125
11-16 19
LA-8070-MS

Informal Report
(ISPO-74)

**Tokai Advanced Safeguards
Technology Exercise Task T-F**

**Study of Selected Capabilities Needed to
Apply DYMACE Principles to Safeguarding
the Tokai Reprocessing Plant**

MASTER

University of California

LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545

USA

IAEA

PROGRAM FOR
TECHNICAL ASSISTANCE
TO IAEA SAFEGUARDS

Department of Energy
Washington, D.C.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

LA-8070-MS
Informal Report
(ISPO-74)
UC-83
Base Technology
Issued: October 1979

**Tokai Advanced Safeguards
Technology Exercise Task T-F**
**Study of Selected Capabilities Needed to
Apply DYMAC Principles to Safeguarding
the Tokai Reprocessing Plant**

Compiled by
Luther L. Lowry
Ronald H. Augustson

Contributors:
Thomas R. Canada
Donald D. Cobb
James P. Shipley
Luther L. Lowry

MASTER

CONTENTS

ABSTRACT	1
I. INTRODUCTION	2
II. INSTRUMENTATION AT PROCESS POINTS INSIDE MBA-3	7
A. X-ray Fluorescence Spectroscopy	8
B. Absorption Edge Densitometry	10
C. Conclusions	18
III. DETERMINATION OF INVENTORY IN SOLVENT-EXTRACTION CONTACTORS	20
A. Techniques for Contactor In-Process Inventory Estimation	22
1. Direct Measurements	22
2. Isotope Tracer Methods	24
3. Contactor Models	26
4. Dynamic State Estimation	32
5. Reduced-Order Dynamic State Estimation	32
B. Conclusion	34
IV. DIVERSION SENSITIVITY ANALYSIS	35
A. Performance Measures	35
B. Performance Assessment	38
C. Synthesis to Meet Specifications	41
D. Application to TOKAI Reprocessing Facility	43
V. IAEA VERIFICATION OF DYMAC-LIKE SAFEGUARD SYSTEMS	46
A. Software Verification	47
B. Automatic Countermeasures	48
C. Human Countermeasures	51
D. Conclusions	51
REFERENCES	52

TOKAI ADVANCED SAFEGUARDS TECHNOLOGY EXERCISE

TASK T-F

STUDY OF SELECTED CAPABILITIES NEEDED TO APPLY DYMACH
PRINCIPLES TO SAFEGUARDING THE TOKAI REPROCESSING PLANT

Compiled by

Luther L. Lowry and Ronald H. Augustson

Contributors:

Thomas R. Canada
Donald D. Cobb
James P. Shipley
Luther L. Lowry

ABSTRACT

We discuss selected technical capabilities needed to apply the DYMACH principles to safeguarding the Tokai reprocessing plant. We are chiefly concerned with the measurements needed to close the mass balance around the process line and the analysis methods for assessing the results. Process conditions at the Tokai plant were used when numerical values were needed to assist the analysis.

A rationale is presented for the selection of instruments (x-ray fluorescence spectrometers, x-ray densitometers, and gamma-ray spectrometers) best suited to establishing plutonium concentrations and, ultimately, inventories in the feed tanks.

The current state of the art in estimating inventory in contactors is reviewed and profitable directions for further work are recommended.

A generalized performance surface has been developed that can measure the diversion sensitivity of the safeguard system when the instrument performance levels, the number of measurements made, and the false alarm probability are specified. An analysis of its application to the Tokai plant is given.

Finally, a conceptual approach to the problem of IAEA safeguards verification is discussed. It appears possible that, in the process of verifying, the full power of the plant operator's safeguard system can be brought to the service of the IAEA.

I. INTRODUCTION

In the spring of 1978, the United States, Japan, and France agreed on a cooperative effort to provide advanced technologies to the Japanese that would help them safeguard the Tokai Nuclear Fuel Reprocessing Facility. The International Atomic Energy Agency (IAEA) was invited to participate. The general name given to these efforts was Tokai Advanced Safeguards Technology Exercise (TASTEX). This report covers Task T-F, one of thirteen tasks in the program.

In this task we hoped to give the Japanese an understanding of the dynamic material accountability system (DYMAC) being installed in LASL's plutonium processing facility. We wanted to describe the basic philosophy that guides the design, to describe the instruments used for safeguards measurements and let them gain some first hand experience by using such instruments, and, finally, to describe the computer and software programs that analyze the measured data.

These goals were reached to a degree by holding a workshop at LASL but further discussions revealed additional problems. The specific problems LASL could help with were defined at a meeting in Vienna in October, 1978. These problems dealt with: (1) instrumentation for the nondestructive assay of plutonium at a series of points in the process flow, (2) the determination of plutonium inventory in the mixer-settlers, (3) data analysis methods that would yield a high sensitivity to diversion, and (4) a discussion of how the IAEA can benefit from the presence of a DYMAC-like safeguards system in a processing plant. These are areas in which LASL has some background so we agreed to examine these problems.

The essence of dynamic material accounting is the ability to draw conclusions about the possible diversion of small amounts of SNM after short time intervals. The upper limit to the time interval is the time required for the diverter to construct a weapon from the diverted material. The material diversion detection limit is vaguely described as a "significant quantity" or the amount required to make "one or some" weapons. These limits have sometimes been specified as 10 days and 8 kg. The accounting process consists of knowing, through measurement, the amount of material in the area at the start of the accounting period, knowing the amount at the end, and knowing how much material has entered and left the area during a short accounting interval. At the present time this interval is measured in months.

After studying the process flow at the Tokai facility, Mr. James E. Lovett concluded that the most convenient time to start (and end) the accounting process was the time when the product evaporator was emptied into an accountability vessel. This process occurs once a day and transfers material out of MBA-3 (see Fig. 1). At this time all other measurements needed to close the balance are made. At the present time, some of the material measurements are made by chemical analysis, which takes about two to three days. Lovett suggested that this procedure be followed for a period of seven days so that by the end of the tenth day all of the analyses could have been made and the computations completed. If, however, the problems addressed in this report can be solved by the approaches suggested, closure of the mass balance could be done each day. It remains to be seen whether or not the measurements are accurate enough to achieve the 8 kg diversion sensitivity.

The Tokai plant flow sheet is shown in Fig. 1. An examination of the figure shows that the plant is divided into eight material balance areas (MBAs). At certain times, a material balance will be struck for each of the MBAs by adding to the material already there, the difference between the material that goes in and the material that goes out. This gives the current inventory. A measurement of this current inventory should reveal whether or not there is material unaccounted for (MUF). The confidence we have in the number we get for MUF depends on the instrument accuracy and on the way we analyze the data.

One MBA, MBA-3, shown in Fig. 2, is of particular importance. It carries out the chemical processes that extract the fission products and separate the uranium and the plutonium from each other. To close the balance around this MBA, it is necessary to know all inputs and outputs and the initial and final inventory. The inputs and product outputs are measured batchwise. Waste output flow is measured continuously and samples are taken for concentration measurements. The rest of the material is in a state of motion, flowing through feed vessels, through mixer-settlers, or into the plutonium product evaporator. The approximate inventories within this MBA are shown in Table I.

To measure the material in the feed vessels, we recommend that additional measurement points be established at D2, D3, and D4 (Fig. 2). The input point to MBA-3, Q1, and the output point, Q9, would be retained. Section II of this report discusses instruments that would be suitable for these measurements.

MBA NUMBER

PNC PROCESSING DIV. 1971

Fig. 1.
Model reprocessing plant.

TABLE I

PLUTONIUM INVENTORY

<u>Location</u>	<u>Grams</u>
1st Feed Vessel	5300
1st Extraction Cycle	1770
2nd Feed Vessel	2440
2nd Extraction Cycle	1360
Pu 3rd Feed Vessel	1430
Pu 3rd Extraction Cycle	760
Pu Evaporator Feed Vessel	3750
Evaporator	10600
Product Receiving Vessel	10600

The accuracies determine the sensitivity of the test. If, however, the numbers from successive closures are combined, a greater sensitivity in the decision can be achieved. Section IV discusses the mathematical techniques of decision theory that can provide this increased detection sensitivity.

The dynamic accountability system that closes MBAs around unit processes and keeps records of all transfers of SNM to various places in the plant is expected to be a great help to the plant operator. The IAEA inspector is charged with the same responsibility of ensuring no diversion of material but his equipment is much less versatile. We would like to see if there is any way the inspector can use the dynamic system while maintaining his independence of plant-operator-produced conclusions. Section V will discuss thoughts on how the inspector can use the system while independently verifying the data and computations.

II. INSTRUMENTATION AT PROCESS POINTS INSIDE MBA-3

T. R. Canada¹

The successful application of dynamic material accountability procedures to any facility is based on the ability to close SNM material balances in a timely fashion. This implies a measurement capability that allows accurate, quick, and reliable SNM concentration determinations for samples drawn from key measuring points in the process.

The key measuring points we are concerned with now are shown in Fig. 3 as Q1, D2, D3, D4, and Q9. The choice of measurement technique and the achievable accuracies are strongly influenced by the characteristics of the material to be measured. These characteristics are shown in the table of Fig. 3 and were provided by T. Koizumi and M. Iwanaga of the Power Reactor and Nuclear

	Q 1	D2	D3	D4	Q9
U CONCENT	180 g/L	55	.8	0	—
PU CONCENT	2 g/L	.6	1.9	15	252
U/PU	90	92	.42	0	0
GAMMA ACTIVITY	340 Ci/L	.3	.0034	00024	TRACE

MBA NUMBER

KEY MEASURING POINT

SAFEGUARDS PROCEDURE
PNC REPROCESSING DIV. 1971
(MODIFIED)

Fig. 3.
Safeguards augmentation in process flow.

Fuel Development Corp., Tokyo, Japan. They were obtained assuming 4% initial enrichment fuel run at a specific power of 35 MWD/MTU, a burnup of 28,000 MWD/MTU, and a 180 day cooling period.

A. X-ray Fluorescence Spectroscopy

At Q1 the gamma-ray activity from the fission products are equivalent to 340 curies/liter (Ci/L). This is far too large a gamma-ray background for any instrument that assays on the basis of passive reception of gamma rays with or without sophisticated spectral analysis. In addition, the large U/Pu ratio rules out the determination of plutonium concentration by the simple application of absorption edge densitometry. (It is assumed here that no effort is made to remove the fission products and/or the uranium from the sample.) At the key measuring point D2, the U/Pu ratio is the same as at Q1 so, again, absorptimetry will not work. And, despite the gamma-ray activity being lower at this point (0.3 Ci/L), it is still too high for passive gamma-ray measurements.

Though gamma-ray analysis won't work with these solutions, x-ray fluorescence spectroscopy will. Both wavelength dispersive and energy dispersive systems have been used to measure uranium and plutonium in such solutions.

The high resolution of wavelength dispersive spectrometers is an asset in the measurement of spent fuels where there is a potential for interference from the multitude of fission product gamma rays. But x-ray tubes are needed to excite the fluorescence enough to overcome the background. An example of this is the instrument used by von Baeckmann, et al.²⁻⁴ to assay solutions having U/Pu ratios of from 50 to 300 with accuracies of about 1% (1 sigma) in 2 to 5 minutes. Solutions having up to 1 Ci/mL were routinely handled. No serious line interference from the fission products was observed.

The high efficiency of the energy dispersive solid state Si(Li) spectrometers, with radioisotopic excitation, is often an asset in measurements of nonradioactive solutions. It can also perform well with intensely radioactive solutions if it is excited by an x-ray tube. The instrument built by Pickles and Cate⁵ is a good example here. They were able to assay solutions having a U/Pu ratio of 400, a uranium concentration of 50 g/L, and a fission product background of about 2 Ci/L. Preliminary tests indicated that accuracies of 1-2% were possible with 10 minute analysis times.

The main consideration in the choice of which emission x-ray line or lines to use in an x-ray fluorescence measurement is the line's intensity above background and how well it is resolved relative to other lines in the spectrum. For uranium and plutonium (and other actinides, as well), the usual choice is the $L\alpha$ line. Figure 4 shows how well these lines can be resolved in the wavelength dispersive fluorescence spectrometer.³ And Fig. 5 shows how well they are resolved with the energy dispersive spectrometer.⁵ (It should be noted that the U/Pu ratio for this spectrum was 250 which explains the relative sizes of the U L and the Pu L peaks.)

It is important to correct the measured results for effects such as variations in x-ray beam fluence and for the absorption of fluorescence x-rays by other elements in the sample. This correction can be made in the wavelength dispersive spectrometer by dissolving a known amount of thorium in the sample solution. The correction is made by dividing the measured uranium or plutonium $L\alpha$ count by the measured thorium $L\alpha$ count and multiplying by the thorium concentration. Figures 6 and 7 show how well this works.³ The corrected calibration curve extends well beyond 30 mg/g of solution. In practice, however, it is recommended that the concentrations be diluted to about 20 mg/g for measurement. This technique can be used either with a sample of solution directly or a sample prepared by drying about 20 μ l of solution on a filter paper. Because of the small amount of material used on the filter paper, fission product activities up to 1 Ci/g can be directly handled and measured.

For energy dispersive spectrometers, the corrections are also made by counting the fluorescence x-rays from a standard. Titanium foils have been used as the standard when the sample is deposited as a dried thin film. In this case, it corrects not only for variations in beam fluence and fluorescence x-ray absorption but also for system dead time, pulse pileup rejection losses, and changes in geometry.⁵

At D3, the key measuring point for the feed tank to the plutonium third extraction cycle, the fission product background is very low and, normally, an x-ray fluorescence spectrometer would not be needed here. But, as the table in Fig. 3, shows, the concentrations of both the uranium and plutonium are very low at this point, 0.6 g U/L and 1.9 g Pu/L. The x-ray fluorescence spectrometer again seems to be the best instrument. At the Los Alamos Scientific Laboratory, R. B. Strittmatter has assayed uranium samples as low as 0.2

Fig. 4.
Spectrum of X-ray L-lines of Th-Cm
with a wavelength dispersive spec-
trometer.

g/L with a precision of 0.5% with a counting time of 1000 seconds. His instrument used an x-ray generator and a Si(Li) detector. He placed a thorium foil back of the sample and obtained absorption correction information by measuring the transmission of thorium fluorescence x-rays.

B. Absorption Edge Densitometry

Although we recommend the use of an x-ray fluorescence spectrometer at D3, it may be possible to use an L-edge densitometer. At the Los Alamos Scientific Laboratory, L-edge has been used to measure uranium concentrations between 2 and 100 g/L with manageable sample thicknesses. And

Fig. 5.
Uranium and plutonium X-ray L-lines
with an energy dispersive spec-
trometer.

Fig. 6.
Calibration curves for direct measurements.

Fig. 7.
Calibration curve for internal standard method.

it may be possible to do better. Since the instrument uses an x-ray tube to excite the fluorescence radiation, it is possible to measure both uranium and plutonium concentrations at the same time. The L-edge densitometer could also work well at D4 where the plutonium concentration is 15 g/L but not at Q9 where it is 252 g/L. The higher concentration attenuates the soft x-rays too much and sample thickness becomes difficult to manage. Of course, sample dilution is a possibility. This problem can be avoided by going to the K-edge densitometer. It can handle these concentrations very well. The K-edge densitometer is somewhat simpler, too, because isotopic sources that emit gamma rays near the plutonium K-edge are available. Consequently, this instrument does not need an x-ray generator.

Before discussing instrument performance, we will digress a moment to mention the more common methods of correcting for the effects of extraneous chemical elements in the sample, the so-called matrix corrections.

The ratio of transmissions for two gamma rays at energies E_U and E_L through an SNM-bearing solution of thickness x is given by

$$T_U/T_L = \exp(-\Delta\mu_s \rho_s x) \exp(-\Delta\mu_m \rho_m x) ,$$

where $\Delta\mu_{iU} = \mu_{iU} - \mu_{iL}$. At the absorption edge, E , of the SNM of interest, μ_m is a monotonically decreasing function of energy. If E_U and E_L are picked to be $E \pm \Delta E$ (where ΔE is small) (Fig. 8), then $\Delta\mu_m \approx 0$ and ρ_s is given by

$$\rho_s \approx -\ln(R)/(\Delta\mu_s x), \quad (1)$$

where $R = T_U/T_L$. As $E \rightarrow 0$, Eq. (1) approaches an equality, i.e., the measurement is specific to the SNM of interest and is independent of the matrix properties. The sensitivity of this technique is determined by the slope of this equation, $1/(\Delta\mu_s x)$, and may be increased by having a larger sample thickness x or by picking an absorption edge for which $\Delta\mu_s$ is larger. A list of the uranium and plutonium K and L absorption-edge energies and their corresponding $\Delta\mu$ values is given in Table II.

Either an x-ray generator or radioactive isotope may be used as the transmission photon source. The former has several advantages: (a) the "tailored" x-ray energy and beam intensity are variable; (b) the energy displacement, ΔE , from the absorption edge is limited only by the detector resolution; and (c) multiple, simultaneous elemental determinations such as plutonium/uranium are possible. Radioisotopic sources, when available with the correct energies, are often convenient and inexpensive. In general, high energy-resolution detection systems are required.

Since the development of this technique, it has been used to assay a variety of elements.⁶⁻¹¹ A majority of the systems have used x-ray generators and high-resolution wavelength-dispersive spectrometers. All have been concerned with techniques which will assure minimum matrix dependence.

Two principal techniques, with slight variations, have been used. In the first, ρ_s is calculated from Eq. (2), which is derived by writing the transmission equations for two x-rays through the sample containing SNM and matrix contaminants and solving for ρ_s .

$$\rho_s = \frac{k \ln T_2 - \ln T_1}{(\mu_{s1} - k \mu_{s2})x} \quad (2)$$

with $k = \mu_{m1}/\mu_{m2}$. The ratio, μ_{m1}/μ_{m2} , is determined either by measurement with a standard containing the known matrix impurities or by calculational estimation. Over limited energy ranges, the ratio, $k = \mu_1/\mu_2$ may

Fig. 8.
Measured plutonium mass-attenuation coefficients. The straight lines are least squares fits to the experimental data whose uncertainties are smaller than the symbols.

TABLE II

<u>Absorption Edge</u>	<u>Element</u>	<u>Edge Energy (keV)</u>	<u>(cm²/g)</u>
L _{III}	U	17.17	54.6
	Pu	18.05	51.9
K	U	115.6	3.65
	Pu	121.76	3.39

be written as a function of the energy: $\log(\mu_1/\mu_2) = m \log(E_2/E_1)$. The parameter m , the slope of this equation, is approximately constant for a wide range of Z at both edges. This permits an estimate of μ_1/μ_2 to be made either by assuming certain matrix contaminants or by measurement with an arbitrary (within limits) matrix blank.¹⁰

In the second approach, an extrapolation to the absorption-edge energy is made either by measuring the transmission at several energies¹¹ or, when known matrix contaminants are present, by calculation.¹² The latter technique is equivalent to that described above, but results in a slightly different expression for ρ_s . For example, transmissions measured near the K-edge

for samples with known ($Z > 50$) matrix contaminants may be extrapolated to the absorption edge:

$$\rho_s = \frac{-1}{\Delta\mu_s} \times \left(\frac{E}{E_U}\right)^{-m} \left[\ln T_U - \frac{E}{E_L} \right]^{-m} \ln T_L, \quad (3)$$

where $\Delta\mu_s$ is measured at the absorption-edge energy, and m is ~ 2.55 . When transmissions are measured at a number of energies close to the absorption edge (see Fig. 8), the data may be fitted by a straight line that is extrapolated to the edge energy.

We can now turn to instrument performance.

L-Edge Densitometry

For L_{III} -edge densitometry a low energy x-ray generator spectrum is required as a transmission source because no adequate radioisotopic sources are available with the proper energies. Figure 9 shows measurements made by Canada, et al.¹² It illustrates the performance of the instrument in assaying a set of uranium-bearing solution standards, using a high resolution Si(Li) detector. The calibration curve obtained from these data is shown at the bottom of the figure. The data points on each side of the absorption edge were fitted to a simple function and extrapolated to the absorption edge to provide a matrix-independent value of R .

Fig. 9.
Upper Curve: X-ray spectra transmitted through solutions containing a range of uranium densities. Lower Curve: The logarithm of the ratio of the transmissions on either side of the L_{III} edge vs the uranium density. The solid line is a least squares fit to the data.

Two uranium solution assay systems have been developed which utilize the L_{III} absorption edge technique. Both use wavelength dispersive spectrometers to detect the transmitted photons. The first system by Wright and Barringer¹³ was calibrated by measuring the transmission of a continuous x-ray spectrum at two discrete energies, one on each side of the absorption edge, through a sample blank containing matrix constituents only and then through a sample blank containing a known concentration of uranium. The second system by Hakkila¹⁴ differed from this in that the measured transmitted photon energies were derived from the fluorescence of a secondary target constructed of molybdenum and niobium. The corresponding K lines at 17.4 keV and 16.6 keV bracket the uranium L_{III} edge (17.17 keV) closely. Both systems report accuracies of about 1% (one sigma) for concentrations ranging from 5 to 35 g/L and sample thicknesses of 1 to 3 cm. Routine calibration assay times were approximately 30 minutes.

In a modification of the procedure, Hakkila, et al¹⁵ used the niobium K and the molybdenum K fluorescence x-rays from the secondary target to bracket the plutonium L_{III} absorption edge (18.05 keV). The system was calibrated by transmitting these lines and the niobium $K\alpha$ x-ray through a single standard solution containing both the matrix impurity elements and a known amount of plutonium and assuming the slope of the matrix absorption function (μ vs E) was constant over the energy range of interest. With sample lengths of 1 to 1.5 cm, plutonium concentrations of 10 to 25 g/L were measured with uncertainties of about 0.6%.

K-Edge Densitometry

K-edge absorption techniques with radioisotopic sources may be conveniently used to assay SNM-bearing solutions with concentrations 100 g/L. In the case of plutonium, the ^{75}Se and ^{57}Co gamma rays at 121.12 and 122.06 keV bracket the plutonium K-edge (121.795 keV) so closely that, in most cases, Eq. (2) may be applied by putting $k = 1$. Figure 10 shows an example of a measured calibration curve using these sources. It also shows a similar curve for the two ^{75}Se lines at 121.12 and 136.00 keV that, as a result of a larger ΔE from the edge, exhibits a smaller slope and a larger matrix sensitivity (a measured intercept greater than one). To demonstrate the matrix sensitivity for these ratios of transmission, measurements were made through a plutonium bearing solution ($\rho_{\text{Pu}} = 67.5$ g/L) and a variety of tin thicknesses. The results are plotted in Fig. 11 as R versus the effective tin density, had it

Fig. 10.

The ratio of transmissions, R , of two gamma rays through 2-cm-thick plutonium solutions vs plutonium density.

Fig. 11.

Logarithm of the measured ratio of gamma-ray transmissions through a plutonium-bearing solution and the (136/121) corrected ratio R_c plotted as a function of tin density in the solution.

been distributed in the solution. These data show the matrix sensitivity for $R(122/121)$ to be reasonably small, whereas that for $R(136/121)$ is significant. Also shown is the result of applying the correction given in Eq. (1) to the ^{75}Se gamma-ray transmissions (136 and 121 keV). The ratio obtained from the corrected transmission values, $R_c(136/121)$, varies only 1.5% with an addition of tin equal to 23 times the plutonium density.

Two high-resolution Ge(Li)-detector-based systems are presently under development that will measure ρ_{Pu} in solutions using $R(122/121)$. One of these,¹² will be placed in a glovebox line and is designed to measure plutonium concentrations between 100 and 400 g/L with a standard deviation between 0.5 and 1%. The gamma-ray transmissions from the two sources are measured separately through the 2.5-cm-thick sample. The second system¹⁶ uses a

mixed ^{75}Se - ^{57}Co source and fits the resulting doublet in a single transmission measurement. The uncertainties and applicable ρ_{Pu} range are equivalent for the two approaches.

No radioisotopic source is available with adequate intensities or peak-to-background ratios which brackets the uranium K-edge (115.6 keV) as closely as the 121.1- and 122.1-keV gamma rays bracket the plutonium K-edge (121.795). An optimum combination now appears to be a mixed ^{57}Co (122.1 keV)- ^{169}Yb (109.8 keV) source. Other possibilities exist, one of which is the ^{169}Yb 109.8 and 130.9 gamma rays. The latter is now used in a Ge(Li) detector-based solution-measurement system¹⁷ to assay the product solutions of a uranium recovery facility. These solutions are well characterized and have been duplicated by a set of standards that have similar matrix properties. Routine assays of 2-cm-thick samples for 1000 seconds yield a standard deviation of 0.5 to 1% over a concentration range of 100 to 400 g/L. The approach is being altered to the extrapolation technique described above to remove the requirement that the standards be close replicas of the samples. In addition to measuring the 109.8- and 130.5-keV gamma-ray transmissions, those of the ^{169}Yb 177.2- and 198.0-keV gamma rays will also be measured. The resulting data will be fitted and extrapolated to the edge energy.

Another combination of gamma rays, ^{57}Co (122 keV) and ^{153}Gd (103 keV), has been used with a NaI detector system and 1.5- to 2.0-cm sample thicknesses to measure lower concentrations of uranium between 1 and 100 g/L.²¹ Empirical correction factors are included in the analysis to account for variation in the matrix density due to acid type and normality. Accuracies of a few percent are claimed for concentrations between 5 and 100 g/L.

As stated above, the continuous bremsstrahlung spectrum from by an x-ray generator has a number of advantages as a transmission source.^{17,18} Figure 12 shows the spectra transmitted through four plutonium-bearing solutions and the resulting calibration curve obtained by taking the ratio of counts on each side of the absorption edge at 121.8 keV. The application of this source to multiple simultaneous elemental determinations is demonstrated in Fig. 13, which shows the x-ray spectrum transmitted through a sample of HTGR fuel. The K-absorption edges of both thorium and uranium ($\rho_{\text{Th}}/\rho_{\text{U}}$ approximately 4/1) are identified, allowing an accurate determination of the respective densities. A similar technique could be used for the simultaneous determinations of uranium and plutonium, provided the concentration ratios are not too extreme.

Fig. 12.

Upper Curve: X-ray spectra transmitted through solutions containing a range of plutonium densities. Lower Curve: The logarithm of the ratio of the transmissions on either side of the K-edge vs the uranium density.

Fig. 13.

X-ray spectrum transmitted through 2 cm of HTGR fuel.

C. Conclusions

1. At the feed vessel to the 1st extraction cycle (Q1) and at the feed vessel to the 2nd extraction cycle (D2), x-ray fluorescence spectroscopy seems to be the only choice because of the high gamma-ray background from the fission products. It is a good choice because the instrument can function very well in such an environment.

2. At the feed vessel to the plutonium 3rd extraction cycle (D3), the instrument choice is dictated by the plutonium concentration and it appears that, again, x-ray fluorescence spectroscopy should be used. We cannot, however, entirely rule out L-edge densitometry.

3. At D4, the feed vessel to the plutonium concentration stage, and at Q9, the product output, both the K-edge densitometer being developed under TASTEX T-G and the passive gamma-ray spectrometer being developed under TASTEX T-H can be used. These instruments have been optimized for measuring product (Q9). It might be desirable to optimize the gamma-ray spectrometer for the lower plutonium concentration (15 g Pu/L) at D4. The principal modification required would be the enlargement of the sample volume. It could still be used for higher concentration measurements by diluting the sample.

III. DETERMINATION OF INVENTORY IN SOLVENT-EXTRACTION CONTACTORS

(D. D. Cobb)

The determination of in-process inventory in process equipment is essential to the development of near-real-time (dynamic) accountability and control systems for nuclear processing plants.¹⁹ For spent-fuel reprocessing plants in particular, the determination of in-process inventories of uranium and plutonium in solvent-extraction contactors is especially troublesome, and is a key requirement for implementing proposed advanced accountability systems.^{20,21}

The problem of contactor inventory was critically examined in the Los Alamos Scientific Laboratory (LASL) report (LA-6881) of dynamic accounting techniques for future industrial-scale reprocessing plants. In that study, the reference chemical-separations process (1500 MTHM/year) includes two cycles of solvent extraction in the plutonium purification process (PPP) area (Fig. 14) following the U-Pu codecontamination and partition cycles. The two plutonium purification cycles consist of a series of four pulsed columns (2A, 2B, 3A, 3B). The nominal plutonium inventory in the four columns was estimated to be 17.6 kg, corresponding to a plutonium residence time of about 8.4 h for a throughput of 50 kg Pu/day.

Steady-state models of the PPP columns were developed to simulate typical column operation (LA-6881, Vol. II, App. G). The loss-detection sensitivity of dynamic accounting in the PPP was investigated as a function of the uncertainty in pulse column plutonium inventory, which was assumed to be in the range 5-20%. The loss-detection sensitivity was found to be limited by the uncertainty in column inventory during accounting periods of one day or less, and had a significant dependence on the column inventory uncertainty for accounting periods up to one week or more. For longer accounting periods, the loss-detection sensitivity was limited by systematic errors in measuring the input-output streams of the PPP.

In smaller reprocessing plants such as Tokai, contactor (mixer-settlers) inventories should affect materials accountability at least as much as in large plants, because the contactor inventory makes a larger contribution to the materials balances relative to the throughput.

For the above reasons, we have initiated a modest effort within existing US Department of Energy-Safeguards and Security funds to identify possible approaches to estimating contactor inventories. Because very limited resources are available for this effort, we are depending on the support of various experts already working in related technical areas.

Fig. 14.
Plutonium purification process block diagram.

The General Atomic Company is supplying experimental data on uranium inventories in pulsed columns from process development work at their Solvent-Extraction Pilot Facility. The GA Pilot Facility is operated by personnel with experience in solvent extraction work dating back to the startup of the Hanford Purex line. This is a unique resource from which we are obtaining essential operating data and experimental verification at a very modest cost. Such data are absolutely essential and, unfortunately, are virtually nonexistent in the open literature.

Researchers at Clemson University have initiated limited studies of the effects of mass-transfer dynamics and chemical kinetics on contactor behavior, and of modern systems identification techniques that may be useful for real-time estimation of contactor inventory. Dr. Lawrence Burkhart and coworkers at Iowa State University and the DOE-Ames Laboratory are engaged in developing improved theoretical models of pulsed column behavior, and are providing valuable consulting services.

It is premature to predict what results might come out of this tentative study of a very complex problem. Certainly we hope to identify credible approaches and the major problems that must be solved. Conceivable options range from direct, high-quality measurements of contactor inventory to real-time estimation of contactor behavior using models that are valid under all possible operating (and upset) conditions. Neither extreme appears feasible in the near future. A logical first step would be to consider combinations of measurement and estimation techniques that could be applied at least over a limited range of typical contactor operation.

A. Techniques for Contactor In-Process Inventory Estimation

The purpose of this discussion is to identify possible techniques for estimating contactor in-process inventory and to discuss their currently perceived advantages and limitations. None of these techniques are developed for contactor inventory estimation as yet. In the following discussion they are referred to as: (1) direct inventory measurements; (2) isotope tracer methods; (3) dynamic state estimation; and (4) "reduced-order" dynamic state estimation. Elements of each technique, separately or in combination, may be applicable to any particular contactor system.

1. Direct Measurements. A variety of process control instrumentation is installed in a chemical separations plant. Much of this plant-grade instrumentation is used to monitor and control the operation of solvent-extraction

contactors. For example, the acid concentrations, flow rates, and temperatures of inlet and outlet streams, the aqueous-organic interface levels, and the pulse amplitudes and frequencies are all monitored and controlled.

Neutron detectors may be used to monitor the contactors for plutonium concentration "bulges", indicative of process upsets and possible criticality excursions. Whereas these neutron sensors are adequate for criticality control, they would not normally be calibrated or shielded for quantitative inventory measurements.

Sample lines may be available from selected mixer-settler stages or from pulsed-column disengagement sections. Where present, such sample lines could provide very useful data for estimating contactor inventories.

Many process control measurements are made typically on input-output streams of the contactors. Stream flow-rate measurements may be obtained from the steam-jet or air-lift transfer systems. Rough calibration of those liquid-transfer systems could result in flow-rate measurements having accuracies of 5-10%. Orifice meters in head pots could also be calibrated in terms of flow rate. If monitors are placed on raffinate and recycle streams, they can be calibrated to provide plutonium concentration measurements that should be adequate (10% or better) for the small concentrations (0.1 g Pu/L) in those streams.²⁰

Of particular interest for contactor inventory estimation would be the availability of high-quality flow and concentration measurements from contactor feed and product streams. If the separations process is designed so that the contactors are separated by buffer tanks, then the feed and product flows and concentrations could be obtained by combining volume measurements with chemical analyses of the tank contents, assuming that the buffer tanks are equipped with appropriate level, density, and sample lines.

This is not the design of the reference plutonium purification process (Fig. 14) in which the four pulsed columns are buffered mainly by the column product-disengagement sections. For the reference process, on-line flow meters or head-pot orifice meters might be used to measure feed and product stream flow rates to a few percent or better. High-quality concentration measurements could be obtained if sample lines were provided from the product disengagement sections. Note that the desired number of sample analyses could be large, probably dictating the use of rapid nondestructive assay (NDA) techniques. Continuous measurements of plutonium concentration in process streams

might also be considered using appropriate NDA techniques and sample loops. Possible near-real-time measurement techniques for process streams are discussed in LA-6881 (see especially Vol. II, Apps. A and B).

If input-output measurements are not available for each contactor, one may be forced to estimate the lumped in-process inventory of a series of contactors bounded by the available input-output accounting measurements. This could seriously complicate the contactor-inventory estimation algorithm, and would probably degrade the materials accounting sensitivity.

In summary, direct, high-quality measurements of contactor inventory are not currently available, and their development likely would result in substantial instrumentation costs and extensive process design modifications. The development of indirect methods of estimating contactor inventory using available signals from plant-grade sensors probably is feasible. Successful development and application of such indirect methods, however, may well require the addition of high-quality flow and concentration measurements on contactor feed and product streams.

2. Isotope Tracer Methods. Isotope tracer methods for inventory determination are divided into two categories, the step-displacement and the tracer-pulse techniques. Both have been proposed as possible methods for estimating the in-process inventory in an entire chemical-separations process without first having to drain and flush the process line.

a. The isotope step-displacement method is discussed in LA-6881, Vol. II, App. D and in the references therein. This method is proposed for relatively small, batch-operated chemical separations processes having little batch mixing, that is, having small heels between batches. When the in-process inventory is to be determined, a special feed batch is prepared having a readily identifiable difference in plutonium isotopic composition from the normal feed batches. Small output batches are collected, sampled, and isotopically analyzed from all product and recycle streams until the analyses indicate that all normal in-process materials initially present have been displaced by the special batch.

The sensitivity of the method primarily depends on the number and size of the small output batches and the maintenance of input-batch integrity throughout the process. For a large reprocessing plant, the number of small batches to be collected and analyzed, the desirability of maintaining isotopically

uniform product, and the loss of batch integrity during normal, continuous operation may preclude applying this technique.

The feasibility of the step-displacement method, however, has been demonstrated at the small, batch-operated Eurochemic and Nuclear Fuel Services reprocessing plants. Although difficulties were encountered in applying the method, a few successful experiments were conducted at these plants resulting in plutonium and uranium inventory estimates to 5% or better.

b. The isotope tracer-pulse method is described in Ref. 22. The method is based on linear systems theory, in which the response of a linear, stationary, stochastic process can be derived solely from input-output observations.

Rigorous application of this theory would require that two assumptions be satisfied: (1) the process is linear and is maintained at steady state during the input-output observations; and (2) the process and measurement statistics are all stationary during the observations. If these assumptions are satisfied, the in-process inventory can be estimated by adding a series of trace quantities (pulses) of isotopically identifiable material to the input stream, and then sampling and isotopically analyzing the output stream. For a single input pulse, the inventory is estimated using the equation,²²

$$I = \frac{F^2}{L} \int_0^T C t dt, \quad (4)$$

where I is the steady-state inventory averaged over time T, F is the flow rate, C is the tracer isotope concentration, and L is the total amount of the tracer isotope.

To apply this method successfully would likely require careful conditioning of the input stream using well-characterized pulses of the tracer isotope and frequent, perhaps continuous, flow-rate and isotopic-assay measurements of the output stream. Because a chemical separations process will disperse the input pulses significantly in time, the output measurements will have to continue for an extended period.

For example, the plutonium residence time in the reference process is on the order of one day, and may be as long as 3h in a single contactor.²⁰ The time integration in Eq. (4), and hence the output measurements, must extend over a period that is significantly longer than the residence time. The resulting inventory estimate would be averaged over this extended period, and steady-state operation would have to be maintained for good results.

It is important to note that, if the tracer-pulse method were to be implemented, the behavior of the process would first have to be analyzed (modeled) to see if the requisite assumptions are satisfied. For the reasons given, we believe that input-output measurements and contactor models could be used more effectively and efficiently to estimate contactor inventory when used in conjunction with dynamic state estimation techniques, in which case, no special conditioning of the input should be required. Such estimation techniques will be discussed after first considering the status of solvent-extraction contactor models.

3. Contactor Models. The most widely used contactor model for solvent-extraction process development is SEPHIS (Solvent Extraction Processing Having Interacting Solutes).²³ SEPHIS was originally developed for a dilute-purex (15% TBP) LMFBR flow sheet. It has since been modified for standard Purex (30% TBP) flow sheets,²⁴ and upgraded by incorporating better equilibrium data with temperature effects and by providing for U-Pu partitioning calculations.²⁵

SEPHIS performs a stage-wise, iterative calculation of the approach to steady state of the U, Pu, and HNO_3 concentrations in a multistage contactor. The contactor is modeled as a series of ideal mixer-settler stages with equilibrium end points. The required number of ideal stages is selected to match the desired overall extraction or separation efficiency. The time lag per stage is just the ideal stage volume divided by the sum of the aqueous and organic flow rates.

Starting with initial U, Pu, and HNO_3 concentrations and feed stream compositions, the SEPHIS program calculates the concentrations in each theoretical stage at the end of each time step. Thus, calculations of transient behavior as well as of steady-state conditions can be made. However, SEPHIS does not include reaction kinetics, mass transfer, and turbulence effects that may be important, particularly during transients. Also, the underlying assumptions more nearly describe the steady-state operation of mixer-settlers than of continuous differential contactors (i. e., columns). Finally, there is little experimental validation of SEPHIS in the published literature, even for steady-state operation.

a. Mixer-Settler Models. It is generally believed that the assumptions of the SEPHIS model most nearly match the conditions in mixer-settler contactors. One comparison of SEPHIS with experiments is given in Refs. 23 and 24 for a

laboratory-scale mixer-settler contactor (3 scrub stages and 8 extraction stages). The feed stream contained 27.7 g U/L and 4.43 g Pu/L in 3.2 M HNO₃. The aqueous/organic flow ratio was 0.72 and the extractant contained 15.3% TBP. By assuming that aqueous and organic phase volumes per stage are constant, the (steady-state) plutonium concentration profiles can be integrated to obtain:

	<u>Aqueous</u>	<u>Organic</u>
Experimental Pu Inventory =	18.87 kg	14.94 kg
Calculated Pu Inventory =	23.26	16.59
% Difference =	+ 23	+ 11

About 99% of the plutonium inventory resides in the three scrub stages plus the first three extraction stages.

Savannah River Laboratory has published²⁶ a few comparisons of measured uranium inventories in their so-called mini-mixer-settlers with calculations using the TRANSIENTS code, apparently a variant of SEPHIS modified for a 7.5% uranium-recovery flow sheet. The compound extraction-scrub mixer-settler banks are center-fed, 16-stage contactors. The stripping mixer-settler banks are 12-stage contactors.

The following comparisons are taken from Ref. 26.

1D Bank (extraction-scrub)

Feed: 21.34 g U/L in 4.06 M HNO₃

Aqueous/Organic flow ratio = 0.47

Experimental U inventory = 4.991 kg

(1.474 kg in aqueous, 3.516 kg in organic)

Calc. U inv. = 5.194 kg

% Difference = +4.1

1E Bank (stripping)

Feed: 7.12 g U/L

Feed/Strip flow ratio = 3.0

Experimental U inventory = 3.532 kg

(3.327 kg in aqueous, 0.206 kg in organic)

Calc. U inv. = 3.467 kg

% Difference = -1.9

Essentially all of the uranium inventory in the D-Bank is in the 8 scrub stages plus the first two extraction stages. Most of the inventory in the E-Bank is in the last 4 stages.

Also in Ref. 26, power-law correlations were established from uranium inventory calculations over ranges of operating parameters. The forms of these power-law fits are given in the equations below, where U = uranium inventory, C = uranium feed concentration, F = feed flow, X = extractant flow, and S = scrub or strip flow. Some dependence on HNO_3 concentration in the various streams was also derived, but it is omitted here. In all cases the dependence of uranium inventory on the uranium concentration in the feed stream is nearly linear.

Extraction-scrub:

$$\ln(U) = a \ln(C) + b \ln(F/X) + c \ln(S/X) + d(S/X)^2 \ln(S/X).$$

Stripping:

$$\ln(U) = a + b \ln(C) + c \ln(F/S) + d(\ln(F/S))^2.$$

Under LASL sponsorship, researchers at Clemson University are developing improved models of mixer-settlers. The effects of nonequilibrium operating conditions are currently under investigation.²⁷ Potential effects of finite mass transfer coefficients (hence, nonequilibrium end points) are shown in Fig. 15. A 16-stage mixer-settler was modeled having the aqueous feed stream

Fig. 15.
Uranium aqueous-phase concentrations at steady state for Stages 9 to 16 of a 16 stage mixer-settler having an aqueous feed stream (10 g U/L) at stage 8; are points assuming a mass-transfer coefficient of 1 and x are points assuming a mass-transfer coefficient of 1000 (SEPHIS equilibrium limit).

(10 g U/L) inlet at stage 8 and the aqueous raffinate outlet at stage 16. A small value of the mass transfer coefficient, that is, a large departure from equilibrium, causes the aqueous phase holdup to increase. Experimental investigation of such effects is urgently needed.

The experimental data needed may be obtainable from the Tokai facility since we understand the mixer-settlers to be quite extensively instrumented for sample taking. To help verify the SEPHIS code and the improved Clemson University code, the following data are needed for each mixer-settler bank:

1. The number of stages in the mixer-settler bank.
2. The volume of each mixer.
3. The volume of each settler.
4. Ratios of the organic-to-aqueous phase volumes for the settlers of each stage.
5. Nitric acid concentration for the scrub solution or stripping solution, depending on the type of the mixer-settler bank.
6. TBP volume percent.
7. Number of feed streams, the stages receiving the feed streams, the flow rates of the feed streams, and the concentration of each component (U, Pu, HNO_3) in the feed streams.
8. Flow rate of the scrub solution and the flow rate of the organic extractant or the stripping solution.
9. If the mixer-settler bank is used for the U-Pu partitioning, the types and concentrations of the reductants must be given.

The programs simulate the transient behavior of all concentrations as well as the steady state concentrations for each stage. Total holdup and holdup in each stage may also be obtained directly from the programs. It would be very helpful to code development if this information could be made available.

b. Pulsed Columns. A set of SEPHIS calculations of plutonium inventory in the 2A column of the PPP (Fig. 14) has been provided to LASL by Allied General Nuclear Services.^{20,28} A total of 87 SEPHIS runs were made varying the feed, scrub, and extractant flow rates and the plutonium concentration in the feed stream over typical operating ranges. The small amount of uranium in the feed was found to have a negligible effect on the plutonium distribution in the column, as does the HNO_3 concentration in the scrub stream which can

be controlled within a narrow range. The effect of varying the HNO_3 concentration in the feed stream (0.9–1.1 M) was also negligible. Temperature variations were not considered. A summary of the SEPHIS 2A calculations is shown in Fig. 16.

A power-law fit to the SEPHIS 2A calculations over a restricted parameter range is given by

$$\text{Pu inventory} = 0.14 C^{1.0} \left(\frac{F + S}{X} \right)^{1.57}, \quad (5)$$

where the inventory is in kg, C is the feed stream concentration in g Pu/L, and F , S , and X are the feed, scrub and extractant flow rates, respectively. A standard error of 3% for the fit was obtained over the parameter ranges $C = 4 - 6$ g Pu/L and aqueous/organic flow ratio = 2.5 - 3.9.

Fig. 16.
SEPHIS 2A calculations.

The calculated 2A column inventory includes the total plutonium in the scrub, extraction, and disengagement sections. For example, the inventory corresponding to nominal flowsheet values²⁰ ($C = 5$ g Pu/L, $F = 430$ L/h, $S = 60$ L/h, and $X = 150$ L/h) is 4.5 kg Pu. Based on our steady-state modeling of the PPP columns (LA-6881, App. G), we estimate that about 90% of this inventory resides in the scrub and product disengagement sections. The plutonium residence time in the 2A column is estimated to be on the order of 2h, the majority of which (about 1.3 h) is in the product disengagement section.

Burkhart and coworkers²⁹ have developed a state-of-the-art simulation model for pulsed columns. The model incorporates several improvements over SEPHIS, based on a critical review of about 160 papers. Calculations are performed in a stage-wise fashion essentially using finite difference equations that include the effects of reaction kinetics, finite mass transfer, backmixing, and new correlations of phase volumes with phase flow rates. In Ref. 29, three versions of the Burkhart model, ranging from essentially a SEPHIS-type version to the most comprehensive version (one "real stage" per plate), are compared with input-output data (presumably steady state) from a Hanford partitioning column. Interestingly, even though all three versions of the model are matched to the same input-output data, the calculated plutonium inventory is more than a factor of two larger using the more sophisticated version of the model versus the SEPHIS-type version. This result calls into question the previous belief that SEPHIS inventory predictions are "on the high side" and therefore could have important consequences for both criticality control and safeguards accountability. Further comparisons of the Burkhart model are underway using experimental data provided by the General Atomic Company.

c. Centrifugal Contactors. Centrifugal contactors minimize the accountability problem because the in-process inventory is much smaller than in either mixer-settlers or pulsed columns for the same throughput. We are unaware of any theoretical modeling work for centrifugal contactors in the published literature. Presumably, one could make stagewise, SEPHIS-type calculations by matching desired end point conditions, but it is likely that the standard modeling assumptions are even less applicable to centrifugal contactors than to pulsed columns.

One recommendation given in LA-6881 of alternative process design considerations for improved materials accountability and control is to use centrifugal

contactors wherever possible to reduce the in-process inventory. For example, AGNS is using a 10-stage centrifugal contactor in the codecontamination cycle. The total centrifugal contactor volume may be on the order of 100 L, which corresponds to a liquid residence time of about 100 seconds. The plutonium inventory should be less than 100 g and would be a negligible contributor to materials balances.

4. Dynamic State Estimation. Modern systems identification and state estimation techniques potentially can provide the best possible estimates of contactor inventory. A framework, such as the Kalman filter, would be used to combine the following elements: (1) a dynamic contactor model that is valid under all operating conditions, ideally including startup, shutdown, and upset transients; (2) a sufficient set of process observables and corresponding measurements, most likely including real-time measurements of contactor inputs and outputs; and (3) the statistics of both the process and the measurements.

Candy and coworkers³⁰ have considered an idealized example in which a stage-wise contactor model served as the basis of an extended-Kalman-filter estimation-detection scheme. An idealized three-stage contactor model was developed using 27 "states," and all 27 states were assumed to be directly observable, that is, measurable. A set of fictitious real-time measurements having errors of 1% or less was invoked. Under these assumptions, the detection algorithm was shown to be very sensitive to unmeasured losses of plutonium from the contactor system.

Although not feasible at present, the future development of dynamic state estimation techniques could significantly enhance materials accountability and control in large-scale reprocessing plants and should therefore be pursued.

5. Reduced-Order Dynamic State Estimation. Because full dynamic state estimation of contactor inventory is not currently feasible, it seems logical, at least as a first step, to consider a restricted approach that may have near-term applicability. We refer to this restricted state estimation scheme as "reduced-order" dynamic state estimation because, although the same three elements (models, measurements, and statistics) would be required for its implementation, the scope of the required input information may be restricted, as necessary, to match the projected near-term availability of the appropriate technologies.

For example, consider a mass balance drawn about one or more contactors in series,

$$I(t_2) = I(t_1) + T(t_1, t_2), \quad (6)$$

where I is the in-process inventory and T is the net transfer of material across the contactor(s) during the time interval t_1 to t_2 , that is, during the accounting period. Then, if the input and output flows and concentrations were measured and the process were operated near steady state, at least at the end points of the accounting period, a near-steady-state contactor model could provide the required inventory estimates at the end points to close the mass balance. If transient (upset) conditions were encountered near the end of the desired accounting period, closure of the balance could be delayed until a steady-state condition is restored.

Depending on the operating range of validity of the contactor model, a series of well-characterized "reference states" of the contactors might be established that would be used for in-process materials control and inventory estimation. Such reference states could also be used as fiducials during periods of start up, shut down, or recovery from upsets.

Near-steady-state contactor models might include relatively straightforward correlations of the in-process inventory with external observables, similar to the power-law relationship expressed previously. For the purpose of illustration, consider the power-law relationship given above in Eq. (5) combined with relative-error standard deviations of 3% for the linear coefficient in (the "contactor system calibration parameter") and 1% for the measurements of input and output flows and concentrations. Then the relative standard deviation of the contactor inventory estimate would be about 4%, using standard, first-order, propagation-of-error theory.

A 5%, or even a 10%, contactor inventory estimate may be adequate for many applications.²⁰ Note, however, that inventory estimates based on simple input-output relationships may also be strongly correlated with the net-transfer terms in the mass-balance equation (Eq. 6). Such correlations would have to be accounted for explicitly when analyzing the sensitivity of a particular algorithm.

B. Conclusion

In conclusion, it must be noted that any contactor inventory estimation technique will probably require experimental validation (calibration) for each contactor system. Furthermore, the reduced-order dynamic state estimation approach should be viewed as a first step toward the development of a more comprehensive dynamic state estimation technique. As discussed above, modern state estimation techniques potentially provide the most accurate inventory estimates and also the possibility of determining contactor inventories in real time.

Because of the safeguards sensitivity of reprocessing plants in general, and the importance of solvent-extraction contactor inventory determination for improved materials accounting and control in particular, we recommend accelerating the development of contactor-inventory estimation techniques. The development of contactor models and estimation algorithms has currently progressed to the point that, without experimental testing and validation, further progress will be seriously hampered.

IV. DIVERSION SENSITIVITY ANALYSIS

(J. P. Shipley and D. D. Cobb)

An essential part of the design of a materials accounting system is the assessment of its performance in detecting possible diversion of nuclear material. Systems performance assessment, in turn, implies a definition of suitable performance measures that can be easily compared to externally generated criteria. Thus, there are two aspects to the analysis problem: first, the definition of performance measures, and second, relating those performance measures to the performance criteria for specific cases.

Generally, the second aspect can be treated in two ways. The first is the "analysis" approach in which a systems design, based on incorporation of specific technology, is postulated and then analyzed to obtain values of the performance levels. The second is the "synthesis" approach in which values of the performance measures are specified, and the technology required to meet the specifications is determined.

We shall discuss both approaches, and the definition of performance measures, in the sequel. These ideas are based on the framework of decision analysis developed in the Systems Studies Group³¹⁻³⁵ and on several internal documents concerning the problem of diversion sensitivity assessment.³⁶⁻³⁸ We shall also give an example of the application of the techniques to the Japanese reprocessing plant at Tokai-Mura.

A. Performance Measures

The primary performance measures embody the concepts of diversion sensitivity and timeliness. Because of the statistical nature of materials accounting, diversion sensitivity can be described appropriately in terms of the probability of detecting some level of diversion, while accepting some probability of a false alarm. Timeliness is quantified by the time required to achieve detection. Thus, the usual procedure is to define three performance measures: the probability of detection, the total diversion, and the time over which the diversion occurs until it is detected. Note that the diversion scenario is unspecified in detail; whether the diversion occurs in an abrupt or protracted fashion, the total amount diverted is the chosen measure of performance.

Using statistical techniques like those of decision analysis, one can show that the three performance measures are interrelated by a family of continuous functions that depend on the details of each particular materials accounting

system. Thus, a convenient way of displaying the capability of a system is a 3-dimensional graph of detection probability vs total loss and time. Such a graph might appear as in Fig. 17 and is called a performance surface.

The shape of the surface will depend not just on the materials measurement system, but also on the statistical techniques used to analyze the materials accounting data. Within the framework of decision analysis, several different tests are available to take advantage of specific diversion scenarios. For example, there are tests tailored to both random and uniform protracted diversion schemes. A performance surface such as Fig. 17 can be obtained for each test, but the surface is only realizable when the actual diversion scenario corresponds to that for which the test is designed. Therefore, to ensure most effective treatment of all possible diversion strategies, all the tests should be used.

The resultant performance surface when all tests are used is a composite of the performance surfaces for each test. It can be found in the following manner. Let L be the total loss and t be the current time; let $s_i(L,t)$ be one strategy for diverting L amount of material in the time interval 0 to t . The set of all $s_i(L,t)$ is $S(L,t)$, and the set of all tests is J . Then the probability of detecting L amount lost in time t under strategy $s_i(L,t)$ by means of test j is $P_D[s_i(L,t);j]$. One way of defining the composite performance surface is

$$P_D(L,t) = \max_J \min_J P_D[s_i(L,t);j] . \quad (7)$$

Fig. 17.
Typical performance surface for a dynamic safeguards accountability system.

That is, each point on the composite surface is found by choosing the worst-case diversion strategy among all the tests and then using the results of the best test. This definition takes no account of corroborating evidence provided by tests other than the best one. It may be possible to use some weighted combination of all the test results, particularly if the tests are independent,³⁹ but that procedure would be more complicated and has not yet been fully developed.

It should be clear that obtaining a performance surface described by Eq. (1) is a complex and time-consuming process, primarily because of the need to treat many diversion scenarios and the appropriate tests. Furthermore, the worst-case diversion scenario is not always obvious, although preliminary work has been done in this area.⁴⁰

Fortunately, there exists one test that does not depend on how the material was lost, but responds only to a total loss L in time interval t ; i.e., the test is scenario-independent. This is the CUSUM (cumulative summation of materials balances) test. If there are N materials balances drawn in time interval t , then the CUSUM statistic at time t , or at balance N , is

$$C(N) = I(0) - I(N) + \sum_{i=0}^{N-1} T(i) , \quad (8)$$

where $I(j)$ is an inventory measurement at balance j and $T(i)$ is a net-transfer measurement just after balance i . The fixed-length CUSUM test used to calculate the performance surface consists of comparing the CUSUM, divided by its standard deviation, to a threshold that depends on the desired false-alarm probability. That is, if we fix the false-alarm probability, P_F , then the threshold value for $C(N)$, $C_T(N)$, is the solution to

$$P_F = \int_{C_T(N)}^{\infty} P[C(N) | H_0] dC(N) , \quad (9)$$

where the integrand is the conditional probability density function of $C(N)$ given that no material is lost, i.e., hypothesis H_0 is true. The resultant probability of detection, the equation for the performance surface, is

$$P_D(L, N) = \int_{C_T(N)}^{\infty} P[C(N) | H_1; L] dC(N) , \quad (10)$$

where the integrand now is the conditional probability density function of $C(n)$ given that L amount of material is lost between balances 0 and N , i.e., hypothesis H_1 is true.

Using the CUSUM test, calculation of a lower bound, Eq. (9), for the performance surface given by Eq. (7) is relatively easy. It should be noted that the CUSUM generally is not optimal for any diversion strategy; one can always obtain better performance if the diversion strategy matches a particular test. However, if the CUSUM test is always among the tests used, the composite performance surface is always at least as good as that for the CUSUM, regardless of diversion scenario. Thus, the CUSUM test provides a good, conservative measure of systems performance.

B. Performance Assessment

The problem of measuring and displaying diversion sensitivity for a particular system has been significantly simplified by choice of the CUSUM test and the 3-dimensional performance surface. However, it would seem that other such analyses would be required for each different accounting system. This is true if one desires to characterize all the details of the performance precisely. For purposes of initial design and evaluation and approximate assessment of systems performance, reasonable simplifying assumptions based on the CUSUM test allow a set of universal performance surfaces to be derived.

Suppose that the variances of the absolute random and calibration errors, $\sigma_{\epsilon T}^2$ and $\sigma_{\epsilon I}^2$, respectively, in the measurements of net transfers are stationary. (This supposition implies no instrument recalibration.) Also, define

$$\sigma_I^2 = [I^2(0) + I^2(N)] \sigma_{\epsilon I}^2 + [I(0) - I(N)]^2 \sigma_{\eta I}^2, \quad (11)$$

where $\sigma_{\epsilon I}^2$ and $\sigma_{\eta I}^2$ are the (constant) random and calibration relative error variances, respectively, of the inventory measurements. Then, the variance of the CUSUM in Eq. (8) is

$$\sigma_C^2(N) = \sigma_I^2 + N\sigma_{\epsilon T}^2 + N^2\sigma_{\eta T}^2. \quad (12)$$

Normalizing with respect to σ_I^2 ,

$$\hat{\sigma}_C^2(N) = 1 + N\hat{\sigma}_{\epsilon T}^2 + N^2\hat{\sigma}_{\eta T}^2 ,$$

where

$$\hat{\sigma}_C^2(N) = \frac{\sigma_C^2(N)}{\sigma_I^2} , \quad \hat{\sigma}_{\epsilon T}^2 = \frac{\sigma_{\epsilon T}^2}{\sigma_I^2} , \quad \hat{\sigma}_{\eta T}^2 = \frac{\sigma_{\eta T}^2}{\sigma_I^2} .$$

Now, observe that

$$N_O \triangleq \frac{\sigma_{\epsilon T}^2}{\sigma_{\eta T}^2} = \frac{\hat{\sigma}_{\epsilon T}^2}{\hat{\sigma}_{\eta T}^2} \quad (13)$$

is the balance number at which the random and calibration errors in the net-transfer measurements make equal contributions to the CUSUM variance. Further, define

$$\hat{N} = \frac{N}{N_O} \quad (14)$$

$$= \left(\frac{\sigma_{\epsilon T}^2}{\sigma_I^2} \right) \left(\frac{\sigma_{\eta T}^2}{\sigma_I^2} \right) . \quad (15)$$

Then, the normalized CUSUM variance is

$$\hat{\sigma}_C^2(N) = 1 + \beta \hat{N}(\hat{N}+1) . \quad (16)$$

Note that this normalized CUSUM variance is independent of both the absolute values of the measurement-error variances and the absolute value of the balance number. Using Eqs. (9) and (10), with appropriate normalized variables, the response surfaces for a range of values of the parameter β can be calculated and displayed. Several such graphs for β values of 0.01, 1 and 100 are shown in Figs. 18-20. Surfaces for other values of β can be easily generated, and portions of the surfaces can be expanded to give more detail. For the surfaces of Figs. 18-20, the false-alarm probability is $P_F = 0.001$.

Fig. 18.
Performance surface for $\beta = 0.01$ and
false alarm probability = 0.001.

Fig. 19.
Performance surface for $\beta = 1.0$ and
false alarm probability = 0.001.

Fig. 20.
Performance surface for $\beta = 100$ and
false alarm probability = 0.001.

Thus, to use the graphs for assessing systems performance, which implies knowledge of σ_I^2 , $\sigma_{\epsilon T}^2$, and $\sigma_{\eta T}^2$, the procedure is as follows:

1. Calculate β to choose the appropriate surface.
2. Scale the total loss in σ_I units, $\hat{L} = \frac{L}{\sigma_I}$.
3. Calculate N_O and scale the time in N_O units, $\hat{N} = \frac{N}{N_O}$.
4. Each point on the surface then relates detection probability to $\hat{L}$ and $\hat{N}$.

These techniques give a method of assessing, approximately, the performance to be expected of a system assuming the capabilities of the individual instruments are known. That is, the techniques treat the analysis problem. The next section deals with the synthesis of materials accounting systems to meet performance specifications.

C. Synthesis to Meet Specifications

In the synthesis problem, desirable values of the performance measures, P_D (and P_F), L , and t are specified, and the required level of technology must be found. If the CUSUM test is used for data analysis, as has been argued to be appropriate, then the level of technology is specified by σ_I^2 , $\sigma_{\epsilon T}^2$, and $\sigma_{\eta T}^2$, as well as the number of balances, N , that occur during the time interval 0 to t . The variable N is subject to auxiliary constraints such as process features that preclude drawing balances as often as might be desired. In addition, N must be greater than zero at time t or no balance can be drawn at that time.

The specifications of P_D , P_F , L , and t are generally given in absolute units, e.g., 50% probability of detecting 8 kg of plutonium missing within 1 week at a false-alarm probability of 0.001. The P_D , P_F , and L , specifications can be translated into a requirement on $\sigma_C^2(N)$, Eq. (12), by means of Eqs. (7) and (10); denote the required value by σ_C^{*2} . Also define

$$\sigma_1^2 = N\sigma_{\epsilon T}^2$$

$$\sigma_2^2 = N^2\sigma_{\eta T}^2.$$

The variances σ_1^2 and σ_2^2 are the total contributions to the CUSUM variance of the random and calibration error components, respectively. Then, the requirement on the measurement-error variances, normalized to σ_C^{*2} , from Eq. (12) is

$$\hat{\sigma}_I^2 + \hat{\sigma}_1^2 + \hat{\sigma}_2^2 \leq 1, \quad (17)$$

where

$$\hat{\sigma}_I = \frac{\sigma_I}{\sigma_C^*}, \quad \hat{\sigma}_1 = \frac{\sigma_1}{\sigma_C^*}, \quad \hat{\sigma}_2 = \frac{\sigma_2}{\sigma_C^*}. \quad (18)$$

Equation (11) describes the first-octant volume of a sphere in $\sigma_I, \sigma_1, \sigma_2$ space. Any point on or inside the sphere (but in the first octant) has coordinates specifying measurement capabilities that are adequate to at least meet the σ_C^{*2} criterion.

The choice of the number of materials balances, N , to occur during time t depends on the individual measurements and measurement devices. In general, $\sigma_I, \sigma_{\epsilon T}$, and $\sigma_{\eta T}$ all depend on the amount of material to be measured and the time span of the measurements, that is, on N and t . As a very simple example, consider a process having M identical items or batches in the initial inventory; all M items are transferred at a constant rate through the process during time t , and the final inventory contains the same M items at time t . Let the absolute measurement error variances for a single item be denoted by $\sigma_{\epsilon I}^2, \sigma_{\eta I}^2, \sigma_{\epsilon T}^2$, and $\sigma_{\eta T}^2$ for random and calibration net-inventory and net-transfer error variances, respectively. Let the time interval be divided into N equal increments, each of length Δt :

$$\Delta t = \frac{t}{N}.$$

The number of items measured (as net transfers) each t interval is

$$n = \frac{M}{t} \Delta t = \frac{M}{N}.$$

Then, the variance of the CUSUM at time t is

$$\begin{aligned} \sigma_C^2(N) &= M\sigma_{\epsilon I}^2 + Nn\sigma_{\epsilon T}^2 + N^2n^2\sigma_{\eta T}^2 \\ &= M\sigma_{\epsilon I}^2 + M\sigma_{\epsilon T}^2 + M^2\sigma_{\eta T}^2, \end{aligned}$$

which is independent of the choice of N, but depends on the throughput, M. This result is a consequence of using the CUSUM test and assuming a simple structure for the measurement errors. For other measurement characteristics (e.g., allowing instrument recalibration) and other tests, such as those based on the Kalman filter, $\sigma_c(N)$ will depend on N.

The important point is that the synthesis can proceed in general way through the use of Eq. (17). Further synthesis depends on the details of the particular system and process, and on the available technology.

D. Application to TOKAI Reprocessing Facility

For the purposes of this simple example, the Tokai process can be viewed as a series of four, identical buffer tanks alternating with three, identical mixer-settler banks. The process throughput is 7.56 kg Pu/day, occurring as two 3.78-kg input batches and one 7.56-kg output batch each day. The inventory in each buffer tank and mixer-settler bank is 3.78 kg Pu for a total inventory of 26.46 kg. The inventories and transfers are assumed constant, and a materials balance is drawn once a week. Table III gives the assumed measurement errors. The question then is how well can we expect the accounting system to perform after one week. Thus, $N = 1$ in Eq. (12).

From Eq. (11), the net-inventory variance is

$$\sigma_I^2 = 14(3.78)^2(0.05)^2 = 0.5,$$

the net-transfer random-error variance is

$$\sigma_{ET}^2 = 14(3.78)^2(0.02)^2 + 7(7.56)^2(0.02)^2 = 0.24 ,$$

TABLE III

ASSUMED MEASUREMENT ERRORS

Item	Random Error (%, 1σ)	Calibration Error (%, 1σ)
Input (per batch)	2	1
Output (per batch)	2	1
Buffer tank (per tank)	5	—*
Mixer-settler bank (per bank)	5	—*

*Inventory calibration errors cancel at steady state.

and the net-transfer calibration-error variance is

$$\sigma_{IT}^2 = (14)^2(3.78)^2(0.01)^2 + (7)^2(7.56)^2(0.01)^2 = 0.56$$

Therefore, the CUSUM variance after one week is

$$\sigma_C^2(1) = \sigma_I^2 + \sigma_{ET}^2 + \sigma_{IT}^2 = 1.3 \quad .$$

The parameter β is

$$\beta = \left(\frac{0.24}{0.5} \right) \left(\frac{0.24}{0.56} \right) = 0.206 \quad ,$$

the values of N_O and N are

$$N_O = \frac{0.24}{0.56} = 0.429 \quad , \quad \hat{N} = \frac{1}{0.429} = 2.33 \quad ,$$

and the normalized CUSUM variance is

$$\hat{\sigma}_C^2(1) = \frac{1.3}{0.5} = 2.6 \quad .$$

Figure 21 is the performance surface corresponding to $\beta = 0.2$. For a diversion of $3\sigma_C(1) = 3.42$ kg at $P_F = 0.001$, Fig. 21 yields $P_D \approx 0.5$, in accord with our prior knowledge. Similarly, if we choose to set the detection level at $5\sigma_C(1) = 5.70$ kg, then Fig. 21 gives $P_D \approx 0.95$.

Now suppose that we wish to detect an 8-kg diversion after one week with $P_D > 0.99$ (i.e., the $6\sigma_C$ level) and $P_F = 0.001$, and let the measurement errors be as specified in Table III, except that the mixer-settler bank inventory error is unknown. We will use the methods of Sec. C to calculate the required value. We have

$$\sigma_C^* = \frac{1}{6}(8) = 1.333 \text{ kg} \quad ,$$

so that

Fig. 21.
Performance surface illustration for
hypothetical Tokai application.

$$\begin{aligned}\sigma_I^2 &= \sigma_C^{*2} - \sigma_1^2 - \sigma_2^2 \\ &= (1.333)^2 - 0.24 - 0.56 = 0.978.\end{aligned}$$

But

$$\sigma_I^2 = \sigma_I^2(\text{tanks}) + \sigma_I^2(\text{mixer-settlers}),$$

and

$$\sigma_I^2(\text{mixer-settlers}) = 0.978 - 0.2858 = 0.692.$$

Therefore, the required inventory measurement relative random-error variance for each mixer-settler bank is

$$\sigma_{IMS}^2 = \frac{0.692}{6(3.78)^2} = 0.008,$$

or the required relative measurement precision is

$$\sigma_{IMS} = 9\%.$$

Other choices of false-alarm and detection probabilities will yield a different requirement.

V. IAEA VERIFICATION OF DYMAL-LIKE SAFEGUARD SYSTEMS

(L. L. Lowry)

The IAEA, as an international body, is responsible for assuring that any diversion of SNM will be discovered; and discovered in a timely manner. It cannot relinquish this responsibility to anyone else. Nor can it accept assertions by any individual of a monitored nation as to the condition of the SNM in a plant.

The IAEA must depend on itself. It must make its own assessment of the reliability of the information it gets. It must make its own interpretation of the significance of the information. And, it must draw its own conclusions as to whether or not SNM has been diverted.

How well can the inspector do this? Compared to the plant operator, who is equipped with a DYMAL-like system, the Agency inspector is at a distinct disadvantage.

The complexity of the inspector's problem varies considerably, depending on whether he is involved with enrichment, fuel fabrication, power production, or reprocessing. Under current conditions, he is totally responsible for his instruments. He is burdened with their portability, occupied with maintaining their calibration and accuracy, and concerned about their reliability. Moreover, access to key measuring points in the plant and time available to him are severely limited. Some of the difficulties could be eliminated and a much greater capability could be provided if the IAEA inspector could have access to the plant operator's dynamic SNM accountability system. In fact, INFCIRC 153, paragraph 31, provides that the Agency "shall make full use of the State's system of accounting for and control of all nuclear material subject to safeguards under the Agreement, and shall avoid unnecessary duplication of the State's accounting and control activities."

But is there any way for the inspector to use plant equipment and still maintain his independence? If there is, the payoff seems large. It may well be worth some hard thinking about how to do this.

We can discuss some preliminary thoughts. None of the ideas have been implemented or even quantitatively analyzed. At the moment, they're just ideas. But if the IAEA has a staff and laboratory on site, some interesting possibilities come to mind.

A. Software Verification

We can use Fig. 22 to illustrate the ideas. In this figure we have represented an instrument that could be a gamma-ray spectrometer. It has a detector, a pulse-height analyzer, an analogue-to-digital converter, its own mini-computer to perform the spectral analysis and control the instrument, and finally a signal conditioner to drive the line to the plant computer.

First, suppose the IAEA had a safeguards computer that could process all the safeguards information just as the plant computer does. Such computers are not excessively expensive and a large part of the software for it would have been developed by the plant operator. True, the IAEA can't simply accept the software program without making certain that it does what is claimed. It must close material balances, make estimates of holdup, file transactions, and maintain a data bank of the quantities, characteristics, and locations of all the SNM in the plant. The Agency must also have the ability to make any other comparisons and analyses it desires. With immediate access to the data bank, it would be able to run historical comparisons, to make consistency checks on instrument readings, to compile inventories, and to make its own diversion probability analysis. This capability may have to be provided by the Agency itself since the things to be done may be different from what is done by the plant operator. But, with its own computer, the Agency could monitor and confirm by comparison every computation made by the plant computer, if that were deemed desirable. The Agency's computational capabilities would not be deficient.

Though the IAEA would have its own computer, it would not have its own instruments; it would use the plant operator's instruments. This would make the same SNM data that is available to the plant operator available to the Agency, as well. As we shall see, however, certain modifications to the instrument will be required to allow the Agency to validate the data. But the combination of a computational capability and valid data allows the Agency's

Fig. 22.
IAEA verification system.

verification activities to keep pace with the operator's dynamic SNM accountability system. Moreover, it gives the Agency a quantitative credibility equal to that of the plant operator's.

We must now think about the problem of validating the data. We must think of ways in which the system might be subverted and then think of ways of preventing it. As it turns out, four subversion methods can be countered automatically. One subversion method will require human countermeasures.

B. Automatic Countermeasures

Subversion Method 1: Falsification at the Minicomputer -- If SNM had been removed from the process material, a sample of it would read low. To make the output of the instrument right so the plant computer would give the right answers, the instrument's minicomputer could adjust the low reading by multiplying it by the ratio of concentrations, which the diverter would know. On the other hand, there will be times when the Agency will run its own standards

on this instrument. At such times, a normal software program would have to be in operation. To know which algorithm to use, the operator would have to be able to automatically distinguish between the Agency's sources and his own. But this could be done by concealing signatures in his source holders for ultrasonic, magnetic, or even optical recognition. With this ability to select the appropriate algorithm in the hands of the operator, the Agency could not rely on data received from the normal output of the instrument.

Countermeasure -- If we take the output of the instrument's detector preamp as the input to the Agency's own pulse height analyzer (as illustrated in Fig. 22), it would no longer be possible for the operator to modify it, since the signal doesn't go through the operator's minicomputer. This means, of course, that the Agency would have to have additional electronic components on the operator's instrument. They would be sealed and the Agency would have to maintain them. In the illustration, the output of the Agency's pulse height analyzer goes to the Agency's computer, rather than to a minicomputer, for gamma-ray spectral analysis. In fact, there may be no advantage for the operator to use a minicomputer, for spectral analysis either. (Minicomputers for data reduction and analysis are useful for stand-alone instruments. For this case, a minicomputer would still be needed for instrument control.)

Subversion Method 2: Biased Calibration -- This may be done in two ways. A set of false standards may be used to indicate a higher mass or the instrument sensitivity may be increased by appropriate adjustment of voltages so that the weaker source would give high count rates. The particular technique used would depend on the characteristics of the instrument. Indeed, since the sensitivity to voltage levels is different for different detectors, the feasibility of making adequate changes in the sensitivity will depend on the instrument.

Countermeasure -- The initial calibration of the instrument in its operational environment should be done jointly by the operator and the IAEA inspectors. The Agency should subject the operator's standards to a verification count in its own instrument designed and calibrated for this purpose. Once the Agency is convinced that the standards are true, the joint calibration can proceed. During the calibration, the Agency should monitor and record all instrument voltages (voltage monitor box in Fig. 22) that could have any influence on the instrument's sensitivity. Immediately after the calibration exercise, the

Agency should run a response curve with its remotely variable source. Variability is achieved by either changing the distance between source and detector as in the Californium Shuffler or by the remote introduction of standard absorbers (these functions are indicated by the response test box in Fig. 22). With this information available, the Agency could run a response check coordinated with an instrument voltage verification any time it wanted to. Moreover, it would be good practice to monitor the instrument voltages each time a measurement is made.

Subversion Method 3: False Data Insertion -- In a TV monitoring system, false data could be inserted by feeding the system from a video disc so that the scene would appear stationary. In a pulse system, electronically generated pulses could be used to bring the reading up to the desired level.

Countermeasure -- This can be detected by exercising the detector. In the case of the TV system, the scene being viewed should be equipped with a modifiable segment. It could be as simple as a light that could be turned on or off from the Agency's computer. In the case of a gamma detector, remotely insertable absorbers should be available; for neutron detectors, arrangements should be made to remotely insert material that would change the neutron spectrum, such as polyethylene, boron, or cadmium. For either of these detectors, the count rate would not change in the proper way if some portion of the count were being generated electronically.

This interference with the gamma-ray and neutron detectors would have to be timed so that corrections could be made for counts collected during these periods.

Subversion Method 4: Instrument Substitution -- A multiplexing system would be used to cycle the computers through the instruments to be read. The Agency's computer may be switched to the wrong or a substitute instrument.

Countermeasure -- To provide instrument identification, the Agency could store a set of random numbers in each instrument. Then, when the Agency's computer is supposed to be connected to a certain instrument, the computer could call for a transmission of a stored number. If the proper number were received, connection to the proper instrument would be assured. Transmission of the random numbers to the instrument's memory could be encrypted so the operator could not learn what they were. The numbers transmitted in response to interrogation would not have to be encrypted since they would be used only once.

C. Human Countermeasures

Subversion Method 1: Augmented Source -- Here some of the SNM has been removed from the process material so that concentrations are too low. To achieve the proper instrument reading, an augmenting source of the same SNM is provided.

Countermeasure -- We must assume that the augmenting source is of the proper isotopic composition. The combination, therefore, is indistinguishable from the correct source and, consequently, appears to the instrument as a valid source.

If the augmentation is through spiking to bring the sample up to strength, or if a sample is drawn from a previously prepared supply, there is no way to discover this at the instrument. To prevent this, IAEA inspection of the chemistry laboratory would have to show that no material was available to allow either of these subterfuges to occur.

If augmentation is by addition of a separate source, such as a foil, to the sample of the real process material, the Agency may inhibit such practice by asking that two samples be made of the process material, or that the Agency be given custody of the measured sample. The Agency would tell the operator that the sample would be used at unannounced times as a secondary standard. There would be no way for the operator to know when to attempt an augmentation.

D. Conclusions

It appears conceptually feasible for the IAEA to have a safeguard capability equal to that of the plant operator. But it will require the agency to have its own computer to verify the plant operator's software; it will require an automatic means of detecting certain kinds of data subversion; and it will require human monitoring to prevent the preparation of fallacious samples.

REFERENCES

1. T. R. Canada, D. G. Langner, and J. L. Parker, "Coordinated Safeguards for Materials Management in a Fuel Reprocessing Plant," Los Alamos Scientific Laboratory report LA-6881, Vol. II, Appendix A. (Sept. 1977).
2. A. von Baeckmann, "Determination of Uranium and Plutonium in Solutions of Spent Nuclear Fuels by X-Ray Fluorescence Analysis," Proc. IAEA Symp. on Safeguard Techniques, Vol. II, 41-45 (1970).
3. A. H. E. von Baeckmann, D. Ertel, and J. Neuber, "Determination of Actinide Elements in Nuclear Fuels by X-ray Analysis," Advan. X-ray Anal. 18, 62-74 (1975).
4. P. A. Pella and A. V. Baeckmann, "The X-Ray Spectrometric Determination of Uranium and Plutonium in Solutions of Spent Nuclear Fuels," Anal. Chim. Acta. 47, 431-438 (1969).
5. W. L. Pickles and J. L. Cate, Jr., "Quantitative Nondispersive X-Ray Fluorescence Analysis of Highly Radioactive Samples for Uranium and Plutonium Concentration," UCRL-74717 (August 1973).
6. E. A. Hakkila and G. R. Waterbury, "X-Ray Absorption Edge Determination of Cobalt in Complex Mixtures," Advan. X-Rays Anal. 5, 379-388 (1961).
7. E. A. Hakkila and G. R. Waterbury, "Applications of X-Ray Absorption Edge Analysis," Devel. Appl. Spect. 2, 297-307 (1963).
8. C. G. Dodd and D. J. Kaup, "Optimum Conditions for Chemical Analysis by X-Ray Absorption-Edge Analysis," Anal. Chem. 36, 2325-2329 (1964).
9. K. T. Knapp, R. H. Lindahl, and A. J. Mabis, "An X-Ray Absorption Method for Elemental Analysis," Advan. X-Ray Anal. 7, 318-324 (1964).
10. E. P. Bertin, R. J. Longoloucco, and R. J. Carver, "A Simplified Routine Method for X-Ray Absorption Edge Spectrometric Analysis," Anal. Chem. 36, 641-655 (1964).
11. H. W. Dunn, "X-Ray Absorption Edge Analysis," Anal. Chem. 34, 116-121 (1961).
12. T. R. Canada, E. R. Martin, J. W. Tape, W. B. Tippens, T. D. Reilly, and J. L. Parker, "Gamma-Ray Densitometry," LA-6675-PR, 19-22 (1977).
13. W. B. Wright and R. E. Barringer, "Uranium Analysis by Monochromatic X-Ray Absorption," Union Carbide Nuclear Company Report Y-1095 (August 1955).
14. E. A. Hakkila, "X-Ray Absorption Edge Determination of Uranium in Complex Mixtures," Anal. Chem. 33, 1012-1015 (1961).
15. E. A. Hakkila, R. G. Hurley, and G. R. Waterbury, "Three-Wavelength X-Ray Absorption Edge Method for Determination of Plutonium in Nitrate Media," Anal. Chem. 38, 425-427 (1966).

16. R. Gunnink, private communication.
17. T. R. Canada, J. L. Parker, and T. D. Reilly, "Total Plutonium and Uranium Determination by Gamma-Ray Densitometry," LA-6142-PR, 10-13 (1975).
18. T. R. Canada, J. L. Parker, and T. D. Reilly, "Total Plutonium and Uranium Determination by Gamma-Ray Densitometry," ANS Transactions 22, 140 (1975).
19. H. A. Dayem, D. D. Cobb, R. J. Dietz, E. A. Hakkila, J. P. Shipley, and D. B. Smith, "Dynamic Materials Accounting in the Back End of the LWR Fuel Cycle," Nuclear Technology, Special Issue on the Back End of the LWR Fuel Cycle, (mid-April, 1979).
20. E. A. Hakkila, D. D. Cobb, H. A. Dayem, R. J. Dietz, E. A. Kern, E. P. Schelonka, J. P. Shipley, D. B. Smith, R. H. Augustson, and J. W. Barnes, "Coordinated Safeguards for Materials Management in a Fuel Reprocessing Plant," Los Alamos Scientific Laboratory report LA-6881 (September 1977).
21. R. H. Augustson, Los Alamos Scientific Laboratory and J. E. Lovett, International Atomic Energy Agency, personal communication concerning TASTEX-Task F, February 1979.
22. S. M. Zivi and W. B. Seefeldt, "Temporal Response Methods for Dynamic Measurement of In-Process Inventory of Dissolved Nuclear Materials," US Nuclear Regulatory Commission report NUREG-0249 (September 1977).
23. W. S. Groenier, "Calculation of the Transient Behavior of a Dilute-Purex Solvent Extraction Process Having Application to the Reprocessing of LMFBR Fuels," Oak Ridge National Laboratory report ORNL-4746 (April 1972).
24. S. B. Watson and R. H. Rainey, "Modifications of the SEPHIS Computer Code for Calculating the Purex Solvent Extraction System," Oak Ridge National Laboratory report ORNL-TM-5123 (December 1975).
25. G. L. Richardson, "Effect of High Solvent Irradiation on TBP Processing of Spent LMFBR Fuels," Hanford Engineering Development Laboratory report HEDL-TME-73-51 (June 1973).
26. M. C. Thompson and R. L. Shankle, "Calculation of Uranium Inventories in Mixer-Settlers During Solvent Extraction with 7.5% TBP," Savannah River Laboratory report DP-1357 (August 1974).
27. J. E. Bennett, Clemson University, personal communication, February 28, 1979.
28. B. M. Legler, Allied-General Nuclear Services, to E. P. Schelonka, Los Alamos Scientific Laboratory, personal communication, May 4, 1977.
29. S. M. Yih and L. Burkhardt, "State-of-the-Art Simulation of a U-Pu Partitioning Column for Nuclear Fuel Reprocessing," ANS Proceedings of the Winter Meeting, TANSO 30, 328 (November 1978).

30. J. V. Candy, R. A. Emmert, and G. K. Patterson, "Material Accounting in a Complex Chemical Processing Unit - An Application of Estimation/Detection," Lawrence Livermore Laboratory preprint UCRL-79607 (October 1977).
31. E. A. Hakkila, D. D. Cobb, H. A. Dayem, R. J. Dietz, E. A. Kern, E. P. Schelonka, J. P. Shipley, D. B. Smith, R. H. Augustson, and J. W. Barnes, "Coordinated Safeguards for Materials Management in a Fuel Reprocessing Plant," Los Alamos Scientific Laboratory report LA-6881, Vol. II, App. E (September 1977).
32. J. P. Shipley, "Decision Analysis in Safeguarding Special Nuclear Material," invited paper, Trans. Am. Nucl. Soc. 27, 178 (1977).
33. J. P. Shipley, "Decision Analysis for Nuclear Safeguards," invited paper presented to the American Chemical Society 1978 Spring Meeting, Anaheim, California, March 12-17, 1978, published in Nuclear Safeguards Analysis - Nondestructive and Analytical Chemical Techniques, E. A. Hakkila, Ed. (American Chemical Society, Washington, D. C., 1978).
34. J. P. Shipley, "Decision Analysis for Dynamic Accounting of Nuclear Material," presented at the American Nuclear Society Topical Meeting, Williamsburg, Virginia, May 15-17.
35. J. P. Shipley, "Efficient Analysis of Materials Accounting Data," presented at the 19th Annual Meeting of the Inst. Nucl. Mater. Manag., Cincinnati, Ohio, June 27-29, 1978.
36. J. P. Shipley, "Evaluating Diversion Sensitivities", Los Alamos Scientific Laboratory internal document Q-4/78-394 (December 26, 1978).
37. J. P. Shipley, "Universal Curves for Diversion Sensitivities", Los Alamos Scientific Laboratory internal document Q-4/79-013 (January 8, 1979).
38. D. D. Cobb, "CUSUM Universal Plots (CUPS)", Los Alamos Scientific Laboratory internal document Q-4/79-015 (January 10, 1979).
39. W. J. Whitty, "Combination of Statistical Tests of Significance," Los Alamos Scientific Laboratory internal document Q-4/79-061 (February 12, 1979).
40. J. T. Markin, "Applications of Dynamic Programming in Nuclear Safeguards," Los Alamos Scientific Laboratory internal document Q-4/Technical Memorandum No. 2 (January 9, 1979).