $^{236}\text{U}$  in sufficient purity, it will be necessary to enrich presently available stocks ( $^{236}\text{U} = 89\%$ ) by a single pass through an enriching calutron operated by Oak Ridge National Laboratory.  $^{244}\text{Pu}$  must be made by first generating it in a reactor, separating plutonium from other products, and enriching the desired isotope on a calutron. This is a costly business, but will pay enormous dividends in the improved accuracy and precision to be obtained from very small samples, thus retaining high quality while reducing transportation costs.

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Table 1. Isotopic Composition of Uranium Internal Standards

Isotope	233	234	235	236	238	233/236
High Purity Spike						
233 Component	99.925	0.0317	0.0032	0.0014	0.038	
236 Component	0.0108	0.0008	0.201	99.674	0.113	
Mixture	46.935	0.0155	0.1084	52.864	0.0771	0.88785
Low Purity Spike						
233 Component	99.528	0.185	0.062	0.015	0.209	
236 Component	0.000	0.119	9.25	89.27	1.32	
Mixture	47.899	0.153	4.903	46.250	0.795	1.03566

Table 2. Isotopic Composition of Plutonium Internal Standard

	238	239	240	241	242	244	242/244
High 242	0.0004	0.0048	0.0518	0.0246	99.853	0.0648	
High 244	0.0015	0.0409	0.0863	0.0008	0.911	98.960	
Mix	0.00056	0.02258	0.06813	0.00892	50.0343	49.8655	1.00339

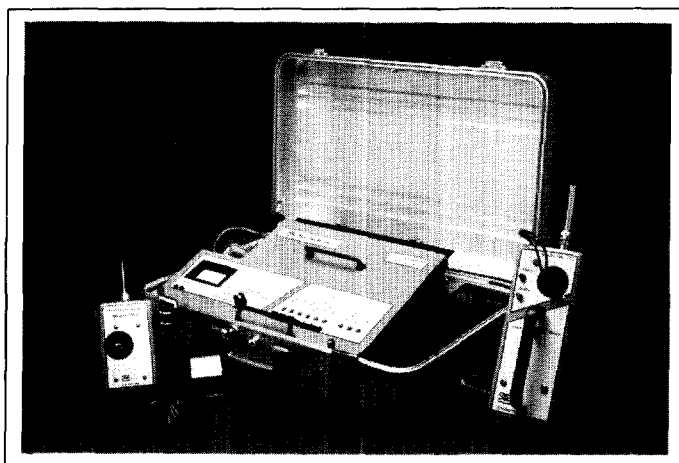
Table 3. Comparison of Results With and Without Use of the Internal Standard

Sample	Analyses	235 U / 238 U		Bias Correction % Mass	235 U / 238 U		Bias Correction % Mass
NBS U010	6	0.012172		0.296	0.012192		0.234
		+0.000043			+0.000009		
SALE H-66	6	0.025770		0.296	0.025702		0.384
		+0.000053			+0.000029		

Table 4. Systems Calibration

NBS Standard	Certified Pct. 235	<u>5/8 Meas.</u> <u>5/8 Theo.</u>	% RSD
005	0.4895	0.9999	0.10
950	0.750	1.0000	0.14
010	1.0037	0.9992	0.11
020	2.038	1.0008	0.14
030	3.046	0.9999	0.11
050	5.010	1.0008	0.03
100	10.190	1.0001	0.05
200	20.013	1.0001	0.08
500	49.696	1.0002	0.07
750	75.357	1.0017	0.04
930	93.336	1.0004	0.08
Avg.		1.0003	0.07

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