KfK 3515 Juli 1983

Overview Report of the International Workshop on the Near-Real-Time Accountancy Measure

D. Gupta [Ed.] Entwicklungsabteilung Kernmaterialsicherung

Kernforschungszentrum Karlsruhe

KERNFORSCHUNGSZENTRUM KARLSRUHE Entwicklungsabteilung Kernmaterialsicherung

KfK 3515

Overview Report of the International Workshop on the Near-Real-Time Accountancy Measure

D. Gupta [Ed]

Based on Contributions from

R. Avenhaus, R. Beedgen, C.A. Bennett, R. Berg, M.J. Canty, H.J. Hein, C.G. Hough, K. Ikawa, J.L. Jaech, M. Kluth, M. Kuechle, E. Leitner, J.E. Lovett, E. Mainka, J.T. Markin, H. Nishimura, D. Sellinschegg J.P. Shipley, G. Spannagel, F. Voss, R. Weh, H. Wuerz

A slightly modified version of this report without the Appendix was presented as paper No. IAEA-SM-260/29 at the International Symposium on Recent Advances in Nuclear Materials Safeguards. IAEA, Vienna Nov. (1982)

Kernforschungszentrum Karlsruhe GmbH, Karlsruhe

Als Manuskript vervielfältigt Für diesen Bericht behalten wir uns alle Rechte vor

Kernforschungszentrum Karlsruhe GmbH ISSN 0303-4003

Organizations, Active Participants, Contributors International Workshop on the Near-Real-Time-Accountancy Measure

Subject	Participants(p)/Contributors(c)	Organizations
l. Reference Reprocessing Facility	R. Weh (c); M. Kluth (c), F. Voss (c); H.J. Hein(c)	DWK KfK WAK (Federal Republic of Germany)
2. Measurement Systems	L. Koch (p); P. Groll (p), E.Mainka (c) M. Kuechle (c), H. Ottmar (p), H. Wuerz (c); R. Berg (c)	EURATOM { KfK (Federal Republic { of Germany) { WAK (Federal Republic of Germany)
3. Simulation Models	<pre>J.E. Lovett (c); K. Ikawa (c); M.J. Canty (c); G. Naegele (p), D. Sellinschegg (c), G. Spannagel (c), F. Voss (c); J.P. Shipley (c); H.J. Hein (c)</pre>	IAEA JAERI (Japan) KFA (Federal Republic of Germany) (KfK (Federal Republic (of Germany) Los Alamos (USA) WAK (Federal Republic of Germany)
4. Statistical Test Procedures	<pre>C.A. Bennett (c); F. Argentesi (p); J.L. Jaech (c); R. Avenhaus (c); C.G. Hough (c), J.E. Lovett (c); K. Ikawa (c), H. Nishimura (c); R. Beedgen (c), D. Sellinschegg (c); J.T.Markin (c), J.P. Shipley (c); H.J. Hein (c)</pre>	Battelle (USA) EURATOM Exxon (USA) Hochschule der Bundeswehr (Federal Republic of Germany) IAEA JAERI (Japan) KfK (Federal Republic of Germany) Los Alamos (USA) WAK (Federal Republic of Germany)

·

ABSTRACT

An International Workshop on the near-real-time accountancy (NRTA) measure was established in December 1980 to investigate the capabilities and limitations of this measure for a large-scale reprocessing facility. The present overview report summarizes the activities and the results of this workshop as of July 1982. After establishing the process and accountancy data-base for a 1000 t HM/a reference reprocessing facility, the workshop developed simulation models for the sequential generation of data for throughput and inventory of plutonium in the process material balance area (MBA). A well defined set of boundary conditions and parameter values for measurement uncertainties and loss patterns was established, on the basis of which a number of sequential statistical test procedures was evaluated. One important condition for the application of the NRTA measure was the stipulation that routinely measured Pu inventories in process tanks only, would be used, since more than 95% of Pu inventories in the process MBA are in these tanks. About 12 kg of Pu, expected to be the normal inventory in six pulse columns, was assumed to be constant. In spite of the simplifications made and the fact that mainly simulated data were used, these investigations permit the conclusion that the NRTA measure provides a greater sensitivity in terms of the amounts which can be detected and the timeliness of detection, than the conventional material accountancy. Since measurements are restricted to process tanks only, routinely available measurement techniques can be used. The main thrust of R & D activities has to lie in the practical demonstrations of this measure under operating conditions, some of which are already under way.

Zusammenfassender Bericht des internationalen Ausschusses zur dynamischen Bilanzierungsmaßnahme

ZUSAMMENFASSUNG

Im Dezember 1980 wurde ein internationaler Workshop gegründet, um die Leistungsfähigkeit und die Grenzen der dynamischen Bilanzierungsmaßnahme (Near-Real-Time Accountancy - NRTA - Measure) zu untersuchen. In dem vorliegenden Bericht sind die Arbeiten und Ergebnisse dieses Workshops bis Juli 1982 zusammengefaßt.

Nach der Festlegung der verfahrenstechnischen und Kernmaterialbilanzierungsdaten für eine 1000 t Schwermetall/Jahr Wiederaufarbeitungsanlage wurden Simulationsmodelle entwickelt, die u.a. das Verhalten des Prozeßinventars feststellen sowie die erforderlichen Plutoniumdurchfluß- und Inventardaten für die Prozeß-MBA sequentiell erzeugen können. Die so erzeugten Datensätze wurden verwendet, um die Aussagefähigkeit von 8 der bisher im Zusammenhang mit der NRTA angewendeten statistischen Testverfahren für einige wohldefinierte Pu-Verlustszenarien aus der Prozeß-MBA zu prüfen.

Trotz einiger vereinfachender Annahmen und der Tatsache, daß nur simulierte Datensätze verwendet wurden, zeigte die in dem Workshop untersuchte NRTA-Maßnahme eine höhere Empfindlichkeit bezüglich der rechtzeitigen Entdeckung einer gegebenen Menge gegenüber der konventionellen Materialbilanzierung. Da die Pu-Messungen in erster Linie auf die Prozeßbehälter beschränkt bleiben (da sich mehr als 95% des Pu-Iventars im Prozeß in diesen Behältern befinden), können die für betriebliche Zwecke verwendeten Meßmethoden für die NRTA-Maßnahme eingesetzt werden.

Die Hauptrichtung der zukünftigen Entwicklungsarbeiten sollte in der praktischen Demonstration dieser Maßnahme unter routinemäßigen Betriebsbedingungen einer Wiederaufarbeitungsanlage liegen. Einige dieser Untersuchungen sind bereits eingeleitet worden. Title of the main paper:

Overview Report of the International Workshop on the Near-Real-Time Accountancy Measure

Appendix from Page 25 onwards.

.

INTRODUCTION

An International Workshop on the near-real-time accountancy (NRTA) measure was established in December 1980 to investigate the capabilities and limitations of this measure for a large-scale reprocessing facility. The present overview report summarizes the activities and the results of this workshop as of July 1982.

NRTA has been considered by many as one of the promising measures which could extend the capabilities of present-day international safeguards. However, the fairly large volume of published information on this subject in the recent past may not always enable the reader to form an objective and unbiased opinion on the applicability of such a measure.

The International Workshop was specially established to develop a set of data base and guidelines with the help of which the capabilities and robustness of the NRTA measure can be investigated.

Organizations and internationally known experts from Japan, the USA and the Federal Republic of Germany, actively engaged in this area, were invited to provide advice and information as well as contribute actively to the workshop. The overview report is prepared on the basis of these contributions.

The contributing organizations and experts actively taking part in the workshop meetings and activities are listed on page I.

The International Workshop has covered up to now the following areas:

- Generation of throughput and inventory data for Pu for a reference layout of a reprocessing facility with a capacity of 1000 t HM/a.
- (2) Measurement and other estimating systems for the generation of material balance data for the reference facility.
- (3) Simulation of relevant process characteristics of the reference facility (in particular the behaviour of inprocess Pu inventory under normal and unstable process conditions).
- (4) Establishment of boundary conditions for comparison of different statistical procedures for the evaluation of the material balance data with regard to different loss patterns including a possible diversion.
- (5) Selection of relevant sequential statistical test procedures.
- (6) Preliminary comparison of different statistical test procedures to evaluate their capability.
- (7) Identification of areas in which specific R & D activities would be required for demonstrating the possibility of implementing NRTA as an international safeguards measure on a routine basis.

In the remainder of this report these subjects are treated one by one.

1. The reference reprocessing facility

An extremely simplified block diagram of the reference reprocessing facility is presented in Fig. 1, with some of the information relevant to NRTA on throughput and inventory of plutonium summarized in Table I |1|. Table II presents data on plutonium inventories in various process tanks.

According to this layout about 95% of the process inventory of Pu is expected to be in about 25 process buffer tanks under normal operating conditions. This has been found to have important simplifying consequences for the NRTA measure. Since the main measurement efforts for assaying Pu process inventory are restricted to these tanks, most of the measurement data can be generated with well-tested measurement methods already existing for operating purposes for Pu assay in accountability tanks in reprocessing facilities. In case the Pu inventories in pulse columns become a relevant problem, simplified simulation methods for estimating these inventories could be used.

2. <u>Measurement systems for the generation of material</u> balance data 2,3

In establishing the requirements and characteristics of measurement systems to be considered for the generation of the required material balance data for the NRTA measure, a number of boundary conditions were stipulated:

- The plutonium amounts in inventories and in flows to and from the process material balance area (MBA) will be determined, as far as practicable, on the basis of measurements carried out in connection with the normal operation of the facility.
- As the first alternative, only the process tanks (and not process equipment or pipelines) will be assayed to establish the plutonium inventory in the process. Such inventories will be established during the operation of the process (physical inventory taking without plant shut-down).
- The plutonium inventories in process equipments and pipelines (which correspond to approximately 6-7% of the total plutonium inventory in the process) will be considered to be approximately constant or fluctuating within a given range (e.g. +10%).
- In most of the cases, the throughput and inventory of Pu will be assayed in tanks. The following steps may be required for a measurement system for determining the Pu amount in such a tank:
 - Homogenization of the solution
 - Measurement of the respective tank volume
 - Sampling from the tank
 - Sample transport to the analytical measurement system

TABLE I. CHARACTERISTICS OF THE 1000 t HM/YEAR REFERENCE REPROCESSING FACILITY

Throughput	1000 t 10 t	HM/a Pu/a	=	5 50	t kg	HM/d Pu/d
Pu inventory (kg)						
Process tanks		· .		52	25	(feed adjustment - product accounting; about 25 process buffer tanks with significant quantities of Pu)
Process equipment						
Pulse columns	12					
Mixer settler and oxidation equipment	6					
Evaporator	12			3 55	5	

TABLE II.

.

Pu INVENTORY IN TANKS (MAIN PROCESS STREAM

		Number	Max.inv./tank (kg)	
lst Cycle	ACC.TANK	2	50	
	ADJ.TANK	4	50	
	HAF-TANK	2	50	
	1BXP-TANK	1	10	1
	1 RP-A-TANK	1	3.4	i.
2nd Cycle	2 AF-TANK	5	17	
	2RP-A-TANK	1	5.7	1
3rd Cycle	3AF-TANK	3	57	
······	3RP-A-TANK	1	5.0	
- - -	3RP-TANK	2	100	
Pu concentrat	ion			
	4AF-TANK	1	25	
	4AC-TANK	2	125	1
	Total	25		

- Sample conditioning
- Measurement of the plutonium concentration
- Calculation of the plutonium inventory in the tank.
- (NOTE: The main investigation was directed to the measurement systems involved in the determination of plutonium concentration, although the step involving sample conditioning could be the decisive one for establishing the suitability of a system. This fact was taken into account in the total time required for a given measurement system).
 - The total measurement time including all the steps should be, whenever possible, less than the residence time of a Pu solution in a tank (e.g. in the range of 8-24 hours). In this manner the frequency of the NRTA will be determined by the inherent process parameter involving the residence time of the Pu-containing solution in a tank and not by the delays in the measurement system. In addition, the verification of the operator's measurement data could be carried out in principle during the time the solution is still in the tank.

The plutonium flows and characteristics of the process materials relevant for NRTA, are presented in Table III.

Process stream	Plutonium flow (kg Pu/a)	Material description (Approximate Plutonium Concentration; activi- ty)
Input tank	10 000	$1-2 \text{ mg/m1} (\alpha, \beta, \gamma)$
End product	10 000	250 mg/m1 (α)
Centrifuge waste	20	50 g/g (α, β, γ)
Leached hulls	. 10	$1 g/g (\alpha, \beta, \gamma)$
High active waste (HAW)	40	$1 g/g (\alpha, \beta, \gamma)$
Medium level/low active wastes (MAW/LAW)	3	$1 g/g (\alpha, \beta, \gamma)$

TABLE III. PLUTONIUM FLOWS, CONCENTRATIONS AND ACTIVITIES IN DIFFERENT PROCESS STREAMS OF THE REFERENCE FACILITY

It is to be noted that:

- Both the Pu-concentrations and the radioactivity in the different process streams undergo variations by several orders of magnitude.
- The sample material in the different process streams in the facility can be broken down into four categories:
 - The input solution,
 - The end product,
 - The liquid waste,
 - The solid waste.

It has been assumed that for the first three categories of solution chemical analytical methods would be routinely used whereas for the fourth category (solid wastes) non-destructive measurement methods would have to be applied.

Different measurement methods were investigated in the context of their application possibilities, for the input product and waste streams.

After taking into consideration the different characteristics of the possible measurement systems, a set of values for measurement errors (1 σ) were established for carrying out the required sensitivity study for the NRTA measure. These measurement errors which are somewhat on the conservative side are presented in Table IV. For the purpose of this study the values for the random (σ_R) and the systematic (σ_S) errors have been assumed to be the same for the reference case.

TABLE IV. $\frac{\text{MEASUREMENT ERRORS FOR THE DIFFERENT}}{\text{FLOWS AND INVENTORIES IN THE}}$ REFERENCE FACILITY ($\sigma_{R} = \sigma_{S}$)

Object of measurement	Measurement errors (1σ) (%)
Input accountability tanks Product output	1.0 0.3
Centrifuge wastes	25
Leached hulls	50
HAW	25
MAW, LAW	25
Inventory in process tanks	1.0

The unmeasured inventory corresponding to an average of 30 kg of plutonium in the different process equipment and pipelines, is assumed either to remain constant or to fluctuate by $\pm 10\%$ around the average for the different case studies.

3. <u>Simulation of relevant plutonium flow and</u> inventory characteristics 4-6

Model simulation has been considered to play an essential role in evaluating the capabilities of the NRTA measure. This is of two types.

The first type deals with the mathematical modelling of the relevant plutonium extraction and purification steps in the reference facility and simulation of the distribution and flows of plutonium in these steps. The input data for such simulation studies are obtained from the data base for the process layout of the reference facility. This type of simulation is expected to indicate the behaviour of the flow and inventory of plutonium in the process MBA under start-up, normal and some abnormal operating conditions. This process model is then used for the simulation of accountancy data for establishing MUF values and their associated uncertainties, for different values of measurement uncertainties, fluctuations in the inventories and for different throughputs and inventories. The accountancy data thus generated in the simulation model are then used for applying different statistical test procedures for generating safeguards-relevant conclusions with regard to the status of the plutonium in the process area. The capabilities of such test procedures can also be evaluated.

This type of simulation model is necessary for analysing the capability and limitations of the NRTA measure under different facility conditions. How much confidence one can place in the result of such models depends on how well they can simulate the actual conditions prevailing in the facility. It is, therefore, important that data based on actual operating conditions in a facility be used in such models. The initial results from such a simulation model are presented elsewhere in these proceedings |4|. It is to be noted that these models are not required for routine implementation purposes of the NRTA measure.

The other type of simulation model which can be a subset of the first type, deals with the estimation of plutonium inventory in process equipment such as pulse columns. Under the present series of investigations, the measured data base are generated mainly from measurements in process tanks. The unmeasured inventories in the process equipment are assumed to remain constant or vary within a limited range of +10%. If this inventory is found to vary much more under routine operating conditions than the assumed rates, the sensitivity of the NRTA measure would automatically go down. For such cases (which might be remote), some possibility of obtaining a value for the plutonium quantity in this equipment on the basis of some derived estimates would be useful. Such models are expected to be as simple as practicable and to be based on a few measurable and verifiable data. If required, they would then form part of the measurement systems for generating the accountancy data on a routine basis for the NRTA measure.

Up to now three different types of model for estimating the plutonium inventories in the pulse columns of the reference facility have been developed.

3.1 Exponential model

The exponential model for prediction holdup of special nuclear material in pulse columns provides a simplified method for approximating the in-column inventories. In this model it is assumed that the plutonium concentration profiles in the extraction section of the column vary exponentially. The model is intended for steady-state operation only and does not take into consideration the many complex variables that affect column performance and holdup.

3.2 Ideal stage model

In the ideal stage model, the holdup of plutonium in a pulse column is estimated by summing up the plutonium amounts per stage (i.e. average plutonium concentration x liquid volume per stage). The holdup of plutonium in this model is ultimately a function of the plutonium concentration and flow rates of the aqueous feed and the organic extractant at the input and output of the column, the separation coefficient for extraction, the number of ideal stages and the total volume of the column. In this model also, complex effects such as back mixing, temperature and molarity dependence of distribution coefficients etc., are neglected.

3.3 Reduced-order linear model

The reduced-order linear model is a linear inventory estimator based on first-order perturbations about an expected steady-state value. The steady-state inventory value is calculated for the expected operational conditions using a detailed chemical model that has been validated experimentally for the particular contactor system. Alternatively, experiments can be performed directly to determine the expected inventory by bringing the contactor to steady state and then draining the contents to holding tanks for measurement.

The column inventory calculations are based on the following assumptions:

- The column is operating near a steady-state operating point.
- The column inventory near the operating point is linear in the concentrations.
- Concentration and flow-rate measurements are available in near-real time.
- The column inventory at the nominal operating point has been previously determined from chemical model calculations and calibration experiments.

A realistic simulation of solvent extraction columns at at least near-equilibrium operation is essential if the respective models are to be of use for in-process holdup estimation or as elements of an overall process simulator.

In the first stage of the investigations, these three models were tested on the basis of the flow sheet data for pulse columns for the reference facility. The exponential model was also tested for the pulse columns of the Barnwell facility, for which experimental data for a uranium stream are available [7]. The responses (for plutonium inventory changes) of these models to 10% increases in feed and extractant flow rates and concentrations, show that the holdup variations are roughly proportional to the changes in input flows and concentrations. Further investigations with realistic operational data are required to demonstrate the usefulness of these models. 4. Boundary conditions and parameters for comparison and evaluation of different statistical test procedures

During the workshop activities a uniform set of boundary conditions and parameters was established for the comparison and evaluation of different statistical test procedures. One of the difficulties in the past in assessing the capability of the NRTA measure using different test procedures, had been the lack of a uniform set of such conditions for comparison.

From a large number of individual conditions |8-11| the more relevant ones are summarized as follows:

4.1 Boundary conditions

4.1.1 The evaluation will consider a series of determinations of the inventories and transfers corresponding to a single MBA. Inventories will be assumed to be taken at times t. (without stopping the operation of the facility) starting at some initial time t and continuing indefinitely. It will be possible to consider:

- a) All the data
- b) All the data subsequent to t
- c) All the data obtained during some fixed period T.
- 4.1.2 The data will be characterized by
- a) Assumed true values μ_1 of the inventories at time t. In simulation studies these are derived from the process simulation.
- b) Assumed errors of determination of net transfers and inventory. These are propagated from an assumed set of measurement errors, accounting procedures and covariance structure.
- c) Assumed losses L. in the periods ∆t = t.-t. between inventories. These may be present in known or unmodelled process errors, long-term or unmodelled measurement biases, or deliberate diversion.

4.1.3 In this exercise the primary concern is to estimate and test hypotheses concerning the L.. The μ_{1} are modelled since (1) assumptions concerning these may effect the ability to estimate the L. and (2) in more general situations it may be desirable to estimate or test inventories as well as losses. 4.1.4 Characteristics of loss patterns are to be considered.

- a) The evaluation is concerned with the total loss M during T which may be, for example, a calendar year or a campaign. The individual loss $L_1 = \gamma_1 M$ within this period are of concern only as the patterns, γ_1 affect the ability to estimate and test M.
- b) Within T, to take account of the nature or timeliness of losses, the time period of the initial loss, the time at which the amount M was assumed to be available and the form of diversion (e.g. abrupt vs protracted) will have to be considered.
- c) Because of externally imposed conditions (legal, administrative) the evaluation procedures have to be concerned with the losses within T from τ_1 to τ_2 following τ_0 . Several sub-cases need to be considered:
 - The existence of a base set of data known to be loss-free.
 - Only the cases L_i>0 will be considered.
 - There exists a time τ_3 by which detection of the losses between τ_1 and τ_2 should be achieved.
- 4.2 Parameters considered for the evaluation of statistical methods

4.2.1 Definitions

The following errors are treated in this section:

- σ_{η} Random error standard deviation in measuring inventory
- $\sigma_{\epsilon} \mbox{-Random error standard deviation in } \\ measuring net transfers or flows } \label{eq:standard}$
- $σ_{\delta}$ Systematic error standard deviation in measuring net transfers.

Two loss patterns were defined. In loss pattern No.1 the loss per time interval is uniform, beginning at interval I (one parameter) and extending over m intervals (a second parameter). In loss pattern No. 2, I and m are defined as in loss pattern 1 except that the loss pattern is not uniform over these m intervals; it alternates by $\pm 50\%$ about the central value, M/m.

The marameter values were defined relative to the value for σ , which is fixed at one unit throughout. Also, σ_{δ} is expressed relative to σ_{ϵ} , i.e. $\sigma_{\delta}/\sigma_{\epsilon}$ is one of the factors varied.

4.2.2 Parameter values

In the first phase of the investigations, the following sets of parameter values were selected for uniform loss patterns (for both abrupt and protracted). The alternative loss patterns (loss pattern No. 2) have not been investigated in detail:

> $\sigma_{\varepsilon} = 0.1, 0.55$ $\sigma_{\delta} / \sigma_{\varepsilon} = 2.5$ M = 15, 25 $I_{o} = 1, 11, 21$ m = 5, 10

The cases investigated for the different statistical test procedures were 24 in number, consisting of all the possible combinations of the above-mentioned parameter sets. In addition, for I = 11, m = 5, and $\sigma_{\rm c}$ = 0.1 and 0.55, two cases were run at M = 0 to determine the values of the false alarm probability, α . The case number identification is given in Table V.

TABLE	V.	CASE	NUI	IBER	IDENTI	FICATION	FOR	THE
		SET	OF	PAR	AMETERS	INVESTI	GATEI)

			(σ _δ /σ _ε	= 2.5	for all the	e case	s)	1999-99-99 - 99 - 99 - 99 - 99 - 99 - 9	9 7 107 1 7 9 1 7 9 1 9 1 9 1 9 1 9 1 9 1 9 1
Case	σ ε	М	I _o	m	Case	σ ε	М	Io	m
1	.1	15	1	5	14	.55	15	1	10
2	. 1	15	1	10	15	.55	15	11	5
3	. 1	15	11	5	16	.55	15	11	10
4	. 1	15	11	10	17	.55	15	21	5
5	.1	15	21	5	18	.55	15	21	10
6	. 1	15	21	10	19	.55	25	1	5
7	. 1	25	1	5	20	.55	25	1	10
8	. 1	25	1	10	21	.55	25	11	5
9	.1	25	11	5	22	.55	25	11	10
10	.1	25	11	10	23	.55	25	21	. 5
11	. 1	25	21	5	24	.55	25	21	10
12	.1	25	21	10	25	. 1	0	11	5
13	.55	15	1	5	26	.55	0	11	5

5. <u>Description of the statistical test procedures</u> investigated

The values realized for accountancy data were generated with the process simulation model for the different parameter sets identified under section 4.2, with the assumed uniform loss patterns for different values of M. Under the present series of investigations these loss patterns were supposed to be detected with the help of different statistical hypothesis testing procedures. The probability of detection P_D was taken to be the indicator for the sensitivity of a test procedure for a given set of accountancy data parameters, loss pattern and the value for α .

— 11 —

Under this approach the observed material accounting data generated sequentially are applied to test the hypothesis H of no material loss against the alternative hypothesis H₁ of material loss. Such tests are of two types: The fixed length test in which a predetermined number N of balances are observed before deciding between H and H₁, and the sequential test in which the possibility of a decision is allowed after each balance is observed.

For the purpose of checking the sensitiveness, eight basic statistical test procedures were selected amongst those normally considered for NRTA measures. A short description of these tests follows |11|.

5.1 MUF

The MUF test is a test on the material balance for a given period. Letting D. be the observed MUF for period i, loss detection is said to occur if D, exceeds some critical value determined by the value of α and the values of the measurement error standard deviations. The MUF test does not take into a account any prior history. It is aimed at detecting an abrupt loss, one that occurs somewhere within the material balance period in question. As a test sequence, the MUF test is applied at each material balance period and loss detection over the P periods occurs if at least one MUF exceeds its critical value. The α value over all P tests is controlled by reducing the size of the significance level for each individual test.

5.2 CUMUF

The test statistic to be applied in period i is denoted by T. and is the sum of the individual observed MUFs beginning at some point in time and extending through period i:

$$\Gamma_{i} = \sum_{j=1}^{i} D_{i}$$

At a given point in time, T. is independent of how the losses are distributed throughout the i periods. This is the cited advantage of the CUMUF test. As a test sequence, the CUMUF test is applied at each material balance period, as is the MUF test. Clearly, there is a close correlation between successive CUMUFs.

In this study, CUMUF is applied in sequence on the one hand, and only at the end of the 35 periods on the other. The single test in this latter instance is, of course, more powerful than is the 35th such test applied as the last test in the sequence. However, this increase in power is counterbalanced by the lack of timeliness, i.e., the inability of the test to detect losses that occur early in the sequence of time periods.

5.3 <u>Uniform diversion</u> (D₁₁)

The test statistic is designed to detect uniform losses. Since uniform losses over a number of successive balance periods were the primary loss patterns studied in this phase, it would be expected that the uniform diversion test statistic would exhibit good detection capabilities in this study.

The linear statistic in question is the minimum variance unbiased estimate of uniform loss. Specifically, in this study, the statistic was defined for each group of four successive MUFs. It is a moving weighted average of four such MUFs, and it is clear that successive test statistics would be closely correlated.

The weighted average is derived as follows. Let

$$T_{i} = a_{1}D_{i}^{+}a_{2}D_{i+1}^{+}a_{3}D_{i+2}^{+}a_{4}D_{i+3}^{+}a_{4}D_{i+3}^{+}a_{5}D_{i+2}^{+}a_{4}D_{i+3}^{+}a_{5}D_{i+5}^{+}a_{5}^$$

where the a,'s sum to 1 for j=1, 2, 3, 4. The a.'s are chosen to minimize the variance of T.. The first test statistic is calculated at the end of the fourth balance period.

When j=1-4 as here, the calculation of the a.'s is quite simple. For more complex cases, calculational algorithms are helpful. The oft-mentioned Kalman filter is a calculational algorithm used in this instance.

5.4 CUMUFR

CUMUFR is an acronym for cumulative sum of standarized MUF residuals. It is designed to detect changes in loss patterns. A uniform loss that occurs in all balance periods would not be detectable with the CUMUFR test.

The MUF residual for period i, MUFR;, is defined as

$$MUFR_{i} = D_{i} - E(D_{i}/D_{1}, D_{2}, ..., D_{i-1})$$

where $E(D_i/D_1, D_2, ..., D_{i-1})$ is an appropriate linear function of $D_1, D_2, ..., D_{i-1}$, chosen such that MUFR, has minimal variance. The standardized MUF residual is found by

dividing MUFR. by its standard deviation σ_{i} , and the CUMUFR test statistic for balance period k is found by summing MUFR./ σ_{i} from 1 to k.

The time series of MUFRs is a linear transformation of the time series of MUFs. They can be calculated exactly by applying this transformation or approximately through use of a Kalman filter.

The CUMUFR test may be applied as a two-sided test or as a one-sided test. In a two-sided test application, periods of losses followed by periods of no losses would also be detectable, whereas for a one-sided test, only periods of losses following periods of no losses would be detectable.

Note that in applying the CUMUFR test sequence, use is always made of all the MUF data extending back to period 1. This is a principal distinction in this study between CUMUFR and the D_n test discussed next.

5.5 <u>D</u>_n_

Like CUMUFR just discussed, D is aimed at detecting changes in loss patterns. Unlike CUMUFR, in this study n was fixed at 5, i.e. at the end of each balance period, and beginning with period 6, the current MUF is compared with some constant β times the sum of the 5 previous MUFs where β is chosen to minimize the variance. Specifically, the test statistic for period i is

 $T_{i} = D_{i+5} - \beta \sum_{\substack{j=1\\j=1}}^{i+4} D_{j}$

where β is a simple function of the error variances in measuring net transfers and inventories.

In this study, the test was applied as a one-sided test. Thus, a period of losses followed by a period of non-losses would not be detectable.

5.6 Sequential probability ratio test

The sequential probability ratio test is related to the CUMUF test in that the test statistic is the cumulative sum of the MUFs. However, the test is now a sequential test in the true sense of the word, as distinguished from a sequence of fixed length tests.

With the sequential test, when the value of the test statistic is calculated at the end of each period, the decision is made to either reject the hypothesis of no loss (i.e. declare that a loss has been detected), accept the hypothesis of no loss, or continue testing. When the hypothesis of no loss is accepted, then the test is restarted, and all prior data are ignored. This restarting of the test and deletion of prior data is what distinguishes the sequential probability ratio test from the CUMUF test described earlier. With the CUMUF test, the MUF data extending backward to period 1 are always retained.

5.7 Modified pages test

The modified pages test is also a sequential test in that the test may be restarted with all prior data eliminated when the accumulated evidence indicates that there has been no loss of material.

For the modified pages test, the test statistic is

$$T_{i} = C_{i} - \min_{j} C_{j}$$

$$\sum_{k=0}^{j} (D_{k} - \delta).$$

where $C_j = \sum_{k=1}^{J} (D_k)$

In effect, the test statistic is the current CUMUF minus the largest previous CUMUF, δ being a constant.

The upper threshold (critical value) is a function of some parameter, A, which controls the false alarm rate, and of the period number i. The lower threshold is zero for the modified pages text.

5.8 Truncated sequential CUMUF

Like the sequential probability ratio test, the basic statistic is the cumulative sum of the MUFs. Also, the test is sequential in nature. This test procedure is called a truncated one because after a fixed number of material balance periods, a decision must be made as to whether or not a loss has occured.

In evaluating this test procedure, a saddle-point solution is also found. The saddle point solution gives a guaranteed efficiency in the sense that it gives the detection probability corresponding to the least favourable loss pattern, i.e. it reacts to a diversion scenario in which the adversary chooses an optimum strategy.

6. Test results

Eleven statistical test procedures were investigated including some variations and comparisons of the eight basic testing procedures described under Section 5 for the 26 sets of parameter variations indicated in Table V. These eleven cases are identified in Table VI.

6.1 Comparison of results

The results of investigations of the eleven statistical test procedures are summarized in Table VII. As mentioned earlier, the investigations in this phase were restricted to the uniform loss patterns.

TABLE VI.

IDENTIFICATION OF STATISTICAL TEST PROCEDURES INVESTIGATED

Case identification	Case type	Reference
TS-1	Standard MUF Test	Jaech 11
TS-2	CUMUF Test	Jaech 11
TS-3	D ₁ Test	Jaech 11
TS-4	D, Test	Jaech 11
TS-5	Třuncated Sequential CUMUF Test	Beedgen 12
TS-6	CUMUFR Test; two- sided sequential test with power one	Sellinschegg 13
TS-7	CUMUF; sequentially performed fixed length test	Sellinschegg 14
TS-8	CUMUF (35); fixed length test at the end of 35 periods	Sellinschegg 14
TS-9	CUMUFR Test; one- sided sequential test with power one	Sellinschegg 14
TS-10	Sequential Probability Ratio Test	Markin 15
TS-11	Modified Pages Test	Markin 15

The results presented in Table VII illustrate some interesting points. Remembering that in this phase the simulation model has assumed that the inventories in the process columns would remain constant and that the systematic error components for the inventory measurements would cancel out (because of the fact that these data are generated as the difference of two measured values), the results are to be considered as indicating the highest sensitivity to be expected from the statistical test procedures, using the measurement data for the reference facility for the assumed uniform lost patterns.

a) The cases 25 and 26 give the actual α -values. When comparing detection probabilities, these differences should be kept in mind. Ideally they should be all about 0.05 for a fair comparison, but it is difficult in some cases to fix α precisely in advance. This would mean that for $\alpha = 0.05$ the P_D values for the test procedures like

TABLE VII.

$\underline{\text{DETECTION PROBABILITIES P}_{D} \text{ FOR PARAMETERS}}$

IDENTIFIED IN TABLE V. AND STATISTICAL TEST

PROCEDURES IDENTIFIED IN TABLE VI.

Case	<u>TS-1</u>	TS-2	<u>TS-3</u>	<u>TS-4</u>	<u>TS-5</u>	<u>TS-6</u>	<u>TS-7</u>	<u>TS-8</u>	<u>TS-9</u>	TS-10	<u>TS-11</u>
1	1.000	.340	1.000	.687	1	1.0	1.0	.52	1.0	.92	1.0
2	.995	.695	.991	.211	1	1.0	1.0	.52	1.0	.27	.94
3	.750	.993	1.000	.682	.978	1.0	.9	.52	1.0	.93	1.0
4	.435	.750	.994	.207	.811	1.0	.72	.52	1.0	.27	.91
5	.299	.993	1.000	.678	.53	1.0	.57	.52	1.0	.92	.96
6	.192	.745	.995	.204	.355	1.0	.48	.52	1.0	.29	.80
7	1.000	.894	1.000	1,000	1	1.0	1.0	.88	1.0	1.0	1.0
8	1.000	.999	1.000	.616	1	1.0	1.0	.88	1.0	.95	1.0
9	1.000	1.000	1.000	1.000	1	1.0	1.0	.88	1.0	1.0	1.0
10	.985	1.000	1.000	.608	1	1.0	.99	.88	1.0	.97	1.0
11	.780	1.000	1.000	1.000	.989	1.0	.96	.88	1.0	1.0	1.0
12	.539	1.000	1.000	.601	.909	1.0	.89	.88	1.0	.96	1.0
13	.301	.038	.296	.257	.583	1.0	.27	.09	.23	.36	.08
14	.103	.043	.094	.120	.20	.99	.07	.09	.10	.17	.05
15	.048	.272	.255	.256	.06	.98	.05	.09	.98	.42	.08
16	.042	.082	.085	.120	.052	.87	.05	.09	.92	.18	.05
17	.033	.270	.246	.256	.052	.96	.05	.09	.97	.42	.08
18	.031	.085	.080	.120	.051	.90	.05	.09	.93	.17	.05
19	.736	.037	.785	.699	.763	1.0	.78	.13	.72	.83	.16
20	.236	.046	.239	.267	.494	1.0	.22	.13	.26	.40	.11
21	.084	.829	.706	.700	.209	1.0	.09	.13	1.0	.85	.12
22	.063	.184	.206	.267	.088	1.0	.08	.13	1.0	.41	.10
23	.046	.829	.710	.700	.064	1.0	.08	.13	1.0	.85	.11
24	.039	.185	.198	.266	.055	1.0	.08	.13	1.0	.41	.10
25	.027	.045	.034	.041	.05	.051	.035	.051	.051	.05	.05
26	.023	.043	.024	.044	.05	.050	.027	.050	.051	.05	.05

16 -

TS-1, TS-3 and TS-7 could be higher than those obtained in the present cases.

- b) It is to be noted that the test sequences TS-1 and TS-7 should give identical results with the TS-1 results calculated by the multivariate normal distribution and the TS-7 results by simulation. Taking into account the differences in the α values, the agreement is good.
- c) The parameter cases 13-18 represent the worst cases considered since the uncertainties with throughput measurements are increased by a factor of about 5 for the same value of M=15 compared to the other cases. There is in general a reduction in P_p values for almost all the test procedures excepting TS-6 (cumulative sum of MUF residuals), in which the detection probabilitites remain fairly high.
- d) Test 6 in fact shows the highest probability values for all the 24 cases investigated in this phase.

6.2 CUMUFR test

Since the particular test procedure TS-6 provided the highest set of probability of detection values investigated so far, this test was investigated in some more detail in the frame of the NRTA Workshop |13|. Using the values of Case 6 in Table VII as a basis, the results of these additional investigations are illustrated in Fig. 2, top diagram, taken from |13|. Converted to the data of the reference facility, Case 6 would correspond approximately to the following absolute values:

	σ _n	-	2.1 kg Pu
	σ	=	0.21 kg Pu
	m	=	l day
	I	=	21 periods
	м .	=	15 (equivalent to ≃30 kg of Pu)
Loss	patterns		(Nos. refer to the Nos. in Fig. 2)
No.	1 2 3 4 5 6 7 8		30 kg diverted over 10 days 20 days 30 days 40 days 50 days 100 days 200 days M = 0 (to obtain α)

Figure 2 illustrates a number of aspects in connection with the test procedures TS-6. They are mainly summarized from |13|.

- a) For abrupt diversion patterns (loss patterns 1, 2, 3) the $P_{\rm D}$ is above 95%.
- b) For protracted diversion patterns (loss patterns 4, 5, 6, 7) the P increases with increasing balancing intervals, i.e. from ≈25% to 78% when the balancing intervals m are increased from 1 day to 10 days.
- c) This sensitivity is obtained by assuming that the loss patterns start after 21 zero loss periods. (I_=21)
- However, the main message which one gets from these d) illustrative examples as well as from those given under Table VII, is the fact that this type of NRTA measure brings about a significant improvement in the capability of material accountancy (in respect of detection probability for a given amount and detection time), over that for the conventional type of material accountancy. This remains valid in spite of the simplified assumptions made in this phase of the investigations. This basic fact is also illustrated through another in-depth work carried out by Ikawa 16 using the Barnwell reprocessing facility (BNFP) as the reference facility. One other important conclusion in that study is the suggestion that if the present chemical process were carried out in two parallel lines with half the processing capacity each, the detection capability for protracted diversion might be significantly improved.

7. Conclusions

The NRTA Workshop has completed its activities involving simplified simulation models of the data for the reference reprocessing facility with 1000 t HM/a. Inspite of the simplified assumptions and the fact that the capability of the NRTA measure could be investigated on the basis of mainly simulated data with little experimental validation, a number of generalized conclusions can be drawn. Some of these conclusions which are similar to those drawn by a group of concultants at an IAEA consultants meeting on this subject and reported upon by Lovett |17|, will not be repeated here.

- 7.1 Measurement systems for the type of NRTA measure investigated in the workshop i.e. sequential generation of the required material accountancy data sets, on the basis of Pu measurements in process tanks only (and not in process equipments), can be based on currently available technology.
- 7.2 Required statistical test methods for evaluating the material accountancy data in generating safeguards-relevant conclusions are available.
- 7.3 The sensitivity of the NRTA measure, in terms of amounts which can be detected with a given set of probability values and timeliness, is higher than that possible for conventional type of material accountancy measure. For

the type of NRTA measure investigated, this sensitivity will go down with high fluctuations of estimated Pu inventories in process equipments.

7.4 The main thrust of the R&D activities have to be in the direction of the validation of simulation models with actual operational data.

The in-depth activities on the basis of which the results presented in this overview report could be generated, were possible only through the support of the involved organizations and the excellent cooperation of the participants.

REFERENCES

- M. Kluth, H.O. Haug, H. Schmieder, Konzept zur verfahrenstechnischen Auslegung einer 1000 jato Purex-Referenzanlage mit Basisdaten für eine Spaltstoffflußkontrolle, KfK 3204, Sept. (1981)
- E. Mainka, Evaluation of Destructive Measuring Techniques Eligible for Plutonium Assay in a Large Reprocessing Plant, Paper presented at the second Meeting of the Workshop at the Karlsruhe Nuclear Research Centre. February (1982), s.p. 93
- 3. M. Kuechle, Note presented at the first meeting of the Workshop at the Karlsruhe Nuclear Research Centre, December (1980), s.p. 47
- M.J. Canty, G. Spannagel, F. Voss, A. Berliner, "Computer Simulation of the Process MBA for a 1000 tonne Reprocessing Facility", Nucl. Safeguards Technology (Proc. Symp. Vienna 1982) I, IAEA (1983), 555-556
- 5. M.J. Canty, H.A. Dayem, E.A. Kern, G. Spannagel, "Simulation of Nuclear Fuel Reprocessing for Safeguards", KFK 3439 (1982)
- J. Shipley et.al., Recommendation of Working Group 2 -Simulation, Paper prepared at the first meeting of the NRTA Workshop, December (1980), s.p. 55
- 7. E.A. Hakkila et al., LA-6881, September (1977)
- 8. C.A. Bennett, R. Avenhaus, Loss Models, Paper prepared at the second International Workshop on NRTA at the Nuclear Research Centre, Karlsruhe, February (1982), s.p. 80
- 9. C.A. Bennett, Parametric Description of Loss Patterns, Paper contributed to the Workshop on NRTA, November (1982), s.p. 161
- D. Sellinschegg, J.L. Jaech, Data Base Variables and Parameters for Data Evaluation Methods, Paper prepared at the second Workshop on NRTA, February (1982), s.p. 83
- J.L. Jaech, Determination of the Detection Probability as a Function of Various Loss Strategies, Phase 2, Part 2, Study Results, Paper prepared for the NRTA Workshop. August (1982), s.p. 197
- R. Beedgen, Truncated Sequential Test Procedure Using the CUMUF Statistic for a Timely Detection of Diversion, Nucl. Safeguards Technology (Proc. Symp. Vienna 1982) II, IAEA (1983), 383-392

- D. Sellinschegg, U. Bicking, MUF-Residuals Tested by a Sequential Test with Power One, Nucl. Safeguards Technology (Proc. Symp. Vienna 1982) II, IAEA (1983), 393-406
- 14. D. Sellinschegg, Paper prepared for the NRTA Workshop, August (1982), unpublished (for theoretical consideration see Journal INMM, Vol.XI,4, pp 48-59, 1982)
- 15. J.T. Markin, Paper prepared for the NRTA Workshop August (1982), s.p.167
- 16. K. Ikawa, Preliminary Evaluation of Near-Real-Time Material Accountancy Models in a Large Scale Reprocessing Plant, Paper presented to the NRTA Workshop August (1982), s.p. 209
- J.E. Lovett, K. Ikawa, J. Shipley, D. Sellinschegg, Near-Real-Time Material Accountancy - A Technical Status Report, Nucl. Safeguards Technology (Proc. Symp. Vienna 1982)I, IAEA (1983), 487-512

Appendix

For contents see p. 25.

Appendix

Some papers and notes, submitted to the NRTA Workshop for discussion or for inclusion in the Overview Report, and not published elsewhere, as well as the minutes of the two main meetings of this workshop, which contain some relevant information, are presented in this Appendix.

Pages No.

Contents

- Minutes of the First Meeting Dec. 8-12, 1980, containing 27-63 among others:
 Estimate of measurement uncertainty for a material
 - balance in a 1000 t HM/yr. reprocessing facility. (Ref. 3 of Overview Report)
 - J. Shipley et al, Recommendation on Simulation Work. (Ref. 6 of Overview Report)
- 2. Minutes of the Second Meeting Feb. 24-26, 1982, containing 65-91 among others:
 - C.A. Bennett; R. Avenhaus, Loss Models. (Ref. 8 of Overview Report)
 - D. Sellinschegg; J.L. Jaech, Data Base Variables and Parameters for Data Evaluation Methods. (Ref. 10 of Overview Report)
- 3. <u>Measurement Systems</u>
 - 3.1 E. Mainka, Evaluation of Destructive Measuring Techniques 93-103 Eligible for Plutonium Assay in a Large Reprocessing Plant. Submitted at the Second Meeting (Ref. 2 of Overview Report).
- 4. Simulation Studies (all papers submitted at the Second Meeting)
 - 4.1 M. Canty, G. Spannagel, F. Voß, Results on Inventory 105-116 Simulation Activities
 - 4.2 G. Naegele, Activities Regarding a Simulation Model. 117-127
 - 4.3 J.E. Lovett, On the Estimation of Solvent Extraction 129-137 Pu-Inventories in NRTA.
- 5. Evaluation Methods and Results
 - 5.1 R. Avenhaus, Criteria for Decision Procedures for a 139-149 Sequence of Inventory Periods. First Meeting.
 - 5.2 J.E. Jaech, Test Descriptions. (Prepared for the Overview 151-156 Report)
 - 5.3 H. Nishimura, On Evaluation Methodology for NRTA. 157-159 Private communication. Second Meeting
 - 5.4 C.A. Bennett, Parametric Description of Loss Patterns. 161-166 Second Meeting

			- 4805 110
	5.5	J.T. Markin, Timely Detection of Materials Loss. Second Meeting. (Includes some results generated subsequent to the meeting.)	167-185
	5.6	J.E. Jaech, Determination of the Detection Probability as a Function of Various Loss Strategies - Phase 2, Part 1: Design of Study. (Prepared for the Overview Report)	187-196
	5.7	J.E. Jaech, Determination of the Detection Probability as a Function of Various Loss Strategies - Phase 2, Part 2: Study Results. (Prepared for the Overview Report)	197-208
6.	K. Acco Subi	Ikawa, Preliminary Evaluation of Near-Real-Time Materials ountancy Models in a Large Scale Reprocessing Plant. mitted to the Rapporteur August 1982.	209–239
7.	к. 1	Ikawa, H. Ihara, H. Nishimura, M. Hirata, H.Sakuragi, M. Ido, Sawahata, M. Tsutsumi, M. Iwanaga, N. Suyama and J.E. Lovett,	

T. Sawahata, M. Tsutsumi, M. Iwanaga, N. Suyama and J.E. Lovett, Final Report of TASTEX Task F: Study of the Application of Near-Real-Time Materials Accountancy to Safeguards for Reprocessing Facilities, to be published as JAERI Report. This publication contains information on measurement techniques to be used for NRTA and on flow and inventory simulation activities for evaluation of NRTA, which were submitted to Workshop Meetings on NRTA for consideration.

not included

Pages No.

1. Minutes of the First Meeting, December 8-12, 1980 (Pages 27-63)

				Page
Annex	I	-	List of Participants	33
Annex	II		Agenda	34
Annex	IIIa	-	Discussion Paper on Required R+D Work	35
Annex	IIIb	1003	Introduction to the Workshop Meeting	42
Annex	IVa		Estimate of Measurement Uncertainty for	
			a Material Balance	47
Annex	IVb	-	Recommendations of Working Group 1:	
			Measurement Techniques	50
Annex	V		Recommendations of Working Group 2:	
			Simulation	55
Annex	VI		Recommendations of Working Group 3:	
			Evaluation Methodology	61

.
. . . .

MINUTES OF THE FIRST MEETING of the Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

> Nuclear Research Center Karlsruhe December 8-12, 1980

1. List of Participants: ANNEX I

2. Agenda: ANNEX II

3. Main Results of the Meeting

3.1.1 Purpose of the Meeting

The purpose and boundary conditions covering the R+D activities for near-real-time accountancy measures (NRTA) were explained by Mr. Gupta. The main points are summarized in ANNEX IIIa,b (a: discussion paper on R+D activities for near-real-time accountancy measures, b: Introduction to the Meeting).

It was emphasized that the capabilities and limitations of the NRTA measures are to be established on the basis of well-founded R+D activities by international expert groups working in this field. These activities would not prejudice in any way the future application possibilities of near-real-time accountancy measures.

3.1.2 Reference Facility

Mr. Voß introduced the flow sheet for the 1000 t U/a reference facility. Mr. Küchle used the throughput and inventory data from this flow sheet to illustrate the influence of different measurement accuracies on the uncertainties of MUF values from such a facility. Only the systematic error components of a measurement system are relevant in this connection. For a wide range of values for systematic errors for the different throughput streams and the inventory, that for the measurement of plutonium in the accountability tank was found to be the most important single value determining the MUF uncertainty.

3.1.3 Previous Experience on NRTA

Mr. Hough (IAEA) described shortly the practice of NRTA at the two Hanford reprocessing facilities (the first facility operating during the period 1952-1962, the second facility operating from about 1958 onward). The inventories of plutonium in the running facilities were established monthly mainly by using measurement data for the process tanks (volume, density and acid concentrations). These measurements were made routinely for process operation. Mr. Hough was responsible for the measurement quality program and MUF evaluation in these facilities.

He also reported on the NRTA method proposed for the AGNS-Barnwell facility. Recently, the operators of this facility described this method in the course of a seminar at the IAEA Headquarters, Vienna. According to them, the process tanks and equipment in the Barnwell facility are instrumented in such a way that the total inventory in the process area can be assayed and registered at frequent intervals without stopping the plant operation. The same measurement data base as in the case of Hanford facilities, i.e. volume, density and acid concentrations, is used for this determination. The procedure has been demonstrated with uranium in the Barnwell facility.

3.2 <u>Recommendations of the Working Groups</u>

Following three working groups were established to identify and prepare recommendations for the R+D work:

	working Group
1	"Measurement Techniques"

2 "Simulation"

. .

3 "Evaluation Methodology"

Rapporteur

M. Küchle (KfK)

J. Shipley (LASL)

R. Avenhaus (Hochschule der Bundeswehr München)

The respective rapporteurs gave a short introduction on their subject at plenary meetings. The participants then separated into working groups to prepare the recommendations. The recommendations were discussed at plenary meetings to incorporate modifications, if any. The introductory remarks by M. Küchle and the recommendations of the working groups are presented in ANNEXES IV a+b (Introductory Remarks, M. Küchle; Measurement Techniques), V (Simulation) and VI (Evaluation Methodologies). A summary of recommended R+D tasks is presented at the end of each of annexes IVb, V and VI.

3.3 Expert Groups Expected To Participate In R+D Activities

- 3.3.1 The organizations and experts invited so far to participate in the different R+D activities were identified.
 - (1) Measurement techniques:IAK, INR, IHCh, IRCh, PWA (KfK); Los Alamos (U.S.A.)
 - (2) Input data for process simulation:DWK; GWK; IHCh (KfK)
 - (3) Data on operating characteristics for the reference facility

 (e.g. fluctuations on process inventories, hidden inventories,
 losses, etc., for plutonium):

 DWK; GWK; IHCh (KfK)
 - (4) Simulation activities:EKS, IDT (KfK); Mr. Canty (KFA); Los Alamos (assistance and advice)
 - (5) Evaluation methodology:
 EKS, IDT (KfK); Mr. Avenhaus (Hochschule der Bundeswehr München);
 Battelle/U.S.A. (C. Bennett); Los Alamos; JRC Ispra (CEC);
 IAEA (Hough); J. Jaech (EXXON)
 - (6) Verification strategies:
 EKS, IDT (KfK); Mr. Avenhaus (Hochschule der Bundeswehr München);
 IAEA (Lovett)
- 3.3.2 The workshop emphasized the need for realistic plant operation data and recommended a close collaboration with the facility designers and operators having experience.

It was also recommended that an active participation of the safeguards organisations IAEA and EURATOM be ensured particularly in the field of evaluation, verification, requirements and strategies. In particular, experienced inspectors should be involved in the workshop.

3.4 <u>Time Schedule</u> (starting point: January 1981)

The specifications for the different R+D activities will have to be completed within the next two months.

- The first model on process simulation should be completed approximately in about 6-8 months time.
- The first screening and evaluation of the different statistical methodologies would have to be completed within the next 2-3 months, before starting detailed actions on a number of specified methods.
- The second meeting of the workshop is expected to be held during the period of early May to middle of May 1981 before the ESARDA symposium in Karlsruhe.

It is expected that the first results of the R+D activities in this area will be presented in a series of papers at the IAEA Safeguards Symposium planned to be scheduled during the fall of 1982.

ANNEX I - Participants

Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Nuclear Research Center Karlsruhe December 8-12, 1980

Name	Affiliation	Plenary	Working Group l	Working Group 2	Working Group 3	Preparation of Minutes
F. Argentesi	JRC Ispra	х		X	X ·	
R. Avenhaus	Hochschule d. Bundeswehr München	Х		Х	X	
R. Beedgen	K£K/IDT	Х			Х	
W. Beyrich	K£K/EKS	Х			Х	
M. Canty	KFA/KuU	Х		х	Х	x
S. Flach	K£K/PWA		Х			
J.E. Foley	LASL	х		Х		
P. Groll	KfK/IHCh	Х	Х			
D. Gupta	K£K/EKS	х		x	Х	х
H. Haug	KfK/IHCh	Х		x		
H.J. Hein	GWK	х	Х	х	Х	
H. Hough	IAEA	Х			Х	x
L. Koch	TU	х	Х			
M. Küchle	K£K/INR	х	Х			
J. Lausch	GWK	х		Х	Х	
E. Leitner	DWK	Х		x	Х	
H.R. Mache	K£K/EKS	Х				
Mrs. E. Mainka	KfK/IRCh	Х	X			
H. Ottmar	K£K/IAK	х	Х	х		
Mrs. S. Schoof	KfK/IRCh	х			Х	
D. Sellinschegg	K£K/EKS	х			X	
J.P. Shipley	LASL	х		Х	Х	
G. Spannagel	K£K/IDT	Х		x	х	х
H. Stöber	K£K/PWA	Х				[
F. Voß	K£K/EKS	Х		X	Х	х

ANNEX II - Agenda

Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Nuclear Research Center Karlsruhe December 8-12, 1980

Date		Time	Subject Discussed
Dec.	8 .	14:00	<pre>Plenary session - Opening remarks - General presentation of the objectives of the workshop meeting</pre>
Dec. Dec.	8 9	15:00 - 17:00 9:00 - 12:00)) Working Group 1: Measurement Techniques
Dec. Dec.	9 10	14:00 - 17:00 9:00 - 12:00)) Working Group 2: Simulation
Dec. Dec.	10 11	14:00 - 17:00 9:00 - 12:00)) Working Group 3: Evaluation Methods
Dec.	11	14:00 - 17:00	 Plenary session Discussions on recommendations of the working groups Future course of work Concluding remarks
Dec.	12	9:00 - 12:00	Preparation of the minutes of the meeting (with limited attendance)

The main workshop meeting concluded on December 11 at 17:00.

- 34 ---

Discussion Paper on Required R+D Work Prepared for Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

D. Gupta

Nuclear Research Center Karlsruhe F.R. Germany

1. General

The purpose of the present meeting is expected to be twofold:

- To identify the area in which R+D work will be required to establish the capability and limitations of the near-real-time accountancy measure under operating conditions in connection with international safeguards for a large scale reference reprocessing facility
- To formulate R+D tasks (objectives, subject matters to be treated, results to strive for, distribution of activities for the identified tasks etc.).

2. General Boundary Conditions

2.1 Reprocessing Facility

The flow sheet and the layout of a 1000 tU/a reprocessing facility, developed by DWK/KfK, F.R. Germany, will form the basis for these investigations.

2.2 The near-real-time accountancy (n.r.t.) measure will be applied only to the process area of the reference facility for the plutonium flows and inventories.

2.3 Determination of Plutonium Amounts

The plutonium amounts in inventories in and in flows to and from the process area will be determined as far as practicable, on the basis of measurements carried out in connection with the operation of the facility.

- 2.3.1 As the first alternative, only the process tanks (and not process equipment or pipelines) will be assayed to establish the plutonium inventory in the process. Such inventories will be established <u>during</u> the operation of the process (physical inventory taking without plant shut down).
- 2.3.2 The plutonium inventories in process equipments and pipelines (which correspond to approximately 6-7 % of the total plutonium inventory in the process) will be considered to be approximately constant fluctuating within a given range (for example ± 5-10 %). In case plutonium amounts will be assayed in process equipments for operational reasons, consideration could be given to the use of such values as a possible alternative for providing credibility to the assumed values for plutonium content in the process equipment and pipelines for n.r.t. accountancy purposes. However, such values could be used only if they could be verified by the safeguards organizations and the use of such values would not cause a hindrance or a disadvantage to the facility operators.

2.4 Independent Verification

In general all the relevant steps required for the implementation of n.r.t. measures on a routine basis (for example measurement systems, calibration methods, statistical evaluation method for data for providing safeguards relevant statements etc.), should be verifiable by safeguards organizations with regard to the credibility.

3. Measurement Basis

In identifying the R+D areas for measurement systems for the n.r.t. accountancy measure, a number of points will be worth considering. Some of them are indicated below:

3.1 Number of Steps in a Measurement System

In most of the cases, the throughput and inventory amount of Pu will be assayed in tanks. Following steps may be required for a measurement system for determining the Pu amount in such a tank:

- Sample taking from a tank
- Sample transfer to the analytical measurement system
- Pretreatment of the sample
- Measurement of Pu concentration (along with the associated calibration procedure)
- Measurement of volume (associated with calibration procedure)
- Calculation of Pu-amount in the tank.

The less the number of intermediate steps, the less also is the:

- Number of the sources of error in the measurement
- The possibility of falsification
- Number of steps requiring verification by safeguards organizations.

3.2 The Measurement Time

The total measurement time including all the steps should be whenever possible, less than the residence time of a Pu-solution in a tank (e.g. in the range of 8-24 hours). In this manner the frequency of the n.r.t. accounting will be determined by the inherent process parameter involving the residence time of the Pu containing solution in a tank and not by the delays in the measurement system. Besides, the verification of the operators measurement data could be carried out in principle, during the time the solution is still in the tank.

3.3 Random and Systematic Errors

Preliminary investigations indicate that the MUF uncertainty (σ MUF) for n.r.t. accountancy is determined mainly by the systematic error component of the measurement uncertainty for Pu amounts in the <u>input accountability</u> <u>tank</u>. The contributions of the random error component of this measurement system as well as the expected systematic and random errors of all the other measurement systems (e.g. inventory, product, waste) to the σ MUF are rather insignificant. At present the systematic error component of around 1 % (1 σ value) at the input accountability tank, has been found mainly to arise from the volumetric and analytical measurement sources. A reduction in the systematic errors at these steps could be extremely useful.

3.4 Calibration Possibility

In principle, the possibility of calibration for all the measurement systems in a n.r.t. accountancy may have to be foreseen. However, the importance of calibration for the different systems may be completely different. for different throughput and inventory measurements. For example recalibration possibility may not be necessary for inventory (process tanks) measurements. A calibration system in case it becomes necessary, must be operable during the operation of the process area and easily verifiable by the safeguards organizations.

3.5 Data Generation and Verification

In establishing the different tasks for creating the measurement basis, main emphasis is to be placed on the possibility of the realization in a facility under routine operation. Besides, the data sets generated have to be available to the safeguards organizations in a verifiable form. A large part of the investigative R+D efforts will have to be directed to this area.

4. Simulation

4.1 <u>Objectives</u>: To simulate the relevant accountancy data after taking into consideration real operating conditions.

4.2 Subjects of Simulation

- 4.2.1 The behaviour of the inprocess inventory such as the range of fluctuation, uncertainty, minimum available inventories under routine operating conditions which may be verified through measurements, influence of the fluctuations of the unmeasured part of inventory on the measured part etc. Such simulations may have to be carried out under start-up, normal operating and abnormal conditions.
- 4.2.2 Simulation of different accountancy data for establishing uncertainty in MUF, trends etc. for different values of measurement uncertainties (both systematic and random), fluctuations in the inventories, absolute amounts of throughputs and inventories etc.; establishment of trends in the uncertainty of data base and different types of influences on the MUF values.

4.2.3 Simulation of different diversion strategies under the given set of process inventory and throughput conditions to establish capabilities and limitations of n.r.t. accountancy measure with regard to these diversions.

4.3 Sources of Information for Simulation

Sources of information on different topics with regard to simulation should be derived to the maximum possible extent from practical experience as well as realistic data from operating facilities.

4.4 Experimental Verification of Simulation Data

The simulated data will have to be validated under real operating conditions. Therefore, a part of the R+D activities may involve possibilities of verifying the simulated data.

5. Statistical Evaluation of the Data Generated

5.1 <u>Objective</u>: On the basis of the data set produced by n.r.t. accountancy measure, the safeguards organizations will have to prepare a statement with regard to a possible diversion. For achieving this objective, the first question to be answered is the form of the final statement, expressed in the context of the proposed goals of the safeguards organization. Since these goals are expressed on the basis of abrupt and protracted diversions, a final statement will have to address to both these possibilities. Therefore, such a statement should be made at least for the following types of diversion strategies.

5.1.1 Abrupt Diversion

A diversion may be considered to be abrupt if an amount M be diverted within a period of 1-2 weeks (or less). This diversion has to be detected within 1-3 weeks after the total amount M has been diverted.

5.1.2 Protracted Diversion

A diversion may be considered to be protracted if this diversion is carried out over a period greater than 2 weeks during a whole calendar year. This diversion has to be detected within 1-3 weeks after the total amount M has been diverted but in any case before the end of the calendar year.

5.2 Evaluation Formalism

The evaluation formalism has to be designed in such a manner as to enable the safeguards organizations to initiate activities in a timely manner or to clarify anomalies which may be caused by different types of process fluctuations and losses or by different types of diversion strategies.

5.3 Probabilities of False Alarms and Non-Detection (α, β)

A number of alternatives may be available for the choice of α and β . They could be for example:

- To establish different α values for different diversion strategies
- To establish the same α value for all the diversion strategies.

The capabilities and limitations of n.r.t. accountancy could be analyzed on the basis of a parametric study for different α β alternatives for the given reference facility.

The possibility of using estimates for determining trends could also be investigated.

5.4 Short Detection Time

A detection time can be considered to be short if a detection can be made within 1-3 weeks after a given amount has been diverted in an abrupt fashion. Short detection time should not be treated <u>separately</u> but together with the diversion strategy to be considered. Therefore, the question in connection with the short detection time would be:

If a given amount M is diverted during 1-2 weeks, what would be the minimum probability of detection with which such a diversion could be expected to be detected for a given false alarm within 1-3 weeks after the diversion of the complete amount of M has taken place. - How could the probability of detection be improved?

5.5 Protracted Diversion

The capability of near-real-time accountancy with regard to protracted diversion could be considered for two boundary cases.

- Case 1

The operator plans to divert a small but the same amount continuously during a given period of time (for example one year). What would be the minimum amount which could be detected as diverted for a given values for α and β ?

How could this amount be reduced?

- Case 2

The plant operator plans to divert a given amount M over a longer period of time than two weeks and distributes this amount randomly over a calendar year. What would be the minimum probability of detecting the amount M within 10-30 days after the diversion of the total amount M has been completed for a given α .

How could the probability of detection be improved?

5.6 Establishing the Areas for Future R+D Tasks

After an agreement is obtained on the types of diversion strategies and principals of methodology to be utilized, it is desirable to have agreement on the specific methods which should be utilized for evaluating the data base obtained by n.r.t. accountancy measures.

ANNEX IIIb

Introduction to the Workshop Meeting on Near-Real-Time Accountancy in Large Reprocessing Facilities

Nuclear Research Center Karlsruhe December 8-12, 1980

D. Gupta Nuclear Research Center Karlsruhe F.R. Germany

PURPOSE OF THE MEETING

- IDENTIFY AND SPECIFY R+D ACTIVITIES WITH EXPERIMENTAL VALIDATION
- TO ESTABLISH
- CAPABILITIES AND LIMITATIONS OF NRT-ACCOUNTANCY MEASURE
 UNDER OPERATING CONDITIONS IN A LARGE-SCALE REPROCESSING
 FACILITY

NEAR-REAL-TIME ACCOUNTANCY

--- 44 ---

- MEASURES TO PROVIDE A VERIFIABLE DATA BASE TO SAFEGUARDS ORGANISATIONS FOR ENSURING CONTINUITY OF KNOWLEDGE ON FLOW AND INVENTORY OF PU IN A LARGE-SCALE REPROCESSING FACILITY
- WITH A VIEW TO
- ENABLE THESE ORGANISATIONS TO MAKE SAFEGUARDS-RELEVANT STATEMENTS ON POSSIBLE <u>DIVERSION STRATEGIES</u> ON THE BASIS OF THIS DATA BASE

BOUNDARY CONDITIONS

- 1000 T U/YR, REFERENCE FACILITY
- MAXIMUM POSSIBLE USE OF MEASUREMENTS MADE IN ANY CASE FOR
 PLANT OPERATION
- PU-INVENTORIES MAINLY IN PROCESS TANKS
- ALL STEPS LEADING TO DATA BASE MUST BE VERIFIABLE BY SAFEGUARDS ORGANISATIONS
- MINIMUM POSSIBLE HINDRANCE TO NORMAL PLANT OPERATION

DIVERSION STRATEGIES AND DETECTION TIMES

– ABRUPT

.

- DIVERSION OF AN AMOUNT M OVER A

SHORT PERIOD: 1-2 WEEKS

- DETECTION: 1-3 WEEKS AFTER M DIVERTED

- PROTRACTED

- DIVERSION OF AN AMOUNT M OVER A

LONGER PERIOD: 2-52 WEEKS

IN A CALENDAR YEAR

- DETECTION: 1-3 WEEKS AFTER M DIVERTED IN ANY CASE BEFORE END OF A CALENDAR YEAR ANNEX IVa

Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Nuclear Research Center Karlsruhe December 8-12, 1980

M. Küchle Nuclear Research Center Karlsruhe F.R. Germany

1000 t/a Reprocessing Plant

Plutonium Inventory in Process Area

Area	St	orage Tanks	Process Equipment
	Number of	total Pu content (kg Pu)	Pu content (kg Pu)
head end	4	50	
1. cycle	8	157	9.6
2. cycle	6	45	7.9
3. cycle	6	177	6.1
Pu concentr.	2	137	2.5
total	[·] 26	566 kg Pu 95.6 %	26.1 kg Pu 4.4 %

1000 t/a Reprocessing Plant

Annual Plutonium Flow and

Estimated Cumulative Measurement Error (Systematic)

Object of measurement	Pu flow Measure		ment error (20)	
	kg Pu/a	7,	kg Pu/a	
input accountability tank	10000	1 (0.5)	100 (50)	
product output	10000	0.3	30	
centrifuge waste	20	50	10	
leached hulls	10	100	10	
HAW	40	50	20	
MAW, LAW	3	50	1.5	

$$\sqrt{\Sigma \Delta M_{i}^{2}} = 107 \text{ kg} (63)$$

if all waste streams <u>+</u> 100 % 114 kg (74)

period:	l day	l week	4 weeks	l year
2σ:	0.54 kg	2.7 kg	10.7 kg	107 kg

Systematic errors in inventory measurements

storage tanks 566 kg Pu process equipment 26 kg Pu

Assumed error (20)

storage tanks		process	equipment	combined error
~	kg Pu	~ ~ ~	kg Pu	kg Pu
2	11.3	10	2.6	11.6
2	11.3	30	7.8	13.7
2	11.3	50	13	17.2
3	17.0	50	13	21.4

.

ANNEX IVb

Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Nuclear Research Center Karlsruhe December 8-12, 1980

Recommendations of Working Group 1: Measurement Techniques

Participants: S. Flach (KfK/PWA); P. Groll (KfK/IHCh); H.J. Hein (GWK); L. Koch (TU); M. Küchle (KfK/INR); E. Mainka (KfK/IRCh); H. Ottmar (KfK/IAK)

1. Running Inventory

1.1 Pu-Inventory in Storage Tanks

A measurement with a systematic error of 2 $\sigma = \pm 2$ % of plutonium concentration and volume of solution in storage tanks is feasible.

Sampling of solution can be made within one hour provided that one sampling head has not to handle more than 6 samples. Thus, with level measurement being continuous the time of inventory is defined well enough. The time required for sample analysis is estimated to be one week or less.

The following diversion scenario was considered to be of primary concern: In view of the fact that the inspector can only see the level indicator but not the real level of the solution falsification of instrument reading and clandestine removal of solution is possible. Some R+D work on verification possibilities of the level in process tanks would be required. As an alternative, an independent measurement of the total liquid flows in the aqueous stream and balance of liquid volumes is proposed for verification of the presence of solution. Neutron monitors or seals shall prevent extraction of plutonium via the organic phase.

This proposal, if implemented, would require design verification of the process area during the plant construction phase. For the highest concentration (50 g Pu/1) 200 l have to be diverted to get 10 kg of plutonium. An abrupt diversion of this volume can already be detected by monitoring a few liquid flows, part of which have free access.

For detection of protracted diversion about twice the number of tanks which are used for plutonium assay have to be monitored.

Alternatively it was proposed to perform the plutonium assay in storage tanks by continuous concentration measurements of the tank input flow which by integration and combination with the level measurement gives a continuous plutonium content reading. The verification problem is similar to the one discussed above, the concentration measurement can be verified by the inspector directly.

As R+D it is proposed to look into the diversion scenario and verification technique on the basis of detailed flow sheets.

A rapid though not very accurate verification of sample analysis is facilitated by the straight_foreward and transparent measurement technique in case γ -absorptiometry or RF-analysis are used. Alternatively ²⁴⁰Pu passive neutron counting by the inspector should be considered. Some R+D work in this area is still required.

1.2 Pu-Inventory in Process Equipment

A more careful inspection of WAK data revealed significant short term fluctuations of acid level in tanks (\simeq 50 %). Similarly significant fluctuations of Pu-inventory in process equipment has to be assumed. Fortunately for criticality reasons just these components that contain large amounts of plutonium are monitored by passive neutron counting. Calibration and verification impose serious problems. Operational experience with a plant running under normal conditions may be helpful. It has to be investigated whether the plutonium accumulation monitors can satisfy safeguards requirements.

Isotope correlation and a moving plutonium isotope composition discontinuity were not considered to be useful tools in the process area because of rework and complicated process streams.

2. Input Accountability

A significant improvement in input accountability tank assay beyond 2 $\sigma = \pm 1 \%$ does not seem feasible.

For verification an independent measurement of the total uranium content combined with a Pu/U-analysis of 1 % accuracy is proposed. Total uranium is determined from the fuel element fabricators data corrected for burn up, losses in leached hulls and losses in centrifuge waste. A 0.5 % accuracy seems obtainable.

An independent uranium assay can be obtained from y-absorptiometry.

Because of the difficulties encountered with the ²³⁸Pu- and ²⁴¹Pu-analysis it is recommended to make the plutonium accountancy via ²⁴⁰Pu instead of total Pu. Conversion to total Pu can finally be made using the input analysis. ²⁴⁰Pu has also the advantage that it is directly measured by passive neutron counting. It is an important R+D effort to look into this proposal in detail.

Abrupt diversion in the head end area could be detected by checking the isotope correlations of 134 Cs/137_{Cs} and 244 Cm/Pu as well as the Xe release. 134 Cs/137_{Cs} is by the burn up correlated with Pu/U which would be changed by removal of Pu. R+D work is needed to assess the accuracy and reliability of this method.

3. Waste Measurements

3.1 Solid Waste

Determination of residual fuel on leached hulls can be done in the following way: With the fuel element monitor neutron emission per gram of uranium is determined and by passive neutron counting at the leached hulls basket the neutron emission rate of the hulls is measured and converted into uranium. It is not clear, however, whether the Cm to Pu ratio of the fuel is the same as on the hulls. To clarify this point is for the moment the most important R+D issue in the area of leached hull monitors. The same is true for the $144Ce to Pu ratio in case of passive <math display="inline">\gamma-\text{counting.}$

Alternatively active neutron interrogation has to be applied by which total fissile content is measured. Significant R+D work is needed to assess the accuracy of this method under field conditions.

Active neutron interrogation also has to be applied to plutonium assay of filters and centrifuge waste. This method being expensive and inaccurate it should be checked whether inductively coupled plasma spectroscopy could be used at least for calibration.

The verification of plutonium content in barrels to which the inspector has full access does not impose a principal problem but has serious practical difficulties.

3.2 Liquid Waste

From the liquid waste only HAW contains enough plutonium to be of relevance for the material balance. Assay should be done by sampling, analysis and volume measurement. Automated α-spectrometry is needed and is under development.

Alternative measurement techniques are inductively coupled plasma and laser technique which require R+D work.

Summary of necessary R+D effort

Working Group 1: Measurement Techniques

1. <u>Running Inventory</u>

1.1 Plutonium inventory in storage tanks

- Investigation of methods for verification of liquid levels in process tanks
- Study of the possibility of combining tank level measurements with continuous concentration measurements of tank input flow for assaying plutonium in storage tanks
- Investigation of possible diversion scenarios based on a detailed process flow sheet for establishing the verification techniques and alternative possibilities (e.g., independent measurement of the total liquid flows in the aqueous streams and balance of liquid volume)
- The use of transparent straight-forward measurement techniques for rough verification of sample analysis: e.g. gamma-absorptiometry, RF analysis, ²⁴⁰Pu neutron counting

1.2 Plutonium inventory in process equipment

- Study of the applicability of in-process plutonium accumulation monitors as an aid to estimate inventories in process equipment with particular attention to the verification possibilities

2. Input Accountability

- Investigation of alternative level indication techniques for high accuracy in input accountability tank (e.g. "Ruska" method)
- Analysis of the sources of <u>systematic error components</u> in the measurement systems used for the input accountability sample assay, with a view to reduce them or to convert them into random error components (e.g. through calibration)
- Study of the feasibility of accounting for ²⁴⁰Pu rather than total plutonium in the process area
- Assessment of accuracy and reliability of isotopic correlations as a means of verifying operator input accountability data (e.g. checking the isotopic correlations of 134 Cs/137 Cs, 244 Cm/Pu as well as the Xe release)

3. Measurements for Solid and Liquid Wastes

- Clarification of the problem of differing Cm to Pu ratio between spent fuel and leached hulls, also for the ratio 144 Ce to Pu in case of passive gamma counting
- Investigation of active neutron interrogation methods for solid wastes
- Investigation of laser and inductively coupled plasma techniques for assaying plutonium in filters, centrifuge waste and liquid HAW
- Further development of the automated alpha-spectrometry for plutonium assay in liquid waste
- Development of different methods for assaying plutonium content in barrels

ANNEX V

Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Nuclear Research Center Karlsruhe December 8-12, 1980

Recommendations of Working Group 2: Simulation

Participants: F. Argentesi (JRC Ispra); R. Awenhaus (Hochschule der Bundeswehr München); M. Canty (KFA); J.E. Foley (LASL); D. Gupta (KfK/EKS); H.J. Hein (GWK); J. Lausch (GWK); E. Leitner (DWK); H. Ottmar (KfK/IAK); J.P. Shipley (LASL); G. Spannagel (KfK/IDT); F. Voß (KfK/EKS)

PART A - MODEL CONSTRUCTION

- I. Process Model
 - A. Information
 - 1. flowsheet values
 - 2. variations
 - 3. operating procedures
 - B. Model Equations
 - 1. tanks
 - 2. other vessels
 - 3. diversion (step III.D)
 - 4. fit the pieces together
 - C. Translate to Computer Program
 - D. Simulate
 - E. Display Results
 - 1. nominal behavior
 - 2. extremes

II. Accounting System Model

- A. Information
 - 1. measurement technology
 - 2. measurement capability
 - a. statistics
 - b. procedures
- B. Model Equations
 - error models
 - 2. materials balance constraints
 - 3. falsification (step III.F)

C. Translate to Computer Program

D. Simulate

- E. Display Results
 - 1. I, T, MUF
 - 2. measurement error variances for the composite I, T, and MUF

III. Diversion Model

- A. Detect Diversion Strategies
 - 1. location
 - 2. time evolution
 - a. abrupt
 - b. protracted
- B. Choose Diversion Amount
- C. Translate to Computer Program
- D. Insert into process model (step I.B.3)
- E. Modify Materials Accounting Data
 - 1. unfalsified (for operator)
 - 2. falsified (reported to inspector)
- F. Insert into Accounting System Model (step II.B.3)

IV. Verification Model

A. Determine Possible Verification Strategies

B. Examine the Feasibility of Verification Activities

C. Quantify the Capability of Individual Verification Activities

.

D. Model Equations

1. verification errors

2. relation to accounting system model

E. Translate to Computer Program

F. Simulate

G. Display Results

- I. Process Model
 - A. Information Sources
 - 1. process operating histories
 - 2. operator experience
 - 3. new experiments
 - B. Areas of Particular Concern
 - 1. unmeasured inventories
 - 2. upset conditions

II. Accounting System Model

- A. Information Sources
 - 1. instrument designers
 - 2. operator and inspector experience
 - 3. new experiments
- B. Areas of Particular Concern
 - 1. measurement control program
 - 2. satisfaction of measurement conditions

III. Diversion Model

- A. Information Sources
 - 1. past experience
 - 2. operators and inspectors
 - 3. new experiments
- B. Areas of Particular Concern
 - 1. feasibility
 - 2. are worst cases included?

- IV. Verification Model
 - A. Information Sources
 - 1. past and present procedures
 - 2. inspectors
 - 3. new experiments
 - B. Areas of Particular Concern
 - 1. feasibility
 - 2. sensitivity to changes in diversion strategy
 - 3. relation to process operating conditions

Summary of necessary R+D effort Working Group 2: Simulation

- Collection and, if necessary, generation of reliable data base corresponding to actual plant operating conditions and measurement capabilities
- 2. Modeling and Validation
 - a. process
 - b. accounting system
 - c. diversion
 - d. verification
- 3. Effects and Treatment of Unmeasured Inventories
- 4. Individual Verification Techniques
- 5. Overall Verification Strategies (related to evaluation methods)

ANNEX VI

.

Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Nuclear Research Center Karlsruhe December 8-12, 1980

Recommendations of Working Group 3: Evaluation Methodology

- Participants: F. Argentesi (JRC Ispra); R. Avenhaus (Hochschule der Bundeswehr München); R. Beedgen (KfK/IDT); W. Beyrich (KfK/EKS); M. Canty (KFA); D. Gupta (KfK/EKS); H.J. Hein (GWK); H. Hough (IAEA); J. Lausch (GWK); E. Leitner (DWK); S. Schoof (KfK/IRCh); D. Sellinschegg (KfK/EKS); J.P. Shipley (LASL); G. Spannagel (KfK/IDT); F. Voß (KfK/EKS)
- We consider that the safeguards approach for detecting protracted diversion
 (i.e. for conventional material accountancy) is established. We will investigate the augmenting of these techniques with methods for detecting abrupt diversion as the other extreme and diversion strategies between those extremes.
- 2. We will fix values for α , and the reference time interval (one year) and investigate the behavior of β as a function of loss amount and detection time.
- 3. We will establish the frequency of drawing materials balances (without shutdown of the facility) to be of the order of the conversion time (less than one month). For abrupt diversion this means that the detection time is essentially the time between materials balances.
- 4. When the testing procedure produces an alarm an investigation at different levels should be initiated as opposed to shutting down the plant. These levels would involve examination of
 - mistakes in the data
 - bias in the measurement system
 - location of unmeasured losses

- location of hidden inventory
- further actions to establish whether there is something missing (e.g. clean-out).
- 5. We recommend investigation of a few evaluation methods deemed to be appropriate by this group including sequential testing procedures. We will consider procedures that make use of data from previous time periods under the constraint that the decisions for the previous calendar years will not be revised.
- As an objective criterion for the optimization of sequential test procedures we take the expected detection time for protracted diversion extending over 12 months or less.
- 7. We recommend starting the analysis of sequential test procedures by using the CUMUF-statistics.
- 8. We will consider the use of specialized estimators in order to investigate the loss pattern and a minimum variance unbiased estimate of the loss amount within the process operation constraints.
- 9. In developing the different test and estimation procedures, it is essential that realistic data should be used. The following types of data may influence the effectiveness of different test and estimation procedures:
 - measurement errors
 - fluctuation of process inventories
 - hidden inventories
 - unmeasured losses.

Appropriate R+D efforts may be necessary to obtain such data.

- 10. The resulting procedures should be documented and demonstrated so that inspector personnel will understand how to use these procedures as well as which data will be required and how to acquire them.
- 11. The question of verification by international organisations has not been covered at present. This point has to be taken up at an appropriate time.

Summary of necessary R+D effort Working Group 3: Evaluation Methodology

The main tasks of the working group are:

- 1. Complete definition of the objectives and boundary conditions as first outlined in the recommendations
- Analysis of sequential test and estimation procedures by using CUMUF
 statistics and a limited number of other evaluation methods and specialized estimators
- 3. Definition of the input data elements required to implement the procedures and acquisition of realistic data sets
- 4. Test the procedures
- 5. Determine the effectiveness of the procedures when the data are influenced by measurement errors, fluctuation of process inventories, hidden inventories and unmeasured losses
- 6. Document the most effective procedures that can be recommended for use by inspectors
- 7. Define the verification methodology based on the results obtained from the other working groups
,

Annex	I		List of Participants	70
Annex	II	-	Agendia	71
Annex	III	-	List of Papers Presented for Discussion	72
Annex	IV	-	Characteristic Data of the Reference Reprocessing	
			Facility RWA-1000	73
Annex	V	-	Working Papers of the Subgroups	79
Annex	Va	-	Loss Models	80
Annex	Vb		Data Base-Variables and Parameters	83
Annex	Vc	-	Statistical Test Procedures	85
Annex	Vd		Process Simulation for a 1000 t Reference	
			Reprocessing Facility	88
Annex	Ve		Possible Contributions of the Measurement	
			Simulation Model Activities to the Planned	
		÷	NRTA Overview Report	89
Annex	Vf	120	Incorporation of Solvent Extraction into	
			Simulation Studies	91

.

MINUTES

of the 2nd Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Nuclear Research Center Karlsruhe February 24-26, 1982

- 1. D. Gupta welcomed the participants (a list of participants is given in Annex I).
- The provisional agenda was amended by including additional presentations (Annex II).
- 3. D. Gupta outlined the objectives of the meeting: a) to establish the state-of-the-art of Near-Real-Time Accountancy for application in reprocessing plants, b) to discuss how to handle the results of the workshop for presentation at the IAEA Symposium, November 1982, Vienna. J. Lovett pointed out that he could not make any commitments in respect of b). He invited proposals until beginning of May 1982.
- 4. Results of studies in three main areas (evaluation, simulation, measurements) were presented and discussed. Most of the presentations were based on working papers distributed during the meeting (a list of these papers is compiled in Annex III).
 - 4.1 Evaluation

This topic began with a presentation (C. Bennett) of the general framework in which evaluation studies should be carried out and in which different methods should be compared, emphasizing the importance of clearly defining the problems under investigation.

A number of statistical test procedures were described and discussed in respect of their underlying assumptions and boundary conditions, i.e. the problems for which they are principally appropriate (J.L. Jaech, D. Sellinschegg, J.T. Markin, R. Avenhaus, N. Nishimura). Several speakers also presented numerical results of applying these tests to specific examples and in one case to real data in order to demonstrate the performance of the procedures. It turned out that these results were not immediately comparable and it was recognized that there is a need to compare the sensitivity of different test procedures on a common basis.

4.2 Simulation

The status of the simulation activities of KfK/KFA was presented and compared to the program outlined during the first workshop. For completeness (and in addition to the distributed working paper) some of the basic features of the reference plant under study are summarized in Annex IV. J. Lovett presented a simple model for estimating the Pu inventory in extractors.

H. Nishimura gave a survey of the simulation activities in Japan.

4.3 Measurements

E. Mainka compared different measurement techniques for input solution, Pu product solution and waste streams in respect of accuracy, effort, costs and verifiability. Several participants emphasized the importance of sampling errors which in operating facilities often dominate the pure measurement errors in a laboratory.

H. Würz presented neutron measurement techniques developed or under development at KfK for application at extractors in a reprocessing plant, These instruments are designed to serve operating purposes and not to quantitatively measure the Pu inventory of extractors. Under special circumstances these methods might be modified to use them for quantitative estimates.

G. Hough confirmed that in the past operators in the US were able to estimate the Pu hold-up in columns using empirical correlations and selected process information.

5. Conclusions

Three subgroup papers related to evaluation procedures (Annex Va-Vc) were drafted. The discussion in the group led to several clarifications and corrections which, however, have not been incorporated in the annexed drafts. They are giving a framework and ranges of parameters for comparing different test procedures on a common basis. This comparison would be carried out under relatively simple assumptions about the process, since a) data from the more detailed process models are not yet available and b) some of the tests, in their present form, are not suited to evaluate the output of such models. The participants concerned will contact each other to decide how to proceed further.

Three other subgroup papers (Annex Vd-Vf) deal with simulation studies. They either list areas where contributions to a possible overview report could be made or indicate where work in the near future should be carried out. It was pointed out that a clear distinction should be made betweem column models used in simulation studies for evaluating different statistical test procedures and simplified column models for estimating the Pu inventories in these columns. It was also recommended that the actual content of contributions from this area to an overview report can be established at a later date.

Annex I

2nd International Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Karlsruhe, February 24-26, 1982

List of Participants

••

Name	Affiliation	Name	Affiliation
R. Avenhaus	HSBwM	H. Ottmar	K£K/IAK
C.A. Bennett	Battelle-HARC	D. Sellinschegg	KfK/EKS
H. Borrmann	KfK/PWA-PL	G. Spannagel	KfK/IDT
M. Canty	KFA/KUU	D. Stünkel	KfK/PKÜ-PL
E. Gabowitsch	KfK/IDT	F. Voss	K£K/EKS
D. Gupta	K£K/EKS	H. Würz	KfK/INR
H.J. Hein	WAK		
G. Hough	IAEA		
J.L. Jaech	Exxon Nuclear		
S. Kaiser	CEC Luxembourg		
U. Knapp	K£K/PKÜ-PL		
J. Lovett	IAEA		
R. Mache	K£K/PKÜ-PL		
Mrs. E. Mainka	KfK/IRCh		
J.T. Markin	LANL		
G. Nägele	K£K/EKS		
H. Nishimura	JAERI		

Annex II

2nd International Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Karlsruhe, February 24-26, 1982

Agenda

February 24	Speaker	References
1. Opening Remarks	D. Gupta	
2. Presentation and Discussion of	C.A. Bennett	1
Results of Evaluation Methodology	J.L. Jaech	2,3
	D. Sellinschegg	4
	J.T. Markin	5,6
	R. Avenhaus	
	H. Nishimura	7,8

3. Subgroup Sessions

February 25

F. Voss	9
M. Canty	9
D. Spannagel	. 9
J. Lovett	10
	F. Voss M. Canty D. Spannagel J. Lovett

- H. Nishimura 11
- G. Nägele 12

5. Subgroup Sessions

February 26

- 6. Presentation and Discussion of ResultsE. Mainka13on Measurement Techniques for NRTAH. Würz
- Presentation and Discussion of the Working Papers of the Subgroups

Annex III

2nd International Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Karlsruhe, February 24-26, 1982

References (Working papers)

- 1. C.A. Bennett, "Parametric Description of Loss Patterns"
- 2. J.L. Jaech, "Determination of the Detection Probability as a Function of Various Loss Strategies: Part 1, Design of Study"
- 3. J.L. Jaech, A. Kraft, "Determination of the Detection Probability as a Function of Various Loss Strategies: Part 2, Study Results"
- 4. D. Sellinschegg, U. Bicking, "The Performance of the Sequential CUR Test with Power One"
- 5. D. Sellinschegg, U. Bicking, "A Statistic Sensitive to Changes in the Loss Scenario"
- 6. J.T. Markin, "Timely Detection of Material Loss"
- 7. R. Beedgen, "Statistical Results in Multiple Materials Balance Models"
- 8. K. Ikawa et al., "An Integrated Near-Real-Time Materials Accountancy Safeguards System"
- 9. H. Nishimura, "On Evaluation Methodology of Near-Real-Time Materials Accountancy"
- 10. M. Canty, G. Spannagel, F. Voß, "Results on Inventory Simulation Activities"
- 11. J.E. Lovett, "On the Estimation of Solvent Extraction Plutonium Inventories in Near-Real-Time Materials Accountancy"
- 12. H. Nishimura, "On Flow and Inventory Simulation Activities for Evaluation of Near-Real-Time Materials Accountancy"
- 13. G. Nägele, "Activities Regarding a Simulation Model for a NRTA Measurement System of a Large Reference Reprocessing Plant"
- 14. E. Mainka, "Evaluation of Destructive Measuring Techniques Eligible for Plutonium Assay in a Large Reprocessing Plant"

Annex IV

Characteristic Data of the Reference Reprocessing Facility RWA-1000

(presented by F. Voss)

.

SIMULTANEOUS MEASUREMENTS AT TIME OF PIT

	NUMBER	EMPTY		VOLUME	SAMPLE
ACC.TANK	2	2			-
ADJ.TANK	4	2			-
HAF-TANK	2	-/1		(1)	-
1BXP-TANK	1	•rgi		1	1
1RP-A-TANK	1	_		1	1
2AF-TANK	5	1/2		1	_
2RP-A-TANK	1	-		1	1
3AF-TANK	3	-/1		1	_
3RP-A-TANK	1			1	1
3RP-TANK	2	-	,	-	-
4AF-TANK	1	_		1	(1)
4AC-TANK	2	1		1	1
	25	6/8		8(9)	5(6)

N.,

FREQUENCY OF INPUTS / OUTPUTS

	TIME BETWEEN TRANSFERS	FREQUENCY
ACCOUNTABILITY TANK	24 H	1.0 / D
4AC-TANK (PU-PROD.)	61 H	0.4 / D
HAW-TANK	6.3 H	3.8 / D
1BXU-TANK	1.3 H	19.0 / D
2AW-TANK	9.0 H	2.7 / D
3AW-TANK	19.3 H	1.2 / D
		28.0 / D

PU-INVENTORY IN TANKS

(SIDESTREAMS)

		NUMBER	MAX.INV,/TANK
1. CYCLE	HAW-TANK	2	40 G
teng Hout titler Allie Lynn gyg, Kall Long,	1BXU-TANK	2	4 G
	1BKW-TANK	1	2.5 G
	BKW-TANK	1	44 G
2. CYCLE	2AW-TANK	2	40 G
with Alba Fills with hing play kind inne	2BW-TANK	2	15 G
	2BKW-TANK	1	6 G
3. CYCLE	3AW-TANK	2	25 G
want tille finn film dans here bill den	3BW-TANK	2	15 G
	2BKW-TANK	1	12 G

16

PU-INVENTORY IN TANKS

(MAIN PROCESS STREAM)

		NUMBER	MAX, INV, /TANK
1. CYCLE	ACC.TANK	2	50 KG
and and and man and and and and	ADJ.TANK	4	50 KG
	HAF-TANK	2	50 KG
	1BXP-TANK	1	10 KG
	1RP-A-TANK	1	3.4 KG
2. CYCLE	2AF-TANK	5	17 KG
	2RP-A-TANK	1	5.7 KG
3. CYCLE	3AF-TANK	3	57 KG
	3RP-A-TANK	1	5.0 KG
	3RP-TANK	2	100 KG
PU-CONCENTRATION	4AF-TANK	1	25 KG
	4AC-TANK	2	125 KG

.

,

,

REFERENCE REPROCESSING FACILITY RWA-1000

THROUGHPUT:

1000 T_{HM}/A ≜ 5 T_{HM}/D

10 T_{PU}/A ≜ 50 KG_{PU}/D

3 DISSOLVER BATCHES/D

1 INPUT BATCH/D

1 OUTPUT BATCH/2.5 D

PU-INVENTORY:

TANKS

525 KG

(FEED ADJ.TANK - PROD.ACC.TANK)

PROCESS

EQUIPMENT	30.2	KG	TOTAL	
	11.6	KG	PULSE	COLUMNS

Annex V

2nd International Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities

Karlsruhe, February 24-26, 1982

Working Papers of the Subgroups

C.A. Bennett R. Avenhaus

<u>Annex Va</u>

1. LOSS MODELS

1.1 Models which describe the assumptions concerning the data to be evaluated are necessary to provide:

1) A framework within which to determine tests that are in some sense "best" or robust".

2) A framework for comparative studies based on simulation procedures.

1.2 The evaluation will consider a series of determinations of the inventories and transfers corresponding to a single MBA. Inventories will be taken at times t_i starting at some initial time t_o and continuing indefinitely. We may wish to consider:

- a) All the data
- b) All the data subsequent to some initial time τ_{o}
- c) All the data obtained during some fixed period T.

1.3 The data will be characterized by

- 1) Assumed true values μ_i of the inventories at time t_i . In simulation studies these are derived from the process simulation.
- Assumed errors of determination of net transfers and inventory. These are propagated from an assumed set of measurement errors and accounting procedures.
- 3) Assumed losses L_i in the periods $\Delta t = t_i t_{i-1}$ between inventories. These may be present in known or unmodeled process errors, long term or unmodeled measurement biases, or deliberate diversion.

1.4 In this exercise the primary concern is to estimate and test hypotheses concerning the L_i . The μ_i are modeled since (1) assumptions concerning these may effect the ability to estimate the L_i , and (2) in more general situations we may wish to estimate or test inventories as well as losses.

1.5 Models of the losses which will be considered in this exercise are the following:

1.5.1 We are concerned with the total loss M during some fixed time period T which may be, for example, a calendar year or a campaign. The individual losses $L_i = \gamma_i M$ within this period are of concern only as the patterns γ_i effect the ability to estimate and test M.

1.5.2 As in 1.5.1, excepted that we are concerned with the nature or timeliness of losses within the fixed period. This may involve:

1.5.2.1 Consideration of the time period of the initial loss

1.5.2.2 Consideration of the time at which the total amount M was available.

1.5.2.3 Consideration of the form of the diversion (e.g., abrupt vs. protracted).

1.5.3 We are concerned with the losses in some fixed period T from τ_1 to τ_2 following τ_0 . This may involve either a large or small fraction of the total time periods under consideration. Several subcases will be considered.

1.5.3.1 The existence of a base set of data known to be loss free. (Equivalently, can we assume $\mu_0 = 0$?)

1.5.3.2 Whether or not only $L_1 \ge 0$ are considered.

1.5.3.3 Whether or not there exists a time τ_3 by which detection of the losses between τ_1 and τ_2 should be achieved.

1.5.4 Within the model developed in 1.5.3, two types of assumptions with respect to the loss pattern during T are of interest. Both involve the expected value L and the variance $\sigma_{\rm L}^2$ of the losses.

1.5.4.1 (Diversion Model) . The pattern of losses such that $E(L_i)=L(\text{or }\Sigma L_i=M)$ is chosen to minimize the probability of detection for the particular test or tests being used. This differs from 1.5.1 because the location and length of the period T are not stipulated.

1.5.4.2 (Process Model) . The losses L_i are assumed to be characteristic of some process characteristics, such as a waste loss, process shift, or measurement bias. Typical models include a basic variability σ_L^2 and an assumed shift or drift in the expected value of L., (e.g. $E(L_i) = \mu_o$, $t_i^{<\tau}$. i $E(L_i) = \mu_o^{+\delta}$, $t_i^{>\tau_1}$).

<u>Annex Vb</u>

Report of Subgroup 2 on Data Evaluation Methods: Data Base-Variables and Parameters

> D. Sellinschegg J. Jaech

February 26, 1982

Two types of parameters are to be considered in the future definition of this study. The <u>error structure</u> and the <u>loss</u> <u>pattern</u> are specified.

Error structure

The notation is consistent with that given in the report "Determination of the Detection Probability as a Function of Various Loss Strategies: Part 1", J.L. Jaech, January, 1982. In addition, the systematic error will be broken down into two components, a long term and a short term systematic error variance as follows:

$$\sigma_{\delta}^2 = \sigma_{\omega}^2 + \sigma_{\theta}^2$$

where

 σ_{ω}^2 = L.T. S.E. variance σ_{θ}^2 = S.T. S.E. variance

The ranges of the error parameters to be investigated in this future work are as follows, with $\sigma_n = 1$ as the base value so that all parameter values are relative to σ_n .

```
\sigma_{\varepsilon}: 0.1 to 10

\sigma_{\delta}/\sigma_{\varepsilon}: 0 to 10

\sigma_{\omega} = \sigma_{\theta}

M: 0 to 24
```

The S.T.S.E. will be shifted every T time intervals. The range on T will be

T: 0.1n to n

Loss patterns

The losses will be presumed to include unmeasured inventories. In the simulation work, the losses can be characterized by either specifying expected losses or actual losses, but in the analytical studies, only actual losses can be input.

In the loss patterns, from time t_0 to t_a , the losses (exclusive of unmeasured inventories) will be zero for all intervals. In the next series of intervals, t_a to t_b , non-zero losses will occur according to patterns defined below. From t_b to t_n , losses will again be zero.

From t_a to t_b, the families of loss patterns will be as follows:

- Family 1: abrupt losses occurring in 1-4 intervals with varied spacings
- Family 2: Linear losses, increasing, decreasing, or uniform patterns: L_{i+1} = C_iL_i

(Note that if C_i is 1 for all i, the loss pattern is uniform. If $C_i > 1$ for all i, the losses are increasing linearly. If $C_i < 1$ for all i, they are decreasing linearly. If $C_i > 1$ for $i=1,2,\ldots,m$ and $C_i < 1$ for i>m, the loss increases initially and then decreases, etc.). J.T. Markin H. Nishimura

<u>Annex Vc</u>

Statistical Test Procedures

The problem of detecting material loss from a nuclear facility is considered in the context of statistical hypothesis testing theory. Under this approach the observed material accounting data are applied to test the hypothesis H_0 of no material loss against the alternative hypothesis H_1 of material loss. Such tests are of two types: The fixed length test in which a predetermined number N of balances are observed before deciding between H_0 and H_1 , and the sequential test in which the possibility of a decision is allowed after each balance is observed. In the case of the fixed length test, where only the probability of detection is of concern, it is known that the cumulative sum of material balances is the optimal test procedure. However, for sequential testing there is no corresponding result, so a number of test procedures are considered.

Sequential testing requires a statistic S(N) of the accounting data, such as the material balance or the cumulative sum of material balances, and upper and lower decision threshold TU(N) and TL(N), respectively. The testing procedure consists of accepting H_1 when $S(N) \ge TU(N)$, accepting H_0 when $S(N) \le TL(N)$, and continuing to test the data otherwise. Acceptance of H_0 implies that the test should be restarted by eliminating all previously acquired data and resetting the thresholds to their value at N = 1. In some cases the lower threshold $TL(N) = -\infty$ so that the test terminates only when H_1 is accepted.

The sequential tests to be considered share the following attributes: 1. Require inventory and transfer measurements and their covariance structure; 2. Assume the validity of the accounting data; 3. Test the hypothesis H_0 of no material loss against the alternative H_1 of material loss; 4. Allow the possibility of a decision about material loss in each balance period; 5. Employ a decision procedure in which a statistic of the material balance data is compared to a threshold to detect material loss. These tests are: 1. Cumulative sum of MUF residuals. This test uses a Kalman filter to estimate the material balance and compares the cumulative sum of the MUF residuals to a power one threshold to detect material loss. This test assumes a uniform loss model. The test statistic is Σ (MB₁-MB₁), where MB₁ is the observed material balance, \widehat{MB}_i in the Kalman filter estimate, and the decision threshold is

$$T(N) = \left[(N+M)(A^2 + Log(N/M+1)) \right]^{1/2}$$

where M and A control the false-alarm rate.

2. CUSUM. The cumulative sum of material balances $C(N) = \sum_{i=1}^{NB} MB_i$ is compared to the upper threshold $K\sigma_{C(N)}$ where K is chosen to bound the falsealarm rate. When $C(N) \ge K\sigma_{C(N)}$ a material loss is indicated. This procudure makes no assumption about the loss scenarios.

N

3. Page's Test. The statistic for this test is S(N) = C(N) - Min C(i), $1 \le i \le N$ where $C(i) = \sum_{j=1}^{\infty} (MB_i - \delta)$, and the upper threshold is h>0. The parameters j=1 δ and h are chosen to bound the false-alarm rate. Material loss is indicated when $S(N) \ge h$. In effect S(N) determines the largest CUSUM over all prior observations.

4. Modified Page's Test. This test uses the Page's statistic S(N) in conjunction with the power one threshold $T(N) = \left[N(A^2 + Log(N))\right]^{1/2}$ where the parameter A controls the false-alarm rate. A material loss is detected when $S(N) \ge T(N)$. For this test the lower threshold is 0 and when S(N) = 0 the test is restarted by eliminating all previous data and resetting the thresholds to their value at time N = 1.

5. Sequential Probability Ratio Test. Under this test procedure the hypothesis H_1 assumes a uniform loss scenario. The test statistic is the cumulative sum of material balances $C(N) = \sum_{\Sigma}^{N} MB_i$. When $C(N) \ge \frac{N\mu_1}{2} + \frac{Log(A)}{\mu_1}$ the hypothesis i=1 H_1 is accepted; when $C(N) \le \frac{N\mu_1}{2} + \frac{Log(B)}{\mu_1}$ the hypothesis H_0 is accepted and the test is restarted; if neither threshold is crossed then an additional observation is taken. The parameters A and B control the error rates. 6. Material Balance Test. Each material balance MB_i is compared to the threshold $K\sigma_{MB_i}$, where K is chosen to bound the false-alarm rate. Material loss is indicated when $MB_i \ge K \sigma_{MB_i}$. This test is sensitive to an abrupt loss.

7. Material Balance and CUSUM Test. The material balance and CUSUM statistics are compared separately to decision threshold. A material loss is indicated when at least one statistic exceeds a threshold. Correlations between these statistics must be considered in setting the thresholds to bound the false-alarm rate.

— 86 —

Variations of this test are constructed by applying the CUSUM only at energy $N^{\mbox{th}}$ balance.

Uniform Diversion Test. This test employs a Kalman filter to estimate the material balance. Material loss is detected when the filter estimate MB_i exceeds $K\sigma_{MB}$. A uniform loss scenario is assumed.

Several of these tests assume a priori that the loss scenario has a specific form. When this assumption is valid these tests perform well in detecting material loss; however, when the actual loss scenario does not agree with the assumed scenario, test performance is degraded. Tests that do not specify a loss scenario should in general be more robust. Among the tests considered the cumulative sum of MUF residuals, uniform diversion test and sequential probability ratio test all assume a uniform loss model under the hypothesis H_1 .

For sequential tests the duration of testing may either be specified a priori or continued until termination occurr when the test statistic crosses a threshold. The Page's test, modified Page's test, and the sequential probability ratio test terminate only when a threshold is crossed. All other tests are terminated after a predetermined number of balances.

While each of the tests detects material loss by sensing an increase in the material balance, the test statistics may express the increase in different forms. The material balance estimates the loss in each balance directly, the CUSUM estimates the cumulative sum of the losses, and the Kalman filter estimates the average loss per balance.

— 87 —

M. Canty

ŀ

<u>Annex Vd</u>

G. Spannagel

Process Simulation for a 1000 t Reference Reprocessing Facility

- Objectives
- Brief description of the process model
 - -- Flowsheet
 - -- Operation strategy
 - -- Model assumptions
 - -- Simulation
 - -- Data handling and transfer
- Selected simulation experiments
 - -- Normal operating conditions
 - -- Changes in burn-up
 - -- Unstable process behavior
 - -- Abnormal utilization of buffer capacity
 - -- Diversion strategies and hidden inventories

- Assessment of results

Annex Ve

Possible Contributions of the Measurement Simulation Model Activities to the Planned NRTA Overview Report

Participants of the working group:

Mr. Hein / WAK

Mr. Kaiser / EURATOM

Mr. Nägele / KfK (EKS)

Conclusions of the discussions:

- The hard core of the planned overview report must be a systematic investigation and comparison of the various NRTA data evaluation procedures proposed.
- Those principal investigations can and should be carried out using the simplified simulation models presently available.
- 3. The realistic and therefore much more complicated simulation programs are not suited nor thought to be used in doing these principal investigations. Their purpose is to demonstrate the robustness of the conclusions derived by these simple simulation models under more realistic assumptions for selected special cases.
- 4. A meaningful contribution of these simulation activities can be made only if these two models can be coupled together with the NRTA data evaluation program to a complete NRTA simulation model. An independent contribution of the measurement system simulation model or of the facility operation simulation model as well would make no sense.

- 89 ---

5. In coupling the currently developed complex simulation models together two problem areas arise. Firstly the facility simulation model has to be still completed, secondly there are some practical, not principal, problems to compute the complete covariance matrix needed as model information by the NRTA data evaluation program.

No problems arise in simulating the accountancy data series to be evaluated.

6. As the covariance matrix is nearly insensitive to fluctuations in the inventory and flow distributions, it is sufficient to calculate it only once and not for every simulation run. Therefore, it should be possible to overcome the practical problems within the time available if either

- the number of material balances is limited to about 50 or

- the covariance matrix is calculated under simplifying assumptions.

It should be left open at the moment which way has to be followed.

7. If the complex facility model cannot be completed within the time available, the simulation runs can also be made with the somewhat simpler model already integrated in the measurement system simulation program. J. Lovett G. Hough

<u>Annex Vf</u>

Incorporation of Solvent Extraction into Simulation Studies

C_f Variation Assumptions:

- mean value varies slowly over a range of ± 40-50 %
- individual batches vary randomly over a range of ± 10-20 %
- no operator intervention
- Case 1: Above variations appear in MUF as a result of assuming that solvent extraction inventory is constant.
- Case 2: Above variations are accounted for by using a simplified model for estimating solvent extraction inventories. Lacking any better model, assume $H = A \cdot C_f$, where A is a constant to \pm 5-10 % and C_f is measured.
- Case 3: As in case 2, but use $H = A \cdot C_f + B \cdot C_w$. This probably is not feasible in terms of Symposium presentations.

For all cases, compare detection sensitivity under a consistent set of values for other parameters, measurement variances, etc.

•

Evaluation of Destructive Measuring Techniques Eligible for Plutonium Assay in a Large Reprocessing Plant

E. Mainka

Kernforschungszentrum Karlsruhe Institut für Radiochemie

I. Introduction

It is the purpose of this paper to evaluate the available destructive methods of measurement which are eligible for plutonium assay in a large reprocessing plant. The system of evaluation relies mainly on the following criteria:

- a) accuracy of the method of measurement
- b) expenditure in terms of work; time required for an assay
- c) costs incurred; costs of equipment and analysis
- d) capability of verifying the method.

The data obtained in this way possibly furnish basic data for elaborating a control system for real-time balancing.

Before we discuss the individual measuring techniques, the boundary conditions of the plant, which are significant for the measurement, will be presented.

General Boundary Conditions for the Measurements

When striking the plutonium balance in a large reprocessing plant, one should rely as far as possible on the measurements performed in the framework of process control. All considerations start from a 1000 te/a plant whose flowsheet and layout have been described in detail in a KfK report / 1 /. The plutonium inventories for the individual process steps, which are evident from this block diagram, have been summarized in Table 1.

Table 1: Compilation of the Plutonium Inventory in the Individual Process Steps Process Area Plutonium Content Apparatus Inventory in the Respective inclusive of Pipelines Tanks kq Pu Head-end 190.0 -----1st cycle 33.5 6.5 2nd cycle 47.0 9.0 3rd cycle 7.0 93.0 Pu-concentrate -240.0 603.5 kg Pu 22.5 kg Pu 3.6% 96.38

With a daily throughput of 48 kg this gives a total stay time of 13 days.

As a result of the agreement saying that in the first line measurements also required in process control should be employed, the aim is pursued of measuring solely the plutonium contents contained in process tanks and not the contents or process apparatuses and process lines, respectively. The plutonium inventory in the latter two process units can be considered to be rather constant in case of trouble-free operation. Variations between + 5-10% are expected.

In Table 1 only the plutonium product streams have been taken into account because the waste streams occurring in the process are of minor importance for nuclear safeguards, above all if they include irrecoverable plutonium material. Deletion of the waste from the books makes it necessary to estimate the plutonium content. Using data gathered in practical application such an estimate has been made for a large plant |2|. The values have been compiled in Table 2 and are to serve as a tool in the discussions relating to the evaluation of measuring techniques in this area.

Table 2: Compilation of the Plutonium Flow in the Individual Process Streams of a 1000 te/a Plant

Process Stream	Plutonium Flow kg Pu/a	Material Description Approximate Plutonium Concentration and Activity
Input tank	∿10 , 000	1-2 mg/ml (α-Pu, γ-activity)
End product	∿10 , 000	~ 250 mg/ml (α-activity)
Centrifuge waste	∿ 20	\sim 50 µg/g (α , P, γ -activity)
Fuel clad wastes	∿ 10	\sim 1 µg/g (α , P, γ -activity)
High level waste (HLW)	∿ 4 0	\sim >1 $\mu g/g$ (α , P and $\gamma\text{-activity})$
Medium level and low level wastes (MLW and LLW)	3	\sim 1 µg/g (α , P and $\gamma\text{-activity})$

The Measurement System

When organizing a complete measurement system the following partial steps must be taken into account:

- Homogenisation of the solution

- measurement of the respective tank volume
- sampling from the tank
- sample transport to the analytical measurement system
- sample conditioning
- measurement of the plutonium concentration
- calculation of the plutonium inventory in this tank.

The discussion in this report relates to the determination of concentration. However, sample conditioning may be the decisive step for perfect measurement. This fact is taken into account here from the point of view of the time required for the assay.

II. Evaluation of Destructive Methods of Measurement

Preliminary remarks on the sample material:

- The plutonium concentration of the individual process steps undergoes variations by at least six orders of magnitude.
- The variations of radioactivity likewise cover several powers of ten.

This is the reason why the sample material in the plant can be broken down into four categories:

- a) the input solution
- b) the end product
- c) the liquid waste
- d) the solid waste.

In this report only the material categories a-c will be treated because the plutonium contents in solid waste samples are measured by non-destructive methods.

A. Analytical Methods for Input Analysis

Particular importance is attributed to the plutonium content in the dissolver and balancing tank, respectively, of a reprocessing plant. Here the first possibility is available of analyzing by destructive techniques the plutonium content in the material after burnup of the fuel elements in the reactor.

The Problem

With the light water reactors primarily operating today, burnups ~ 3 %, a uranium to plutonium ratio of about 100:1 must be expected at this point. For 1 g of fuel the $\beta-\gamma$ activity of such a material is 1 Ci of fission products after about two years of decay time. This means that the analyses cannot be performed until appropriate shielding measures have been taken. Moreover, plutonium must be assayed besides an approximately 100 times excess in uranium.

Under the prevailing conditions efforts are being taken to further improve the plutonium assay. An alternative considered consists in improving fission product balancing by increasing the accuracy of the uranium assay. Then, the plutonium concentration could possibly be determined from measurements of the U/Pu-ratio which can be done more conveniently.

Based on the criteria of evaluation indicated at the beginning, the following methods of measurement have been selected to fulfil this task. Methods of Input Analysis Plutonium concentration 1 - 2 g/l

Analytical Method	Variation Co- efficient of the Method (1σ)	Number of Measurements per day (8 hours)	Costs of Analysis per Sample (DM)	Capability of Verification
Isotope dilution analysis	∿1.28	4	1000	expensive, special lab required
X-ray fluorescence spectroscopy	< 2%	24	20	simple, with standard samples
Isotope correlation technique	∿18	1-16 1)	501000	expensive or simple ¹⁾
Gamma absorptiometry	0.2-0.38 ²⁾	∿16	50	simple, with calibration standards
REDOX titration	<0.5% ²⁾	4	250	expensive, special lab required
Emission spectroscopy	∿1୫	∿30	25	simple, with calibration standards
Alpha-spectros- copy	2-3%	2	100	expensive, special lab required
Laser RAMAN	5-20%	∿30	? ³⁾	? ²⁾

Depending on the element used for establishing the correlation.

²⁾Only uranium assay possible.

³⁾Data taken from the literature: proposed as an in-line method.

Analytical Methods for Plutonium End Product Assay

The plutonium containing end product in a reprocessing plant is obtained in most cases as an acid nitrate solution with a concentration of about 200 g/l (30). This is a "highly pure material" whose total trace content is specified to be <500 ppm. On account of the high plutonium content extremely stringent requirements for the accuracy of the assay have been made at this point. The following measuring techniques have been selected under this aspect. The high accuracy could be achieved by good and properly trained personnel.

Methods of End Product Assay Plutonium concentration 100 - 150 g/kg

Analytical Method	Variation Co- efficient of the Method (1g)	Number of Measurements per day (8 hours)	Costs of Analysis per Sample (DM)	Capability of Verification
REDOX titration	0.1-0.3%	4	250	expensive, special lab required
Coulometry	0.1-0.3%	4	250	expensive, special lab required
Gamma ab- sorptiometry	0.2-0.3%	16	50	simple, with calibration standards
X-ray fluorescence spectroscopy	∿18	24	50	simple, with calibration standards
Gravimetry	O.18	2	50	simple, although requiring special lab
Density/acid correlation	∿O . 5%	8	150	simple, at special lab

— 99 —
Analytical Methods for Plutonium Assay in Waste Solutions

In the waste from a reprocessing plant the plutonium content undergoes variations over a large range. Typical plutonium contents in the waste lie between 10^{-6} to 10^{-1} g/l |39|. On account of the low concentration also the requirements for accuracy are less stringent.

The following measuring techniques are applied in this case: Methods of Waste Analysis Plutonium contents vary in the µg-range

Analytical Method	Variation Co- efficient of the Method (1g)	Number of Measurements per day (8 hours)	Costs of Analysis per Sample (DM)	Capability of Verification
Spectral photo- metry < 1 mg/l	∿10%	4–16	50-250	simple, with calibration standard
Extraction followed by α -spectroscopy and counting, resp. < 10^{-6} mg/l	∿ 88	∿ 4	250	expensive, special lab required
Emission spectros- copy after plasma excitation < 0,1 mg/1	∿10%	∿16	50	simple, with calibration standard
X-ray fluorescence spectroscopy < 5 mg/l	∿10–20%	∿16	50	simple, with calibration standard
Correlation of specific α-activity and α-activity ratio (Bq/g Pu / <u>Pu 238</u> Pu-239+240	∿1–5%	∿ 4	250	expensive, special lab required
< 10 mg/1				

— 100 —

Summary

The majority of analytical techniques cited in this report have been tested and are measuring methods proven in practical application. Part of them are being used already now in process control.

Remark:

I wish to thank Mr. R. Berg (GWK) for valuable discussions.

Literatur

- 1. M. Kluth, H.O. Haug und H. Schmieder: KfK 3204 (1981)
- 2. M. Küchle (Privatmitteilung)
- 3. Webster et.al.: Anal. Chim. Acta 24 371 (1961)
- 4. Bokelund: ETR-217 (1967)
- 5. ASTM E 266
- 6. W. Beyrich und E. Drosselmeyer: KFK 1905/1 und 1905/2
- 7. W. Lycke, M. Gallet, F. Peetermans, R. Damen, E. Bouwmeester,P. De Bièvre und U. van Andenvove, CBNM/MS/76/79
- 8. Ertel: KfK 77, KfK 1121
- 9. v. Baeckmann: IAEA STI/Pub/337 (1972) 33
- 10. R. Kraemer und W. Beyrich ed.: KfK 1100 Chapt. 7
- 11. L. Koch, The Isotope Correlation Experiment, 2nd Annual Symp. on Safeguards and Nuclear Material Management, Edinburgh, Scotland, March 1980, p. 392
- 12. H.J. Arenz, G. Hough und L. Koch, Experience for Safeguards Gained in the Isotope Correlation Experiment, 2nd Annual Symp. on Safeguards and Nuclear Material Management, Edinburgh, Scotland, March 1980, p. 339
- 13. S.M. Schoof und W.L. Zijp, How good are Isotope Correlations Compared to Conventional Statistical Techniques, 2nd Annual Symp. on Safeguards and Nuclear Material Management, Edinburgh, Scotland, March 1980, p. 342
- 14. D.E. Christensen, R.A. Schneider und K.B. Stewart, Summary of Experience with the Use of Isotopic Correlation Safeguards Techniques, BNWL-SA-4274 (March 20, 1972) presented at the IAEA Working Group Meeting on the Use of Isotopic Composition Data in Safeguards, Vienna, Austria, April 10-14, 1972
- 15. French Contribution to the Research Coordination Meeting on Application of Isotopic Correlation Techniques to International Safeguards, IAEA, Vienna, 12-14 November 1979
- 16. C. Beets, Editor "Contributions to the Joint Safeguards Experiment Mol IV at the Eurochemic Reprocessing Plant Mol-Belgium", BLG. 486

- 17. H. Eberle, P. Matussek, I. Michel-Piper, H. Ottmar, Proceedings of the 2nd ESARDA Symp. on Safeguards and Nuclear Material Management, Edinburgh, Scotland, March 26-28, 1980, report ESARDA 11 (1980) edited by Joint Research Centre, Ispra, Italy
- 18. H. Eberle, P. Matussek, H. Ottmar, I. Michel-Piper, M.R. Iyer, P.P. Chakraborty, Nuclear Safeguards Technology, 1978, Vol. II, International Atomic Energy Agency, Vienna, 1979, 27-50
- 19. W.M.J. Veigele, Atomic Data Tables 5 (1973) 51-111
- 20. T.R. Canada, R.C. Bearse, J. Tape, Report, LA-6675-PR (1977) 22-24
- 21. H. Eberle, P. Matussek, I. Michel-Piper, H. Ottmar, Proceedings of the 3rd Annual Symp. on Safeguards and Nuclear Material Management, Karlsruhe, 6-8 Mai 1981
- 22. W. Davis und W. Gray: Talanta 11 1203 (1964)
- 23. A.R. Eberle, M.W. Lerner, C.G. Goldbeck und C.J. Rodden NBL 252 (1970)
- 24. Lindner und A.v. Baeckmann: KfK 701 (1967)
- 25. J. Slanina, F. Bakker, A.J. P. Groen und W.A. Lingerak: Z. Anal. Chem. 289 102 (1978)
- 26. W. Coerdt, S. Flach, W. König, E. Mainka, K. Matern, J. Neuber und H.G. Müller: EUR-6629 EN 89 (1979)
- 27. M. Edelson und V.A. Fassel: ESARDA 13 97 (1981)
- 28. R. Berg: EUR 6629 EN 119 (1979)
- 29. J.P. Hohimer: SAND 78-1522
- 30. R. Berg (Privatmitteilung)
- 31. Shults und Thomason: Anal. Chem. 35 921/ (1963)
- 32. G.U.C. Milner, G. Phillips und Judge: Talanta 15 1241 (1968)
- 33. G. Phillips, D. Crossby: Proceedings: Nuclear Safeguards Technology 1978, IAEA-SM-231/56
- 34. D.D. Jackson, R.M. Hollen, E.R. Roensch und J.E. Rein: Analytica Chem. Acta 117 205/215 (1980)
- 35. K.A. Swinburn und J.R. McGowan: BNFL-Rep. 205 (W)
- 36. J.R. McGowan, Johnson und K.A. Swinburn, IAEA STI/PUB 291 (192)3
- 37. L. Lindner und A.v.Baeckmann: KfK 701 (1967)
- 38. R. Berg: IAEA SM-231 (1979) 661/668
- 39. R. Berg: EUR 6629 EN 119/126 (1979)

.

KERNFORSCHUNGSZENTRUM KARLSRUHE

Institut für Datenverarbeitung in der Technik Entwicklungsabteilung Kernmaterialsicherung

Results on Inventory Simulation Activities

von

M.J. Canty (KFA), G. Spannagel, F. Voß

Kernforschungszentrum Karlsruhe GmbH, Karlsruhe

Introduction

At the workshop we had in December 1980 our opinion was confirmed that R+D work related to Near-Real-Time Accountancy should be started with simulated process data. With the assistance of Dr. Shipley from LANL we elaborated a basic working schedule during that workshop (see Minutes of the meeting/Annex V). Part A of that schedule refers to "model construction": Part B refers to "model validation". In these days we still have to finish Part A. Although the basic concept of the process model has been worked out, anomalous process behavior has not yet been covered. The results obtained in the main areas of interest ("Base Data for the Reference Facility", "Solvent Extractor Modelling", "Results of Simulation") will be described below.

1. Base Data for the Reference Facility

The basis for the simulation studies is a 1000 $t_{\rm HM}^{\prime}/a$ reference reprocessing facility*. The flow sheet of the main process stages containing the bulk of the Pu inventory has been presented at the first meeting of the work-shop together with nominal flow rates, Pu concentrations and Pu inventories.

1.1 Tanks

All tanks are assumed to be equipped with capabilities for volume measurement, homogenization and sample taking. Since all transfers are measured batchwise in tanks, no on-line instruments for flow and concentration measurements are required.

Preliminary values have been established for those parameters which determine the working cycle of tanks but which cannot be derived from the flow sheet. Starting from these values inventories and transfers have been calculated for given time intervals (assuming constant flow rates) in order to study the influence of varying liquid volumes on the sensitivity of NRTA while the total inventory remains constant.

1.2 Solvent Extractors

No measurements of pulse column inventories are foreseen in the present studies.

We have started to estimate the variability of (unmeasured) column inventories under normal operating conditions on the basis of simplified assumptions.

M. Kluth, H.O. Haug und H. Schmieder KfK-Report, KfK 3204, September 1981

2. Solvent Extractor Modelling

In order to develop a computer program to simulate the flow of Pu through the extraction and purification sections of a reprocessing facility, mathematical models for the various process components are needed. Since the associated algorithms are called a very large number of times in the course of a simulation run, the models must be kept as simple as possible, but at the same time reproduce in a reasonable way the behavior of the actual components. In this contribution we describe a model for pulsed columns similar to one developed at Los Alamos¹ and incorporated into the simulation program developed in the F.R. Germany for a looo t reference reprocessing facility.

The model is depicted in Fig. 1. The C_1 represent Pu concentrations (in g/l) and the F_1 flow rates (in 1/hr). V_1 and V_3 are disengagement volumes, V is the total volume of liquid in the mixing section and V_{12} is the volume of the liquid phase entering in streams F_1 and F_2 . For example, for A-type columns, F_1 , F_2 and F_3 would correspond respectively to feed, scrub and extractant flow rates, F_4 and F_5 to the product and waste streams, V_1 and V_3 to product and waste disengagement volumes and V_{12} to the volume of the aqueous phase in the extraction section. This model has been used to simulate the HA, HS, 1BX, 1BS, 2A, 2AS, 2B, 3A, 3AS and 3B columns of the reference flow sheet.

The main assumptions made in deriving the model equations are:

- (i) constant volumes $V_1^{}$, $V_3^{}$ and $V_3^{}$
- (ii) exponential Pu concentration profiles in the mixing section
- (iii) perfect mixing of streams 1 and 2
 - (iv) proportionality between the mass flow in streams 5 to that in streams 1 and 2.

'E.A. Hakkila et alii,

[&]quot;Coordinated Safeguards for Materials Management in a Fuel Reprocessing Plant", Los Alamos National Laboratory report LA-6881 (1977)



Fig. 1: Pulse Column Model

These assumptions lead to the following equations for the Pu concentrations in the two phases of the mixing section

$$c_{12}(y) = C_{12} e^{-\alpha y}$$

$$c_{3}(y) = C_{3} + (F_{1}C_{1} + F_{2}C_{2})(e^{-\alpha y} - e^{-\alpha L})/F_{3}$$
with $C_{12} = (C_{1}F_{1} + C_{2}F_{2})/(F_{1} + F_{2})$

and $a = -\ln (C_5/C_{12}) / L = -\ln k/L$

Where k is the constant of proportionality for assumption (iv) above.

These equations are integrated to determine the Pu hold-ups in the mixing section:

$$H_{12} = V_{12} C_{12} (k-1) / lnk$$

$$H_{3} = (V-V_{12}) C_{3}/(1-k) + C_{3}/lnk - C_{4}k/(1-k) - C_{4}/lnk$$

The hold-ups in the disengagement sections are simply

$$H_4 = V_1 C_4$$

 $H_5 = V_3 C_5 = V_3 C_{12} k$

and the total hold-up in the pulsed column at equilibrium is

$$H = H_{12} + H_3 + H_4 + H_5$$

We consider the flow and concentrations to be slowly varying functions of time t and use the notation $F(1) = F(t=t_1)$ etc,

$$H(1) = C_{12}(1) (a_1 + a_5) + C_3(1)a_2 + C_4(1) (a_4 - a_3)$$

where the a_i are in litres and are given by

$$a_{1} = V_{12} (k-1)/lnk$$

$$a_{2} = (V-V_{12}) (l+lnk-k) / (l-k)lnk$$

$$a_{3} = (V-V_{12}) (l+klnk-k) / (l-k)lnk$$

$$a_{4} = V_{1}$$

$$a_{5} = V_{3}k$$

The equation of continuity requires that

$$\frac{\Delta H}{\Delta t} = \frac{H(2) - H(1)}{\Delta t} = \langle C_1 F_1 \rangle + \langle C_2 F_2 \rangle + \langle C_3 F_3 \rangle - \langle C_4 F_4 \rangle - \langle C_5 F_5 \rangle$$
where $\langle CF \rangle = \int_{t_1}^{t_2} C(t)F(t)dt / \Delta t$

For small time increments this integral is approximately

$$\langle CF \rangle = \{C(1)F(1) + C(2)F(2)\} / 3 + \{C(1)F(2)+C(2)F(1)\} / 6$$

The calculation of column hold-up and output flows and concentrations for successive time increments in the process similation proceeds as follows:

All quantities at time t=t, are known -

$$V_{12}(1)$$
, H(1), $C_i(1)$, $F_i(1)$ i = 1...5

Input flows and concentrations at time t=t₂ are either taken from the output of preceding process units or are generated stochastically with a random walk procedure -

$$C_{i}(2)$$
, $F_{i}(2)$ $i = 1, 2, 3$

The phase volume V_{12} is also treated as a slowly varying stochastic variable -

$$v_{12}(2)$$

The output flows $F_4(2)$ and $F_5(2)$ at $t=t_2$ are then given by the volume conservation condition. For example

$$F_{4}(2) = F_{3}(1) + F_{3}(2) - F_{4}(1) + 2(V_{12}(2) - V_{12}(1)) / \Delta t$$

Finally, the output concentrations are given by

$$C_{5}(2) = k C_{12}(2)$$

$$C_{4}(2) = \left\{ (\langle C_{1}F_{1} \rangle + \langle C_{2}F_{2} \rangle) (1-k) + \langle C_{3}F_{3} \rangle - C_{4}(1)F_{4}(1)/3 - C_{4}(1)F_{4}(2)/6 - C_{12}(2)(a_{1}+a_{5})/\Delta t - C_{3}(2)a_{2}/t + H(1)/\Delta t \right\} \right|$$

$$\left\{ (a_{4}-a_{3})/\Delta t + F_{4}(2)/3 + F_{4}(1)/6 \right\}$$

In preparation for the next step,

$$C_{i}(1) := C_{i}(2)$$
, $F_{i}(1) := F_{i}(2)$, $H_{i}(1) := H_{i}(2)$
 $V_{12}(1) := V_{12}(2)$

and

3. Results of Simulation

Based on the general input data and on the model equations described above, our simulation model has been developed. With this simulation we assume that each cycle is surveilled by an operator and that the operators are controlled by a supervisor. Operators and supervisor essentially have to handle information supplied by the instruments in the central control room. In addition, now and then a note will be available on the positive result of an analysis carried out on the contents of a full tank. In a rather simplified manner the operators' duty consists in two major tasks:

(1) They try to keep to nominal values of all the process variables.(2) They handle transfers, for example between tanks.

As to (1), certain process fluctuations will occur because it usually will take some time until

- an operator will recognize a deviation from nominal behavior
- a counteraction has taken place
- the process starts to return to nominal behavior.

As to (2), the operators have to communicate before a transfer can be performed. For example, the contents of a tank can only be released if

- all valve settings have been checked
- the contents of the tank have been stirred for a time long enough to homogenize the inventory for sample taking
- the sample has been taken and analyzed, and the result of this analysis allows the respective transfer
- the volume of the tank filling is known.

Summing up it can be stated that simulation must cover the different response properties of the operators and that it has to perform all these tedious steps related, for example, to a transfer. Of course, in order to get the estimates of the Pu inventories in the tanks of the whole plant, simulation must follow the interactions of all changes in flow-rates and in concentrations.

For translation into the computer program we applied a special language called SIMULA. This language was developed around 1966 by the Norwegian Computing Center in Oslo. The experts told us that SIMULA offered some flexibility which might be necessary if, for process operation, we have to accomodate strategies different from those described above. We wish to point out again that, for the time being, no special process states have been considered; the results given below were obtained under steady-state conditions.

The main program runs on the central computer of KfK. Using some very special equipment of KfK/IDT it is possible to transfer selected simulation results to the IDT computer area, where further handling (for example graphical display or plotting) is possible.

To test our simulation model we have selected the 2nd Pu cycle of our reference facility; this proved to be a reasonable decision. However, this cycle is not adequate for presenting exciting results. The lst U cycle offers much more possibilities for a demonstration; but our current simulation covers only the 2nd Pu cycle.

Figs. 2 through 4 present results of simulation obtained for the Pu-inventories of the extractors 2A and 2B and of the 2B mixer-settler during a 30 hour interval. The inventory variation is mainly induced by the variation of the feed of the 2A extractor.



114 -



Fig. 3: Pu Inventory of the 2 B Extractor versus Time (h)

- 115 -



Fig. 4: Pu Inventory of the 2 B Mixer-Settler versus Time (h)

116 —

Activities Regarding a Simulation Model for the NRTA Measurement System of a Large Reference Reprocessing Plant

G. Nägele

Kernforschungszentrum Karlsruhe, EKS

Paper presented at the NRTA-Workshop in Karlsruhe, February 24-26, 1982

I. Objectives

1. The Task

Starting from a set of "true data" on the flow and inventory distributions in the reference reprocessing facility, it is the task of the simulation program under discussion to simulate the effect of the NRTA measurement system on these data as well as those parts of the accountancy system necessary to transform the measurement data into a time series of MUFvalues, which then will be an input for the sophisticated NRTA data evaluation procedures developed by D. Sellinschegg. Besides this simulated time series of MUF-values, the program has also to provide the information about the covariance structure of these MUF-data needed by the data evaluation procedures.

The "true" set of data will be generated by the simulation models for facility operation developed by Canty, Spannagel and Voss.

2. Intentions

Currently, the characteristics of NRTA Data Evaluation Procedures - i.e. false alarm probability, sensitivity and timeliness of detection - are investigated in Karlsruhe as a function of various diversion scenarios and facility parameters by Monte Carlo simulation runs using a rather simple simulation model. Before such procedures can become a routine safeguards measure, it will be necessary to demonstrate their good performance and the robustness of the derived conclusions under more realistic conditions. The implications of the simulation model are twofold: on the one side, for application of NRTA evaluation procedures a covariance structure of the data series to be analyzed has to be assumed in the procedure, on the other side the data series directly reflects the underlying covariance structure. Our intentions with the measurement simulation program under development are therefore:

- To find out how various features of the measurement system and error model affect the covariance structure of NRTA data series.
- (ii) Investigate the sensitivity of the evaluation procedures versus changes in the covariance structure.
- (iii) Test the robustness of the procedures in case of discrepancies between assumed and "real" covariance structure.

These intentions imply some constraints for the measurement simulation program to be developed: It should allow to model a NRTA measurement system for the reference reprocessing facility as realistic as possible especially allow for all such error features which might play a role in reality. On the other side, it should be so flexible to allow for design changes in the facility and the measurement system as well as the treatment of simplified models. Finally, it should be fast enough to allow Monte Carlo simulation runs.

II. The Basic Elements of the Model

Three basic elements are used in modeling the NRTA measurement system of the reference reprocessing plant:

- (i) the error model for an individual measurement,
- (ii) the measurement model which describes how the accountancy data are determined from measurement values,
- (iii) the basic structure for the measurement system and its operation.

1. The Error Model

The error model for an individual measurement is like usual assumed to be a linear function of mutually independent normally distributed error components (δ). In order to allow for correlations between different measurements with the same instrument or procedure, we distinguish systematic ($\boldsymbol{\delta}_{_{\mathbf{C}}}$) and random (δ_{Z}) components. Random errors are newly generated for every measurement, whereas the systematic contributions are newly generated only upon recalibration of the instrument and remain constant within one calibration period. The possibility of a non-zero expectation value for systematic contributions allows to introduce long-term systematic effects. Because we mainly deal with the measurement of variables of a broad range of values including zero, both systematic and random errors may have additive as well as multiplicative contributions as function of the true value (T) of the measured quantity. I.e. we use the general model:

$$X = T + (\delta_{SA} + \delta_{SM} \cdot T) + (\delta_{ZA} + T \cdot \delta_{ZM})$$

2. The Measurement Model

In general, the plutonium content (Y) of a batch of nuclear material cannot be directly measured but will be determined from a series of measured quantities $(X_i, i \in \{1, ..., n\})$ related to Y - e.g. Pu-concentration and volume. Because batch to batch correlations will be - in general - different for the various quantities to be measured, we have to take this into account and consider the following general model:

$$Y = F(X_1, \ldots, X_n).$$

I.e. Y is a function of the vector X formed by the complete set of quantities (more exactly types of quantities like Puconcentration, volume, etc.) measured for Pu-determination in the plant. In principle, F might be specific for each specific batch entering the material balance. We assume, however, that there will be only a small number of functions used in the NRTA measurement system and only the indication, which quantities are to be measured and which of this set of functions is to use for Pu-determination will be batch specific.

For the determination of the covariance structure, the linear error propagation model of Gauss is used:

$$\Delta Y = Y - F(T_1, \dots, T_n) \sim \sum_{i=1}^{n} \left(\frac{\partial F}{\partial X_i} \right) \cdot \left(\left(\delta_{SA} + \delta_{ZA} \right) + T_i \left(\delta_{SM} + \delta_{ZM} \right) \right) \\ \left(X_1, \dots, X_n \right) = \left(T_1, \dots, T_n \right)$$

3. Basic Structure of the Measurement System Model

Measurement errors are assumed to be characteristic for a specific set of measurement equipments and/or procedures. This implies the following structure of the measurement system: For each type of quantity, which will be measured at some place or time for NRTA purposes, a set of measurement instruments or procedures (at least one) must be defined. This is done by specifying the parameters in the general error model specifically for each individual "instrument". In addition, for every instrument it has to be specified at what points in time during a simulation run it will be "recalibrated", i.e. a new realization for the systematic error contributions will be generated. Currently only periodic recalibration strategies are foreseen.

This set of instruments is operating on the nuclear material distributed over and flowing through the facility's process area. In our model we assume that - at the time of measurement - nuclear material inventories as well as transfer flows are structured in "batches" defined by containments, e.g. buffer and process tanks. For accountancy purposes we distinguish between inventory and transfer batches. Output transfer batches are identified by a negative sign of the material amount. For clarity and to simplify a possible later introduction of subbalance equations the set of material batches is further structured in "process areas" or "transfer streams", respectively, and "key measurement points". In our model, a batch is realized by a specification of the set $X = (X_1, \dots, X_n)$ of quantities available for measurement. Further, the time has to be specified when the batch "existed" and was available for measurement. For inventory batches, this is the time of inventory taking, for transfer batches the time when the transfer actually (or formally) occurred.

The correlation between the two structures of the model, the batch structure on the one and the "measurement instruments" on the other side, is assumed to be batch specific. I.e. for every potentially existing batch it is specified which of the quantities X_1, \ldots, X_n are to be measured and with which specific "instrument". This also defines function $F(X_1, \ldots, X_n)$ to be used to calculate the Pu-content of the batch.

III. The Computer Program

1. The Function and the Output of the Program

The functions of the measurement simulation program and its output data are defined by the input requests of the NRTA data evaluation programs developed by D. Sellinschegg. One function is quite obvious, namely to provide simulated time series of MUF-data for NRTA data evaluation. It should be possible to manipulate these data in such a way as to reflect fluctuations due to facility operation and realistic effects of the measurement system in order to study the performance and robustness of the proposed evaluation procedures.

The other function is to derive from the measurement system model the parameters for the stochastic model assumed in the data evaluation procedure. The data evaluation procedure proposed by D. Sellinschegg is based on the CUR test statistic. Meanwhile, Mr. Sellinschegg developed two completely different approaches to determine this test statistic from the original MUF-data time series, which also require completely different model parameters as input.

The first approach he developed was based on Kalman filters. As it is well known in this approach a Gauss-Markov process is assumed as stochastic model. In this model the subsequent state is completely determined by the preceding state and the transition matrix between these two states. Systematic errors, however, can be introduced only by a special treatment (i.e. inclusion in the state equation of the system).

The measurement system simulation program in the Kalman filter version has, therefore, to provide as model parameters the error variances and covariances for inventory and for net transfer and separately for the systematic and random, additive and multiplicative components of the error model. However, it is sufficient to provide only the covariances between two succeeding material balance periods. In the second approach, now followed by Mr. Sellinschegg, the matrix which transforms the series of MUF-data into the vector of measurement residuums is directly determined in a recursive way, starting from the covariance matrix of the MUF-data series. Systematic errors no longer require a special treatment, therefore the various error contributions can be combined. But instead the complete covariance matrix for the MUF-data series has to be provided by the measurement simulation program. It turned out that this cannot be done by a simple extension of the Kalman filter version because of excessive storage requirements. A new version of the measurement simulation program is in work to overcome this problem.

2. The Interface with the Facility Operation Simulation Program

For the measurement simulation program, the facility operation is reduced to the generation of sets of (physical) source data for batches of nuclear material. Because these batches are defined by the physical containments in the facility (e.g. buffer or process tanks), no principal interface problems should exist between facility operation model and measurement simulation model as long as both assume an identical facility design.

Because of the complexity of the simulation model for the chemical process in a reference reprocessing plant being developed by Canty, Spannagel and Voss, we found it preferable to have a simpler model of the facility operation available for test and special simulation runs. For test purposes a very simple static facility model was integrated in the measurement simulation program. This model assumes a static **inventory in all containments. Only in input** and output **buffer tanks** is the inventory determined as to maintain validity of the material balance equation. This model was further developed by F. Voss so that it is now able to simulate the filling and emptying of all buffer and process tanks of the facility, assuming a constant inventory for all process components. It is now available as a subroutine to the measurement simulation model. The interface of the complex process simulation model will be realized by data exchange via external disc storage.

Common to all three facility operation models now available or under development is that they provide only the solution volume and the Pu-volume concentration as source data of a batch and thereby limit the inherent possibilities of the measurement model to this simple case.

3. The Program Structure and Operation

In the measurement simulation program (in its version for Kalman filter evaluation and using the stationary facility operation model by F. Voss) the facility operation model and the measurement system model (i.e. the set of available "instruments" and the specification of the error model for them) are realized by two subroutines WAAMOD and MESSIM, respectively, whereas it is the function of the main program to establish the coordination between both models. Within this structure further subroutines are used to realize special functions as convenient. Those three main program elements are themselves subdivided each in an initializing part and an computational or simulation part. This is realized for the two subroutines by means of a second entry directly leading to the computational part. The operational sequence of the program system is as follows:

First the model structure, i.e. the data fields and their (maximal) dimensions, is defined. Then these models, i.e. their actual dimensions and data values, are specified by means of input data. Included in this specification part are the definition of the balancing strategy (i.e. balance period and number of balances to consider) as well as the periods for recalibration of the individual "measurement instruments". The computational part of the program is formulated for one material balance and will be iterated as often as requested. It starts with a section where the data of the last 3 balance periods, as far as required for determination of the covariance of MUF(k) and MUF(k-1), are saved. Then the facility model subroutine is called which determines transfers and transfer times within the period and the true batch source data for transfer and for inventory batches. Afterwards the transfer batches are ordered according to measurement time and in a Do-loop over all batches, transfers and inventory, the subroutine MESSIM is called to simulate the measurement of the batch source data. Then the Pu-content in the batch and the failure propagation coefficients for this determination are calculated by corresponding subroutines.

In the next step the true and the measured value of the Puinventory are summed up separately to the net transfer T_K and to the inventory I_K (where K denotes the balance period number) over all batches. At the same time the various contributions to the covariances of T_K and I_K with the corresponding terms: T_K , I_K , T_{K-1} , I_{K-1} , T_{K-2} , I_{K-2} are summed up separately for the four error components of the error model.

Finally, the true and measured value of MUF and the covariances of MUF(K) with MUF(L), L = K and K-1, are computed and the calculational results written on a TSO output data set for use by the data evaluation program.

IV. First Results

Until now no realistic simulation runs with the measurement simulation program have been carried out combined with a subsequent NRTA data evaluation run. Nevertheless some qualitative conclusions which were not expected in this clarity, can already be drawn from the test runs made so far, though no realistic failure estimates but only invented numbers which appeared not too unrealistic, have been used.

The first surprising result were the small values for the error variances of inventory and net transfer as well as of MUF-values. This in spite of the fact that just to limit this effect, relatively large additive contributions to systematic as well as random errors had been chosen. It turned out that in case of no recalibration the additive systematic contributions cancelled better than the multiplicative ones, whereas the random errors were clearly dominated by the additive contributions.

The next unexpected result concerned the covariance structure itself. Covariance terms revealed as dominating which were expected to be neglectable and vice versa. However, this turned out to be not a stable effect but due to peculiar assumptions concerning error model specification and recalibration strategy. As the NRTA data evaluation strongly depends on the covariance structure of the data to be analyzed, these influencing factors have to be further investigated.

In all cases treated so far, the covariance structure was remarkably simple and determinated always by some few dominating terms. All the complicated correlations taken into account in the model, even inventory changes by up to 30 %, resulted only in small fluctuations of this main structure. This gives rise to the hope that it might be possible to describe adequately the real covariance structure by a rather simple model. The application of NRTA evaluation procedures

STR-116

ON THE ESTIMATION OF SOLVENT PLUTONIUM INVENTORIES IN NEAR-REAL-TIME MATERIALS ACCOUNTANTY

J. E. Lovett

System Studies Section Division of Development and Technical Support Department of Safeguards Interntional Atomic Energy Agency Vienna, Austria

June 1982

STR-116 June 1982

ON THE ESTIMATION OF SOLVENT EXTRACTION PLUTONIUM INVENTORIES IN NEAR-REAL-TIME MATERIALS ACCOUNTANCY

J.E. Lovett International Atomic Energy Agency Vienna, Austria

1. Are Estimates Necessary?

The essence of near-real-time materials accountancy is that physical inventories should be taken at relatively frequent intervals, such as daily or weekly, on an in-process basis, and that the resulting MUF data should be analyzed using techniques which recognize its time sequential nature. Since the inventories are taken on an in-process basis, it rarely is possible to measure all nuclear material physically present. The unmeasured material appears as MUF during the first material balance period; thereafter only variations in the quantity of unmeasured material appear as MUF.

For reprocessing facilities, it has been suggested that n.r.t. accountancy could be implemented by including only buffer storage tanks in the measured in-process inventories. The solvent extraction systems, in this simplified model, would be treated as containing a constant but unmeasured quantity of plutonium. The in-process inventories would be scheduled to coincide with the routine emptying of the product evaporator so that the evaporator inventory also would be small and constant, if not actually zero. Such a model, it is argued, would involve little or no increased effort on the part of facility operators, and would avoid political questions as to how much information relative to solvent extraction systems should be released to inspectors. It is also argued that this simplified model should produce results which would be essentially equivalent to those based on a more complex model in which solvent extraction system inventories are measured or estimated.

Obviously, the success of this simplified model is heavily dependent on the extent to which the solvent extraction system plutonium inventory is truly constant. If, as some claim, solvent extraction system plutonium inventories vary no more than \pm 5% or so during normal operation, then obtaining a more precise direct estimate might be of questionable value. There is no published data to support such a claim, however, and the next section argues that in fact the plutonium inventory probably will vary over a much wider range. Since a process variation of, say, 5000 gms Pu, has the same effect on MUF as a measurement with a standard deviation of 5000 gms Pu, the question is not trivial.

2. How Constant Are Solvent Extraction System Inventories

2.1 Variables Affecting Pu Inventory

The plutonium inventory in a solvent extraction system is a function of the following variables:

a) <u>type of contactor used</u>. Centrifugal contactors have very small residence times, and therefore very small plutonium inventories. Pulse columns and mixer-settlers have significantly longer residence times, and therefore correspondingly larger inventory holdups. For a given plant capacity pulse columns have the largest holdup.

b) <u>plant capacity</u>. For a given type of contactor increasing a plant's design capacity translates more or less linearly into a corresponding increase in the size of the solvent extraction systems, and therefore the plutonium inventory.

c) <u>burnup level of spent fuel processed</u>. The primary component of spent fuel is uranium, which has a solubility limit in nitric acid solution in the range of 200 g/l. Most flow sheets are designed to operate at about 180 g/l. At 30 000 MWd/t the corresponding plutonium concentration is on the order of 2 g/l. At 10 000 MWd/t it may be only 1 g/l. In the first extraction cycle these variations translate directly into inventory variations. Later extraction cycles may be somewhat less affected, but to a first approximation variations in feed concentration are transmitted through all cycles until the product evaporator is reached.

d) <u>operator-controlled process variables.</u> The process operator has the ability to vary a number of parameters which will (or at least may) affect the total plutonium inventory. Examples include varying the flow rates of aqueous feed, organic extractant, nitric acid scrub, or aqueous strip solutions. Since a solvent extraction system operates full (to overflow lines) at all times, varying one flow rate does not necessarily require varying a second, and generalities as to the effect on inventory are difficult.

e) <u>distribution coefficient changes.</u> A fresh sysem, i.e., one that has recently been cleaned out and charged with clean organic extractant, has a distribution coefficient which is determined by the nature and concentration of the organic extractant, the nitric acid concentration, temperature, and the competing influence of fission products. As this system continues to operate, crud formation occurs, leading to a gradual deterioration of the distribution coefficient. (Crud is the technical term used to describe a solid phase, usually finely suspended in the organic phase. It consists primarily of fission products, but it decreases the ability of the organic phase to extract uranium and plutonium.) Deterioration of the distribution coefficient in turn leads to a flattening of the aqueous/organic distribution curve, and to a gradual increase in the plutonium inventory.

f) <u>non-equilibrium operation</u>. If the process operator has varied one or more process parameters to too great an extent, any of several process upset conditions may occur. The most common is flooding, which usually is caused by an insufficient pulse stroke relative to input flow rates. The

— 132 —

result, in terms of materials accountancy, is that the plutonium inventory first increases significantly, then decreases as the accumulated plutonium leaves the system via the aqueous waste stream.

2.2 Inventory Variation Under "Normal" Conditions

If a solvent extraction system is operating "normally", most of the variables listed above are fixed, either by plant design or by operator control. Type of contactor is fixed, plant capacity is fixed, burnup level remains relatively constant over periods of at least a few weeks, and the operator is presumed to make relatively few adjustments to process variables for fear of upsetting a somewhat unstable equilibrium.

This analysis unfortunately omits one very important factor, the quantity of fuel charged to a dissolver and the time allowed for dissolution to proceed. Data collected during n.r.t. accountancy field testing indicate that this input Pu concentration may vary over a range of at least 1.87 (maximum/minimum), not counting rinse batches which are more dilute by a factor of at least four. Adjacent input batches often differ from each other by as much as 15%, even though the fuel dissolved is nominally identical. Within one reactor discharge input concentration variations as large as 1.50 (maximum/minimum) have been observed.

Since this feed solution is transferred more or less directly to the first extraction cycle, inventory variations in the first cycle may be presumed to be approximately of the same magnitude. In the absence of intermediate concentration steps, the same variations will be transmitted to the second and third cycles.

2.2 <u>Inventory Variation Under Non-Normal Conditions</u>

No published data is known to exist concerning the extent of crud formation in solvent extraction systems. It cannot be avoided in any strict sense, especially in the first cycle where both radiation levels and fission product concentrations are high. Since crud is in some way a function of radiation damage to the organic solvent, one may suppose that facility

— 133 —

specific questions of organic purification and recycling will significantly affect actual crud formation.

To the extent that crud does form, the effect is one of reducing the distribution coefficient, which in turn means that more solvent extraction stages are needed to achieve the same levels of purification. Reasonable crud levels normally are not an actual operating problem, because extra stages are included in the design for exactly that reason, but the flattening of the distribution curve leads to a gradual increase in the plutonium inventory.

Turning the question around, assume that n.r.t. accountancy is implemented without estimating the plutonium inventory in solvent extraction systems, and that some sequential data test "detects" a constant MUF of perhaps a few hundred grams per week. If the operator "explains" the apparent MUF loss by arguing that crud formation is leading to a gradual increase in the solvent extraction system plutonium inventory, how is the inspector to know whether he should accept this explanation?

The operation of a solvent extraction system is to a significant extent an art rather than a science. In cold scrap recovery or other purification processes where pulse columns can be made of glass and the operator can directly see what is happening, process upsets can be rare occurrences. In spent fuel reprocessing, where the operator has no chance for even momentary glimpses of actual operation, occasional process upsets must be accepted as unavoidable.

The most common process upset is termed flooding. It can occur under either of two circumstances, where the pulse stroke is inadequate to counter the downward flow of the aqueous phase, or where the pulse stroke is excessive, leading to emulsion formation. In either case the plutonium inventory first increases, as aqueous flow continues without a balancing outflow of plutonium-rich organic, then decreases as unextracted plutonium flows out in the aqueous waste. Corrective measures are virtually nonexistent; the operator must shut the system down, wait for the phases to settle out, and then carefully restart the system. Once back in operation, there is also a quantity of plutonium in aqueous waste to be recovered. If the operator is monitoring his aqueous waste carefully, frequently, and on a real time basis, it is possible to detect conditions leading to flooding before the column actually floods. In this case it may be possible to change the flow rates or the pulse stroke, or both, in time to prevent an actual upset.

3. <u>A Simplified Inventory Model</u>

Consider a solvent extraction system which is totally empty. At time t_o a flow of V_f litres per minute of aqueous feed containing C_f grams per litre of plutonium is started and, simultaneously, a flow of organic solvent is also started. Flow continues until at some later time t_1 the system becomes full and both aqueous and organic phases begin to flow out of their respective outlets. At the instant t_1 the plutonium inventory in the system is given by

$$H = V_f \times C_f \times (t_1 - t_0) \qquad (eq. 1)$$

This result is totally independent of whether extraction occurs. However, it does assume that neither C_f nor V_f changed during the time period $t_1 - t_0 = R$ (defined as the system residence time).

If at time t_1 a steady state equilibrium exists, and if neither C_f nor V_f change as a function of time, equation 1 will continue to hold for any time after t_1 . These are, of course, exactly the conditions which the system operator would like to achieve, but there are many secondary factors which can affect system operation, and maintaining a steady state equilibrium for a long period of time is easier said than done. It is, therefore, necessary to consider the effect on plutonium inventory of various deviations from desired behaviour.

If C_f varies over time but extraction conditions do not change, equation 1 still remains at least theoretically valid. If the change occurred at least R minutes prior to the time in question, the system may be
treated as having "settled in" on a new C_f , and equation 1 is valid. If the change occurred more recently some approximation presumably can be reached using an average C_c .

Similar arguments hold with respect to V_f . If the system has had time to settle in on new operating conditions then equation 1 is valid. If the system is still in a transition state then either the equation must be treated as a time integral or some approximation must be found.

In any operating solvent extraction system only some fraction of the stages are actually needed for extraction. The remaining stages serve important functions, notably raffinate washing and organic scrubbing. Of primary importance here, some extra stages are usually provided to ensure that extraction will be complete even if less than ideal conditions lead to a deterioration of the effective distribution coefficient.

It is not necessary for safeguards that the exact number of stages needed for complete extraction be known, so long as the "knee" in the aqueous plutonium concentration curve remains constant. Indeed, in most systems the definition of a "stage" is more theoretical than physical, and the operator himself may have only a general knowledge of the exact location of this knee.

Of more importance is the possibility that crud formation, or some other secondary factor, has led to an alteration of the shape of the extraction curve, shifting the knee in the aqueous plutonium concentration to a later stage. If the alteration is significant it should also lead to an increase in the plutonium concentration in the aqueous waste. This latter effect may not be measurable if enough extra stages have been included, but one may hypothesize that if the effect is not measurable it may also not be significant. Thus it seems logical to suggest that a better representation of the plutonium inventory in a solvent extraction system can be given by:

 $H = [A \times C_{f}] + [B \times C_{w}]$ (eq. 2)

— 136 —

where $A = V_f \times R$ $C_w = Pu$ concentration in aqueous waste B = constant or function to be defined

The first part of this equation, $A \ge C_f$, represents the plutonium inventory under the assumption of steady state equilibrium conditions. The second part, $B \ge C_w$, represents a correction for non-equilibrium operating conditions.

To date no one has suggested a physical definition for the parameter B. Indeed, no one has demonstrated that practical variations in extraction system operation can be measured by measuring the waste concentration. Theoretically the effect must be there, but practically speaking it may not be detectable.

4. Experimental Data

Relatively little experimental data is available, at least partly because relatively little reprocessing is occurring. As previously noted, field test data which are available indicate that input Pu concentrations are likely to vary by as much as a factor of two under normal operations, and by larger factors when dissolver rinsing occurs.

Equation 1 has been used to compute solvent extraction system inventories for the same field test data. The mean inventory (calculated) was 3092 gms Pu, and the standard deviation was \pm 1601 gms Pu, or about 52%. This compares with a "guesstimated" constant inventory of 2500 gms used in earlier results. Introduction of these inventory calculations into the n.r.t. accountancy data results in a qualitatively noticeable smoothing of the CUMUF graph, but it is not known whether the calculated values truly represent the actual Pu solvent extraction system inventory. It is also not known whether the system was at equilibrium at all times (indeed, it is strongly suspected that it was not always at or near equilibrium). Obviously, further work is needed.

Criteria for Decision Procedures for a Sequence of Inventory Periods

- Topics for an Experts' Meeting -

by

Rudolf Avenhaus

.

Hochschule der Bundeswehr München Neubiberg, Federal Republic of Germany

November 1980

Introduction

The application of the principle of material accountancy to international nuclear material safeguards has been laid down in the model agreement between the IAEA and the states subject to the Non-Proliferation Treaty (IAEA 1970); the statistical problems, which are connected with its use and which arise from the inevitable measurement uncertainties, have been analyzed since more than 20 years (Stewart 1958). Nevertheless, there still exists a number of open questions which should be answered as soon as possible in view of some recent developments, both on the "political-administrative" side, e.g.

- emphasis on short detection time,
- emphasis on a high probability of detection for specific diversion strategies ("abrupt", "protracted"),

as well as on the "scientific-analytical" side, e.g.

- emphasis on "batteries of tests", each constructed for a specific diversion strategy (Shipley 1980),
- emphasis on the use of estimation theories, especially Kalman Filters (Pike and Morrison 1975).

At the moment, the situation may be characterized by a certain confusion: There are many objectives, boundaries and qualitative constraints on one hand and many test and estimation procedures on the other hand, however, there is no agreement on

- the quantitative formulation of objectives and boundary conditions,
- the priorities of objectives in case they are in conflict to each other (which actually happens),
- the diversion strategies which have to be taken into account, and
- the appropriate choice of statistical evaluation procedures (or, more modestly expressed, the direction into which the development of procedures should be guided).

It is the idea that a limited number of experts in this field shall discuss these questions and work out a kind of program which can help to structure the future

international development. In the following, the pending problems shall be discussed in more detail; at the end of this paper a list of questions is given which may serve as a guide for the experts' discussion.

General Formulation of the Problem

Let us first establish the data base and furthermore, let us quantitatively formulate the problems to be solved:

We consider a well-defined material balance area and a reference time interval $[t_0, t_n]$, e.g. one year. At time points t_0, t_1, \ldots, t_n physical inventories are taken, and for the intervals $[t_0, t_1]$, $[t_1, t_2], \ldots, [t_{n-1}, t_n]$ the net transfers T_1, T_2, \ldots, T_n (inputs minus outputs) are determined. The distributions of the measurement errors of these quantities are assumed to be known. Under the null hypothesis H₀ the expected values of the MUF₁-variables, defined by

 $MUF_{i} = I_{i-1} + T_{i} - I_{i}, i=1,...,n,$

are assumed to be zero.

It should be noted that there may be plant internal losses which cause non-zero expected MUF values. We will come back to this point; for the sake of simplicity, however, we assume for the moment that such losses do not exist.

The general problem is to make with the help of the 2n+1 observed quantities $I_0, I_1, \ldots, I_n, T_1, \ldots, T_n$ statements about the possible diversion of nuclear material during the inventory periods $[t_0, t_1], \ldots, [t_{n-1}, t_n]$: In case of diversion of the amount M_i in the interval of time $[t_{i-1}, t_i]$, $i=1, \ldots, n$, the expected value of $I_{i-1} + T_i - I_i$ is M_i, therefore, a decision has to be made if a non-zero observed value of $I_{i-1} + T_i - I_i$ can be explained by measurement errors, or if material has been diverted.

Naturally, part of the problem is to determine the appropriate number n of inventories during the reference time $[t_0, t_n]$.

For one inventory period, the solution of the decision problem is simple: the null hypothesis H_0 is (if we omit the index i)

$$H_{o}: E(MUF) = 0,$$

and the alternative hypothesis is

$$H_1: E(MUF) = M > 0.$$

The decision procedure is, if MUF is the observed value of the random variable MUF,

 $\hat{MUF} \leq s: H_{o} true$ $\hat{MUF} > s: H_{1} true.$

The relation between the significance threshold s and the false alarm probability α is

$$1-\alpha = \Phi(\frac{s}{\alpha}),$$

where $\sigma^2 := var(MUF)$, and where Φ is the normal distribution function. The probability of detection $1-\beta$ as function of the false alarm probability is

$$1-\beta = \Phi(\frac{M}{\sigma} - U_{1-\alpha}),$$

where U is the inverse of Φ .

Objectives and Boundaries

Any statistical decision permits the possibility of a false accusation. Therefore, first it has to be decided which false alarm probability can be tolerated and to which area and interval of time its value shall be related.

It has been proposed at several occasions that it shall be related to one material balance area and that also in case of sequential decision procedures it shall be fixed for a well defined reference time interval under consideration. For a given value of the false alarm probability for one material balance area and the reference time interval $[t_0, t_n]$ it has been shown under very general assumptions (Avenhaus and Frick 1974, Frick 1979, Avenhaus 1980) that it is best in the sense of the *overall probability of detecting a diversion of the total amount M of nuclear material* not to take into account the in-between inventories I_1, \ldots, I_{n-1} , but to test only the overall balance

$$\sum_{i}^{\text{MUF}} = I_0 + T_1 - I_1 + I_1 + T_2 - I_2 + \dots + I_{n-1} + T_n - I_n = I_0 + \sum_{i}^{\text{T}} I_n - I_n$$

It should be noted, however, that it is assumed that this is the best inspection strategy against *any* diversion strategy leading to the total diverted amount M. If the inspector knew that the operator, e.g., will divert the same amount $\frac{1}{n} \cdot M$ per inventory period or, e.g., that he will divert the total amount M with probability q_i during the i-th inventory period, i=1,...,n, then a different test procedure than that based on the global balance would lead to a higher overall probability of detection.

The objective "short detection time" poses several problems. First, this objective cannot be formulated quantitatively so easily as the one discussed so far: As there exists a non-zero probability that a diversion will not be detected during the reference time, we cannot simply take as a criterion the expected detection time

$$\begin{array}{c}
n\\ \Sigma \quad i \cdot p_i,\\i=1
\end{array}$$

where p_i is the probability of detecting a diversion for the first time at the end of the i-th inventory period, as we have

$$\sum_{i=1}^{n} p_i < 1.$$

One possibility would be to take the expected detection time under the condition that detection actually takes place during the reference time.

Another difficulty of the conditional expected detection time is that it is not necessarily a monotone function of the number of inventory periods per reference time (Avenhaus and Frick 1974) and that the minimum depends on the numerical data and cannot be determined analytically which means that simplified "fist formulae" cannot be derived.

Let us assume that a quantitatively formulated objective "short detection time" has been agreed. As both criteria, high probability of detection and short detection time, may be conflicting it has to be decided in which way the tradeoff between these two objectives shall be resolved. The only theoretically satisfying answer to this difficult problem is to define *payoff parameters* for both the inspector and the operator for the different outcomes of the "inspection game" (detected or not detected diversion during the i-th inventory period) which take into account the risk as well as the time aspect (Avenhaus 1980) and to analyze the appropriate game theoretical model. Naturally, the values of these parameters can hardly be estimated, therefore, a conclusion should be drawn if at least ratios of such payoff parameters can be estimated or, if even this is not possible, in which way the different objectives shall be weighted.

In 1979, a Peer Review Group was established by the USNRC in order to answer five questions concerning the applicability of game theoretical models to material accountancy problems (Bennett et al. 1979). The group members based their findings primarily on one specific paper (Siri et al. 1978) which did not address to those concrete questions which are discussed here thus, their results are only of general value for our purposes.

It follows already from the fact that the expected detection time - contrary to the overall probability of detection - cannot be derived from a game theoretical model that the expected detection time does not represent a natural objective.

It has already been mentioned that recently specific diversion strategies have been discussed which shall be detected with as high a probability of detection as possible. In principle, one can construct tests which are best for a uniform (protracted) diversion or which are best for an abrupt diversion, but naturally there exists no test which is best for both these extremes and their mixtures.

One solution to the problem of finding the best test procedure against any of these diversion strategies would be to use all the single best tests simultaneously. This, however, poses the difficult analytical problem of determining the overall false alarm probability: As in all tests the same data are used, the different test statistics are highly dependent. Another possibility would be to treat the two problems of detecting an abrupt and a protracted diversion as completely different inspection problems which would mean that different false alarm probabilities would be fixed for the two test procedures, and the fact would be ignored that the same data are used for both tests.

— 145 —

This idea is an extension of the idea of separating the *physical protection* problem (detection of a diversion on the subnational level) from the *international safeguards* problem, even though in practice many measures may serve both purposes at the same time.

In addition to the objectives and boundaries discussed so far, there exist further constraints which frequently cannot be formulated quantitatively ("non-intrusiveness", "minimization of plant operations disturbances" etc.). It should be attempted, however, to establish a list of these constraints and to give arguments for possible consequences of these constraints to the test procedures to be developed.

Role of Estimation Procedures

The detection of a diversion of nuclear material can principally be achieved only by means of *test procedures*. Nevertheless, in the last years *estimation procedures* have been discussed at length also in connection with international nuclear material safeguards, even though it is not their objective to estimate the diverted amount but only to detect it.

One objective for the development of such procedures is the estimation of plantinternal losses. This is important also for international nuclear material safeguards, because these losses contribute to a non-zero MUF-value, therefore, their distribution has to be known in order that they can be separated from an eventual diversion. We demonstrate this for one inventory period (again omitting the index i):

The null hypothesis H now is given by

 H_{o} : MUF = e+L,

where e is the total measurement error and ℓ the loss, with known moments

$$E(e) = 0$$
, $var(e) = \sigma^2$, $E(l) = L$, $var(l) = \sigma_l^2$.

Furthermore, the alternative hypothesis H, is given by

$$H_1: MUF = M+e+\ell$$
,

where M is the diversion. Then the decision is

$$\frac{\hat{\text{MUF-L}}}{\hat{\text{MUF-L}}} \le s: H_0 \text{ true}$$

$$\frac{\hat{\text{MUF-L}}}{\hat{\text{MUF-L}}} > s: H_1 \text{ true},$$

and the probability of detection as function of the false alarm probability is

$$1-\beta = \Phi\left(\frac{M}{\sqrt{\sigma^2 + \sigma_{\rho}^2}} - U_{1-\alpha}\right).$$

Another objective for the development of estimation procedures is the expectation that one might arrive at this way at "best" test procedures in cases where optimal tests cannot be constructed directly. In fact, there exist many relations between so-called "sufficient" statistics and best tests (see e.g. Witting 1980). The idea is that the operator uses a certain diversion strategy during the reference time which can be "revealed" in the first inventory periods, and that this information can be used in later periods for the application of appropriate tests. However, this does not work necessarily: Stewart's estimate of the starting inventory does *not* lead to the best test in the sense of the overall probability of detection.

Analysis of Test Procedures

. ...

Let us assume that the objectives and boundaries have been agreed upon. Let us assume, furthermore, that it has been agreed that one should try to establish a theoretically fully satisfying game theoretical model of the problem, but that in addition to this simple and practical test procedures shall be developed without the use of payoff parameters. Which way should one proceed?

First, it has to be clarified whether or not tests with *indifference regions* make sense and also whether or not *sequential procedures* shall be taken into account (Shipley 1980).

Both questions can be answered only if it is clear which *action levels* follow after a specific decision: Shall the plant be shut down in case of a significant MUF value? Shall different actions be taken in the cases of "indifference" and of "significance"?

Thereafter, it shall be decided if "batteries of tests" shall be envisaged which means to solve difficult analytical problems, or if single tests shall be applied which may be best for one specific diversion strategy and hopefully not so bad for others, or if completely separated procedures shall be applied, independently of their statistical dependencies because the same data base is used in all procedures.

Conclusion: List of Questions

In order to structure the discussion of the experts about the topics outlined so far, a list of questions is formulated which should be answered as precisely as possible. Naturally, this list cannot be exhaustive (e.g., the important aspect of data verification has not been mentioned at all), nor can it be expected that all questions will find a satisfying answer.

- 1. To which framework in space and time shall the value of the false alarm probability be related?
- 2. Which quantitative criterion for the objective "short detection time" shall be used?
- 3. Do models which use payoff parameters make more sense than a purely theoretical one?
- 4. How can the tradeoff between the two objectives "high probability of detection" and "short expected detection time" be resolved in a pragmatical way,

i.e. without the use of payoff parameters?

- 5. Which boundary conditions have to be taken into account? Can they be formulated quantitatively? Which consequences do they have?
- 6. Which diversion strategies have to be taken into account? Abrupt diversion? Protracted diversion? Mixed strategies? Which priorities can be formulated with respect to these strategies?
- 7. What is the purpose of estimating the amounts of diverted material in the framework of international nuclear material safeguards?
- 8. Do tests with indifference regions make sense, and do sequential tests make sense in view of the action levels which have been formulated so far?
- 9. In which direction shall research and development in this area go: Shall batteries of tests be developed or shall one concentrate on one test which is optimal for one diversion strategy and not so bad for others, or shall one envisage completely separated procedures without taking into account the dependencies of the statistics?

Finally, an important organisational question is posed:

10. Which steps can be taken in order to discuss the findings of this meeting with the international nuclear material safeguards community and finally to reach a consensus?

Literature

- Avenhaus, R. and H. Frick (1974), Game Theoretical Treatment of Material Accountability Problems, Part II. International Institute for Applied Systems Analysis, RR-74-21.
- Avenhaus, R. (1980), Best Tests for Means of Multivariate Normally Distributed Random Variables and their Application to Material Accountability Problems, To be published.
- Bennett, C.A.; Goldman, A.J.; Higinbotham, W.A.; Jaech, J.L.; Lucas, W.F. and R.F. Lumb (1979), A Review of the Application of Strategic Analysis to Material Accounting. USNRC-Report NUREG/CR-0950.
- Frick, H. (1979), On an Application of Game Theory to a Problem of Testing Statistical Hypothesis. International Journal of Game Theory, Vol. 8, Issue 3, pp. 175-92.
- International Atomic Energy Agency (1971), The Structure and Content of Agreements between the Agency and States Required in Connection with the Treaty on the Non-Proliferation of Nuclear Weapons. INFCIRC/153.
- Pike, D.H. and G.W. Morrison (1977), A New Approach to Safeguards Accounting. ORNL/GSD/TM-25.
- Shipley, J. (1980), Notes for the Seminar on Data Generation and Evaluation for Safeguards from July 7 to 9, 1980 at LASL, Los Alamos, N.M., Unpublished.
- Siri, W.E.; Ruderman, H. and M. Dresher (1978), The Application of Game Theory to Nuclear Material Accounting. USNRC-Report NUREG/CR-0490.

Stewart, K.B. (1958), A New Weighted Estimate. Technometrics 12, pp. 247-58.

Witting, H. (1978), Mathematische Statistik - Eine Einführung in Theorie und Methoden, 3. Auflage, B.G. Teubner, Stuttgart.

EXON NUCLEAR COMPANY, Inc.

600 - 108th Avenue N.E., C-00777, Bellevue, Washington 98009, Telephone (206) 453-4300

August 27, 1982

Dr. Dipak Gupta EKS Kernforschungzentrum Karlsruhe Postfach 3640 D-7500 Karlsruhe Federal Republic of Germany

Dear Dipak:

At the July 27, 1982 meeting between us and Carl Bennett, you had asked that I draft Section 6 of the Overview report, the section dealing with descriptions of the 8 tests. It was my understanding that these descriptions be largely non-mathematical, pointing out in narrative form the differences between, and similarities of, the various tests.

Enclosed you will find my draft. I would suppose that some editing will be required to fit this section in with the balance of the report. Please feel free to edit accordingly. Except for the required editing, I trust that this draft is generally responsive to what you had in mind. I am also sending this to Beedgen, Markin, and Sellinschegg to give them the opportunity to review and edit if what I have written is not properly descriptive of their test procedures. By this copy, I ask that they send any comments directly to you.

I look forward to seeing you at the Symposium in November.

Best regards,

John L. Jaech, Consultant Statistics/Safeguards

JLJ:jes

Enclosure

cc: (w/att.)

R. Beedgen C.A. Bennett J.T. Markin D. Sellinschegg H.E. Williamson File/LB

AFFILIATE OF EXXON CORPORATION

In this section, a description is given of the eight basic statistical test sequences investigated in this study.

MUF

The MUF test is a test on the material balance for a given period. Letting D_i be the observed MUF for period i, loss detection is said to occur if D_i exceeds some critical value determined by the value of α and the values of the measurement error standard deviations. The MUF test does not take into account any prior history. It is aimed at detecting an abrupt loss, one that occurs somewhere within the material balance period in question. As a test sequence, the MUF test is applied at each material balance period and loss detection over the P periods occurs if at least one of the MUF tests returns a significant result, i.e., if at least one MUF exceeds its critical value. The α value over all P tests is controlled by reducing the size of the significance level for each individual test.

CUMUF

The test statistic to be applied in period i is denoted by T_i and is the sum of the individual observed MUF's beginning at some point in time and extending through period i:

$$T_{i} = \sum_{\substack{j=1\\j=1}}^{i} D_{i}$$

At a given point in time, T_i is independent of how the losses are distributed throughout the i periods. This is the cited advantage of the CUMUF test. As a test sequence, the CUMUF test is applied at each material

balance period, as is the MUF test. Clearly, there is a close correlation between successive CUMUF's.

--- 153 ----

In this study, CUMUF is applied in sequence on the one hand, and only at the end of the 35 periods on the other. The single test in this latter instance is, of course, more powerful than is the 35th such test applied as the last test in the sequence. However, this increase in power is counterbalanced by the lack of timeliness, i.e., the inability of the test to detect losses that occur early in the sequence of time periods. Since the MUF test and the CUMUF test are directed at quite different loss patterns, in Phase 1 of the study for one test sequence under investigation, both the MUF and CUMUF tests were applied at the end of each material balance period.

Uniform Diversion

The test statistic is designed to detect uniform losses. Since uniform losses over a number of successive balance periods were the primary loss patterns studied in Phase 2, it would be expected that the uniform diversion test statistic would exhibit good detection capabilities in this study.

The linear statistic in question is the minimum variance unbiased estimate of uniform loss. Specifically, in this study, the statistic was defined for each group of four successive MUF's. It is a moving weighted average of four such MUF's, and it is clear that successive test statistics would be closely correlated.

The weighted average is derived as follows. Let

 $T_i = a_1 D_i + a_2 D_{i+1} + a_3 D_{i+2} + a_4 D_{i+3}$

where the a_j 's sum to 1 for j=1, 2, 3, 4. The a_j 's are chosen to minimize

-2-

the variance of T_i . The first test statistic is calculated at the end of the fourth balance period.

When j=1-4 as here, the calculation of the a_j 's is quite simple. For more complex cases, calculational algorithms are helpful. The oft-mentioned Kalman filter is a calculational algorithm used in this instance.

CUMUFR

CUMUFR is an acronym for <u>cumulative</u> sum of standardized <u>MUF</u> residuals. It is designed to detect changes in loss patterns. A uniform loss that occurs in all balance periods would not be detectable with the CUMUFR test.

The MUF residual for period i, MUFR;, is defined as

 $MUFR_i = D_i - E(D_i | D_1, D_2, \dots, D_{i-1})$

where $E(D_i | D_1, D_2, \ldots, D_{i-1})$ is an appropriate linear function of $D_1, D_2, \ldots, D_{i-1}$, so chosen such that MUFR_i has minimal variance. The standardized MUF residual is found by dividing MUFR_i by its standard deviation, σ_i and the CUMUFR test statistic for balance period k is found by summing MUFR_i/ σ_i from 1 to k.

The time series of MUFR's is a linear transformation of the time series of MUF's. They can be calculated exactly by applying this transformation or approximately through use of a Kalman filter.

The CUMUFR test may be applied as a two-sided test or as a one-sided test. In a two-sided test application, periods of losses followed by periods of no losses would also be detectable, whereas for a one-sided test, only periods of losses following periods of no losses would be detectable.

-3-

Note that in applying the CUMUFR test sequence, use is always made of all the MUF data extending back to period 1. This is a principal distinction in this study between CUMUFR and the D_n test discussed next.

Dn

Like CUMUFR just discussed, D_n is aimed at detecting changes in loss patterns. Unlike CUMUFR, in this study n was fixed at 5, i.e., at the end of each balance period, and beginning with period 6, the current MUF is compared with some constant β times the sum of the 5 previous MUF's, where β is chosen to minimize the variance. Specifically, the test statistic for period i is

$$T_i = D_{i+5} - \beta \sum_{j=1}^{i+4} D_j$$

where β is a simple function of the error variances in measuring net transfers and inventories.

In this study, the test was applied as a one-sided test. Thus, a period of losses followed by a period of non-losses would not be detectable.

Sequential Probability Ratio Test

The sequential probability ratio test is related to the CUMUF test in that the test statistic is the cumulative sum of the MUF's. However, the test is now a sequential test in the true sense of the word, as distinguished from a sequence of fixed length tests.

With a sequential test, when the value of the test statistic is calculated at the end of each period, the decision is made to either reject the hypothesis of no loss (i.e., declare that a loss has been detected), accept the hypothesis of no loss, or continue testing. When the hypothesis of no loss is accepted, then the test is restarted, and all prior data are ignored. This restarting of the test and deletion of prior data is what distinguishes the sequential probability ratio test from the CUMUF test described earlier. With the CUMUF test, the MUF data extending

- 155 -

-4-

backward to period 1 are always retained.

Modified Pages Test

The Modified Pages test is also a sequential test in that the test may be restarted with all prior data eliminated when the accumulated evidence indicates that there has been no loss of material.

- 156 —

For the Modified Pages test, the test statistic is

$$T_{i} = C_{i} - \min C_{j}$$

$$j$$
where $C_{j} = \int_{k=1}^{j} (D_{k} - \delta)$

In effect, the test statistic is the current CUMUF minus the largest previous CUMUF, δ being a constant.

The upper threshold (critical value) is a function of some parameter, A, which controls the false alarm rate, and of the period number, i. The lower threshold is zero for the Modified Pages test.

Truncated Sequential CUMUF

Like the sequential probability ratio test, the basic statistic is the cumulative sum of the MUF's. Also, the test is sequential in nature. This test procedure is called a truncated one because after a fixed number of material balance periods, a decision must be made as to whether or not a loss has occurred.

In evaluating this test procedure, a saddle-point solution is also found. The saddle point solution gives a guaranteed efficiency in the sense that it gives the detection probability corresponding to the least favorable loss pattern, i.e., it reacts to a diversion scenario in which the adversary chooses an optimum strategy.

-5- '

On Evaluation Methodology for Near-Real-Time Materials Accountancy +)

Prepared for the second International Workshop on Near-Real-Time Accountancy in Large Reprocessing Facilities, 24th to 26th February 1982, KfK, Karlsruhe, F.R. Germany

by

Hideo Nishimura, Japan Atomic Energy Research Institute, Tokai, Japan

Summary

In the TASTEX Task-F study, simulated material balance data and experimental MUF values were analyzed using the statistical evaluation procedures which have been developed at the Los Alamos National Laboratory.

For this purpose a computer code "SADAC" (SAfeguards Data Analysis Code) has been developed.

Following descriptions are referred to: JAERI-memo 9532, STUDY OF THE APPLICATION OF NEAR-REAL-TIME MATERIALS ACCOUNTANCY TO SAFEGUARDING REPROCESSING PLANTS, K. Ikawa, e.al, May 1981.

+) submitted as private communication

Statistical Evaluation Procedures

Statistical evaluation procedures used in the study are essentially those developed at the Los Alamos National Laboratory. The complete computer package includes four statistical tests, a straightforward cumulative sum (CUSUM) test, a uniform diversion test based on the Kalman filter statistic, a variance test, and a two directional test based on two Kalman filter models operating in opposite directions. The purposes of these tests are as follows :

- (a) CUSUM this statistic provides a relatively powerful test which is in general not dependent on an assumed diversion pattern,
- (b) uniform diversion as the name implies, this test is more sensitive when the divertor follows the nominally optimum strategy of diverting a uniformly small quantity during each material balance period,
- (c) variance test if a would-be divertor attempts to defeat the statistical tests by diverting in a random manner, the observed variance of the MUF data will be significantly larger than the variance derived from the measurement uncertainty for each material balance. The variance test is designed to give an increased detection sensitivity against randomized diversions by detecting this increased variance in the data,
- (d) two-directional test this test recognizes that a revised estimate of the inventory at any earlier point in time can be derived from a consideration of subsequent flow and inventory data. In borderline situations it is expected that a two-directional test would be more sensitive to possible abrupt diversions.

Although early feasibility studies using simulated data considered all four of these tests, most work has been with only the first two. It seems likely that in actual practice primary reliance will be on the CUSUM and uniform diversion tests, and that the other tests will be used only to provide supplementary evidence, or to suggest the need for further investigation of possible borderline situations.

Development of Computer Code

In order to analyze material balance data, a computer code "SADAC" (<u>SAfeguards Data Analysis Code</u>) has been developed using the data analysis and sequential decision techniques.

The sequential decision procedure adopted in SADAC has a small addition to the original one. These additions are as follows;

- (1) SADAC tests against both gain and loss, and it indicates an alphabetical symbol if the material unbalance is positive, while indicates a numerical, if the unbalance is negative.
- (2) When a decision test obtains a result with a false alarm probabillity greater than 0.5 at the first point of a subsequence, i.e., at the point of $\{r_1, r_2\} \mid r_1 = r_2\}$, the decision test indicates the symbol 'T' at this point on the alarm-sequence chart and is terminated immediately in the original decision test procedure. On the contrary, in SADAC, the decision test continues examination of the subsequent material balance data so long as the false-alarm probabilities of succeeding tests continues to decrease.

.

C.A. Bennett, Battelle-HARC, Seattle, USA 11/15/82

PARAMETRIC DESCRIPTION OF LOSS PATTERNS

1. Scope of the Description

1.1 A description is needed to provide

- (a) a space of alternate hypotheses for applying methods of determining "best" tests;
- (b) as a framework for comparative studies based on simulation procedures.

1.2 Consideration will be restricted to a single MBA. The most general problem in this case considers a continuing sequence of inventories y_i at times t_i , $i = 0, \pm 1, \pm 2$, and the corresponding net transfers x_i between times t_i and t_i .

 $y_{i} = \mu_{i} + \eta_{i} , \quad i = 0, \dots, n$ $x_{i} = \mu_{i} - \mu_{i-1} + L_{i} + \epsilon_{i} , \quad i = 1, \dots, n$ $D_{i} = y_{i-1} + x_{i} - y_{i} = L_{i} + \eta_{i-1} - \eta_{i} + \epsilon_{i}$

1.3 Consideration will be stricted to random errors η_i und ε_i with E(η_i) = E_i(ε_i) = 0 and known distributional properties. This means that we are not interested here in describing or testing the nature of the random errors in the determination of the values of x_i and y_i . Variance components corresponding to measurement biases will usually be assumed to be described by the variance-covariance matrix of these observations, but in some instances the existence of constant biases will be postulated. 1.4 The concern here is therefore with characterization of the expected values μ_i of the inventory and L_i of the inventory differences.

2. Extent and Nature of Available Observations

2.1 First consider limitations on the sequence of observations to be considered. In general, we can assume that:

2.1.1 The sequence extends from initiation of facility to present, i.e., from some initial inventory $\mu_0 = 0$ to a sequentially increasing value of i. (We may choose to base our estimation procedures on a subset of these observations, but are not restricted from doing so.)

2.1.2 However, for operational or legal reasons we can be restricted to either:

2.1.2.1 the observations subsequent to some initial time T_{o} ; or

2.1.2.2 the observations during a fixed period of time (i.e., a calendar year).

2.2 There is no inherent reason why the times t_i of inventory taking should be equally spaced. Hypotheses (assumptions) concerning the general loss pattern L_i may involve the time periods $\Delta_i = t_i - t_{i-1}$ between inventories.

3. Whether or not the sequence of observations is limited, we may wish to limit our consideration of possible losses to some subset of periods. There are two reasons for this:

3.1 For legal or administrative reasons we may wish to limit our consideration only to those losses which occurred during a predetermined time period.

— 162 —

3.2 If the total loss is of concern we may wish to prevent the loss rate from becoming arbitrarily small. This has been traditionally done by assuming that after some period of time possible prior losses should be neglected. This can also be done by weighting the importance of past losses by an exponentially or geometrically decreasing function of the elapsed time since the loss is assumed to have occurred. In either case the assumption is with respect to a period prior to the present time, not a fixed period as in 3.1.

4. Sensible descriptions of the loss patterns and the form of the associated estimates or decision procedures depend on whether the losses are assumed to originate from normal material handling and processing or from deliberate diversion.

4.1 For process losses:

4.1.1 There is no reason to arbitrarily limit the number of observations to be considered.

4.1.2 There is no reason to consider any statute of limitations on losses, especially since consequences may be latent and/or cumulative.

4.1.3 Loss mechanisms can be assumed not to depend on any deliberate choice of an optimum loss strategy, but to be the result of some independent process characteristic or operational procedure (e.g., hold up or unmeasured waste).

4.2 For diversion:

4.2.1 The losses, and hence <u>de facto</u> the observations, to be considered may be limited.

4.2.2 Loss mechanisms must be assumed to be deliberately chosen to minimize the chances of detection. Several cases:

4.2.2.1 Knowledge of detection procedure and past data available to the divertor.

4.2.2.2 Past data but not detection procedures known to divertor.

4.2.2.3 Neither past data nor detection procedures available.

4.3 In either case, the absence of consequences may be considered as prima facie evidence of the absence of diversion in past periods.

5. For the case of process losses neither arbitrary limitations on the length of the sequence of observation or the nature of the losses makes any sense. There are three models of interest, all with parallels in the usual methods of process control:

5.1 A constant long term loss rate, characterized by an expected loss L per unit of time and a variance σ_L^2 about this fixed rate associated with a specified time unit. This model best characterizes unmeasured process losses, waste losses or unknown measurement biases. Note that the loss may be proportional to throughput, time between inventories, or both.

5.2 Drifts in loss rate, similar to models for tool wear. Simplest model is linear increase in loss rate with time, starting at some initial point.

5.3 Shifts in expected loss rate due to process changes or changes in the character of the material or equipment.

6. For deliberate diversion, the possibilities can be described as follows:

6.1 A single acquisition over a fixed period. The divertor strategy with respect to the loss pattern over this period will depend on: 6.1.1 Whether the data on which the decision is based is limited to an established period. If so, the acquisition will be:

- 6.1.1.1 optimally allocated with respect to successive data sets
 (optimum timing);
- 6.1.1.2 within each data set, the total diversion will be distributed over the period of acquisition (e.g., distribution of the required diversion in equal amounts over k of the n periods in the data set (optimum loss pattern).

6.1.2 If the diversion is not tied to a fixed data set, then must consider:

6.1.2.1 Balance between rate of diversion and timeliness of acquisition.

6.1.2.2 For a fixed period of acquisition, timing with respect to individual material balance periods, and either random or systematic variation of rate.

6.2 When considering a fixed long term rate of diversion, the strategy must take into account:

6.2.1 The optimum induced random variability in the loss rate.

6.2.2 The possible feedback of information into the detection strategy.

7. There are two specific cases which seem to bracket the patterns of interest:

7.1 The concern is with a loss restricted to a fixed period (e.g., a given calendar year). Corresponding to this fixed period we have n + 1 observed inventories and n observed net transfers. We wish to detect the attempt to divert a fixed total amount M during the year based on these

2n + 1 observations. Testing can be carried out following any inventory and net transfer determination based on all data available up to the time t_i of the ith inventory. Under these circumstances studies of optimum decision procedures require the ability to determine for a fixed testing procedure with an established false alarm rate (probability of "detection" when diversion has occured): (1) what is the divertor strategy which minimizes the chances of detection by the end of the year of an amount M at some value of i; (2) the increased probability of detection as a function of the increased time between acquisition of the total amount and detection. This is the classic case of abrupt vs. protracted diversion over a fixed one-year period where protracted means throughout the year but not necessarily constant. Conversely, for fixed diversion strategies or classes of strategies it may be possible to establish test procedures which have optimum properties, or whose effectiveness is independent of the assumed loss pattern.

7.2 In the absence of administrative and legal considerations the problem that should be considered is the prompt detection of the introduction of some loss pattern. The simplest cases are those in which the shift to a constant loss of varying size is to be detected. The shift is described by a single parameter L. If the expected number of periods required to detect the shift is used as a measure of the effectiveness of the test, then the optimum choice of L should maximize the difference between this number and the number of periods M/L required to accumulate a fixed quality M at a loss rate L. A more complex formulation would consider both the nature of the shift in the expectation L; of D; and the deliberate variation σ_{L} in the i-th loss which would maximize this expected time from accumulation of an amount L, to detection. In particular, one could study the effect of an additional variability σ_{T} associated with a constant shift, as well as additional tests to determine the shift in variability.

— 166 —

TIMELY DETECTION OF MATERIALS LOSS

J. T. Markin Los Alamos National Laboratory, Los Alamos, USA

Timely detection of materials loss from a high-throughput nuclear facility is not attainable under the practice of closing a materials balance at 6-month or yearly intervals. However, where a near-real-time accounting system is in place to gather process measurement data and sequential statistical testing procedures are applied to these data, the possibility of timely detection is improved (see, for example, Refs. 1, 2). This paper compares several sequential testing procedures using expected time to detect a uniform materials loss and the probability of detecting an abrupt loss as the measures of test performance. Sequential tests were selected for evaluation because they (1) allow an immediate decision about materials loss as accounting data are acquired, (2) are sensitive to both low-level materials loss over extended periods and abrupt loss in a single period, (3) signal a loss more quickly on the average than fixed length tests, (4) have bounded false-alarm rates, and (5) allow past data to be eliminated when the procedure decides no prior materials loss.

Detection of materials loss is modeled as the statistical problem of deciding between two hypotheses about the mean of a probability distribution given observations from that distribution. In materials accounting the materials balance mean μ is to be determined by deciding between the hypothesis H_0 of no materials loss ($\mu = 0$) against the alternative hypothesis H_1 of materials loss ($\mu > 0$). Sequential procedures for deciding between H_0 and H_1 consist of a statistic that depends on the observed materials balances, and upper and lower decision thresholds TU and TL.³ At each observation the statistic is compared to the thresholds and a decision is made according to the rule: accept H_0 when $S \leq TL$, accept H_1 when $S \geq TU$, and continue testing

otherwise. Acceptance of H₀ causes prior observations to be removed from consideration and the test to be restarted. Where procedural or legal requirements require a decision at a particular time, test truncation rules allow a decision when the test statistic has not crossed a threshold. Type I and Type II errors for sequential tests are controlled by appropriate choice of the decision thresholds.

The sequential tests to be compared are Page's test, Page's test with a modified decision threshold, and the sequential probability ratio test. Page's statistic⁴ is defined as

$$S(N) = C(N) - Min C(i)$$
$$1 \le i \le N$$

where C(i) is the cumulative sum $\sum_{j=1}^{N} (MB_j - \delta)$ where δ is a constant parameter to be determined. The decision rule for this test is to accept H_0 when S(N) = 0, accept H_1 when $S(N) \ge h > 0$, and continue testing otherwise. The false-alarm rate and detection probability for this test are controlled through the parameters δ and h.

Recently hypothesis testing procedures of power 1 have been developed in which a threshold of the form $T(N) = {N[A^2 + Log(N)]}^{1/2}$ is used to test for an increase in the mean of |C(N)|, where A is a parameter_controlling the false-alarm rate ε .^{5,6} Under this procedure when H₀ is true,

 $P[|C(N)| > T(N), N > 1] = \varepsilon$

and under H₁

P[|C(N)| > T(N), N > 1] = 1.

Because |C(N)| has the same probability distribution as Page's statistic S(N) under the hypothesis H_0 (Ref. 7), the threshold T(N) has been adapted to Page's test. The test procedure consists of accepting H_0 when S(N) = 0, accepting H_1 when $S(N) \ge T(N)$, and continuing testing otherwise. When H_0 is accepted, the test is restarted by eliminating all previous data and restarting the threshold at its initial value.

The sequential probability ratio test³ is based on the probability ratio that is a measure of the relative likelihood of each hypothesis under the observations. The test statistic is the cumulative sum of materials balances C(N) and the test thresholds are

$$TL(N) = \frac{N\mu_1}{2} + \frac{Log(B)}{\mu_1}$$
$$TU(N) = \frac{N\mu_1}{2} + \frac{Log(A)}{\mu_1}$$

where A and B determine the false-alarm and detection probabilities. The test procedure is to accept H_0 when $C(N) \ge TU(N)$, accept H_1 when $C(N) \le TL(N)$, and continue testing otherwise. Note that this test requires the value of μ_1 to be specified, unlike the Page's test where the hypothesis H_1 only specified that $\mu > 0$.

Test performances against both uniform and abrupt losses are evaluated with simulated materials balance data. Materials balances are assumed to be distributed as N(0,1) under H₀ and as N(μ ,1), μ > 0 under H₁. Each balance is assumed uncorrelated with all other balances. For Page's test the parameter values are h = 5.0 and δ = 0.5, for the Modified Page's test they are A² = 2.9 and δ = 0, and for the SPRT they are A = 2.94, B = -2.94, and μ_1 = 0.38. These parameter values were chosen by simulation to attain a 0.05 false-alarm rate per year when balances are drawn weekly. For a uniform loss rate of 0.5 per balance, Figures 1 and 2 compare the runlength distribution of the Modified Page's test with Page's test and the SPRT, respectively. These results imply that the average run length to detect a uniform materials loss is significantly smaller for the Modified Page's test compared with the other tests.

Test performance in detecting an abrupt materials loss is compared by simulating abrupt diversions in the last balance period of balance sequences of length 10 and 25. Detection probabilities given in Tables I and II show the Modified Page's test to be the best for runs of length 10 balance periods and Pages's test to be the best over 25 balance periods.

The Modified Page's test has been applied to materials accounting data generated with the model process diagrammed in Figure 3. Process measurement errors and their uncertainties are given in Tables III and IV, where correlated errors have been set to zero to achieve independent materials balances. The operation of the Modified Page's test on a representative materials balance sequence from the model process appears in Figure 4. These data represent normal operation during balances 1-50 and a uniform loss of 0.8 per balance during balances 51-100. Note that each time S(N) = 0 the threshold is returned to its initial value. Detection of materials loss is in balance 99 when S(N) crosses the upper threshold.

REFERENCES

- 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. I and II (September 1977).
- 2. J. P. Shipley, "Decision Analysis for Nuclear Safeguards," in <u>Nuclear Safeguards Analysis, Nondestructive and Analytical</u> <u>Chemical Techniques</u>, E. A. Hakkila, Ed. (American Chemical Society, Washington, DC, 1978), pp. 34-64.

- 3. A. Wald, <u>Sequential Analysis</u> (John Wiley and Sons, Inc., New York, 1947).
- E. Page, "Continuous Inspection Schemes," Biometrika <u>41</u>, 100 (1954).
- 5. H. Robbins and D. Siegmund, "Boundary Crossing Probabilities For the Wiener Process and Sample Sums," Annals of Math. Stat. <u>41</u>, 1410-1429 (1970).
- H. Robbins, "Statistical Methods Related to the Law of the Iterated Logarithm," Annals of Math. Stat. <u>41</u>, 1397-1409 (1970).
- 7. S. Karlin, <u>A First Course in Stochastic Process</u> (Academic Press, New York, 1966).


RELATIVE FREQUENCY

— 172 —

FIGURE 2



- 173 -



BLOCK DIAGRAM OF THE PU-SEPARATION PROCESS OF A REPROCESSING FACILITY USED AS BASIS FOR THE NUMERICAL MODEL

.

- 174 ---

.



FIGURE 4

PAGES STATISTIC

- 175 -

TABLE I

DETECTION PROBABILITY FOR ABRUPT MATERIAL LOSS* (10 BALANCES)

MATERIAL AMOUNT	TEST					
	MODIFIED	PAGES	SPRT			
2.0	. 16	.02	.01			
4.0	.65	.20	.05			
6.0	.91	.78	.31			

*ABRUPT DIVERSION IN LAST BALANCE PERIOD

Los Alamos

TABLE II DETECTION PROBABILITY FOR ABRUPT MATERIAL LOSS" (25 BALANCES)

	TEST				
MATERIAL	MODIFIED PAGES	PAGES	SPRT		
2.0	.11	.02	.01		
4.0	.45	.20	.05		
6.0	.70	.78	.26		

*MATERIAL LOSS IN LAST BALANCE PERIOD

TABLE III

	PLUTONIUM/ INVENTORY (KG)	RELATIV RANDOM	E STD. DEV.
HEADEND	196.5	0.01	0.0
1 ^{*†} Pu CYCLE	7.6	0.01	0.0
2 nd Pu CYCLE	50.	0.005	0.0
3 rd Pu CYCLE	134.	0.005	0.0
Pu-CONCENTRATION	62.5	0.005	0.0
Теленалар, должно и на	$\Sigma = 450.6$		

PLUTONIUM INVENTORY OF MODEL FACILITY AND CORRESPONDING MEASUREMENT UNCERTAINTIES

- 178 --

TABLE IV

	PLUTONIUM/	RELATIVE STD. DEV			
	BATCH (KG)	RANDOM	SYSTEMATIC		
INPUT	16.73	0.01	0.0		
PRODUCT OUTPUT	25.	0.002	0.0		
WASTE OUTPUT	0.2	0.25	0.0		

PLUTONIUM CONTENT OF INPUT AND OUTPUT BATCHES AND CORRESPONDING MEASUREMENT UNCERTAINTIES, THREE INPUT BATCHES, TWO PRODUCT OUTPUT BATCHES AND ONE WASTE OUTPUT BATCH PER DAY ARE ASSUMED AND A PLANT OPERATION OF 200 DAYS PER YEAR



Los Alamos National Laboratory Los Alamos, New Mexico 87545 DATE: August 26, 1982 IN REPLY REFER TO: Q-4/82-417 MAIL STOP: E541 TELEPHONE: (505) 667-7777 (FTS) 843-7777

Energy Division Safeguards Systems Group, Q-4

Mr. Dipak Gupta Kernforschungszentrum Karlsruhe GmbH Postfach 3640 D 7500 Karlsruhe 1, Federal Republic of Germany 7826484A KFK D

Dear Gupta:

I am enclosing some test results for use in the study being done by the International Workshop on Near-Real-Time Accounting. The tests are the modified Pages test and the sequential probability ratio test with decision thresholds adjusted for a .05 false alarm rate. They were applied to uniform loss scenarios under the conditions described in the tables.

Best wishes,

Joh Markin Jack Markin

JM:ew

Enc.: a/s

cy: CRMO, MS A150 (2) file $\sigma_{\epsilon} = 0.1$; $\sigma_{\delta}/\sigma_{\epsilon} = 2.5$

М	oI	m	Detection Probability	
15	1 11 11 21 21	5 10 5 10 5 10	1.0 .94 1.0 .91 .96 .80	
25	1 11 11 21 21	5 10 5 10 5 10	·· 1.0 1.0 1.0 1.0 1.0 1.0 1.0	

MODIFIED PAGES TEST

$$\sigma_{\varepsilon} = .55$$
; $\sigma_{\varepsilon}/\sigma_{\delta} = 2.5$

М	Io	m	Detection Probability	
15		5	.08	
	1	10	.05	
	11	5	.08	
	11	10	. 05	
	21	5	.08	
	21	10	.05	
25	1	5	.16	
	1	10	.11	
	11	5	.12	
	11	10	.10	
	21	5	. 11	
	21	10	.10	
М	total amount	diverted		
Io	initial bala	nce for mater:	ial loss	
m	number of ba.	lances in which	ch loss occurs	
σ _ε	standard dev:	iation of net	transfer random error	

 σ_δ standard deviation of net transfer systematic error

.

Modified Page Test Results

Sigma <u>Epsilon</u>	Sigma Epsilon Sigma Delta	Uniform Loss			Abrupt Loss <u>lst Balance</u>		Last Balance			
		<u>5</u>	<u>15</u>	25	<u>5</u>	<u>15</u>	25	<u>5</u>	<u>15</u>	25
0.1	1.5	0.05	0.25	0.79	0.83	1.0	1.0	0.25	0.74	1.0
	4.5	0.05	0.20	0.48	0.75	1.0	1.0	0.20	0.70	1.0
	9.0	0.05	0.11	0.20	0.08	0.92	1.0	0.08	0.46	0.69
1.0	1.5	0.5	0.08	0.11	0.06	0.10	0.92	0.05	0.08	0.46
	4.5	0.05	0.05	0.07	0.05	0.06	0.07	0.05	0.05	0.07
	9.0	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
10.0	1.5	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
	4.5	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
	9.0	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05

SEQUENTIAL PROBABILITY RATIO TEST

 $\sigma_{\varepsilon} = 0.1$; $\sigma_{\varepsilon}/\sigma_{\delta} = 2.5$

М	Io	m	Detection Probability	
15	1	5	.92	
	1	10	.27	
	11	10	。93	
	11	10	.27	
	21	5	.92	
	21	10	.29	
25	1	5	1.0	
	1	10	.95	
	11	5	1.0	
	11	10	.97	
	21	5	1.0	
	21	10	.96	

SEQUENTIAL PROBABILITY RATIO TEST

 $\sigma_{\epsilon} = .55$; $\sigma_{\epsilon}/\sigma_{\delta} = 2.5$

М	Io	m	Detection Probability			
15		5	.36			
	1	10	.17			
	11	5	. 42			
	11	10	.18			
	21	5	.42			
	21	10	.17			
25	1 .	5	.83			
	1	10	. 40			
	11	5	.85			
	11	10	.41			
	21	5	.85			
	21	10	.41			
M I _O	total amount diverted initial balance for material loss					

 $\sigma_{\rm E}$ standard deviation of net transfer random error σ_{δ} standard deviation of net transfer systematic error

Sequential Probability Test Results

Sigma Epsilon	Sigma Epsilon Sigma Delta		Unifo	orm Los	S	Abrupt 1st Ba	Loss lance	Last	Balan	ce
		5	15	25	5	<u>15</u>	25	<u>5</u>	<u>15</u>	25
0.1	1.5	0.07	0.15	0.21	0.71	1.0	1.0	0.62	1.0	1.0
	4.5	0.05	0.12	0.18	0.64	1.0	1.0	0.56	1.0	1.0
	9.0	0.05	0.10	0.15	0.30	1.0	1.0	0.27	1.0	1.0
1.0	1.5	0.5	0.07	0.09	0.24	1.0	1.0	0.20	0.98	1.0
	4.5	0.05	0.05	0.07	0.09	0.60	0.99	0.05	0.49	0.91
	9.0	0.05	0.05	0.05	0.05	0.10	0.42	0.05	0.06	0.30
10.0	1.5	0.05	0.05	0.05	0.05	0.06	0.07	0.05	0.05	0.05
	4.5	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
	9.0	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05

· 、

Determination of the Detection Probability as a Function of Various Loss Strategies--Phase 2 Part 1: Design of Study

> John L. Jaech Exxon Nuclear Co. Inc. Bellevue, WA 98009

April, 1982

Introduction

Phase 2 of the subject study extends the ranges of the model parameters, introduces additional test statistics, and changes the loss patterns from the initial study phase. This report presents the study to be performed at Exxon Nuclear Company on Phase 2. The Mathematical model and notation are contained in my earlier report, same title exclusive of "Phase 2". The notation will be extended as necessary.

Loss Pattern

The loss patterns to be studied in Phase 2 are as follows. To correspond to computer notation, L_i is replaced by L(I) in the description of the loss patterns.

Loss Pattern 1:

 $L(I) = \frac{M}{m} \text{ for } I = I_0, I_0 + 1, \dots I_0 + m - 1$ = 0 elsewhere

The quantities M, m and I_0 are model parameters to be varied in the study. The loss pattern is a uniform pattern beginning at period I_0 and continuing through m periods.

Loss Pattern 2:

For even m,
$$L(I) = \frac{0.5M}{m}$$
 for $I = I_0$, I_0+2 , I_0+4 , ..., I_0+m-2
$$= \frac{1.5M}{m}$$
 for $I = I_0+1$, I_0+3 , I_0+5 , ..., I_0+m-1
$$= 0$$
 elsewhere

For odd m,
$$L(I) = \frac{0.5M}{m}$$
 for $I = I_0$, I_0+2 , I_0+4 , ..., I_0+m-3
= $\frac{1.5M}{m}$ for $I = I_0+1$, I_0+3 , I_0+5 , ..., I_0+m-2
= $\frac{M}{m}$ for $I = I_0+m-1$
= 0 elsewhere

These are periodic loss functions for losses occurring during the m successive periods. More generally, the coefficients 0.5 and 1.5 could be replaced by any two quantities that sum to 2, including the possibility that one coefficient could be negative.

Test Statistics

Four sets of test statistics will be investigated. Calling the test statistics, T_1 , T_2 , ..., T_k these are defined for the four sets of test statistics.

<u>Set 1</u>. Cumulative MUF test applied at the end of each material balance period. $T_i = \sum_{j=1}^{i} D_j$; i=1, 2, ..., k <u>Set 2</u>. Dn statistic proposed in my 1976 INMM paper, "Can the Effects of Systematic Errors on LE-MUF be Reduced?", with n = 5.

$$T_{i} = D_{i+5} - \beta \sum_{\substack{j=1 \\ j=1}}^{i+4} D_{j}$$
$$\beta = \frac{-\sigma_{\eta}^{2} + 5\sigma_{\delta}^{2}}{2\sigma_{\eta}^{2} + 5\sigma_{\epsilon}^{2} + 25\sigma_{\delta}^{2}}$$

where

The first test is not made until the end of the sixth material balance period.

<u>Set 3.</u> The uniform diversion test statistic calculated over each set of four successive material balance periods. This test statistic reduces to the following form, as is shown in the appendix.

$$T_i = a_1(D_i + D_{i+3}) + a_2(D_{i+1} + D_{i+2})$$

where

$$a_{1} = \frac{2\sigma_{\eta}^{2} + \sigma_{\varepsilon}^{2}}{2(5\sigma_{\eta}^{2} + 2\sigma_{\varepsilon}^{2})}$$

$$a_{2} = \frac{3 \sigma_{n}^{2} + \sigma_{\varepsilon}^{2}}{2(5\sigma_{n}^{2} + 2\sigma_{\varepsilon}^{2})}$$

Note that a_1 and a_2 sum to one-half.

Set 4. The simple MUF test

 $T_i = D_i$ for all i.

Experimental Design

A fractional factorial 3⁵ design will be utilized for each combination of loss patterns and test sequences. The basic 3⁵ fractional design involves 41 runs. An additional 9 runs will be made at other selected points in the design space, giving a total of 50 MULNR runs per loss pattern, test sequence combination. There being 8 such combinations, the total number of MULNR runs is 400. The total number of response surfaces is eight. (In Phase 1, 150 MULNR runs were made.)

The five factors to be varied are the following. (As in Phase 1, σ_η is fixed at 1 unit so that all values involving quantities of material are relative to σ).

		<u>Values in Test Design</u>
Factor 1:	σ _ε	0.1, 1, 10
Factor 2:	σδ/σε	1.5, 4.5, 7.5 in basic design
		3, 6, 9 in additional runs
Factor 3:	Μ	5, 15, 25 in basic design
		0, 10, 20 in additional runs
Factor 4:	Io	1, 11, 21
Factor 5:	m	5, 10, 15 in basic design
		1, 20, 35 in additional runs

The specific design matrix for a given combination of loss pattern and test statistics is in Table 1. The first 41 runs are from the basic fractional factorial design with runs 42-50 being the supplemental runs made to explore other regions in the design space.

Table 1

Design Matrix

Case or Run	σ <u>ε</u>	σδ/σε	M	I _o	<u>m</u>
1	10	7.5	15	11	10
2	10	1.5	15	11	10
3	.1	7.5	15	11	10
4	.1	1.5	15	11	10
5	1	4.5	25	21	10
6	1	4.5	25	1	10
7	1	4.5	5	21	10
8	1	4.5	5	1	10
9	1	7.5	15	11	15
10	1	7.5	15	11	5
11	1	1.5	15	11	15
12	1	1.5	15	11	5
13	10	4.5	25	11	10
14	10	4.5	5	11	10
15	.1	4,5	25	11	10
16	.1	4.5	5	11	10
17	1	4.5	15	21	15
18	1	4.5	15	21	5
19	1	4.5	15	1	15
20	1	4,5	15	1	5
21	1	7.5	25	11	10
22	1.	7.5	5	11	10
23	1	1.5	25	11	10
24	1	1.5	5	11	10
25	10	4.5	15	21	10
26	10	4.5	15	1	10
27	.1	4.5	15	21	10
28	.1	4.5	15	1	10
29	1	4.5	25	11	15
30	1	4.5	25	11	5
31	1	4.5	5	11	15
32	1	4.5	5	11	5
33	1	4.5	15	11	15
34	1	4.5	15	11	5
35	1	4.5	15	11	15
36	1	4.5	15	11	5
37	1	7.5	15	21	10
38	1	7.5	15	1	10
39	1	1.5	15	21	10
40	1	1.5	15	1	10
41	1	4.5	15	11	10
42	1	3	0	11	1
43	1	6	10	11	1
44	1	9	20	11	1
45	1	6	0	11	20
46	1	9	10	11	20
47	1	3	20	11	20
48	1	9	0	1	35
49	1	3	10	1	35
50	1	6	20	1	35

Inputs to MULNR

The basic MULNR program deck was given to the sponsor, Karlsruhe Research Centre, as part of the Phase 1 product. In this section of the report, the inputs to the program are given for the four test sequences under study in Phase 2.

Program inputs include a specification of the following quantities:

K = number of tests made
S(1,0) = 1 (at all times)
Z(I,0) for I = 1, 2, ..., K
R(I,J,0) for I = 1, 2, K-1
and J > I

In all cases, Z (I,0) is defined by

$$Z(I,0) = \frac{M(I)}{\sqrt{V(I)}} - C(S)$$

and R(I,J,O) by

$$R(I,J,0) = \frac{COV(I,J)}{V(I) \cdot V(J)}$$

The quantities C(S), M(I), V(I), and COV(I,J) are defined for the four test sequences. C(S) is defined in general by

$$\frac{1}{\sqrt{2\pi}} \int_{C(S)}^{\infty} \exp(-x^{2/2}) dx = 1 - (0.95)^{1/k}$$

and is the critical value per test to give an overall α of 0.05 assuming the tests are independent. M(I) is the mean value for the test statistic, V(I) its variance, and COV(I,J) the covariance between tests I and J. — 193 —

K = 35 C(S) = 2.975424 $M(I) = \prod_{J=1}^{I} L(J)$ $V(I) = 2 VAR(1) + I \cdot VAR(2) + I^{2} \cdot VAR(3)$ $COV(I,J) = VAR(1) + I \cdot VAR(2) + I \cdot J \cdot VAR(3)$ where VAR (1) = σ_{η}^{2} $VAR(2) = \sigma_{\varepsilon}^{2}$ $VAR(3) = \sigma_{\delta}^{2}$

Test Sequence 2

Test Sequence 1

K = 30

(The first test is applied at the close of the sixth material balance period.)

C(S) = 2.927865M(I) = L(I+5) - $\beta \sum_{I=1}^{I+4} L(J)$

where

 $\beta = \frac{-VAR(1) + 5 VAR(3)}{2 VAR(1) + 5 VAR(2) + 25 VAR(3)}$

$$V(I) = A(1+5 \beta^2) - 2 \beta(B_1+4B_2) + 4 \beta^2(2B_1+3 B_2)$$

where

A = 2 VAR(1) + VAR(2) + VAR(3) $B_1 = - VAR(1) + VAR(3)$ $B_2 = VAR(3)$

 $COV(I,I+1) = B_1 - \beta (A+B_1+8B_2) + \beta^2(8B_1+4A+13 B_2)$ $COV(I,I+2) = B_1 - \beta (A+2B_1+7B_2) + \beta^2(6B_1+3A+16B_2)$ $COV(I,I+3) = B_2 - \beta (A+2B_1+7B_2) + \beta^2(4B_1+2A+19 B_2)$ $COV(I,I+4) = B_2 - \beta (A+2B_1+yB_2) + \beta^2(2B_1+A+22B_2)$

 $COV(I,I+5) = B_2 - \beta (A+B_1+8 B_2) + \beta^2 (B_1+24 B_2)$ $COV(I,I+6) = B_2 - \beta (B_1+9 B_2) + 25 B_2 \beta^2$ $COV(I,J) = B_2 - 10 B_2 \beta + 25 B_2 \beta^2 FOR J > (I+6)$

Test Sequence 3

K = 32 (The first test is applied at the close of the fourth material balance period.) C(S) = 2.947855

$$M(I) = a_1[L(I)+L(I+3)] + a_2[L(I+1)+L(I+2)]$$

where

$$a_1 = \frac{D}{2(2D-E)}$$

$$a_2 = \frac{(D-E)}{2(2D-E)}$$

and where

$$D = 2 \text{ VAR}(1) + \text{VAR}(2)$$

$$E = -\text{VAR}(1)$$

$$V(I) = \frac{(D^2 + DE - E^2)}{2(2D - E)} + \text{VAR}(3)$$

$$COV(I, I+1) = \frac{3D^3 + 2D^2E - 5DE^2 + 2E^3}{4(2D - E)^2} + \text{VAR}(3)$$

$$COV(I, I+2) = \frac{2D^3 + 2D^2E - 4DE^2 + E^3}{4(2D - E)^2} + \text{VAR}(3)$$

$$COV(I, I+3) = \frac{D^3 + 2D^2E - 2DE^2}{4(2D - E)^2} + \text{VAR}(3)$$

$$COV(I, I+4) = \frac{D^2E}{4(2D - E)^2} + \text{VAR}(3)$$

$$COV(I,J) = VAR(3)$$
 for $J \ge 5$

K	=	35
C(S)	=	2.975424
M(I)	=	L(I)
V(I)	=	2VAR(1)+VAR(2)+VAR(3)
COV(I,I+1)	=	-VAR(1)+VAR(3)
COV(I,J)	=	VAR(3) for $J \neq I+1$

Appendix

We include here a derivation of test statistic 3. $T_{\rm i}$ is defined by

$$T_i = a_1 D_i + a_2 D_{i+1} + a_3 D_{i+2} + a_4 D_{i+3}$$

Since $a_4 = 1 - a_1 - a_2 - a_3$, T_i may be rewritten

$$T_i = a_1(D_i - D_{i+3}) + a_2(D_{i+1} - D_{i+3}) + a_3(D_{i+2} - D_{i+3}) + D_{i+3}$$

The problem is to choose a_1 , a_2 and a_3 to minimize the variance of T_i . To simplify notation, let

 $D = 2 \sigma_{\eta}^{2} + \sigma_{\varepsilon}^{2} \qquad E = -\sigma_{\eta}^{2}$ then it can easily be shown that

VAR
$$T_i = 2D a_1^2 + 2D a_2^2 + 2(D-E) a_3^2$$

+ $(D+\sigma_{\delta}^2) + 2(D+E) a_1a_2 + 2(D-E) a_1a_3$
- 2 $Da_1 + 2D a_2a_3 - 2Da_2 - 2(D-E)a_3$

The partial derivations of VAR T_i are taken with respect to a_1, a_2 , and a_3 ; they are equated to zero and solved simultaneously.

0

$$\frac{\delta \text{ VAR } \text{T}_{i}}{\delta a_{1}} = 4\text{D}a_{1} + 2(\text{D}+\text{E})a_{2} + 2(\text{D}-\text{E})a_{3} - 2\text{D} = 0$$

$$\frac{\delta \text{ VAR } \text{T}_{i}}{\delta a_{2}} = 2(\text{D}+\text{E})a_{1} + 4\text{D}a_{2} + 2\text{D}a_{3} - 2\text{D} = 0$$

$$\frac{\delta \text{ VAR } \text{T}_{i}}{\delta a_{2}} = 2(\text{D}-\text{E})a_{1} + 2\text{D}a_{2} + 4(\text{d}-\text{E})a_{3} - 2(\text{D}-\text{E}) = \frac{\delta a_{3}}{\delta a_{3}}$$

The solutions for a_1 , a_2 , and a_3 are the solutions to the matrix equation

$$\begin{pmatrix} 2D & (D+E) & (D-E) \\ (D+E) & 2D & D \\ (D-E) & D & 2(D-E) \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \\ a_3 \end{pmatrix} = \begin{pmatrix} D \\ D \\ D-E \end{pmatrix}$$

After some algebra, the solutions are

$$a_1 = \frac{D}{2(2D-E)}$$
$$a_2 = a_3 = \frac{D-E}{2(2D-E)}$$

and since the a's must sum to 1,

$$a_4 = \frac{D}{2(2D-E)}$$
 which completes the derivation

John L. Jaech Exxon Nuclear Co. Inc. Bellevue, WA 98009

August, 1982

Introduction:

Phase 2 of the subject study extends the ranges of the model parameters, introduces additional test statistics, and changes the loss patterns from the initial study phase. In a previous report, which bears the same title as this one except that "Part 2: Study Results" is replaced by "Part 1: Design of Study", the details of the Phase 2 study were given. That is, the loss patterns were specified, the test statistics defined explicitly, and the design matrix was given. The initial design matrix was extended to include additional combinations of the factors as will be indicated later in this report. The reference report also provided the inputs to the MULNR computer program, including the variance of each test statistic and the covariances between all pairs.

All the computer output was sent to D. Sellinschegg at Karlsruhe on August 24, 1982 along with a cover letter that describes it. The cover letter also summarizes the information about the error parameters, losses, loss patterns, and test statistics in both phases of the study.

The purpose of this report is to present the results of the Phase 2 study. Emphasis is concentrated on the detection probability as of the end of time interval 35 as the response variable. The computer output gives the cumulative detection probability as of the end of each time interval. It would be of interest to relate the cumulative detection probabilities to the loss patterns for the various test statistics, but the limited resources did not permit such a study at this time. The study results for Phase 2 are divided into 3 sets. Set 1 refers to the Table 1 Design Matrix given in the reference document for both the uniform and up and down or periodic loss function. Set 2 refers to additional runs with the parameter space redefined because of very small detection probabilities for so many Set 1 cases. Set 3 refers to a limited number of additional runs made to permit direct comparison of our results with those reported by other participants in this study.

For Set 1, recall from the referenced document that the values for the 5 factors in the basic design matrix were as follows, using the notation of the referenced document:

σε		0.1, 1.0, 10
^σ δ ^{/σ} ε	27	1.5, 4.5, 7.5
М		5, 15, 25
Io		1, 11, 21
m	=	5, 10, 15

In addition to the 41 cases in the basic design, 9 more were run to test the adequacy of the empirical model to be fit to the results. For these additional cases, σ_{ϵ} was fixed at 1 unit while the other factors assumed one of the following values for each case:

 $\sigma_{\delta} / \sigma_{\varepsilon} = 3, 6, 9$ M = 0, 10, 20 $I_{0} = 1, 11$ m = 1, 20, 35

Table 1 below gives the detection probabilities for both the uniform and up and down loss patterns and for the four test sequences identified in the reference report. To briefly review the four test sequences (TS): TS-1: Cumulative MUF test applied at the end of each interval

- TS-2: The D_n statistic proposed in my 1976 INMM paper, "Can the Effects of Systematic Errors on LE-MUF be Reduced?", with n = 5
- TS-3: The uniform diversion test statistic calculated over each set of four successive material balance periods.

TS-4: The simple MUF test applied at the end of each interval.

The cases in Table 1 are those identified in Table 1 of the reference document and will not be repeated here. The column headed "U" refers to the uniform loss pattern and that headed "U-D" to the up and down pattern.

Tab	le	1

Detection I	Probabilities	for	Set	1	Cases
-------------	---------------	-----	-----	---	-------

	TS	-1	Т	'S-2	T	<u>S-3</u>	<u></u> TS2	<u>ا</u>
Case	U	U-D	U	<u>U-D</u>	<u><u>u</u></u>	U-D	<u>u</u>	<u>U-</u> D
1	.023	.023	.051	.051	.023	.023	.023	.023
2	.024	.024	.051	.052	.025	.025	.033	.033
3	.071	.070	.109	.171	. 255	. 257	.189	.263
4	.964	.962	. 994	. 997	1.000	1.000	.207	.317
5	.026	.025	.136	. 242	.039	.039	.045	.052
6	.049	.047	.040	.076	.042	.042	.048	.056
7	023	.023	050	.052	.025	.025	.029	.029
8	026	026	043	044	025	.025	.029	.029
9	.024	.024	.055	.066	.026	.026	.027	.028
10	.025	.025	186	.330	.028	.028	.030	.031
11	036	036	062	076	054	.054	.086	.096
12	045	045	210	368	197	. 222	. 203	. 303
13	023	023	052	054	024	.024	.024	.024
14	023	023	050	050	024	023	024	.024
15	361	356	.000	836	999	999	573	.819
16	045	045	.000	071	088	088	063	067
17	024	.045	.007	.071	027	027	033	034
18	024	.024	188	.000	.027	036	042	049
10	.025	.025	.188	.555	.035	.030	034	035
20	.032	.032	.042	.049	.030	.030	045	.053
20	.040	.040	.039	.039	.037	.039	.045	.03/
21	.027	.027	.130	.230	.030	.030	.032	025
22	.023	.023	.047	.049	.024	170	.025	.025
23	.057	.057	, 152	.200	, 109	.170	.217	. 322
24	.020	.027	.050	.052	.032	.032	.034	.055
25	.025	.023	.053	.055	.023	.023	.024	.024
20	.024	.023	.048	.049	.023	,023	.024	.024
21	.074	.073	.219	. 319	.002	.005	.204	. 299
20	,004	. 636	.146	.230	.080	.000	.207	. 303
29	.029	.029	.076	,118	.035	.035	.041	.043
21	,032	.032	.015	.000	.000	.070	.001	. 122
21	.024	.024	.046	.047	.025	.025	.028	.029
22	.024	.024	.055	.060	.025	.025	.029	.029
22	.023	.023	.051	.051	.023	.025	.024	.024
24	.023	.023	.053	.054	.023	.023	.024	.024
30	, 105	. 104	. 149	.187	. 328	. 334	.138	.173
20	.250	.254	.645	.842	.999	1,000	.618	.820
27	.023	.023	.073	.098	.026	.026	.028	.028
20	.030	.029	.040	.051	.027	.027	.028	.029
39	.030	.030	.081	.111	.070	.070	.104	,128
40	.089	.083	.045	.060	.080	.081	.104	.128
41	.027	.027	.069	.095	.031	.031	.035	.037
42	.023	.023	.045	.045	.023	.023	.031	.031
45	.025	.025	. 990	. 990	.029	.029	.084	.084
44	.026	.026	1,000	1,000	.034	.034	.202	. 202
45	.023	.023	.045	.045	.023	.023	.025	.025
40	.023	.023	.050	.054	.024	.024	.025	.025
4/	.029	.029	.061	.077	.036	.036	.049	.052
48	.022	.022	.045	.045	.023	.023	.024	.024
49	.027	.027	.046	.048	.028	.028	.038	.038
50	.027	.027	.046	.052	.027	.027	.030	.030

The following observations are made on these Table 1 results:

- 1. For test sequences 1 and 3, it makes very little difference whether the loss pattern is uniform or up and down. Both test sequences are directed at protracted losses, and how the protracted losses occur is not too important.
- 2. Test sequence 2 detects changes in loss patterns, and hence there are factor combinations for which the detection probability is larger for the up and down loss pattern than for the uniform loss pattern.
- 3. There are also factor combinations for which the simple MUF test (TS-4) detects the up and down loss pattern with larger probability than it does the uniform loss pattern.
- 4. Attempts were made to fix the α error probabilities at around 0.05 by assuming independence of the tests. In fact, the α -values that resulted are a function of the factor combinations. For the M = 0 values (cases 42, 45, 48), α is about 0.023 for TS-1, and TS-3, 0.045 for TS-2, and 0.025 for TS-4.
- 5. There is no one test sequence that is generally superior to the others, the ability of a given test sequence to detect losses being dependent upon the loss patterns.
- 6. Many of the detection probabilities in Table 1 are quite small. Because of this, the ranges on three of the factors were changed and a second set of cases were run. This is discussed in the next section.

Set 2 Results

In this second set of runs, only the uniform loss pattern is considered. The design matrix for the first 41 cases is the same as that given in Table 1 of the referenced document except for the following changes in the values of

 σ_{ϵ} , $\sigma_{\delta} \beta_{\epsilon}$ σ :

, and M. change 1 to 0.55 change 10 to 1

σsβε

: change 1.5 to 1 change 4.5 to 2.5 change 7.5 to 4

M: change 5 to 15 change 15 to 25 change 25 to 35

Five additional cases were run with $\sigma_{\epsilon} = 0.55$, $\sigma_{\delta} = 2.5$, M=25, m-10, and I₀ = 1, 3, 5, 7, and 9 for cases 42, 43, 44, 45, and 46 respectively.

The detection probabilities for Set 2 are given in Table 2. Two columns of probabilities are given. Column 1 gives the detection probability as calculated by MULNR, and column 2 gives the detection probability calculated by an empirical second order polynomial model fit through these data, using the same type of approach as was used in Phase 1 of the study. As in Phase 1, the response variable, y, was defined by

Detection Probability =
$$\frac{1}{\sqrt{2\pi}} \int_{0}^{y} \exp(-x^{2}/2) dx$$

The 22 parameter empirical model for a given test sequence was of the form

$$y = b_0 + \sum_{i=1}^{5} b_i x_i + \sum_{i=1}^{5} b_{ii} x_i^2 + \frac{4}{\sum_{i=1}^{5} \sum_{j>i}} b_{ij} x_i x_j$$

— 203 —

where

$$x_{1} = (\sigma_{\varepsilon} - .55)/.45$$

$$x_{2} = (\sigma_{\delta}/\sigma_{\varepsilon} - 2.5)/1.5$$

$$x_{3} = (M-25)/10$$

$$x_{4} = (I_{0}-11)/10$$

$$x_{5} = (m-10)/5$$

As is noted from Table 2, the second order polymomial empirical model does not provide a very close fit. In this phase of the study, no further attempts were made to provide a closer fit due to resource limitations. Such an effort, possibly incorporating results from test statistics investigated by other study participants as well, is deferred for the present.

Detection Probabilities for Set 2 Cases

Case	T	S-1	T:	S-2	TS-3		Т	S-4
	MULNR	EMPIRICAL	MULNR	EMPIRICAL	MULNR	EMPIRICAL	MULNR	EMPIRICAL
1	.031	.108	.133	. 336	.044	.012	.053	.019
2	.089	.030	.185	.092	. 354	.745	.321	.353
3	.463	.714	.748	. 941	1.000	1.000	.584	.624
4	1.000	1.000	1.000	1.000	1.000	1.000	.621	.829
5	.052	.050	.400	.655	. 396	.580	.494	.512
6	.436	.689	.051	.156	.462	.630	.494	.516
7	.031	.009	.085	.038	.080	.068	.120	.080
8	.103	.143	.043	.016	.095	.075	.120	.082
9	.038	.015	.087	.059	.061	.211	.090	.139
10	.050	.040	.801	.621	. 289	.198	. 350	.453
11	.157	.187	.220	. 247	.538	.490	.287	. 207
12	.448	.609	.977	. 960	1.000	1.000	.986	.974
13	.049	.006	.278	.140	.126	.108	.173	.141
14	.031	.116	.072	.154	.043	,022	.060	.069
15	1,000	1,000	1.000	1.000	1,000	1.000	.968	.965
16	.435	.662	.750	.903	. 994	.997	. 207	.258
17	.034	.027	.096	.030	.099	.127	.173	. 140
18	.046	.026	.829	.928	,710	.766	. 700	. 784
19	.128	.153	.048	.057	.125	.147	.174	.143
20	.736	.719	.037	.269	.785	.806	.699	.787
21	.054	.046	.374	. 338	.161	,086	.226	. 233
22	.033	.009	.076	.036	.048	.124	.068	.060
23	.457	.665	.740	.845	1.000	1.000	.858	. 853
24	.102	.122	.153	.178	.413	.357	.188	.159
25	.029	.101	.141	.171	.070	.059	.100	.088
26	.091	.041	.042	.022	.083	.076	.104	.088
27	.539	.739	1.000	.998	1,000	1.000	.601	.691
28	1.000	1.000	.999	. 986	1.000	1.000	.616	.699
29	.072	.090	.164	.113	.194	.205	. 295	.243
30	.144	.341	. 998	.992	. 966	.976	.961	.977
31	.038	.010	.062	.084	.059	.049	.096	.095
32	.045	.046	.272	,234	,255	.260	.256	. 395
33	.036	.063	.080	.115	.053	.020	.077	.232
34	.045	.019	.631	.528	.212	.372	.259	.227
35	.793	.889	.933	.975	1.000	1.000	. 313	, 225
36	1,000	1.000	1.000	1.000	1,000	1.000	1.000	. 999
37	.031	.021	.171	.152	.086	.109	.125	.111
38	.110	.102	.040	.020	,103	. 148	.129	. 115
39	.113	.090	. 388	.498	.935	.945	.498	.481
40	.907	.907	. 206	. 222	. 923	.944	. 503	.484
41	.062	.062	.184	.184	. 206	. 206	.267	. 267
42	.201	. 249	.046	.089	. 202	.220	.238	.239
43	.119	.198	.059	.104	.188	.217	. 238	. 249
44	.093	.154	.105	, 121	.183	. 214	.238	.257
45	.077	.117	.153	.140	.180	.212	. 238	.262
46	.067	.086	.153	.161	.178	. 209	. 238	.266

Keeping in mind the limitations of the empirical fits, some computer plots were made of detection probability versus various parameters. Twelve such plots are included in this report primarily to illustrate the motivation behind the attempts at empirical model building. If future plans call for refining the models, such plots and variations thereof would be the final output for the various test sequences. They would be very useful in comparing the various test sequences in different regions of the factors space.

Figures 1-4 give detection probability versus I_0 for fixed σ_{ϵ} , $\sigma_{\delta}/\sigma_{\epsilon}$, and m; for 3 values of m; and for test sequences 1-4 respectively. Figures 5-8 give detection probability versus σ_{ϵ} for fixed M, I, and m; for 3 values of $\sigma_{\delta}/\sigma_{\epsilon}$; and for test sequences 1-4 **respectively**. Figures 9-12 give detection probability versus M for fixed σ_{ϵ} , I, and m; for 3 values of $\sigma_{\delta}/\sigma_{\epsilon}$; and for test sequences 1-4 respectively.

Some unusual behavior in these curves is due to model inadequacy. For example, in Figure 5, it appears that detection probability begins to increase for $\sigma_{\delta}/\sigma_{\epsilon} = 4$ with increasing σ_{ϵ} after a certain point. This, of course, is not correct; detection probability would continue to decrease. Set 3 Results

In discussions with D. Gupta and C.A. Bennett on July 27, 1982, a third set of factor combinations was defined in order to compare the test sequences considered here and by other study participants at common sets of input values. The cases consisted of all 24 combinations of the following, with $\sigma_{\delta}/\sigma_{\epsilon}$ fixed at 2.5.

 $\sigma_{\varepsilon} = 0.1, 0.55$ M = 15, 25 $I_{o} = 1, 11, 21$ m = 5, 10

In addition, for $I_0 = 11$, m = 5, and $\sigma_{\epsilon} = 0.1$ and 0.55, two cases were run at M = 0 to determine the α values. The case number identification is given

in Table 3. Both the uniform and up and down loss patterns were studied here, but since only the uniform loss pattern was studied by the other participants, the Set 3 results are restricted to this loss pattern.

Table 3 Set 3 Cases

.

Case	σε	M	Io	m	Case	σε	M	<u>I</u> o	m
1	.1	15	1	5	14	.55	15	1	10
2	.1	15	. 1	10	15	.55	15	11	5
3	.1	15	11	5	16	.55	15	11	10
4	.1	15	11	10	17	.55	15	21	5
5	.1	15	21	5	18	.55	15	21	10
6	.1	15	21	10	. 19	.55	25	1	5
7	.1	25	1	5	20	.55	25	1	10
8	.1	25	1	10	21	.55	25	11	5
9	.1	25	11	5	22	.55	25	11	10
10	.1	25	11	10	23	.55	25	21	5
11	.1	25	21	5	24	.55	25	21	10
12	.1	25	21	10	25	.1	0	11	5
13	.55	15	1	5	26	.55	0	11	5

A comparison of the results from the tests is given in Tabale 4. The Table 4 results are for the uniform loss pattern. The ll test sequences in Table 4 are identified as follows:

TS-1 to TS-4:	Described in the <u>Set 1 Results</u> Section of this report
TS-5 :	Truncated sequential CUMUF test (Beedgen)
TS-6:	CUMUFR test; two-sided sequential test with power one
	(Sellinschegg)
TS-7:	CUMUF; sequentially performed fixed-length test (Sellinschegg)
TS-8:	CUMUF(35); fixed length test at the end of 35 periods
	(Sellinschegg)
TS-9:	Same as TS-6 but one-sided (Sellinschegg)
TS-10:	Sequential Probability Ratio Test (Markin)
TS-11:	Modified Pages Test (Markin)

Tab	le	4
		•

Detection Probabilities for Table 3 Cases

Case	<u>TS-1</u>	TS-2	TS-3	TS-4	TS-5	TS-6	<u>TS-7</u>	TS-8	TS-9	<u>TS-10</u>	<u>TS-11</u>
1	1.000	. 340	1.000	. 687	1	1.0	1.0	. 52	1.0	. 92	1.0
2	.995	.695	.991	.211	1	1.0	1.0	.52	1.0	. 27	.94
3	.750	.993	1.000	,682	.978	1.0	. 9	.52	1.0	.93	1.0
4	.435	.750	.994	. 207	.811	1.0	.72	.52	1.0	. 27	.91
5	. 299	.993	1.000	,678	.53	1.0	. 57	.52	1.0	. 92	.96
6	.192	, 745	.995	. 204	.355	1.0	.48	.52	1.0	.29	.80
7	1.000	.894	1.000	1.000	1	1.0	1.0	.88	1.0	1.0	1.0
8	1,000	. 999	1.000	.616	1	1.0	1.0	.88	1.0	. 95	1.0
9	1.000	1.000	1.000	1.000	1	1.0	1.0	.88	1.0	1.0	1.0
10	.985	1.000	1.000	.608	1	1.0	. 99	.88	1.0	.97	1.0
11	.780	1.000	1.000	1.000	.989	1.0	.96	. 88	1.0	1.0	1.0
12	. 539	1.000	1.000	.601	.909	1.0	.89	. 88	1.0	.96	1.0
13-	.301	.038	. 296	.257	.583	1.0	.27	.09	.23	. 36	.08
14	.103	.043	.094	.120	. 20	. 99	.07	.09	.10	.17	.05
15	.048	.272	.255	.256	.06	. 98	.05	.09	. 98	.42	.08
16	.042	.082	.085	.120	.052	.87	.05	.09	.92	.18	.05
17	.033	.270	.246	,256	.052	. 96	.05	.09	. 97	.42	.08
18	.031	.085	.080	.120	.051	. 90	.05	.09	.93	.17	.05
19	.736	.037	.785	.699	.763	1.0	.78	.13	.72	.83	.16
20	.236	.046	.239	.267	.494	1.0	.22	.13	.26	.40	.11
21	.084	.829	.706	.700	.209	1.0	.09	.13	1.0	.85	.12
22	.063	.184	. 206	.267	.088	1.0	.08	.13	1.0	.41	.10
23	.046	.829	.710	.700	.064	1.0	.08	.13	1.0	.85	.11
24	.039	.185	.198	.266	.055	1.0	.08	.13	1.0	.41	.10
25	.027	.045	.034	.041	.05	.051	.035	.051	.051	.05	.05
26	.023	.043	.024	.044	.05	.050	.027	.050	.051	.05	.05
Note that cases 25 and 26 give the actual α - values. When comparing detection probabilities, these differences in the α - values should be kept in mind. Ideally, they should all be about 0.05 for a fair comparison, but it is difficult in some instances to fix α precisely in advance.

Note that test sequences 1 and 7 should give identical results, with the TS-1 results calculated by the multivariate normal distribution and the TS-7 results by simulation. Taking into account the differences in the α values, the agreement is good.

The results of Table 4 are plotted in a series of figures, 13-23. These results were plotted as straight line segments rather than as smoothed curves to emphasize that what is plotted are the first 24 case results of Table 4 with no model building involved. The straight-line interpolation between $I_0 = 1$ and $I_0 = 11$ would be especially misleading for some of the test sequences if one were to focus on I_0 values between $I_0 = 1$ and $I_0 = 11$ rather than on the two point results. For test sequences 1-4, and for $\sigma_{\varepsilon} = .55$, M = 25, m = 10, the detection probabilities for $I_0 = 1,3,5,7$, and 9 are given as cases 42-46 in Table 2, and are plotted as open circles in Figures 13-16.

Preliminary Evaluation of Near-Real-Time Materials Accountancy Models in A Large-scale Reprocessing Plant

Koji Ikawa

Japan Atomic Energy Research Institute

Submitted to:

Dr. D. Gupta who is acting as a rapporteur of the paper on the status of development of the n.r.t. accountancy system to present it to the International Symposium on Recent Advances in Nuclear Material Safeguards to be held in Vienna from 8 to 12 November 1982.

Preliminary Evaluation of Near-Real-Time Materials Accountancy Models in A Large-scale Reprocessing Plant

K. Ikawa Japan Atomic Energy Research Institute

ABSTRACT

The capability of near-real-time materials accountancy models as applied to a large-scale reprocessing plant has been evaluated using the computer simulation technology. In this study three different material balance periods, i.e., 8 hours, 2 days, and 1 week, were assumed and two structures of material balance areas were considered for comparison purposes. Assumed modes of diversion are abrupt diversion of 8 kgs Pu per 2 weeks, protracted diversion of the diversion rates of 52 kgs, 32 24 kgs, 16 kgs and 8 kgs plutonium per year. Simulation kgs, calculations covered four months of plant operation. The preliminary result shows that abrupt diversions of 8 kgs Pu / 2 weeks can be detected in every cases before such diversions are completed. On the other hand, detection capabilities for protracted diversions varies from case to case . The results suggested that more long-term simulation calculations are neccessary to obtain reliable conclusions. It also suggested that if the present chemical process line is divided into two parallel lines of a half process capacity, the detection capability for protracted diversion might be significantly improved. These problems will be investigated in the next step of this study.

1. Introduction

A study of the feasibility of applying the concept of near-realtime materials accountancy (n.r.t. accountancy) to large scale spent fuel reprocessing facilities has been investigated using the Allied-General Nuclear Services fuel reprocessing plant at Barnwell (BNFP) as the reference facility. This study has been carried out basing on the experience obtained by the past study of the n.r.t. accountancy system using the existing Tokai Reprocessing Plant under the TASTEX (Tokai Advanced Safeguards Technology Exercise) programme. Although a similar study had been performed at the Los Alamos National Laboratory (8), this study has been carried out for the following two major purposes:

- (i) to investigate the feasibility of applying the ten-daydetection-time model, which was developed for a medium sized reprocessing plant under the TASTEX programme, to a large-scale reprocessing facility, and
- (ii) to investigate the practical detection goals such as detection goal quantities as a function of significant quantities, detection times and probabilities related to detection capability and false alarm rates.

Now this study is on an early stage and have given us a very limited result which can not permit us to get clearcut conclusions for the problems above mentioned. Therefore this paper has a characteristics to describe a preliminary result of our investigation.

2. Model Plant

In this study the Allied-General Nuclear Services fuel reprocessing plant at Barnwell (BNFP) was used as the reference facility. The BNFP is designed to process spent fuel at rates up to 5 ton (heavy metal) / day. The facility uses conventional PUREX recovery process which is designed to process 1500 ton heavy metal per year of nuclear fuel and to recover 15 ton / year of plutonium as the nitrate solution. The process flow through the plant and basic process data were referred to Ref (8).

3. Operator's Materials Accountancy System

3.1. Conventional Materials Accounting System

The conventional materials accountancy is assumed to be based on the periodic clean-out physical inventory takings and measurements of all input into and output from the material balance area. In this study seven material balance areas (MBA) with sixteen key measurement points (KMP) as illustrated in Fig. 1 were assumed for the conventional materials accountancy. Nuclear materials and types of measurements at flow and inventory key measurement points are, respectively, shown in Tables 1 and 2 with the associated measurement accuracies. Although the model facility is divided into seven MBA's as shown in Fig. 1, MBA 6 and MBA 7 can be treated as a single other facility which is independent from the reprocessing facility, because these are waste storage areas of temporary use.

Among these seven MBA's, only MBA 3 is a pure MUF-MBA and all the others are S/RD MBA's or pure storage areas. Characteristics and activities of these MBA's are as follows :

(A) MBA 1 : Spent Fuel Receiving and Storage

Since MBA 1 includes only the cask-unloading and spent fuel storage pond, this is a pure storage area, i.e., non-MUF and non-S/RD area, and therefore the item accountancy shall be adopted to take a material balance in this area. However, when NDA methods for verifying declared burnup and cooling time and for assaying fissile content of spent fuel with sufficiently high accuracies were developed, characteristics of materials accountancy in MBA 1 may become different ones. If such NDA methods became available for the use at KMP 2, shipper / receiver differences could be determined within MBA In this case MBA 1 shall be treated as an S/RD area, and 1. the MBA structure could be modified by making the chop and leach area (present MBA 2) and the chemical separation area (present MBA 3) into a single process MBA.

(B) MBA 2 : Chop and Leach

A chop and leach area is in usual case included either in the front area, i.e., MBA 1 in this study, or in the succeeding

area, i.e. the chemical separation process area. In this study, however, this is treated as an independent material balance area, where the shipper/receiver difference can be determined by applying the gravimetric method. Pu/U ratio is measured at the dissolver tank for this purpose. Therefore, MBA 2 is a pure S/RD MBA. If MBA 1 is used as a S/RD-MBA as previousely described, however, MBA 2 can be included in the succeeding chemical separation process area or could be left order to make the reliability of S/RD it is in as determination higher by the duplicate determinations of an S/RD.

(C) MBA 3 : Chemical Separation Process

MBA 3 is a pure MUF-MBA, which includes the solvent extraction processes from the input accountability tank to the product tanks. Plutonium nitrate product solution is transferred to the plutonium product storage area, MBA 4, through KMP 7 by batches (\sim 400 L, \sim 250 gPu/L). Uranium nitrate product batches (\sim 4460 L, \sim 380 gU/L) are transferred to the uranium product storage area through KMP 9.

(D) MBA 4 : Plutonium Nitrate Storage

This MBA is assumed to be an S/RD MBA in this study. The input measurement is made at the boundary between MBA3 and MBA 4, i.e., KMP 7, which produces shipper's data for MBA4. The shippment of the plutonium product is made through KMP 13, where the accountability measurement is expected to be carried If a plutonium-nitrate-to-oxide conversion plant is out. located at the next door of this reprocessing plant, however, the measurement for accountability purpose at KMP 13 could be replaced by the input accountability measurement at the entrance of the conversion plant. When the accountability measurement at KMP 13 is eliminated, suitable containment/surveillance measures should be provided in MBA 4 to assure the integrity of the material accountancy data in this area.

- (E) MBA 5 : Uranium Nitrate Storage This MBA has similar characteristics with MBA 4.
- (F) MBA 6 and MBA 7 : Waste Storage These MBA's are used to store waste from MBA 2 and MBA 3. The accountability measurements of waste are made at KMP's 5, 11 and 12. After the quantities of plutonium and uranium are determined, waste are sealed drums and stored. Item accountancy can be made by checking seals provided and summing up the data recorded. Therfore this MBA is a special storage area in which the containment and survaillance is used as a principal safeguards measures.

3.2. Near-Real-Time Materials Accountancy in The Plutonium Purification Process

In this study a near-real-time materials accountancy system is assumed in the plutonium purification process of the chemical separation process, MBA 3, in order to evaluate its effectiveness from the view point of the materials accountancy for safeguards. At an early stage of this study, it was considered that the area to be coverred by the n.r.t. accountancy should be taken as the whole area of MBA 3 in order to get generic conclusions on the effectiveness of n.r.t. accountancy. Α simulation programme for such a purpose was developed and tested. The result of test calculations indicated that a long-term simulation calculation of the whole material balance area requires very much computer time. Since the budget of this study was restricted, the time and space covered by simulation programme had to be limited in shorter and smaller ones, i.e., four months and the plutonium purification process instead of the whole processes.

Main conditions of the n.r.t. accountancy are as follows :

(A) Sub-MBA structures

Two set of sub-MBA structures for the n.r.t. accountancy system were considered. These are shown in Fig. 2 by names of sub-MBA model. These sub-MBA's require additional measurement points to get near-real-time material balances in these areas. Those points are listed in Tables 1 and 2. (B) Material Balance Periods

The following three cases were assumed for the comparison purpose.

- case 1 : 8 hrs
 case 2 : 2 days (48 hrs)
 case 3 : 1 week (168 hrs), which corresponds to 'the
 ten-day-detection-time model' developed in TASTEX
 project.
- (C) Frequencies of Calibration of Analysis and Flow Meters Two cases were assumed as follows : case 1 : every 24 hrs case 2 : every week
- (D) Measurement Accuracies of n.r.t. accounting In this study only one set of data of measurement accuracies were assumed and used for simulation calculations. These data are shown in Table 3.

4. Effectiveness of N.R.T. Materials Accountancy Models

4.1 Effectiveness Evaluation Method

Effectiveness of a materials accountancy system of an actual operating reprocessing plant could be evaluated, if sufficient data relating the materials accountancy system were obtained and became available for analysists to make thier evaluation analyses of effectiveness. In an actual situation, however, operating histories from an existing plant have not always been available, and therefore it is not easy to carry out such effectiveness evaluation analyses. Difficulties of this kind will become more significant if the plant is still under the design stage.

In such a case, it is very convenient to utilize the simulation technology. Simulated data are useful for evaluation of a material measurement and accountancy system. For this purpose, a dynamic mathematical model of the reference plant was developed. The model includes almost all major processes in the plant, but some parts, e.g., waste disposal processes, were excluded because of their less importance from the viewpoint of safeguards. Based on this model, a simulation system "DYSAS-R II" (<u>Dynamic Safeguard Simulation code for Reprocessing</u> facilities) was developed. Experiences obtained by performing TASTEX project had been fully utilized for this development work.

Using this simulation code, process materials flows and holdups at various operating conditions can be simulated. Operations of start-up, flush-out, clean-out and diversions can also be included. Measurement simulations can be made by a code "SIMAC" (Simulation of Measurements and Accountancy), and simulated measurement data can be analyzed by statistical and sequential-decision techniques, which were programmed by "SADAC" (Safeguards Data Analysis Code). These two codes were developed and used in TASTEX project.

4.2 Outputs of Simulation Runs

Simulations of materials flows were made only for the plutonium purification process where equilibrium conditions were assumed. Simulated plutonium holdups and plutonium concentrations of solutions in major tanks, columns and concentrators are shown in Figs. 3 and 4. These data are used in the measurement simulation code where they are treated as true values of corresponding measurement data. Material balance data are calculated by these simulated measurement data. Simulation runs of 43 cases, 18 no-diversion cases and 25 diversion cases, have been carried out. All computer outputs from simulation runs were re-treated for graphical expressions. Detailed analyses on capability of each of the n.r.t. accountancy models can easily be made by these graphic expressions. In this paper a few examples of them are shown by Figs. 5 to 7.

Simulation conditions are as follows :

N.R.T. MBA Model 2 : shown in Fig. 2

N.R.T. MB Period Fig. 5 - 8 hrs Fig. 6 - 2 days Fig. 7 - 1 week

Diversion Mode Abrupt : 8 kgPu/2 weeks (720 hr - 1056 hr)

	Material Balance No.	Material Balance No.
	of Initiation	of Cease
Fig. 5	91	132
Fig. 6	16	22
Fig. 7	5	7

Calibration Period 24 hrs

Simulation covered 2 months

The top of each figure is a material balance (Shewhart) chart. For each chart, n.r.t. material balances (MUF) are plotted sequentially with 1 *O* error bars. An alarm chart was produced for every Shewharts. However, almost all of alarm charts could indicate small number of material balance alarms in comparison with other type of alarm charts as described later. Therefore, the alarm charts associated with Shewhart were not cited in this paper.

The middle of each figure is a CUMUF (CUSUM) chart, for which cumulative summations of n.r.t. material balances are plotted sequentially with 1 6 error bars. The associated alarm chart indicates letter symbols which mean the length and significance of sequences of n.r.t. material balances that generate alarms. Since the theoretical base of decision analysis were introduced from Los Alamos National Laboratory (8,9), the definition of letter symbols are identical with those of LANL except some minor points as indicated in the following table.

Sym	001	False-Alarm Probability
$MUF \ge 0$	MUF < 0	Taise Alaim Hobability
A	1	10^{-2} 5 x 10^{-3}
В	2	5×10^{-3} 10^{-3}
С	- 3	10^{-3} 5 x 10^{-4}
D	4	5×10^{-4} 10^{-4}
Е	5	10^{-4} 10^{-5}
F	6	10^{-5} 10^{-8}
G	7	10 ⁻⁸
т	т	0.5

- 217 -

The CUMUF chart of Fig. 5 indicates a significant number of alarms. The first alarm appears at the 104 material balance number with symbol 'A' and symbol 'E' appears at 126 material balance number.

The bottom of each figure is a Kalman-filter estimates of the average amounts of missing material per balance period which are plotted sequentially with 16 error bars.

The Kalman-filter estimates of Fig. 5 shows that it has a capability to detect abrupt diversion assumed in this simulation, although it is, in principle, suitable to detect protracted diversions.

The results of simulation runs mentioned as examples show that the n.r.t. accountancy models of material balance periods less than two days can detect an abrupt diversion of 8 kgPu/2 weeks, and that of material balance period of one week may detect such a diversion.

4.3 Characteristics of the N.R.T. Accountancy Models

Characteristics of the n.r.t. accountancy models are summarized in Table 4. These are obtained by four months simulation calculations under the normal equilibrium operations. Figures of CUSUM and are calculated for four months plant equilibrium operation which does not include any start-up and clean-out operations.

From this table, any significant difference is not seen between the two n.r.t. sub-MBA models. When graphical simulation outputs were carefully analysed, it became clear that the appearance of false-alarms increases if the calibration period was taken to be less than the n.r.t. material balance period. To avoid this problem, twenty-four hours-calibration period is used as a fixed value in the succeeding diversion sensitivity analyses.

4.4. Results of Diversion Sensitivity Analyses

The srsults of simulation runs for diversion sensitivity analyses are summarized in Table 5. In this study, the term 'diversion' is defined as 'to remove nuclear material from the process line without any declaration'. Such a removal is assumed at the point of flow from 3P concentrator (3PCP), where the most concentrated plutonium nitrate solution in the plutonium purification process is produced. Diversion modes assumed are as follows:

	Abrupt Diversion	Protracted Diversion
Diversion Time	from 720 hr to 1056 hr	from 0 hr to 2880 hr
Diversion Rates	8 kgs/2 weeks	8 kgsPu/year 16 kgsPu/year 24 kgsPu/year 32 kgsPu/year 52 kgsPu/year

Essences of Table 5 are as follows :

(A) Capability to Detect Abrupt Diversions

n.r.t. MBP	Time required for detection	Total amount diverted before detection
8 hrs	136 hrs = 5.7 days	3.2 kgPu
2 days	192 hrs = 8 days	4.6 kgPu
1 week	288 hrs = 12 days	6.9 kgPu

- (1) In all cases simulated, the abrupt diversion can be detected before the total plutonium amount diverted does not exceed 8 kgs.
- (2) So long as the results of simulations up to now are evaluated, the following is suggested; A n.r.t. accountancy model of a single n.r.t. MBA for the whole plutonium purification process with a weekly in-process inventory ,i.e., weekly material balance, may meet IAEA provisional criteria for detecting the abrupt diversion of 8 kg of plutonium.
- (3) However, if the weekly material balance is adopted, there may be possibility that an order of 7 kg of plutonium could be diverted without detection, because the number of alarms may be so small and

their level of significance may be so low that an inspector may not decide that the diversion has occured. An example of this case is shown in Fig. 7.

(4) If the chemical separation process is divided into two separate parallel lines, however, longer (than 7 days) material balance period may be sufficient to counteract the abrupt diversion strategy.

(B) Capability to detect protracted diversions

- In case of the 8 hrs-material balance period, the diversion of the minimum diversion rate, i.e., 8 kgPu per year, could be detected at 3.5 months after the diversion was initiated. The total plutonium amount diverted did not exceed 2.3 kgs when detected (Case P-19).
- (2) In case of the 2 days-material balance period, the n.r.t. accountancy model of the MBA model 1 could detect the diversion of the rate of 52 kgPu per year at 36 days after the initiation of the diversion, and the total plutonium amount diverted was restricted to 5.2 kgs (Case P-2). If the MBA model 2 was adopted instead of the model 1, the diversion of the same rate could be detected at 3.8 months after its initiation, and the total amount at detection was restricted to 16.4 kgs (Case P-5).
- (3) In case of the weekly material balance, the diversion of the rate of 52 kgPu per year could be detected at 4 months later, and the total amount diverted was restricted to 17 kgs (Cases P-3b, P-6b).

4.5. Evaluation of the Results of Simulations

- (1) The time covered by the simulation runs is too short to bring a clearcut conclusion on the capability to detect the protracted diversion. Longer simulations (6 months ~ 1 year) are desirable.
- (2) The capability of the n.r.t. accountancy should evaluated for the case that the plant consists of two parallel process lines, taking correlations between the two lines into consideration.
- (3) The capability should also be evaluated when the n.r.t. accountancy system is extended to cover the whole chemical separation process. Correlations between two material balances of the dissolution-coseparation process and the succeeding plutonium purification process could be effectively utilized for safeguards purpose.

5. Conclusion

The study of the feasibility of applying the n.r.t. accounatncy system to safeguarding large-scale reprocessing plants is still on an early stage. Quantitative evaluations of the capability of attaining detection goals by the n.r.t. accountancy system has just been started in Japan. From the study performed up to now several useful suggestions were obtained, and they can be utilized in the future study.

TABLE 1

FLOW KEY MEASUREMENT POINTS FOR CONVENTIONAL MATERIALS ACCOUNTING IN THE MODEL FACILITY (Ref. 8)

KMP	Measurement Point	Material Description	Measurement Type	Instrument Precision (% 1σ)	Calibration Error (% lo)
1	Cask-unloading pool	Irradiated fuel assemblies ∿l% U-235, ∿l% Pu	Idéntification Item accounting		
2	Fuel transfer pool		Identification Item accounting		
3a	Accountability tank	Dissolver solution 300 g U/L 3 g Pu/L	Volume Mass spectrometry Mass spectrometry	0.3 1 1	0.1 0.2 0.3
3b	MBA 2 laboratory samples	U, Pu, FP in HNO3	Chemical analysis		
4	Dissolver acid surge tank	HNO3 (Recycle Acid) Trace of U	Volume; Fluorimetry or	2 20	3 10
		Trace of Pu	NDA, a	10	5
5	Leached hull basket	S.S., Zr Traces of U, Pu, FP	NDA		
6	Inspection sample Receipt/Shippment	Inspection sample			
7a _,	Pu product sample tank	Plutonium nitrate 250 g Pu/L	Volume Amperometry or coulometry	1 0.2	0.5 0.1
7b	Pu product interim storage tanks (3)	Plutonium nitrate 250 g Pu/L	Volume Amperometry or coulometry	0.3 0.2	0.1 0.1
68	Pu rework tank	Plutonium nitrate 250 g Pu/L	Volume Amperometry or coulometry	1 0.2	0.5 0.1
8b	Laboratory samples	Plutonium nitrate	Chemical analysis		
9	U product sample tank	Uranyl nitrate 370 g U/L	Volume Gravimetry	0.3 0.25	0.1

TABLE 1 (cont)

KMP	Measurement Point	Material Description	Measurement Type	Instrument Precision (% lo)	Calibration Error (% lo)
10a	U rework tank	Uranyl nitrate 370 g U/L	Volume Gravimetry	0.5 0.25	0.5 0.1
10Ъ	Laboratory samples	Uranyl nitrate	Chemical analysis.		
11	Solid-waste drums	Very low-level solid waste Traces of U, Pu	NDA Y,n	50	10
12a	HLW sample tank	Concentrated high-level waste 3 g U/L 0.l g Pu/L	Volume; Mass spectrometry Mass spectrometry	5 1 1	3 0.5 0.5
12Ъ	General process waste check tank	Concentrated low-level waste 13 g U/L Trace of Pu	Volume; Mass spectrometry Mass spectrometry	5 1 1	0.5 0.5 0.5
12c	Solvent-burner feed tank	Waste solvent Trace of U	Volume Fluorimetry or spectrophotometry	1 20	0.5 10
		Trace of Pu	NDA, a	10	5
12d	Central stack	Off-gas Traces of U, Pu	Volume NDA ?	20 40	10 20
13	Pu nitrate output accountability	Plutonium nitrate	Volume Amperometry or coulometry	0.2 0.2	0.1 0.1
14	V nitrate output accountability tank	Uranyl nitrate	Volume Gravimetry	0.3 0.25	0.1
15	Solid-waste drums	Very low-level waste	Identification Item counting	50	10
16	Liquid-waste drums	Condenced high-level waste Condenced low-level waste Waste solvent	Identification Item counting	5 10	2 5

FP = fission products. S.S. = stainless steel.

- 223 -

TABLE 2

INVENTORY KEY MEASUREMENT POINTS FOR CONVENTIONAL MATERIALS ACCOUNTING IN THE MODEL FACILITY (Ref. 8)

KMP	Measurement Point	Material Description	Measurement Type	Instrument Precision (%_lo)	Calibration Error (% lg)
Α	Spent-fuel pool	Irradiated fuel assemblies	Identification Item accounting		
В	Dissolver tank	Dissolver solution 310 g Pu/L 3 g Pu/L	Gravimetric (Pu/U ratio)		
C1	Feed-adjust tanks (2)	U, Pu, FP in HNO3 300 g U/L 3 g Pu/L	Volume Mass spectrometry Mass spectrometry	1 1 1	0.5 0.2 0.3
C2	1BP surge tank	U, Pu, residual FP in HNO3	Volume	1	0.5
		Material DescriptionMeasurement Typeradiated fuel assembliesIdentification Item accountingssolver solution 0 g Pu/LGravimetric (Pu/U ratio)Pu, FP in HNO3 0 g U/LVolume Mass spectrometry Mass spectrometry or coulometryg Pu/LVolume Mass spectrometry or coulometryf-spec uranyl nitrate 0 g U/LVolume Gravimetry or coulometryf-spec plutonium nitrate 0 g Pu/LVolume Mass spectrometry or coulometryscellaneous solutions ace of U ace of Pu he following tanks contain gligible quantities of U hovent, and miscellaneous blutions.Volume Mass spectrometry Mass spectrometry Volume Traces of U by fluorimetry or spectrophotometTraces of Pu by NDA, αTraces of Pu by NDA, α	1	0.25	
C3	Off-spec product tank	Off-spec uranyl nitrate 370 g U/L	Volume Gravimetry	0.5 0.25	0.5 0.1
C4	Pu rework tank	Off-spec plutonium nitrate 250 g Pu/L	Volume Amperometry or coulometry	1 0.2	0.5 0.1
C5	lSF tank	Miscellaneous solutions Trace of U Trace of Pu	Volume Mass spectrometry; Mass spectrometry	1 1 1	0.5 0.1 0.25
C6	LAWB check	The following tanks contain	Volume	1-5	0.5-3
C7	Recovered-acid storage	and Pu in recovered acid, solvent, and miscellaneous solutions.	Traces of U by fluorimetry or spectrophotometry	1-40	0.5-20
C8	Solvent-system feed (2)		Traces of Pu by NDA, α	10	5
C9	Solvent-batch strip				
C10	Service-concentrator feed				
C11	Service-concentrator check				
C12	Sump collection				
D	Laboratory	Assorted samples			

TABLE 2 (cont)

<u>KMP</u>	Measurement Point	Material Description	Measurement Type	Instrument Precision (% lo)	Calibration Error (%_lg)
B	Plutonium nitrate storage tank	Plutonium nitrate 250 g Pu/L	Volume Amperometry or coulometry	0.5 0.2	0.2 0.1
F	Uranium nitrate storage tank	Uranium nitrate	Volume Gravimetry	0.5 0.25	0.5 0.1
G	Solid-waste storage	Very low-level waste	Item accounting		
H	Liquid-waste storage	Condenced high-level waste Condenced low-level waste Waste solvent	Item accounting		

FP = fission products.

TABLE 3

MEASUREMENTS ADDED FOR n.r.t. ACCOUNTABILITY IN THE PLUTONIUM PURIFICATION PROCESS OF THE CHEMICAL SEPARATIONS PROCESS (MBA3) (Ref. 8)

Measurement Deint	Material Description	More uromonte muno	Instrument Precision	Calibration Error
Measurement Point	Material Description	Measurement Type	(8 10)	(8 10)
1BP stream	U, Pu, residual FP in HNO3 400 L/h 5 g Pu/L	Flow meter Absorption-edge densitometry	1	0.5 0.3
1BP surge tank	U, Pu, residual FP in HNO3 5 g Pu/L	Volume Density	3 3	
2A column	U, Pu, residual FP in aqueous, organic phases; Pu inventory		5-20	
2AW stream	U, Pu, residual FP in HNO3 500 L/h <0.1 g Pu/L	Flow meter NDA, α	5 10	1 2
2B column	U, Pu, trace FP in aqueous, organic phases, Pu inventory		5-20	
2BW stream	U, trace Pu in solvent 150 L/h Trace Pu	Flow meter NDA, a	5 10	1 2
3A column	U, Pu, trace FP in aqueous, organic phases, Pu inventory		5-20	
3AW stream	U, Pu, trace FP in HNO ₃ 215 L/h <0.l g Pu/L	Flow meter NDA, a	5 10	1 2
3B column	U, Pu in aqueous, organic phases; Pu inventory		5-20	
3BW stream	U, trace Pu in solvent 105 L/h Trace Pu	Flow meter NDA, a	5 10	1 2
3PS diluent-wash	Pu in aqueous phase, trace Pu in organic phase; Pu inventory		5-20	~-

TABLE	3	(cont)
-------	---	--------

Mea	surement Point	Material Description	Measurement Type	Instrument Precision (% 1σ)	Calibration Error (% lo)
3P (concentrator	Concentrated plutonium nitrate 250 g Pu/L	Volume (constant)	 1.5	
3PD	stream	Residual Pu in HNO3 32 L/h <0.l g Pu/L	Flow meter NDA, α	5 10	1 2
3PC	P stream	Plutonium-nitrate product 8 L/h 250 g Pu/L	Flow meter Absorption-edge densitometry	1 1	0.5 0.3

Саяе	nr t *		0.1.1	Throughput	Averaged over n.r.t. MBP σ_{MUF}				After 4 months	
Vase	MBA	MBP	tion ***	N.R.T MBP	Transfer	In-process	σ _M	UFd	σcusum	CUSUM
		(hrs)	(hrs)	(Kg)	component (Kg)	component (Kg)	Absolute (Kg)	Ratio to throughput (%:1a)	(Kg)	(Kg)
N – 1	Model 1	8	24	57.474	0.308	1.019	1.471	2.56	6. 590	8.408
2	^♪		168	57.474	0.309	1.019	1.471	2.56	10.429	6.660
3		48	24	101.456	0.831	1.002	1.634	1.61	6,546	8.520
4	-		168	101.456	0.923	1.002	1.682	1.66	10.347	7.874
5		168	24	355.097	1.556	0.996	2.098	0.59	6.576	8.000
6			168	355.097	2.499	0.996	2.868	0.81	10.420	6,641
7	Model 2	8	24	41.391	0 1 4 8	0.995	1.413	3.41	4.864	2.880
8	⚠∼⚠		168	41.391	0.149	0.995	1.413	3.41	12.109	1.960
9		48	24	101.455	0.599	1.002	1.528	1.51	4.830	3.121
10			168	101.456	0.813	1.002	1.624	1.60	12.007	2.592
11		168	24	355.097	1.124	0.993	1.799	0.59	4.853	3,207
12			168	355.097	2.912	0.993	3.232	0.91	12.105	. 2.771
13	Model 3	8	24	16.414	0.305	0.216	0 432	2.63	6.352	7.505
14	<u>&~A</u>		168	16 414	0.306	0.216	0.432	2.63	10.030	5.138
15		48	24	98.484	0.821	0.0	0.821	0.83	6.318	6,154
16			168	98.484	0.907	0.0	0.907	0.92	9.953	4.871
17		168	24	344.694	1.538	00	1.538	0.45	6.345	7.357
18			168	344.694	2.427	0.0	2.427	0.70	10.026	5.258

Table 4Characteristics of n.r.t. materials accountancy in the plutonium
purification process

* n.r.t. sub-MBA

** n.r.t. material balance period (n.r.t. MBP)

*** calibration periods for analyses and flow meters

- 228 -

Table 5 Capability of the n.r.t. materials accountacy in the chemical separation process of the model facility (1500 t/a) - Simulation study -

Case	Diversion	N. R. T.	MBA	N. R. T. MBP	Calibration period	Simulation covered	Average ⁰ млға	At the of sim CUSUM	end ulation CUSUM	Detection time	Total at detection	Reference
A - 1	Abrupt Div.	Model:1	1-2	6 hrs	24 hra	2 months	1.476 9	10.258 9	4.767 9	136 hrs	324 Kp	
A - 2	8 Kp / 2 Week	6		48	-		1,640	9,928	4.682	192	4.57	
A - 3a				168	~	-	2,105	12.325	4.612	288	6.86	
A - 36				168	168	~	2.872	16.028	7,205	_	343	t
A - 4		Model:2	$\Delta - \Delta$	6	24	*	2 419	7.726	3.567	120	2 86	
A - 5				48	~	~	1.533	7.389	3.523	192	4 57	
A - 6a	-			168	~	~	1.809	7.393	3.490	288	6.85	
A - 66				168	168	. .	3, 233	11.113	8,350		343	¢
P - 1	Protracted Div.	Model:1	A-A	8	24	4 months	1, 470	8.743	6,576	712	4 27	·
P - 2	52 Kp/year			48	~	~	1.633	8,389	6.533	864	5.18	
P - 3a				168	-	-	2.096	9.053	6.562	1.008	6 05	
P - 36		L		168	168	~	2.866	10,436	10,411		17.3	t
P-4		Model:2	$\Delta \overline{\Delta}$	8	24	-	1, 413	14.266	4.858	456	2.74	
P - 5				48	~	~	1, 528	13.782	4.824	2 736	16 4	
P - 6a				168	-	- '	1,798	13,844	4.847		17.3	\$
P - 66				168	158	~	3. 229	14.286	12.088		17.3	£ .
P - 7	32 Kg/year	Model:1	$\Delta - \Delta$	8	24	~	1. 413	8.019	4.860	840	3.08	
P-0				48	~	~	1. 528	7.627	4.826	_]	10.7	\$
P - 9				168	-	~	1, 798	7.631	4,849		10.7	8
P - 10	24Kp/year ·	Model:1	\$-₽	8	~	~	1. 471	- 222	6.583	880	2.42	
P - 11				48	-	· ۲	1. 633	- 450	6,540	_	8.0	t
P - 12				168	~	~	2.097	130	6,569	·	80	\$
P - 13		Model:2	$\Delta - \Delta$	8	•	-	1.413	5,314	4.861	.2.480	6.81	
P - 14				48	-	~	1.528	4.961	4.827	·	8.0	¢
<u>P - 15</u>				168	~	-	1.798	4,942	4,850	·	8.0	¢ l
P · 16	16 ^R P/year	Model:2	$\Delta \sim \Delta$	8	~	~	1. 413	2.600	4,862	2,496	4 57	longor torm
P - 17				48	~	-	1, 528	2,284	4,828	·	5.3	simulation is
P - 18				168	-	-	1.798	2.244	4.851		5.3	necessary
P - 19	8kp/year	Model :2	$\Delta - \Delta$	8	~	~	1, 413	- 140	4.863	2.512	2.30	longor
P - 20				48	~	-	1, 528	- 420	4.829	·	2.7	simulation is
P - 21		L		168	~	~	1.798	- 483	4.852		2.7	necessary

* Total plutonium amount diverted in the simulation period exceeded 8 kgs.



Fig. 1 The MBA structure of the model facility

- 230



Fig. 2. Plutonium purification process block diagram and sub-MBA structures



Fig. 3 Plutonium holdup in the plutonium purification process



Fig. 3 (Cont¹d)

-

- 233 ---



Fig. 4 Plutonium concentration in major processes in the plutonium purification process

- 234 -



Fig. 5 Diversion sensitivity analysis (Case : A-4)



BALANCE 0 01



Fig. 6 Diversion sensitivity analysis (Case : A-5)





Fig. 7 Diversion sensitivity analysis (Case : A-6a)

.

REFERENCES

- TASTEX; Tokai Advanced Safeguards Technology Exercise, International Atomic Energy Agency, Technical Reports Series No.213, Vienna 1982.
- 2. Ikawa, K.; Ihara, H.; Nishimura, H.; Hirata, M.; Sakuragi, H.; and Ido, M.; "Final Report of TASTEX task F - Study of the Application of Near-Real-Time Materials Accountancy to Safeguarding Reprocessing plants", Japan Atomic Energy Research Institute report JAERI-memo-9532 (May 1981). (Note: JAERI-memo-9532, strictly speaking should not be referenced. However, an openly available version is planned. The reference should be corrected later.)
- 3. Lovett, J.E.; Hirata, M.; Ikawa, K.; Augustson, R.H., "Application of the Basic concepts of Dynamic Materials Accountancy to the Tokai Spent Fuel Reprocessing Facility - A Feasibility Study", International Atomic Energy Agency Department of Safeguards internal report STR-94 (January 1980). Also published in Japan as JAERI-M-9186 (November 1980).
- 4. Lowry, L.L.; and Augustson, R.H.; comp.; "TASTEX task F, Study of Selected Capabilities Needed to Apply DYMAC Principles to Safeguarding the Tokai Reprocessing Plant", Los Alamos Scientific Laboratory report LA-8070-MS (date?).
- 5. Lovett, J.E.; Hirata, M.; Augustson, R.H.; "Application of the Basic Concepts of Dynamic Materials Accountancy to the Tokai Spent Fuel Reprocessing Facility - a Feasibility Study", Proceedings, First Annual (ESARDA) Symposium on Safeguards and Nuclear Material Management, Brussels, April 1979.
- 6. Ikawa, K.; et al.; "Study of the Application of Semi-Dynamic Material Control Concepts to Safeguarding Spent Fuel Reprocessing Plants", in NBS Special Publication 582, Report of the Kiawah Island Topical Meeting, <u>Measurement Technology for Safeguards</u> <u>Materials Control</u>, (meeting date November 1979, publication date June 1980), pp. 730-739.

- 7. Ikawa, K.; Ihara. H.; Nishimura, H.; Hirata, M.; and Ido, M.; "An Integrated Near-Real-Time Materials Accountancy Safeguards System", Journal of the Institute of Nuclear Materials Management, Proceedings Issue, 22nd annual meeting, San Francisco, pp. 390-401 (July 1981).
- 8. 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, J.W. Barnes; <u>Coordinated Safeguards for materials management in a Fuel</u> <u>Reprocessing Plant</u>, Los Alamos Scientific Laboratory Report LA-6881, Vol.I,II (September 1977)
- 9. Hakkila, E.A., et al., "Materials Management in an Internationally Safeguarded Fuels Reprocessing Plant", LA-8042 (April 1980)