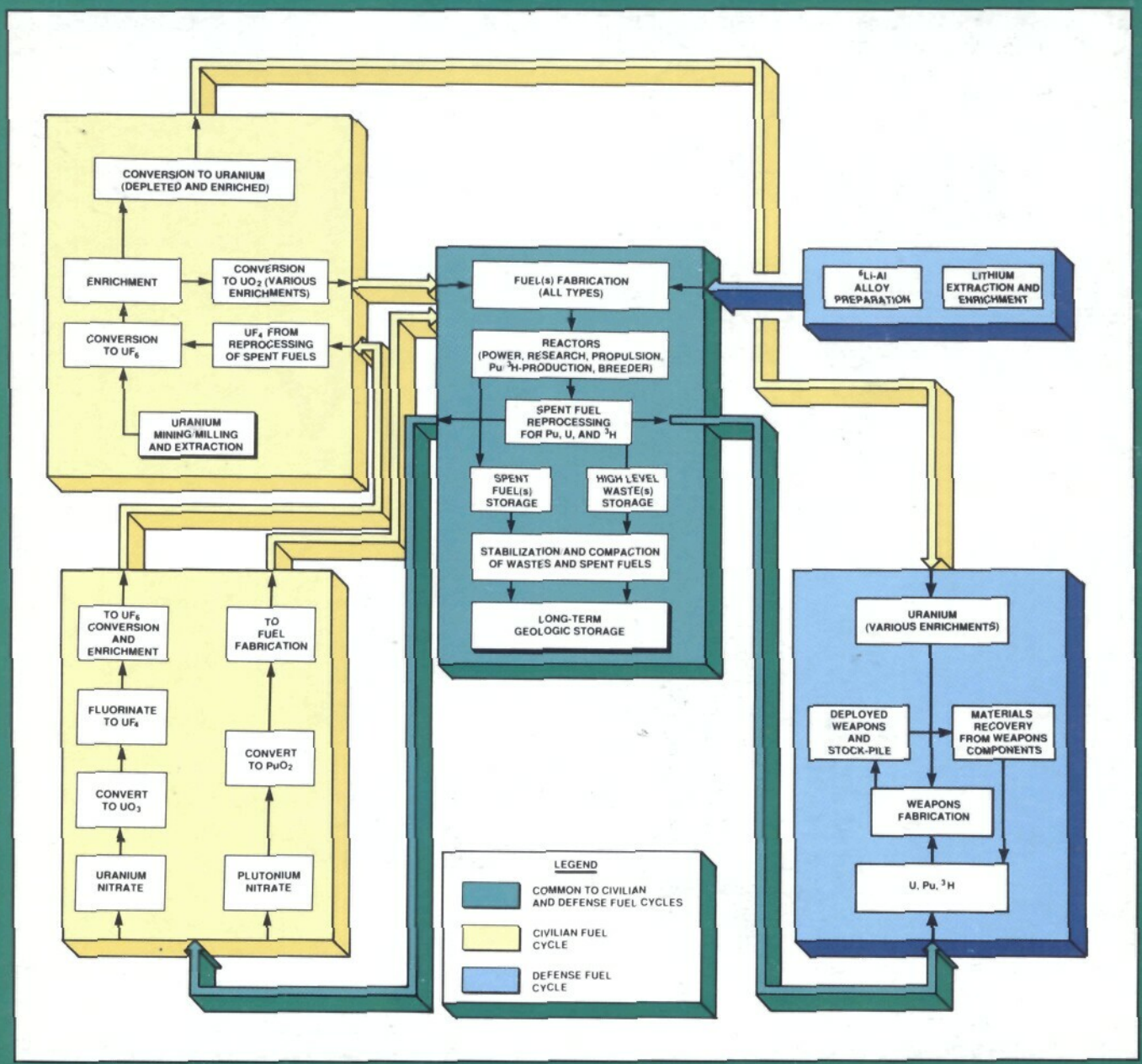
JNMM
JOURNAL OF
THE INSTITUTE OF
NUCLEAR MATERIALS MANAGEMENT

Process Holdup of Special Nuclear Materials



Volume XVI, Number 4 • July 1988

CONTENTS

Volume XVI, Number 4 • July 1988

PAPERS

Selected Results for Measuring Holdup in Oxide Hoods and Transfer Lines at Hanford
R.A. Jones and R.E. Kerns 6

Measurement Campaigns for Holdup Estimation
R.R. Picard 9

The Importance of Understanding Process Holdup — A Department of Energy View
Glenn A. Hammond and Ron L. Hawkins 12

Process Holdup — The Regulatory View
E. William Brach 15

Holdup Related Issues in Safeguarding Nuclear Materials
K.K.S. Pillay 18

An Overview of Holdup Measurement Technology
Martin S. Zucker 21

Total Neutron-Counting Plutonium Inventory Measurement Systems and Their Potential Application to Near-Real-Time Materials Accountancy
I. Driscall, G.H. Fox, C.H. Orr, and K.R. Whitehouse 26

NUMATH: A Nuclear Material Holdup Estimator
Alan B. Krichinsky 30

Literature Survey
K.K.S. Pillay 33

EDITORIALS

Technical Editor's Note 2
INMM Chairman's Message 2
JNMM Comment 3

DEPARTMENTS

Books 37
IAEA Employment Opportunities 42
Literature 42
Equipment, Materials & Services 43
Calendar 44
Advertiser Index 44

On the Cover:
Illustration of the combined nuclear fuel cycle. Design by Sam Pillay, Los Alamos National Laboratory, Safeguards Systems Group; graphic illustration by Teri A. Ortiz and Robert E. Davis, Los Alamos National Laboratory, Graphic Illustrations Group.

JNMM is published four times a year by the Institute of Nuclear Materials Management, Inc., a not-for-profit membership organization with the purpose of advancing and promoting efficient management and safeguards of nuclear materials.

SUBSCRIPTION RATES: Annual (U.S., Canada and Mexico) $100.00, annual (other countries) $135.00; (shipped via air mail printed matter); single copy regular issues (U.S. and other countries) $25.00; single copy of the proceedings of the annual meeting (U.S. and other countries) $65.00. Mail subscription requests to *JNMM*, 60 Revere Drive, Suite 500, Northbrook, Illinois 60062 U.S.A. Make checks payable to INMM.

ADVERTISING, distribution and delivery inquiries should be directed to *JNMM*, 60 Revere Drive, Suite 500, Northbrook, Illinois 60062 U.S.A. or contact Greg Schultz at (312) 480-9573, telex 910-221-5870, telecopy (312) 480-9282. Allow eight weeks for a change of address to be implemented.

ISSN 0893-6188

SOCIETY OF NATIONAL ASSOCIATION PUBLICATIONS

INMM
INSTITUTE OF NUCLEAR MATERIALS MANAGEMENT

TECHNICAL EDITOR'S NOTE

Making Progress Measuring Holdup

We are now publishing four Journal issues a year in addition to the proceedings of the annual meeting. The papers for an annual meeting are selected by a "papers committee" on the basis of abstracts and short summaries submitted well in advance of the meeting. The authors are required to submit final drafts, suitable for offset printing. By making a special effort, our dedicated and competent staff publish the Proceedings within a reasonably short time. The Proceedings papers are not edited, although they number far more than those in the regular Journal issues. Our assumption is that selection for presentation at an annual meeting should ensure that the topics are relevant and that speedy publication is more important than reviewing and editing so many papers.

Most of the papers in the regular issues of the Journal now are taken from the several topical meetings and workshops which the Institute sponsors each year. In such cases, those responsible for organizing these meetings make the selections and help with the editing. This means that the associate editors, listed on the masthead, are not responsible for their accuracy or clarity.

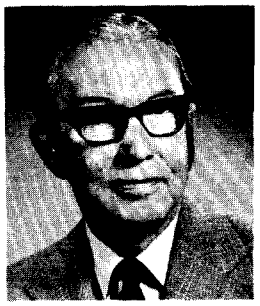
This procedure has some advantages and some disadvantages. An advantage is that it assures us of having several high-quality technical papers in each of these issues. This is important for our members who wish to keep up with the technical developments in the field, and to encourage libraries to subscribe to and keep the Journal on file. Libraries are interested in the technical issues which have a lasting value for future reference, not the editorials and committee reports. To be of interest to libraries and abstracting services, four issues a year with several technical articles each is a minimum.

The disadvantage of this system is that the topics represented do not cover all of the areas of interest to our members. Just about every area of interest to our members is included in the annual meetings. Still, it would be good to have more voluntary contributions for regular JNMM issues.

The papers in this issue were selected from those presented at the Technical Workshop on Process Holdup of Special Nuclear Materials, which took place in Rockville, Md., March 2-4. This was an extremely well-organized and well-managed workshop. Each of the authors was asked to prepare a summary of the paper to be presented, and these summaries were printed in a neat report which was handed out at the meeting. This made it possible to plan ahead for the discussions which followed each session and obviated the need to take notes while attempting to understand the presentations.

Holdup of nuclear materials in equipment has been with us as long as there have been nuclear materials to process. Considerable progress has been made in locating and "measuring" it during the last 20 years; but more progress is needed. K.K.S. Pillay, the chairman of the organizing committee, provided a bibliography with 71 items in the workshop program, which is not a large number of items for a subject so varied, complex, and universal. Some of the authors requested and received constructive suggestions as to how to make better assessments. That the workshop was useful, there can be no doubt. That more discussion and exchanges are merited, is also obvious. Comments on these papers, and on this topic in general will be welcomed.

William A. Higinbotham
Brookhaven National Laboratory
Upton, New York

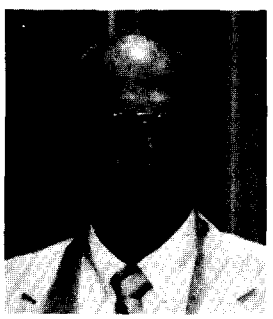


CHAIRMAN'S MESSAGE

Facing Change with an Eye on Quality

Greetings fellow members and supporters of the INMM. For the past two years I have been sharing my views and ideas as Chairman of the Institute. This has been a pleasure, and I have enjoyed discussions with many of you.

During the past two years, the Institute has continued to uphold its reputation as a serving and caring professional organization. At the same time, the INMM has been facing transition, as change is a way of life. Yet, there has been a steady improvement in virtually every area of INMM activity. This trend began several years ago when we implemented long range planning.



In looking back over the past years' successes, I see two common elements: the quality of the people and the quality of the programs. The Institute is fortunate to have a membership composed of such high quality professionals. This association is the major reason that it has been an honor and privilege to serve as Chairman of the Institute.

In the coming years, the INMM will continue to face challenges, and it must continue to handle change. Planning for the future — long range planning — will continue to be a key element in the task of keeping the INMM a viable organization. The Institute must accept the challenges of globalization and work to provide the quality programs that are needed, at the proper cost. At the same time, the Institute must carefully guard its personal character and professional

quality, traceable to our roots over a quarter century ago.

Finally, I would like to thank you for all your support. I encourage each member to continue your involvement in INMM activities and, where possible, expand them. Also, you must continue to support your leadership; be encouraging, and let them know your desires and concerns.

Charles M. Vaughan
GE Nuclear Energy
Wilmington, North Carolina

A Summary of Discussions at the Technical Workshop on Process Holdup of Special Nuclear Materials

The INMM-sponsored technical workshop on process holdup of special nuclear materials (SNM) held at Rockville, Md., March 2-4, 1988, provided a forum for timely discussion of ideas and exchange of information among persons concerned with the problems of holdup at nuclear materials processing facilities in the U.S., two from the U.K., and representatives of the Nuclear Regulatory Commission (NRC) and the Office of Safeguards and Security of the Department of Energy (DOE) attended the workshop. Formal presentations of papers at each session were followed by lively, responsive, and constructive exchanges among the participants. Individual perspectives on problems and participation in discussions of issues related to holdup varied with each participant's background, past experiences, and current responsibilities. There were good-natured confessions of inadequate attention as well as serious expressions of frustrations in dealing with holdup problems.

The 26 formal presentations at this workshop and extended discussions that followed demonstrated that holdup is considered an insidious problem by facility operators and regulators alike. The first two sessions reviewed safeguards-related issues such as the impact of holdup on inventory differences (IDs) and the frustrations and rewards in attempting to estimate hidden inventories through nonintrusive measurements.

Papers representing a variety of processes and plant experiences resulted in exchange of new ideas and information among all the participants.

One of the formal presentations of the first day described the experiences in designing a new facility with holdup estimation in mind. A second paper from a highly enriched uranium recovery operation gave a "lessons learned" type presentation covering a wide range of plant experiences. Two papers addressed experiences at a plutonium facility with extremely challenging holdup measurement problems. The authors described the innovative approaches they had taken to estimate holdup in a variety of contaminated canyon floors and process equipment. Another paper provided insights into holdup problems at a mixed oxide plutonium processing facility, the compounding of ID problems, and attempts to identify and measure holdup. Several papers in the first day's sessions reviewed problems unique to uranium enrichment plants. The capability for orderly shutdown of processes to estimate holdup at most bulk handling facilities was contrasted against the need to make such estimates in a dynamic mode at gaseous diffusion plants. An unusual experience involving a 15 Mt uranium holdup in a twenty foot long pipe at a low-enrichment gaseous diffusion plant and approaches taken by the plant operators to address the problem received special attention.

The second day's activities started out with "Unresolved Issues Regarding Holdup," where two senior representatives from U.S. federal agencies presented their views of holdup problems. These presentations were complemented by presentations of issues common to all facilities as well as those of special concern to NRC licensees and DOE production facilities. The lively discussion that followed clarified many issues as well as underlined the need for regulatory reforms and new perspectives on operational and safeguards practices at processing facilities. Recently issued amendments to the NRC's materials control and accounting (MC&A) requirements and upcoming new orders from the DOE were extensively discussed. The impacts of performance-oriented MC&A requirements issued by the NRC and graded safeguards in the new DOE orders were explored.

Holdup assay techniques were discussed during several presentations and at a dedicated session. A number of papers based on neutron and gamma measurements reviewed progress made in holdup measurements over the years, and presented new and innovative ideas for using available technologies to measure holdup at operating facilities. A unique application of in-situ neutron activation analysis to estimate SNM dispersed in a hostile environment was the subject of one of the presentations. A paper from British Nuclear Fuels, plc., reviewed the applications of multiple neutron detectors placed throughout a plutonium finishing plant to estimate in-plant plutonium accumulations on a near-real-time basis. The importance of understanding physical models of radiation transport and detection to develop mathematical expressions and their potential value to instrument calibrations was also discussed at this session. The advantages and limitations of NDA techniques and the need for innovations in planning of holdup measurements, designing portable instruments, and preparing geometrically and chemically compatible standards for noninvasive, nondestructive holdup measurements were highlighted during this session as well as others. In general, all discussions on holdup measurements emphasized the objective of achieving improved measurement accuracy and the need to establish credibility to support material balance inventory data.

The final session was dedicated to the use of statistical sampling and modeling in holdup estimation. Three papers demonstrated the potential value of models in estimating holdup. In the opening paper, holdup models were developed for a new facility using published data from experimental studies. For the process of interest, anticipated holdup was believed to conform to a negative exponential growth curve. Such modeling efforts have value in evaluating process and equipment design choices. In the second paper, computational issues related to developing model-based in-process inventories on a near-real-time basis were discussed. A third paper addressed issues related to tritium holdup and brought out experiences in tritium inventory control. The importance of detailed knowledge of the process operation for model development and validation was emphasized. The use of formal sampling plans to provide technically defensible holdup estimates was the subject of yet another paper, where unique aspects of holdup relative to the application of textbook sampling principles were reviewed.

Eight selected papers from the workshop have been prepared for publication in this issue of the *Journal* of the INMM. A separate publication issued by the INMM provides summaries of 23 of the presentations at this workshop.

This workshop on holdup was the first forum for open discussion of concerns among responsible people, and it has paved the way for renewed efforts to develop practical solutions to holdup problems that have been haunting the nuclear process industry from its inception.

K.K.S. Pillay
Los Alamos National Laboratory
Los Alamos, New Mexico

T.L. Brumfield
Martin Marietta Energy Systems, Inc.
Oak Ridge, Tennessee

Workshop Program Co-Chairmen

Selected Results for Measuring Holdup in Oxide Hoods and Transfer Lines at Hanford

R.A. Jones and R.E. Kerns
Westinghouse Hanford Company
Richland, Washington U.S.A.

ABSTRACT

Physical inventories require quantifying the holdup in process equipment; one practical way of accomplishing this is measuring the gamma ray from plutonium (^{239}Pu). Field measurements of plutonium holdup in dry plutonium oxide hoods or process transfer lines is frequently biased low, due to the possibility of heavy loading causing severe self-attenuation of the gamma rays. An experiment was performed to determine the allowable loading limits for measuring plutonium holdup in dry oxide hoods. Field measurements of transfer lines in a plutonium production facility are frequently biased low due to the self shielding of the holdup material. Fixed and portable gamma ray measurements were made of 6-ft-long sections of pipe. The results of this comparison are being used as a correction factor for routine field measurements of piping in the production facility.

INTRODUCTION

By procedure, the maximum quantity of residual plutonium in a working dry oxide hood is specified in criticality limits; thus, the quantity of plutonium allowed far exceeds the quantity that can be realistically measured by portable gamma nondestructive assay (NDA). An experiment was performed in a process hood in the oxide conversion area of the Plutonium-Uranium Extraction (PUREX) Plant at the Hanford Site, Washington, to investigate plutonium measurement in dry hoods. Known aliquots of oxide powder were added to the floor, and portable NDA measurements were taken after each successive addition. Sufficient measurement points were used to ensure that the entire floor of the hood was measured. The 413.7-keV ^{239}Pu peak was measured, using a shielded intrinsic germanium detector and portable MCA, and total plutonium was calculated based on the known isotopic abundances of the material. The gram values determined by NDA for each oxide lot were compared to the gram values determined by using wet chemical analytical methods on the same aliquot. The results were used to estimate allowed plutonium loading when performing portable gamma NDA measurements on oxide hoods at the PUREX Plant.

In a process plant the sheer number and lengths of transfer lines (solution, vacuum, and exhaust), their frequently awkward locations, and their dynamic loading has made measuring the self-attenuation of the plutonium gamma ray for each measurement point along these pipes quite impractical. Yet, it is known that the self-attenuation of the holdup material can be considerable in the size of piping and loading typically encountered. A method was needed to estimate the self-attenuation in piping. An estimate was derived, based on a comparison of portable NDA with fixed gamma NDA measured values for 6-ft-long sections of pipe.

OXIDE HOOD DETAILS

The hood section used was 36 in. wide and 6 ft. long at the floor level. The physical space constraints required that the detector be no more than 12 in. from the walls of the hood. Measurement points were chosen every 12 in. along both sides of the hood to ensure full overlap, and the measurement was extended one position beyond the active area of the hood on each end. The detector was placed on the floor for each shot, shooting parallel to the floor with the active detector centered 2½ in. above floor level. The shielding for this detector caused the response to drop to 50% at an angle of 26½ degrees from the center line; thus, the 12-in. spacing ensured full overlap at the near wall. The hood walls are formed of 3/16-in.-thick stainless steel, with 2-in.-thick Benelex shielding. The attenuation of samples of Benelex were measured and the results used to correct the measurement data for all inventory work as well as for this test.

During the measurement process, leaded gloves in the hood were moved before Run 4 and a length of channel iron was discovered under them. The data was corrected for the attenuation caused by the channel iron and both corrected and uncorrected data were turned over to an independent statistics team for analysis. A beginning background run and five data runs were made as material was added and subtracted from the hood in measured amounts. The measurement team was not informed of the amount or if it was an addition or subtraction until the

data was turned in. The background run showed 470 g of plutonium uncorrected and 504 g of plutonium corrected for the channel iron. Table 1 and Figure 1 summarize the net amounts found for the next five runs compared to the sum of the amounts actually added.

Table 1. Net Amounts of Plutonium (g).

Amount	Run (% of actual)				
	1	2	3	4	5*
Actual	922	922	1,172	649	337
Uncorrected	866 (94%)	869 (94%)	1,155 (99%)	745 (115%)	371 (110%)
Corrected	951 (103%)	929 (101%)	1,241 (106%)	817 (126%)	419 (124%)

*New detector

Recoveries were excellent for both corrected and uncorrected data when the hood configuration was not disturbed. In Run 2 no material was added, but the existing material was rearranged into a nonuniform distribution, with piles scattered around the hood and close to the edges. In Run 4, when the leaded gloves were removed, a significant error was introduced. Between Runs 4 and 5 the intrinsic germanium detector failed and was replaced by the backup detector. This did not significantly affect the results.

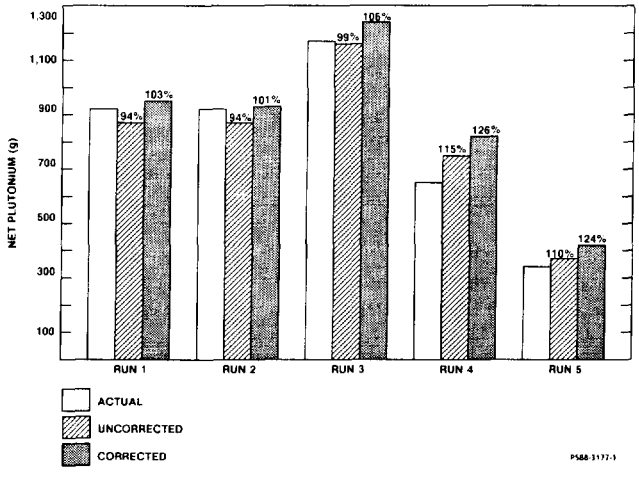


Figure 1. Recoveries of Added Oxide Measurements.

OXIDE HOOD CONCLUSIONS

Experimental results indicate that portable gamma NDA in oxide hoods is reliable at levels to at least 70 g of plutonium/ft^2. Problems due to self-attenuation at these levels are minor compared to problems caused by changes in the hood configuration. This underlines the importance of bringing the hoods to a standard repeatable configuration for each inventory. Runs 1 and 2 contained the same material, but in Run 2 it was rearranged into a nonuniform distribution, with piles scattered around the hood and close to the edges. This caused only minor errors, due to the large amount of overlap between adjacent shots; thus, distribution errors appear to be small when the hood can be shot from both sides and all areas of the hood are overlapped between adjacent shots.

TRANSFER LINE DETAILS

The data generated for the comparison was from 6-ft-long sections of a 6-in-inside diameter pipe. The sections were so heavily caked with plutonium that they had to be scraped before the data could be obtained. The following procedure was used to obtain the data. The pipe was gamma assayed by portable NDA (all NDA measurements were taken from the side of the pipe) for its plutonium holdup. All removable plutonium was sent to the Plutonium Finishing Plant NDA Laboratory where the plutonium content was measured by a Segmented Gamma Scan Assay System (SGSAS). The cleaned 6-ft-long pipe sections were again assayed by portable NDA for any residual plutonium. The difference between the first portable NDA and the second portable NDA was compared to the SGSAS-measured amounts removed from each section. These results are shown in Table 2.

Table 2. Plutonium Holdup Measurements.

First NDA (g)	Second NDA (g)	Delta mass (g)	Measured SGSAS (g)	Ratio
103	9	94	217	2.31
102	6	96	195	2.03
120	10	110	277	2.52
146	23	123	247	2.01
155	18	137	317	2.31
168	25	143	249	2.06
150	5	145	263	1.81
182	8	174	297	1.71
196	14	182	357	1.96

NDA = Nondestructive assay.
SGSAS = Segmented Gamma Scan Assay System.

The portable NDA instrumentation consisted of an ^{241}Am-doped NaI detector connected to two single channel analyzers. One analyzer was used to measure the plutonium peak (375 to 500 keV) while the other analyzer was used to measure the ^{241}Am doping peak. Measurement of the doping peak was used as a quality control check on the detector calibration. The SGSAS was a commercially available unit from Canberra, Inc.

PIPE TEST CONCLUSIONS

The spread in observed ratio seems to have no correlation to the amount of material in the pipe, which seems reasonable considering the uncertainty of the analytical mea-

surements, self-attenuation, and nonuniformity of the plutonium. The SGSAS is also a gamma-ray measuring system and, as such, is affected to some extent by the same self-shielding and attenuation factors which affect the portable NDA systems. Due to the limited data, no detailed evaluation was done.

Based on the bias observed (detailed in Table 2), current practice at this facility is to double the portable NDA-measured amount of plutonium for piping of less than 10 in. in diameter and 6 ft. in length, with assay values greater than 50 g. The portable NDA team feels this practice gives an improved estimate of the plutonium holdup in pipes. As more test results become available, this practice will be reevaluated.

SIGNIFICANCE

The methods developed are useful in accounting for self-attenuation of plutonium when measuring holdup in material balance areas. There is a tendency for holdup quantities to increase with throughput in a processing plant. The implementation of these methods was necessary to prevent holdup measurements from adding a significant bias to the accounting of special nuclear materials at the Hanford site plants.

DISCLAIMER

Roger A. Jones earned his M.S. degree in nuclear chemistry from Oregon State University at Corvallis, Oregon in 1976. He accepted a position at the Hanford Site in 1978 and has been responsible for the technical support to a counting room responsible for analysis of effluent, waste, and research support samples from throughout the site. In 1980 he became a member of the Portable NDA team that measures holdup at production facilities; in 1982 he became a team leader of other chemists involved in these roles.

Robert E. Kerns earned his B.S. degree in math/physics from Whitman College at Walla Walla, Washington in 1969, and an M.A.T. degree in physics from Kansas University at Lawrence, Kansas in 1971. After several years of service as a math/science teacher, he accepted a position at the Hanford Site in 1980, setting up the PUREX Plant counting room preparatory to plant restart. His interest include portable NDA, automation, and data base maintenance and design.

Measurement Campaigns for Holdup Estimation

R.R. Picard
Los Alamos National Laboratory
Los Alamos, New Mexico U.S.A.

ABSTRACT

The derivation of technically defensible holdup estimates is described. Considerations important in the planning of measurement campaigns to provide necessary data are reviewed and the role of statistical sampling is discussed. By design, the presentation is nonmathematical and intended for a general audience.

INTRODUCTION

Though clearly important, use of sampling principles in the planning of holdup-related activities is sometimes viewed with apprehension. Holdup is often poorly understood to begin with, and the incorporation of other esoteric matters only adds to an image problem.

Unfortunately, there are no painless options. In many operating facilities, surface areas on which holdup has accumulated amount to many square miles. It is not practical to pursue 100% measurement of all such surface areas. Thus, some portion is measured — constituting a "sample," whether obtained by a formal procedure or not. Understanding the principles behind sampling is important in planning and in developing legitimate holdup estimates. Although derivation of legitimate, facility-wide holdup estimates is not currently mandated by Department of Energy regulatory requirements, the related activities would greatly advance the present state of knowledge.

PLANNING OF A MEASUREMENT CAMPAIGN: PRELIMINARIES

Before a measurement campaign begins, reliable measurement techniques must be available. As with nondestructive assay (NDA) measurements used for accountability, calibrations must be developed to convert observed responses to material amounts. This may require careful construction of special standards, interpretation of cleanout data from pieces of equipment previously measured, and/or use of results from controlled experiments. Also, it is important that measurement uncertainties be determined at this stage, and that those uncertainties not be "picked out of the air" but be representative of achieved accuracies. Interested bystanders, such as regulatory bodies, may want to be convinced of the validity of measurement procedures and claimed uncertainties before lending credence to the results.

There are many cases where direct holdup measurements are possible on a periodic basis. Here, the term "direct measurement" usually means NDA because results can be obtained quickly and with a minimum of intrusiveness on operations. Measurement campaigns based on direct measurements can be designed through the application of sampling theory. Holdup estimates and associated uncertainties are obtainable via the usual procedures.

Given necessary input information, development of a cost-effective, technically sound sampling plan for a measurement campaign is straightforward. The corresponding mathematical optimization reduces to the propagation of measurement and sampling errors together with integer programming, the details of which are not of concern here. An artificial example from the world of systems studies, illustrating the calculations for stratified sampling, has appeared[1] in the literature. In that paper, an efficient measurement campaign was derived using parameters concerning a (hypothetical) operating facility. I am not aware of any similar work in the open literature, perhaps owing to the specialized nature of the problem and/or restrictions concerning classified data.

Principles common to other applications of sampling — uses ranging from Nielsen television ratings to projection of election outcomes — are long established and widely understood. Unfortunately, applications to holdup estimation are, in some ways, unique. Consider the example where material has plated out within the pipework of a large facility. The total amount of piping (perhaps measured in the tens or hundreds of miles) may be difficult to determine because facility blueprints are out of date or awkward to read. Moreover, access to some of the piping for measurement may not come easily. A textbook approach to holdup monitoring — "measure a random sample of pipe sections" — may be difficult to implement. Nevertheless, such sampling is the basis for the meth-

Table 1
Major Factors in the Planning of Measurement Campaigns

Factor	*Importance*	*Practical Considerations*
anticipated amount of holdup in each type of equipment, on average	want to devote most effort, all other things equal, to areas containing the most holdup	cleanout data and results from previous measurement campaigns (if any) can be used here
homogeneity in amounts of holdup for each type of equipment	similar to sampling of bulk materials; i.e., if amounts are very homogeneous, sample sizes can be small	impacts sampling error contribution to overall uncertainty
costs	can't effectively propose or allocate a budget if specific costs are unknown	costs can be large when there is interference with operations; some costs are hard to estimate initially
sampling frame (i.e., the comprehensive list of equipment containing holdup)	individual items to be measured constitute a sample (possibly stratified or clustered) from this list	collecting this information can be very tedious (e.g., *exactly* how many miles of piping exist in a large plant?); facility blueprints helpful here
measurement uncertainties	can't propagate errors without knowledge of these	cleanout data, holdup experiments, and (in principle) measurement control efforts based on realistic standards can supply information
uncertainty in estimated holdup	crucial for interpretation of bottom line results; σ vs. cost curve is helpful for budget allocation	depends on measurement errors (for measured items) and sampling errors (for extrapolating results to unmeasured items); uncertainty can be large for both reasons

odological purity at the heart of defensible holdup estimates.

PLANNING OF A MEASUREMENT CAMPAIGN: SPECIFIC CONSIDERATIONS

Many factors influence the planning of a measurement campaign; see Table 1 for summary. Quantitative information on these factors is necessary input for a serious design effort and for sample size determination.

Perhaps the single most important consideration is the anticipated holdup in different pieces of equipment. The amount of holdup being measured in any given instance can vary greatly depending on the situation. From the standpoint of planning, it is desirable to concentrate most effort, all other things equal, where the most material is.

As with generic sample survey design, planning is circular and prone to problems with 20/20 hindsight. Efficient surveys exploit the known demographics of the population of interest; however, if the demographics were known **completely**, there would be no need to conduct a survey.

For example, a measurement campaign might be designed calling for extensive monitoring on a certain type of equipment that is felt, a priori, to contain much holdup. It could turn out, upon actually making the measurements, that holdup in the equipment was less than anticipated. In the extreme, the resource allocation could be inefficient because of the initial misconceptions. There's not much to be done about this problem except to recognize that it exists and that planned effectiveness can be different than actual effectiveness. Obviously, interpretation of results from previous measurement campaigns (if any), preliminary NDA surveys, and controlled experiments can be useful in the planning stage.

Homogeneity in amounts of holdup for a given type of equipment can also impact sample design. Return to the example where holdup has plated out inside some pipework. From section to section along the length of pipework, amounts of holdup in those sections could be roughly the same or, if there were low spots or other points of accumulations, the amounts from section to section could be more variable. This factor affects sampling errors similar to the way material homogeneity affects bulk sampling of materials. That is, if there has been uniform plating and the pipe sections contain very nearly the same amounts of holdup, it is not necessary to measure too many sections to get a good idea of the holdup in the entire length of piping. Conversely, if holdup in pipe sections were less uniform, more measurements would be needed to give the same quality estimate.

The standard deviation of the estimated holdup depends on two elements. The first is the effect of measurement uncertainties for those items that are measured and the second is the effect of sampling errors that result from extrapolating results from the measured items to the items that are unmeasured. Both contributors can be disturbingly large. Through error propagation, a legitimate estimate of uncertainty can be obtained. Such estimates are not achievable in the absence of the formal sampling plan.

(Aside. Although overall facility holdup is relevant to the facility's cumulative ID, the ID for an individual accounting period is affected by the change in holdup during the period. In some cases, NDA count rates can be fairly reproducible if there has been little or no change in the holdup for a particular item. If so, changes in holdup may be estimable with small absolute error even though the amount(s) of holdup involved may not be well known.) Although space constraints for this paper pre-

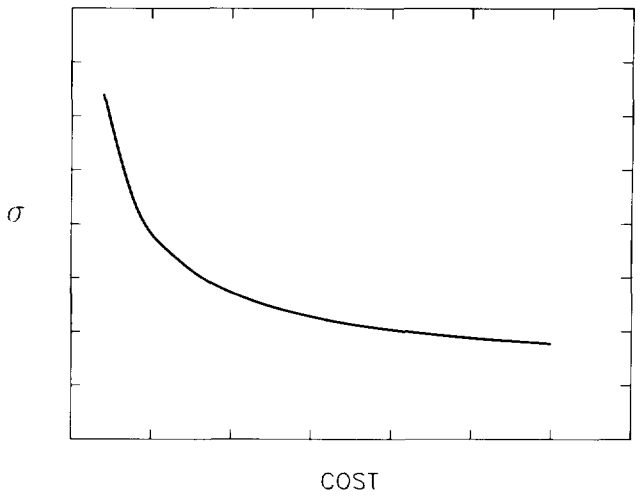


Figure 1. Typical shape of σ (uncertainty in estimated holdup) vs. cost (of measurement campaign) curve.

clude the detailed treatment of a realistic numerical example, Figure 1 displays the typical shape of a σ vs. cost curve, where σ denotes the standard deviation of the estimated holdup corresponding to the optimal sampling plan. Note that the curve has a diminishing returns shape. Its asymptote corresponds to the limiting accuracy that can be achieved given measurement uncertainties were there to be infinite resources and 100% sampling carried out.

Derivation of Figure 1 for a specific application is extremely useful in planning, even if the calculations are based on rough estimates for costs, measurement uncertainties, and so on. Results indicate what kind of performance can be anticipated for given expenditures. Determining a "best" amount of resources to expend on a measurement campaign is rather subjective, involving the tradeoff between increased cost and increased accuracy of holdup estimation. Other factors, such as the negative publicity that could arise from holdup-related ID problems and potential difficulties posed by re-assignment of measurement personnel, may also require subjective evaluation.

Several reasons exist for the diminishing returns phenomenon. One is that sampling errors behave this way; for example, exit polls to project election outcomes don't survey millions of people, but usually less than 2000. Indeed, although this is counter-intuitive to some, the absolute number of people sampled is much more relevant to overall survey accuracy than the fraction of the population sampled. Secondly, the effects of so-called systematic measurement errors persist and are not diminished by increasing the sample size. And third, the incremental component of overall costs is often linear in the sample size. For large populations, carrying out too many measurements (i.e., thousands) can be time-consuming, expensive, and unnecessary for stated purposes.

PLANNING OF EXPERIMENTS

Controlled experiments are an essential part of understanding holdup. Historical data are often fraught with shortcomings — corresponding measurements may not be of adequate quality to determine the nature of material deposition, relevant factors affecting holdup may not be controlled or recorded, and confirmatory information (cleanout data, precise knowledge of material input to the equipment) may be unavailable. Collection of data as part of a hasty response to an ID crisis has occasionally exaggerated these problems. As such, the value of historical data for holdup estimation is often limited and controlled experiments are quite useful.

The first step in planning an experiment is to determine the objective(s). Many things could be of interest, such as perfecting techniques for routine measurements to be made during future measurement campaigns, obtaining uncertainty estimates appropriate for those measurements, characterizing holdup accumulation with respect to throughput in a piece of equipment for certain operating conditions, characterizing holdup accumulation in the form of a spatial profile, or determining which operating conditions affect holdup and which do not. Different objectives naturally lead to different experiments.

Modeling of experimental results can have many benefits. The mere act of developing and validating the model leads to improved understanding of the dynamics of holdup accumulation. Moreover, estimation can be pursued in a more timely and cost-effective manner; indeed, it may be possible to estimate holdup at certain times without resort to direct measurement at those times. The related subject matter, primarily time series modeling and/or response surface methodology, is extensively discussed in the statistical literature. Successful applications of model development for holdup estimation have been reported (e.g., Ref. 2).

Of course, limitations must also be recognized. If estimation of holdup were an easy problem, it would already be solved. In some instances, it is impractical to develop models, such as when negligible amounts of material are involved, high quality measurements to determine the specific form of the model are not achievable, acceleration of time is difficult (that is, reproducing effects of long-term process operation from short-term experiments), or costs of experimentation exceed benefits.

Nonetheless, there is no substitute for tackling the holdup problem head on. The alternative is the status quo, where holdup is often a mystery.

REFERENCES

1. R.R. Picard and K.K.S. Pillay, "Statistical Sampling for Holdup Measurement," Trans. Amer. Nucl. Soc. 53, 193-194 (1986).
2. K.K.S. Pillay and R.R. Picard, "Estimation Methods for Special Nuclear Materials Holdup," Nucl. Mater. Manage. XIII, 60-65 (1984).

Rick Picard joined the Los Alamos National Laboratory in 1981 after earning his B.A. in mathematics from Carleton College and Ph.D. in applied statistics from the University of Minnesota. He has since worked on a variety of safeguards problems with the Safeguards Systems Group and on arms control problems with the Statistics Group.

The Importance of Understanding Process Holdup A Department of Energy (DOE) View

Glenn A. Hammond
Office of Safeguards and Security
U.S. Department of Energy
Washington, D.C. U.S.A.

Ron L. Hawkins
Lamb Associates, Inc.
a subcontractor to Meridian Corporation
Rockville, Maryland U.S.A.

ABSTRACT

Residual material in processing equipment is today and will in the future continue to be one of the major problems in controlling and accounting for nuclear material. Existing facilities were designed for product quantity, quality, and safety; not for minimization and quantification of residual material in process. With the development of measurement systems, and with enhanced material control and accounting practices and procedures, inventory differences have been emphasized. The improvement in processing input and output measurements has highlighted the problems of quantifying residual process material. The primary purpose for quantifying material held up in process is to determine the inventory difference and its related uncertainties or statistical variances. This quantification and its associated problems must be addressed if we are to prevent, deter, and detect theft and/or diversion of nuclear materials.

INTRODUCTION

Recently revised DEO Order 5633.3, "Control and Accountability of Nuclear Material", defines holdup as "the amount of nuclear material remaining in process equipment and facilities after the in-process material, stored materials, and product have been removed." It further states that "Estimates or measured values of materials in holdup may be reflected in the facility's inventory records."

In the DOE complex, processing facilities, such as scrap recovery, oxide-to-button lines, and processing facilities employ large and complicated equipment (dissolvers, precipitators, extraction columns, mixer-settlers) to attain DOE-scale throughputs. Process holdup in this equipment during inventory, and any variation between inventories can be an important contributor to the inventory differences and their control limits. Recognizing the large uncertainties usually associated with holdup estimates or measurements, biases can significantly reduce or mask the real Inventory Difference (ID). As a result, DOE is concerned that ID's may soon become an endangered species. Ensuring that ID's continue to be a valid indicator for detecting possible theft or diversion should be one of our primary concerns.

A prime example of the complexities of process holdup is a glovebox and its associated filtering system. Most gloveboxes have square corners that are difficult to reach from the glove ports. At the time of inventory, the bulk of residual material can be removed by sweeping and shovelling until only gram amounts are left as contamination and accumulation in the corners. The problem then becomes one of determining how many grams remain and how accurately the quantity is known. You may find that the residual material is less than 20 grams, but the uncertainty associated with the material is greater than 20 grams. If a material balance area (MBA) has many gloveboxes, the uncertainty associated with this residual material may be a significant contributor to the inventory variance. It can be a greater contributor than kg quantities of material that are in an accurately measurable form.

The significance of the glovebox holdup is augmented by the ventilation system and associated filters. Normally the filtration system (e.g., HEPA filter) is located on or near the glovebox. The ventilation system has been designed to minimize the release of SNM when a filter is changed, but generally is not designed to minimize holdup or facilitate its removal.

The holdup quantities and uncertainties for the HEPA filters are similar to the glovebox. One difference is that if a filter is left in place for a relatively long period, then the accumulated quantities are greater and associated measurement uncertainties are much greater due to unknown absorption effects. When a filter is removed, material is introduced into a ventilation system, resulting in less total holdup but a larger measurement uncertainty due to the number and type of measurements required in accounting for the distributed material.

In uranium processing, the UF_6 to U_3O_8 or UO_2 processing activities illustrate many of the problems encountered in monitoring or measuring holdup. The physical form of the SNM changes from a solid to a gas to a liquid back to a solid, and chemical composition changes approximately

five times. In addition to the problems with gloveboxes and ventilation systems discussed above, in this flow we have: (1) a very reactive form of SNM, UF_6, that has to be heated to be removed from its container and can react with the moisture in air to form a solid or can plate out on cool surfaces; (2) hundreds of linear feet of piping and associated valves and tank storage systems that have large surface areas for "film" holdup, drainage problems, and are prone to leaks; (3) SNM in a solution that can react with the metal and the binder in concrete to cause containment leaks; and (4) SNM calcining systems that are prone to holdup and which cannot be disassembled for cleaning without major expenditures in time and labor.

STUDIES AND DEVELOPMENTS

DOE has funded the development of equipment and performance studies to identify, quantify, and resolve problems associated with the control and accounting for holdup. In the area of equipment, significant advances have been attained. Stabilized portable single channel analyzers, portable multichannel analyzers and associated NaI and GeLi detectors have been developed that allow measurement systems to be taken into a process area, to the point of holdup locations, to make measurements.

Two of the holdup measurement problems that have not been solved are the acrobatic skill and endurance required of personnel in the measurement of holdup. The holdup version of "Murphy's Rule" is "SNM holdup seeks an area with highest background, the most inaccessible location, and the point with the greatest shielding, while maintaining a configuration that produces the greatest possible self attenuation." A third problem which has always been an obstacle is unique to the quantification of holdup. It is the general misconception and lack of understanding of holdup measurements. As Jim Sprinkle of the Los Alamos National Laboratory (LANL) noted recently, "Some people consider holdup measurements to be a black art. I prefer the description of holdup measurements as lots of hard work requiring extensive training and careful observation."

BASIC APPROACHES: LEARNING TO LIVE WITH HOLDUP

While studies have not always provided results immediately applicable to the problem, they have provided insight to the problems and have pointed the way for future developments. To date, there are several aspects of holdup that are well known and documented. Primary among these is that today's facilities were not designed with the minimization, quantification or removal of holdup in mind. Additionally, we know that over time SNM accumulates in most areas of a facility. Often, the accumulation can be considered to be deminimus in relation to a specific inventory difference. Holdup has been found in the concrete in dike areas, in solid concrete block, on roofs, outside doors to process areas, in demister filters, in muffle furnaces, adhered to clothing worn in process areas, as residue in containers previously used to hold process materials, on the Raschig rings in tanks of process solution, or in equipment removed from a processing area.

Secondly, measuring holdup in all pieces of equipment and associated facilities is not a viable option in terms of time, money and radiation exposure to personnel. Removal of holdup and processing it to a measurable form is also not a viable option in terms of production constraints. Product quality is often related to impurity content. Equipment that is coated with a film of SNM introduces less impurities in the product than does cleaned equipment. Thus, we have to find a way to "Live with Holdup." Studies such as the one performed by K.K.S. Pillay and R.R. Picard of the Los Alamos National Laboratory, entitled "Learning to Live with Holdup" will be of special importance in the design and initial start-up of new facilities of modification of existing facilities.[1]

There are five basic approaches to living with holdup that are used currently:

1. *Equilibrium* — Seek a state of equilibrium such that process holdup is quantified only when it is removed from the process, either as a consequence of operational actions or as a result of criticality concerns. In this approach, reduction of process holdup is not attempted and process holdup quantities are not, normally, booked. The approach is based on the assumption that process holdup is the result of a trickle loss from the process flow, the quantities are not applicable to any one inventory period, and the increase of holdup for each inventory period is less than the measurement uncertainty.
2. *Clean-out* — Clean-out and process all SNM to accurately measurable forms. Measurable quantities of SNM are not allowed to remain in process equipment. All holdup is removed and accounted for in the appropriate inventory period. This approach is the most work intensive and costly. It generates excessive "down-time" for processing equipment and often causes the subsequent generation of "off-spec" product (result of cleaning and removal of SNM coating).
3. *Monitor and Quantify* — Develop target values for the quantities of SNM allowed to remain in process equipment. The target values should be deminimus quantities of SNM; or at a level that can be accurately quantified; or at a level that does not significantly contribute to the inventory difference or its uncertainty. This approach is in wide use and lends itself to engineering efforts to reduce holdup.
4. *Historical In-Growth (Modeling)* — Determine holdup growth rates based on a history and study of recovered quantities and of processing activities. This approach assumes that the rate of growth of holdup is proportional to either throughput or time. Thus, the distribution of holdup can be modeled and quantities can be determined mathematically either from the throughput or the elapsed time since the last clean-out in relation to historical throughput and recovered holdup. A variation of this approach incorporates process holdup measurements. In this approach, the distribution of holdup is known or has been modeled. The rela-

tionship of the holdup at a specific point or group of points to the total holdup is known. The total holdup can thus be evaluated based on a limited number of measurements. The limitation of the historical in-growth/modeling approach is that a segment of the process has to be cleaned and the recovered material accurately quantified prior to implementing the approach.

5. *Sampling* — Determine quantities on a statistical sampling basis. This approach assumes that holdup patterns are known or are at least similar for similar equipment. Thus, the measurements of a few items in a population allow the extrapolation of measurement values to all equipment.

All five of these approaches have limitations, but there are examples of processing equipment for which each is considered appropriate. A combination of these approaches is probably the most desirable when one considers production requirements in relation to material control and accounting requirements for existing facilities. The most practical approach for controlling holdup is to incorporate the concern in the design of the facility and/or its subsequent modifications. Equipment can be designed to minimize the potential and to facilitate the removal of holdup. These types of design concerns have been successfully implemented for many years in the areas of health and safety and the prevention of criticality. If we are to "live with holdup", we must give it a similar priority.

CONCLUSIONS

A review of practices and holdup studies performed by the national laboratories and plant operators leads to the following general conclusions:

1. A rapid survey for hot spots is feasible and cost effective.
2. Plant operators should develop an overall strategy for holdup quantification.
3. The more experienced the staff, the higher the quality of results.
4. The measurement program staff will be challenged by the extensive documentation, calibration, and training requirements.
5. The quality of a holdup program is determined by the clean out and recovery efforts in validating results.

If progress is to be made in minimizing insider vulnerabilities, ways must be found to accurately control and quantify holdup. If this cannot be done to acceptable levels, then means must be developed to minimize its variability and monitor the boundaries of its containment. For cost-effectiveness, better measurement, estimation, and control methods must be developed to reduce the contribution of holdup to inventory differences to aid in countering insider vulnerabilities.

From a graded safeguards point of view, there is no gain in striving for the third and fourth decimal places of feed and product measurements when the control limits may be dominated by process holdup variability. Usually, one knows how to attack the third and fourth decimal places for high quality materials (better measurement control, improved standards, more training, and so forth.) With process holdup, one often does not really know how to attack the problem. This is where cooperation between the R&D labs, the Field Offices, and the facility operations remain essential.

Finally, the new DOE Order 5633.3 places a premium on the timeliness of detection and performance criteria. Obviously, these objectives go far beyond the variability of holdup at inventory time. To implement the order, one must attack the problem of holdup quantification on an hour-by-hour basis or even shorter! Nor is this unreasonable: health and safety systems already demand continuous control of hazardous quantities of SNM in process to protect personnel and sustain operational viability. Motivated by the requirements of the revised DOE orders, holdup technology must advance in parallel with the Department's ability to control the process operation to meet all the health, safety, efficiency, economy, quality assurance and safeguards requirements.

REFERENCES

1. K.K.S. Pillay and R.R. Picard, "Learning to Live With Holdup," *Nuclear Materials Management Journal. XV, 109-114* (June 1986).

Glenn A. Hammond is Director, Division of Safeguards, Office of Safeguards and Security (Defense Programs), at the Department of Energy. He has served over 30 years in various technical positions at both field and Headquarters in nuclear materials production, quality assurance, and management; and in safeguards and security programs involving policy, procedures, standards, and R&D for both domestic and international safeguards. He has been a member of the INMM for the last 20 years with primary involvement in the technical program and long-range planning committees, presentations, as a voting representative of INMM-sponsored ANSI standards, and was Executive Committee Member-At-Large during 1982-1983. He is a Senior Member of the Institute and was elected Fellow in 1985 by the Executive Committee.

Ron L. Hawkins is a Senior Nuclear and Quality Assurance Engineer with Lamb Associates, Inc. He has seventeen years of experience in the nuclear field in various technical positions. He has served as a laboratory instructor in the area of health physics; developed NDA measurement methods; provided consulting services in areas of health physics, nuclear fuel quality assurance, and safeguards; and most recently served as safeguards inspector with the International Atomic Energy Agency. Mr. Hawkins is a senior member of the INMM and was certified as a "Safeguards Specialist" in 1981.

Process Holdup — The Regulatory View

E. William Brach
U.S. Nuclear Regulatory Commission
Washington, D.C. U.S.A.

ABSTRACT

Performance-oriented safeguards regulations reflecting the different strategic value of special nuclear material were recently published as final rules. These rules have a significant impact on the requirements for holdup calculations and measurements. For licensees processing low enriched uranium, it is expected that few, if any, holdup measurements will be made for safeguards purposes. However, for licensees processing high enriched uranium or plutonium, the new regulations require extensive analysis of holdup measurements and estimations in modeling process flows, holdup, and yields for individual unit processes. Good quality holdup measurements and estimations will be required for both physical inventory and process monitoring purposes.

INTRODUCTION

This paper will discuss current requirements for process holdup measurements as viewed by the regulator of the licensed industry and it will discuss the impact of the recently published performance-oriented safeguards rules on holdup measurements. In the past few years, performance-oriented material control and accounting (MC&A) regulations that reflect the different strategic value of special nuclear material, for example different enrichments of uranium, were published.[1,2] These rules impact six low-enriched uranium and four high-enriched uranium fuel processing licensees. If licensed plutonium fuel fabrication becomes a reality, the facility would have to follow the high-enriched uranium rules. The Nuclear Regulatory Commission (NRC) has now completed the review of all six low-enriched uranium licensees' plans for implementing the new rule and these licensees are in the final stages of implementing the rule. The NRC is currently reviewing the initial submittals by the licensees authorized to process high enriched uranium.

In many respects, the traditional safeguards material control and accounting practices are retained in the new performance-oriented rules. For example, licensees are required to have measured material balances based on measured values of uranium and U-235. However, for the low-enriched uranium licensees,[3] it is expected that few, if any, holdup measurements for MC&A purposes will be conducted. Ventilation equipment may be the most common item measured for holdup, and then only for physical inventory. Because of the small quantities of uranium involved, only a minimal measurement control program is needed. Equipment which is cleaned out can be assumed to have a zero value for MC&A purposes (exclusive of any health and safety requirements) even though some residue may remain in the equipment.

However, the new rules for licensees processing high-enriched uranium[4] change considerably because of the material's high strategic value. There is a strong emphasis on more timely and localized loss detection. To achieve these detection objectives, the MC&A measurement and control programs need to be much more vigorous.

The new regulations establish requirements for unit process monitoring to detect abrupt and protracted losses of material. The new regulations also reduce the physical inventory frequency from two to six month intervals. With regard to the treatment of holdup, there are two distinct aspects of the rule. The first case is measurement of holdup for periodic physical inventories; the second case is the estimation (based on historical performance or engineering estimates) and actual measurement of holdup in the unit process monitoring systems.

PHYSICAL INVENTORY HOLDUP MEASUREMENTS

For physical inventories, holdup measurements must be made with systems calibrated using standards that are traceable through an unbroken chain of comparisons, including the overall uncertainty in each, to a national standard. The performance of each of the holdup measurement systems has to be monitored using standards that are representative of the material being measured with respect to geometric distribution, matrix, and uranium content. Standards must be constructed and controlled to ensure the continued validity of the standards' assigned values. Control limits with levels of significance of 0.05 and 0.001 are used to evaluate the reliability of the holdup measurement system. Control data exceeding the 0.05 limit must be investigated and corrective action taken. Whenever the control data exceeds the 0.001 limit, the holdup measurement system must not be used for MC&A purposes until it has been brought into control at the 0.05 limit. Control limits are sometimes exceeded because of instrument drift

or failure or someone changing instrument settings. To bring the system back into control, recalibration is often needed. Bias corrections have to be applied to individual measured holdup areas if for any measurement system: (1) the relative bias estimate exceeds twice the standard deviation of its estimator, (2) the absolute bias estimate exceeds 50 grams of SNMM when applied across all measured areas, and (3) the absolute bias estimate on an individual measurement basis exceeds the rounding error of effected measurements.

PROCESS MONITORING HOLDUP ESTIMATES

The new regulations require licensees to subdivide the operations into unit processes. Each of these unit processes is subjected to periodic tests and evaluations to identify and to localize losses of strategic special nuclear material.

The criteria or threshold for alarm in each unit process is based on input and output measurements, yield versus expected or predicted yield of unit process product, side streams and holdup.

The determination of holdup for unit process monitoring differs markedly from the requirements for holdup measurements at the time of physical inventory (as discussed above). For process monitoring purposes, holdup is not required to be measured for each abrupt or protracted material loss test. Rather the process holdup can be estimated based on engineering analysis and verified by actual holdup measurements and/or historical cleanout data. Depending on the strategic attractiveness of the material processed, the abrupt loss test is required to be conducted every 3 or 7 days while the protracted loss test would be conducted on a longer period of time and this test would analyze trends in process performance.

Licensees will be expected to prepare engineering models for each unit process to characterize the holdup estimates to be used in performing abrupt and protracted loss tests. For example, historical data can be used in the model to reflect actual quantities of material recovered from previous process cleanouts. These data, correlated with engineering estimates of material accumulated in the process as a function of throughput and time and other factors), could be used to model the holdup estimate. It is expected that the holdup measurements and the process/equipment cleanout activities conducted during the semiannual physical inventories will provide additional data to evaluate the accuracy of the holdup estimates used for process monitoring purposes. Further, any changes in the process operations will require reanalysis of the engineering model used for the process units affected by the change.

Alarm response and resolution are key requirements of the new regulation. The importance of sound engineering models for each process unit and its associated estimate of holdup are highlighted by the specific alarm response actions required by the regulation. For example, if an abrupt loss detection estimate exceeds five formula kilograms of strategic special nuclear material, then processing operations must be suspended and the NRC must be notified. Both actions are to be completed within 24 hours of the occurrence of the alarm. Inaccurate engineering estimates of holdup should become evident quickly if the holdup estimates are identified as the cause of repeated alarms. In most cases, NRC expects that the licensee's initial response to an abrupt loss alarm or a trend indicative of a protracted loss in a process unit would include review and reexamination of records of input and output measurements and an assessment and reverification of the continued validity of the process unit holdup estimates.

QUALITY OF HOLDUP MEASUREMENTS/ESTIMATES

As noted above, it is important to have sound and good quality engineering estimates of process holdup to avoid unnecessary investigations or shutdown of process units to resolve loss alarms caused by inaccurate estimates of holdup. Process and engineering analyses and tests should be performed to evaluate holdup quantities and distribution and how they change relative to operational parameters. These analyses and tests should be continually reviewed, evaluated, and updated to reflect the results of process cleanout and actual holdup measurements taken, for example, at physical inventory. Further, the quality of holdup measurements at time of physical inventory is also important. All program data, including holdup measurements performed for physical inventories, must be controlled so that they do not cause the standard error of the inventory difference (SEID) to exceed 0.1 percent of the active inventory. Active inventory means the sum of additions to, beginning, ending, and removals from inventory, after all items which appear in the active inventory calculation more than once and come from the same measurement system are excluded. Holdup measurement uncertainty could be a problem in facilities with varying and large holdup quantities.

Inherently, there are large uncertainties associated with typical holdup measurements and estimates because of difficulties in obtaining representative standards. Problems with accessibility to in-process equipment for holdup measurement and analysis is likewise a major contributing factor to the uncertainty of holdup measurements and estimates. Large variations in holdup and SSNM inventory in process ventilation equipment, gloveboxes, liquid handling equipment, and furnaces have traditionally been the problem areas in holdup measurements and estimates. A number of cases have been identified where unaccounted-for-material has been found in ductwork, which caused both nuclear safety and safeguards concerns. In some cases, this happened because facility operators did not believe that uranium would be present, trapped or accumulated in ductwork. Quality holdup measurements and sound engineering analyses would most likely have detected or predicted the material buildup earlier so that the material could have been properly accounted for on inventory. Furnaces, especially large manual or semiautomatic load furnaces, pose unique problems of their own in determining holdup. Liquid systems, for example scrubbers, often accumulate solids while in other liquid process streams, solids have accumulated in unpredicted locations. Washing, and sometimes

disassembly of the system, is often necessary to determine the process holdup.

Whether measuring process holdup for physical inventory or estimating process holdup in conjunction with unit process monitoring, experience to date indicates that holdup problems are generally minimized when the process is cleaned to the same, or nearly same, level for each physical inventory and when the process is operated in a steady state mode. In the latter case, there are generally fewer variables to influence or alter the established engineering models for estimating process holdup.

USE OF CURRENT TECHNOLOGY/ TECHNIQUES

The new performance-oriented safeguards regulations do not mandate new techniques for holdup measurement and do not require the development of new technology for holdup determinations. The most commonly used holdup measurement technique by NRC licensees is passive gamma counting. NRC expects licensees to continue to use their current holdup measurement techniques to meet the new requirements. Historical process and equipment cleanout data will be used in conjunction with engineering analyses to estimate process holdup to be included in material control test models for individual process units. While new developments in process holdup determination are not required, it is expected however that holdup measurements and estimates will play a more crucial role in process difference and inventory difference alarm response and resolution.

REFERENCES

1. Title 10 Code of Federal Regulations Part 74 Subpart C — Special Nuclear Material of Low Strategic Significance 50 FR 7579, February 25, 1985.
2. Title 10 Code of Federal Regulations Part 74 Subpart E — Formula Quantities of Strategic Special Nuclear Material 52 FR 10040, March 30, 1987.
3. C.W. Emeigh, G.E. Gundersen and C.J. Withee, *Acceptance Criteria for the Low Enriched Uranium Reform Amendments* NUREG-1065, Revision 1, U.S. Nuclear Regulatory Commission, Washington, D.C., April 1985.
4. C.W. Emeigh, *Standard Format and Content/Acceptance Criteria for the Material Control and Accounting Reform Amendment*, NUREG-1280, U.S. Nuclear Regulatory Commission, Washington, D.C., March 1987.

E. William Brach joined the AEC/NRC in 1971 as a safeguards inspector and subsequently transferred to NRC headquarters safeguards staff. During Mr. Brach's career, he has served with the IAEA in Vienna, Austria as a senior expert on safeguards technologies, and within the NRC as Chief, Technical Assistant to the Director of Inspection and Enforcement, senior quality assurance engineer, and the Executive Coordinator for Regional Operations. In 1987, Mr. Brach was appointed to his current position, Chief, Licensing Branch, Office of Nuclear Material Safety and Safeguards, responsible for domestic safeguards licensing for fuel facilities and U.S. implementation of international safeguards and technical support to the IAEA.

Mr. Brach received his B.S. degree in mathematics from the University of Tennessee, and completed graduate work in computer science and statistics.

Holdup—Related Issues in Safeguarding of Nuclear Materials

K.K.S. Pillay
Los Alamos National Laboratory
Los Alamos, New Mexico U.S.A.

ABSTRACT

Residual inventories of special nuclear materials (SNM) remaining in processing facilities (holdup) are recognized as an insidious problem for both safety and safeguards. This paper identifies some of the issues that are of concern to the safeguards community at-large that are related to holdup of SNM in large-scale process equipment. These issues range from basic technologies of SNM production to changing regulatory requirements to meet the needs of safeguarding nuclear materials. Although there are no magic formulas to resolve these issues, there are several initiatives that could be taken in areas of facility design, plant operation, personnel training, SNM monitoring, and regulatory guidelines to minimize the problems of holdup and thereby improve both safety and safeguards at nuclear material processing plants.

I. INTRODUCTION

In recent years, because of the increasing concerns over theft/diversion of special nuclear materials (SNM) for clandestine applications, regulatory requirements for safeguarding SNM have become very stringent, almost to the point of seriously affecting the main missions of production facilities. An important feature of current regulations in the U.S. is the requirement to report inventory differences (IDs) periodically to regulatory agencies, congressional oversight committees, and the public. This requirement, which came into effect in 1977, did further encourage bulk-handling facilities, where potentials for SNM holdup are generally high, to examine ways to reduce holdup and thereby decrease their IDs.

Holdup of materials in process equipment is not unique to SNM processing. But uncontrolled accumulation of materials within process equipment is both a safety and safeguards concern at nuclear material processing facilities. Holdup of SNM in process facilities can result from both normal and abnormal operations of the plant. From a detailed knowledge of the process chemistry and behavior of material forms, it is possible to make reasonable predictions about regions of holdup as well as the relative magnitude of holdup in several kinds of process equipment during normal operations. However, it would be extremely difficult to speculate on the magnitude of holdup during abnormal conditions resulting from process upsets and/or improper plant operations.

II. THE ISSUES

The role of hidden inventories, or holdup, as a safeguards problem is now recognized by almost everyone interested in establishing effective safeguards for SNM. As part of the effort to organize the first INMM-sponsored workshop on process holdup of SNM, a literature survey identified over 70 publications in the open literature relevant to this subject.[1] Some of the major issues related to holdup that are of concern at present to the safeguards community are identified in the following paragraphs along with some personal thoughts on the issues themselves and possible approaches to addressing them.

1. Impact of Plant and Process Designs on Holdup

The influence of plant design on SNM holdup at bulk-handling facilities has long been recognized. In the mid-1970s, the U.S. Atomic Energy Commission issued three specific guidelines [2-4] describing desirable design features of bulk-handling facilities for minimizing holdup. Process selection and equipment design do influence holdup, and in many instances there are alternatives that can be chosen to minimize holdup. This latter approach has not yet received the attention it deserves. An illustrative example of alternative process design is the use of microwave heating for the direct conversion of uranyl nitrate to uranium oxide. This operation is still carried out in a sequence of steps involving ammonium diuranate precipitation, filtration, drying, calcination to U_3O_8 and subsequent reduction to UO_2. A significant reduction in holdup can be achieved in this process through the direct denitration and oxidation of uranyl nitrate to UO_2. Similar process changes can minimize holdup during large-scale processing of all SNM.

A second example is in the use of new materials in facility construction and fabrication of process equipment. Synthetic polymers, such as chlorinated polyvinyl chloride (CPVC), polyvinyl diene fluoride (PVDF) such as "KYNAR," fiberglass reinforced plastics (FRPs), and corrosion-

resistant alloys, such as Hastalloy, are now available for large-scale plant construction and equipment fabrication. In addition, pure metals, such as tantalum and surface liners made of high-temperature plastics, are now available at reasonable cost for process facility applications requiring reduced corrosion and surface adhesion.

2. Impact of Holdup on IDS

It is extremely difficult to locate and measure all hidden inventories of SNM in a large plant, and this is the primary reason for large inventory differences (IDs) at some of the bulk-handling facilities in the U.S. A rough estimate of the amount of plutonium or uranium required to create a thousandth of an inch (0.025 mm) coating in about 500 miles (800 km) of 1-inch (2.54 cm) diameter pipes (the length of pipe ordinarily found in several large processing facilities) is over 540 kg. This estimate assumes that the deposit has a density of 1 gm/cm^3 and that only a third of the deposit is the elemental form of the SNM. In many SNM processing facilities, there are many pieces of equipment with much larger surface areas and those with greater potentials for heavier material deposition.

Large inventory differences have resulted in many kinds of regulatory actions at a number of nuclear material processing facilities in the U.S. They have ranged from a temporary halting of facility operations to complete cessation of operations and decommissioning of facilities. The regulatory agencies have begun to recognize the seriousness of this problem. Beginning in January 1986, the U.S. DOE adopted a new format for presenting its semi-annual report on strategic SNM inventory differences.[5] The new format clearly demonstrates a recognition of the contributions of process holdup to inventory differences. Of the nine recognized categories of IDs, two of them, "process holdup difference" and "equipment holdup difference," account for a major fraction of total IDs at several bulk-handling facilities.

3. Impact of Holdup on Plant Operations

In addition to large IDs, unidentified holdup of SNM in process equipment is a serious safety problem due to its potential for criticality events. A variety of factors contribute to this situation, including the details of the process, equipment design, personnel training, process upsets, and the facility management philosophy. It is possible to identify and address these issues and take corrective measures to minimize the impact on both safety and safeguards.

4. Periodic Cleanout of Equipment for Materials Accounting

Although periodic termination of process operations to cleanout equipment for materials accounting is a desirable goal for good materials accounting, this practice is counter-productive to the primary missions of such facilities, namely the production of nuclear materials economically and efficiently. Therefore, such practices are frowned upon by managers of process facilities. As a result, most holdup measurements done to-date have been in response to large IDs or criticality safety concerns. Prudent management of facilities processing large amounts of highly enriched uranium and separated plutonium should include scheduled cleanout operations.

5. Advantages and Limitations of Nondestructive Assay (NDA) Techniques for Holdup Estimation

NDA techniques for measuring plutonium and uranium, using passive gamma and neutron measurements, have continually improved over the past two decades. There are, however, fundamental limitations to these NDA techniques that will continue to affect holdup measurements. The crowded environments of process facilities, combined with non-stoichiometry and nonuniformity of holdup residuals and the inadequacies of calibration standards and measurement equipment designs, will continue to offer challenges to NDA measurements of holdup.

6. Development of NDA Instruments and Standards for Holdup Measurement

Developing suitable standards for NDA of radioactive materials is always a challenge. However, in the case of holdup measurements, the problem becomes extremely complex due to the nonhomogeneity of the sample to be assayed, its unknown distribution pattern, varying chemical composition, the complex geometry of the equipment in which the materials reside, the attenuation of radiations by the equipment and the matrix, and the high background radiation levels in processing areas. Ideally, it is desirable to have calibration sources closely simulating the actual holdup deposits to be assayed. The present practices are to use point sources, line sources, or uniform flat sources as calibration standards and extrapolate the results to other complex geometries. Although this approach is adequate in many instances and is a desirable compromise at others, there is much that can be done to improve calibration standards for holdup measurements and thus minimize the uncertainties of such estimates.[6,7]

7. Regulatory Reforms to Accommodate Holdup Estimates to Reduce IDs

Although safeguarding of SNM was always considered important from the early days of nuclear technology development, regulatory guidelines to achieve this objective have been in a state of evolution. There is a growing recognition that a significant part of the IDs at the bulk-handling facilities is due to unmeasured inventories and/or holdup. In addition to regulatory pressures to reduce IDs, there are several safety and safeguards issues related to cumulative effects of SNM holdup. Because of the uniqueness of nuclear criticality safety, many holdup-related safety issues are addressed during plant and process designs. However, in the past, safeguards issues seldom influenced facility design, and they are often difficult to resolve later.

8. Pragmatic Alternatives to Plant-Wide Holdup Measurements

A prevailing view is that facility-wide holdup estimates would have very large measurement and sampling errors; thus, adjusting an ID to reflect an estimated change in

holdup would create a new quantity with uncertainty so large as to render the quantity meaningless. An objective re-examination of this view as well as current regulations and the development of pragmatic approaches to include sound estimates of holdup in calculating IDs would be a step in the right direction.

Regulatory reforms encouraging the use of modern tools, such as sampling and modeling, to maximize the use of resources and develop technically sound estimates of holdup without interrupting production schedules would encourage facility operators to invest resources to address this problem. Although indirect methods of holdup estimation[8] using tracer techniques and mathematical modeling have been demonstrated to be viable and less instrusive, there have not yet been any plant-wide applications of these approaches.

9. Costs and Benefits

Current regulatory guidelines do not offer sufficient incentives to facility operators to invest resources to estimate holdup. Because various nuclear fuel cycle facilities contain materials of various attractiveness, the safeguards and resource requirements do differ considerably across the nuclear materials production complex. Large-scale resource investments to minimize holdup and improve safeguards at fuel cycle facilities handling materials of low attractiveness would not be prudent. At the same time, it would be a good investment to spend adequate resources to minimize holdup and improve safeguards by addressing all the issues discussed in this paper, including a scheduled, planned shut-down for a complete cleanout inventory.

10. Research Efforts to Address Specific Holdup Problems

In the nuclear fuel cycle, there are very few areas where holdup of SNM is not a problem. However, the issues are of great concern when the materials are in chemically pure and isotopically enriched form due to attractiveness of materials as well as their potential for causing criticality accidents. Presently, there are several on-going projects in the U.S. to refurbish and renovate aging production facilities as well as to build new ones. These projects offer unique opportunities to incorporate state of the art technologies that would minimize holdup problems and improve overall efficiency of materials production. In areas of holdup measurement, there is a crying need to develop specially designed monitoring equipment and calibration standards to meet the special needs of holdup measurement. Indirect measurement capabilities, innovative calibration techniques, portable assay equipment, and proper personnel training can go a long way in alleviating holdup-related problems at SNM production facilities.

III. SUMMARY

There is a growing recognition that holdup of SNM in processing facilities is deleterious not only to safety but to the safeguarding of such materials. Both facility operators and regulatory agencies are beginning to address the complex issues that have been kept in abeyance for a long time. Presently, there are new opportunities and challenges to develop and apply new processes, equipment designs and materials, innovative plant layouts, and specially designed radiation measurement instruments and calibration standards. Open discussion of issues identified here is an essential step in addressing holdup-related issues rationally and to develop satisfactory solutions to a problem that has plagued the nuclear process industry for the past four decades.

IV. REFERENCES

1. K.K.S. Pillay, "A Survey of Open Literature Publications on Process Holdup of Special Nuclear Materials," LA-UR-87-4185, JNMM Vol. 16 No. 4 (July 1988), (INMM, Northbrook, IL, March 1988).
2. Regulatory Guide 5.8, "Design Considerations for Minimizing Residual Holdup of Special Nuclear Material in Drying and Fluidized Bed Operations," US Atomic Energy Commission (May 1974).
3. Regulatory Guide 5.25, "Design Considerations for Minimizing Residual Holdup of Special Nuclear Material in Equipment for Wet Processing Operations," US Atomic Energy Commission (June 1974).
4. Regulatory Guide 5.42, "Design Considerations for Minimizing Residual Holdup of Special Nuclear Material in Equipment for Dry Process Operations," US Atomic Energy Commission (January 1975).
5. "Semiannual Report on Strategic Special Nuclear Material Inventory Difference," DOE/DP-001/17 (January 1986).
6. K.K.S. Pillay, "Tracer Techniques in Estimating Nuclear Materials Holdup," J. Radioanal. Nucl. Chem (1988) (to be published).
7. S.F. Marsh, "Nondestructive Assay of Plutonium Residue in Horizontal Storage Tanks," in "Analytical Chemistry Instrumentation," Ed. W. R. Laing, Lewis Publishers, New York, NY, 91 (1986).
8. K.K.S. Pillay, R.R. Picard, and R.S. Marshall, "Estimation Methods for Process Holdup of Special Nuclear Materials," LA-10038, Los Alamos National Laboratory, Los Alamos, NM (June 1984).

Dr. K.K.S. Pillay is a member of the staff of the Nuclear Technology and Engineering Division of the Los Alamos National Laboratory. He received his Ph.D. in Nuclear and Radiochemistry from Penn State. His present interests are in safeguards system design, process facility safety, safeguards education, and nuclear materials production limitations for arms control.

His previous work experiences include his association with the Argonne National Laboratory, the Nuclear Research Center at the State University of New York at Buffalo, and the Pennsylvania State University.

Dr. Pillay is a Fellow of the American Nuclear Society and the American Institute of Chemists. He is an active member of several other professional organizations, including INMM, ACS, HPS, the NY Academy of Sciences, and the American Association for the Advancement of Science.

An Overview of Holdup Measurement Technology

Martin S. Zucker
Brookhaven National Laboratory
Department of Nuclear Energy
Upton, New York U.S.A.

ABSTRACT

The origin of holdup, where it occurs in plants, and the methods of assay thus far found useful under conditions often found in nuclear fuel manufacturing installations are discussed.

INTRODUCTION

"Holdup" refers to special nuclear material (SNM) unintentionally deposited in or on process equipment or plant structure. Monitoring and assay of holdup is necessary for criticality control, calculating material balances, to gauge manufacturing efficiency, and to protect the environment, as well as for safeguards. Aside from answering the basic question of how much material is held up and where it is located, the purpose of holdup assay may also be to determine the ease or difficulty of recovery. This information can have an appreciable financial impact on the facility, either because of the intrinsic value of the material, or because of the extent of security required for a decommissioned plant.

Holdup arises through imperfections in the manufacturing process, accidents or failures of equipment, and deficiencies in plant and process equipment design. Since nuclear fuel manufacturing involves finely divided powders, reactive liquid and gaseous chemicals, and grinding and sintering processes, the generation of holdup in plant and process equipment is inevitable, and assaying for it, and recovery where practical, has to be considered a part of the normal manufacturing process.

It is quite apparent in retrospect that many if not all existing manufacturing plants and process equipment could have been designed in ways that would have significantly reduced the generation of holdup and made it easier to assay and recover such holdup as did occur.

Setting up an effective measurement campaign or procedure for holdup assay first involves some study of the particular plant manufacturing process, including equipment, floor layout, material handling procedures and flow patterns, and the history of recent and past operations, since these provide clues as to where to look most effectively for holdup and what techniques will be useful. In addition to plant documentation, interviews with key personnel (foremen, health physicists, supervisors, etc.) can be most helpful in determining the best allocation of resources, manpower, and time.

Certain holdup producing situations seem to occur regularly in the nuclear fuel manufacturing industry, for example:

- It must be taken for granted that any process equipment containing liquids will leak on some occasion and perhaps chronically. This includes in particular, pumps and pump shaft packings, valves, and even the catch basins (supposed to help contain the leaks) which sometimes overflow.
- Nuclear material will often accumulate in ductwork supplying a negative pressure to glove boxes or other process equipment in places where there are abrupt changes in direction or cross section, or simply because airflow rate is insufficient to keep particles in suspension. On the other hand, too high an airflow rate is often the cause of dust particles being swept out of process equipment and brought into the ductwork.
- Cleaning up the inevitable accidental liquid or solid spills on the concrete floors and concrete block walls typical of plant construction has a tendency to drive some fraction of the material into this porous media. Repeated cleanup operations tend to diffuse new material into the media and cause further penetration of the old material.
- Any process involving sintered material being transported in open trays on a conveyer belt in a tunnel between process equipment seems inevitably to give rise to particulates ranging from dust to pellet fragments which deposit in the tunnel.
- Blending and grinding operations by their nature produce large amounts of SNM dust and fragments. For some reason sintering furnaces also do.
- Chemical process equipment and piping accumulates deposits or plating out of nuclear materials that cannot be completely flushed out even using powerful reagents. Repeated flushing of process lines sometimes

causes leaks, giving rise to holdup in another form.
- Air filter holders, either because of occassional failures of filters or during the process of changing them, material is released, also allow accumulations to occur.

A feature of holdup assay that is different from other types of nuclear material assay is that the more extensively material is looked for the more material is uncovered. Therefore, there is a built in bias in that the assay results in a lower limit estimate of the total amount actually present. At some point, a judgement has to be made that any increase in measurement effort will involve an unacceptable rise in cost.

The value of the recoverable portion of the material uncovered should bear some relation to the cost of the assay. However, it does seem that in some situations the criterion of cost effectiveness is overruled by other considerations: social, political, or security.

Holdup assay has involved detection of characteristic radiation, usually gamma rays, sometimes neutrons, and (more rarely) alpha particles and x-rays.

Holdup measurements are made difficult by several factors: The measurements are usually made under adverse environmental conditions such as extremes of temperature and humidity, noise, and contaminated surroundings requiring uncomfortable protective clothing and occasionally respirators. These poor conditions make it difficult for personnel to perform efficiently and effectively, and put stress on the radiation detection instrumentation.

Holdup material is usually distributed with a very low density, hence count rates and therefore statistical precision is poor. In the relatively few cases where there are sizeable concentrations of material, self-absorption of the characteristic radiation leads to uncertainties. In either situation, there is usually poor control over the measurement geometry. Often there is interference from sources other than the one being assayed.

Relating the apparent intensity of the characteristic radiation to the amount of special nuclear material present is not always straightforward and may even be misleading. Uncertainties in chemical composition, which influence (α, n) neutron production also impact on using neutrons as a signature. Calculation for geometry and allowing for attenuation due to walls, pipes, etc. interposed between the nuclear material and the detector is another source of uncertainty.

For the above reasons, assay of holdup is inherently less accurate and precise than other types of nuclear measurements. The most careful nuclear physics or nuclear chemistry measurements are often in the 0.1 to 1% range of precision and accuracy. Non-destructive assays of nuclear material that is well characterized will often fall in the one to 10% range. Holdup measurements in contrast will only rarely fall into the few percent range; more likely they will be of the order of 25 to 50% in precision and accuracy.

Even in those cases where the holdup in a plant is distributed in a very diffuse way, the total amounts of material may be important by any of the usually applied criteria; financial, safeguards, or environmental. Therefore, even though the measurements lack precision and accuracy by usual standards, the potentially significant amounts of material involved require that one strive for an unbiased estimate.

INSTRUMENTS

Only the simplest, most portable types of instruments have proven useful under the harsh conditions under which most holdup measurements are made. Three particular electronic packages, used with a variety of detector heads, have been proven useful.

The earliest was the Eberline Corporation's SAM[1] unit, adapted by that company from a health physics instrument at the request of the Los Alamos National Laboratory safeguards group, which then pioneered in developing safeguards applications using it. This instrument, basically, is a portable two channel analyzer, complete with bias supply for a detector, amplifier for nuclear detector pulses, and a scaler readout. It could be battery powered. An important feature is the use of a NaI-photomultiplier detector head with an internal Am^{241} alpha source to provide a source of reference pulses to stabilize the gain of the instrument. This stabilization is essential for NaI detectors, especially those used under the harsh conditions met in portable, field operations.

Limitations in this instrument led to the development at Brookhaven National Laboratory of an improved instrument. This battery operated portable and ruggedized unit improved on the stabilization scheme of the Eberline SAM and has many automatic features, including precalibration for nuclides of interest (^{235}U, Pu, etc.), self-checking features and a built-in programmable calculator for immediate data reduction, e.g., statistics and enrichment.

The third of these instrumentation packages is a miniature battery-operated, microprocessor-driven multichannel analyzer. The original concept was by a Los Alamos health physics group. Their design was improved on by the Los Alamos safeguards group and is available commercially. The instrument incorporates the BNL NaI stabilization scheme. The microprocessor, using Los Alamos-developed software tailored for safeguards applications, together with the flexibility afforded by the multichannel analyzer capability, makes this portable instrument (weighing roughly the same as either of the previous two instruments) the most versatile and powerful electronics package for holdup measurement. Despite being push-button operated and menu driven, it is however more difficult to use than either of the previous devices under field conditions and requires considerably more expertise and training of the operator.

While the usual application of these three electronic packages is with NaI detectors, each can also be used with other detector heads. There are two of particular interest. One is a neutron detector using a He^3 proportional counter and shielding arranged to produce a directional effect in the sensivity. Other arrays using BF_3 or He^3 proportional counters can be custom made to suit particular applications and can be used with each of these electronic packages. The second is a hand-held instrinsic germanium diode gamma detector. The higher resolution of these de-

tectors compared to NaI enables them to resolve interfering gamma lines. Such detectors are now commercially available and will work with either the BSAM or the mini MCA.

CALIBRATING GAMMA RAY INSTRUMENTS

In assaying holdup under field conditions, it is of course necessary to relate the intensity of radiation received from an object to the amount of special nuclear material contained in or on the object. To accomplish this it is sometimes necessary to idealize the object's geometry as a point, line, or area source of radiation. For examples, a glove box wall can be idealized as nuclear material distributed over an area; a pipe would be idealized as a linear source.

Having idealized these objects, one then has the choice of relating the response of the detector to such an object mathematically or experimentally.

Neglecting absorption, and to the extent that the detector can be considered a point (i.e., its dimensions being small compared to other distances involved in the measurement geometry) the radiation from a point source will vary inversely as the square of the distance between detector and source, from a line source inversely as the distance, while from a plane, it would be ideally independent of the distance. (This assumes the line and plane dimensions are large compared to the distance between detector and object).

An alternative procedure, preferred because it can be made quite realistic and not dependent on idealizations, is to use synthetic point, line, or area sources to calibrate the detector.

A point source of ^{235}U radiation is often simulated using a small disk of high-enriched uranium metal, laminated between thin plastic sheets to make it robust and contamination-free. Self absorption must be allowed for in relating the amount of ^{235}U to the instrument response. An encapsulated pellet of PuO_2 of known isotopic composition, with similar allowances for self absorption, can be used for Pu.

The effect of a line source on the detector can be synthesized by displacing the point source along a line perpendicular to the detector axis and summing the contributions. The response to a uniformly dispersed area source can be simulated in the same manner, by measuring the response to a point source at successively larger distances orthoganal to the detector axis and multiplying each of the readings by a weighting factor which is proportional to the area of the annular segment of the surface in which it is located. Alternatively, an area source may be fabricated in the following manner:

Sheets of transparent plastic with adhesive on one side are commercially available. The adhesive is protected with paper which can be peeled off. This protective paper is scored with the dimensions of the desired area source, and this area is peeled off leaving a protected border. Then (for a ^{235}U source) powdered UO_2 is sprinkled and brushed uniformly over the exposed sticky area. The protected border is then peeled off and another sheet of transparent plastic, sticky side down, is placed over the first, sealing the UO_2 powder between two sheets of plastic and within a frame free of UO_2. This transparent sandwich can then be visually examined for uniformity.

Weight measurements of the plastic sheet and paper backing, before and after loading the sticky side with UO_2, gives the weight of deposited SNM material per unit area. This area source exposed to the detector at a distance such that the detector cannot see the source edges, can then be used to calibrate the response of the instrument in counts per second per gram ^{235}U per unit area.

EXAMPLES OF ENRICHED URANIUM HOLDUP MEASUREMENTS

The concrete floors and cement block walls in areas of nuclear fuel manufacturing plants that have been intensively utilized, particularly for chemical processing, will often become impregnated with nuclear material. While the radiation levels may be low enough from the point of view of safety, the contaminated materials may need to be recovered or guarded when the building is decommissioned.

Plant history and a preliminary radiation survey should indicate which sections of the floors or walls deserve attention. Then a grid is down on the surface to be surveyed with chalk, for example, and the cordinates inscribed. The grid spacings should relate to the anticipated concentration of SNM. Typically they range from one to three feet on a side.

Floor assays can be accomplished with the aid of one of the portable detector-instrumentation packages described above, mounted on a wheeled device resembling a golf bag cart (the "floor sweeper"). The collimated detector (typically a 3/4" by 3/4" NaI) is printed to the floor, a fixed distance above it. From one to five separate counts are taken depending on the apparent intensity and variability within the grid square, and the results noted on a floor map. The detector is calibrated by noting the response to an area source placed on a clean area of the floor.

Guided by the results of the floor sweeper survey, drill dust samples (made with a rotary hammer drill with carbide bit) are taken, again at one to five locations within the grid square. Hole depths are typically three to five inches, based on the apparent radioactivity encountered, measured as described below. It has been found possible to recover 98% to 99% of the powder generated in the drilling process. This is done with the aid of a small ($\approx$ 1 foot diameter) cyclone separator operating from a small shop vacuum cleaner. The cement powder is sucked out of the hole, enters the separator and then falls down through the bottom of the separator into plastic vials. The vacuum cleaner exit is equipped with absolute filters. A motor driven eccentric weight sets up a vibration in the separator, which prevents the powder from sticking to the walls. The vials, labelled with the grid coordinates where the samples originated, are assayed in a NaI well counter. They may also be analyzed by a wet chemistry procedure and, if desired by mass spectroscopy.

Thus in principle, a correlation can be established between the "floor sweeper," the gamma assayed drill samples, and the chemically recovered material. In practice,

the correlation between the NaI well counter assay and chemical/mass spectrographic assay of the recovered dust samples is fair to good, while the correlation of floor sweeper results with the other two is usually poor, because of the poor control of geometry and the variable and uncertain amounts of absorption of radiation by the cement matrix. Nevertheless, the floor sweeper is still valuable because it shows where the more accurate but more laborious assay procedures should be carried out and thereby indicates what the proper allocation of time and effort should be.

Plant walls are typically of hollow, cement or cinder, block construction. It is therefore difficult to drill into them to recover material, and nuclear material on one side of the wall will interfere with assay of the other side. Fortunately, with some noteworthy exceptions, walls and ceilings hold much less material than floors. Usually, assays of walls and ceilings are done with a device analogous to the floor sweeper, descriptively termed "the counter telescope." This is a large (3" × 5") collimated NaI detector that can be aimed at any angle from horizontal to vertical and is calibrated using area sources.

Duct work often contains holdup. The value of this material, the fact that it is in an easily recoverable form and that significant amounts are often involved, makes it particularly important to calibrate detectors accurately. This is often done using mockups of ductwork containing nuclear material. Samples of the ductwork identical or similar to that used in the plant are obtained with the cooperation of plant engineering or maintenance personnel who seem invariably to have samples or "leftovers" of the ductwork used in fabricating the process line. The inner surfaces of sections of this duct can be covered with area standard sources, to calibrate the detector for the actual measurement geometry, including attenuation.

In assaying the ductwork, it is often necessary to place a lead sheet of flexible lead blanket over the far outside of the duct in order to prevent radiation from other sources from interfering with the assay. At a particular point along the length of the duct, assays are taken at four mutually perpendicular or opposite directions to compensate for unequal distribution of material over the inside duct surface. Assay results are noted on a map of the ductwork according to position. The positions should be chosen at intervals consistent with the apparent intensity of the radiation, except for places where the ductwork abruptly changes direction or cross section. These should be treated separately, since unusually large concentrations are often found there.

Glove boxes ranging from desk-sized to multistoried structures form another important holdup source, from the equipment within and on the inner surfaces of the box structure. Prior to assay, window ports should be specially cleaned or replaced if possible, and gloves replaced. Then the detector can view the opposite walls or objects within the glove box through the clean windows and the detector can be inserted into the gloves while keeping it in clean surroundings.

Objects inside the glove box are idealized as point, line, or area sources and are assayed using the respective detector calibrations. Where necessary, attenuations are calculated from knowledge of the material and thickness of the objects being viewed. Calibrations are also possible *in situ* by introducing (bagged) standard sources. Interference from neighboring objects is prevented using bagged-in shielding, or shields placed outside the opposite wall. As in the case of floor or duct assay, it is helpful to have a sketch of the box and its contents on which the various assay results can be noted and tabulated.

Furnaces and ovens are places where holdup is often found in significant quantities. With these objects there is the additional problem that their walls are so thick that calculations of attenuation are hopelessly inaccurate. In these cases, it has been found possible to send a bagged detector without collimation into the cooled-down furnace at the end of a suitable long cable, carried on the conveyor belt used to feed the furnace.

MEASUREMENT OF PLUTONIUM HOLDUP

Plutonium isotopes emit gamma-rays of many energies, some of which are considerably more penetrating than the 186 keV gamma ray of U-235. However, the isotopic composition is variable and it changes as Pu-241 decays to Am-241. The calibration methods described above may be used, though multiple enclosures for sources are advisable because of the much greater radio-toxicity of plutonium.

There is however an additional possibility for plutonium assay in that neutrons emitted by spontaneous fission are in principle also available for assay. A serious complication, however, is that neutrons are produced by α, n reactions with light element impurities as well as by spontaneous fission of the even isotopes. The plutonium isotopic ratios and the light element concentrations may not be well established. There is also the problem that the detection efficiency for neutrons is dependent on the neutron energy, which in turn is strongly influenced by the presence of moderators. This effect is difficult to compute in the complicated geometries found in holdup measurement situations. It is also difficult (compared to the corresponding gamma calculations) to calculate neutron absorption.

However, neutrons do tend to be more penetrating than gammas, and this makes them useful at least as an auxiliary assay signiture. By the same token, it is harder to collimate neutron detectors or to shield them against other sources near the one being assayed.

Deliberately non-directional neutron detectors[2] with flat energy response have been used to sample the total neutron production in a given room, with the hope that this constitutes a signature for the total amount of plutonium holdup in the room and its equipment. The measurements are often interpreted using computer code calculations of what the response of the detector should be. Aside from uncertainties in the relative (α, n) *versus* fission neutron contributions, the assumption is being made that the room is isolated from neighboring areas, a condition which is valid only if solid concrete (not cinder or cement block) walls are the room boundaries.

CONCLUSION

The picture of holdup assay as presented above should be seen as a conglomerate of relatively simple common-sense procedures involving only low technology instrumentation and a will to do the best job under often highly adverse circumstances.

The techniques described above were developed by industry, the national laboratories, and the regulatory agencies, who have all cooperated in this difficult area of nuclear technology. Though the views expressed are my own, my thanks and acknowledgements go to my colleagues and coworkers from all these sectors of the nuclear field.

*Research carried out under the auspices of the U.S. Department of Energy Under Contract No. DE-AC02-76CH00016.

REFERENCES

1. Eberline Instrument Co., Santa Fe, New Mexico.
2. Tape, S.W., et al: Total Room Holdup of Plutonium Measured with a Large Area Detector, Proc. 17th Annual Meeting, INMM, Vol. V, No. III, Jour. INMM, pp. 533-539, 1976.
3. Further references may be found in the excellent compilation by K.K.S. Pillay in the proceedings of this meeting.

Martin S. Zucker is formally educated in nuclear physics with a background in instrumentation and applied mathematics. Dr. Zucker has been at Brookhaven National Laboratory since 1958 in a variety of positions in physics and nuclear science and technology, and engages in private consulting work for the nuclear industry.

Total Neutron-Counting Plutonium Inventory Measurement Systems (PIMS) — and their Potential Application to Near Real Time Materials Accountancy (NRTMA)

I. Driscall, G.H. Fox, C.H. Orr, and K.R. Whitehouse
Physical Science and Engineering Development Group
British Nuclear Fuels plc (BNFL)
Sellafield, UK

ABSTRACT

A radiometric method of determining the inventory of an operating plutonium plant is described. An array of total neutron counters distributed across the plant is used to estimate hold-up at each plant item. Corrections for the sensitivity of detectors to plutonium in adjacent plant items are achieved through a matrix approach.

This paper describes our experience in design, calibration and operation of a Plutonium Inventory Measurement System (PIMS) on an oxalate precipitation plutonium finishing line. Data from a recent trial of Near-Real-Time Materials Accounting (NRTMA) using the PIMS are presented and used to illustrate its present performance and problem areas. The reader is asked to consider what role PIMS might have in future accountancy systems.

PHYSICAL PRINCIPLES

1. The penetrating nature of neutrons makes them less susceptible than gamma rays to attenuation in bulk fissile materials or glovebox construction materials. Inventory measurements in a plutonium plant using total neutron counting (TNC) are therefore less likely to underestimate the inventory than gamma counting. TNC also has the advantage of simplicity, but is subject to the following restrictions:

(i) the isotopic and chemical composition of material at all points in the plant must be well characterized and

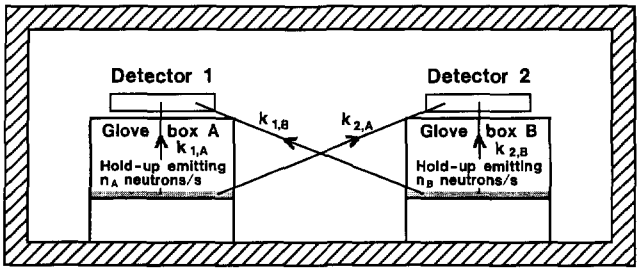
(ii) correction for multiplication effects in bulk product material is required.

2. Neutrons are emitted from plutonium with energies of 1-3 MeV and will undergo many interactions with their surroundings, particularly concrete building materials, before becoming thermalised and absorbed. After a small number of interactions, the point of origin of a neutron is lost. The ideal plutonium inventory measurement system (PIMS) based on Total Neutron Counting would use detectors sensitive only to 1-3 MeV neutrons so that maximum information on the mass distribution of hold-up would be obtained. In practice, such detectors would be unacceptably bulky, and smaller devices with an intermediate energy response must be accepted, together with a means of correctly assigning detected neutrons to the originating sources. We have developed a simple matrix approach (Fig. 1) to achieve this. The counting rate from a detector is the sum of the response of that detector to all sources for which it has significant sensitivity. Provided that all such sensitivity factors can be measured, the matrix equation is easily solved for the neutron emission from each plant area. Correction for multiplication effects, and division by the specific neutron emission appropriate to the chemical and isotopic form of material at the point monitored, gives plutonium mass directly.

3. The performance of a PIMS is determined by the care with which individual detectors are sited. Criteria for siting should be to:

(i) maximise detection efficiency to the target item, while minimising efficiencies to other nearby items,

(ii) minimise efficiency variation for possible plutonium distributions in the target item, and

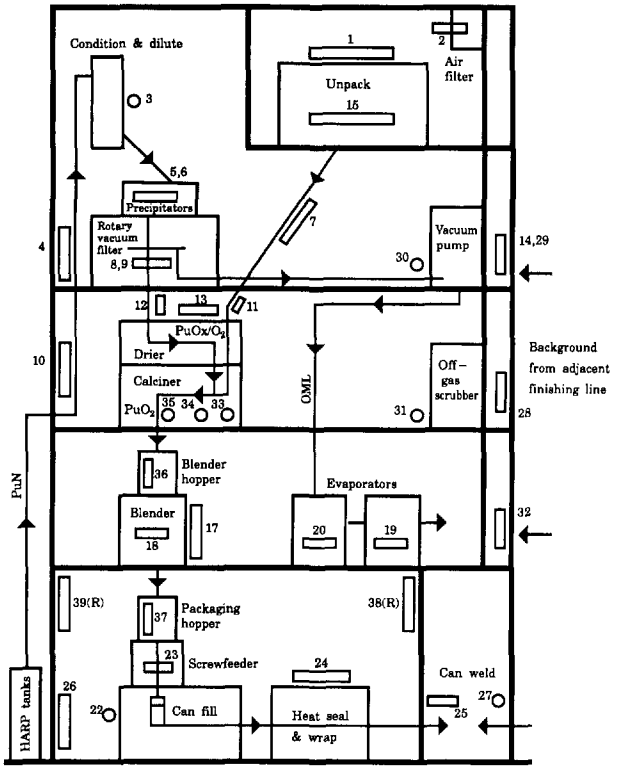


Countrate $C_1 = k_{1,A} \cdot n_A + k_{1,B} \cdot n_B$
$C_2 = k_{2,A} \cdot n_A + k_{2,B} \cdot n_B$

Where the $k_{1,A}$ is the sensitivity (detection efficiency) of detector 1 to hold-up in glovebox A etc.

or $\begin{bmatrix} C_1 \\ C_2 \end{bmatrix} = \begin{bmatrix} k_{1,A} & k_{1,B} \\ k_{2,A} & k_{2,B} \end{bmatrix} \begin{bmatrix} n_A \\ n_B \end{bmatrix}$

Figure 1. Matrix approach



Detectors are numbered 1–39 (with some gaps)
(R) signifies thermal neutron room-monitors
PuN – plutonium nitrate, PuOx – plutonium oxalate, PuO_2 – plutonium oxide
OML – oxalate mother liquor
Can weld area is shared with adjacent finishing line

Figure 2. PIMS experimental system

(iii) consider the maximum countrates likely and any resulting deadtime losses.

4. In designing a PIMS, considerable effort should be put into experimentally simulating source-detector geometries to achieve objectives (3.i) and (3.ii). Relationships to all concrete surfaces and other moderators or reflectors should be accurately simulated, but steelwork can be ignored. It is, however, impractical to simulate interactions with nearby sources accurately — instead, more limited experimental work and available data on, for example, neutron attentuation by concrete has to be used. Theoretical modelling also has been of some use for this purpose.

5. Once detectors are installed, the system must be calibrated before the plant goes active to enable accurate detector sensitivities to be determined. The calibration source used must be physically small for accurate positioning and easy access inside plant items, but with a high neutron emission to minimise count times. Ideally, a plutonium source of characteristics similar to the hold-up expected at the particular calibration point should be used. All possible hold-up positions must be covered by this point calibration exercise, and the data must be combined in such a way that the expected normal plutonium distribution is closely simulated. The importance of minimising efficiency variations within a target item is clear.

6. It is important to define detector positions correctly at the outset, as subsequent modification of detector positions will involve recalibration under active plant conditions. Definition at an early stage in plant design also allows provision for re-entrant tubes to mount detectors if this is desirable.

EXPERIMENTAL PIMS

1. Our experimental system is installed on an operating plutonium finishing line. This consists of a glovebox line arranged vertically in five concrete cells as shown in Fig. 2. Plutonium nitrate is pumped to the top of the plant, then oxalic acid is added to precipitate plutonium oxalate, which is filtered off, decomposed to oxide in a drier furnace, calcined, blended (optional), and packed in all-welded steel cans.

2. The locations of PIMS detectors are also shown in Fig. 2. There are a total of 29 in-process detectors, mostly single detectors monitoring individual gloveboxes. Limited use of 2- or 3-detector arrays is made to give much reduced efficiency variation in key areas (e.g., detectors 33-35 under the calciner). All but two of the in-process detectors are 50-mm diameter boron trifluoride (BF_3) neutron counters enclosed in a 25-mm thick polythene moderator annulus, which itself is enclosed in a 0.5-mm cadmium neutron shield. This package has a maximum efficiency at 10 eV (3-4 times the efficiency at 1 MeV) and the efficiency falls off rapidly below 0.5 eV. The other two in-process detectors (detectors 36 and 37) are 25-mm -diameter BF_3 counters located within the 300-mm thick neutron shielding that surrounds the two bulk hoppers. A 0.5-mm cadmium shield is positioned over the center of the detectors to flatten the response variation over the anticipated vertical height of product material.

3. Two more 50-mm BF_3 counters (detectors 38 and 39) are mounted bare in the packaging cell. These room-monitors measure the thermal flux in the cell and will ultimately be used to crosscheck the inventory determined from the in-process detectors.

4. A particular difficulty with the experimental PIMS is its proximity to a second plutonium finishing line. Corrections are made for varying backgrounds from this source. A total of 6 detectors (14, 26, 27, 28, 29, and 32), identical in design to the in-process detectors, are provided for this purpose. Background corrections over most of the plant are small, and are based on a ratio correction that assumes the background detectors to be sensitive only to the interfering plant (i.e., a one-way interaction). In the packaging area, background effects are more significant — full calibration of the interfering plant was therefore carried out, and detector 27 was included in the matrix so that two-way correction is available.

5. Data outputs from the experimental system are displayed directly as countrates and archived on a data-logger. Countrate information has proved invaluable to the plant operators for process monitoring and control and in providing early warning of blockage problems. Processing the data to produce the plutonium-mass distribution is done off-line at present, but work is underway to enable all data processing to be carried out on-line, giving the plant operators direct readout of plutonium-mass-distribution on a whole-plant, individual cell, or plant item basis.

Table 1
PIMS inventory data (selected)

Day	1	2	3	·····	14	15	16
Whole Plant	84.1	62.9	103.0		98.0	95.3	101.7
Cell Totals							
Unpack	7.2	7.3	7.1		7.2	7.2	7.4
RVF & precip.	10.7	14.0	14.0		17.0	17.3	15.4
Furnace	21.5	16.1	21.0		24.2	23.6	25.0
Blender	3.5	4.7	2.9		2.8	3.1	3.1
Packaging	41.3	20.8	57.8		46.8	44.0	50.9
Packaging Cell							
Packaging hopper	26.8	0.4	28.7		34.0	14.6	20.9
Dispense screwfeeder	16.2	-2.0	10.7		14.9	14.9	15.9
Dispense head	-1.5	24.0	20.1		-2.0	10.9	4.9
Heat seal box	-0.2	-2.0	-2.0		-0.1	4.3	11.8
Can weld area	0.0	0.4	0.4		0.0	-0.8	-2.5

Data normalized to average plant stock of 100

PIMS PERFORMANCE

1. The calibration matrix for any PIMS is diagonal in nature because detectors are predominantly sensitive to items in their own cell area and in cells immediately adjacent. The leading diagonal efficiencies should be the maximum terms in their row and column. Figures of merit that measure the strength of the leading diagonal can be used as indicators of the quality of system design. Leading diagonal terms are typically 5 to 10 times larger than their nearest-neighbour off-diagonal terms, although in some areas factors of only 2 times are obtained.

2. Results of a recent trial of near-real-time materials accountancy (NRTMA) using the PIMS giving a good idea of its potential. PIMS assessments were carried out daily at midnight, with the plant operating but with no product can movements during the measurement. Taking the PIMS assessment for the previous day as an opening stock, a closing stock can be calculated daily from receipts and issues data obtained by conventional means. This can then be compared with the day's PIMS assessment to derive a material unaccounted for (MUF) (or inventory difference (ID) in the normal way.

3. Table 1 shows some of the data obtained from a 16-day trial. Data have been normalised to an average plant stock over the trial of 100 units. PIMS data are shown for the whole plant, for individual cells, and for individual items within the packaging cell where the bulk of the inventory variation occurs.

4. The PIMS data within the packaging cell include some negative masses, which result from misallocation in the matrix approach. Although obviously undesirable, there is no justification for excluding these negative values from any summation of data for the cells and the whole plant. Whole-plant data have been validated against calculated book stocks for a period of a year from plant start-up, and the agreement obtained was typically ±10%.

5. Figure 3 plots MUF and CUMUF time-series for the NRTMA trial. The PIMS clearly follows changes in plant stock — Agreement is again typically within ±10% resulting in small daily MUFs. One instance of a large positive MUF followed by large negative MUF the next day is discounted as due to timing differences in data collection. Figure 4 shows a regression plot of the change in PIMS against (receipts-issues) for all available trial data. The calculated slope of $0.82 \pm 0.08 (2\sigma)$ suggests some residual bias in the PIMS which leads it to slightly under-estimate plant inventory.

PROBLEMS

1. A number of deficiencies are recognized in the experimental PIMS — in particular, because the decision to install it was taken at a very late stage in plant design, little

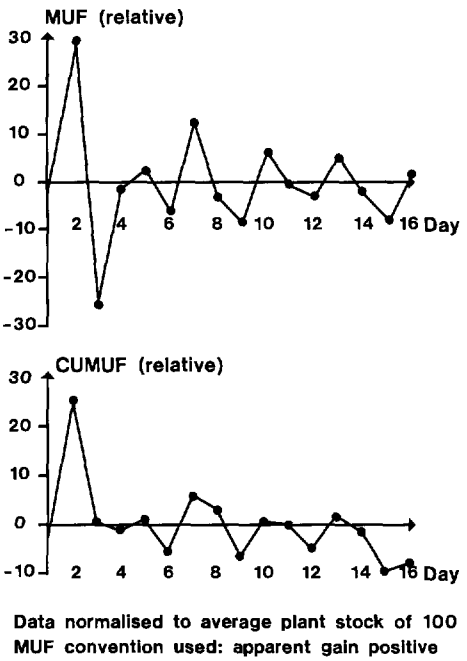


Figure 3. Results of 16-day NRTMA trial

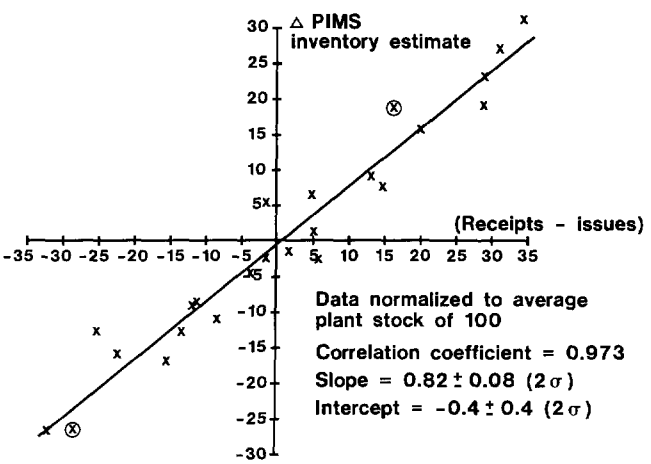


Figure 4. PIMS performance

optimization of detector positions was possible. This will contribute to bias in the results. Other potential sources of bias include:

(i) The combination of point-calibration data may be inappropriate for some plant states. In areas where discrete bulk source movements occur (e.g., packaging) it may be possible to monitor the plant state through its control systems and select a matrix appropriate to that plant state. This was done manually in our NRTMA trials.

(ii) The assumed isotopic and chemical compositions may not be appropriate at all points. The isotopic match may be particularly poor when the plant handles a wide range of isotopic compositions, especially if measurements are carried out at plant clean-out where it is not clear that hold-up derives from the last batch processed. High resolution gamma spectrometry is to be investigated as a means of obtaining periodic hold-up isotopics.

(iii) The feed pipe inventory is included in the book stock calculation but only partly in the PIMS assessment. This may give a small bias under continuous operating conditions.

(iv) Errors may be present in the initial point calibration due to position mismatch, spectral differences (if non-plutonium sources were used) or multiplication errors (if plutonium sources were used).

(v) Multiplication corrections may be needed in bulk sources. To date this has not been attempted other than to position hopper detectors in such a way as to minimise non-linear response. Non-linearity will have to be assessed by careful correlation of estimated plutonium mass from operational data with corrected neutron emission detected by the PIMS. Correction for non-linearity can then be done after matrix solution. Modelling multiplication using a neutronics code may also be helpful.

2. More fundamental problems of PIMS development yet to be tackled are:

(i) the elimination of negative hold-up masses by inclusion of extra neutron shielding between plant items or by compromising on detector dimensions.

(ii) error analysis — because the major error source is in the assumptions made as to the distribution of material, the starting point is to establish some limits on this. A probability distribution can then be defined covering these limits for each sensitivity term. By repeatedly sampling all the distributions to obtain new calibration matrices which are then solved, error bounds on each hold-up mass can be derived.

(iii) validation of the initial calibration and subsequent calibration checks — arrangements have yet to be discussed with inspection agencies as to how these could be tackled.

(iv) measurement control — this is limited at present to periodic standardization by manually attaching a standard source to each detector in turn. Measurement control algorithms will be incorporated to analyse this data. More frequent but less rigorous checks could also be done by periodically injecting into each channel a known signal and solving the matrix equation. This would test all parts of the system except the detectors and their associated cabling.

CONCLUSION

In a short paper such as this, it is only possible to give a flavour of this work. We feel the potential of the system has been clearly demonstrated, and are confident of further improvements in performance given greater emphasis on improved detector siting. Design work is in progress on a number of new plant installations in parallel with further development of the experimental system. We would ask that the reader consider the role of PIMS in future accountancy systems.

Ian Driscall graduated with a BSc (Hons) in Chemistry/Geochemistry from the University of Leicester in 1980. He then undertook research into radiometric methods of measuring ^{238}U-series disequilibrium in environmental samples and subsequently joined Research and Development Department, BNFL Sellafield in 1984. He has since worked on development of radiometric instrumentation for plutonium plants — principally total neutron counting systems for inventory determination.

George Fox graduated from the University of Durham in 1960 with a BSc (Hons) in Chemistry and Physics. He joined UKAEA as a radiometric physicist and subsequently transferred to BNFL. Since 1966, he has worked widely on development of special radiometric instrumentation for on-plant measurements at all stages of the Magnox and oxide reprocessing cycles.

Chris Orr graduated in 1974 from the University of Bath with a BSc (Hons) in Applied Physics. He joined BNFL as a radiometric physicist and has worked on the development of radiometric systems for measurement of plutonium and uranium products, residues and wastes. His principal area of interest involve application of high and low resolution gamma spectrometry, total and coincident neutron detection techniques.

Kevin Whitehouse graduated from the University of Birmingham in 1983 with a BSc (Hons) in Physics, specializing in nuclear and quantum physics. He has worked for two years in radiometric physics at BNFL Sellafield — principally on the experimental plutonium inventory measurement system.

NUMATH: A Nuclear Material Holdup Estimator

Alan M. Krichinsky
Oak Ridge National Laboratory
Oak Ridge, Tennessee U.S.A.

ABSTRACT

NUMATH is a computer program which provides near-real-time estimations of material compositions and inventories from previous inventory measurements, operating data, and available on-line process measurements — in a manner that is transparent to the computer terminal operator. In steady-state simulated-run testing, NUMATH produced estimates within 10% of the measured inventories for accountable materials.

INTRODUCTION

In response to heightened attention to nuclear material safeguards and the desirability of providing real-time nuclear material inventories, a modular FORTRAN computer program, NUMATH (*Nu*clear *Ma*terial *H*oldup Estimator),[1] has been developed to provide near-real-time estimations of material compositions and inventories from previous inventory measurements, operating data, and available on-line process measurements — in a manner that is transparent to the computer terminal operator. This program has been tested in the remotely operated ^{233}U Radiochemical Processing Plant (RPP) at Oak Ridge National Laboratory where, in addition to liquid and solids blending, the unit operations employed include solids dissolution, solvent extraction, ion exchange, evaporation, precipitation, centrifugation, and calcination.

Since the compositions are used for inventory estimation, the results are cataloged in container-oriented files[2] to represent material accumulated in applicable vessels — including consideration for material previously logged in these vessels. When an operator prompts a user-friendly, material-transfer monitoring program[2] to terminate the active use of any vessel in the process, NUMATH is automatically invoked to:

1. Collect pertinent data.
2. Estimate the present composition of material in every vessel (except source vessels, which maintain their material characteristics) involved in the unit operation based on each vessel's process measurements (i.e., volume and density), previous vessel compositions (calculated or, if available, analyzed), unit operation characteristics (e.g., flow rates, vessel connections, transfer characteristics), and selected models.
3. Store the estimates in the container files (i.e., the physical inventory) and other appropriate files in the system.

Records in which estimates are stored are marked as containing estimated compositions until samples are analyzed and the measured values (input via a user-friendly analytical data base access program[2]) supersede the estimates.

NUMATH DESCRIPTION

The inventories of individual vessels involved in a unit operation may be estimated based on those vessels' functions in the unit operation. Based on data from plant operating history and the degree of accuracy required for the estimates (within 10% of analyzed values for fissile and fertile materials), the assumption has been made that the models applicable to operations in the subject plant are weakly dependent on source material compositions and that resultant material compositions may, therefore, be calculated without iteration. For steady-rate operations, three basic models evolve from the above considerations: the blending model, the separation model, and the conversion model.

The blending model uses a perfect mixing algorithm to describe the combination of bulk quantities (Q^s in weight or volume units) of materials of different element (j) compositions (C_j^s in units of g/L or g/g) from several sources (s = 1, 2, 3, ... ,n). The element composition resulting in the subject vessel (v) may then be found from:

$$C_j^v = \frac{\sum^s Q^s C_j^s}{\sum^s Q^s} .$$

It should be noted that Q^s, the bulk quantity contributed from each source, may represent either the net quantity transferred or the bulk quantity flow rate depending on

the subject vessel's transfer characteristics. This feature is provided to permit composition estimates for material residing only briefly in process vessels (e.g., continuously operated, flow-through columns) in addition to composition estimates for material accumulating in process surge and storage vessels.

Isotopic contributions to the inventory are maintained in units of weight fraction. The weight fraction of the i^{th} isotope resulting in the subject vessel (F_i^s) may be calculated from the isotopic weight fraction (F_i^s) from the source s:

$$F_i^v = \frac{\sum^s Q^s C_j^s F_i^s}{\sum^s Q^s C_j^s} .$$

The separation model uses an equilibrium relationship, the "distribution law" equation,[3] to describe the resulting distribution of components between phases for processes in which equilibrium changes occur:

$$C_j^v = \frac{\sum^s Q^s D_j^s C_j^s}{\sum^s Q^s} .$$

where D_j^s is the distribution factor for element j from vessel s. It is important to note that material is accounted for by containers in a container data base (CDB)[2] and that multiple, immiscible phases (capable of bearing accountable quantities of nuclear material) exist as multiple containers in the CDB. (The isotopic weight fraction resulting from such material distribution, and for the following conversion model, have been developed in a manner similar to that for the blending model.)

The conversion model is used to describe the change in composition for processes in which completion or near-completion changes occur. This model is an impirical relation to adjust elemental compositions of material as it enters the reaction vessel:

$$C_j^v = A_j + \sum^s B_j^s C_j^s \frac{Q^s}{Q^v} ,$$

where A_j and B_j^s are constants describing element j composition adjustments. Notice that B_j^s permits a partial composition adjustment of element j from vessel s, while the ratio of quantities of Q^s and Q^v describes a dilution effect. For a flow mode, Q^v is replaced by Q^s.

The distribution and conversion factors may be empirically determined from process operating histories or from laboratory testing. These factors and the vessel interrelationships are obtained from a special ASCII process definition file[1] that catalogues transfer characteristics of every vessel involved in the process.

Estimated compositions resulting from the three basic models are subjected to a *specific gravity adjustment*. For any vessels having specific gravity (SpG) instrumentation, the estimated elemental compositions are adjusted to expected levels based on the measured SpG. This adjustment is determined by applying a correction factor to the estimated compositions, C_j^v.

adjusted composition = estimated composition • factor.

The empirical correction factor is determined by the ratio of an expected sum of compositions (based on the measured SpG) to the sum of compositions estimated by models mentioned above. This factor adjusts the compositions while preserving the molar ratios of the estimated species.

NUMATH is not restricted to the three aforementioned basic models, but may be provided with any number of user-defined vessel models or, perhaps, rigorous estimators tailored to specific unit operations.

NUMATH IMPLEMENTATION

The composition of material in vessels involved in any unit operation within the subject plant may be estimated by a combination of one or more of the basic models described above. This concept may be best illustrated using a block diagram of a simplified separation process (Fig. 1).

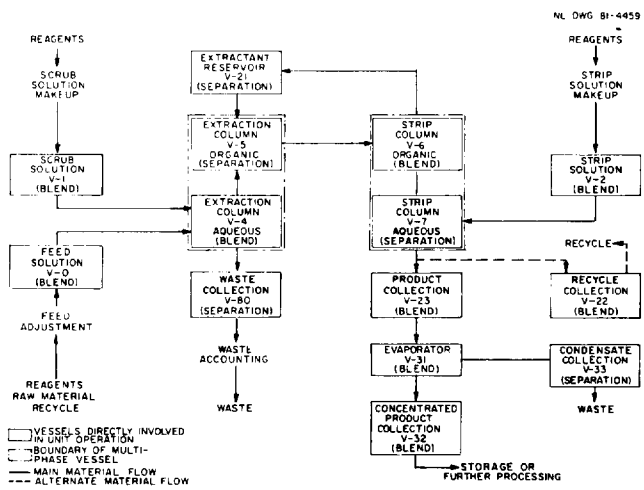


Figure 1. Block diagram of a single-cycle solvent extraction system.

This diagram depicts all vessels directly involved in a single-cycle solvent extraction separation system in which uranium is recovered from a uranium-thorium mixture. The models used to estimate the contents, based on source materials, are shown in parentheses in each block. To accommodate the aqueous and organic phases coexistent in the contacting columns, the extraction/scrubbing column and the strip column have been represented as two vessels each.

NUMATH SIMULATION RESULTS

Trial NUMATH estimations were performed by simulation of solvent extraction operations. Initial conditions (including feed compositions) were entered into applicable vessels, and the unit operations transfers were performed

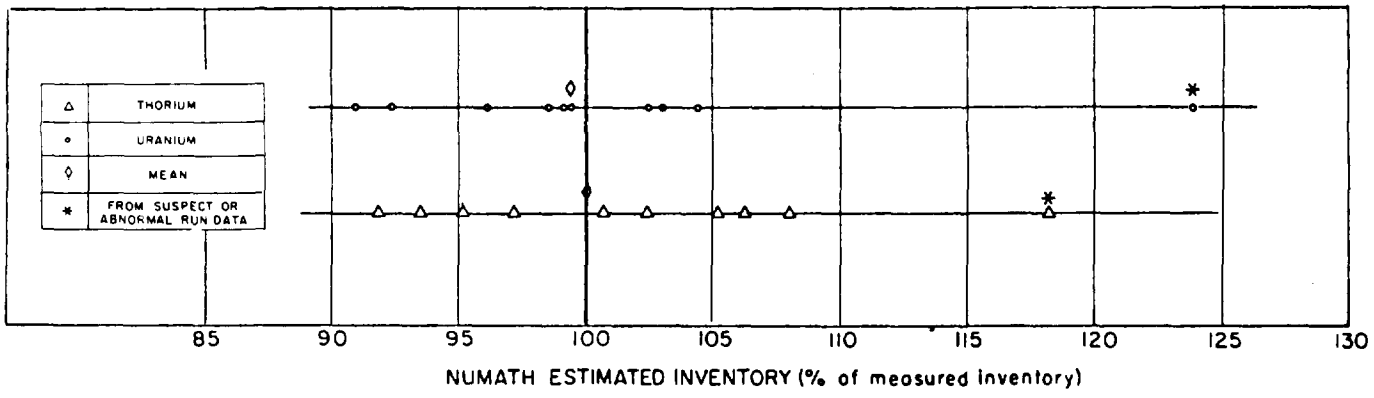


Figure 2. NUMATH performance relative to measured inventories for solvent extraction simulation.

on the computer as they would be for actual runs. NUMATH was automatically invoked by the transfer operations. Estimated composition data were accumulated and evaluated.

The overall performance of NUMATH may be best evaluated by how well its composition estimates yield final inventory totals in relation to the measured final inventory totals for each run (Fig. 2).

For the solvent extraction simulated runs, all but one of the estimated inventories were within 10% of the measured inventories for uranium and for thorium. One run exceeded the ± 10% criterion established earlier for fissile inventory estimates possibly due to the use of off-specification feed. (In such cases, thorium competes with uranium for the unused extraction sites.) The excessive thorium inventory in another run appeared to have been caused by a suspiciously high specific gravity measurement which factored into the NUMATH composition estimates.

CONCLUSIONS

The following conclusions may be drawn from this study with regard to estimation of nuclear material residing in process vessels during process operations:

1. NUMATH yielded inventory estimates within 12% of measured values for the accountable materials collected during a series of steady-state process operations using normal-range feed.
2. The estimated inventories appeared to be biased slightly low for uranium in the simulated solvent extraction runs. A refinement of model coefficients may reduce this bias.
3. Specific gravity instrument-calibration shifts may lead to erroneous estimates when factored into the NUMATH calculations.

REFERENCES

1. A.M. Krichinsky, NUMATH: *Nuclear material Holdup Estimator for Unit Operations and Chemical Processes*, ORNL/TM-8175 (February 1983).
2. J.A. McEvers, A.M. Krichinsky, L.R. Layman, T.H. Dunnigan, R.M. Tuft, and W.P. Murray, "Implementation of the Facility-Integrated Computer System (FICS) for Nuclear Material Accountability," *Proceedings of the 21st Annual Meeting of the Institute of Nuclear Materials Management*, 9: 680-87 (1980).
3. J.R. Welty, C. E. Wicks, and R.E. Wilson, *Fundamentals of Momentum, Heat, and Mass Transfer*, John Wiley & Sons, Inc., New York, 1969, p 565.

Alan B. Krichinsky is the Technical and Operating Group Leader at Chemical Technology Divisions's Radiochemical Processing Plant at the Oak Ridge National Laboratory and Project Manager for the upgrade of the Metallurgical Examination Hot Cells at the laboratory. Mr. Krichinsky earned his B.S. degree in chemical engineering from the University of Maryland in 1974 and his M.S. degree in chemical engineering from the University of Tennessee in 1981. He has been employed at Oak Ridge National Laboratory since 1974 and has been involved in design, construction, operation and evaluation of nuclear fuel plant processes. In addition, he currently serves as the Plant Safety Features section editor for Nuclear Safety and is involved in rewriting an ANSI standard on nuclear material control.

Literature Survey: A Survey of Open Literature On Process Holdup of Special Nuclear Materials

K.K.S. Pillay
Los Alamos National Laboratory
Los Alamos, New Mexico U.S.A.

The residual amount(s) of fissionable materials remaining in process equipment after the runout of bulk materials processed is referred to as "Process Holdup of Special Nuclear Materials." Locating regions of holdup and estimating the quantity of fissile materials remaining as holdup are important not only to materials accountability but also to process safety. Holdup is often referred to as a "Hidden Inventory." In materials accounting terminology, hidden inventories are part of "Materials Unaccounted For" (MUF) or "Inventory Difference" (ID). MUF or ID could be construed as "Loss" or "Diversion." From a safeguards perspective, all these designations are undesirable. Another terminology that is relevant to holdup is "In-Process Inventory." During process operations and temporary shutdown, the holdup within the facility is also known as the in-process inventory. Estimating this inventory is just as challenging as residuals after process runout.

The role of hidden inventories, or holdup, as a safeguards problem is now recognized by almost everyone interested in establishing effective safeguards for special nuclear materials. As part of this effort to organize the first INMM-sponsored Technical Workshop on Process Holdup of Special Nuclear Materials, an attempt was made to update an earlier survey of open literature publications of relevance to holdup.[1] The publications identified during this survey are listed below. The list includes only those publications that are generally available through the resources of a good technical library. An attempt was made to exclude from this list those documents recognized as internal documents, progress reports, preliminary reports, abstracts, etc.

The following list of publications is arranged in the order of their appearance in the open literature.

List of Publications

1. M. H. Ehinger, "Operating Experience with Pulsed Column Holdup Estimator," Nuclear Safeguards Technology 1986, Proceedings of an International Symposium on Nuclear Material Safeguards, Vol. I, 139-149, IAEA, Vienna, Austria (1987).

2. E. A. Griffin, J. K. Young, B. W. Smith, and R. J. Brouns, "Material Control and Accounting (MC&A) Loss Detection During Transition Periods and Process Upset Conditions," NUREG/CR-4695 (PNL 5890), Pacific Northwest Laboratory, Richland, Washington (March 1987).

3. P. A. Russo, J. K. Sprinkle, Jr., and T. H. Elmont, "Holdup Measurements of the Rocky Flats Plant 371 Precipitator Canyons," LA-10967-MS, Los Alamos National Laboratory, Los Alamos, NM (April 1987).

4. M. Canty, "Influence of Contactor and Evaporator Holdups on Detection Sensitivity," Nucl. Mater. Manage. XV(2), 17-22 (January 1987).

5. R. P. Gardner, M. N. Iqbal, and K. Verghese, "Dynamic Model for Radiotracer Determination of Solute Residence Time Distribution in Counter-Current, Pulse Column Solvent Extraction Process," Sep. Sci. Technol. 22(2/3), 269-280 (1987).

6. C. Bonner and R. S. Marshall, "The Use of Storage Tank Holdup Measurements to Reduce Inventory Differences in an Ion Exchange Process," Nucl. Mater. Manage. XV, 102-104 (June 1986).

7. D. E. Boyd, and R. R. Miller, "Characterization of Process Holdup Material at the Portsmouth Gaseous Diffusion Plant," Nucl. Mater. Manage. XV, 84-88 (June 1986).

8. R. E. Ellefson and J. T. Gill, "Tritium Inventory Differences: Sampling and Uranium Getter Pump Holdup," Nucl. Mater. Manage. XV, 89-93, (June 1986).

9. J. T. Gill, R. E. Ellefson, and W. M. Rutherford, "Tritium Inventory Differences: Molecular Sieve Holdup," Nucl. Mater. Manage. XV, 94-101 (June 1986).

10. K.K.S. Pillay and R. R. Picard, "Learning to Live with Holdup," Nucl. Mater. Manage. XV, 109-114 (June 1986).

11. R. R. Picard, and K. K. S. Pillay, "Statistical Sampling for Holdup Measurements," Trans. Am. Nucl. Soc. 53, 193-194 (1986).

12. T. H. Elmont and V. Maurello-Scott, "A Quantitative Estimate of Holdup in a Plutonium Processing Facility," Trans. Am. Nucl. Soc. 53, 194-195 (1986).

13. T. L. Hardt and M. P. Dedo, "Uranium Holdup in Concrete Floors: A Comparison of Nondestructive Methods," Trans. Am. Nucl. Soc. 53, 195 (1986).

14. M. S. Zucker, "The Assay of Holdup in Nuclear Plants," Trans. Am. Nucl. Soc. 53, 192 (1986).

15. S. F. Marsh, "Nondestructive Assay of Plutonium Residue in Horizontal Storage Tanks," in "Analytical Chemistry Instrumentation," Proceedings of the 28th Conference on Analytical Chemistry in Energy Technology, Knoxville, TN, Ed. W. R. Laing, Lewis Publishers, Inc., Chelsea, MI (October 1985).

16. P. A. Russo et al., "Automated Instruments for In-Line Accounting of Highly Enriched Uranium at the Oak Ridge Y-12 Plant," LA-10243-MS, Los Alamos National Laboratory, Los Alamos, New Mexico (1985).

17. D. A. Close, J. C. Pratt, and H. F. Atwater, "Development of Enrichment Measurement Technique and its Application to Enrichment Verification of Gaseous UF_6," Nucl. Inst. and Meth. in Physics Research A240, 398-405 (1985).

18. J. N. Cooley, L. W. Fields, and D. W. Swindle, Jr., "Results from Uranium Deposition Studies for Development of a Limited Frequency Unannounced Access Inspection Strategy for Gas Centrifuge Plants," Nucl. Mater. Manage. XIV(3), Proceedings Issue, 272-77 (1985).

19. K.K.S. Pillay, R. R. Picard, and R. S. Marshall, "Estimation Methods for Process Holdup of Special Nuclear Materials," NUREG/CR-3678, LA-10038, Los Alamos National Laboratory, Los Alamos, New Mexico (June 1984).

20. A. D. Turner and J. J. Dalton, "Review of the Application of Temporary and Permanent Coatings for the Reduction of Activity Hold-up and Increased Ease of Decontamination," AERE-R-11054, U.K. Atomic Energy Agency, Harwell, England (February 1984).

21. Regulatory Guide 5.23, "In-Situ Assay of Plutonium Residual Holdup," U.S. Atomic Energy Commission (May 1974) and proposed revision to Regulatory Guide 5.23, Rev. 1 (February 1984).

22. M.D.H. Amey and W. J. Symons, "Comparison of Nondestructive Techniques for Assay of Plutonium Wastes Arisings and 'Hold-up' in Glove Boxes," Proceedings of the Sixth Annual ESARDA Symposium on Safeguards and Nuclear Materials Management (European Safeguards Research and Development Association, Ispra, Italy, 1984), ESARDA 17, 359-363.

23. D. A. Close, J. C. Pratt, J. J. Malanify, and H. F. Atwater, "X-ray Fluorescent Determination of Uranium in the Gaseous Phase," Nucl. Inst. and Meth. in Physics Research A234, 556-561 (1984).

24. K.K.S. Pillay, "Use of Tracers In Materials Holdup Study," Nucl. Mater. Manage. XII (Proceedings Issue), 182-186 (1984).

25. J. Razvi, K.K.S. Pillay, R. R. Picard, and R. S. Marshall, "Uranium Holdup Measurements for the Development of Predictive Models," Trans. Am. Nucl. Soc., 47, 172-173 (1984).

26. R. F. Eggers et al., "Examples of MC&A Systems to Meet Prompt Accountability Specifications," NURG/CR-2936 (PNL-4336), Pacific Northwest Laboratory, Richland, Washington (1983).

27. Regulatory Guide 5.37, "In-Situ Assay of Enriched Uranium Residual Holdup," U.S. Atomic Energy Commission (April 1974) and proposed revision to Regulatory Guide 5.37, Rev. 1 (November 1983).

28. R. H. Augustson, R. B. Walton. W. Harbarger, J. Hicks, G. Timmons, D. Shisslaer, R. Tayloe, S. Jones, R. Harris, and L. Fields. "Measurement of Uranium Holdup in an Operating Gaseous Diffusion Plant," CONF-831106, Proceedings of the Conference on Safeguards Technology: The Process Safeguards Interface, Hilton Head, SC (November-December 1983), pp. 77-88.

29. F. X. Hass, J. E. Gilmer, G. R. Goebel, J. L. Lawless, J. L. Pigg, D. L. Scott, and A. L. Tindall, "Implementation of Nondestructive Assay Instrumentation in the Rocky Flats Processing Facility," CONF-831106, Proceedings of the Conference on Safeguards Technology: The Process Safeguards Interface, Hilton Head, SC (November-December 1983), pp. 106-111.

30. A. G. Hamlin. "In-Process Holdup as a Measure of Safeguards Significance," Nuclear Safeguards Technology 1982, Proceedings of an International Symposium on Recent Advances in Nuclear Material Safeguards, IAEA, Vienna, Austria (November 1983), pp. 541-554.

31. R. R. Picard and R. S. Marshall, "Uranium Holdup Modeling," NUREG/CR-3448, LA-9853-MS, Los Alamos National Laboratory, Los Alamos, New Mexico (November 1983).

32. P. A. Russo, R. B. Strittmatter, E. L. Sandford, I.W. Jetter, E. McCollough, and G. L. Bowers, "Operation of Automated NDA Instruments for In-Line HEU Accounting at Y-12," Proceedings of the ANS Topical Conference on Safeguards Technology: The Process Safeguards Interface, Hilton Head Island, South Carolina (November 1983), pp. 112-134.

33. A. M. Krichinsky, "NUMATH: A Nuclear Material Holdup Estimator for Unit Operations and Chemical Processes," ORNL/TM-8175, Oak Ridge National Laboratory, Oak Ridge, Tennessee (February 1983).

34. R. Abedin-Zadeh, T. Beetle, G. Busca, S. Guardini, E. Kuhn, D. Terrey, and S. Turel, "Preparation of Plant-Specific NDA Reference Material," in Nuclear Safeguards Technology 1982, Vol. II, 57-63 (IAEA, Vienna, 1983).

35. R. L. Bramblett, "Passive Nondestructive Assay Methods," in Handbook of Nuclear Safeguards Measurement Methods, D. R. Rogers, Ed. (U.S. Nuclear Regulatory Commission, Washington, DC, 1983: NUREO/CR-2078, MLM-2855). Chap. V.

36. R. W. Hass, "Development and Verification of a Mathematical Model for a Pulsed Extraction Column," M.S. Thesis in Chemical Engineering, Iowa State University, Ames, Iowa (1983).

37. M. C. Moxon and E. W. Lees, "Non-Invasive Determination of the Uranium Content in a Centrifuge Plant Dump Trap," in Nuclear Safeguards Technology 1982, Vol. II. 57-63 (IAEA, Vienna, 1983).

38. D. B. Armstrong et al., "Controllable Unit Approach: An Application Manual," MLM-2881, Monsanto Research Corporation, Miamisburg, Ohio (1982).

39. M. S. Zucker, "Standards for Holdup Measurement," BNL-31549, Brookhaven National Laboratory, Upton, New York (1982).

40. M. S. Zucker, "Holdup Measurements for Nuclear-Fuel Manufacturing Plants," BNL-30261, Brookhaven National Laboratory, Upton, New York (1982).

41. M. S. Zucker, M. Degen, I. Cohen, A. Gody, R. Summers, P. Bisset, E. Shaub, and D. Holody, "Holdup Measurement for Nuclear Fuel Manufacturing Plants," Nucl. Mater. Manage. X, 239-251 (July 1981).

42. R.S.T. Shaw, "The Investigation of Industrial Plant Process Using Radioactive Tracer," J. Radioanl. Chem. 64, 337-349 (1981).

43. A. F. Cermak, "Pilot-Scale Pulsed Column Profile and Holdup Studies," AGNS-35900-3.1-111, Allied-General Nuclear Services (November 1980).

44. D. D. Cobb, L. E. Burkhart, and A. L. Beyerlein, "In-Process Inventory Estimation for Pulsed Columns and Mixer-Settlers," Proceedings of the 2nd Annual Symp. on Safeguards and Nuclear Material Management (European Safeguards Research and Development Association, Ispra, Italy, 1980), ESARDA 11, pp. 145-151.

45. T. K. Li and R. S. Marshall, "An In-Line Plutonium Glove-Box Filter Holdup Monitor," NBS Special Publication No. 582, Measurement Technology for Safeguards and Materials Control, NBS, Washington, DC (1980), pp. 308-312.

46. N. M. Trahey, "Reference Materials for Nuclear Safeguards Measurements—A Forecast for the 1980's," Nucl. Mater. Manage. IX (Proceedings Issue), 697-707 (1980).

47. I. A. Vasalos, D. N. Rundell, K. E. Megiris, and G. J. Tjatjopoulos, "Holdup Correlations In Slurry-solid Fluidized Beds," DOE/ET/10149-T3, Amoco Research Center, Naperville, Illinois (1980).

48. T. L. Hardt, M. B. Hughes, and M. P. Dedo, "Nondestructive Analysis at B&W's Uranium Conversion Plant," Nucl. Mater. Manage. V(8), 468-473 (July 1979).

49. G. E. Westsik, "Field Nondestructive Assay Measurements as Applied to Process Inventories," Nucl. Mater. Manage. VIII, 139-147 (July 1979).

50. L. Burkhart, "A Survey of Simulation Methods for Modeling Pulsed Sieve-Plate Extraction Columns," UCRL-15101, Ames Laboratory (March 1979).

51. I. Kiss and S. Vass, "A Possible Estimation of In-Process Inventory in Enrichment Cascades," Nucl. Sci. and Eng. 69(1), 1-5 (1979).

52. R. B. Walton, "Feasibility of Nondestructive Assay Measurements in Uranium Enrichment Plants, LA-7212-MS, Los Alamos Scientific Laboratory, Los Alamos, NM (April 1978).

53. H. C. Woodsum, "Summary Report of Phase I Residual Holdup Measurements for a Mixed Oxide Fuel Fabrication Facillty," WCAP-9278, Westinghouse Electric Corporation, Pittsburgh, Pennsylvania (March 1978).

54. D. J. Downing, D. H. Pike, and G. W. Morrison. "Application of the Kalman Filter to Inventory Control," Technometrics 22, 17-22 (1978).

55. S. B. Brumbach, "Experimental Program for Development and Evaluation of Nondestructive Assay Techniques for Plutonium Holdup," ANL-77-23/NUREG-0236, Argonne National Laboratory, Argonne, Illinois (May 1977).

56. B. F. Disselhorst, J. E. Glancy, and D. S. Brush, "Survey Techniques for Measuring Uranium-235," Nucl. Mater. Manage. VI(III), 411-423 (1977).

57. T. Gozani, "Review of Evaluation of Holdup Measurements," Nucl. Mater. Manage. VI(3), 424-433 (1977).

58. A. R. Anderson, R. H. James and F. Morgan, "Hidden Inventory and Safety Considerations," Nucl. Mater. Manage. V(3), 525-532 (1976).

59. R. Froelich, G. Kussmaul, and P. Schmuk, "Analysis of Hypothetical Nuclear Excursions in the Nuclear Holdup-System External to the Reservoir," (in German) Reaktortag., (Fachvortr.) 288-289 (1976).

60. C. H. Kindle, "In Situ Measurements of Residual Plutonium," Nucl. Mater.Manage. V(3), 540-549 (1976).

61. J. W. Tape, D. A. Close, and R. B. Walton, "Total Room Holdup of Plutonium Measured with a Large-Area Neutron Detector," Nucl. Mater. Manage. V(3), 533-539 (1976).

62. Regulatory Guide 5.42, "Design Considerations for Minimizing Residual Holdup of Special Nuclear Materials in Equipment for Dry Process Operations," (January 1975).

63. Regulatory Guide 5.25, "Design Considerations for Minimizing Residual Holdup of Special Nuclear Material for Wet Process Operations," (June 1974).

64. Regulatory Guide 5.8, "Design Considerations for Minimizing Residual Holdup of Special Nuclear Material in Drying and Fluidized Bed Operations," U.S. Atomic Energy Commission (May 1974).

65. Regulatory Guide 5.13, "Conduct of Nuclear Material Physical Inventories," U.S. Atomic Energy Commission (May 1973).

66. W. D. Reed. Jr., J. P. Andrews, and H. C. Keller, "A Method for Surveying for ^{235}U with Limit of Error Analysis," Nucl. Mater. Manage. II, 395-414 (1973).

67. "Selected Measurement Methods for Plutonium and Uranium in the Nuclear Fuel Cycle," 2nd Ed., C. J. Rodden, Ed., TID-7029, U.S. Atomic Energy Commission (1972).

68. "Evaluation of Minor Isotope Safeguard Techniques (MIST) in Reactor Fuel Reprocessing," WASH-1154, U.S. Atomic Energy Commission (February 1970).

69. H. J. Kouts and J. M. Williams, "U.S. Safeguards Systems Studies," in "Safeguards Systems Analysis of Nuclear Fuel Cycles," WASH-1140, U.S. Atomic Energy Commission (October 1969).

70. G. A. Schmel and A. L. Babb, "Holdup Studies in a Pulsed Sieve-Plate Solvent Extraction Column," Ind. Eng. Chem., Process Design Development, 2, 38-42 (1963).

71. T.E. Gier and J 0. Hougen ,"Concentration Gradients in Spray and Packed Extraction Columns," Ind. Eng. Chem. 45, 1362-1370 (1953).

1. K. K. S. Pillay, R. R. Picard, and R. S. Marshall, "Estimation Methods for Process Holdup of Special Nuclear Materials," NUREG/CR-3678, LA-10038, Los Alamos National Laboratory, Los Alamos, New Mexico (June 1984).

Imperative Ink

Nuclear Imperatives and Public Trust
Luther J. Carter
Resources for the Future, Inc.
Distributed by
Johns Hopkins University Press
Washington, D.C., 1987,
cloth, $25.00

When I received an invitation to write this review, my first reaction was dismay. The first thought was: this must be another book written by a frustrated sociology student, unable to obtain gainful employment, and peddling some one-dimensional view of the world. The words "Imperative" and "Trust" in the title triggered my apprehension. Later, I found myself reading it over a couple of nights as a thriller, and then reading it again carefully to write this review. My first reaction was wrong.

This book presents a quite useful overview of the field. There are some curious little biases and omissions, to which we shall refer later. However, on the whole, it contains a wealth of information not readily available to non-insiders.

High level waste management is such a complex cluster of issues, technical, political, social, economic, bureaucratic and so on, that we should keep in mind the story of the blind men and the elephant. None of us, blind men, can really get a broad perspective of the whole problem. My several years of work in evaluation of high level waste management programs has resulted in a rather humble attitude and a nagging feeling that at any given time I am not in possession of all the relevant and necessary facts. When somebody starts to tell me about the whole elephant, I get suspicious of getting just a projection of the elephant on some political plane.

Reality is rather delicate and elusive, and it is clearly felt only in those things with which we have direct and personal experience. Truth, therefore, in the sense of conformity with reality, in a document covering a broad range of issues beyond my direct experience, is difficult to verify. I have used for this review a rule of thumb that has served me well over the years: Look for those statements about which I have direct experience; if one statement is wrong, then reject the whole document; if no statement is clearly wrong, then assume tentatively that the document is right. I have not found a clearly wrong statement by the author, but I will point to some opinions with which I disagree in the body of the review.

It must be clear by now that I approached this review with great trepidation, but that I came to feel comfortable with the main message of the book. So much for a partial statement of prejudices.

The book is broken into four parts: Part 1, Sources of Public Unease; Part 2, Searching for a Waste Policy; Part 3, Europe, Japan, and the International Waste Problem; and Part 4, A Time to Act.

In the first part, it develops the theme of the two "imperatives, safeguards of SNM and containment of radioactivity," by describing the light water reactor fuel cycle, including decommissioning, decontamination of reactors, reprocessing and geological disposal. Along the way it states that borosilicate glass offers little or no advantage over spent fuel in terms of leach resistance and radionuclide retention. This is a view that is not supported by present work. Since this opinion appears frequently in the rest of the book as part of the argument against reprocessing, it is useful to elaborate the point. Borosilicate glass is not as good as some of the other waste forms that were considered and dismissed in the early eighties, but leach rates of radioisotopes from glass are much more predictable than releases from spent fuel.

Spent fuel has outside contamination, a gap inventory of fission products (in gas and mobile solids form) ready to be released when the cladding fails, and pellets of uranium dioxide that are thermodynamically unstable, tending to loose structural properties. In waste package designs offering containment for 1,000 years, glass comes very close to complying with NRC regulations for controlled release in some environments and by itself. The real technical problem is in the design of a good waste package for spent fuel.

It is curious that work on alternative waste forms that could provide better performance than glass is not mentioned in this part, particularly because such waste forms were being talked about at the time of the promulgation of the NRC regulations in 10/CFR 60.

It is also curious that the problem of low level waste resulting from medical applications of radioisotopes is ignored.

In describing Standards and Regulations for geologic isolation of nuclear wastes, the author mixes the requirements of the NRC in 10/CFR 60 with those of the EPA in 10/CFR 191. Perhaps it would have been useful to lead the reader briefly through the development of those standards, the concept of reasonable assurance, and the concept of multiple barriers. On page 419, near the end of the book, the author states, "... present regulations prescribe that the waste package shall contain the radioactivity after repository closure for 'not less than 300 years nor more than a 1,000 years' ...". As presented in the book it reads as if the regulations call for a beginning of releases at 1,000 years. What the regulations say is that the NRC should not require less than 300 years nor more than 1,000 years of containment. This mistake in interpretation is unfortunately too frequently made. If carried to its logical conclusion it would lead one to assume that the Swedish design is not acceptable in this country because it is expected to contain the activity for more than 1,000 years! Furthermore, the overwhelming of

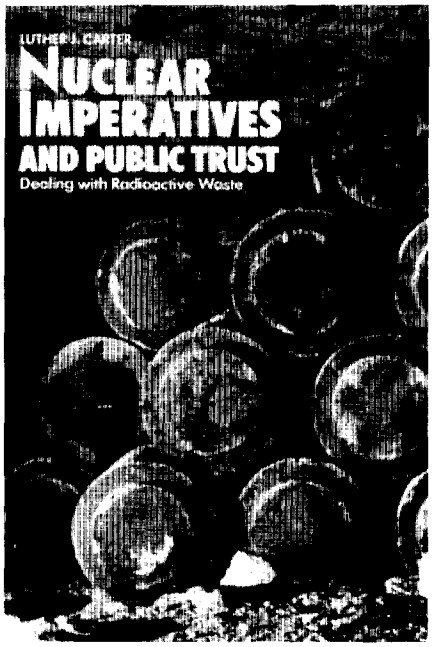
uncertainty advocated by Carter at the end of the book actually implies design goals of containment longer than 1,000 years. In my experience, trying to paraphrase regulations is dangerous, but is done all the time. This point about the regulations is a bit of nit-picking, since this is not used by Carter in the main thrust of his argument.

The following chapter under the title "Technology Ahead of Itself" develops the point that from the early days the AEC/ERDA/DOE tried to rush technology into applications with bad results. A description of the forces in action, such as public vs. private utilities, the push for the breeder, the anti-nuclear movement, military applications vs. civilian applications, the Price-Anderson Bill, and others may better explain the dynamics of the development. As it is, the kinetics of the evolution are very well described, and most people should find many interesting episodes, some amusing and some distressing.

Part 1 ends with the chapter "The Reprocessing Dilemma", a description of the evolution of commercial reprocessing of spent fuel, and safeguards issues. It follows the initial successful experiences of Hanford and Savannah River, and the unfortunate experiences of West Valley, Morris, and Barnwell. In doing so, it follows the differences in plant design philosophy, and the evolution of requirements. It paints an industry trying to cut costs, and being urged by the AEC to do so, competing with paper designs of competitors, in a rapidly changing regulatory and public opinion climate.

Carter's conclusion, "In sum, by the mid 1980's prospects for commercial reprocessing had further deteriorated, whether viewed from a technical, economic, regulatory, or political point of view," is too unqualified and strong. I would agree to it in the following form: By the mid 1980's prospects for commercial processing in the U.S., in the near future, had further deteriorated because of a combination of technical, economic, regulatory, and political reasons. The distinctions are important because they bear on the applicability of the American experience to other countries.

Part 2 begins with the chapter "Policy Struggles in the Bureaucracy". Here, Carter takes up again the thread of the evolution of official waste policy, which started in the sixties, and was described in chapter 2.



The time now in the narrative is the middle seventies and the main issues are beginning to take form: the controversy between looking at many sites on a broad geographical basis and selecting the best, and choosing either Nevada or Handford because they are already government sites; the question of whether to characterize many repositories or only one; the not-in-my-backyard attitude of the States; the choice of reliance on the geology versus reliance on the engineered barrier system to assure performance; the loss of power of the DOE to recommend a solution and have it accepted; the uncertainties of prediction in geology; the question of retrievability of the waste; the horse race between the field managers of the projects. The weave of conflicts of special interests and agendas described here is mind boggling. There is no way to summarize this chapter, you have to read it. It is hard reading but very rewarding.

The next chapter, "Conflict in the Host States," develops the geological characteristics of the various repository sites proposed at one time or another and the theme of not-in-my-backyard in each case. The chapter is long but is worth reading with care because of the number of cases reviewed, and because one can understand the birth of many sacred cows. The discussion of the Yucca Mountain site in Nevada sticks out a little and leaves a faint hint that Carter personally likes the Yucca site.

The conclusions are that environmental and land use conflicts should be avoided, that being able to thoroughly investigate a site helps a lot, that an independent well-financed review group increases the credibility of the site characterization effort, and that there should be incentives to the local population. We can have no argument with those.

Part 2 ends with the chapter "The Nuclear Waste Policy Act" which takes the reader through an inside view of the congressional debates that finally led to the Nuclear Waste Policy Act of 1982, which up to the end of 1987 was the law of the land. Carter refers to "The old adage that there are two things best not directly observed, one the making of sausage, the other the making of laws ...". I agree that the scene described in this chapter was quite disgusting, and tender youths should not read it in civics classes, but adults ought to read it. It leads to an understanding of the fate of the Monitored Retrievable Storage (MRS) and why the Richton Dome was eliminated from the selection.

At this point Carter breaks the

story off abruptly to go into Part 3, an overview of the international scene called "Europe, Japan and the International Waste Problem". I recommend that the reader skip this part and go directly to Part 4 where the yarn left hanging in Part 2 continues. You can treat Part 3 as an appendix.

Part 3 is an extensive review of the reprocessing and waste situations in the United Kingdom, Germany, Sweden, France, Japan, and Austria. As a summary overview it would have been useful to include Canada in the list, particularly because the Canadians were the first to adopt a throw-away fuel cycle. Also for completeness, a review of the USSR program could have helped. Other than that, the review is quite extensive and even touches on such arcana as the study on Palmyra island. The conclusions in this part are that international solutions to spent fuel and waste management are needed, that retrievable storage of spent fuel would alleviate the growing backlog of it, and that practical demonstration of retrievable disposal would facilitate future international waste management arrangements.

Part 4, "A Time to Act", consists of only Chapter 13, "Common Ground." Here Carter describes the controversy that attended the site selection, in the first round when the DOE selects Hanford over Richton, and in the second round when the selection of a site in the East is indefinitely postponed. The narrative stops at about the fall of 1986. He then proceeds to list some rather sensible lessons learned in this exercise. I will not summarize the lessons because by now the reader of this review should be able to guess what they are and because I want you to read the book.

Carter does offer some solutions. He proposes to focus the effort on a single repository site, given that siting the first repository is enough of a problem, and choosing between several sites involves choices between not really commensurable problems.

He suggests adopting a Swedish approach of overwhelming the uncertainties of site characteristics with conservative and robust designs of the engineered barrier system, to ensure containment for far longer than would be possible by just relying on the geologic barriers. He reminds us that this strategy was at the heart of the Interagency Review Group recommendations in 1979, and that this strategy contributed to the broad consensus in the scientific community that geologic isolation was technically feasible.

He proposes a new openness, encouraging broad discussion of the technical issues by independent experts, and discussion of methods by which the repository offers benefits to the local population.

He finds a role for the National Academy of Science in choosing one site for the repository, identifying waste package design concepts capable of overwhelming uncertainty, suggesting methods to be used at a test and evaluation facility to optimize system containment, and to help in setting up a credible process of peer review.

At the time of this review, February 1988, we have seen the Nuclear Waste Policy Amendments Act of 1987 passed by Congress. In it, Yucca Mountain is selected as the only site to be studied, discontinuing all site-specific work in Hanford, Deaf Smith County, and crystalline rock. There are benefit arrangements for the recipients of the repository, to be negotiated by a "Negotiator", and a Nuclear Waste Technical Review Board consisting of 11 members nominated by the National Academy of Sciences which shall evaluate the technical activities undertaken by the Secretary of Energy.

The Congressional Record of the House of December 21, 1987, includes a statement by Representative Snowe saying: "As a journalist, Luther Carter, predicted earlier this year, 'When Congress revisits the Nuclear Waste Policy Act, as it surely will have to do ... the policy should aim for early identification of a site that is technically suitable and relatively free of conflicts, and it should avoid vain and far-flung site-screening attempts that commit DOE ... to a punishing procedural marathon that goes nowhere.' Mr. Carter's prediction has come true."

In fact Carter suggested several things and Congress apparently listened. The suggestion of overwhelming uncertainty by a very conservative design of the waste package unfortunately does not appear in the new Act. It is to be hoped that it does not get lost in the shuffle, because a non-conservative design can easily lead to an endless period of controversy.

Reviewed by
Cesar Sastre
Brookhaven National Laboratory
Upton, New York

Posts Vacant in the IAEA

The Department of State, the U.S. Arms Control and Disarmament Agency and the Department of Energy have initiated a program to improve recruitment of U.S. nationals for employment in the IAEA.

In an effort to support this program, *JNMM* will publish IAEA vacancies.

Department of Safeguards

Division: Safeguards Information Treatment *Section:* Data Processing Development *Position:* Systems Analyst *Grade:* P-4 *Vacancy* #88/024 *Opened:* 15 March 1988 *Closing:* 15 July 1988

Division: Safeguards Information Treatment *Section:* Data Processing Development *Position:* Unit Leader/Systems Analyst *Grade:* P-4 *Vacancy* #88/021 *Opened:* 8 March 1988 *Closing:* 8 July 1988

Division: Operations B *Section:* OB1 *Position:* Senior Inspector - Group Leader *Grade:* P-5 *Vacancy* #88/019 *Opened:* 8 March 1988 *Closing:* 8 July 1988

Division: Safeguards Information Treatment *Section:* Data Processing Systems *Position:* Data Base Analyst *Grade:* P-3 *Vacancy* #88/017 *Opened:* 1 March 1988 *Closing:* 1 July 1988

Division: Operations *Section:* Nuclear Safeguards Inspector *Position:* P-4 (several positions) *Grade:* 87/SGO-4 *Vacancy* #9 June 1987 *Opened:* Continuous recruitment will be carried out until 31 December 1988

Division: Operations *Section:* Nuclear Safeguards Inspector *Position:* P-3 (several positions) *Grade:* 87/SGO-3 *Vacancy* #9 June 1987 *Opened:* Continuous recruitment will be carried out until 31 December 1988

Department of Technical Cooperation

Division: Technical Assistance and Co-operation *Section:* Latin America *Position:* Regional Projects Coordinator *Grade:* P-5 *Vacancy* #88/027 *Opened:* 12 April 1988 *Closing:* 12 August 1988

Department of Nuclear Energy and Safety

Division: Scientific and Technical Information *Section:* Computer *Position:* Systems Analyst *Grade:* P-3 *Vacancy* #88/025 *Opened:* 15 March 1988 *Closing:* 15 July 1988

Division: Scientific and Technical Information *Section:* INIS *Position:* Technical Information Officer *Grade:* P-4 *Vacancy* #88/022 *Opened:* 8 March 1988 *Closing:* 8 July 1988

How to Apply

Applications must include a vacancy notice number, and should be mailed to the United States Mission to the International Atomic Energy Agency, Kundmanngasse 21, 1030 Vienna, Austria (Attention Ronald Bartell). After U.S. Government endorsement is given, the Mission will forward the application to the Division of Personnel at the IAEA.

U.S. Candidates must also send a photocopy of the original application to: (for positions in the Department of Safeguards) P.O. Box 650, Brookhaven National Laboratory, Upton, N.Y. 11973, (for all other positions) IO/T/SCT, Rm. 5336, Department of State, Washington, D.C. 20520.

For more information contact Mr. W. Porter, Department of Energy, FTS 586-6175. Potential applicants should leave their name, address, and position in which they are interested. DOE will then forward a package of information on the IAEA and the position they wish to apply for.

Spent Fuel Seminar Proceedings Available

The proceedings of the fifth Institute of Nuclear Materials Management Spent Fuel Management Seminar are now available. These proceedings are a valuable reference, containing the complete text of the 23 papers presented at the seminar, held Jan. 20-22, 1988 in Washington, D.C.

The papers represent the current state of technology and regulation in Institutional Issues; Developments in Dry Storage Technology; Developments in Consolidation of Spent Fuel; Reactor Storage/DOE Transportation System Interface Considerations; and Technical Issues and Programs.

Copies are available for $200. For information contact Beth Perry, INMM, 60 Revere Dr., Suite 500, Northbrook, Ill. 60062 U.S.A. Telephone 312/480-9573.

Victoreen Publishes New Catalog

Victoreen, Inc. has published a new 168-page nuclear products catalog covering instruments and systems. There are selections for GM/Scintillation survey meters, ion chamber survey meters, personal dosimetry, area minitots, calibration and analysis instruments, quality assurance/radiology and more. Contact Victoreen, 6000 Cochran Rd., Cleveland, OH 44139-3395. Telephone (216) 248-9300.

Guide to Background Investigations Available

The *1988 Guide to Background Investigations* is a comprehensive source directory for employee screening for internal security.

Using the *Guide,* employers can verify such information as criminal convictions, college degree falsifications, workers compensation claims and driving records. The *Guide* suggests what to look for in each catagory, and explains in detail how to use each section.

Included with the *Guide* is a separate 13-page Social Security Number Guide, a booklet that explains the Social Security numbering system and explains how to discover abuses of the system.

The *Guide* includes all 50 states and the District of Columbia, permitting detailed demographic checks. The majority of offices holding public records will search their files for little or no charge. The *Guide* lists procedures, fees and laws regarding the release of information on a state-by-state basis.

The *Guide* is available on a subscription basis for $124.95 for one year (two issues), or $199 for two years (four issues). Single issues are available for $95. Contact: National Employment Screening Services, Inc., 8801 S. Yale, Tulsa, OK 74137-3575. Telephone (916) 491-9936.

CALENDAR

August 1-19, 1988
Seventh Annual Battelle International Program in R&D Management, Battelle Memorial Institute, Columbus, Ohio U.S.A. *Sponsor:* Battelle Memorial Institute *Contact:* Dr. William D. Hitt, Director, Battelle International Program in R&D Management, Battelle Memorial Institute, 505 King Dr., Columbus, OH 43201-2693.

August 21-24, 1988
International Reprocessing and Waste Management Symposium, Denver, Colorado U.S.A. *Sponsor:* Nuclear Engineering Division, American Institute of Chemical Engineers *Contact:* Wayne Freeby, Bechtel National, Inc., 50 Beale St., P.O. Box 3965, San Francisco, CA 94119

September 11-15, 1988
International Topical Meeting on Nuclear and Hazardous Waste Management: Spectrum 88, Pasco, Washington U.S.A. *Sponsor:* American Nuclear Society, Atomic Energy Society of Japan, Canadian Nuclear Society, American Society of Mechanical Engineers, European Nuclear Society *Contact:* Joan M. Tenorio, (509) 376-2979, Assistant General Chairman, Spectrum 88, P.O. Box 159, Richland, WA 99352-0159

September 20-22, 1988
HAZTECH International '88, Cleveland, OH U.S.A. *Sponsor:* HAZTECH International *Contact:*Northwest Center for Professional Education, (206) 746-4173

September 25-27, 1988
Brookhaven National Laboratory, Technical Support Organization, 20th Anniversary Symposium (By Invitation Only) *Sponsor:* Brookhaven National Laboratory *Contact:* Lois Marascia, Symposium Coordinator, Technical Support Organization, Brookhaven National Laboratory, Building 197C, Upton, NY 11973 U.S.A.

September 25-28, 1988
Uranium Seminar '88, Tucson, Arizona U.S.A. *Sponsor:* Atomic Industrial Forum *Contact:* Conference Office, Atomic Industrial Forum, Inc., 7101 Wisconsin Ave., Bethesda, MD 20814-4891

October 30-November 4, 1988
International Conference of the American Nuclear Society, Washington, D.C. U.S.A. *Sponsors:* American Nuclear Society, European Nuclear Society *Contact:* Myron B. Kratzer, (301) 261-1501,1635 Orchard Dr., Annapolis, MD 21401

January 11-13, 1989
INMM Spent Fuel Management Seminar VI, Loew's L'Enfant Plaza, Washington, D.C. U.S.A. *Sponsor:* Institute of Nuclear Materials Management *Contact:* Beth Perry, (312) 480-9573, INMM, 60 Revere Dr., Suite 500, Northbrook, Ill. 60062

June 11-16, 1989
9th International Symposium on the Packaging and Transportation of Radioactive Materials (PATRAM '89), Washington, D.C. U.S.A. *Sponsor:* U.S. Department of Energy and the International Atomic Energy Agency *Contact:* Judith Gale, (301) 986-4870, 7101 Wisconsin Ave., Suite 610, Bethesda, MD 20814

October 23-28, 1989
1989 Joint International Waste Management Conference, Kyoto, Japan *Sponsor:* ASME, JSME, AESJ *Contact:* To submit papers on high-level waste contact S.C. Slate, (509) 376-1867, Battelle, P.O. Box 999, Richland, WA 99352; to submit papers on low-level waste contact F. Fiezollahi, (415) 768-1234, Bethtel National, 50 Beale St., P.O. Box 3965, San Francisco, CA 94119

The events listed in this calendar were provided by Institute members or taken from widely available public listings. We urge INMM members, especially those from countries outside the United States, to send notices of other meetings, workshops or courses to INMM headquarters.

ADVERTISER INDEX

Morgan Technical Service ..... IFC
D.S. Davidson Co. 3
Hirsch Electronics Corp. ..... IBC
INMM Annual Meeting
Call for Papers ..... BC
INMM Integrated Safeguards
Workshop 36
INMM Membership Application .. 39+40
INMM Spent Fuel Management 8
Jomar Systems 17
NUC Services 25
Ralph Lumb Associates 43
Teledyne Isotopes 5
TSA Systems 44