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Materials Accounting Considerations for International Safeguards in a Light-Water Reactor Fuels Reprocessing Plant

by

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ABSTRACT

This paper summarizes the requirements and functions of materials measurement and accounting systems applicable to large (1500 metric tonnes heavy metal per year - MTHM/yr) future reprocessing facilities as well as small (210 MTHM/yr) plants that are presently under IAEA safeguards. The effectiveness of conventional and proposed improved measurement and accounting systems were compared using modeling, simulation, and analysis procedures. The study showed that conventional accountability can meet IAEA goal quantities and detection times in these reference facilities only for low-enriched uranium. Dynamic materials accounting may meet IAEA goals for detecting abrupt (1-3 wks) diversion of 8 kg of plutonium. Current or projected techniques cannot meet the one year protracted diversion goal for plutonium if this goal is based on an absolute 8 kg quantity.

KEYWORDS: Nuclear safeguards; dynamic accounting; fuel reprocessing

I. INTRODUCTION

This study attempts to identify problems and proposes solutions involved in nuclear materials accountability for internationally safeguarding light-water-reactor spent fuel reprocessing plants. The problem was addressed by studying a large reprocessing facility that may be on stream in the 1990s time frame as well as a small plant representative of facilities presently under IAEA safeguards. Near-real-time materials measurement and accounting concepts previously proposed for a State's accounting system[1] were extended to include the problems associated with international verification.

II. BASIS FOR INTERNATIONAL SAFEGUARDS

The basis for most current international safeguards agreements is the 1968 Treaty on the Non-Proliferation of Nuclear Weapons (NPT) agreed to by over 100 signatory nations. The detailed terms and conditions under which specific facilities are safeguarded are negotiated with the International Atomic Energy Agency (IAEA) in accord with the general conditions of Article III of the NPT as set forth in the IAEA document INFCIRC/153.[2]

The objective of international safeguards, as declared by these documents, is the "...timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities... ." The emphasis is on "...the use of materials accountancy as a safeguards measure of fundamental importance, with containment and surveillance as important complementary measures... ."

INFCIRC/153, para. 31 also requires that the IAEA "shall make full use of the State's system of accounting for and control of all nuclear material subject to safeguards under the Agreement, and shall avoid unnecessary duplication of the State's accounting and control activities." In the case of reprocessing plants, the materials balance closing is determined by computing the material unaccounted for and its limit of error based on a measured, verified materials balance. The uncertainty associated with the nuclear materials balance depends fundamentally on the measurement system uncertainties, on the plant throughput, and on the beginning and ending inventories for the materials balance period.

The application of international safeguards is negotiated between the IAEA (Agency) and the State (operator) on a case-by-case basis. "Goal quantities" for the detection of diversion have been proposed by the IAEA, but have not been generally accepted by Member States. These "goals" are related to the quantities of nuclear materials required to produce an explosive device and the time necessary to convert these materials to that purpose. The goals include the detection of the diversion of:

- 75 kg of uranium-235 contained in low-enriched uranium over a period of one year.
- 8 kg of plutonium in 1-3 weeks ("abrupt diversion").
- 8 kg of plutonium over an entire year ("protracted diversion").

The agency verification of the State's accounting system consists of three steps:

- Examination of the information provided in the Design Information Questionaire and in subsequent routine and special accounting reports;
- Collection of independent information by the IAEA in inspections;
- Evaluation of the information provided by the State and collected in inspections for the purpose of determining the completeness, accuracy, and validity of the information provided by the State.

Inspection activity as defined in INFCIRC/153 permits approximately 3700 man hours (18 man years) and 1400 man hours (7 man years) of annual inspection, respectively, for plants having annual throughputs of 1500 and 210 MTHM.

III. REFERENCE FACILITIES

In this study we have used the Allied-General Nuclear Services (AGNS) Barnwell plant as a reference facility for the high-throughput plant and the PNC pilot facility at Tokai-mura, Japan (Tokai) as the reference facility for the smaller plant. Both reprocessing plants use conventional Purex technology to reprocess LWR reactor fuel having a nominal plutonium concentration of approximately 1%. The following differences in process design or operation could be important for materials accounting.

- The AGNS plant uses a centrifugal contactor for initial fission product decontamination, with pulsed columns for all subsequent extraction, scrub, and strip operations. The Tokai facility employs mixer-settlers throughout.
- The centrifuge for solids removal (fission product metallic ingots, Zircaloy fines) is located between the accountability tank and process feed tank at AGNS and between the dissolver and accountability tank at Tokai.
- An additional scrub section in the Tokai plant between the fission product decontamination and the uranium-plutonium partition steps provides an additional 10 to 100-fold improvement in fission product decontamination before the plutonium purification cycle.
- Buffer tanks are included between the decontamination and partition cycles and between the partition and plutonium purification cycles in the Tokai design.

IV. MBA STRUCTURE FOR CONVENTIONAL AND DYNAMIC MATERIALS ACCOUNTING

Both the State's system of accounting and control and the international safeguards system depend fundamentally on the definitions of process areas about which materials balances are to be drawn. We have examined several strategies for drawing these balances for the large and small reprocessing facilities and the conversion process.

We term conventional any materials accounting scheme in which balances are drawn solely on the basis of physical inventories. Under this kind of strategy, the facility customarily is divided into a number of materials balance areas (MBAs) such as those shown in Fig. 1. A balance is drawn about each MBA coincident with a physical inventory of that MBA. Thus, the timeliness of a conventional accounting system is limited by the physical inventory frequency, which in turn is severely constrained by the economics of process operation.

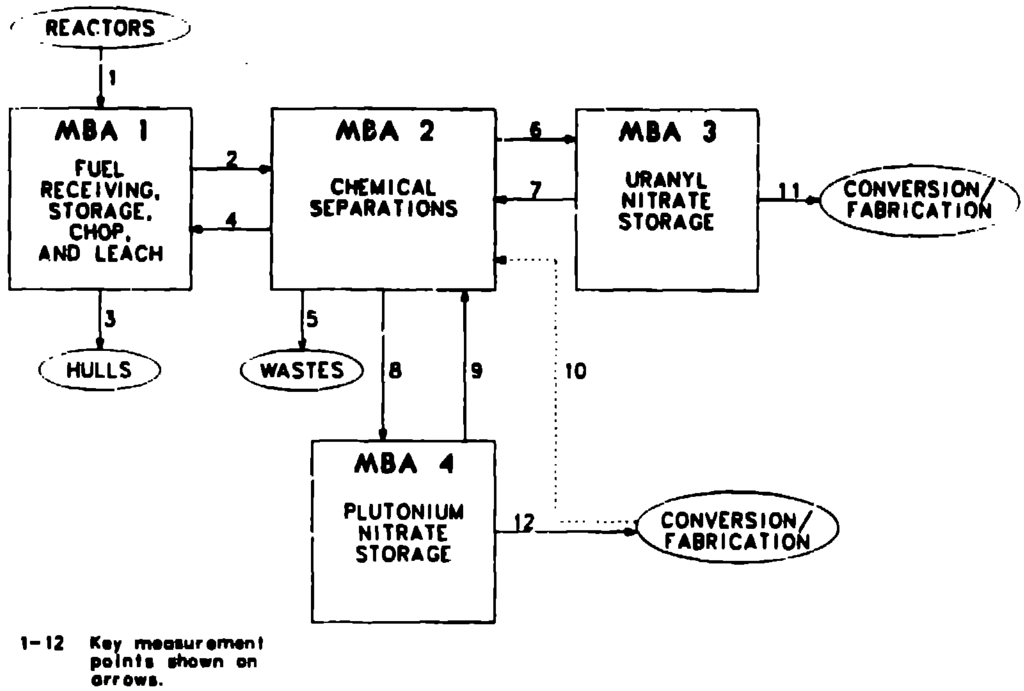

Fig. 1. MBAs for reprocessing facilities.

Near-real-time (or dynamic) materials accounting may be thought of as an augmentation of conventional materials accounting in which additional materials balances are drawn between physical inventories. The physical inventory measurements are replaced by measurements, or estimates, of the in-process inventory using on-line or at-line instrumentation and sophisticated data analysis methods. The drawing of such dynamic materials balances sometimes is facilitated by subdividing the MBAs into unit process accounting areas (UPAAs) that are closely related in time and space through process structures and operating procedures. The use of near-real-time accounting based on the UPAA subdivision generally provides improved sensitivity, in time, location, and amount, to diversion of nuclear material. We have considered mixtures of these strategies for the reference facilities.

A. Large Reference Facility

High-throughput facilities, such as the AGNS plant, will be of increasing safeguards interest in the next few years. Therefore, we have studied the safeguards aspects of such a reference facility based on the AGNS design as the best example currently available.

1. Conventional Materials Accounting

Conventional materials accounting relies on discrete-item counting and materials-balance closure following periodic shutdown, cleanout, and physical inventory. For this study, the baseline facilities are divided into four MBAs. An MBA is generally a physical area that is identified such that the quantity of nuclear materials moving into or out of the MBA can be measured. The input, output, and inventory measurement points for these MBAs are called key measurement points (KMPs).

As shown in Fig. 1, the four MBAs are fuel receiving, storage, chop, and leach (MBA 1), separations process area (MBA 2), uranium product storage area (MBA 3), and plutonium-nitrate storage area (MBA 4). MBAs 1, 3, and 4, are shipper/receiver MBAs while MBA 2 is a process MBA. Each of the MBAs is described in the following text.

a. MBA 1--fuel receiving, storage, chop, and leach. The fuel receiving, storage, chop, and leach MBA includes the cask-unloading and spent fuel pools, the shearing operation, and the dissolution process. The flow KMPs are:

- KMP 1 - receipt of irradiated fuel,
- KMP 2 - transfers from MBA 1 to MBA 2 (chemical separations MBA),
- KMP 3 - measured discards (hulls), and
- KMP 4 - recycle from MBA 2.

The inventory KMP is located in the spent fuel pool.

A shipper/receiver difference can be closed about MBA 1 after each campaign (approximately every 5 days) when the dissolver tanks, hull-rinse tanks, and associated piping are drained and flushed into the accountability tank. This flush-out between batches from different customers results in a more accurate shipper/receiver difference because it minimizes contamination from previous customer batches. The shipper/receiver difference is obtained by adding the shipper's values for a number of fuel batches (KMP 1) to the corresponding number of batches of recycled acid (KMP 4) and subtracting the accountability tank and laboratory vial batches (KMP 2) and the leached hull batches (KMP 3). Inventory verification in MBA 1 is based on piece count and identification of the fuel assembly fabrication serial numbers.

b. MBA 2--chemical separations process. This MBA includes the solvent-extraction operations from the accountability tank to the uranyl-nitrate and plutonium-nitrate product sample tanks. The flow KMPs are:

- KMP 2 - transfers to MBA 2 from MBA 1,
- KMP 4 - recycle to MBA 1,
- KMP 5 - measured discards and retained waste,
- KMP 6 - transfers from MBA 2 to MBA 3 (uranyl-nitrate storage),
- KMP 7 - recycle from MBA 3,
- KMP 8 - transfers from MBA 2 to MBA 4 (plutonium-nitrate storage),
- KMP 9 - recycle from MBA 4, and
- KMP 10 - transfers to MBA 2 from the conversion process.

The inventory KMPs are the analytical laboratory and those tanks in which reliable volume measurements can be made when the process is drained and flushed.

A physical inventory in MBA 2 includes a shutdown and flushout of the separations process area, and a cleanout of extraneous samples and a piece-count verification of remaining materials in the laboratory. The process line is drained and flushed into approximately 26 primary accountability tanks that have been calibrated so that reliable volume measurements can be made and samples can be taken for analysis.

A materials balance is taken after each physical inventory by adding all measured receipts (KMPs 2, 7, 9, and 10) to the initial inventory and subtracting all measured removals (KMPs 4, 5, 6, and 8) and the final inventory.

c. MBA 3--uranyl nitrate product. The uranyl-nitrate product MBA is a shipper/receiver MBA. The shipper's value is accepted under KMP 6 and is obtained from chemical analysis of a sample and volume measurement of the uranium product sample tank. The receiver's value is accepted under KMP 11 and consists of chemical analysis of a sample and volume measurement of the uranyl nitrate accountability tank at the headend of the UF_6 facility. This MBA has no inventory because solution is transferred directly from the uranium product tank in the chemical separations area (MBA 2) to the collocated UF_6 facility.

d. MBA 4--plutonium nitrate product storage. The plutonium nitrate product storage MBA contains slab tanks that are capable of storing 42 000 L of plutonium nitrate at a concentration of 250 g Pu/L. This MBA is a shipper/receiver MBA. The plutonium-nitrate solution transferred from the plutonium-product measuring tank to the plutonium-nitrate storage-facility slab tanks through KMP 8 constitutes the shipper's value. The nitrate product transferred to the receipt tanks in the collocated oxide-conversion plant constitutes the output of MBA 4. The receiver's value is determined by volume measurements and samples taken for chemical analysis in the receipt tanks. Alternatively, plutonium-nitrate product that does not meet specifications can be recycled through KMP 9 from the slab tanks back through the separations process area (MBA-2) on a campaign basis. In this case, the receiver's value is determined in the plutonium rework tank in MBA 2 using volume measurements and chemical analysis.

A physical inventory in MBA 4 requires volume measurements, sampling, and analysis of all solutions in the storage area or, alternatively, confirmation that tamper-safe seals are intact and the prior measurements are still valid.

2. Dynamic Materials Accounting

Dynamic materials accounting can provide significant improvement in the chemical separations process MBA. The chemical separations process area can be treated either as a single UPAA or as two UPAAs: a codecontamination-partitioning process UPAA (UPAA 1) and a plutonium-purification process UPAA (UPAA 2). This UPAA structure is complementary because dynamic materials balances can be taken about the chemical separations area in two ways.

a. UPAA 1 2--chemical separations process. The chemical separations process MBA can be treated as a single UPAA (UPAA 12) if measurements of the in-process inventory are made on each of the major process vessels in the process area. The inventory measurements must be added to the inventory KMPs.

In-process inventory measurements can be combined with flow KMPs 2, 4, 5, 6, 7, 8, 9, and 10 to form a dynamic materials balance approximately every two days. Because most of the material is transferred through the feed and product KMPs, the frequency of taking materials balances is governed by the feed and product batch frequencies. Under normal operating conditions, two and one-half accountability batches and one product batch are processed every day. Therefore, process logic dictates that a materials balance can be taken every two days to include an integral number of feed and product batches. Smaller batches, for example waste batches to high-level waste, are included in the materials balances when the measurements become available.

Alternatively, a materials balance could be taken around UPAA 1 2 after each feed batch (approximately every 9.6 h) if an on-line plutonium product measurement is added. The product measurement would consist of flow and concentration measurements.

b. UPAA 1--codecontamination-partitioning processes. A separate UPAA can be formed around the codecontamination-partitioning processes if flow and concentration measurements are added to the 1BP, 1SP, and POR streams. A dynamic materials balance can be taken about UPAA 1 for each feed accountability batch (every 9.6 h) by combining measurements of the concentration and volume of the feed batch, the concentration and flow in the 1BP, 1SP, and POR streams, the initial and final in-process inventories in the process vessels, and the concentration and volume of the high-activity waste (HAW) sample tank solution.

c. UPAA 2--plutonium purification process. Dynamic materials balances can be taken about the plutonium purification process if flow and concentration measurements are added to the aqueous and organic recycle streams (2AW, 2BW, 3AW, 3BW, and 3PD), and in-process inventory in contactors and the evaporator can be estimated. The balances can be taken using one of two product measurements, the daily batch in the plutonium sample tank or the on-line flow and concentration measurements on the concentrator product (3PCP) stream. Contactor in-process inventory may be estimated using process operating data.[3]

B. Small Reprocessing Plant

Many commercial reprocessing plants that are currently operating have capacities of less than 300 MTHM/year. Therefore, a materials measurement and accounting system that would be more typical of presently operating reprocessing plants was evaluated using the Tokai reprocessing plant as the reference facility.

1. Conventional Materials Accounting

The physical inventory accounting system structure in a small plant is identical to that of the large plant.

2. Dynamic Materials Accounting

Near-real-time accounting of plutonium can be applied to the chemical separations area, as a single UPAA, without additional measurement points by periodically sampling for chemical analysis and measuring the volume of each of the process vessels, and estimating the in-process inventory in each mixer-settler bank. These measurements are necessary for determining the in-process inventory. The UPAA boundaries are the accountability tank, the plutonium receiver tank, and the waste and recycle acid tanks. A dynamic materials balance can be drawn after any integral combination of feed and product batches; i.e., a materials balance could be taken as often as once a day (two feed batches and one product batch).

As shown in Fig. 2, the near-real-time accounting system could be extended to include three UPAAs, and combinations thereof, within the chemical separations area. The UPAAs within the chemical separations area would be codecontamination-partitioning, UPAA 1; codecontamination, UPAA 1A; partitioning, UPAA 1B; and plutonium purification, UPAA 2. The codecontamination-partitioning can be divided into two UPAAs because of the buffer tanks that are between the first and second extraction cycles. This option is lacking in the large chemical separations plant where such a division is not possible. Added measurements include flow and concentration in the streams between the UPAAs, as well as on-line or at-line concentration measurements for determining in-process inventories. The feed and product batch measurements rely on the traditional installed volume measurements coupled with chemical analysis. In-process inventory volume measurements are also in place.

V. MODELING, SIMULATION, AND ANALYSIS TECHNIQUES

A. Modeling and Simulation Approach

The design and evaluation of the accounting systems are based on computer simulations of the reference facilities because these facilities have either not been built or have not been operated in a full production mode. Additionally, alternative operating, measurement, and accounting strategies can be readily compared.

The modeling and simulation approach requires (1) a detailed dynamic model of the process based on actual design data and operator experience; (2) simulation of the model process on a digital computer; (3) a dynamic model of each measurement system based on best estimates of instrument performance and behavior; (4) simulation of accountability measurements applied to nuclear materials flow and in-process inventory data generated by the model process simulation; and (5) evaluation of simulated materials balance data from various materials accounting strategies.[4]

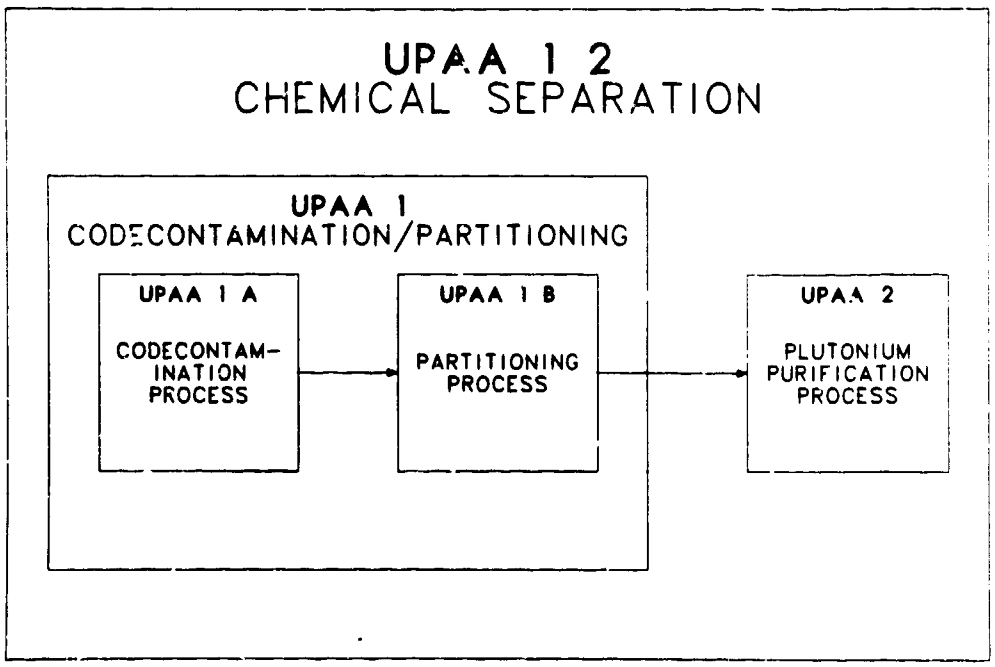

Fig. 2. UPAAs in the reference small chemical separations facility.

B. Data Analysis Techniques

Analysis of materials accounting data for detection of possible nuclear materials diversion is one of the major functions of the MMAS. Diversion may occur in two basic patterns: abrupt diversion (the single theft of a relatively large amount of nuclear materials) and protracted diversion (repeated thefts of nuclear materials on a scale too small to be detected in a single materials balance because of measurement uncertainties).

The use of unit-process accounting and dynamic materials balances enhances the ability to detect such diversions, but it also means that the operator of the safeguards system will be inundated with materials accounting data.

Decision analysis (see Refs. 5-9), which combines techniques from estimation theory, decision theory, and systems analysis, has been developed as a logical framework of tools for statistical treatment of the dynamic materials accounting data that become available sequentially in time. Its primary goals are (1) detection of the event(s) that nuclear materials has been diverted, (2) estimation of the amount(s) diverted, and (3) determination of the significance of the estimates.

The decision analysis algorithms include the Shewhart chart, cusum, uniform diversion test (UDT), sequential variance test (SVT), smoothed materials balance test (SMBT), and Wilcoxon rank sum test. The algorithms for the Shewhart chart, cusum, UDT, SVT, and SMBT are structured to account for correlated data (so-called systematic errors) so that correct variances are computed for the associated decision tests.

C. Data Analysis Graphic Aids

The decision tests must examine all possible sequences of the available materials balance data because, in practice, the time at which a sequence of diversions begins is never known beforehand. Furthermore, to ensure uniform application and interpretation, each test should be performed at several levels of significance. Thus, a graphical display that indicates those sequences that cause alarms, specifying each by its length, time of occurrence, and significance, is essential. One such tool is the alarm-sequence chart,[10] a type of pattern recognition device that has proven very useful for summarizing the results of the various tests and for identifying trends.

D. Systems Performance Analysis

One essential part of designing nuclear materials accounting systems is analyzing their expected performance in detecting losses of nuclear material.[11] Systems performance analysis, in turn, implies the definition of suitable performance measures that can be easily related to externally established criteria. Thus, there are two aspects of the analysis problem: first, defining performance measures, and second, relating those measures to established, quantitative performance criteria.

Performance measures for any nuclear materials accounting system embody the concepts of loss-detection sensitivity and loss-detection time. Because of the statistical nature of materials accounting, loss-detection sensitivity can be described in terms of the probability of detecting some amount of loss while accepting some probability of a false alarm. Loss-detection time is the time required by the accounting system to reach some specified level of loss-detection sensitivity. Note that the loss scenario is not specified; that is, whether the loss occurs in an abrupt or in a protracted fashion, the total amount of loss is the measure of performance. Note also that loss-detection time only refers to the internal response time of the accounting system.

Intuitively, the performance of any accounting system is describable by some function

$$P\,[L,N,\alpha] \quad , \tag{1}$$

where P is the accounting system's probability of loss detection, L is the total amount of loss over a period of N balances, and α is the false-alarm probability. Thus, a convenient way of displaying system performance would be a three-dimensional graph of the surface P versus L and N for some specified value of α. These graphical displays, called <u>performance surfaces</u>, portray the expected performance of an accounting system as a function of the three performance measures, loss, time, and detection probability, rather than as a single point.

VI. EFFECTIVENESS OF THE OPERATOR'S MATERIALS MEASUREMENT AND ACCOUNTING SYSTEM

The operator's material measurement and accounting system for an internationally verifiable safeguards system, including location and types of flow and concentration sensors, has been described previously.[1,12,13] The effectiveness of the accounting system that uses conventional and dynamic accounting for detecting abrupt and protracted diversion of uranium and plutonium were evaluated for the MBA structures described in Sec. III using modeling, simulation, and analysis techniques discussed in Sec. IV.[13]

Table I lists materials balance standard deviations for conventional materials accounting in the process MBAs of the reference facilities. These materials accounting sensitivities will be degraded if high-quality measurements cannot be obtained. Conversely, the sensitivities could be improved if measurement errors can be controlled. Measurement errors can be controlled by identifying the dominant error sources and establishing effective measurement control procedures. Note that the diversion detection sensitivity is at least 3.3 times the materials balance standard deviation for a 95% detection probability and a false alarm probability of 5%. From our analysis, we conclude that:

- For ^{235}U the proposed IAEA criteria for diversion sensitivity and timeliness probably are attainable by conventional materials accountability if rigorous materials measurement control programs are instituted.
- For plutonium, the proposed IAEA criteria for sensitivity and timeliness cannot be met by conventional materials accountability.

Near-real-time materials accounting techniques were applied to the process MBAs in an effort to meet the proposed IAEA criteria. Materials balance uncertainties for the reference facilities are summarized in Table II. In each case, a range of uncertainties is given for the largest UPAA that was considered--the entire process area for each facility. The cases considered range from best-case estimates of contactor in-process inventories with two-day recalibrations of input-output flow and concentration measuring instruments, to worst case estimates of contactor in-process inventories with no recalibrations within the accounting periods. Note that the diversion detection sensitivity is at least 3.3 times the materials balance standard deviation for a 95% detection probability and a false-alarm probability of 5%.

In examining both the conventional (Table I) and the dynamic (Table II) materials accounting sensitivities, we further conclude that for plutonium:

- In the <u>large</u> chemical separations process area, the proposed IAEA criteria for detecting <u>abrupt</u> diversion can probably be met if a rigorous measurement control program is undertaken.
- In the <u>large</u> chemical separations process area, the proposed IAEA criteria for detecting <u>protracted</u> diversion cannot be met by any known system; the goal quantity is only 0.05% of the annual plant throughput.

TABLE I

CONVENTIONAL MATERIALS ACCOUNTING IN THE REFERENCE FACILITIES

Accounting Period (months)	Materials Balance Standard Deviations (kg)			
	Large Reference Facility		Small Reference Facility	
	U-235	Pu	U-235	Pu
3	10.4	13.4	1.4	1.9
6	20.3	26.2	2.8	3.7
12	40.1	52.1	5.7	7.3

TABLE II

DYNAMIC MATERIALS ACCOUNTING IN THE REFERENCE FACILITIES

Accounting Period	Materials Balance Standard Deviations (kg Pu)	
	Large Reference Facility[a]	Small Reference Facility[b]
1 balance[c]	2.0-2.3	0.26-0.37
1 day	---	0.26-0.37
2 days	2.1-2.4	---
1 week	2.5-3.4	0.32-0.43
2 weeks	3.0-5.3	0.37-0.57
1 month	3.9-9.5	0.53-0.83

[a]Ranges are given from two-day recalibration, 5% estimates of contactor in-process inventories to no recalibration, 10% estimates of contactor in-process inventories.

[b]Ranges are given from two-day recalibrations, 10% estimates of contactor in-process inventories to no recalibrations, 20% estimate of contactor in-process inventories.

[c]A materials balance is taken every 9.6 h in the large chemical separations process and one day in the small chemical separations process.

- In the small chemical separations process area, proposed IAEA criteria for abrupt diversion probably can be met.
- In the small chemical separations process area, the proposed IAEA criteria for protracted diversion may be achievable.

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