LA-7521

C.3

Coordinated Safeguards for Materials Management in a Uranium-Plutonium Nitrate-to-Oxide Coconversion Facility: Coprecal

University of California

LOS ALAMOS SCIENTIFIC LABORATORY
Post Office Box 1663 Los Alamos, New Mexico 87545

Work supported by the US Department of Energy,
Office of Safeguards and Security.

UNITED STATES
DEPARTMENT OF ENERGY
CONTRACT W-7405-ENG. 36

LA-7521
UC-15 and UC-83
Issued: February 1979

Coordinated Safeguards for Materials Management in a Uranium-Plutonium Nitrate-to-Oxide Coconversion Facility: Coprecal

H. A. Dayem
D. D. Cobb
R. J. Dietz
E. A. Hakkila
E. A. Kern
E. P. Schelonka
J. P. Shipley
D. B. Smith

EXECUTIVE SUMMARY

This report describes the sixth in a series of studies for applying advanced nuclear materials accountability techniques to high-throughput nuclear fuel-cycle facilities. The purpose of this study is to provide safeguards design concepts for a coconversion facility that converts mixed plutonium-uranium nitrate solution to mixed-oxide feed material for a nuclear fuel refabrication plant. The reference coconversion facility is based on the pilot-demonstrated Coprecal process and has sufficient capacity to convert the plutonium output of approximately 100 light-water reactors of 1000 MW electrical power capacity. The process design and operating procedures provided for the reference facility are tentative and incomplete and have not been previously evaluated for their safeguardability.

Conceptual design efforts for nuclear materials accountability systems are a major portion of an integrated safeguards systems study program that is implemented by the Safeguards Systems Group (Q-4) of the Los Alamos Scientific Laboratory (LASL), at the direction of the US Department of Energy's Office of Safeguards and Security (DOE-OSS). These conceptual designs invoke materials measurement and control technology that has been demonstrated or can be projected in a time frame consistent with the construction schedule of future nuclear fuel-cycle facilities. The studies are intended to define systems concepts, to develop methods for evaluating safeguards systems and the data they produce, and to stimulate further development of the facilities, processes, systems, and instrumentation needed for improving nuclear materials accountability, thus providing more effective safeguards.

Previous reports in this series addressed the materials management requirements of mixed-oxide fuel refabrication facilities (LA-6536), spent-fuel reprocessing plants (LA-6881), fast-critical facilities (LA-7315), thorium-uranium fuel reprocessing plants (LA-7411-MS), and plutonium nitrate-to-oxide conversion facilities (LA-7011). The intent of this report is to evaluate the safeguards materials accountability requirements of a typical coprocessing option by using a reference uranium-plutonium nitrate coconversion facility having the same capacity as the pure plutonium nitrate conversion facility described in LA-7011. This coprocessing study is the third in a series of alternative fuel-cycle safeguards studies performed in support of the Savannah River Laboratory's Alternative Fuel Cycle Technology (AFCT) Program. Previous reports in the AFCT series treated the thorium fuel-cycle option.

Safeguards effectiveness evaluations of complex, high-throughput fuel-cycle facilities require intimate knowledge of the facility, its processes, and its operational details. In addition to these requirements, there are others introduced by the unique characteristics of a dynamic materials management system that subdivides the process into individual unit process accounting areas (UPAA). To satisfy the requirements of both process and materials control, it is necessary to use computerized techniques to analyze the large quantities of process and materials-balance data produced by those systems. Such an analysis has been done in this study by modeling and simulating plant operations, materials flows, and the functions of a materials measurement system, and by computerizing the routine aspects of the decision process. The development and standardization of methods for decision analysis as well as safeguards effectiveness evaluation are increasingly important components of these studies and have helped immensely in defining areas where process and measurement improvements are needed.

The reference coconversion facility is based on a formerly proprietary process (Coprecal) developed by the General Electric Company (G.E.) and has been scaled to accept the output of a 10-tonne-per-day light-water-reactor spent-fuel reprocessing facility. Heavy-metal product solution from the reprocessing plant is supplied at a nominal uranium/plutonium ratio of 8 to 1 and is diluted with natural uranium to a 10% plutonium-90% uranium-nitrate feed solution. Because of a concern for product and isotopic uniformity, G.E. elected to design the process to operate continuously on a campaign basis for over two weeks, using three parallel process lines. This process strategy, coupled with the need for pre-diluting the plutonium solution with uranium, requires an extremely large feed and blend capacity (144,000 L) to ensure homogeneity and process continuity during the extended campaign.

Large equipment, continuous processing, the presence of overwhelming quantities of uranium, and the need for handling ten times as much material contribute to the difficulty of safeguarding this process relative to the pure-plutonium reference process (Oxalate III) previously studied. A direct comparison is complicated by the absence of a comparable blending and mixing function in the Oxalate (III) process, which does not include provisions for homogenization and isotopic blending. To address this problem, we have used several materials accounting strategies in our treatment of the Coprecal process, both with and without large feed-blend tanks, to attempt a comparative evaluation of the conversion and coconversion processes. The influence of the large feed-blend tanks in Coprecal is best shown by treating them as a separate unit process accounting area (UPAA 1). The detection sensitivity of an advanced materials accounting system for any one of the three parallel feed-blend unit processes is given below for time intervals near the beginning of a campaign, when measurement uncertainty is greatest.

COPRECAL
DETECTION SENSITIVITY
STRATEGY 1, UPAA 1

Detection Time	Average Diversion per Balance (2 h) (kg Pu)	Total at Time of Detection (kg Pu)
Protracted diversion		
1 day	1.4	6.8
1 week	0.12	10.1
1 campaign	0.10	20.4
Abrupt Diversion		
2 hours	26.7	26.7
1 day	11.0	11.0
1 week	4.3	4.3
1 campaign	2.8	2.8

If the feeding and blending operation could be modified, for example, by feeding small intermediate aliquot tanks having 2-h operating capacities, the large measurement errors associated with the feed-blend tank would no longer dominate the detection sensitivities. With the exclusion of the large feed-blend tanks from the process simulation, the detection sensitivities of the Coprecal and the Oxalate (III) processes may then be compared on a consistent basis, as below, again for a single process line.

DETECTION SENSITIVITIES
COPRECAL STRATEGY 4, UPAA 23 (BEST CASE)
AND OXALATE (III) STRATEGY 2

	Coprecal		Oxalate (III)	
Detection Time	Average per Balance (2 h) (kg Pu)	Total at Detection (kg Pu)	Average per Balance (1.3 h) (kg Pu)	Total at Detection (kg Pu)
1 balance	1.2	1.2	0.4	0.4
1 day	0.13	1.6	0.02	0.5
1 week	0.04	3.7	0.01	1.7
1 month	0.03	8.4	0.007	3.9

The better detection sensitivity attainable in the Oxalate (III) conversion process is a result of its smaller in-process inventory and the improved precipitator-feed measurements that are made possible because of batch feeding from a small tank.

The two tables above show that the relatively large absolute errors in the feed-blend tank inventory measurements preclude effective materials accountability in the Coprecal process unless the feed-blend tanks can be separated, for accounting purposes, from the rest of the process. Detection sensitivities approaching those estimated for the conversion process should be obtained for the Coprecal process if aliquot tanks are inserted after the feed-blend tanks in the precipitator-feed streams.

Diversion sensitivities for a variety of measurement strategies and unit-process configurations were determined from a series of computer-generated process histories using decision-analysis algorithms developed to handle the large quantities of materials measurement data characteristic of the near-real-time accountability systems invoked in this study. The diversion sensitivity values summarized for a single operating process line above should be compared to the current regulatory requirement for a detection limit of 33 kg plutonium, determined every two months for the entire facility.

In addition to having degraded detection sensitivity, the reference Coprecal process is generally more complicated and has more process equipment than the Oxalate (III) process (excluding any recycle). For instance, the number of accountability instruments required by Coprecal also is large when compared to the Oxalate (III) process. It is estimated that 63 neutron detectors are required for the Coprecal process, most of which must operate in a high-temperature environment for holdup measurements, whereas 19 neutron detectors operating at room temperature are required for the conversion process. Special instrumentation problems are also encountered in Coprecal because of the high operating temperatures of the calciners and filters, and the geometry and size of some process vessels, such as the large calciner primary filters (annulus 7.6 cm thick, 3.5 m o.d., 1.2 m high).

This analysis is restricted to the materials accountability portion of a safeguards system for a free-standing facility based on the reference Coprecal coconversion process. Total installed cost of such a system is estimated at $9.2 million and the annual operating cost is expected to be somewhat less than one-third that amount. Costs would be significantly less if the conversion process dynamic accountability system were incorporated into the overall safeguards system of a larger collocated nuclear complex of the type that has been suggested for both domestic and international commercial reprocessing facilities. In any case, these costs are a small fraction of the total plant capitalization, which has been estimated at a few hundred million dollars for the coconversion module of a multi-billion-dollar, collocated complex.

The relatively high overall materials balance uncertainties obtained for the reference coconversion process, including the feed-blend tanks, should not be regarded as an indictment of the basic Coprecal process or of coconversion in general. This lack of diversion sensitivity in the reference process is attributed to the particular process configuration and operating sequence chosen, which were not influenced by safeguards considerations until this study. Although increased process volumes and decreased plutonium measurement sensitivities are an inherent consequence of uranium dilution of the product, their deleterious effects should be considerably lessened in a process design that incorporates good safeguards practice. The reference Coprecal process is currently being modified in accord with the recommendations of this report and will be evaluated in a subsequent study.

CONTENTS

FIGURES

TABLES

COORDINATED SAFEGUARDS FOR MATERIALS MANAGEMENT IN A URANIUM-PLUTONIUM NITRATE-TO-OXIDE COCONVERSION FACILITY: COPRECAL

by

H. A. Dayem, D. D. Cobb, R. J. Dietz, E. A. Hakkila,
E. A. Kern, E. P. Schelonka, J. P. Shipley, and D. B. Smith

ABSTRACT

This report describes the conceptual design of an advanced materials-management system for safeguarding special nuclear materials in a uranium-plutonium nitrate-to-oxide coconversion facility based on the Coprecal process. Design concepts are presented for near real-time (dynamic) accountability by forming dynamic materials balances from information provided by chemical and nondestructive analyses and from process-control instrumentation. Modeling and simulation techniques are used to compare the sensitivities of proposed dynamic materials accounting strategies to both abrupt and protracted diversion. The safeguards implications of coconversion as well as some unique features of the reference process are discussed and design criteria are identified to improve the safeguardability of the Coprecal coconversion process.

I. INTRODUCTION

Conceptual designs of advanced materials-management systems for safeguarding special nuclear materials (SNM) in the three major components at the back end of the uranium-plutonium fuel cycle (chemical separations,[1] plutonium nitrate-to-oxide conversion,[2] and mixed-oxide fuel fabrication[3]) have been developed and evaluated. When these designs are combined with advanced physical protection systems developed by Sandia Laboratories, they provide effective safeguards concepts for future industrial-scale uranium-plutonium fuel-cycle facilities.

This report describes the first in a series of conceptual designs of advanced materials management systems for safeguarding SNM in proposed alternative fuel-cycle facilities. As in the previous studies, this conceptual design is based on measurement and control technology that has been demonstrated or that can be reasonably projected for the early 1980s. The purpose of the report is to identify concepts for dynamic materials accounting and control in a uranium-plutonium nitrate-to-oxide coconversion facility based on the Coprecal (coprecipitation and calcination) process adapted for light-water reactor fuel requirements.

The reference conceptual process design (Coprecal) chosen for this study was developed by the General Electric Company (G.E.) for the Alternative Fuel Cycles Technologies (AFCT) program. The basic process design (three parallel process lines) and throughput are similar to the plutonium nitrate-to-oxide conversion facility addressed in Ref. 2. As in the Oxalate (III) conversion process, the industrial-scale Coprecal process is in the conceptual design stage. Therefore, specific equipment has not been selected, process control measurements have not been specified, and holdup, scrap, and waste data are not available. Although all these are significant factors in the design of materials accounting and control systems, the conceptual design can be used to compare the similar portions of the Coprecal coconversion and the Oxalate (III) conversion process.

A. Conversion Processes

Nitrate conversion processes are essential to the commercial production of plutonium oxide for nuclear reactor fuels, either as pure oxide or coprocessed with a uranium diluent. The normal product of any spent-fuel reprocessing plant based on the Purex process or any of its variants is a concentrated aqueous solution of plutonium nitrate, $Pu(NO_3)_4$. Industrial-scale plutonium use is usually restricted to the metal or oxide, so efficient large-scale processes are necessary for converting the nitrate solution to the oxide, either for direct use or as an intermediate to metal production.

Various end uses require plutonium product properties that differ greatly and in fact may be mutually exclusive, leading to a profusion of conversion processes intended to optimize the properties required of the final product (see Ref. 2, Sec. I). Historically, the conversion (or reconversion) process has been treated as the initial step of a fuel-fabrication or metal-reduction sequence starting with the stored nitrate product of the separations plant. Regulatory changes effective in early 1978[4] prohibit the transportation of plutonium-nitrate solutions in the United States and require that conversion become the final step in the separations process rather than the initial step in fabrication.

This restriction introduces important modifications in the objectives of the conversion process and the properties desired of the oxide product. High purity, including freedom from decay products, remains a goal for the oxide product. However, decoupling of the conversion process from the end use of the product makes it impossible to provide the required purity, oxide powder morphology, ceramic activity, and chemical reactivity simultaneously using a single product-finishing step, unless the separations plant is coupled directly to a specific fabrication process.

Important reasons for storing and shipping plutonium-nitrate solutions include convenience in handling, blending, and analysis. This mode of operation also has facilitated selection of a conversion process most amenable to the end use and has kept the plutonium in a convenient form for purification from trace contaminations caused by storage and decay immediately before, or during, conversion.

Future major suppliers of plutonium or uranium-plutonium blends may need to convert, redissolve, and reconvert their product to meet both transportation requirements and product specifications. The consequence of delays between purification and use and the conflicting requirements of stability for shipping versus the necessary chemical and ceramic reactivity of the fabrication feed material may require redissolution and reblending of the oxide product at its destination. This step might be coupled with repurification of the nitrate solution before its reconversion to a customized fabrication feed material.

A conversion process must meet several stringent technical requirements. The product must have the appropriate purity, particle size, and morphology so that it can be blended and sintered to provide a homogeneous mixture that can be pelleted as a reactor fuel. Pellet density is critical because it determines the thermal conductivity of the fuel, which in turn controls such parameters as fuel melting, fission-product migration, plutonium and uranium redistribution, and reactor kinetics.

Several industrial-scale conversion processes have been reviewed in a previous report[2] that treats the safeguards aspects of pure plutonium-nitrate conversion processes in detail. Of the processes reviewed, only three, direct denitration, the Coprecal process, and the sol-gel process, are suitable for the coconversion of mixed uranium-plutonium nitrate solutions to mixed oxides. This report supplements Ref. 2 by treating coconversion as a separate subject, using a reference facility based on the Coprecal process.

Coprecal was conceived as a proprietary process by G.E. in 1969 to address specific requirements for producing homogeneous blends of mixed-oxide powders having controlled ceramic properties. The process (described briefly in Sec. I.C and in greater detail in App. A) was evaluated initially using uranium nitrate. A plutonium pilot plant having a heavy-metal capacity of 0.5 kg/h was built for further evaluation. In 1975 G.E. halted plutonium fuels development work because of uncertainties in the regulatory environment, and the Coprecal process was transferred to government ownership. Subsequent development, including uranium-plutonium start-up of the pilot line in June 1977, has been under US Government sponsorship.

Although the process was originally intended to address a purely commercial requirement, interest in Coprecal has been extensively renewed by safeguards and nonproliferation considerations. This interest is based on the premise that mixed uranium-plutonium powders and solutions afford some intrinsic protection against diversion that pure plutonium products do not afford. The coconversion process would also be directly compatible with chemical separations plants operated in a coprocessing mode. In this mode, pure plutonium would never be isolated and the mixed uranium-plutonium nitrate product from chemical separations would be feed material for Coprecal.

In the Coprecal reference process a uranium-plutonium-nitrate master-mix solution (10% Pu) is injected continuously into a concentrated ammonium-hydroxide solution, where it immediately precipitates as an unfilterable, thixotropic slurry. The entire slurry is pumped to a fluidized-bed calciner that converts it to mixed UO_3 and PuO_2 releasing nitrogen and water vapor (nitrates are reduced by excess ammonia). The dry powders are then batch-fed to a reduction-stabilization unit that reduces the UO_3 to UO_2 using an H_2-N_2 mixture and stabilizes it with CO_2.

The resulting mixed-oxide powder is homogeneous, very finely divided, and ceramically reactive. It dissolves readily in HNO_3; however, the uranium cannot be separated from the plutonium solely by this technique. Ease of dissolution is particularly important because it allows the process sequence to be repeated conveniently to satisfy solids shipping requirements, to permit solution blending of subsequent custom blends, and to modify ceramic properties as needed. For these reasons the Coprecal process could be used to couple continuous fuel reprocessing and fabrication facilities or as the product end of a reprocessing plant and the head end of a remotely located fuel fabrication facility.

Coprecal does not provide the liquid-solid phase separations characteristic of other precipitation-based conversion processes, hence all nonvolatile materials that are dissolved and suspended in solution are retained in the product. Therefore, unlike most other precipitation conversion processes, Coprecal affords no additional purification of the feed solution, a disadvantage in the case of off-specification and aged material containing americium and other daughter products. Any contaminants of this type can be eliminated only by redissolution and repurification of the mixed oxide. However, the absence of a liquid-solid phase separation can be regarded as a process advantage in Coprecal because it eliminates the need for filtrate recycle.

B. Safeguarding Conversion Processes

Plutonium-nitrate conversion processes provide a unique safeguards challenge. Unlike the situation that exists at other points in the fuel cycle, input, in-process, and product materials are all attractive, high-purity, concentrated targets for diversion, unhampered by high-level radiation, heavy shielding barriers, or impractically low concentration levels. Also, the conversion process naturally tends to become the process buffer between the loosely coupled functions of plutonium separation (chemical reprocessing) and plutonium use (fuel fabrication) that characterize the commercial fuel cycle. In this buffer function the conversion process is bracketed physically by significant inventories of extremely attractive feed and product materials. These materials have the greatest diversion potential in any domestic fuel cycle and are an important proliferation risk in the nuclear fuel cycle of any nonweapons state.

The pivotal role of the conversion facility in a safeguarded fuel cycle suggests that any enhanced safeguards or nonproliferation strategy first should be applied directly to the conversion plant and subsequently expanded to include the adjacent functions of separation (or coseparation) and fuel fabrication, starting with the critical areas of product and feed storage and inventory control. This could be done best in future facilities by expanding the conversion facility to include product storage for the separations plant, solution blending or early dilution if plutonium partitioning is used, coconversion, custom blending of mixed-oxide powders, and feed storage for the fabrication plant. Collocation of these crucial functions under a single controlling authority in a facility inside or contiguous to the separations plant has been suggested in the "Bonded Crucial Facility" (BCF) concept proposed as a nonproliferation strategy for foreign fuel cycle facilities[2] (Fig. 1). In this way, the safeguards controlling authority can monitor and verify production and consumption rates and can maintain cognizance of the disposition of all fissile products produced by the complex, thus ensuring that no significant quantities of undiluted plutonium leave the complex undetected.

Coconversion, as in the reference Coprecal process, is directly compatible with the BCF nonproliferation strategy, as well as with the requirements of chemical separations facilities in which coprocessing is used for enhanced safeguards. The major safeguards advantage of coprocessing and coconversion is early dilution, which requires that more

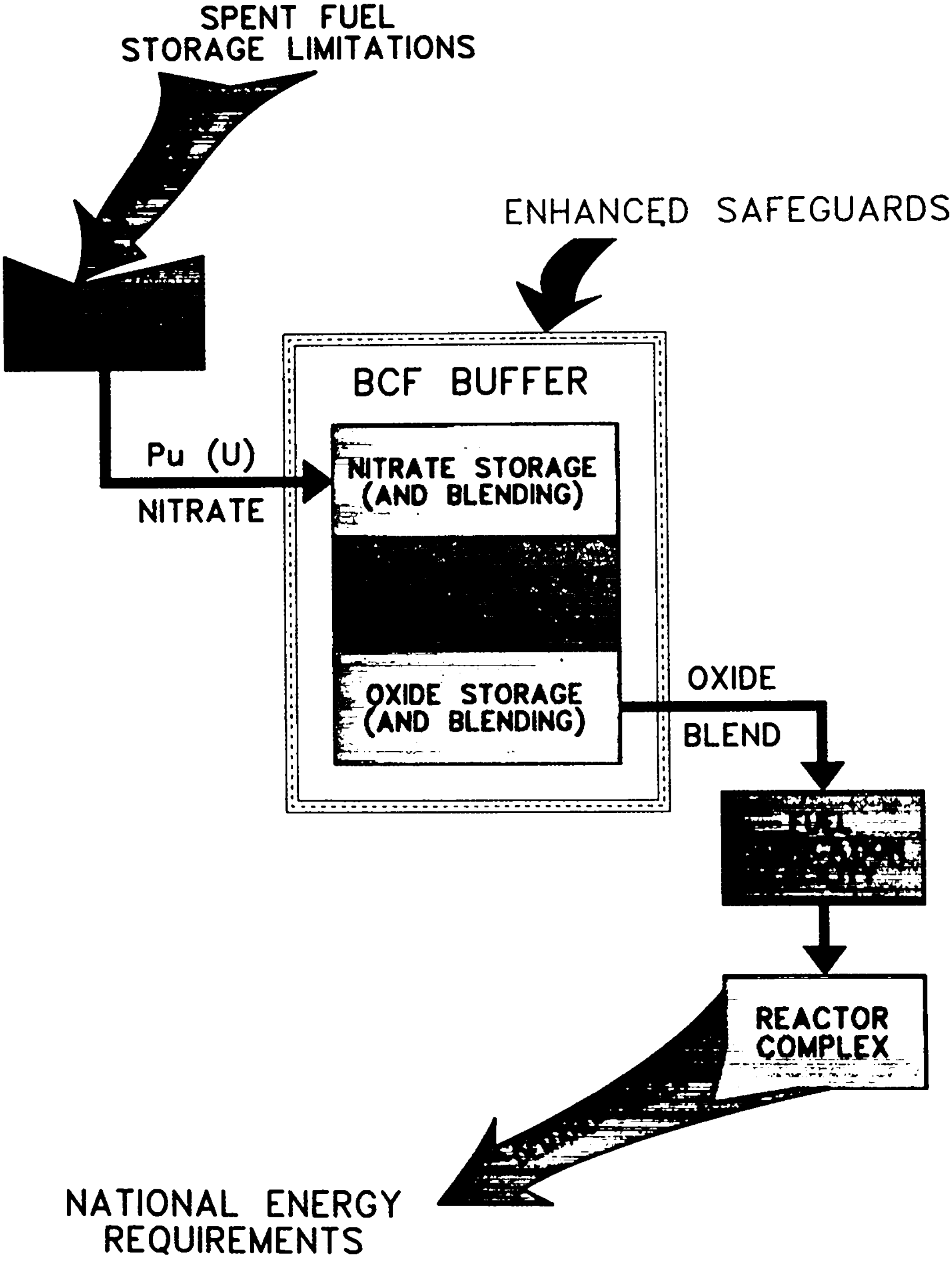


Fig. 1.
The bonded crucial facility.

material (a factor of 10 in the reference facility) be diverted to obtain significant quantities of weapons-usable materials. These materials must then be subsequently processed to separate the plutonium.

Coprocessing in any of its forms provides more assurance against domestic diversion threats than against national proliferation. When national resources can be applied to the repartitioning of any mixed product stream, as by several of the conventional conversion processes discussed in Ref. 2, the benefits of coprocessing are seriously reduced.

C. The Reference Coconversion Facility

The reference nitrate-to-oxide coconversion facility was designed by G.E. for the continuous coprecipitation and calcination of a blended uranium- and plutonium-nitrate solution.[5] The facility was designed to convert 117 kg of plutonium per day: the 100-kg plutonium product from a 10-tonne-per-day Light Water Reactor (LWR) chemical separations facility plus 17 kg of plutonium (as a uranium-plutonium-nitrate solution) from scrap recovery.

Three parallel process lines are required to meet the design-basis throughput in reasonably sized, criticality-safe process equipment. Figure 2 shows a simplified block diagram of the process. Table I lists the flow rates for each stream integrated over all three process lines for the continuous portion of the process, and Table II lists the amount of material transferred per batch for the batch portion of the process. Each line comprises three feed-blend tanks, a single precipitator, four calciners, a single primary filter, a single secondary filter, four parallel reduction-stabilization stations each containing a primary and a final filter, and a single screening station.

Uranium-plutonium solution is received from the solvent-extraction facility as a concentrated nitrate solution containing ∿400 g of heavy metal/L. The uranium-to-plutonium ratio in the solution is nominally 8 to 1 and is not less than 5 to 1. Before coconversion the solution is blended with a natural-uranyl-nitrate solution (400 g uranium/L) to obtain a 10% plutonium solution.

Three parallel feed-blend tanks are required for each process line to permit the continuous processing of large homogeneous batches, which is an important design feature of Coprecal. Each of the nine tanks has a working volume of 16 000 L and can feed a single process line for ∿2 weeks (one campaign).

One feed-blend tank continuously feeds the precipitator where ammonium hydroxide is added to produce a slurry of ammonium diuranate and plutonium hydroxide. The precipitator is a cylinder 15 cm in diameter and 2.5 m high with a 35-L minimum

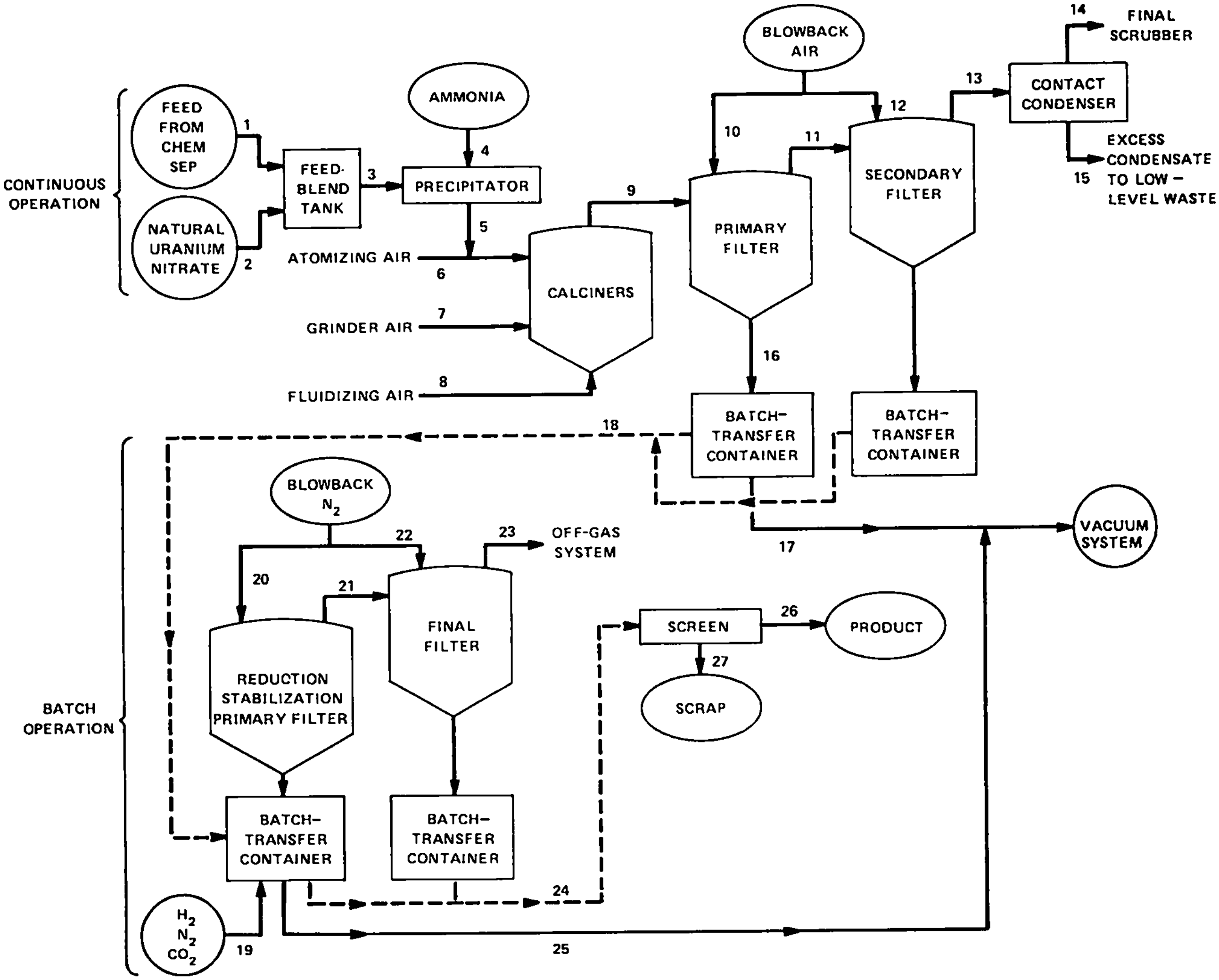


Fig. 2.
Coprecal simplified flow diagram.

working volume. A short residence time and high pH are sufficient in Coprecal to produce a finely divided slurry, in contrast to the usual precipitator goals of promoting crystal growth and producing a filterable material.

The entire slurry from the precipitator is fed continuously to a fluidized-bed calciner system to produce mixed uranium-plutonium-oxide powder. Each calciner system contains four parallel manifolded fluidized beds (15 cm in diameter and 1 m high). The slurry metered into each bed is calcined to a powder; the powder is eluted from the bed by the fluidizing and decomposition gases and steam, which carry it to the primary filter.

TABLE I

COPRECAL CONTINUOUS FLOW STREAMS

Stream No.[a]	Total Flow Rate[b] (g/min)	Plutonium Flow Rate[b] (g/min)
1	397 500[c]	12 000
2	397 500[c]	---
3	3 209	81.27
4	1 288	---
5	4 498	81.27
6	222	---
7	232	---
8	1 054	---
9	5 998	81.27
10	3[d]	---
11	5 031	0.04
12	0.3[d]	---
13	5 031	trace[e]
14	1 968	trace[f]
15	3 064	trace[g]
16	970	81.27
17	1	0.09
19	5 897	---
20	39[d]	---
21	5 981	0.04
22	39[d]	---
23	6 020	0.004

---

[a] See Fig. 1 for stream identification.

[b] Flow rates may not balance exactly because of roundoff error.

[c] Flow rate when material is recovered from chemical separations.

[d] Average flow rate--periodically pulsed to blowback filters.

[e] 0.60 g/day.

[f] 0.25 g/day.

[g] 0.35 g/day.

TABLE II

COPRECAL BATCH TRANSFERS

Stream No.[a]	Total[b,c] kg/batch	Plutonium[b,c] kg/batch
18	38.79	3.25
24	36.66	3.23
25	0.26	0.02
26	36.38	3.21
27	0.28	0.02

---

[a] See Fig. 1 for stream identification.

[b] Flow rates may not balance exactly because of round-off error.

[c] A batch is processed every 40 min.

The four calciners discharge into a single primary filter. The filter contains 43 elements made of porous Inconel metal with a 5-μm rating. The primary filter is an annulus 7.6 cm thick and 1.2 m high with a 3.5 m o.d. Powder collected on the filter is removed by periodically blowing back a few elements at a time. The powder collects in the bottom of the filter chamber and is discharged approximately every 0.5 h through a valve to a batch-transfer container. The batch-transfer container is removed and sent to a reduction-stabilization station every 2 h. From this point the process line operates in batch mode.

Gas from the calciner primary filter is discharged into a secondary filter similar in construction to the primary filter. The powder content of the entering gas is very low; therefore, the batch-transfer container from the secondary filter is removed only once every operating campaign.

Gas from the calciner secondary filter is discharged to a contact-condenser and a gas-scrubbing system and then is discharged to a final scrubber. Excess condensate is discharged to the low-level waste treatment portion of the reprocessing complex.

The batch-transfer container filled with mixed UO_3-PuO_2 powder is transferred from the primary filter to one of four parallel reduction-stabilization stations. Each reduction-stabilization station contains a primary and a final filter. These filters are cylindrical chambers 30.5 cm in diameter and 1.2 m high. They contain 11 elements similar to those in the calciner filters.

The batch-transfer container is connected to the reduction-stabilization station where gas is passed up through the powder at velocities sufficient to "turn over" the powder. During reduction-stabilization, UO_3 is reduced by hot hydrogen gas (6% H_2 in N_2) to UO_2 and is stabilized using hot CO_2 to produce an equilibrated powder ($UO_{2.07}$) that does not readily reoxidize in air. The product is then cooled to room temperature by CO_2. Almost all the powder entrained in the gas is removed by filters that are blown back periodically. Gas from the primary reduction-stabilization filter is discharged through a final filter to the off-gas treatment system. At the completion of the reduction-stabilization cycle (8 h), the batch-transfer container is removed and transferred to the screening station. The transfer container on the reduction-stabilization final filter will probably be removed at each physical inventory.

At the screening station, the stabilized powder is removed from the batch-transfer container and passed to a screen system to remove any foreign particles and oxide agglomerates exceeding 0.15 mm in diameter. The powder passing through the screen is collected in a tared storage can. When all powder from a transfer container has been screened, material on the screen is dumped into a scrap container, which is sent to scrap recovery after filling. The product storage can is removed, sampled, weighed, sealed, and transferred to a storage vault.

II. SAFEGUARDS SYSTEMS

A . Introduction

Current trends in safeguards systems development result from the perceived threats of nuclear materials diversion and the technology available to combat those threats. Both the perceived threat and the technology are increasing in scope and sophistication. To be effective, safeguards systems must address the entire spectrum of threats, requiring that the most relevant safeguards techniques be applied to each diversion threat. From the points of view of both safeguards and the economics of plant operations, it is vitally important that a coherent approach be taken to designing safeguards systems that are capable, make optimum use of the technology, and interface positively with the host facility. The coherent systems approach, with its implied tradeoffs between opposing requirements, can assure safeguards effectiveness without resorting to a hodgepodge overlay of every conceivable countermeasure against every imaginable threat.

This section describes the general features and structure of a coherent domestic, or State's, safeguards system. In addition, the topics of international safeguards and interactions between a host nation and the International Atomic Energy Agency (IAEA) are discussed briefly.

B. Structure of Domestic Safeguards Systems

Any comprehensive safeguards strategy includes four functions:

- exclusion of all unauthorized persons from the facility and selective exclusion of others from sensitive areas within the plant;
- monitoring of all activities involving SNM to determine whether each activity is consistent with safeguards requirements and with normal, expected facility operation;
- accounting for all SNM in the facility to determine whether the correct amounts of all materials are present in their proper locations; and
- response to and reporting of the facility's safeguards status.

At the same time, the safeguards system must not violate any of the following constraints.

- It must be economical.
- It must be based on current technology or reasonable extrapolations thereof.
- It must not disrupt process operations unnecessarily.
- It must not compromise safety or infringe on employee working conditions.

These constraints support the principle that the fundamental purpose of any nuclear fuel cycle facility is to process nuclear material. Safeguards are vitally important, but the relationships among sometimes conflicting objectives must be kept in perspective. Coordination between process and facility designers and safeguards system designers at the earliest stages forms the most effective means of achieving this goal. The following describes a systems structure developed through numerous interactions with the nuclear industry and the safeguards community.

The basic management, control, and coordination structure of safeguards systems for domestic nuclear fuel cycle facilities has been described in earlier reports.[1-3,6-10] The general block diagram of a facility and its safeguards system is shown in Fig. 3. Functions directly related to the process are enclosed in heavily outlined boxes.

The safeguards system, through the safeguards coordination unit (SCU), and the plant management are responsible to a higher authority: the Department of Energy (DOE)/Office of Safeguards and Security for DOE-operated facilities and the Nuclear Regulatory Commission (NRC) for commercial facilities. The safeguards system must (1) provide timely, accurate reports on the safeguards status of the facility, (2) implement safeguards requirements imposed by the regulatory authority, and (3) initiate and coordinate external responses to possible safeguards breaches. In the international environment, the safeguards system must interact similarly with the IAEA and with the host nation's regulatory body.

An effective safeguards system comprises several subsystems, including (1) the safeguards coordination unit, (2) the physical protection system (PPS), (3) the materials measurement and accounting system (MMAS), (4) the process monitoring system (PMS), and (5) the safeguards computer system (SCS). These subsystems and the related facility functions of process control and plant management are discussed below.

1. Safeguards Coordination Unit. The SCU supervises SNM safeguarding in the facility. As the focal point for safeguards decisions, the unit interacts with management and the process-control coordination unit (PCCU) to ensure effective safeguards while minimizing process disruptions. The SCU has three primary functions: (1) data collection and processing, which are required for (2) safeguards condition assessment, which in turn is the basis for (3) the response determination decision. A structural diagram of the SCU is shown in Fig. 4.

To make good decisions, the SCU must have access to all pertinent safeguards data, management input, and process-control information. Current safeguards data are available from the PPS, PMS, and MMAS, and the safeguards operating history is stored

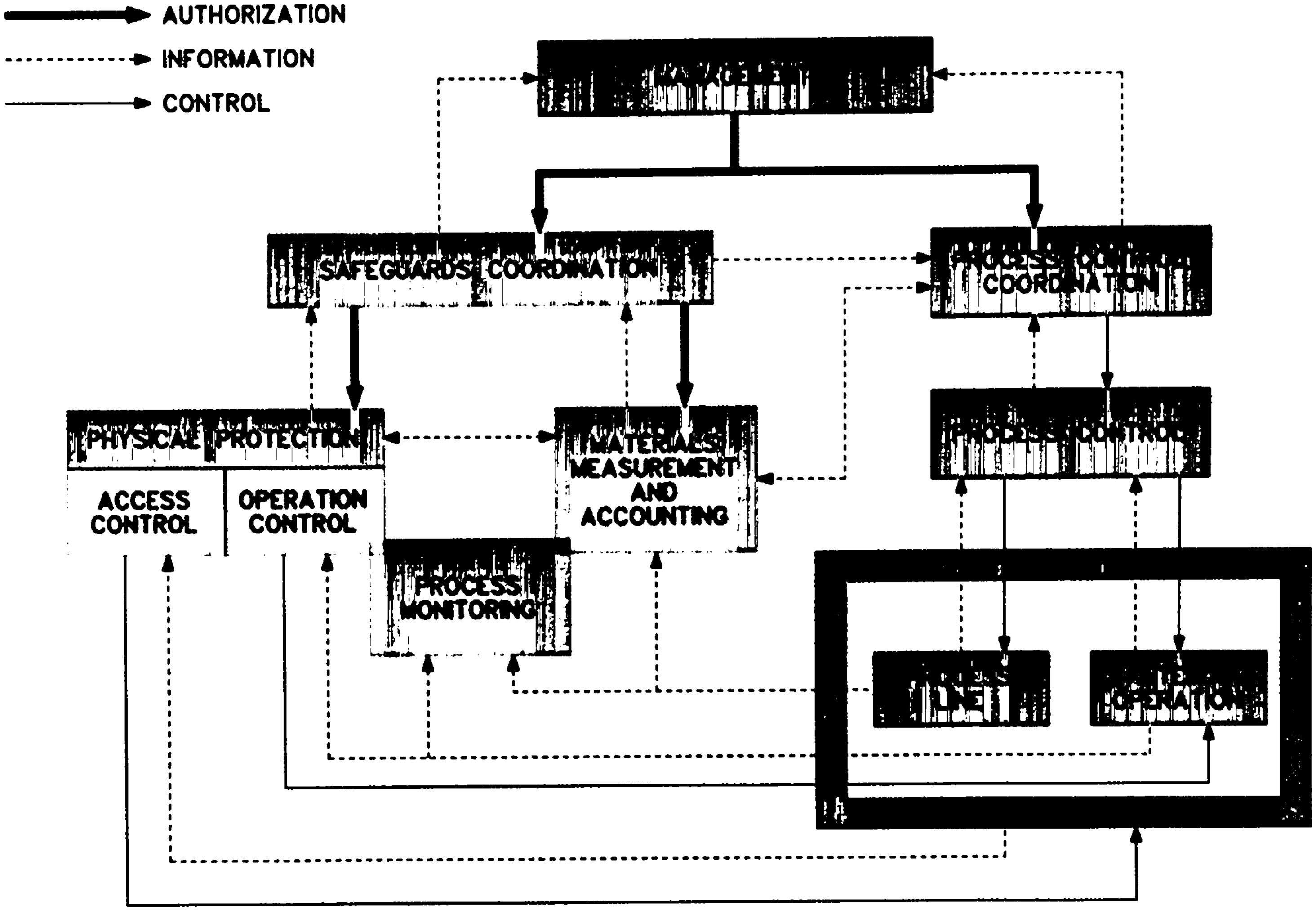


Fig. 3.
Structure of the safeguards system.

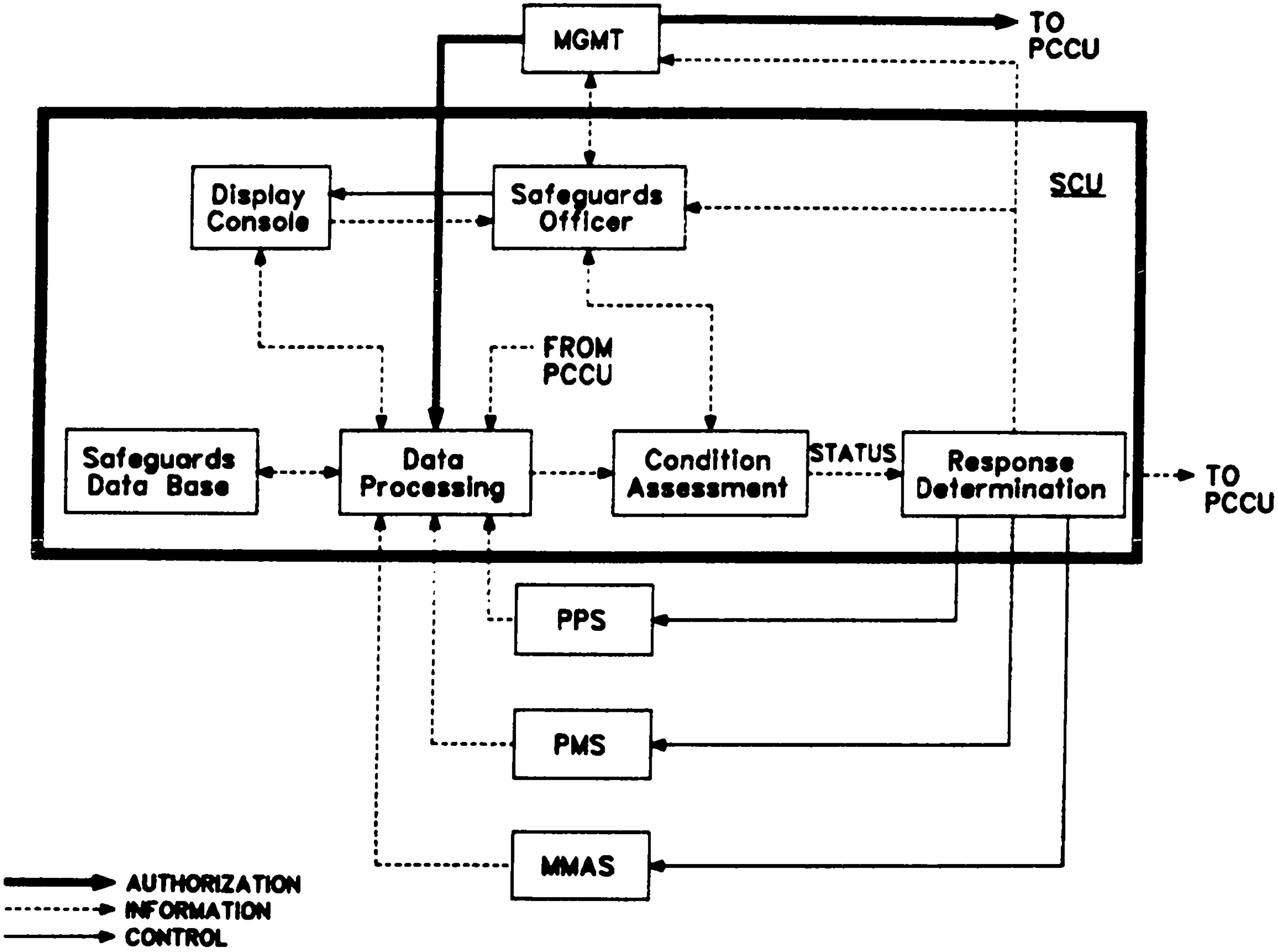


Fig. 4.
SCU structure.

in the data base of the safeguards information system. All management authorizations are sent in parallel to the SCU and to their primary destinations, and the process-control data summary can be acquired from the process information system.

Much of the information available to the SCU is not in a form suitable for decision making. The data-processing block summarizes and formats the data for output at the display console. In addition, it generates the safeguards reports required by management and the regulatory authority.

The Safeguards Officer uses the processed data to assess the plant condition. The result of the condition assessment is the plant's status with respect to safeguards requirements. Many minor condition assessments can be automated with manual override, but others require direct action by the Safeguards Officer. Incoming SCU data are machine monitored for possible indications of status changes (for example, by observing materials imbalances or equipment failures), and a response to each possible change is requested from the Safeguards Officer. He then evaluates the plant safeguards status and determines appropriate responses for the PPS, PMS, and MMAS, taking into account process operations. These responses are transmitted to management and the PCCU, along with recommendations for modified process authorizations.

As many status/response situations as possible are standardized in a manual of operating procedures. This manual (or appropriate portions thereof) is available to plant personnel on a need-to-know basis. Of course, standard procedures may not have been written for many situations, and that is the main reason for the Safeguards Officer. This scheme combines the efficiency of a machine in data handling with the adaptability of a human supervisor in making decisions.

In assessing the plant condition, the Safeguards Officer uses reports from various parts of the plant. Taken together, these status reports constitute the plant status, which is stored in great detail in the safeguards and process information systems. However, the information normally displayed to the Safeguards Officer is condensed for quick assessment, with nonstandard situations flagged to indicate areas that should be investigated. The Safeguards Officer can ask for more detail, either in response to a flag or of his own volition.

The SCU recommendations can range from no recommendation to the extremes of process shutdown and plant evacuation. If the safeguards system is successful, most frequently there will be no recommendation at all. In abnormal safeguards situations, the course of action is chosen by agreement among the Safeguards Officer, plant management, and PCCU, although some responses may be dictated by regulation. In emergency situations, the PCCU must act quickly to prevent safety problems and must inform plant management and the SCU of his actions.

2. Physical Protection System. The PPS controls personnel entry and exit for the facility and for restricted areas inside. The system includes automated equipment and enough guards to provide effective response in an emergency. It expands the conventional security functions, such as access control, to include control of item-handling operations (operations control). This arrangement provides more effective protection through remote, overriding control of discrete material items in handling and storage. The concept is applied only to those portions of the facility that are outside the closely coupled process line, such as the load-out area, where materials flow is not critical to smooth process operations.

The PPS provides appropriate information to the safeguards system and it

- excludes all unauthorized persons and contraband from the facility,
- admits only essential persons to selected areas, and
- precludes unauthorized activities involving nuclear material and vital equipment.

Important objectives in the PPS design are to automate its functions whenever possible and to harden the system against subversion. These objectives are consistent with the stated goal of reducing dependence on personnel reliability.

In the current concept of domestic safeguards, physical protection and materials accounting complement and reinforce each other. In particular, the PPS protects not just nuclear material, but the integrity of the MMAS as well. Conversely, the MMAS confirms the effectiveness of the PPS. For international safeguards, the PPS is part of the hostile environment and might be used actively to subvert the materials accounting and containment/surveillance measures that form the backbone of IAEA safeguards. This fact mandates an IAEA safeguards system that compensates, perhaps through self-protection techniques, for the potential deficiencies or malevolence of the PPS.

The design and evaluation of the PPS for these facilities are the responsibility of Sandia Laboratories. This area is discussed in detail in Refs. 7-9.

3. Materials Measurement and Accounting System. The MMAS is an implementation of the DYMAC concept[11,12] and is similar in principle to that already described for several types of facilities.[1-3] It combines conventional chemical analysis, weighing, and volume measurement with the near-real-time measurement and surveillance capabilities provided by nondestructive assay (NDA) instrumentation to enhance rapid and accurate assessment of the location and amount of SNM in a facility.

Three major MMAS functions in SNM accounting are

- data collection (including measurements),
- data analysis (for diversion detection), and
- data dissemination or reporting.

As currently performed by conventional safeguards inventory control, these tasks rely heavily on materials balance accounting following periodic shutdown, cleanout, and physical inventory. The classical materials balance associated with this system is drawn around the entire plant or a major portion of the process. The balance is formed by adding all measured receipts to the initial measured inventory and subtracting from that sum all measured removals and the final measured inventory. During routine production, materials control is vested largely in administrative and process controls, augmented by secure storage for discrete items.

Although conventional materials balance accounting is essential to safeguards control of nuclear material, it has inherent limitations in sensitivity and timeliness. Sensitivity is limited by measurement uncertainties that desensitize the system to losses of trigger quantities of SNM for large-throughput plants. Timeliness is limited by the frequency of physical inventories. There are practical limits to how often a facility can shut down its process and still be productive. On the other hand, periodic conventional physical inventories including process shutdown and cleanout may be necessary to establish reference points for the dynamic accountability system.

Recently developed NDA technology, state-of-the-art conventional measurement methods, special in-plant sensors, plant instrumentation signals, and the most effective statistical data-analysis techniques combined with supportive computer and data-base-management technology make several more effective methods feasible. For example, conventional methods can be augmented by unit-process accounting,[1-3,11,12] where the facility is partitioned into discrete accounting envelopes called unit-process accounting areas (UPAAs). A unit process can be one or more chemical or physical processes and is chosen on the basis of process logic and whether a materials balance can be drawn around it. By dividing a facility into unit processes and measuring all significant materials transfers, quantities of material much smaller than the total plant inventory can be controlled. Furthermore, any discrepancies are localized to that portion of the process contained in the UPAA.

Materials balances drawn around UPAAs during plant operation are called dynamic materials balances to distinguish them from balances drawn after a cleanout and physical inventory. Ideally, all dynamic materials balances would be zero unless nuclear material had been diverted. In practice, they are never zero, for two reasons. First, measured values are never exact because of the errors inherent in any measuring procedure. Second, constraints on cost or effects on materials processing operations may dictate that not all components of a materials balance be measured equally often; therefore, even if the measurements were exact, the materials balance values would not be zero until closed by additional measurements. In the interim, it is sometimes possible to use historical data as estimates of unmeasured material and then to update the estimates when additional measurements become available.

A consequence of unit-process accounting, the concept of overlapping UPAAs, derives from the fact that two adjacent UPAAs can be combined to form one larger UPAA, eliminating the intervening transfer measurement. This combination may be useful for cross checking, and it allows continued materials accounting (at reduced sensitivity and timeliness) if instruments malfunction.

These ideas must be used flexibly throughout the facility. Their application should be graded according to the safeguards strategic value and vulnerability of the material, hence, the concept of graded safeguards. For example, in the coconversion facility, uranium-plutonium-oxide powder at the product load-out area would be somewhat more desirable to a potential divertor than the relatively dilute (∿40 g plutonium/L, ∿400 g heavy metal/L) uranium-plutonium-nitrate solution in the feed-blend tanks, and both would be more desirable than the slurry (∿25 g plutonium/L) inside the precipitators.

The Coprecal MMAS is described in more detail in Sec. III.

4. Process-Monitoring System. The chemically hostile environment of a conversion plant requires that instrumentation be designed for physical ruggedness and reliability, which often significantly compromises measurement ability. Thus, there is a need for a limited set of on-line, plant-grade measurement equipment and other simple, reliable, process-monitoring devices specifically designed to detect an abnormal situation, with less regard for the measurement precision and accuracy traditionally required for materials accounting.

The PMS combines elements of both the PPS and MMAS and provides supplementary information to each regarding compliance of actual process operating modes with approved procedures. The concept may be regarded as an extension of physical protection monitoring and surveillance functions into the process line, and as an upgrading or appropriate placement of the monitoring devices to allow gross materials accounting.

The PMS collects timely information to detect a theft in progress. The system uses plant instrumentation wherever possible to assess approximate materials balances on transfers between tanks and across vessels. Similarly, an overall plutonium balance can be maintained. This balance is crude by accounting standards but has the advantage of near-real-time availability.

To illustrate, consider a typical process tank that has an inlet, an outlet controlled by a valve and transfer jet, a liquid-level probe, a specific-gravity probe, a sampler, and an air-sparge line.[13] All valve positions are monitored, pressures in all transfer-line pressures are measured, and each line has an SNM sensor (a simple go-no-go device). The PMS checks to see that all these variables are behaving in accordance with approved procedures. For example, a drop in tank level when all valves are closed, or a negative pressure in a probe line, would indicate an attempt to remove SNM from the tank.

Such a system can provide nearly immediate detection of diversion attempts by continuously comparing actual operating conditions with those expected. However, it must always be supplemented by materials accounting to indicate how well it has worked during the last accounting period. This accounting is especially important if there has been some malfunction or if some part of the PMS has been subverted.

5. Safeguards Computer System. The SCS plays an essential role in implementing effective safeguards by collecting safeguards-related data and maintaining and controlling the safeguards information system. A major part of this role is the protection of SNM; an equally important part is the operational effect of the computer system on the processing of nuclear material. This effect occurs because information provided through the computer system forms the basis for all safeguards decisions, which may have varying degrees of effect on the process. Erroneous or unavailable information can degrade decision quality and cause unnecessary process disruptions. Thus, the reliability and integrity of the computer system directly affect economical operation of the process.

In conventional safeguards systems, filling out and transmitting many materials accounting forms requires much time and effort. These forms include records of receipts, shipments, internal transfers, and accounting measurement data. There are numerous possibilities for human error, either unintentional or malicious, and inefficiency in data management is unavoidable.

Such problems can be alleviated by implementing the MMAS through computerized data acquisition and data-base management, with the conventional system retained as a backup in case of malfunction. Under this scheme, most instruments are interfaced directly to the SCS, and the use of computer terminals for data input is minimized. Security problems are eased somewhat, and a self-verification capability is provided easily by designing instrumentation for periodic on-line recalibration under computer control. Data from sources not connected to the SCS may still be entered through a minimal number of terminals.

The SCS acts as the central data manager for the MMAS and serves as a powerful tool in analyzing accounting data for possible SNM diversion. It generates all permanent records of materials quantities, locations, and movements and, on demand, can provide reports required for effectiveness assessment, an assessment of the current status of the safeguards system, and other relevant information. In addition, the SCS interacts with the other safeguards elements to ensure most efficient use of all safeguards data. Furthermore, data may be exchanged as required with the PCCU to improve both safeguards and process control.

If all elements of the safeguards system are assisted by computerized operations, their combined capabilities can improve security and reliability significantly. Possible configurations for the computer network are discussed in Refs. 3 and 14, and Secs. II.D and II.E. discuss security and reliability. In addition, Refs. 15 and 16 report recent work in this area by the TRW Defense and Space Systems Group.

6. Process Control. The primary requirement of process control is optimal operation of the process line. This requirement has three important aspects: (1) economical operation, (2) health and safety considerations, and (3) effective safeguarding of SNM.

Obviously, the economic factor must be emphasized in a privately operated commercial facility. Economy of operation is accomplished by designing the process control to maximize throughput while consuming minimum resources. This condition requires that the process be operated at the design rate with as few interruptions as possible.

The adequate safeguarding of SNM could have an adverse impact on the economical operation of the plant. Consider the case of a small amount of plutonium lost from the process at some intermediate point. Normal process control would probably ignore this event; indeed, it would probably be unaware of it. However, the safeguards system, through its MMAS, is expected to detect the loss and ask for an investigation, and some interruption of operations may be necessary while the source of the problem is determined. If the loss is the result of malicious, purposeful diversion of SNM or is a false alarm, process control tends to regard the interruption as a nuisance. On the other hand, if the loss is benign (for example, an equipment malfunction or SNM leak), process control receives useful and otherwise unavailable information from the safeguards system. This is also true during normal operations because the MMAS monitors the materials flows in greater detail and with more accuracy than the process-control system. The materials flow data are accessible to process control through the adaptive data link to the MMAS (Fig. 5).

These considerations result in a slightly different concept of process control than that normally followed. The process-control function must be willing and able to interact with the health and safety and safeguards functions so that all three operational requirements are satisfied.

The process line is divided into several unit processes, each having its own unit process controller (UPC), which reports to one of the control subsystems (Fig. 5). UPC actions are supervised by PCCU. In addition to the control subsystems, the PCCU also contains other subsystems not directly concerned with process control.

The PCCU is responsible for the coherent operation of the entire process line--it performs a supervisory function. It determines operating levels and sequences for each UPC so that they all work together. This form of hierarchical control, called set point control, is the traditional method of controlling complex systems. It has proved effective and is well understood in the process-control field.

The control hierarchy allows each UPC to be devoted to one relatively simple unit process, thereby simplifying the UPC design because interactions with other unit processes are handled by PCCU. The control may be manual, automatic with manual override, or a combination of these.

The PCCU is also responsible for implementing safeguards-related recommendations that affect process operations. This implementation is necessary to ensure effective compliance with both safeguards and process control. The MMAS and the PCCU also exchange process-related information to improve process operation and safeguards effectiveness.

7. Plant Management. The plant management structure is straightforward and similar to traditional configurations. The major difference is the presence of the safeguards system, with the requirement that the plant and its management be responsive to safeguards considerations. Figure 6 shows the management structure and the authorization, information, and control data paths.

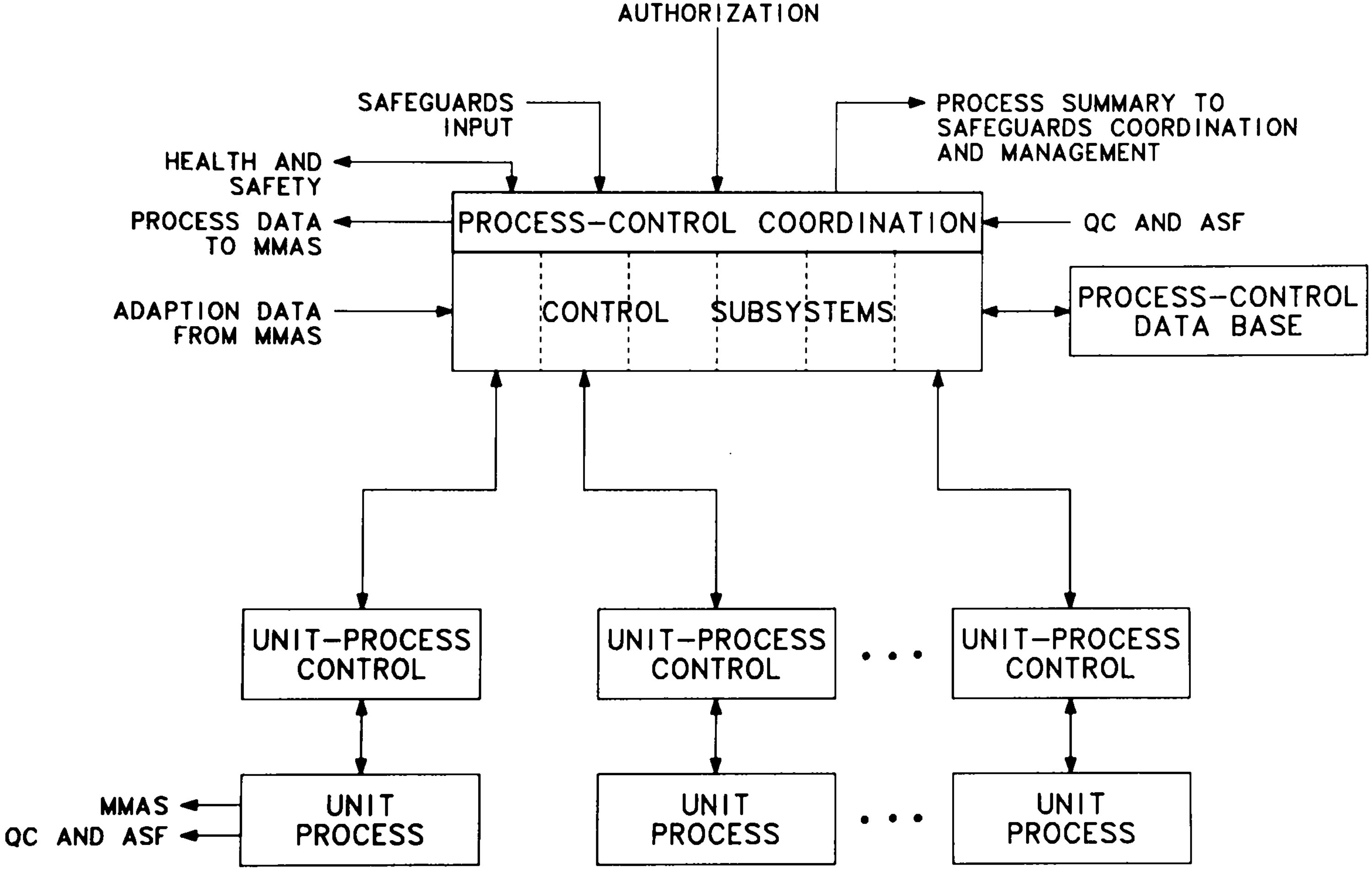


Fig. 5.
Process-control hierarchy.

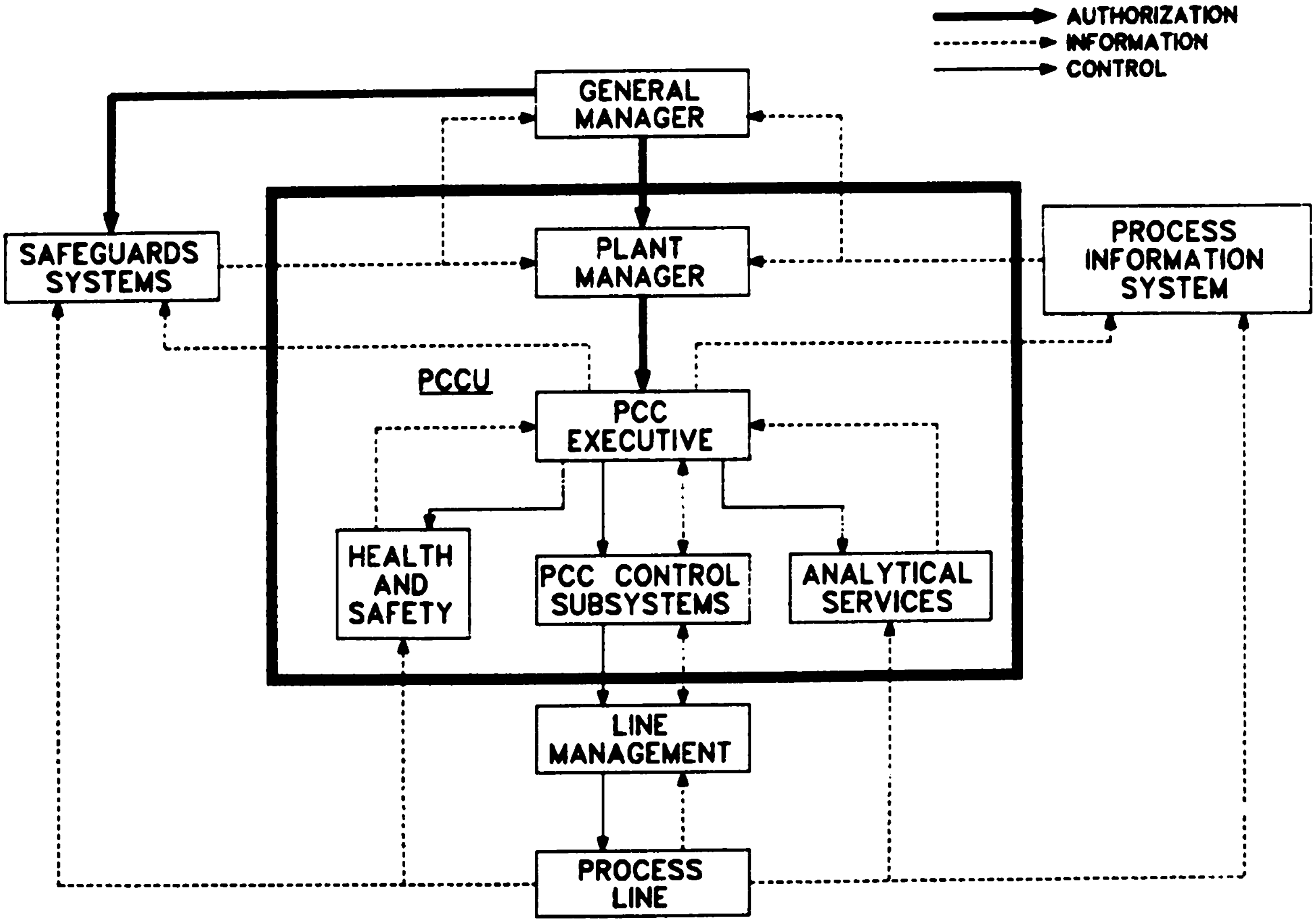


Fig. 6.
Plant management structure.

Management operations consist of the following steps.

(1) The general manager decides on a fuel contract and authorizes processing.

(2) The general manager's fuel projects staff defines the scope and issues a work order to the plant manager.

(3) The plant manager and his staff, using the process information system, plan and schedule the work load.

(4) The plant manager combines the work plan with any safeguards constraints and authorizes the start of work. A shop order describes the technical requirements and scheduling. This same information is transmitted to the safeguards system.

(5) The plant manager continually reviews the shop order status and safeguards system input and initiates any necessary action.

(6) Line management, with the help of the PCCU, organizes the work load and executes process operations on the basis of feedback from the process line, quality control (QC), analytical services facility (ASF), and health and safety, and on the basis of data from the process information system.

(7) QC maintains surveillance of product quality, analytical instrumentation calibration and inspections, and analytical data.

(8) Health and safety continually monitors all plant safety requirements, including criticality-related materials transfers.

(9) The safeguards system continually assesses the plant status and makes action recommendations to the general manager and plant manager in case of possible safeguards breaches.

C. Systems Tradeoffs

The obvious functional overlaps among the safeguards system components and between the safeguards and process-control systems lead naturally to consideration of possible tradeoffs. Certainly, increased capability in one system can be partially compensated by requiring less capability in another, but the relationship is not simple, nor does a given level of safeguards effectiveness have a corresponding unique system configuration.

Clearly, all components discussed in Sec. II.B are necessary in an effective safeguards system. Emphasis on one component does not preclude the need for another. For example, it cannot be assumed that all the nuclear material "is in there somewhere" just because none has been observed leaving through an elaborate perimeter PPS. An efficient materials accounting system must give assurance that the PPS is working. On the other hand, the balance between the several subsystems may depend on the requirements of a particular process area.

For example, containment and surveillance, including the presence of an inspector and video tapes from a closed-circuit TV system, are important in all parts of the process, but they are probably most effective in the shipping, receiving, and product storage areas. It is particularly important that bulky items, such as waste containers and decommissioned equipment, be examined carefully before removal. Monitoring of process variables for unauthorized or nonstandard materials movements best complements materials accounting in those areas where materials balance uncertainties are larger than desirable or where SNM resides as discrete items.

The possibility of instrument tampering requires that all safeguards instrumentation be hardened and tamper-indicating. This means that direct safeguards (or IAEA) supervision of the measurement control and maintenance programs is necessary.

The PPS (or containment and surveillance system) also assists in detecting instrument tampering through direct observation of the instrument environment and the use of seals. This task is much easier if most instruments are automated and interfaced directly to the materials accounting computer system; in that case, the plant operator has no reason to interact with the instrument, except for maintenance and calibration, in which safeguards personnel would also participate.

Much of the instrumentation required for materials accounting and process monitoring (or containment and surveillance) would also be useful for process control and, in fact, would significantly expand process-control capabilities. Likewise, the historical data on process operations maintained in the safeguards data base have proved to be important to the process, as shown during the brief operation period of the DYMAC demonstration in the new Plutonium Processing Facility at Los Alamos.[17-19]

Therefore, it may be desirable for the safeguards system to provide some of the necessary process-control measurements and the historical process operating data. Process control would need some means of assurance that the required measurements would always be available. This step would lift some capital and overhead burden from the facility operator and would constitute a direct benefit from the safeguards system.

D. System Security

Projected nuclear-materials management and control systems for future fuel-cycle facilities range from totally manual to fully automated. Traditional personnel-operated manual and semiautomated information systems, although more familiar, are demonstrably more vulnerable than the computerized systems[20] used in a wide variety of sensitive applications. A preoccupation with the possible complexity and vulnerability of computerized safeguards systems is largely unwarranted and, for the most part, reflects the conservatism of the nuclear process industry and its unfamiliarity with secure data acquisition and distribution systems.

The exhaustive literature devoted to studies and comprehensive analyses of data and computer security risks[21] has emphasized

1. insuring MMAS integrity,
2. protecting the data base from unauthorized modification and disclosure, and
3. protecting the safeguards control functions.

Initial information-system action usually takes place at the source by transferring digital data directly from interfaced instruments. Human interactions, such as data entry through computer terminals, must be kept to the absolute minimum. Subnational fraud is controlled by interactive personnel authentication techniques and the use of two-person concurrence on SNM transactions. To control national or facility-wide subversion, the system is programmed to provide entry error detection through reasonableness bounds and comparisons. Data-base integrity is maintained by establishing and tracing manually verifiable audit trails available for internal facility and external IAEA Inspector audits and by providing secure input for random interrogation and independent verification of the data base by the inspectorate.

Information transmission to the safeguards information center or data bank can be secured by the application of encryption and data-authentication techniques. A system known as TRUST, developed in 1968 by Sandia Laboratories for the Arms Control and Disarmament Agency (ACDA) and the IAEA, provided complete authentication, integrity verification, and fraud detection in data from operating CANDU reactors during normal and refueling operations.[22] This system is extremely difficult to break even though the adversary has total knowledge of the data being transmitted. Its primary limitations are the amount and rate of data transmission. More compact and sophisticated derivatives of this system are being proposed for international use in unattended sensor stations for the proposed Comprehensive Test Ban Treaty (CTBT).[23] The current ACDA-sponsored program, called RECOVER, is being designed to provide secure data transmission from nuclear facilities to IAEA headquarters.

The National Bureau of Standards (NBS) Encryption Standard has been accepted widely within the United States.[20] The three-stage NBS key system would require $\$3 \times 10^{37}$ worth of modern computer processing time to break the code. Additional layers of hardware protection using pseudo-random-number-generator techniques and dual-channel fiber-optics communication links are available.

Data-base integrity is maintained through personnel authentication at LOGIN and file-access points. Covert data collection is inhibited through the use of metallic room and component shields designed to attenuate both near-field and far-field electromagnetic emanations from terminals, printers, and computer main frames as is currently done in secure communications centers. Time-domain reflectometry for detecting taps or more complex tap-resistant, dual-channel fiber-optics communication links can be used. In addition to data authentication and encryption, random character stuffing in the data stream is used to inhibit signal-pattern recognition.

Data are transmitted from computers to off-site data repositories by the message-formatting, error-detection, and error-correction methods of ARPANET for communicating through noisy land-line or satellite channels.[24] An encryption interface has been incorporated into ARPANET that allows computers to intercommunicate at US defense security levels at least as high as SECRET. Classified data packets travel through the network simultaneously with unclassified data packets. The proposed RECOVER system sponsored by ACDA is intended for remote read-out of nuclear facility information system computer data and will use the simpler NBS Encryption Standard.

A safeguards control system encompassing a broad range of sensors, displays, alarms, and enable/disable functions may use any or all of the previously described authentication, encryption, and encoding techniques. Essential control signals within the facility are routed through protected wire lines sealed in conduit. Secure computer operating systems that inhibit penetration have been implemented on large main-frame computers[25] and medium-scale minicomputers.[26]

The most credible threat to the security system would be a computer operator who could exercise essentially unlimited privileges within the machine and its resources. The computer operator must be severely restricted in his functions and closely supervised or employed by the appropriate safeguards authority.

The extent to which data- and computer-security countermeasures need be incorporated into the overall system depends upon the specific installation, the type of fuel cycle, and the threat environment. The computer-security simulation code SECSIM provides a structured set of methods and measurement criteria for determining security effectiveness.[27] Graded technological, administrative, and physical-security strategies are modeled and evaluated, allowing successive designs to converge toward a systems architecture having an acceptable level of security effectiveness.

Current technology provides the means to ensure that valid safeguards information system security controls are in effect. Proper use of this technology ensures that the safeguards system can detect in a timely manner violations of safeguards controls at the national and subnational level with a high degree of confidence. Existing and proposed computerized data-acquisition systems to be operated in known hostile environments provide levels of protection far in excess of the level required for international safeguards, where relatively simple tamper-indicating systems may provide adequate verification of integrity and sufficient indication of national diversion.

E. System Reliability

As previously discussed here and elsewhere,[1-3,14-16] safeguards system reliability is extremely important because it affects both safeguards effectiveness and process operations. Safeguards system reliability has two important aspects: how often can a safeguards failure be expected and how long can a failure be expected to last. The first aspect is described by the mean time between failure (MTBF); the second, by the mean time to repair (MTTR).

Data on failure rates of individual devices such as resistors, capacitors, inductors, diodes, transistors, and integrated circuits are used to predict the reliability of an assembly of such devices. When the devices are assembled into components, manual calculations or computer programs tabulate device interconnections for required functions and the associated probability of failure is calculated for each required mode of operation. From these probabilities an overall, theoretical MTBF is determined for each component as a measure of its reliability. If failures occur randomly in time, the probability P that a component will function normally for any length of time t is given by

$$P = \exp(-t/\mathrm{MTBF}) .$$

For example, if the time since the last failure is equal to the MTBF, there is a 36.8% probability that the component has not failed.

After several components have been assembled, life-test data can be accumulated to determine an experimentally derived MTBF value, and comparisons with the theoretical values are made. Often thermal and overvoltage stresses are applied in accelerated life tests to obtain preliminary MTBF estimates as early as possible.

The MTTR is the time required to return a component to service. Values of MTTR can extend from fractions of a second in the case of automated repair to days if personnel or logistical delays are encountered.

Availability AV is the ratio of time a computer system is functioning normally to the total time the system is in demand. Availability is also expressed in terms of MTBF and MTTR as

$$AV = \frac{MTBF}{MTBF + MTTR} .$$

The downtime per year, DTY, is expressed in terms of availability as

$$DTY = (1 - AV)8760 \text{ h/yr.}$$

To assess the impact of safeguards-related failures, particularly upon the process, both MTBF and MTTR must be known. Because of the relatively long transient response of the process, especially during start-up and shutdown, the differential effect between one comparatively long-lasting failure and several shorter-lived ones of the same total duration can be large. Thus, the downtime must be constrained.

System reliability can be calculated with the computer code RELSIM, which uses probabilistic relationships to decompose complex component networks in an orderly fashion. Initially a two-dimensional, hierarchical network ordering system containing up to several hundred units in both directions is used. Units connected in tandem are converted to an equivalent unit at the lowest level occupied in the original configuration. The equivalent availability is expressed as the product of the individual units. System components connected in parallel with a specified survival criterion of at least K of N are decomposed into an equivalent unit of that availability using the above relationships, and the entire process is repeated at each level until a single equivalent system unit is obtained. Further, as failures occur the computations can be repeated for the new set of conditions and the probability estimates revised. This feature has even broader implications on a total systems level in which, as computer failures occur within a redundant network, the RELSIM model can be updated and a new projection of the availability computed in near real time to give the current dynamic vulnerability to equipment unreliability.

In addition to allowing complete generality in simulating networks, the incorporation of nonidentical, redundant components into RELSIM allows simulations of systems in which incremental upgrading is taking place. These upgrades retain the prior computers but augment them with more modern units. Also, nonidentical units are desirable to avoid common-mode software failures. Because the failure statistics are different for hardware and software, it is possible that all identical, redundant units will experience the same set of input data and internal software conditions, thus causing simultaneous crashes. This added protection of dissimilar units backing up one another must be weighed against the burden of increased workload required to maintain the units.

Because the plants are similar, the reliability of the safeguards system for the reference coconversion plant ideally should not differ greatly from that for the conversion facility;[2] that is, an MTBF of at least 3 months and an MTTR of less than 5 h are desirable. However, because of the larger number of instruments necessitated by the complexity of the reference process design (Sec. III.C), these goals will be somewhat more difficult to achieve.

F. Structure of International Safeguards Systems

International safeguards objectives, as set forth by the IAEA in INFCIRC/153, par. 28,[28] are the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities and the deterrence of such diversion solely by the risk of early detection.

This section addresses the problem of covert diversion by a host nation operating a coconversion facility (national diversion). Overt diversion should be detectable by the IAEA safeguards system if the techniques for detecting covert diversion are effective.

Detecting covert national diversion under the conditions of international safeguards agreements is a significantly different problem than that of detecting covert diversion in a domestic facility. In domestic facilities, the diversion threat is subnational, which implies constraints on manpower and resources within the facility. For national diversion, it is conservative to assume that the host nation can command whatever manpower and resources might be required to reach the diversion objective. As already pointed out, the international safeguards system also cannot rely on the PPS, which is part of the State's safeguards system.

The IAEA considers materials accounting "a safeguards measure of fundamental importance" and containment and surveillance "important complementary measures" (INFCIRC/153, par. 29). The key importance of materials accounting stems from its ability to quantify the diversion and its significance and to provide continuity of knowledge about the state of nuclear material, both in time and in location within the nuclear fuel cycle. Containment and surveillance techniques are directed at prompt detection of illicit activities involving nuclear materials and confirmation of any diversion detected by materials accounting.

INFCIRC/153, par. 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." All these considerations mandate a structure for the international safeguards system like that in Fig. 7. The State's safeguards system should include many (if not all) of the features just described for a domestic safeguards system, and the international safeguards system would form a verification overlay.

By materials accountability the IAEA seeks to obtain assurance to a satisfactory degree of confidence (now 95%) that a significant amount of nuclear material is not diverted from a materials balance area during a certain period. In the case of conversion 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 and on the plant throughput and inventories for the materials balance period.

If dynamic materials accounting (Sec. II.B.3) is implemented by the host nation as the State's accounting system, then the IAEA inspectorate must be able to verify the results independently. This function could be performed as currently proposed for a conventional State's accounting system, that is, by independent measurements linked to the physical inventory structure. In this case, the verification procedures are no different than for a conventional accounting system. However, dynamic materials accounting offers the possibility of more extensive verification activities, particularly if resident inspectors are allowed and are equipped with suitable instruments. In these circumstances, there are more measurement opportunities, both in time and in location. Furthermore, the much more comprehensive data from the dynamic materials accounting system facilitate checks of internal consistency of the State's accounting data between physical inventories.

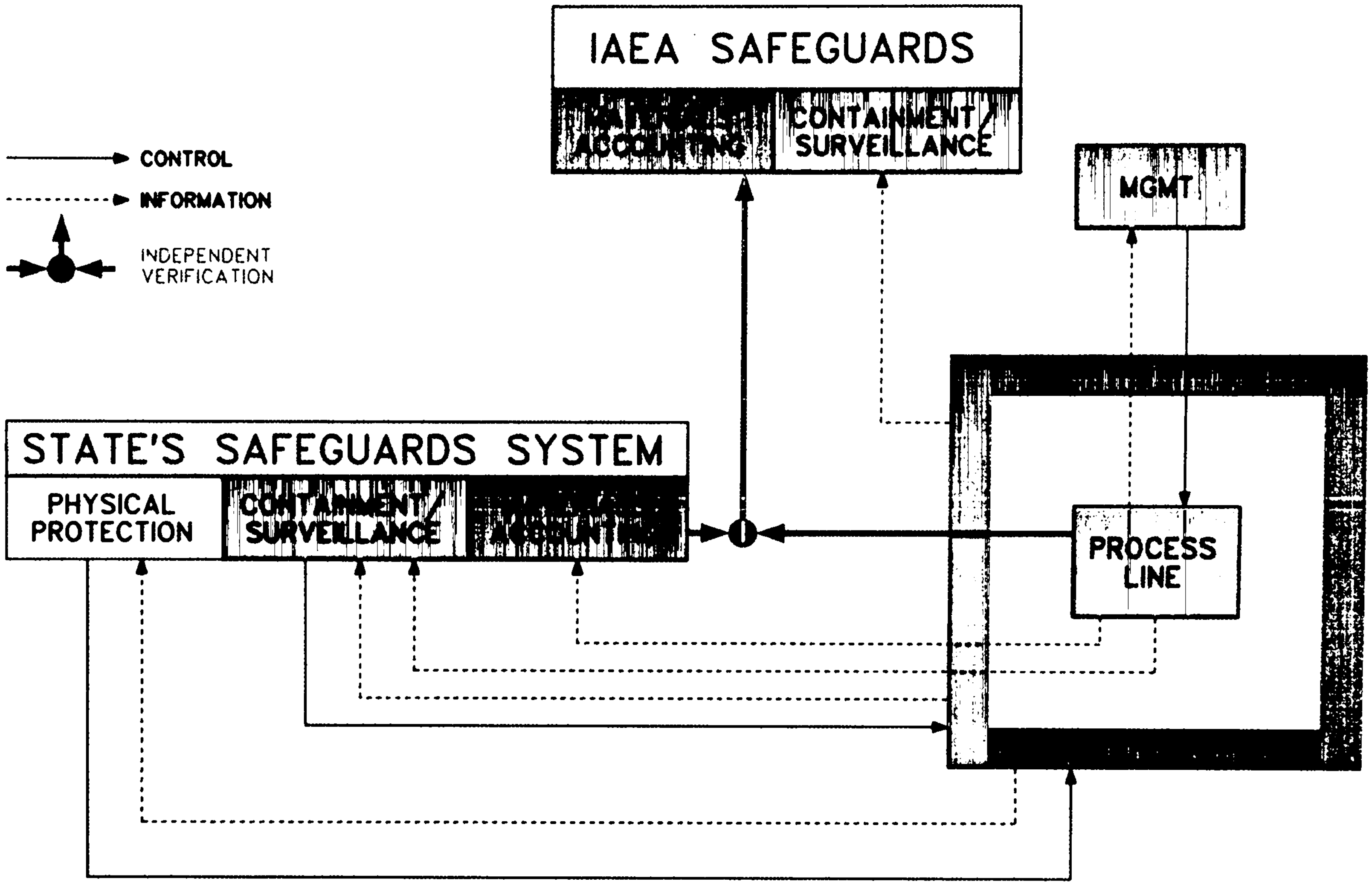


Fig. 7.
Structure of the international safeguards system.

Containment and surveillance devices are used either to detect unusual or inadequate operator's actions or to give assurance for the full period that the inventory has not changed in areas where no changes are supposed to occur. Positive indications by containment and surveillance devices of such actions or changes would be grounds for a new inventory verification by the IAEA inspectors. From the point of view of safeguards effectiveness, incomplete information from containment and surveillance devices or their failure to give assurance for the full period is also significant.

For the successful application of containment and surveillance measures, diversion paths should be analysed carefully. Taking diversion scenarios into account, containment and surveillance devices should be applied to close all diversion paths that might permit the diversion of significant quantities of nuclear material.

In particular, containment and surveillance measures may serve the following safeguards purposes:

(a) to detect that undeclared movement of nuclear material in or out of surveillance areas has taken place or might have taken place;

(b) to detect undeclared operational activities and to provide evidence for confirming the explanations for declared operational activities in the surveillance area;

(c) to deter diversion of nuclear material or any undeclared actions by the risk of early detection;

(d) to effect and complement the validity of materials accounting; and

(e) to simplify procedures for taking physical inventories.

The monitoring of process parameters such as flow rates, temperature, tank levels, and valve status, an important part of the containment and surveillance system, assures that the process is being operated in the design mode. Output of the containment and surveillance sensors is monitored by the safeguards computer network.

Effective materials accounting for both national and international safeguards systems will rely ultimately on a combination of conventional accounting including periodic flush-out inventories and dynamic materials accountability. The measures and features discussed in Sec. II.B have been designed for and are related to domestic safeguards needs. For international safeguards use, the IAEA needs to verify independently the fissile content of processing streams by tamper-indicating monitoring systems. This capability is still being developed.

III. MATERIALS MEASUREMENT AND ACCOUNTING SYSTEM

A. General Considerations

The safeguards system structure is described in Sec. II. In this section design concepts for the application of dynamic materials accountability to the reference Coprecal process (App. A) are considered. Accountability measurements and measurement points are proposed and safeguards staffing and costs are estimated.

For both safeguards and nonproliferation, the conversion of fissile material from the nitrate form to the oxide form is one of the most important processes in the commercial light-water and fast-breeder reactor fuel cycles. As described in a previous Los Alamos Scientific Laboratory (LASL) study,[2] the conversion process links spent-fuel reprocessing with fuel fabrication, and large quantities of relatively pure SNM in both liquid and solid forms are processed.

The variety of chemical and physical forms containing plutonium is less than in a reprocessing plant,[1] primarily because of the absence of fission products, but is somewhat greater than in a mixed-oxide fuel fabrication facility,[3] because of the wet front end of the conversion process. The major difference between the reference coconversion process and the conversion process studied in Ref. 2 is the presence of uranium; the total heavy metal content is 90% uranium and 10% plutonium.

Although all forms of SNM in the reference coconversion process are attractive diversion targets and therefore are important for safeguards, certain process design considerations suggest that some forms may be somewhat more attractive to a potential divertor than others.

For example, consider the amount and form of material required to divert 3 kg of plutonium. Plutonium concentration in the feed-blend tanks is 40 g/L, so 75 L (19 gal) would be needed to obtain 3 kg of plutonium mixed with 27 kg of uranium. On the other hand, one product canister holds 3.2 kg of plutonium (as oxide) diluted with 33 kg of uranium oxide in a volume of 30 L (7.5 gal). Therefore, the product powder is more attractive for diversion than the feed solution in terms of the bulk quantity that would have to be taken. To obtain pure plutonium, the divertor would have to dissolve the mixed-oxide powder, chemically extract the plutonium, and reconvert the plutonium solution. This process is simple relative to the chemical separation of fission products, but it contains one additional step that would be unnecessary for mixed uranium-plutonium-nitrate solution. The concept of graded safeguards provides little guidance in this case. The divertor of 3 kg of plutonium could take 75 L of feed solution and would need to do less chemical processing, or he could take 30 L of product powder and would have to perform additional wet chemistry. Because extracting plutonium from the powder is easy, the powder may be the more desirable form to a divertor.

Process design has a direct impact on the application of dynamic accountability. In the reference process precipitation and calcination are operated continuously during 16.5-day campaigns, while reduction, stabilization, and screening are batch operations. Therefore, batch integrity is not maintained throughout the process, and dynamic accounting requires on-line measurements of flows in continuous process streams.

Another feature that makes materials accounting difficult is the presence of the large (16 000-L) feed-blend tanks. Each tank feeds the process continuously during a campaign. Inventory measurements of such large vessels, although relatively precise and accurate, can result in large absolute uncertainties in the amount of contained SNM.

Holdup of powders in process equipment is a generic materials accountability problem for both conversion and fuels refabrication processes.[3] Relatively simple NDA holdup monitors may be necessary for process equipment and for vacuum system filters. A more thorough evaluation of the engineering designs of specific process equipment will be required to specify the types and the locations of such holdup monitors. Portable NDA holdup monitors will be necessary for use during a physical inventory after the process line is cleaned out. Holdup monitors have been designed for similar application (App. D) and, in general, they are available at low cost and should make little impact on cost and process design.

B. Materials Measurement

Measurements of liquids, solids, and mixed liquid-solids are required for application of dynamic accountability to the Coprecal process. Coconversion requires more and larger process equipment than does conversion[2] to achieve the same fissile throughput because 10 times the amount of material must be processed. This requirement exacerbates the problem of measuring in-process inventory and results in more costly instrumentation. On the other hand, the required measurements of mixed uranium and plutonium materials in the coconversion process are not expected to be fundamentally different or more complex than those of materials in the conversion process (App. D). Therefore, we can use the guidance provided by the extensive experience acquired in measuring uranium and plutonium materials to select appropriate measurement techniques and instrument types and to anticipate their quality and sensitivity.

Table III contains a list of proposed key measurement points, measurement techniques, and their estimated uncertainties for dynamic materials accounting in the main coconversion process.[5] Except for inventory measurements of the feed-blend tanks, all requisite measurements are based on NDA techniques.

Estimates of measurement errors have been grouped in two categories. Instrument precision represents the estimated scatter in a set of individual raw measurements (for example, the uncertainty caused by counting statistics in NDA measurements). Calibration error represents the uncertainty in converting raw, measured values to the quantity of interest, for example, converting raw counts to plutonium mass for NDA measurements. Calibration errors are the most difficult to estimate because they include uncertainties in standards, calibration parameters, instrument environment, and measurement controls. We must assume that appropriate standardization techniques will be available. No calibration errors are quoted for precipitator, calciner, or filter inventory measurements or for holdup measurements because these errors approximately cancel in the error models for materials balances and cumulative summations of materials balances (cusums) (Sec. IV.B).

The measurement of inventory in the feed-blend tanks combines a mass or volume measurement with a concentration measurement. There is some question as to whether mass or volume measurement is more appropriate. Mass measurement, for example, by load cell, for such large vessels may be difficult because of hysteresis effects induced by piping connections to the tank. Similarly, volume measurements may be degraded by flexing and thermal expansion of the tank walls and by the presence of poison rods for criticality control. Furthermore, both measurement methods would be seriously impaired if the tank solution is continuously recirculated to assure complete mixing; however, we assume that, whenever either measurement is made, the recirculation system is turned off to allow the solution to quiet down. These factors imply some uncertainty about how well the receipt tank inventory can be measured and which method, volume or mass, should be used. Therefore, Table III shows error ranges of 0.1-1% (1σ) for both methods; corresponding simulation results appear in Sec. V.

Plutonium concentration in the receipt tanks is measured primarily by analytical chemistry. Appropriate analytical methods for determining isotopic and total element composition are given in App. D. The resulting concentration value can also be assigned to the process feed stream. We recommend that an in-line NDA technique be used to supplement and provide timely confirmation of the concentration measurement. One effective method is absorption-edge densitometry (App. D), an element-specific NDA method that can be applied on-line or at-line in most areas amenable to gross

TABLE III

MEASUREMENTS FOR DYNAMIC ACCOUNTABILITY IN THE COPRECAL PROCESS

Measurement Point	Number of Measurement Points	Material Description	Measurement Type	Instrument Precision [%(1 σ)]	Calibration Error [%(1 σ)]	Calibration Period
Receipt-feed-blend inventory	9	U-Pu in HNO_3 400 g HM/L 10% Pu	Mass or volume Concentration (chem. anal. and abs.-edge densitometry)	0.1-1 0.2	0.1-1 0.2	Campaign Campaign
Precipitator input	3	U-Pu in HNO_3 1 L/min	Flow meter	1.5	1	Campaign
Precipitator inventory	3	Slurry 1 kg Pu	NDA, neutron	10	---	Physical inventory
Calciner inventory	12	PuO_2, UO_3 0.9 kg Pu	NDA, neutron	10	---	Physical inventory
Primary filter holdup	3	PuO_2, UO_3 0.75 kg Pu	NDA, neutron	10	---	Physical inventory
Secondary filter holdup	3	PuO_2, UO_3 0.75 kg Pu	NDA, neutron	10	---	Physical inventory
Reduction container	3	PuO_2, UO_3 3.3 kg Pu	NDA, neutron	2	1	Day
Reduction-stabilization holdup	12	PuO_2, UO_2 0.2 kg PU	NDA, neutron	10	---	Physical inventory
Final filter holdup	12	PuO_2, UO_2 0.2 kg Pu	NDA, neutron	10	---	Physical inventory
Scrap	3	PuO_2, UO_2	NDA, neutron	2	5-10	Day
Vacuum	3	PuO_2, UO_2	NDA, neutron	2	5	Day
Product	3	PuO_2, UO_2 3.2 kg Pu 37 kg HM	NDA, neutron or calorimeter	1	0.5	Day

absorption measurements. Although gross gamma-ray and x-ray densitometry may be suitable for process control, they are susceptible to errors caused by the presence of medium- or high-atomic-number elements, which would be included in the total plutonium analysis. With proper choice of cell path length and either K- or L_{III}-absorption edges, plutonium concentrations between ∿5 and 400 g/L can be measured to a precision of better than 1% (1 σ) in the presence of impurities. Furthermore, the technique is well suited to simultaneous measurement of both plutonium and uranium in a coprocessing mode, and such measurements have been made in the laboratory (Ref. 1, App. A).

In practice, quantitative measurements of calciner in-process inventory may be difficult for three reasons. First, the calciners operate at high temperatures. The NDA neutron detectors will have to operate reliably in a relatively high temperature environment. Second, the proximity of the calciners means that there may be interferences among the neutron counters. Third, the crowded environment of the instruments will complicate instrument calibration and maintenance procedures.

Accurate measurements of inventory and holdup in the precipitators and filters also are difficult because of material form and equipment geometry. The methods chosen require that the chemical and isotopic compositions be available from analysis of samples taken from the process.

In general, each NDA instrument must be designed for its specific application. Equipment design and operating features must be considered in terms of accessibility for measurement, maintenance, and calibration and of background, shielding, and multiplication factors. Specific instrument systems must be evaluated for reliability, sensitivity, and operational acceptability under field conditions.

Although it is not yet part of the conversion process design, there will almost certainly be some solid, low-level waste to be packaged in 200-L (55-gal) drums for disposal. These drums should be analyzed to prevent their use for illicit removal or concealment of materials. A drum scanner measuring the 414-keV gamma ray from ^{239}Pu can detect as little as 1 g of ^{239}Pu in a 5-min scan time.[4] The accuracy for measuring ∿10 g of ^{239}Pu can be as good as 10% for matrices of low-atomic-number combustible waste and can be as poor as 50% for unknown matrices. The drum scanner is in an advanced stage of development, but still requires field testing and evaluation for this application.

C. Safeguards Staffing

The safeguards staff estimated for the reference coconversion facility (Table IV), excluding the security force, should be the same as for the conversion process.[2] Many support functions for the reference plant (for example, those for process control) can support the safeguards system as well with only a small increase in personnel, especially if standard equipment is used throughout. The staffing numbers do not reflect the possibility that the coconversion process may be part of a larger facility. In that case, the safeguards staffing burden on the coconversion process should be much smaller.

D. Safeguards Costs

Safeguards costs (Table V) are estimates of the costs of instrumentation required for dynamic accountability and to perform physical inventories. Costs of volume-measuring and other devices necessary for process control are not included, nor

TABLE IV

SAFEGUARDS STAFFING FOR THE CONVERSION PLANT

Supervisory Personnel	
Materials-management manager	1
Data-evaluation specialist	1
Senior-level professionals	4
Shift Personnel (Total for four shifts)	
Safeguards officer	4
Safeguards assistant	4
Data analyst	4
Computer staff	12
Instrumentation technician	4
Safeguards line inspector	8
TOTAL	42

TABLE V

SAFEGUARDS COSTS

Item	Unit Cost ($ thousand)	Number	Extension ($ thousand)
Safeguards computer and information system hardware	500	1	500
Spare parts (at 25%)	--	--	125
Software development (at 300%)	--	--	1875
Absorption-edge densitometer (plus one spare)	100	4	400
Neutron counter (plus five spares)	40	56	2240
Large neutron well counter for primary and secondary filters (plus one spare)	100	7	700
Precision flow meter (plus one spare)	10	4	40
Mass or volume measurement upgrade	20	9	180
Miscellaneous portable instruments			100
SUBTOTAL			6160
Equipment engineering design and procurement (at 15%)			924
Contingency (at 30%)			2125
TOTAL			9209

are data on the physical protection or process-monitoring systems. Allowances are made for floor space, engineering planning (architectural engineering), installation, equipment engineering design and procurement, and contingency.

The instrument costs and numbers shown in the table are conservative, and no credit is taken for instrumentation of this type that may be required for process control. In some cases, costs will decrease as instrument development proceeds. This is especially true of the seven neutron detection systems proposed for the primary and secondary filters and of the neutron detection systems proposed for the in-process inventory in the calciners and precipitators because such systems have not yet been built and tested.

The hardware cost for the safeguards computer and information system is estimated at $100 thousand more than that for the conversion facility.[2] The reason is the much larger number of instruments and the concomitant interface and data communications requirements.

Safeguards staffing costs are assumed to be $20 thousand/man-yr with 150% overhead, for a total (see Table IV) of $2.1 million. Yearly software maintenance and modification are taken to be 10% of the original software cost, or about $150 thousand. Hardware maintenance and supplies, at 10% of the original hardware cost, consume about $430 thousand/yr. Thus, the operating cost for this part of the safeguards system should be about $2.6 million/yr.

IV. MODELING, SIMULATION, AND ANALYSIS TECHNIQUES

A. Introduction

The Coprecal process has not yet been used in a full-scale conversion facility. Required operating data for this study were obtained from a computerized dynamic model of the Coprecal process based on actual design data for a proposed, industrial-scale facility.[5] This model, called COPSIM (COPrecal SIMulation), is described in App. B. The simulated data from COPSIM represent the materials flows and inventories under expected normal conditions, when the process is operating near steady state. These simulated data are based on and are consistent with current best estimates of the expected performance of the process.

The conceptual MMAS for the Coprecal process is described in Sec. III. Promising strategies for instrumentation and accounting procedures are evaluated in Sec. V using techniques developed during previous safeguards studies.[1-3] The resulting quantitative estimates of diversion sensitivity are the basis for the conclusions and recommendations given in Sec. VI.

Models of accountability measurements and associated measurement errors are described in Sec. IV.B. Equations for dynamic materials balances and cusums of dynamic balances are given (see also App. C), along with models for their uncertainties that are caused by measurement errors. The model measurements are applied to the simulated process-flow and in-process inventory data using the Monte Carlo computer code, COPMEAS (COPrecal MEASurements), developed for that purpose.

The measured values are combined to form dynamic materials balances under various strategies for dynamic materials accounting. Measurement points, appropriate instrumentation, and associated measurement errors are described in Sec. III.B. In many cases, the measurement models are based on the performance of similar instrumentation characterized in both laboratory and field applications with similar materials.

The effectiveness of proposed dynamic accounting strategies is evaluated by applying decision-analysis techniques to the simulated accounting data. The general framework of data-analysis and decision methods developed for earlier studies[3-6] has been expanded and is described in Sec. IV.C. Sensitivity to diversion and effective false-alarm and detection probabilities are estimated (Sec. V) by examining test results from many sets of materials accounting data derived from the process and measurement models.

B. Measurement and Error Models

1. Measurements. Operation of the MMAS is simulated using COPMEAS. This code simulates measurement of the true materials flow and in-process inventory data generated by the process model COPSIM (App. B) and transmits appropriate measured values and their computed uncertainties to DECANAL, the safeguards data-analysis code. Each proposed accounting strategy requires a specialized version of COPMEAS. COPMEAS incorporates both additive and multiplicative measurement-error models,[29] described below.

a. Additive Model. In this model, the measured value M of a true quantity μ is given by

$$M = \mu + \varepsilon + \eta \quad , \tag{1}$$

where ε is the error caused by instrument imprecision and η is the error produced by uncertainty in the instrument calibration. Both errors are assumed to be independent and normally distributed with mean zero and variances σ_ε^2 and σ_η^2, respectively. The variance σ_M^2 of M is given by

$$\sigma_M^2 = \sigma_\varepsilon^2 + \sigma_\eta^2 \quad . \tag{2}$$

All measurements obtained from a given instrument using the same set of values for the calibration parameters are correlated through the calibration error η. The covariance σ_{ij} between the i^{th} and j^{th} measured values is given by

$$\sigma_{ij} = \sigma_\eta^2 \quad . \tag{3}$$

b. Multiplicative Model. In this model, the measured value, variance, and covariance are given, respectively, by

$$M = \mu(1 + \varepsilon + \eta) \quad , \tag{4}$$

$$\sigma_M^2 = \mu^2\left(\sigma_\varepsilon^2 + \sigma_\eta^2\right) \quad , \text{ and} \tag{5}$$

$$\sigma_{ij} = \mu_i \; \mu_j \; \sigma_\eta^2 \quad , \tag{6}$$

where the true value μ and the error components ε and η are defined as above, except that ε and η are now expressed as relative errors.

In simulations, a value for η is sampled periodically from the appropriate distribution to coincide with the frequency of instrument recalibration. A value for ε is sampled for each measurement. Variance and covariance terms are estimated by replacing the true quantities μ in Eqs. (5) and (6) by the appropriate measured quantities. The precisions assigned to the measurements required for the Coprecal MMAS are given in Table III.

2. Materials Balances. Each materials balance MB is a linear combination of measured quantities P_i of plutonium.

$$MB = \sum_{i=1}^{n} c_i P_i \quad , \tag{7}$$

where c_i is +1 (-1) if P_i is an input or an initial in-process inventory (output or final in-process inventory) quantity for the accounting area. Often, the measured mass of plutonium is given by the product P of two different measured quantities.

$$P = xy \quad , \tag{8}$$

where x is either liquid volume or mass, and y is either plutonium concentration or mass fraction. The measurement-error model for both x and y is similar to that given in Eq. (1) or Eq. (4).

$$x = X + \varepsilon_x + \eta_x \quad \text{and} \quad y = Y + \varepsilon_y + \eta_y \tag{9}$$

for the additive model or

$$x = X(1 + \varepsilon_x + \eta_x) \quad \text{and} \quad y = Y(1 + \varepsilon_y + \eta_y) \tag{10}$$

for the multiplicative model, where X and Y are true values, and the error components are defined as in Eq. (1) or Eq. (4). In some cases, the additive model may be appropriate for one of the measured quantities (X or Y), whereas the multiplicative model is appropriate for the other. In such cases, a combined model is used.

The variance σ^2_{MB} in the computed value of a materials balance is a combination of the uncertainties in the contributing measured values. Ordinarily, several measurements will be made using a given instrument before it is recalibrated, and appropriate correlation terms must be included in the computation of the materials balance variance. The general form of the equation used to compute the approximate variance of each materials balance is

$$\begin{aligned} \sigma^2_{MB} &= \sum_{i=1}^{n} X_i^2 Y_i^2 \left(\sigma^2_{\varepsilon_x} + \sigma^2_{\varepsilon_y} \right) \\ &+ \sum_{i=1}^{n} \sum_{j=1}^{n} c_i c_j \; X_i X_j \; Y_i Y_j \left(q_{ij} \sigma^2_{\eta_x} + p_{ij} \sigma^2_{\eta_y} \right) \quad . \end{aligned} \tag{11}$$

The quantity q_{ij} is +1 if X_i and X_j have been measured with a common value of η_x (that is, using the same instrument calibration) and is zero otherwise. Similarly, p_{ij} is +1 or 0, depending on whether a common value of η_y was used in the measurements of Y_i and Y_j. Each $i = j$ term in the double sum in Eq. (11) is the component of variance owing to calibration uncertainty for a single measurement; these terms are present even if all measurements of X or Y are uncorrelated.

Equation (11) is written for the case where the multiplicative model is appropriate for the measurement of both X and Y. If the additive model applies to either measurement, the corresponding quantity (X or Y) is set to unity.

Measured values of net materials transfers and initial and final in-process inventories, along with appropriate components of variance and covariance, are passed to DECANAL for each materials balance period. These quantities are sufficient for computation of materials balances, cusums, and the other test statistics described in Sec. IV.C.

In terms of the net transfer T, initial inventory I_i, and final inventory I_f, the materials balance equation becomes

$$MB = T + I_i - I_f \quad . \tag{12}$$

In some of the proposed accounting strategies (see Sec. V and App. C), the terms in the computation of the materials balance variance [Eq. (11)] arising from calibration errors in the inventory measurements approximately cancel because two inventory measurements appear with opposite signs in each materials balance equation. The magnitude of these terms is $(I_i - I_f)^2 \sigma_\eta^2$. In such cases, if the model process is operated near steady state so that $I_i \simeq I_f$, the contribution to σ_{MB}^2 is relatively small; that is, calibration errors in the inventory measurements nearly cancel. In the error model, perfect cancellation is assumed, and σ_η is set to zero for the associated in-process inventory measurements. Note that this cancellation does not occur in the feed-blend tanks because the in-process inventory changes.

3. Cusums. A cusum, the sum of all materials balances for the unit process since the beginning of the accounting period, is computed after each materials balance period. The cusum variance is a complex combination of the variances of individual materials balances, as these balances usually are not independent. There are two principal sources

of correlation between materials balances. The first is the correlation, discussed previously, between measurement results obtained using a common instrument calibration. The magnitudes of the associated covariance terms depend on the magnitude of the calibration error and the frequency of each instrument recalibration; omission of these terms can cause gross underestimation of the cusum variance. The second source of correlation between materials balances is the occurrence, with opposite signs, of each measured value of in-process inventory in two adjacent materials balances. As a result, only the first and last measurements of in-process inventory appear in the cusum, and only the corresponding variances appear in the cusum variance. The cusum variance is computed using an equation having the same form as Eq. (11). Detailed equations for materials balances, cusums, and their associated variances are given in App. C.

C. Data Analysis for Diversion Detection

Analysis of materials accounting data for possible SNM diversion is a major function of the MMAS. Diversion may occur in two basic patterns: abrupt diversion (the single theft of a relatively large amount of SNM) and protracted diversion (repeated thefts of SNM 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 (see Sec. II.B.3) enhances the ability to detect such diversions, but it also inundates the safeguards system operator with materials accounting data. Furthermore, although these data contain much potentially useful information concerning both safeguards and process control, the significance of any isolated set of measurements is seldom readily apparent and may change from day to day, depending on plant operating conditions. Thus, the safeguards system operator is presented with an overwhelmingly complex body of information from which he must repeatedly determine the plant's safeguards status. Clearly, he must be assisted by a coherent, logical framework of tools that address these problems.

Decision analysis,[30-34] which combines techniques from estimation theory, decision theory, and systems analysis, is such a framework, and it is well suited for statistical treatment of the imperfect dynamic materials accounting data that become available sequentially in time. Its primary goals are (1) detection of the event(s) that SNM has been diverted, (2) estimation of the amount(s) diverted, and (3) determination of the significance of the estimates.

The application of decision analysis to nuclear materials accounting has been reported previously,[30-32] and only a brief overview is given here. The detection and estimation functions of decision analysis are based on classical hypothesis testing and modern state-variable estimation techniques. The systems analysis portion attempts to set thresholds for the hypothesis tests in a rational fashion, for example, by using utility theory to determine desirable false-alarm and detection probabilites.

The detection function is based on acceptance of the hypothesis that some (initially unknown) amount of SNM is missing versus the hypothesis that all SNM is present. One useful kind of decision test compares a likelihood ratio to a threshold. The likelihood ratio is roughly the ratio of the probability that SNM is missing to the probability that it is not, and the threshold is determined by the desired false-alarm and detection probabilities. This structure can accommodate both parametric tests, which require detailed knowledge of measurement error statistics, and nonparametric tests, which do not. Furthermore, the set of tests can search for diversion that may have occurred in any pattern.

The decision-analysis algorithms are applied to the accounting data in the code DECANAL. Interpretation of the results is aided by use of the computerized alarm-sequence charts[1,3,10] developed for earlier studies.

DECANAL has been extended to include the sequential variance test (SVT) and smoothed materials balance test (SMBT), in addition to the Shewhart chart, cusum, uniform diversion test (UDT), and Wilcoxon rank sum test it already contained. All these tests are combined with alarm-sequence charts discussed below.

The Shewhart chart is the oldest graphic-display tool widely used by industry for process control.[29,35] In its standard form, it is a sequential plot of measured data on a chart with warning and action limits usually set at the 2-σ and 3-σ levels, respectively. In safeguards applications, it is a sequential plot of the materials balance data with 1-σ error bars.

The cusum chart, developed in England, was used first during the early 1950s for improved process control.[36] It and its near relative, the Wald test for a shift in location, developed in the United States during World War II, have found wide acceptance in industry because, unlike the Shewhart chart, they are sensitive to small, persistent shifts in process parameters. The cusum is very easy to calculate; it is simply the unweighted cumulative summation of the raw data. The cusum chart is a sequential plot of the cusum values. In safeguards it is a sequential plot of the cusum of dynamic materials balances and its standard deviation, which are calculated from the raw materials balance data.[1-3]

The Kalman filter has found wide application to communications and control systems for signal processing in stochastic environments. It is a powerful tool for extracting weak signals embedded in noise. Its application to safeguards is relatively new[30-32,37-40] and has arisen because dynamic accountability systems will rapidly generate large quantities of data that may contain "weak signals" caused by repeated, small diversions embedded in the "noise" produced by measurement errors. The Kalman filter is the basis for the UDT, the SVT, and the SMBT.

The UDT is designed to detect a small, constant diversion during each materials balance period. Minimum-variance, unbiased estimates of the diversion and the inventory at each time are given by the Kalman filter described in Ref. 30, which also gives a method for correctly treating correlated measurement errors. Similar, but less general, formulations are reported in Refs. 37-40.

The cusum and the UDT are complementary in several respects. The cusum estimates the total amount of missing SNM at the current time, and its standard deviation is taken as the 1-σ error in the estimate of the total. The UDT, on the other hand, estimates the average amount of SNM missing from each materials balance, and its standard deviation estimate is taken as the 1-σ error in the estimate of the average. Thus, both the cusum and the UDT search for a persistent, positive shift of the materials balance data, the cusum by estimating the total and the UDT by estimating the average.

However, the UDT has two advantages over the cusum. First, it provides a better indication of missing SNM because it makes more efficient use of available information; tests based on the Kalman filter estimates are more discriminating. Second, certain forms of the Kalman filter provide improved estimates of in-process inventory, useful for both process control and safeguards. Disadvantages of the UDT are its relative complexity and the fact that its application requires considerable care.

Both the cusum and UDT tests are performed sequentially, facilitating their implementation on a small computer. Furthermore, the accounting data can be examined for any pattern of diversion (random, uniform, etc.) with only minor changes in the algorithms.

Application of the nonparametric Wilcoxon test and the associated rank-sum chart to safeguards data is described in Ref. 1. This test has been used in other fields to analyze data for which the underlying distribution of the measurement uncertainty is unknown. Like the cusum, the Wilcoxon test is very easy to implement; it calculates a weighted sum of the number of positive values in sets of materials balance data.

We expect a larger materials balance error variance when diversion is present than when diversion is absent. The SVT uses two Kalman filters, each similar to that for the UDT, to calculate the materials balance error variances for diversion and for no diversion. The result is roughly equivalent to a sequential formulation of the well-known F test for variances. The corresponding assumption on the diversion scenario is that the diversion during each materials balance period is a Gaussian random variable having constant mean and variance, which are a priori unknown. Maximum-likelihood estimates of the mean and variance are computed sequentially from the likelihood ratio as the data are received.

The diversion pattern assumed for the SVT is much less restrictive than that for the UDT because almost any set of diversions could have been drawn from a white, Gaussian, random process, even if the diversion were constant or intermittent. The only real restrictions are that the mean and variance be constant over the test interval. However, the test procedure covers all possible intervals, so that this assumption is less restrictive than it might seem.

A similar estimation algorithm is described in Refs. 37-40, but no procedures for obtaining the diversion mean and variance are given. In addition, it is not clear what decision test is to be used.

As with the UDT, the SVT provides estimates of both the missing material and the inventory at each time. However, the total amount of missing material over the test interval is also computed by subtracting the last inventory estimate from the first inventory measurement and adding in the intervening net transfers. This estimate of the total diversion is more indicative of the materials accounting situation. Note that the alarm-sequence chart refers not to the missing-material estimates, but to a possible shift in materials balance error variance.

Stewart[41] noted earlier that better (smaller variance) materials balances could be drawn if past data were used to calculate the beginning inventory of the current materials balance. He proposed the equivalent of a Kalman filter, assuming no diversion before the current time, for performing the calculation. This technique can be extended if one is willing to consider deferred decisions. That is, if we have data from N materials balance periods and we wish to compute the materials balance at time k, where k lies between 1 and N, then we can (1) run a "forward" Kalman filter from time 1 to k to estimate the k^{th} beginning inventory, (2) run a "backward" Kalman filter from time N to k + 1 to estimate the k^{th} ending inventory, and (3) subtract the result of (2) from that of (1) and add the intervening transfer measurement to find the smoothed materials balance at time k based on the data from time 1 to N. The procedure can also be done for any number of intervening materials balance periods, and it includes Stewart's method as a special case.

Significant improvements in single materials balance uncertainties may be obtained with the SMBT; the price is a delayed decision. Care must be taken when applying the test to intervals in which several diversions may have occurred; that situation violates the no-diversion assumption on which the filters are based and can cause incorrect materials balance estimates.

The algorithms for the Shewhart chart, cusum, UDT, SVT, and SMBT are structured to account for correlated data so that correct variances are computed for the associated decision tests. The actual false-alarm and detection probabilities for the Wilcoxon test depend on the degree of data correlation. If correlations are large, the Wilcoxon test performance will suffer unless corrective measures are taken.

The Shewhart chart is included in the examples (Sec. V.E) to illustrate the improved detection of long-term diversion that is possible with more sophisticated data-analysis techniques. Although useful for detection of single, large thefts, the Shewhart chart tends to obscure trends in the materials balance data, such as those caused by long-term diversion.

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 graphic display indicating 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,[35] a type of pattern recognition device very useful for summarizing the results of the various tests and for identifying trends.

To generate the alarm-sequence chart, each sequence causing an alarm is assigned a descriptor that classifies the alarm according to its significance (false-alarm probability) and a pair of integers (r_1,r_2) that are, respectively, the indexes of the initial and final materials balances in the sequence. The alarm-sequence chart is a point plot of r_1 vs r_2 for each sequence that caused an alarm, with the significance range of each point indicated by the plotting symbol. The correspondence of plotting symbol to significance is given in Table VI. The symbol T denotes sequences of such low significance that to examine their extensions would be fruitless; the position of the symbol T on the chart indicates the termination point.

TABLE VI

ALARM CLASSIFICATION FOR THE ALARM-SEQUENCE CHART

Classification (Plotting Symbol)	False-Alarm Probability
A	10^{-2} to 5×10^{-3}
B	5×10^{-3} to 10^{-3}
C	10^{-3} to 5×10^{-4}
D	5×10^{-4} to 10^{-4}
E	10^{-4} to 10^{-5}
F	$<10^{-5}$
T	~ 0.5

For example, consider a sequence of materials balance data beginning at balance number 12, and suppose that one of the tests gives an alarm with a false-alarm probability of 2×10^{-4} at balance number 19. On the alarm-sequence chart for that test, the letter D would appear at the point (12,19). This procedure continues for all possible sequences of the available materials balances. Clearly, it is always true that $r_1 \leq r_2$ so that all symbols lie to the right of the line $r_1 = r_2$ through the origin. Persistent data trends (repeated diversions) cause long alarm sequences ($r_1 << r_2$), and the associated symbols on the alarm chart extend far to the right of the line $r_1 = r_2$.

Decision analysis based on mathematically derived decision functions is appealing because it can quantify intuitive feelings and condense large data collections to a smaller set of more easily understood descriptors (statistics). It can also eliminate personal biases and other errors caused by subjective evaluation of data while providing a degree of consistency for the decision process.

The safeguards system operator must be able to apply the tests quickly and easily in whatever fashion seems most appropriate at the moment, within the limit of good statistical-analysis practice, with reasonable assurance that he can understand the meaning of the results. It is unrealistic to expect all users to be equally proficient in test applications and interpretations. Careful design of the human-engineering aspects of the tests minimizes these difficulties and enhances the utility of the diversion-detection function.

Sample results from sensitivity studies made using these decision-analysis techniques are given in Sec. V.E.

V. DYNAMIC MATERIALS ACCOUNTING STRATEGIES

The reference coconversion process (Sec. I.C and App. A) consists of three identical, independent, parallel process lines, in which one product batch of mixed-oxide powder containing nominally 38 kg of mixed oxide (∿3.2 kg of plutonium) is produced every 2 h. The three lines are instrumented identically (Table III), and dynamic accountability techniques are applied independently to each. A block diagram of a single process line is shown in Fig. 8. The Coprecal model process is described in App. B.

In this section, four strategies for the application of dynamic materials accounting to each process line are compared. The same sets (or a subset) of measurement points and measurements of materials flow, in-process inventory, and holdup are used in each strategy. Dynamic materials balances are formed for each strategy from these measurements. The basic materials accounting equations are given in App. C.

A. Strategy 1

In the first accounting strategy each process line is divided into three UPAAs. As shown in Table VII, Strategy 1 is the basic accounting strategy because Strategies 2, 3, and 4 are combinations of the three UPAAs in Strategy 1. The three UPAAs correspond to unit operations performed in each process line (Fig. 9): (1) feed receiving and blending (batch operations), (2) precipitation and calcination (semicontinuous operations), and (3) reduction, stabilization, and screening (batch operations). In each process line the first and third unit operations are carried out in parallel vessels to achieve the desired throughput (Fig. 8).

Strategy 1 comprises eight dynamic materials balance areas. UPAA 1 (feed receiving and blending) consists of three parallel feed-blend tanks. Dynamic materials balances are drawn every 2 h for process feed transfers from each tank. UPAA 2 consists of one precipitator, four calciners, one primary filter, and one secondary filter. Processing is semicontinuous and a dynamic materials balance is drawn every 2 h. UPAA 3 consists of four parallel reduction-stabilization stations and one screening station. Dynamic materials balances are drawn for batches from each reduction-stabilization station. Thus, the four dynamic materials balance areas in UPAA 3 overlap at the common screening station.

1. UPAA 1 (Feed-Blend Tanks).

Each feed-blend tank sequentially performs three process steps: feed receiving and blending, sampling and standby to feed the process line, and feeding the process line. Materials accounting and control are maintained at each step.

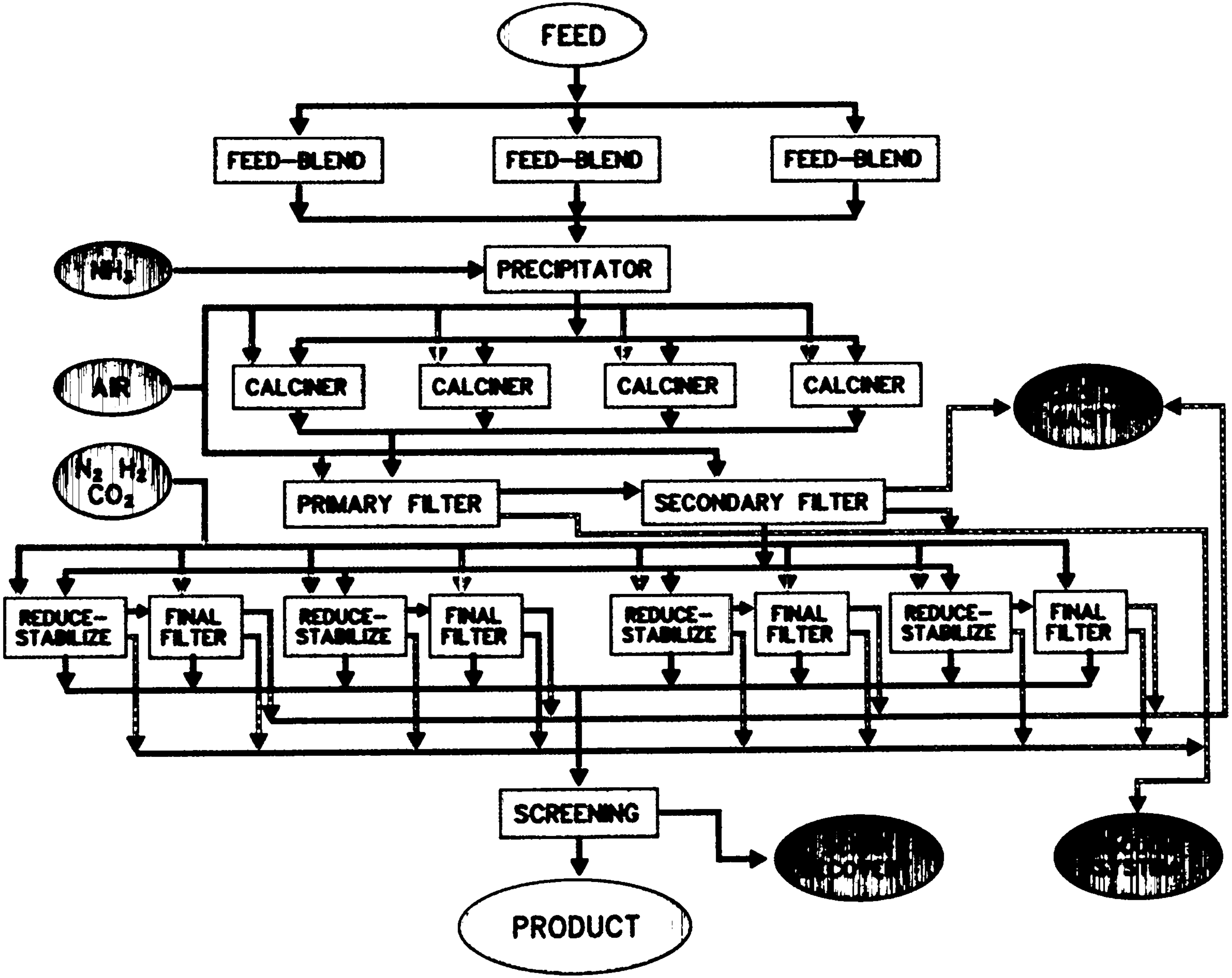


Fig. 8.
Single Coprecal process line.

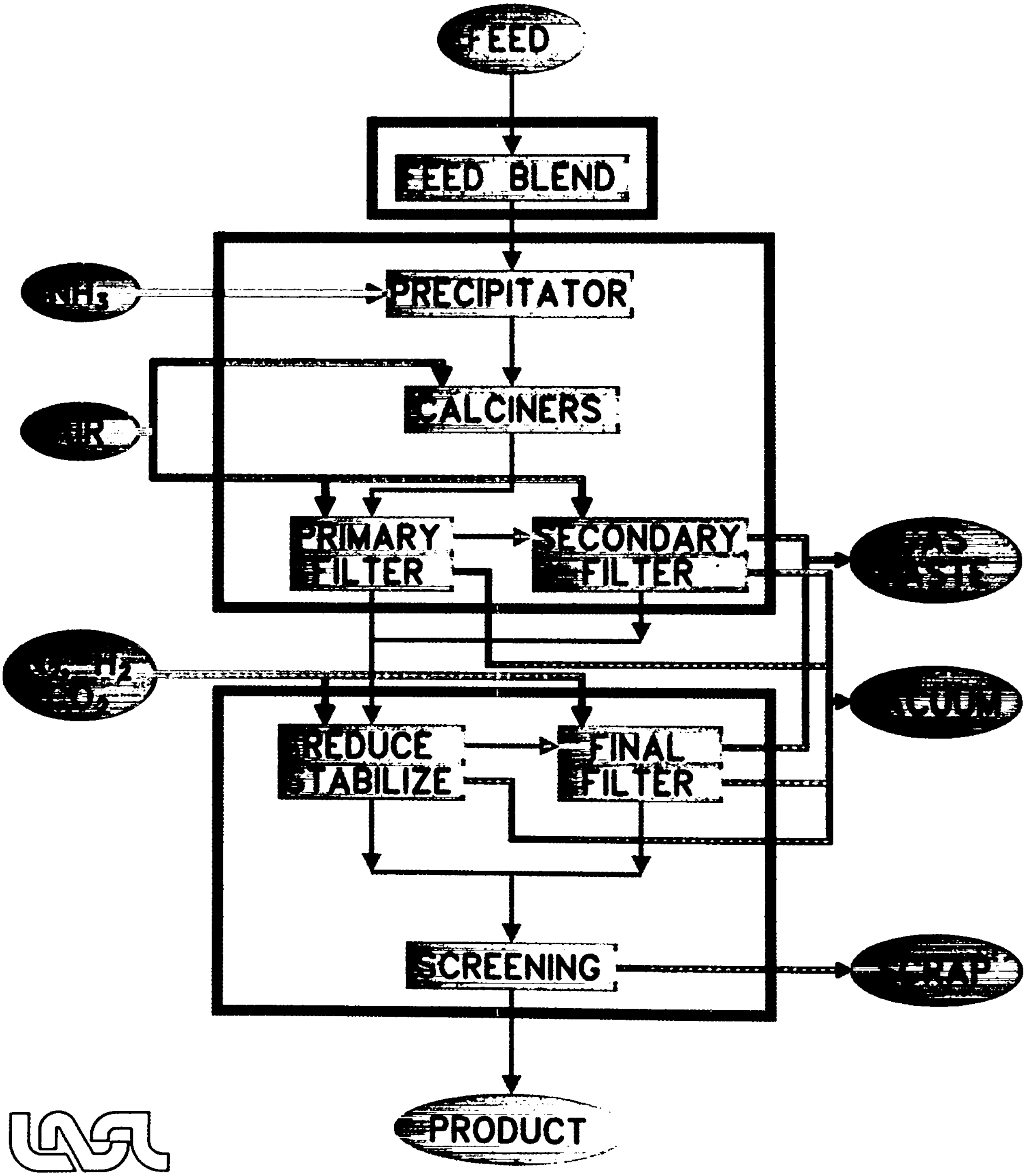


Fig. 9
Coprecal UPAAs.

TABLE VII

ACCOUNTING STRATEGIES

Accounting Strategy	Feed Receiving and Blending	Precipitation and Calcination	Reduction, Stabilization, and Screening
1	UPAA 1	UPAA 2	UPAA 3
2	UPAA 12		UPAA 3
3	UPAA 123		
4	UPAA 1	UPAA 23	

a. Feed Receiving and Blending. Product solution from the chemical separations facility and batch make-up solution are transferred to the feed-blend tanks at the nominal rate of 250 L/min. Each time a tank is filled, a materials balance can be drawn about the entire transfer by combining initial and final in-process inventory measurements of the chemical separations product-storage tank and the coconversion feed-blend tank.

If we assume that the product-storage and feed-blend tanks have similar capacities, 13 000 L of solution containing 625 kg of plutonium would be transferred from one product-storage tank to fill one feed-blend tank. The materials balance standard deviation for such a transfer is estimated to be ∿4.4 kg of plutonium, assuming that the uncertainty in the measured plutonium content of each tank is ∿0.5%. The materials balance standard deviation is proportional to the amount of material transferred. Therefore, if the 13 000-L transfer were divided into smaller transfers and a materials balance were drawn for each small transfer, the uncertainty in each transfer measurement would be reduced. The improvement in materials control obtained by dividing the large transfers into smaller ones would be further enhanced by applying the decision-analysis algorithms described in Sec. IV.C, especially the Kalman filter algorithms, to the small transfer measurements.

b. Standby. After a feed-blend tank has been filled and the solution has been adjusted to the desired plutonium concentration, it is sampled for chemical analysis and certification. Certified tanks are isolated and monitored continuously to preclude the possibility of tampering with the tank or its contents.

c. Feeding the Process. One feed-blend tank feeds the process line until a heel of ∿160 L (∿6.4 kg plutonium) is reached. Each tank feeds the process continuously for one campaign (16.5 days) at a nominal rate of 0.67 L/min. Dynamic materials balances can be drawn by combining the plutonium concentration obtained from chemical analysis with volume or mass and flow rate measurements. The flow rate in the process-feed stream is measured every 0.5 h and the solution volume or mass in the tank is measured every 2 h.

Accounting for materials in large tanks is difficult because small relative errors in the concentration and the volume or mass measurements can result in large absolute errors in the measured SNM content. For this reason, both mass and volume measurements were considered for dynamic materials accounting in the feed-blend UPAA. Measurement precisions and accuracies of 0.1-1% were assumed for each measurement type. The resulting materials control sensitivities were compared and the results are summarized in Table VIII. The separate contributions to the materials balance standard deviations from tank-inventory and feed-transfer measurements are shown. Results are given for a 2-h accounting period and for an entire campaign (16.5 days).

The results indicate that 2-h materials balances based on volume measurements become much more sensitive than mass measurements as the tank is emptied. It must be emphasized that this effect is produced by the models assumed to describe the mass and volume measurements. An additive (absolute error) model is assumed to describe the mass measurements,[3] and a multiplicative (relative error) model is assumed to describe the volume measurements.[1] These models approximate real measurement systems satisfactorily in many cases,[29] but they are idealized limits to the actual measurement behavior. The relative merits of mass and volume measurements must be determined by tests and evaluations of actual measurement systems.

At present, there is much more experience in nuclear process applications with volume measurements than with solution mass measurements (Ref. 1, App. C). Precision and accuracy for state-of-the-art volume measurements are estimated to be 0.1-0.5% when tanks are measured in static mode. A feed-blend tank feeding the process continuously is expected to have larger uncertainties in the dynamic volume (or mass) measurements than there would be in static tank measurements. In the rest of the analysis we assume that dynamic volume measurements having 1% relative instrument precision and 1% relative calibration are made on the feed-blend tanks every 2 h.

TABLE VIII

COMPARISON OF VOLUME AND MASS MEASUREMENTS FOR THE FEED-BLEND TANKS (UPAA 1)

	Standard Deviation (kg Pu)			
	Mass		Volume	
	0.1%	1.0%	0.1%	1.0%
Tank inventory				
Full (625 kg Pu)	0.9	9.1	0.9	9.1
Heel (6.4 kg Pu)	0.9	8.9	0.02	0.09
2-h accounting period				
Full				
Pu flow	0.04	0.04	0.04	0.04
Change in tank inventory	0.9	8.9	0.9	8.9
Materials balance	0.9	8.9	0.9	8.9
Heel				
Pu flow	0.04	0.04	0.04	0.04
Change in tank inventory	0.9	8.9	0.01	0.1
Materials balance	0.9	8.9	0.04	0.1
Campaign accounting period				
Pu flow	6.2	6.2	6.2	6.2
Change in tank inventory	2.0	9.1	2.0	9.1
Materials balance	6.5	11.0	6.5	11.0

Either volume or mass measurements would be facilitated and measurement errors would be reduced if the feed-blend tanks could be isolated for each measurement. The following alternative arrangement of process equipment at the head end might be considered. If two small aliquot tanks each with, say, 2-h operating capacities (80 L) were inserted in the process feed stream after the feed-blend tanks, one tank could be filling while the other is feeding the process. Materials balances could be formed, therefore, by combining static tank measurements (volume or mass) for each 2-h transfer. This arrangement precludes the need for flow measurements in the process feed stream, albeit at the expense of added tankage, piping, and transfer valving. With this arrangement and assuming 0.3% volume measurements (precision and accuracy), the 2-h materials balance standard deviations are estimated to be 2.7 kg of plutonium at the beginning and 0.04 kg of plutonium at the end of a campaign.

2. UPAA 2 (Precipitation and Calcination). The second UPAA includes the precipitator, the four parallel calciners, the primary filter, and the secondary filter (Fig. 9). The process is semicontinuous, producing batches at the primary filter every 2 h. A product batch consists of ∿38 kg of mixed oxide containing ∿3.2 kg of plutonium. A dynamic materials balance is formed for each batch when it is removed from the primary filter by combining measurements of plutonium concentration and bulk flow in the process feed stream from UPAA 1 with measurements of plutonium in the filter batch and measurements of the in-process inventory in each vessel of UPAA 2. Minor sidestreams to the vacuum and off-gas treatment systems are not measured in Strategy 1.

Materials balance standard deviations for 2 h (1 balance), 1 day (12 balances), 1 week (84 balances), and 1 month (336 balances) are given in Table IX. The effect of improving the relative precision of the in-process inventory measurements from 10 to 5% is shown. The effect is small because the materials balance uncertainties are dominated by covariances between the process feed measurements, especially covariances between the flow measurements. In Strategy 1 the flow measurements are recalibrated only at the beginning of each campaign, and all the flow measurements within a campaign are correlated.

3. UPAA 3 (Reduction, Stabilization, and Screening). The third UPAA consists of four parallel reduction-stabilization stations and a single screening station. Each reduction-stabilization station discharges a batch every 8 h. Processing is sequenced through the four stations so that a batch is produced every 2 h. The screen is cleaned into a scrap container after each batch.

TABLE IX

MATERIALS BALANCE UNCERTAINTY
STRATEGY 1, UPAA 2

Accounting Period	Standard Deviation (kg Pu)	
	10%	5%
2 hours	0.34	0.27
1 day	0.70	0.67
1 week	3.2	3.2
1 month	8.6	8.6

A dynamic materials balance can be formed every 8 h for batches from each reduction-stabilization station. Each materials balance is formed by combining measurements of the plutonium holdup in the reduction-stabilization filters before and after the batch is processed with measurements of the plutonium contents of the batch before reduction-stabilization and after screening. The scrap from each batch also is measured.

Materials-balance standard deviations for UPAA 3 are shown in Table X. Materials balance uncertainties for 8 h (1 batch), 1 day (3 batches), 1 week (21 batches), and 1 month (84 batches) are shown. The materials balance uncertainties in UPAA 3 are much smaller than those of UPAA 1 and 2 because the in-process inventory is smaller and the batch measuring instruments are recalibrated daily.

B. Strategies 2 and 3

In the second accounting strategy UPAA 1 and UPAA 2 are combined to form a single feed-precipitation-calcination UPAA denoted by UPAA 12 (Table VII). Dynamic materials balances are drawn for UPAA 12 every 2 h by combining the appropriate measurements from UPAA 1 and UPAA 2. Flow measurements of the process feed stream are not required for this strategy. In Strategy 2, the reduction-stabilization UPAA (UPAA 3) is the same as in Strategy 1 (Table X).

In the third accounting strategy, the three UPAAs are combined into UPAA 123 consisting of one entire process line (Table VII). Dynamic materials balances are formed every 2 h. None of the transfer measurements between the UPAAs are used in Strategy 3.

TABLE X

MATERIALS BALANCE UNCERTAINTY
STRATEGY 1, UPAA 3

Accounting Period	Standard Deviation (kg Pu)
8 hours	0.10
1 day	0.17
1 week	0.44
1 month	0.89

Materials balance standard deviations for Strategies 2 and 3 are given in Table XI. In all cases the materials balance uncertainties are dominated by the uncertainties in the feed-blend tank volume measurements. Note that results are given for full and nearly empty tanks. The effect of the multiplicative measurement-error model assumed for the volume measurements is evident. Materials accounting sensitivities for Strategies 2 and 3 can be improved primarily by reducing the feed-blend tank measurement errors.

C. Strategy 4

The fourth accounting strategy separates the feed-blend tanks (UPAA 1), as in Strategy 1, and places the rest of the process line into UPAA 23 (Table VII). Measurements of the plutonium sidestream to the vacuum system are added.

TABLE XI

MATERIALS BALANCE UNCERTAINTY
STRATEGIES 2 and 3

	Standard Deviation (kg Pu)	
	Strategy 2 UPAA 12	Strategy 3 UPAA 123
In-Process inventory		
Full	9.1	9.1
Heel	0.25	0.29
2-h accounting period		
Full		
Net transfer	0.07	0.04
Inventory change	8.9	8.9
Materials balance	8.9	8.9
Heel		
Net Transfer	0.07	0.04
Inventory Change	0.35	0.41
Materials balance	0.36	0.41
Campaign accounting period		
Net Transfer	1.9	0.9
Inventory Change	9.1	9.1
Materials balance	9.3	9.1

Materials-balance standard deviations for four accounting periods are given in Table XII. Under the column labeled "base case," the 1-week and 1-month uncertainties are dominated by covariances between the feed-stream flow measurements. To illustrate the effect of these covariances, the flowmeter was recalibrated daily instead of at the beginning of each campaign. Materials-balance standard deviations are given in Table XII for this "best case." The materials-balance uncertainties for 2-h and 1-day accounting periods are unchanged, but the errors for 1-week and 1-month periods are reduced significantly. These results emphasize that all instruments at key measurement points should be designed and installed for ease of recalibration. The recalibration of on-line instruments such as flowmeters may require the development and use of dynamic calibration techniques or the capability for switching instruments.

D. Diversion-Detection Sensitivities

The ability to detect diversion of plutonium from the Coprecal process by the application of dynamic materials accounting concepts has been estimated for Strategies 1 and 4 using modeling and simulation techniques. The evaluation methodology is discussed in Sec. IV. Results were obtained for the detection of abrupt diversion (large single theft) and of protracted diversion (repeated small thefts). False-alarm probabilities are <5% in all cases.

TABLE XII

MATERIALS BALANCE UNCERTAINTY
STRATEGY 4, UPAA 23

	Standard Deviation (kg Pu)	
Accounting Period	Base Case	Daily Flow Recalibration
2 hours	0.40	0.40
1 day	0.61	0.61
1 week	2.9	1.3
1 month	8.2	2.5

1. Strategy 1, UPAA 1. Detection sensitivities are given in Table XIII for detection times of 2 h (1 balance), 1 day (12 balances), 1 week (84 balances), and 1 campaign (198 balances). Sensitivity ranges are given over a campaign, that is, over the contents of one feed-blend tank. The first value in each sensitivity range is the detection sensitivity for the first day or the first week of a campaign, and the second value is the sensitivity for the last day or the last week of a campaign. The sensitivity improves during a campaign because the tank-volume measurement error decreases as the tank is emptied. Note that the abrupt-diversion-detection sensitivities are averages over the detection times. The sensitivity to abrupt diversion improves as the detection time increases primarily because the SMBT (Sec. IV.C) examines all available data in searching for a single theft.

2. Strategy 1, UPAA 2. Detection sensitivities estimated for UPAA 2 are given in Table XIV for detection times of 2 h (1 balance), 1 day (12 balances), 1 week (84 balances), and 1 month (336 balances).

3. Strategy 1, UPAA 3. Detection sensitivities estimated for UPAA 3 are given in Table XV for detection times of 8 h (1 balance), 1 day (3 balances), 1 week (21 balances), and 1 month (84 balances).

TABLE XIII

DETECTION SENSITIVITY
STRATEGY 1, UPAA 1

Detection Time	Average Diversion per Balance (2 h) (kg Pu)	Total at Time of Detection (kg Pu)
Protracted diversion		
1 day	1.4 -0.13	6.8-1.6
1 week	0.12-0.10	10.1-8.4
1 campaign	0.10	20.4
Abrupt Diversion		
2 hours	26.7 -0.3	26.7-0.3
1 day	11.0 -0.9	11.0-0.9
1 week	4.3 -1.8	4.3-1.8
1 campaign	2.8	2.8

TABLE XIV

DETECTION SENSITIVITY
STRATEGY 1, UPAA 2

Detection Time	Average Diversion per Balance (2 h) (kg Pu)	Total at Time of Detection (kg Pu)
Protracted Diversion		
1 day	0.17	2.0
1 week	0.09	7.2
1 month	0.08	25.
Abrupt Diversion		
2 hours	1.0	1.0
1 day	0.7	0.7
1 week	0.7	0.7
1 month	0.7	0.7

TABLE XV

DETECTION SENSITIVITY
STRATEGY 1, UPAA 3

Detection Time	Average Diversion per Balance (8 h) (kg Pu)	Total at Time of Detection (kg Pu)
Protracted diversion		
1 day	0.17	0.5
1 week	0.06	1.3
1 month	0.03	2.5
Abrupt diversion		
8 hours	0.3	0.3
1 day	0.3	0.3
1 week	0.3	0.3
1 month	0.3	0.3

4. Strategy 4, UPAA 23. The ability of dynamic materials accounting to detect diversion from UPAA 23 is estimated in Table XVI for the base case and the best case, defined previously. Note that in the best case the flow measurements in the feed stream are recalibrated daily and the improvement in sensitivity to protracted diversion obtained by recalibrating is evident.

E. Sample Results

Sample diversion-detection simulation results are given in Figs. 10-37. Each figure shows results obtained with one of the decision-analysis tests described in Sec. IV.C. The four tests included are materials balance, cusum, UDT (labeled average missing material), and SMBT. Each figure shows plots of the test statistic and the corresponding alarm chart for three cases: no diversion (upper), protracted (uniform) diversion (middle), and abrupt (single) diversion (lower). In all cases the diversion levels are the estimated detection sensitivities given in Tables XIII through XVI.

One set of four figures, corresponding to the four tests, is given for each of the following.

Strategy 1, UPAA 1, one campaign of 2-h balances, Figs. 10-13.
Strategy 1, UPAA 2, 1 month of 2-h balances, Figs. 14-17.
Strategy 1, UPAA 3, 1 month of 8-h balances, Figs. 18-21.
Strategy 4, UPAA 23, 1 month of 2-h balances (base case), Figs. 22-25.
Strategy 4, UPAA 23, 1 month of 2-h balances (best case), Figs. 26-29
Strategy 4, UPAA 23, 1 week of 2-h balances (best case), Figs. 30-33.
Strategy 4, UPAA 23, 1 day of 2-h balances (best case), Figs. 34-37.

TABLE XVI

DETECTION SENSITIVITY
STRATEGY 4, UPAA 23

	Base Case		Best Case	
Detection Time	Average Diversion per Balance (2 h) (kg Pu)	Total at Time of Detection (kg Pu)	Average Diversion per Balance (kg Pu)	Total at Time of Detection (kg Pu)
Protracted Diversion				
1 day	0.13	1.6	0.13	1.6
1 week	0.08	6.7	0.04	3.7
1 month	0.07	23.	0.03	8.4
Abrupt Diversion				
2 hours	1.2	1.2	1.2	1.2
1 day	0.7	0.7	0.7	0.7
1 week	0.7	0.7	0.7	0.7
1 month	0.7	0.7	0.7	0.7

For each materials balance (Shewhart) chart, dynamic materials balances are plotted sequentially with 1-σ error bars. The associated alarm charts can indicate only single materials balance alarms. If the materials-balance charts were not labeled, it would be impossible in most cases to tell whether there had been diversion. Remember that the safeguards officer does not have the luxury of comparing charts with and without diversion for the same time period, and, of course, he would not know beforehand if diversion had occurred. The materials-balance charts are limited in their capability to detect diversion except in case of a very large abrupt diversion. This is the main reason why decision-analysis algorithms and alarm sequence charts have been developed.

For each cusum chart, cumulative summations of dynamic materials balances are plotted sequentially with 1-σ error bars. Letter symbols on the associated alarm-sequence charts indicate the length and significance of sequences of dynamic materials balances that generate alarms (Table VI).

Kalman filter estimates of the average amounts of missing material per balance period are plotted sequentially with 1-σ error bars, along with their associated alarm charts. Note that the UDT is more sensitive generally than the cusum test, that is, the UDT gives more alarms having higher significance in the diversion cases. Smoothed materials-balance estimates are also plotted with 1-σ error bars. The SMBT is especially helpful in locating an abrupt diversion.

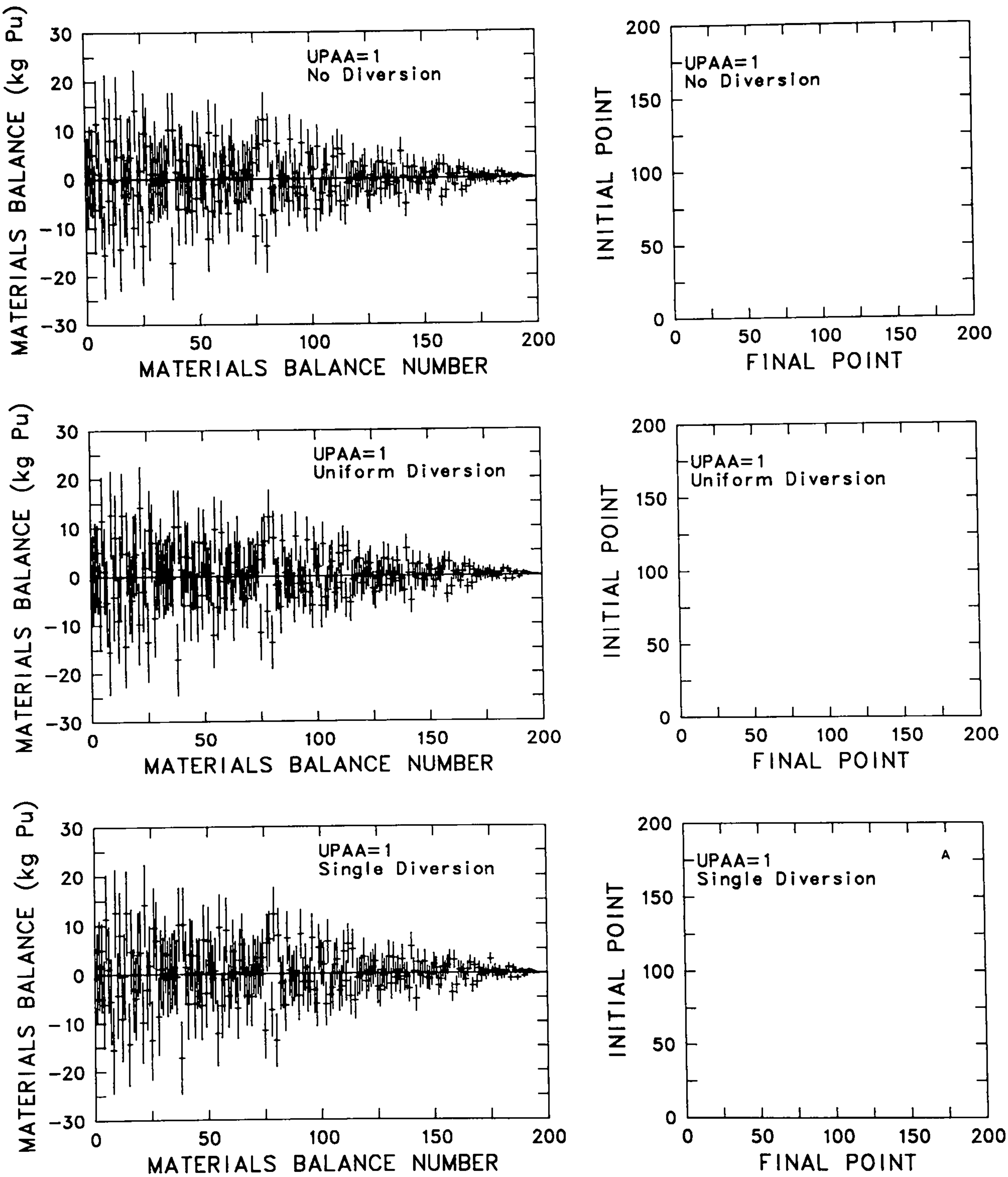


Fig. 10.
Strategy 1, UPAA 1, one campaign: materials balance and alarm charts.

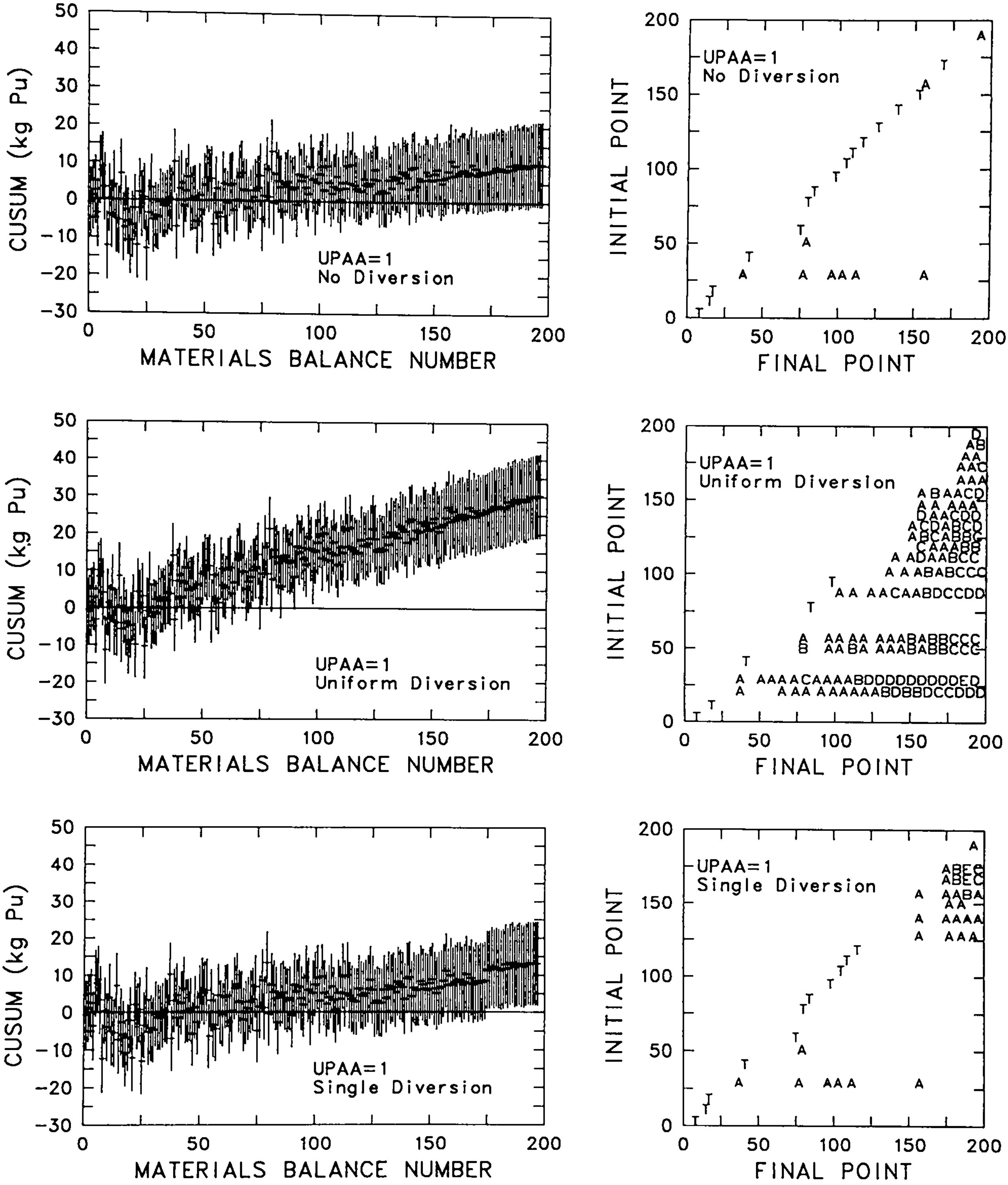


Fig. 11.
Strategy 1, UPAA 1, one campaign: cusum and alarm charts.

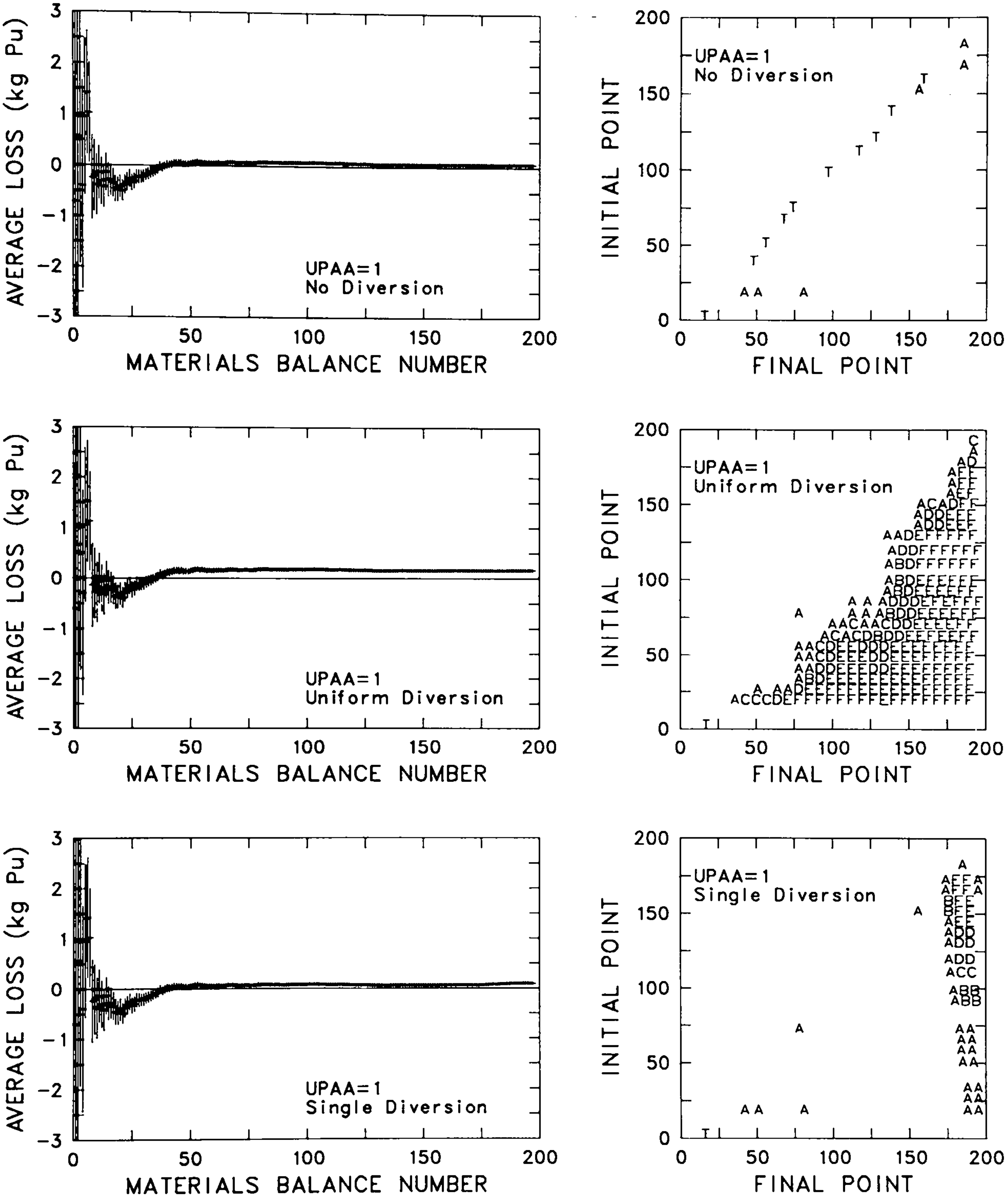


Fig. 12.
Strategy 1, UPAA 1, one campaign: UDT and alarm charts.

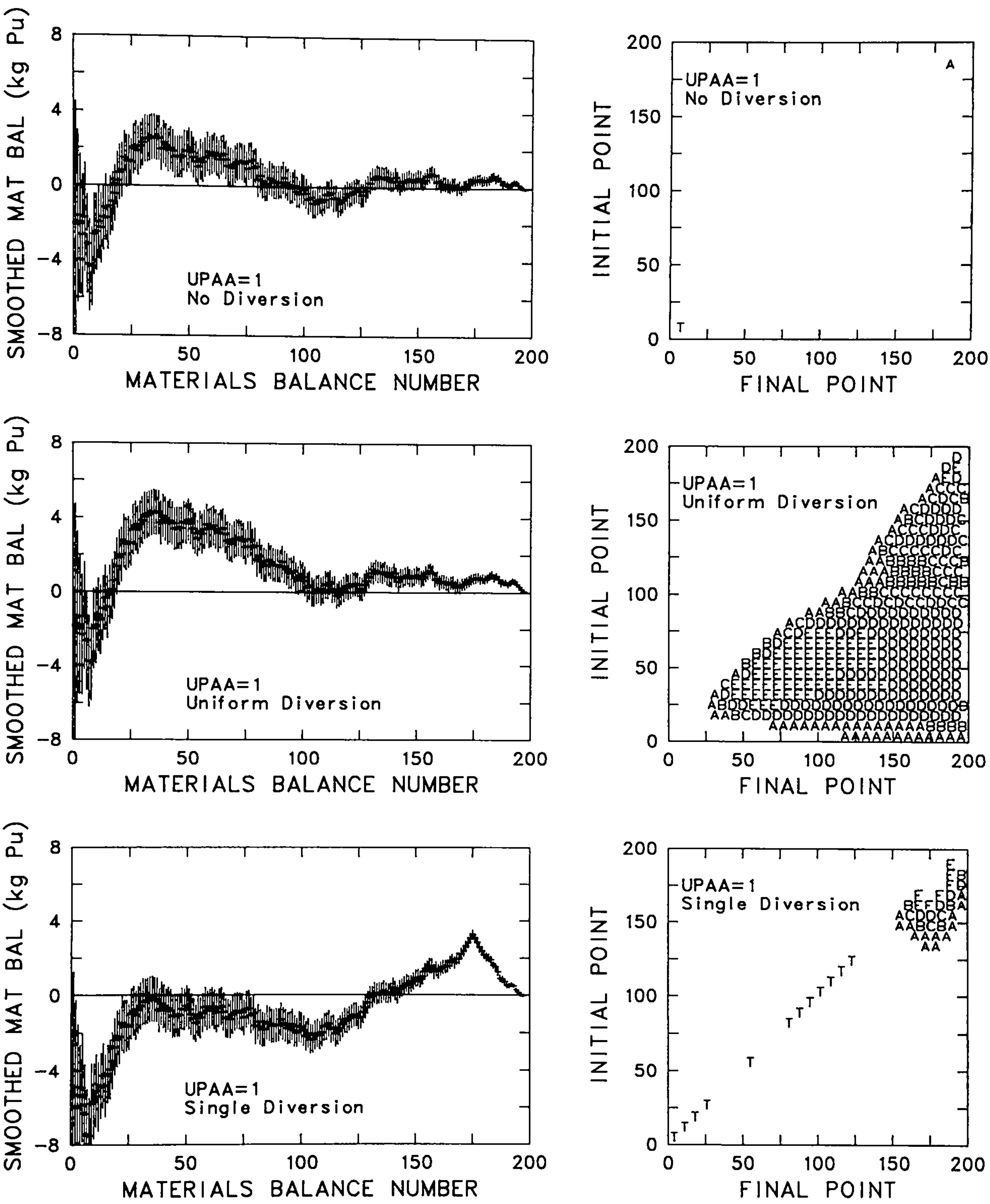


Fig. 13.
Strategy 1, UPAA 1, one campaign: SMBT and alarm charts.

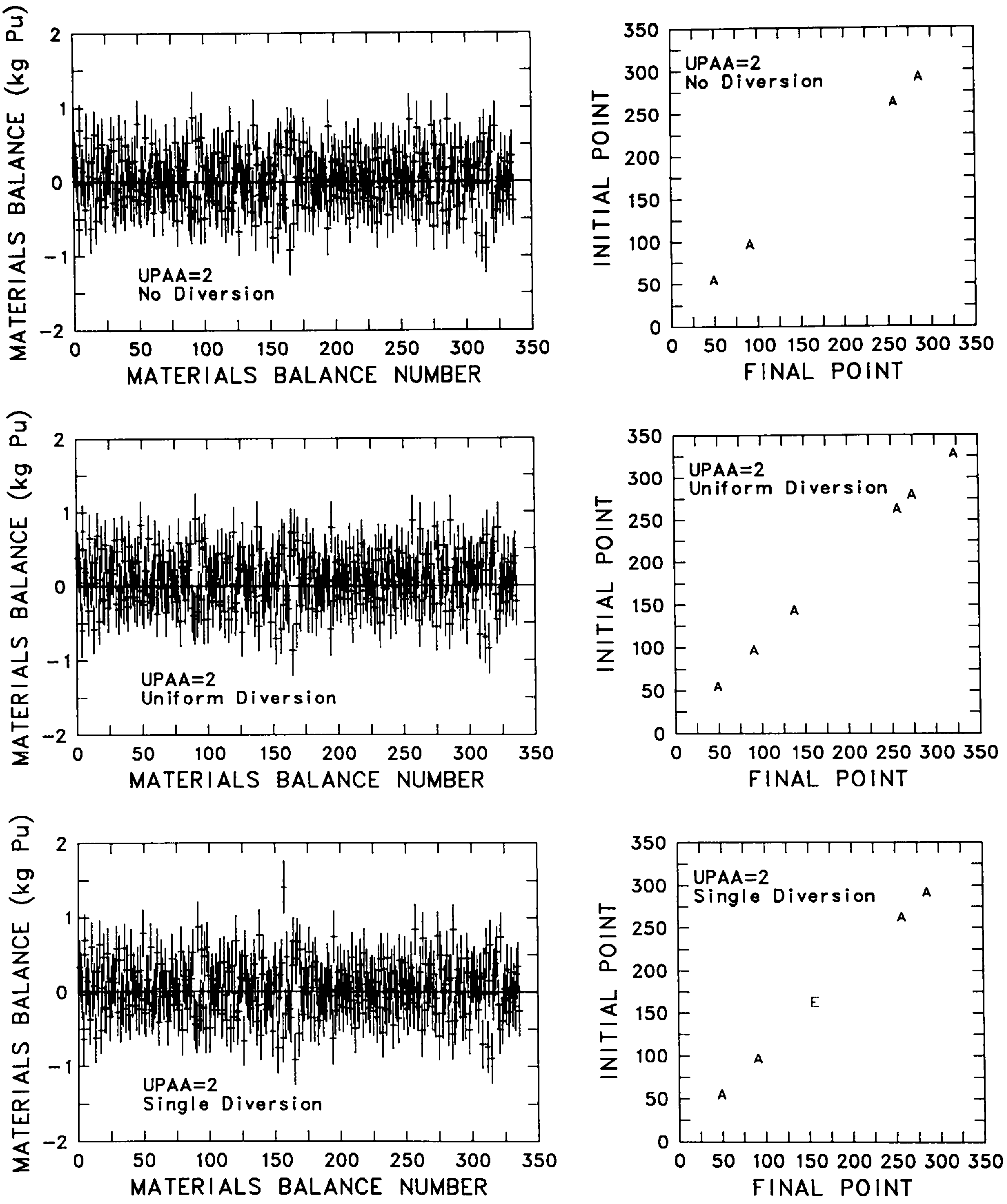


Fig. 14.
Strategy 1, UPAA 2, 1 month: materials balance and alarm charts.

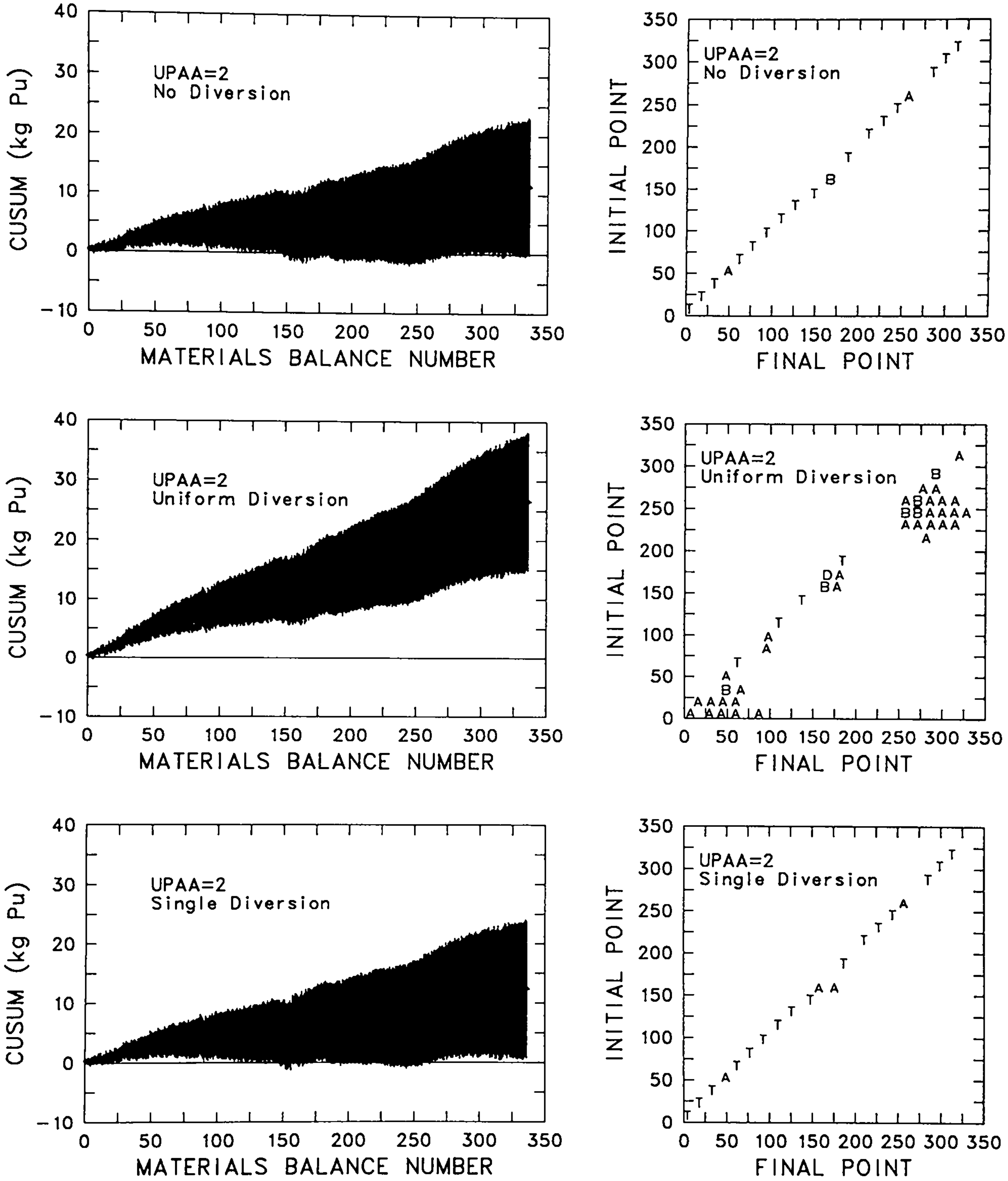


Fig. 15.
Strategy 1, UPAA 2, 1 month: cusum and alarm charts.

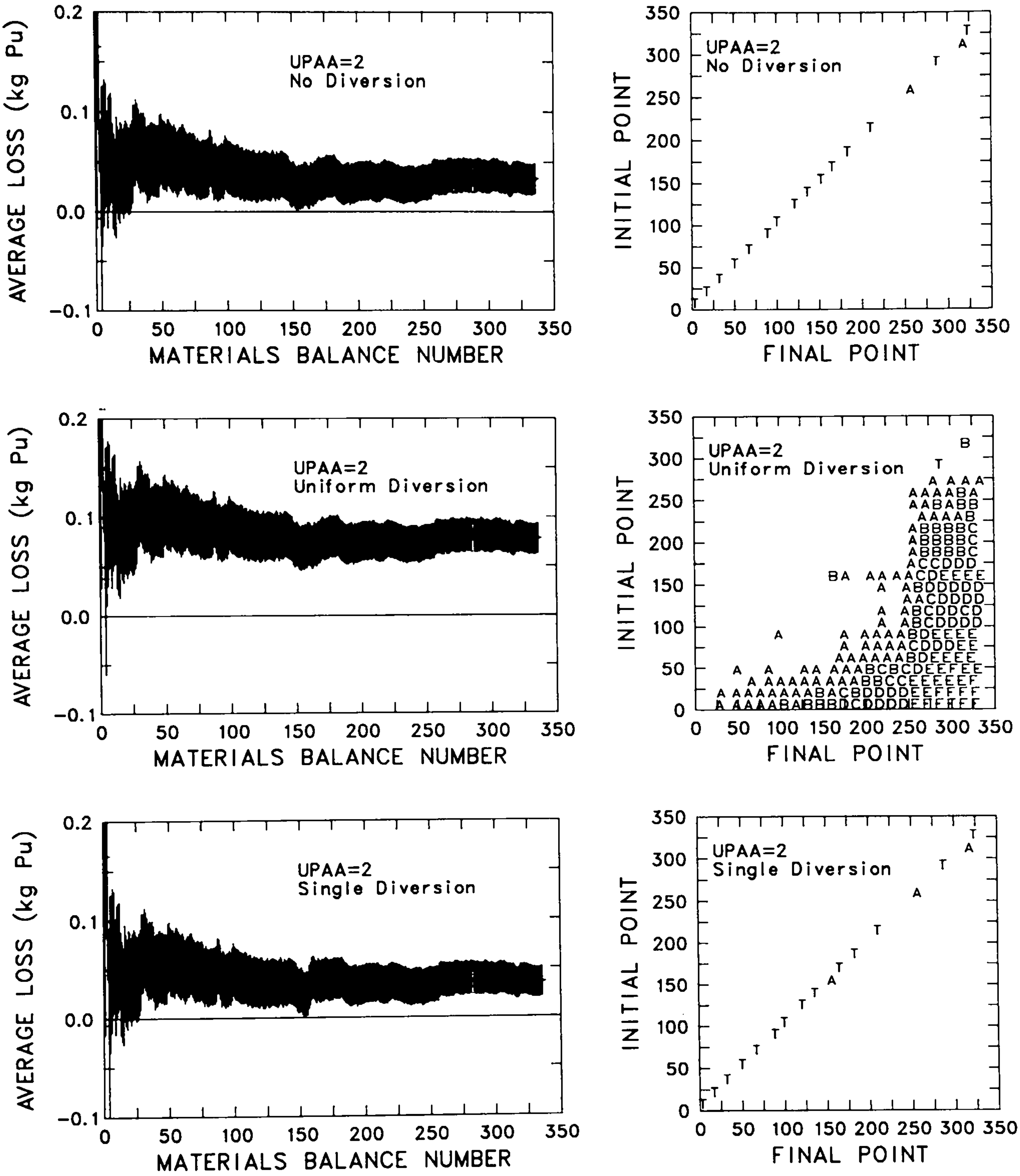


Fig. 16.
Strategy 1, UPAA 2, 1 month: UDT and alarm charts.

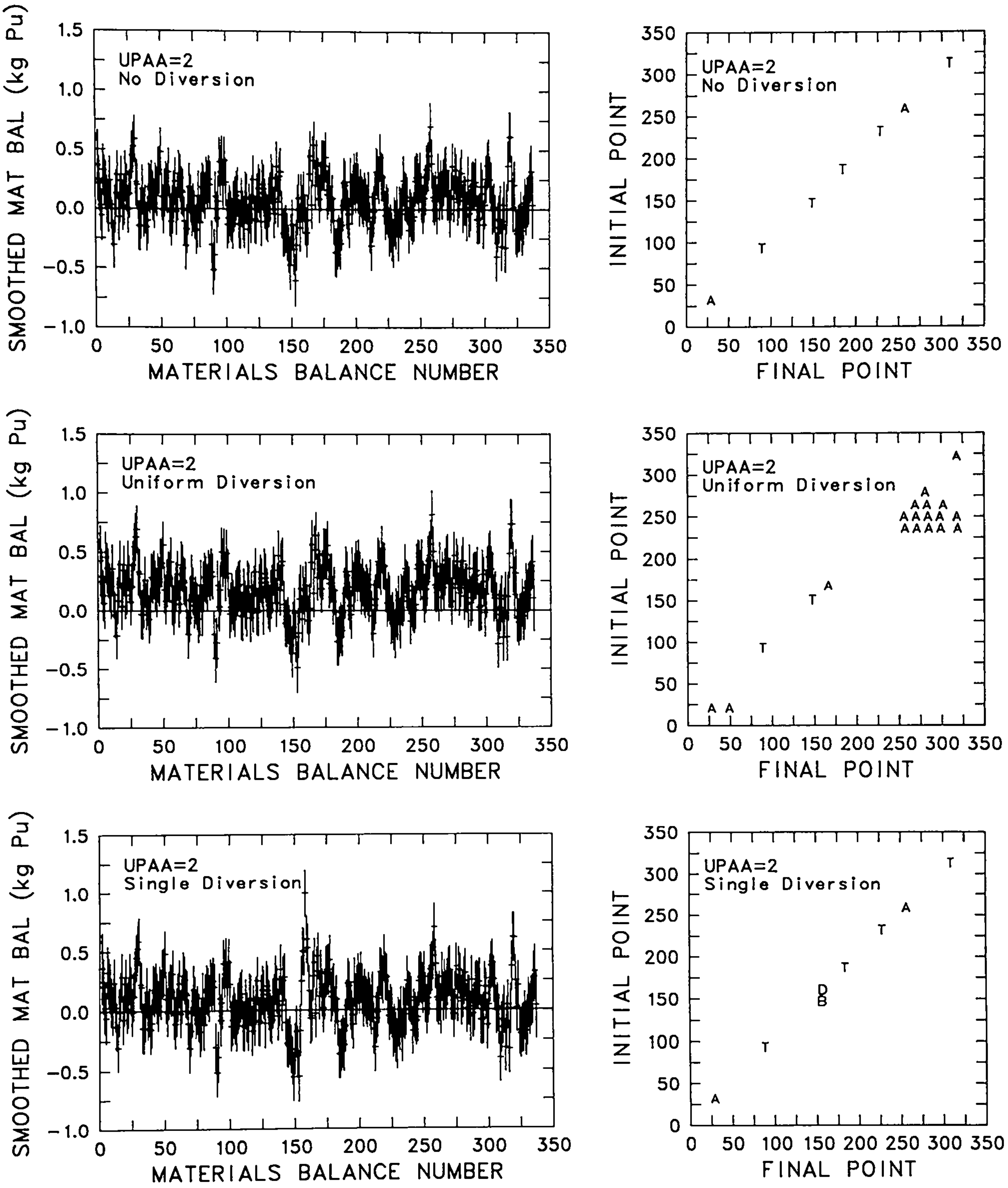


Fig. 17.
Strategy 1, UPAA 2, 1 month: SMBT and alarm charts.

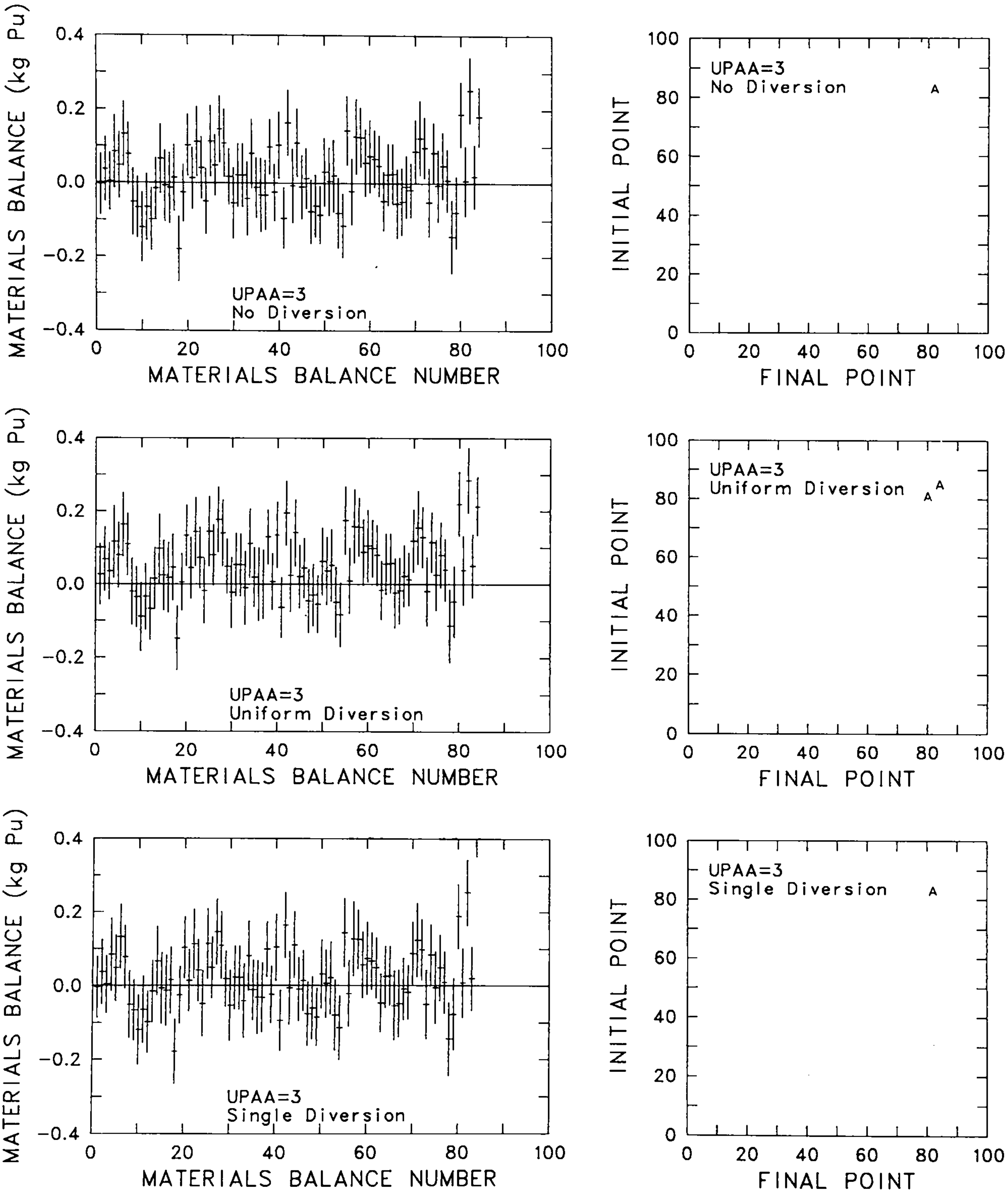


Fig. 18.
Strategy 1, UPAA 3, 1 month: materials balance and alarm charts.

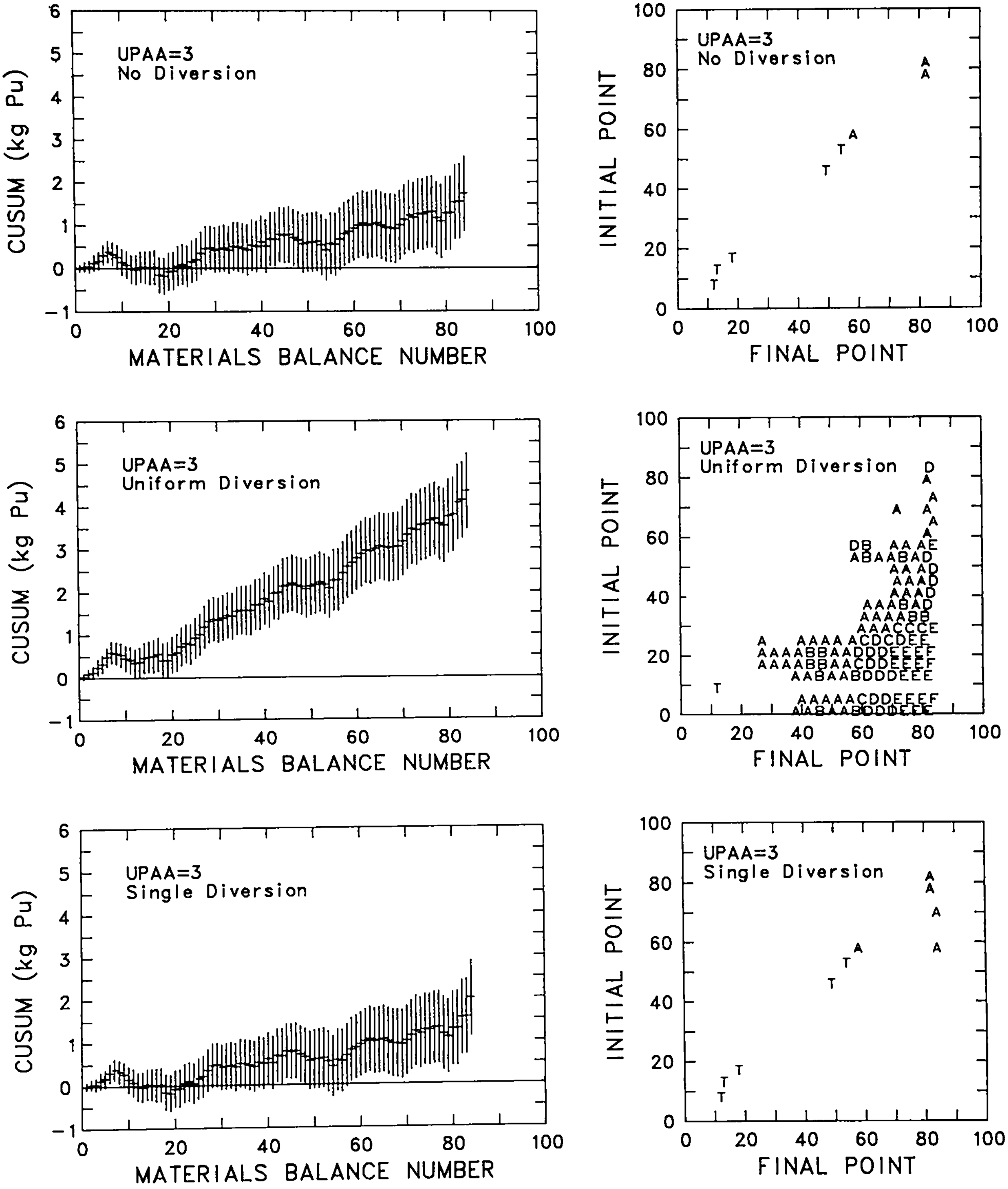


Fig. 19.
Strategy 1, UPAA 3, 1 month: cusum and alarm charts.

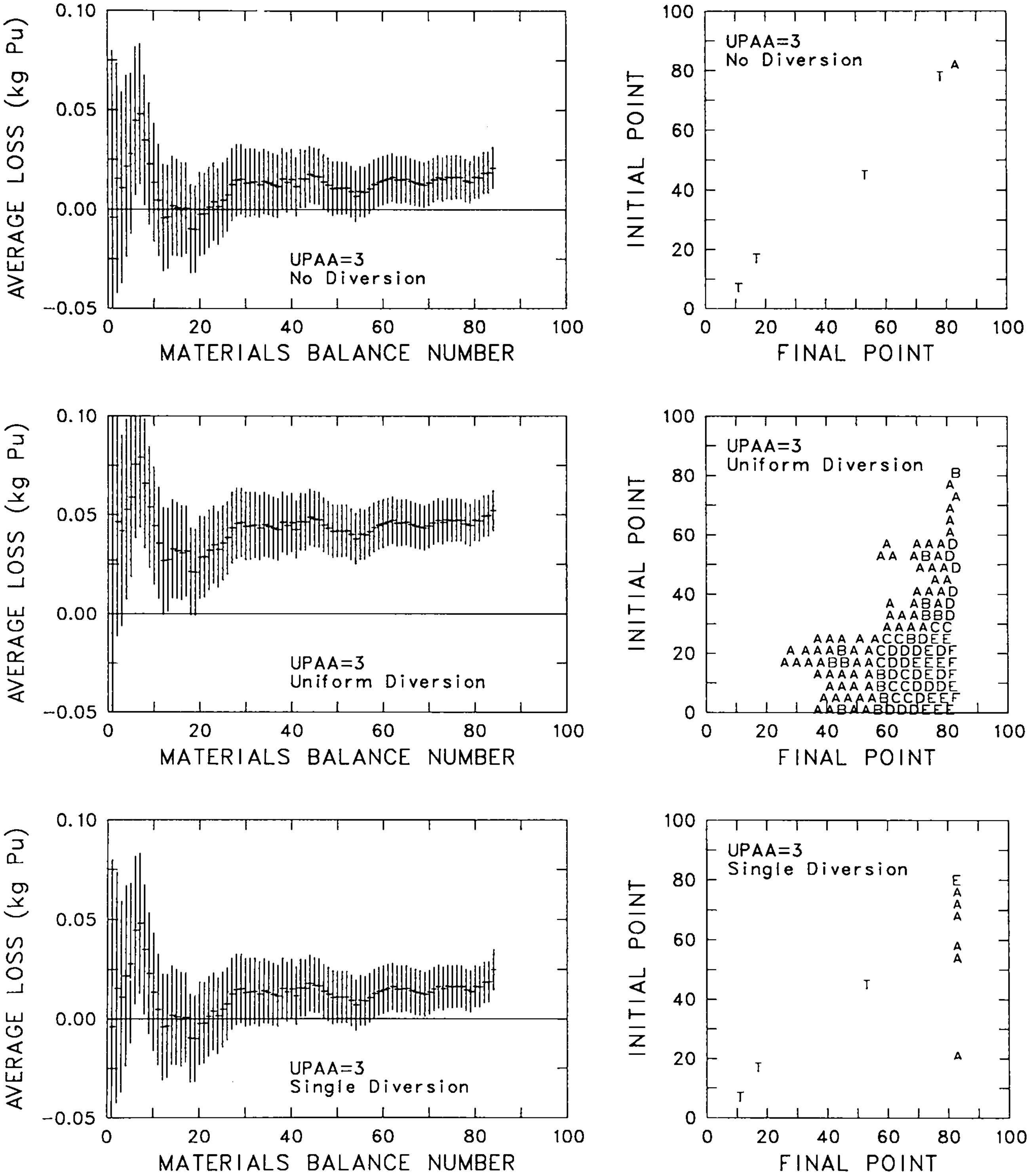


Fig. 20.
Strategy 1, UPAA 3, 1 month: UDT and alarm charts.

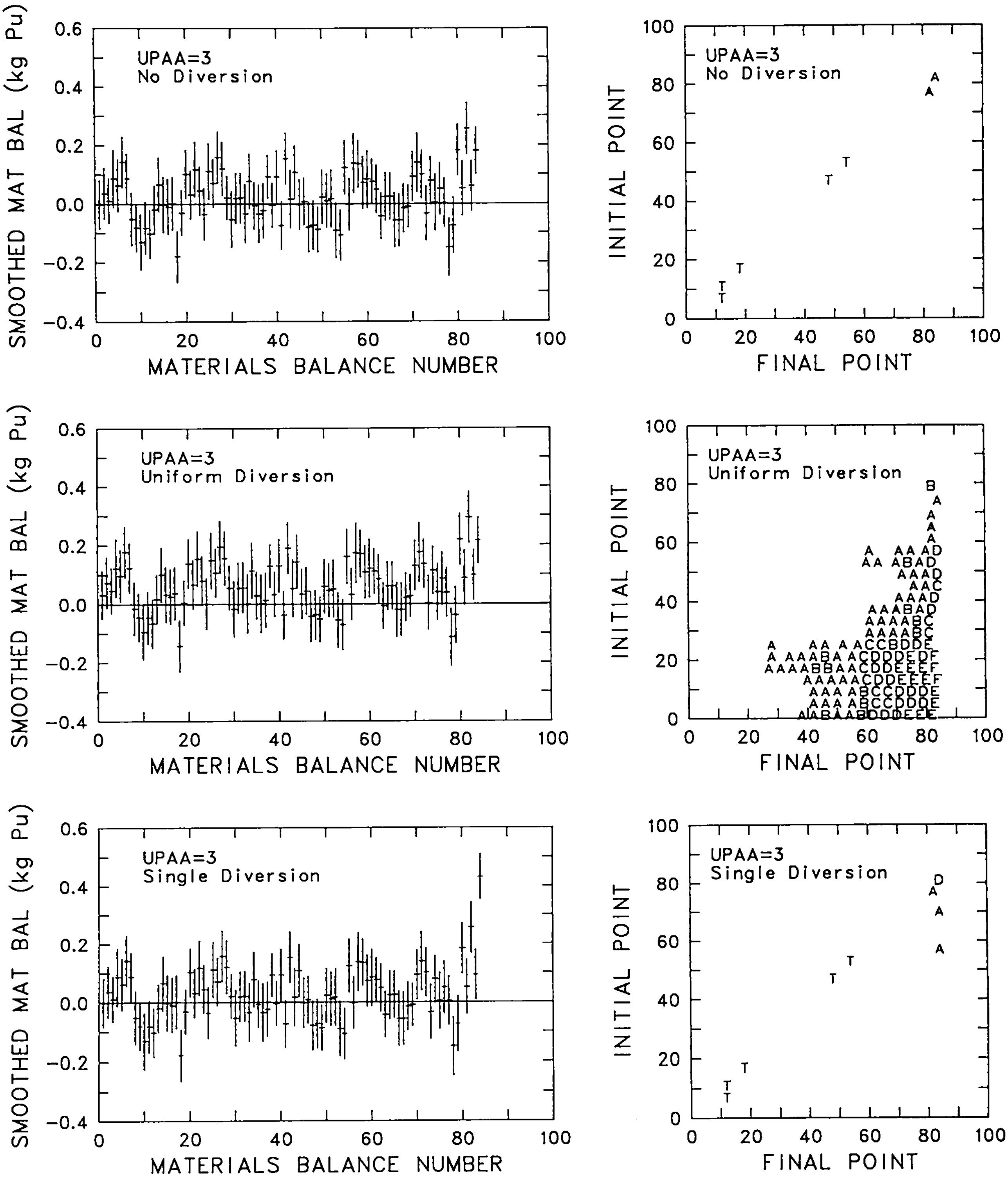


Fig. 21.
Strategy 1, UPAA 3, 1 month: SMBT and alarm charts.

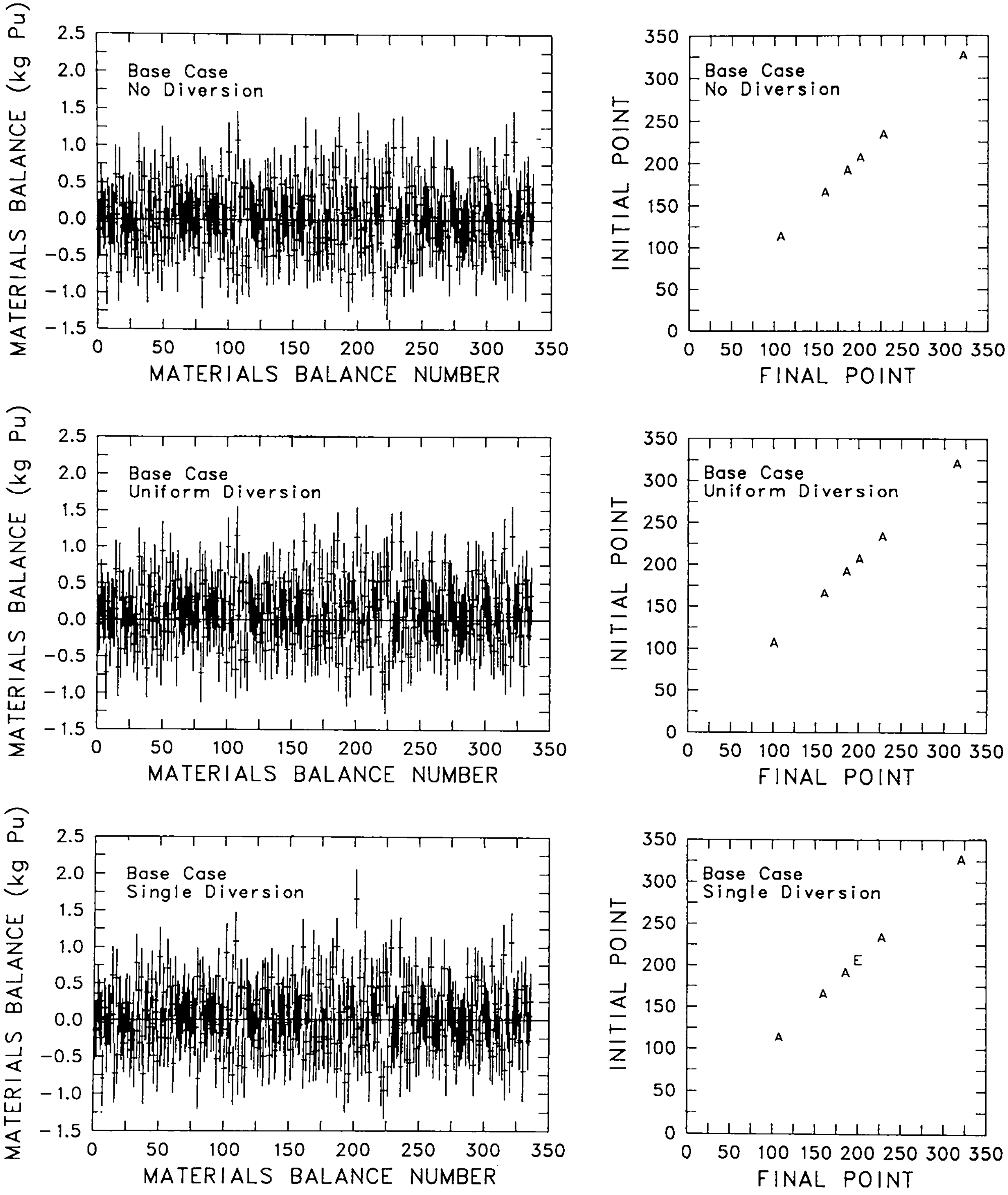


Fig. 22.
Strategy 4, UPAA 23(base case), 1 month:
materials balance and alarm charts.

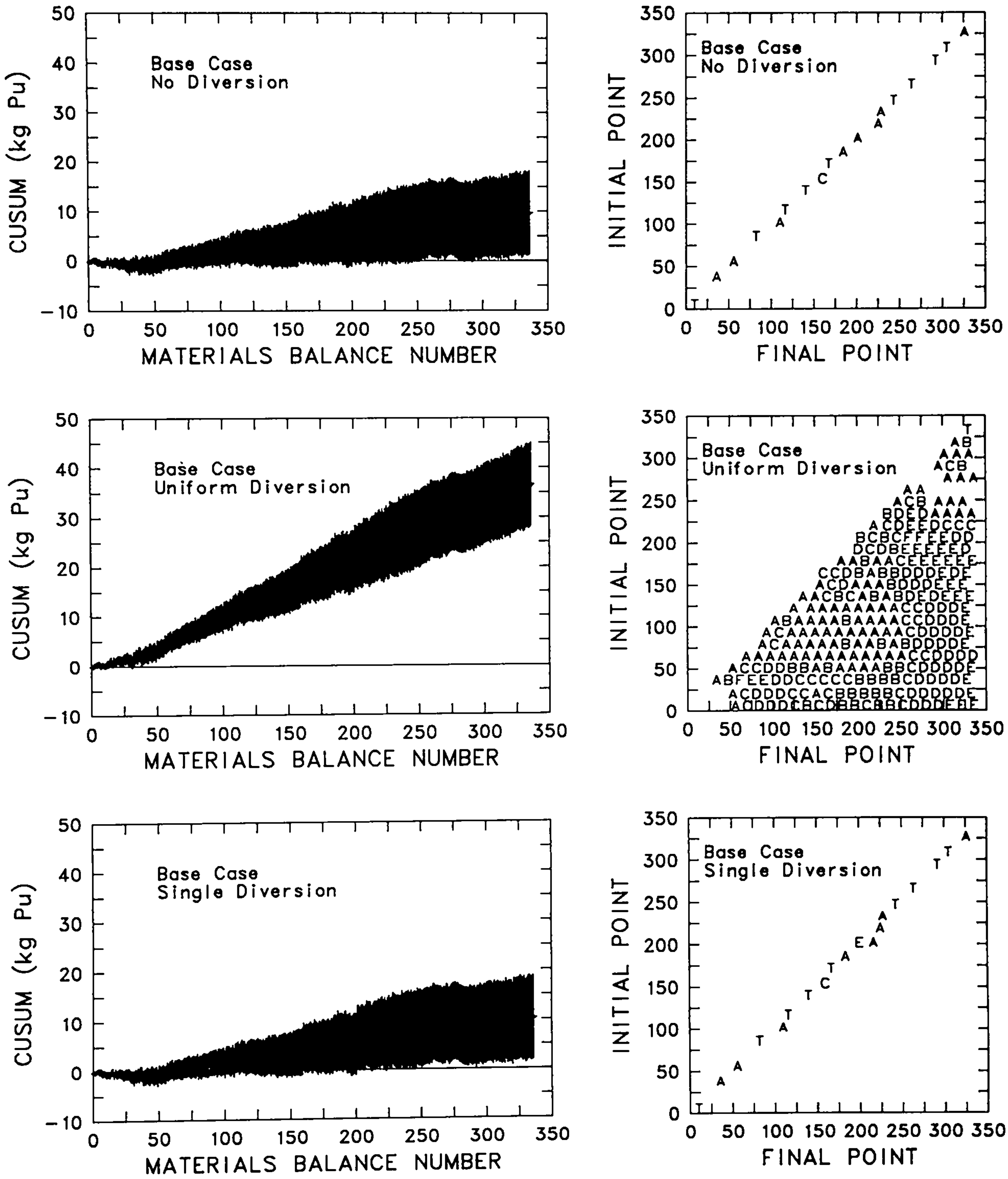


Fig. 23.
Strategy 4, UPAA 23(base case), 1 month: cusum and alarm charts.

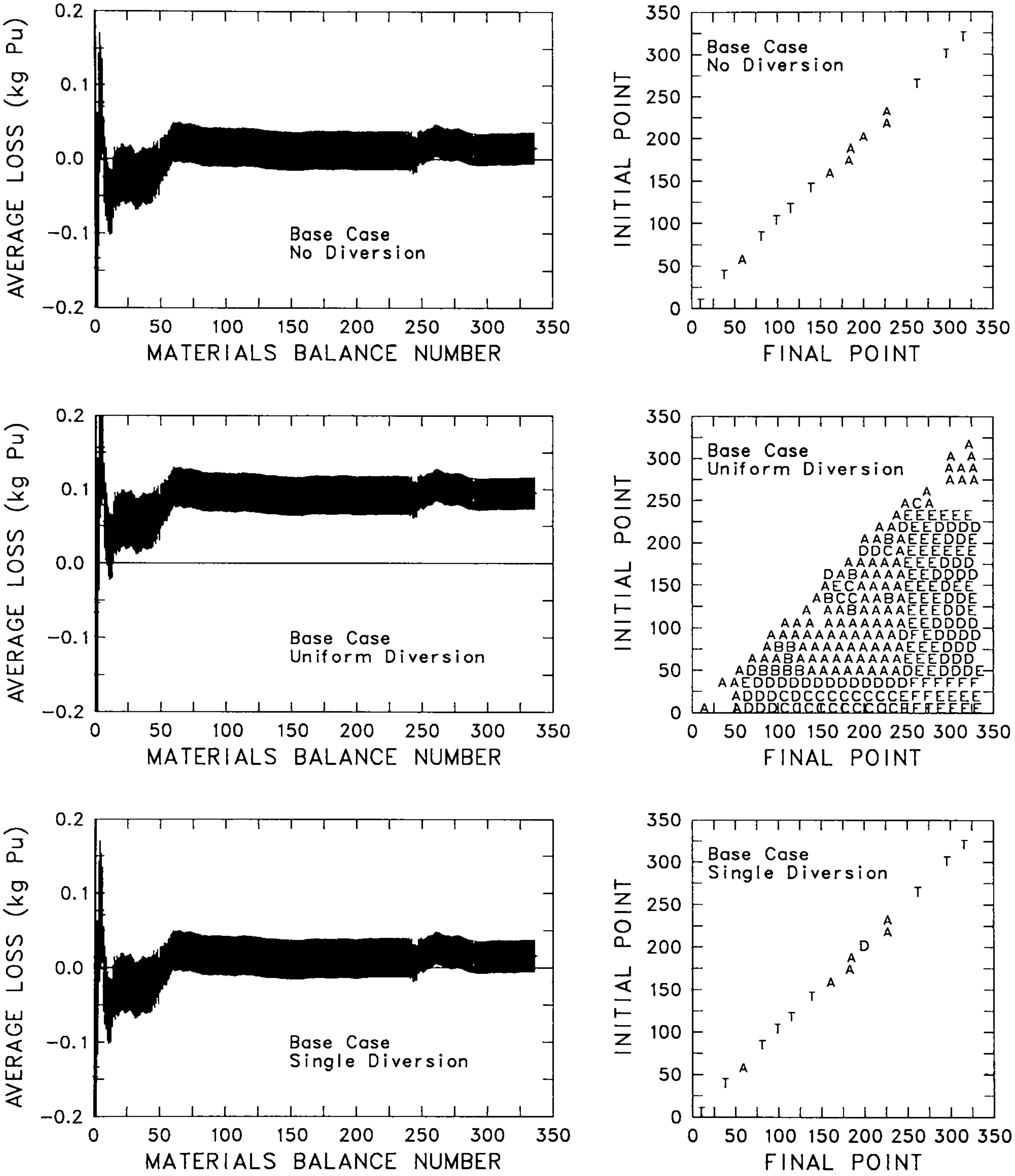


Fig. 24.
Strategy 4, UPAA 23(base case), 1 month: UDT and alarm charts.

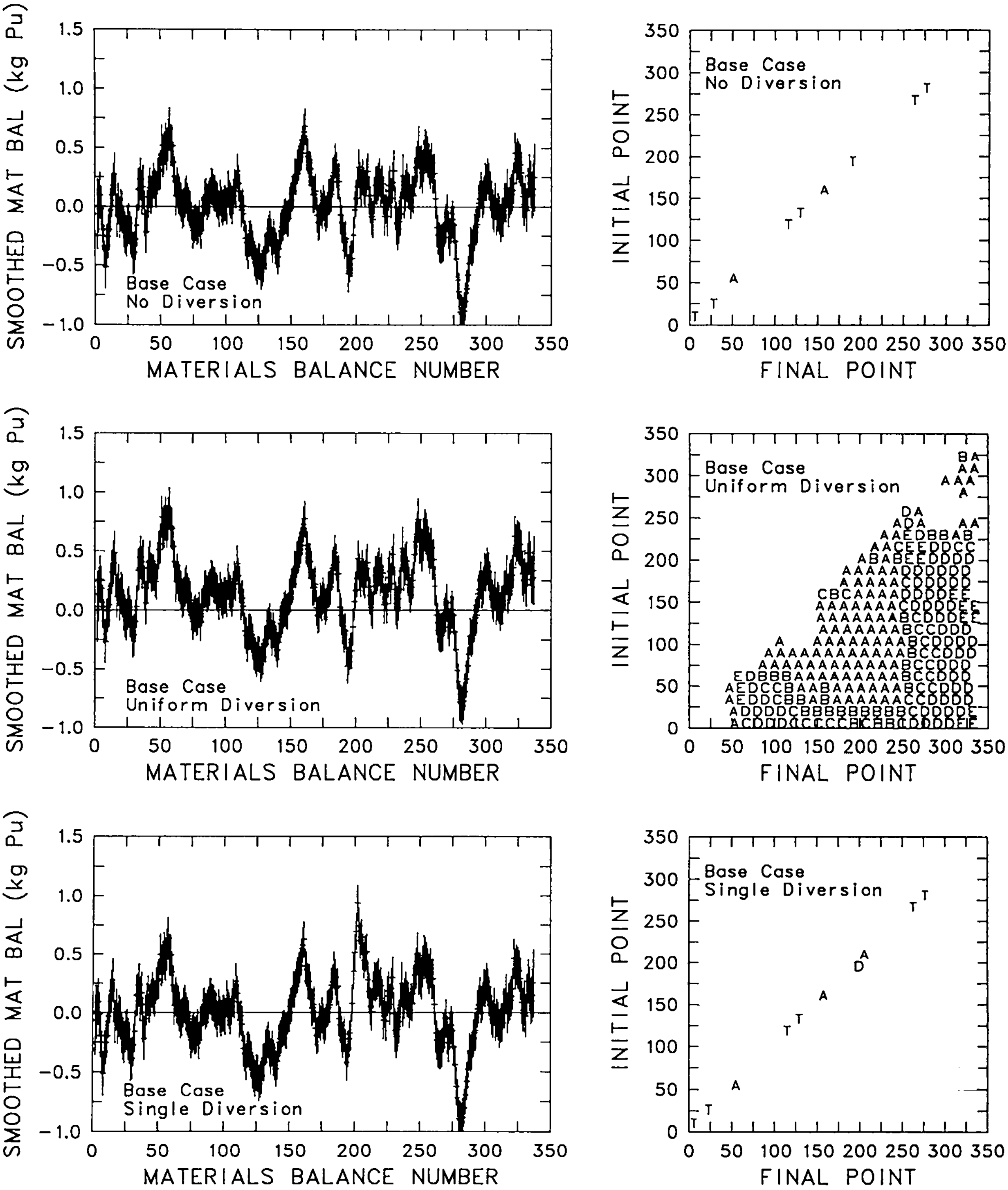


Fig. 25.
Strategy 4, UPAA 23 (base case), 1 month: SMBT and alarm charts.

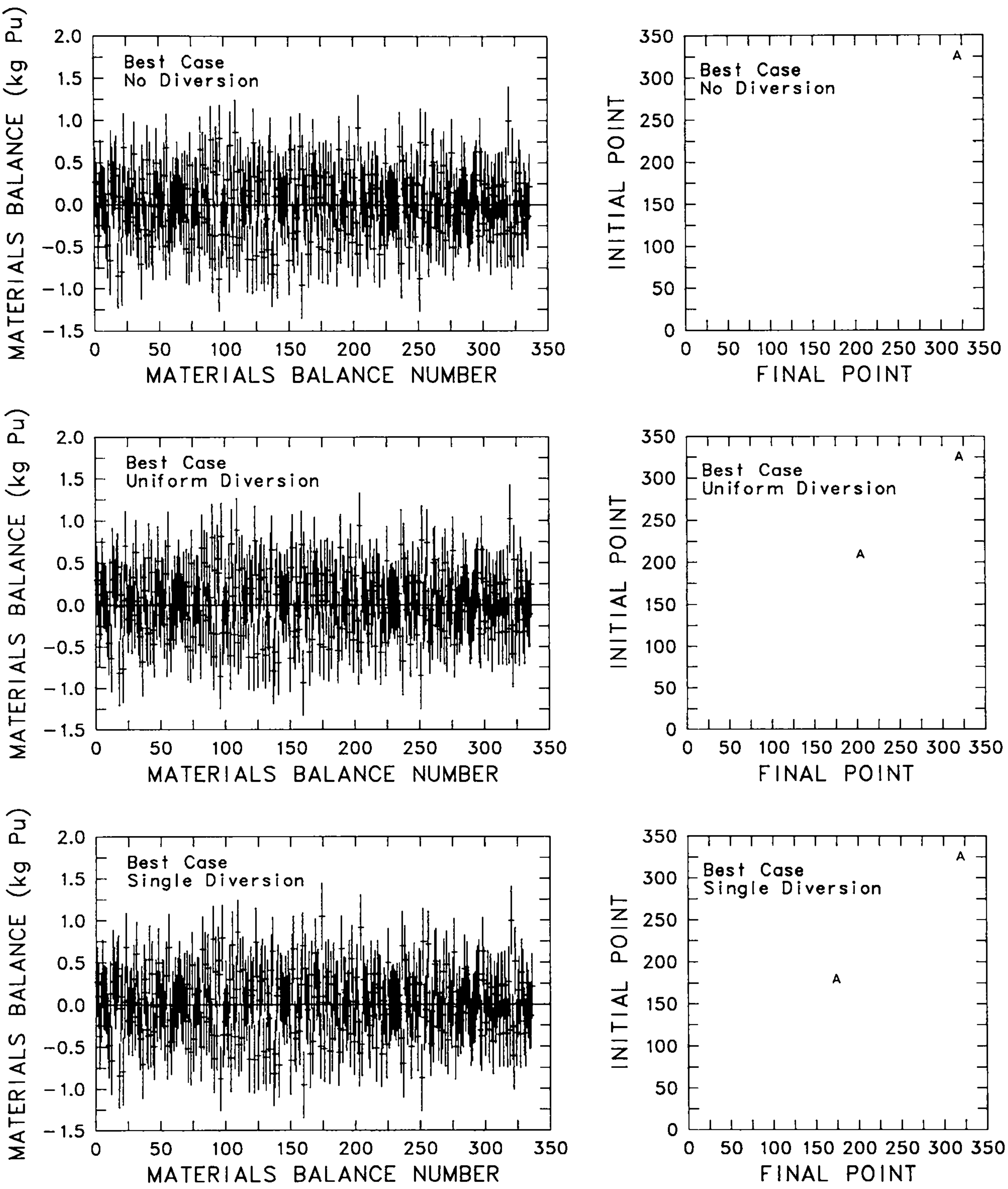


Fig. 26.
Strategy 4, UPAA 23(best case), 1 month:
materials balance and alarm charts.

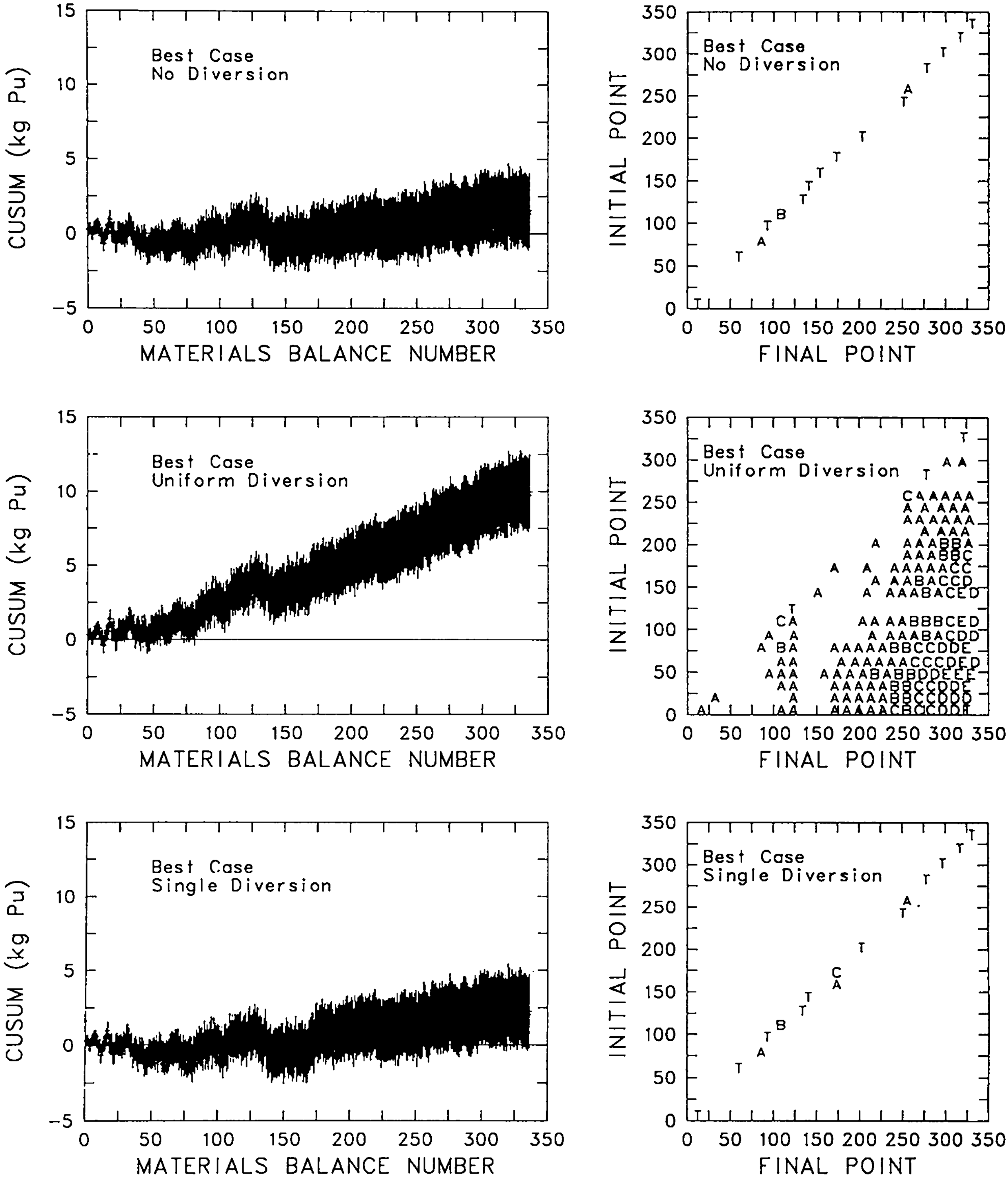


Fig. 27.
Strategy 4, UPAA 23(best case), 1 month: cusum and alarm charts.

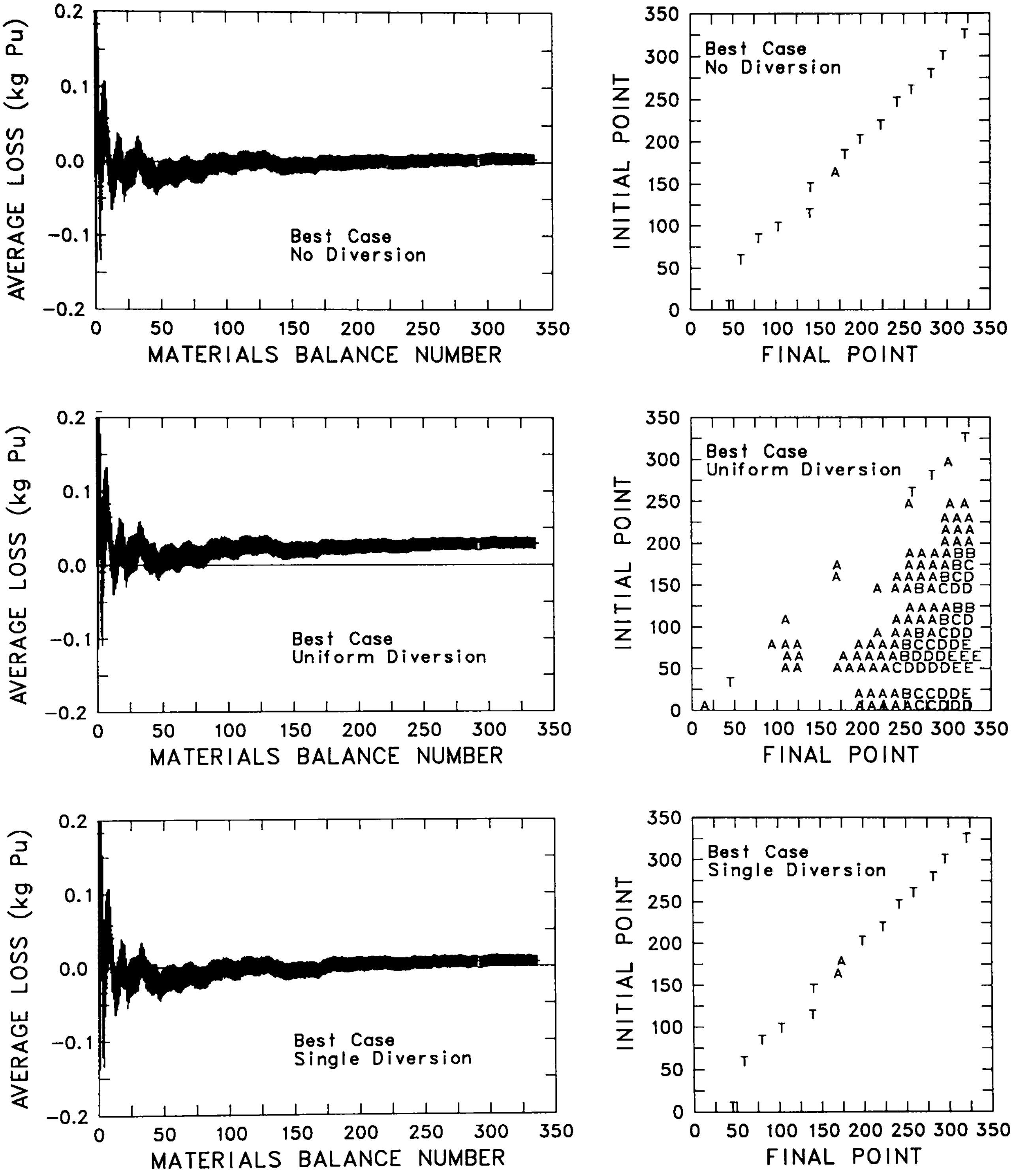


Fig. 28.
Strategy 4, UPAA 23(best case), 1 month: UDT and alarm charts.

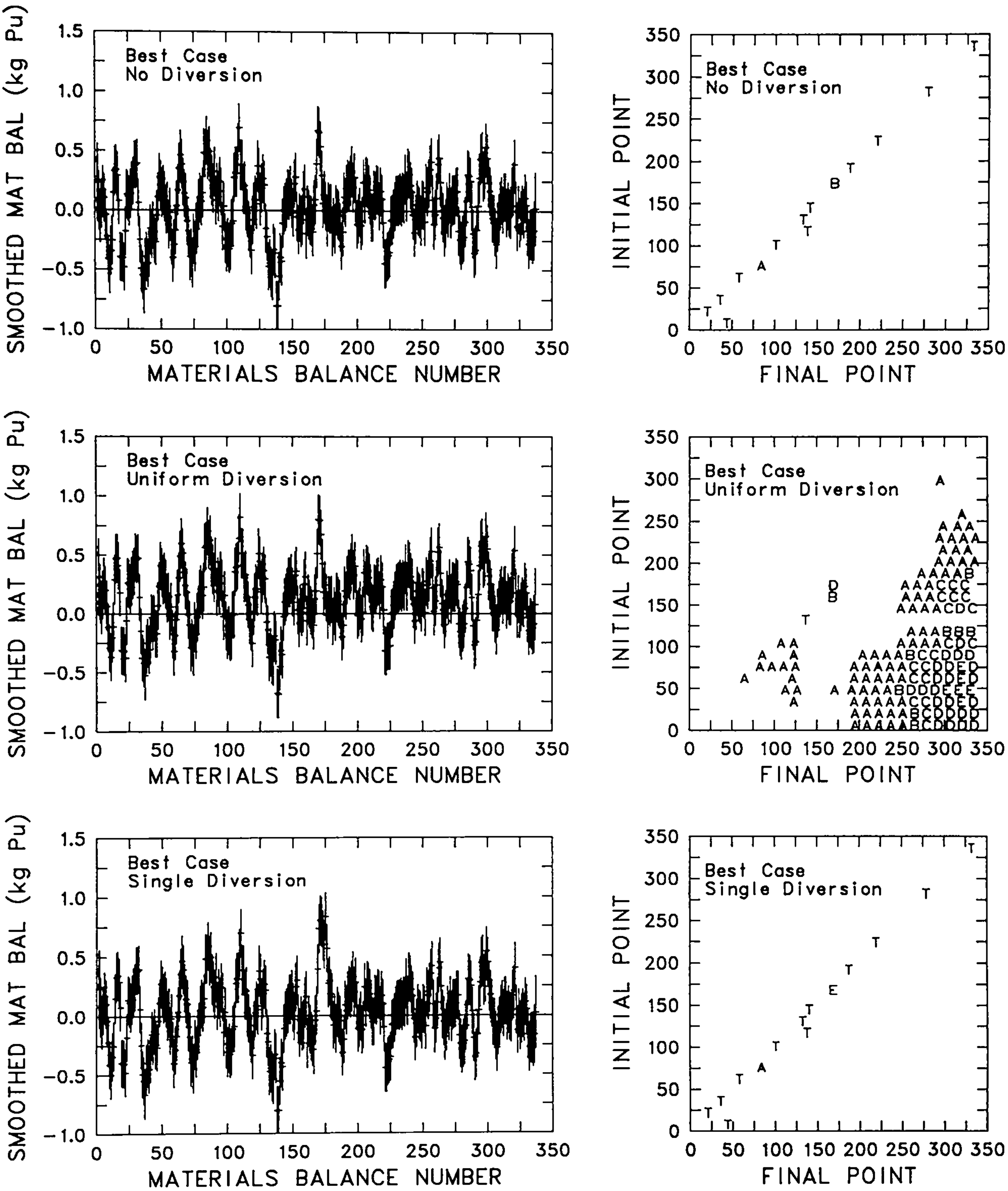


Fig. 29.
Strategy 4, UPAA 23(best case), 1 month: SMBT and alarm charts.

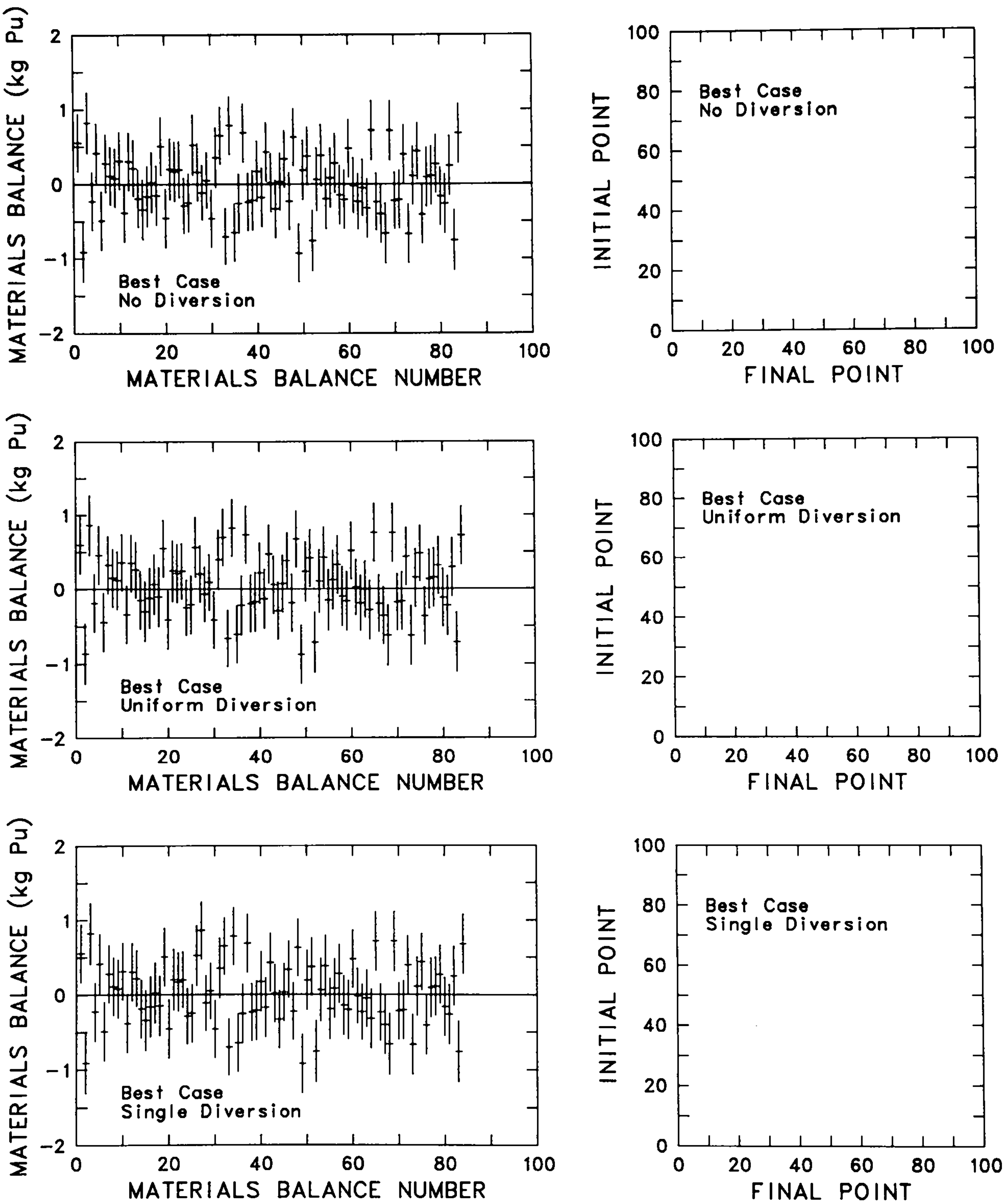


Fig. 30.
Strategy 4, UPAA 23(best case), 1 week:
materials balance and alarm charts.

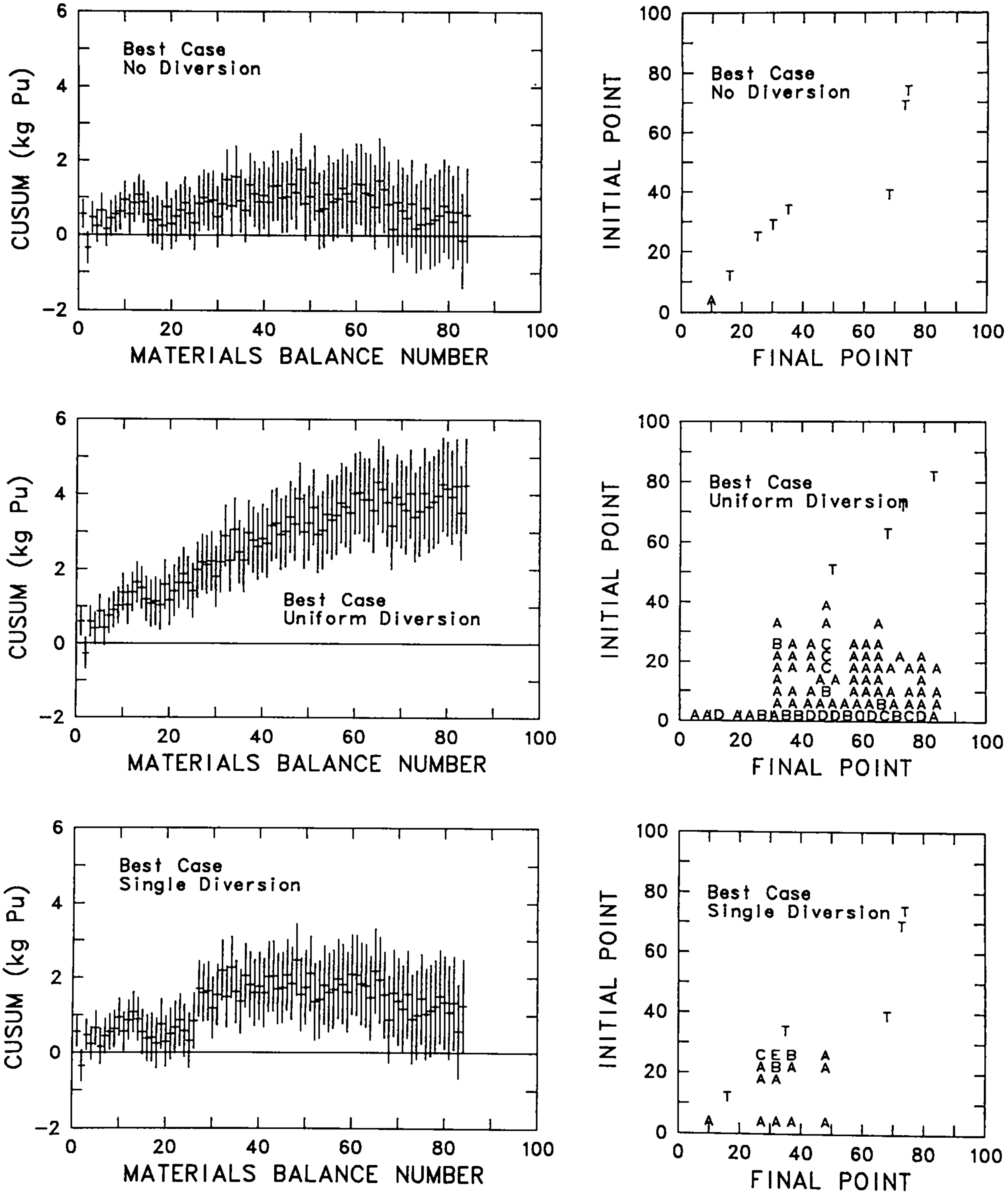


Fig. 31.
Strategy 4, UPAA 23(best case), 1 week: cusum and alarm charts.

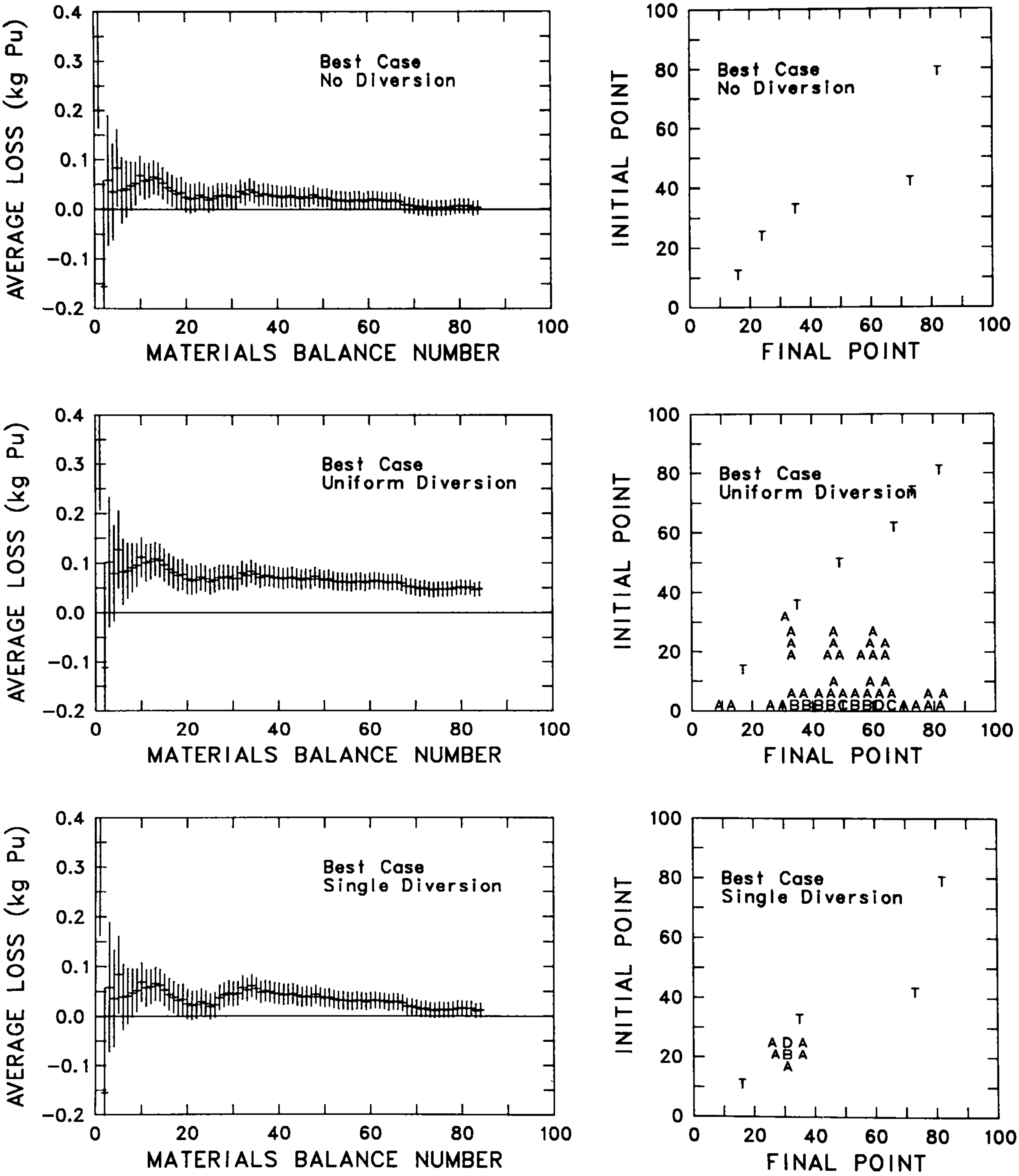


Fig. 32.
Strategy 4, UPAA 23(best case), 1 week: UDT and alarm charts.

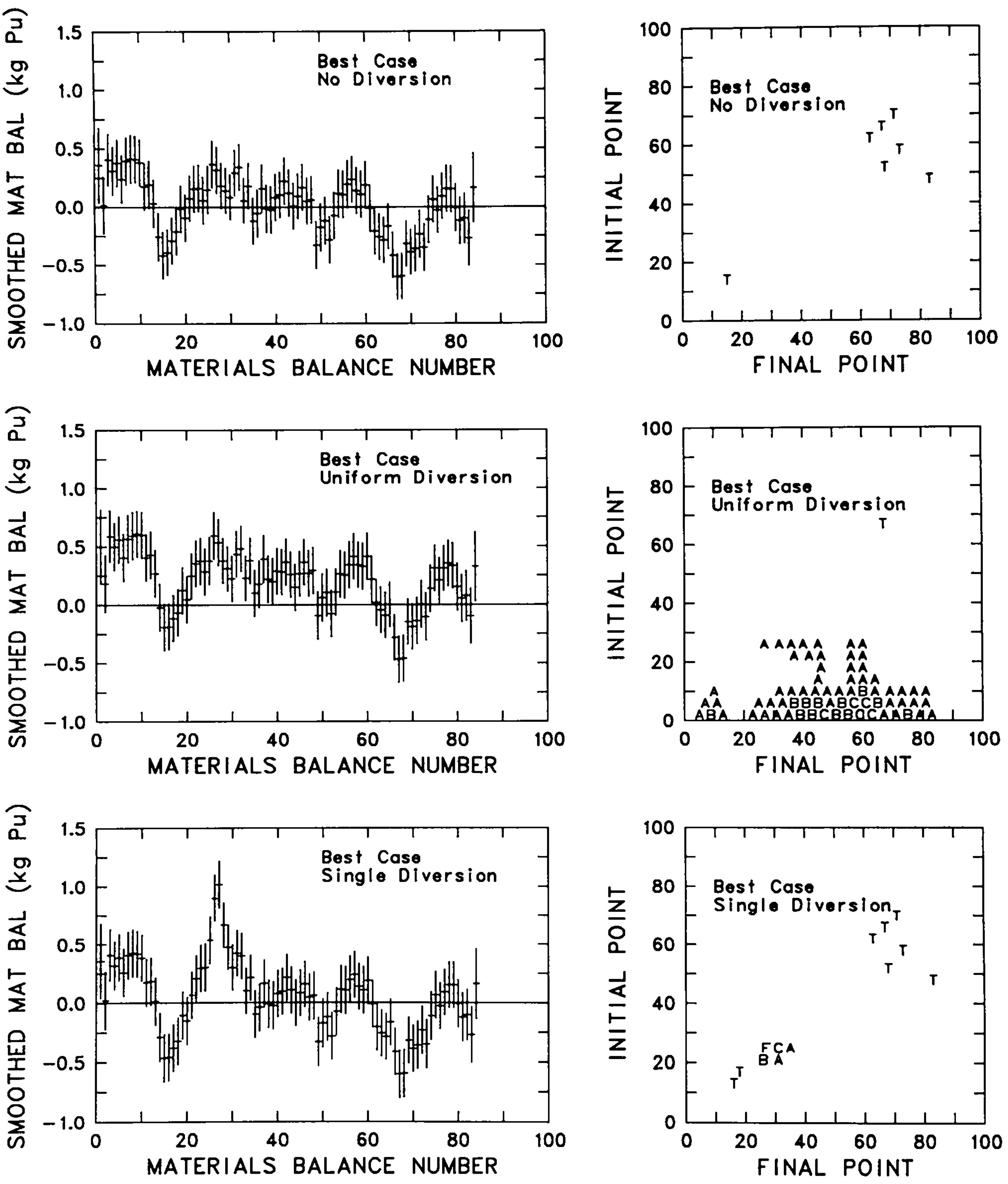


Fig. 33.
Strategy 4, UPAA 23(best case), 1 week: SMBT and alarm charts.

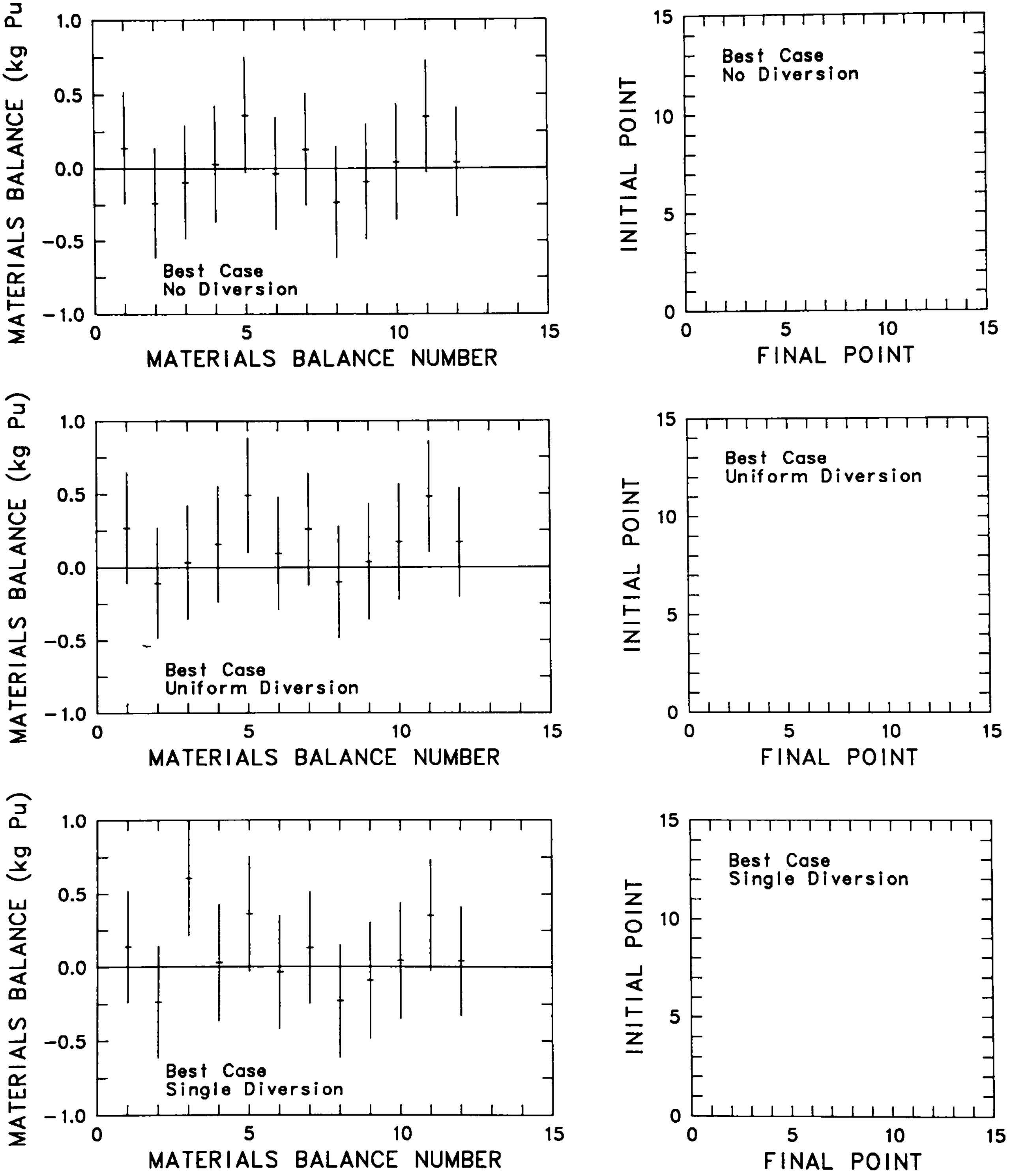


Fig. 34.
Strategy 4, UPAA 23(best case), 1 day:
materials balance and alarm charts.

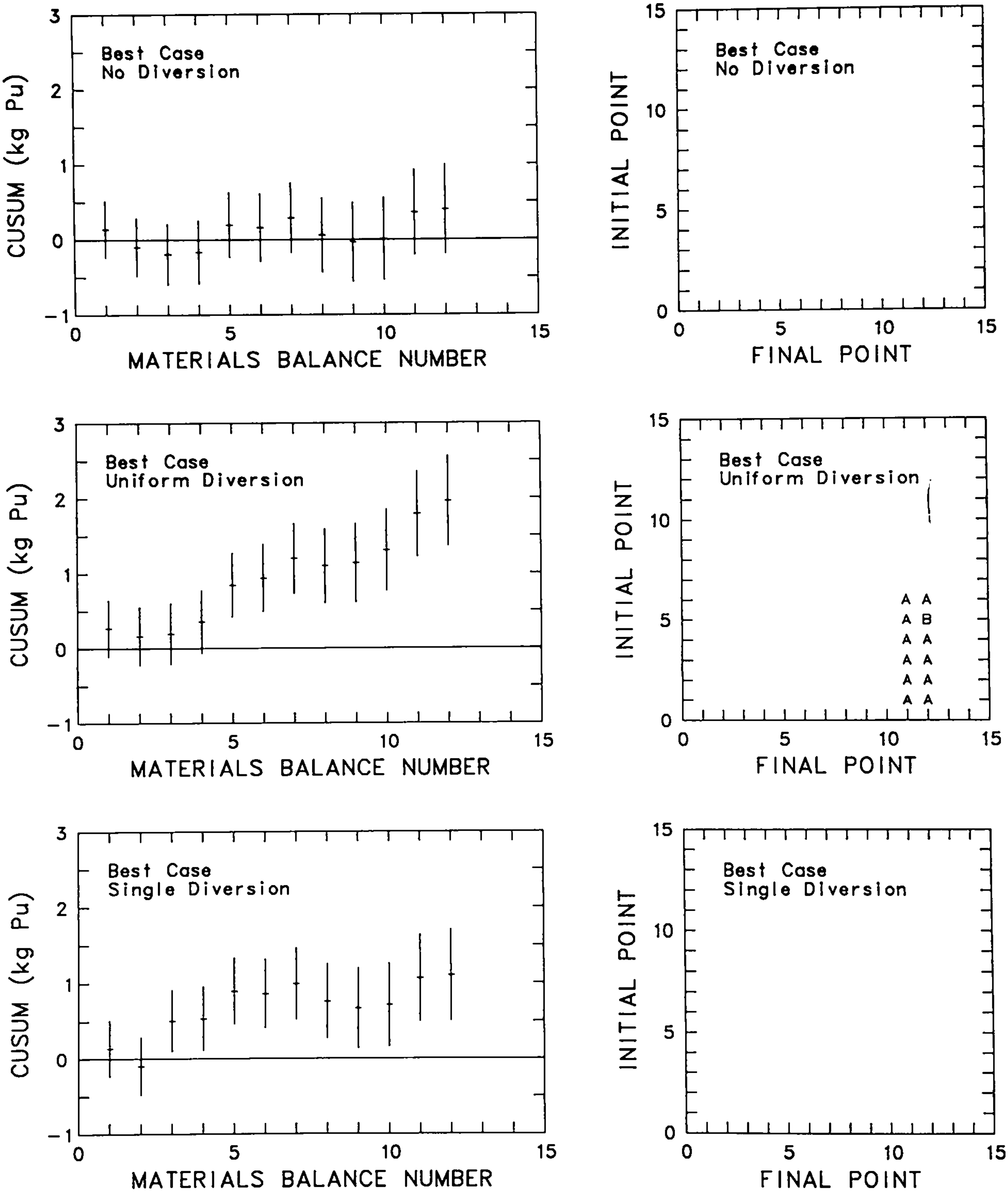


Fig. 35.
Strategy 4, UPAA 23(best case), 1 day: cusum and alarm charts.

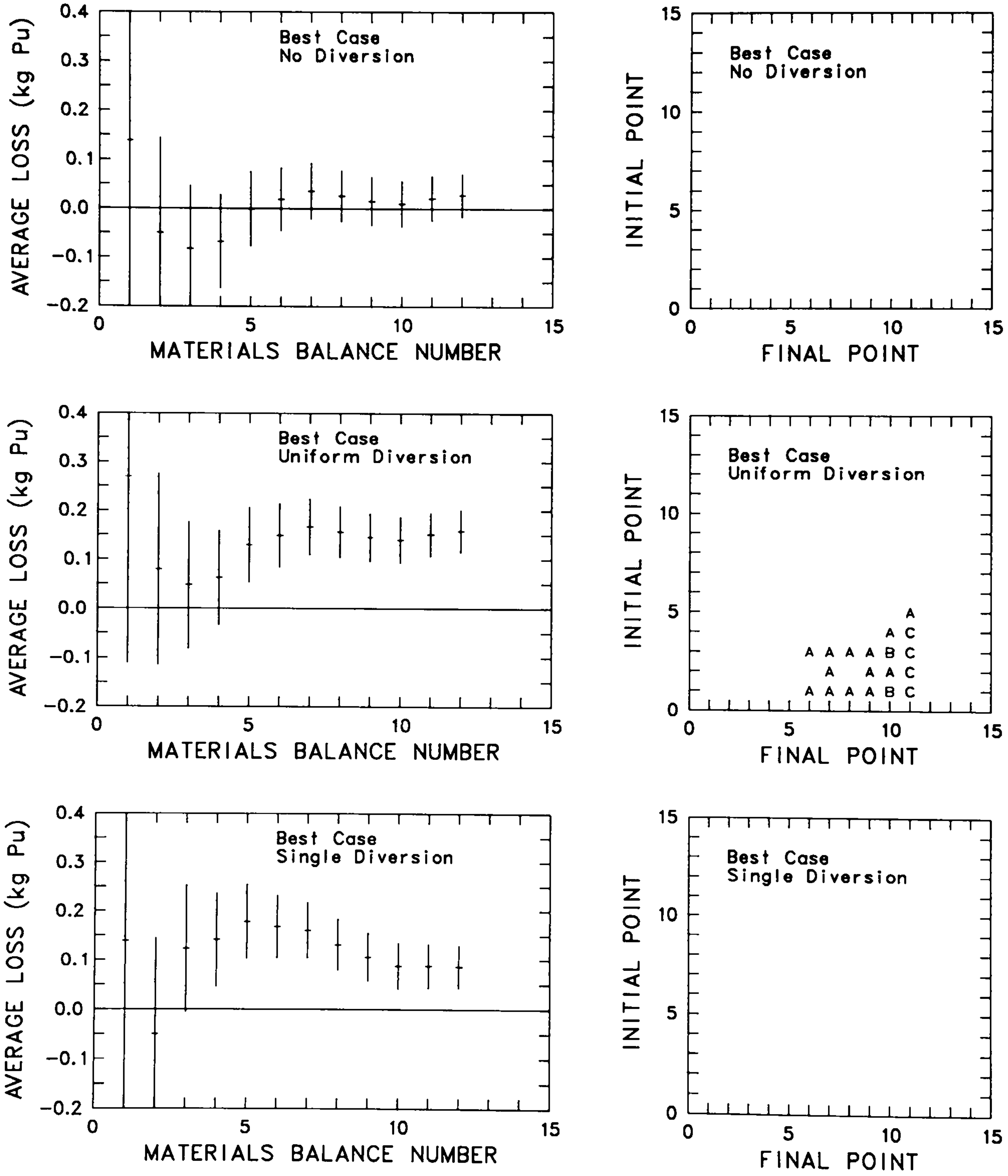


Fig. 36.
Strategy 4, UPAA 23(best case), 1 day: UDT and alarm charts.

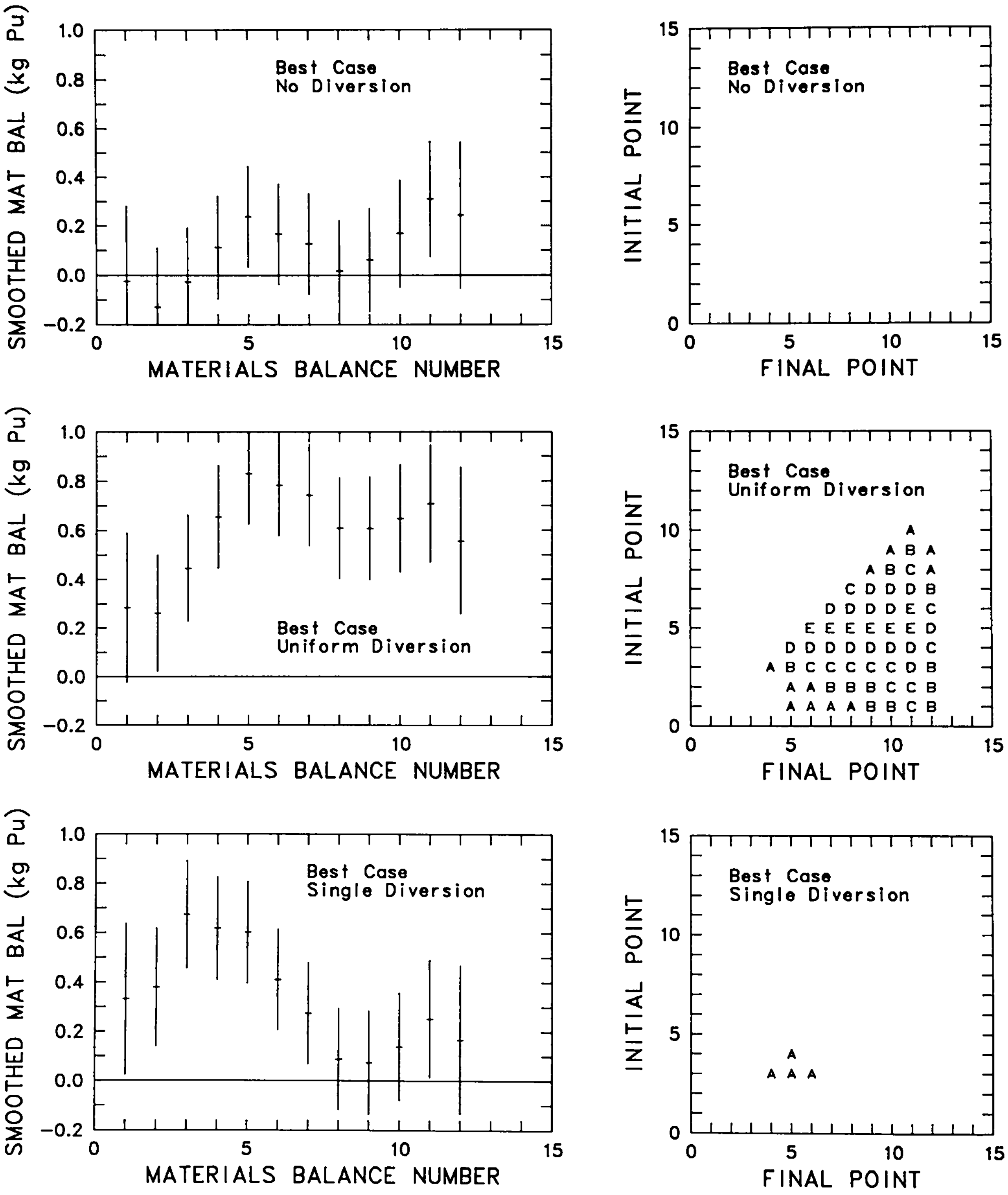


Fig. 37.
Strategy 4, UPAA 23(best case), 1 day: SMBT and alarm charts.

VI. RESULTS AND CONCLUSIONS

A. Results

1. Reference Coprecal Coconversion Process. Advanced concepts for materials management in a Coprecal coconversion facility have been investigated in this first detailed study of advanced materials-management systems for alternative fuel cycle facilities. Previous studies have considered advanced safeguards systems for the reference separated-stream uranium-plutonium fuel cycle[1-3] and have described some preliminary concepts and measurement requirements for the thorium-uranium fuel cycle.[42,43] The present study derives from the Alternative Fuel Cycles Technologies Program (AFCT) and, in particular, from the development by G.E. for Savannah River Laboratory (SRL) of a coconversion process design based on the G.E. Coprecal process.[5]

The reference Coprecal process design, described briefly in Sec. I.C and in detail in App. A, consists of three parallel process lines having a total design throughput of 117 kg of plutonium per day. For LWR fuel feed material, the uranium-to-plutonium ratio in the coprocessing streams is 9/1.

The structure of an advanced safeguards system generally applicable to the domestic safeguarding of any fuel cycle facility is described in Sec. II. The safeguards system comprises several subsystems such as the safeguards coordination unit (including the safeguards management, data collection, and data analysis functions), and subsystems for materials measurement and accounting, physical protection, process monitoring, and the safeguards computer. The functions of these subsystems and their interfaces with each other and the related facility functions of process control and plant management are described, and important related considerations such as system security and reliability are discussed. Possible relationships between a domestic safeguards system and a counterpart international safeguards system are explored generally. International safeguards and nonproliferation features for national fuel cycles will be treated separately in subsequent reports.

The conceptual design of an MMAS for the reference Coprecal process is described in Sec. III. The proposed MMAS is based on near-real-time (dynamic) accountability concepts in which the process is partitioned into discrete accounting areas called unit process accounting areas (UPAAs). Timely and sensitive materials accounting is achieved by forming dynamic materials balances in each UPAA from a combination of state-of-the-art conventional and NDA measurements. As in the previous studies, the proposed measurements and estimates of their precisions and accuracies are based on experience with currently available instrumentation or on modest extrapolations of current measurement technology (App. D). Conventional process cleanout and physical inventory is performed periodically to establish fiducials or reference points for the near-real-time accounting system.

Key measurement points for process streams are the feed-blend tanks, the reduction-stabilization process containers, and the product canisters. The plutonium contents of the feed-blend tanks are obtained from state-of-the-art volume or mass measurements and conventional sampling and chemical assay techniques. An at-line technique such as absorption-edge densitometry is proposed to verify the plutonium concentration in the process feed stream during the 16.5-day campaigns. The reduction-stabilization containers are measured by neutron coincidence counting, and the product canisters can be measured by coincidence counting or possibly by calorimetry.

Because of the high plutonium throughput of this facility and the large size of the process vessels necessary to accommodate the diluted coprocessed material, key measurement points for in-process inventory must include all principal process equipment. Measurement systems based on neutron NDA techniques probably can be designed and built to measure the in-process inventory of equipment items, such as the large filters, to 10% or better. The proposed MMAS requires many such measurements, and these systems represent a significant safeguards cost (Sec. III). Compared with separated plutonium conversion processes (excluding recycle processes), coconversion processes invariably will require considerably more process equipment with greater operating capacities because of the order-of-magnitude dilution of the fissile material.

The key dynamic accountability measurements require supplementary measurements of isotopic composition by gamma-ray or mass spectrometry. Secondary accountability measurements of plutonium holdup and process waste streams are also required.

Four strategies for dynamic accountability are considered. The basic strategy is obtained by dividing each of the three parallel process lines into three UPAAs: (1) feed receiving and blending, (2) precipitation and calcination, and (3) reduction, stabilization, and screening. The four dynamic accountability strategies comprise all possible combinations of the three UPAAs and each strategy incorporates the same set of key measurement points. In practice, therefore, each strategy can be implemented within the same set of accounting measurements by using appropriate analysis software. The safeguards advantages of this analysis procedure are obvious and should be exploited.

The diversion-detection capability of the proposed MMAS is evaluated using modeling, simulation, and advanced data analysis techniques (Sec. IV). COPSIM, a computerized model of the Coprecal process (App. B), was developed. Simulated accountability measurements were applied to process flows and in-process inventories using the computer code COPMEAS (Sec. IV.C and App. C). The effectiveness of each dynamic accountability strategy was evaluated by applying advanced data-analysis algorithms using the computer code DECANAL. DECANAL comprises decision-analysis and graphic-display techniques developed to analyze the many data generated by near-real-time accounting systems.

Sample results of diversion-detection simulation studies are given in Tables XIII through XVI (Sec. V). Detection sensitivities are shown for abrupt and protracted diversion strategies. The empirical false alarm probability determined from many simulation runs is <5%.

In Table XIII, detection sensitivity ranges are given for UPAA 1 corresponding to the variation in the uncertainty of volume measurements made on the large feed-blend tanks; that is, the volume measurement error is assumed to be proportional to the tank inventory. Hence, the absolute error in these measurements is much larger at the beginning of a campaign when the tank is nearly full than at the end of a campaign when the tank is nearly empty.

In Table XVI detection sensitivities are given for UPAA 23 (UPAA 2 and UPAA 3 combined). Results are shown for the two cases called base and best, which differ in the assumed frequency of recalibration of the flowmeter in the process feed stream: once per campaign for the base case and once per day for the best case. Daily recalibration provides significant improvement in the sensitivity to protracted diversion.

As in previous studies, estimates of safeguards system reliability and costs and of the required safeguards staffing have been made. In reliability terms an MTBF of ∿3 months and an MTTR of ∿5 h are judged to be reasonable and desirable design goals for the safeguards system. The computer code RELSIM was developed to determine whether these goals are met by specific choices of system architecture and components.

The capital cost of the safeguards computer and information systems and of the MMAS hardware is estimated to be ∿\$9 million, of which ∿\$3 million is for the neutron detection systems and ∿\$2.5 million is for computer hardware and software development. Annual operating cost for the computer, information, and MMAS systems is estimated to be ∿\$2.6 million. Staffing estimates include 6 supervisory and 36 nonsupervisory personnel.

No credit is taken in these estimates for benefits that would accrue from applying data from the safeguards system to other necessary facility functions. Moreover, safeguards costs and staffing chargeable directly to the coconversion facility would be significantly lower if the facility were collocated with chemical separations and/or fuels refabrication facilities.

2. Comparison with the Reference Oxalate (III) Conversion Process. In this section the results of the Coprecal process safeguards study reported here are compared with the results of the Oxalate (III) conversion process safeguards study reported in Ref. 2.

a. The Processes. Both processes have the same design basis throughput (∿117 kg plutonium/day) and achieve this throughput in parallel operating lines. However, the process-line structure differs in the two facilities.

The Coprecal facility consists of three parallel, completely independent process lines. Each process line consists of three parallel feed-blend tanks, one precipitator, four parallel calciners, one calciner primary filter, one calciner secondary filter, four parallel reduction-stabilization stations each having a primary and a final filter, and one screening station.

The Oxalate (III) facility has four parallel precipitator lines, three of which are operated simultaneously while the fourth is being flushed. The three active precipitator lines are fed alternately from three parallel feed tanks. Each precipitator filter boat passes through one of three parallel tunnel furnaces. The calcined product is unloaded into product canisters and assayed in one of two parallel dump-and-assay stations. Thus, although the precipitator lines are unique, the feed tanks, furnaces, and dump-assay stations are used in common. It was recommended in Ref. 2 that process-line integrity be maintained throughout by adding a furnace and two dump-and-assay stations to enhance materials control and accounting capabilities.

The Coprecal process is nearly continuously operated, whereas the Oxalate (III) process is batch operated. Differences in the methods of feeding the process lines in the two facilities cause major differences in the diversion-detection sensitivities that can be achieved by the two materials accountability systems. Feed tanks for the two processes are compared in Table XVII. In principle, the use of independent feed tanks for each Coprecal process line should enhance materials accountability. However, the large tank size and the fact that each tank feeds continuously during a campaign have a severe adverse impact on detection sensitivity. The large tank volume and the nonstatic state of the contents are expected to result in relatively large measurement errors in the absolute plutonium quantities. Continuous feeding of the process necessitates flow measurements in the process feed stream, and correlations between these measurements increase materials balance uncertainties. The use of common feed tanks in the

TABLE XVII

COPRECAL AND OXALATE (III)
FEED-TANK CHARACTERISTICS

	Coprecal	Oxalate (III)
Numbers of tanks	3 per line	3 for the entire facility
Tank capacity (L)	18 000	260
Normal working volume (L)	16 000	200
Pu concentration (g/L)	40	30
Pu tank content (kg)		
Full	640	6
Heel	6	0
Feeding time (h)	396	1.3
Dynamic accountability measurements		
Volume		
Precision (% 1 σ)	1.	0.2
Calibration (% 1 σ)	1.	0.1
Concentration		
Precision (% 1 σ)	0.2	1.
Calibration (% 1 σ)	0.2	0.3
Combined measurement uncertainty (kg Pu 1 σ)		
Full	9.1	0.06
Heel	0.09	0

conversion facility does not adversely affect the detection sensitivity because the feed is transferred in relatively small batches to each process line.

Because the Oxalate (III) tanks are smaller and the contents are static while measurements are being made, much better measurements are possible. Also, because relatively small batches are dispensed, a precipitator-feed flow measurement is not required, thereby reducing transfer measurement correlations.

The precipitators in the two processes are also operated differently. In the Coprecal process, a relatively short precipitator residence time is sufficient to produce a finely divided slurry. In the Oxalate (III) process, the precipitator has a relatively large working volume (Table XVIII) to provide a residence time sufficiently long to promote crystal growth. In the Coprecal process, the integrity of relatively small batches is maintained only in the reduction, stabilization, and screening operations. In the Oxalate (III) process, the precipitator is batch fed from two parallel valence-adjust tanks, and the product is batched from the two filtering stations. The integrity of relatively small batches is maintained except during precipitation. Processing of relatively small batches generally facilitates materials accounting.

In the Coprecal process, four parallel calciners in each process line discharge continuously into a large filter. In the Oxalate (III) process, batches are transferred through a single tunnel furnace for each process line (Table XIX).

In all coconversion processes, reduction and stabilization are required after calcination to reduce the UO_3 to UO_2. These extra steps significantly increase the residence time of material within the process and the in-process plutonium inventory.

As shown in Table XX, the Coprecal process produces fewer and larger batches per day than the Oxalate (III) process. Instrument precision and accuracy are the same in both processes.

The Coprecal process is generally more complicated and has more process equipment than the Oxalate (III) process (excluding any recycle). Special instrumentation problems are also encountered in Coprecal because of the geometry and size of some process vessels, such as the large calciner primary filters (annulus 7.6 cm thick, 3.5-m o.d., 1.2 m tall), and the high operating temperatures of the calciners and filters.

The number of accountability instruments required by Coprecal is also large when compared to the Oxalate (III) process. It is estimated that 63 neutron detectors are required for the Coprecal process, most of which must operate in a high-temperature environment, whereas 19 neutron detectors operating at room temperature are required for the conversion process.

b. Dynamic Materials Accounting. Two dynamic materials accounting strategies were investigated for the Oxalate (III) process. As in the present Coprecal study, dynamic accountability was applied independently to each process line. In the Oxalate (III) study, Strategy 1 treats a process line as two UPAAs. The first UPAA includes the receipt tanks and a precipitator; the second UPAA includes a furnace and a product dump-and-assay station. Strategy 2 treats the entire process line as a single UPAA.

TABLE XVIII

COPRECAL AND OXALATE (III) PRECIPITATORS

	Coprecal	Oxalate (III)
Number per line	1	1
Working volume (L)	40	90
Nominal Pu inventory (kg)	1	2
Dynamic accountability measurements		
Inventory precision (% 1 σ)	10	2
kg Pu (1 σ)	0.1	0.04

TABLE XIX

COPRECAL AND OXALATE (III) CALCINERS

	Coprecal	Oxalate (III)
Number per line	4	1
Nominal Pu inventory (kg)	5	10
Operational mode	continuous	batch
Dynamic accountability measurements		
Inventory precision (% 1 σ)	10	5
kg Pu (1 σ)	0.5	0.5

TABLE XX

COPRECAL AND OXALATE (III) PRODUCT BATCHES

	Coprecal	Oxalate (III)
Batch frequency per line	1 every 2 h	1 every 1.3 hr
Pu content (kg)	3.2	2.
Total mass (kg)	38.	2.
Dynamic accountability measurements		
Precision (% 1 σ)	1.	1.
Calibration (% 1 σ)	0.5	0.5
Combined measurement uncertainty (kg Pu 1 σ)	0.036	0.022

Both Coprecal Strategy 2 and Oxalate (III) Strategy 1 treat each process line as two UPAAs. However, Coprecal UPAA 12 includes calcination, whereas Oxalate (III) UPAA 1 does not.

Coprecal Strategy 3 and Oxalate (III) Strategy 2 are exactly the same because each entire process line is treated as a single UPAA. These strategies are compared in Table XXI. Coprecal exhibits the poorer sensitivity primarily because the inventory of its feed-blend tank, even as the tank nears its heel, is larger than the inventory of the Oxalate (III) receipt tank, resulting in a much larger materials balance uncertainty. Since the feed-blend tank measurements dominate the uncertainties for the Coprecal strategy in this comparison, further comparison of detection sensitivities is fruitless.

If we assume that the feed-blend tanks are dealt with in some satisfactory manner, then it is useful to compare Coprecal Strategy 4, UPAA 23 (line not including feed-blend tanks) with Oxalate (III) Strategy 2 (whole line). (See Table XXI.) The detection sensitivity in the Coprecal facility is improved in this comparison because separating the feed-blend tanks greatly reduces the in-process inventory, and daily recalibration of the flowmeter reduces the correlations between feed flow measurements. However, the estimated materials-balance uncertainties for the Coprecal process are still somewhat larger because the Coprecal in-process inventory is still about twice that of the

TABLE XXI

COPRECAL UPAA 123 AND UPAA 23 (BEST CASE) AND OXALATE (III) STRATEGY 2

	Standard Deviation (kg Pu)		
	Coprecal		Oxalate (III)
	UPAA 123[a]	UPAA 23 Best Case	Strategy 2
Net transfer	0.04	0.05	0.03
In-process inventory	9.1-0.29	0.3	0.09
Materials balance			
1 balance	8.9-0.41	0.4	0.13
1 week	7.9-4.2	1.3	0.7

---

[a] Range during a campaign.

conversion process and the Coprecal feed flow measurement errors are larger than the conversion process receipt-tank batch transfer measurement errors.

This comparison is extended in Table XXII where the estimated detection sensitivities for Coprecal Strategy 4, UPAA 23, best case, and Oxalate (III) Strategy 2 are given. The better detection sensitivity attainable in the conversion process is a result of (1) its smaller in-process inventory and (2) the improved precipitator-feed measurements possible because of batch feeding from a small tank. Detection sensitivities approaching those estimated for the conversion process should be obtained for the Coprecal process if aliquot tanks are inserted after the feed-blend tanks in the precipitator-feed streams in place of flowmeters.

B. Conclusions

We have identified three essential criteria for effective dynamic materials accountability in the reference Coprecal process.

- The relatively large absolute errors in the feed-blend tank inventory measurements preclude effective materials accountability in the Coprecal process unless the feed-blend tanks can be separated, for accounting purposes, from the rest of the process. This is the reason for placing flowmeters in the process feed streams after the feed-blend tanks.

TABLE XXII

DETECTION SENSITIVITIES
COPRECAL STRATEGY 4, UPAA 23 (BEST CASE)
AND OXALATE (III) STRATEGY 2

	Coprecal		Oxalate (III)[a]	
Detection Time	Average per Balance (2 h) (kg Pu)	Total at Detection (kg Pu)	Average per Balance (1.3 h) (kg Pu)	Total at Detection (kg Pu)
1 balance	1.2	1.2	0.4	0.4
1 day	0.13	1.6	0.02	0.5
1 week	0.04	3.7	0.01	1.7
1 month	0.03	8.4	0.007	3.9

---

[a] The detection sensitivities shown here are not identical to those in Ref. 2 because analysis techniques have been improved.

- Provision must be made for frequent recalibration of accountability instruments at key materials flow measurement points. Otherwise, correlations between flow measurements can degrade the detection sensitivities to unacceptably low levels. Computer-controlled dynamic calibration techniques should be investigated for on-line instrumentation.
- Provision must be made for measuring the plutonium in-process inventory in process equipment. In-process inventory measurements on the order of 10% or better should be satisfactory.

These criteria are especially relevant at the head end of the reference Coprecal process where the problem of satisfying them is exacerbated by the continuous flow of feed material from the large feed-blend tanks. This flow makes frequent measurement and recalibration difficult. This has led us to consider alternative arrangements of the head-end process equipment.

One possible alternative arrangement is to place two small aliquot tanks after the feed-blend tanks—an arrangement similar to the receipt tanks at the head end of the reference Oxalate (III) process. One aliquot tank would feed the process while the other was being filled from a certified feed-blend tank. Transfers of solution from the feed-blend tanks to the aliquot tanks would be verified by static tank measurements, volume or mass, with provision made for frequent recalibration of the tank measurements using techniques such as check weights for mass measurements or the "piston-prover" method[44] for volume measurements. An NDA technique such as absorption-edge densitometry could be used to verify the plutonium concentration in the aliquot tank solutions. Use of the aliquot tanks would preclude the necessity for flowmeters in the process feed streams.

Additional recommendations are listed below.

- If the feed-blend tanks are separated for accounting purposes, the materials balance uncertainty for the rest of the process line is dominated by measurement errors associated with determining the large in-process inventory. Careful consideration should be given to reducing this inventory.
- Additional work is required to develop and evaluate instrumentation that has been recommended for safeguards measurements. Absorption-edge densitometry is an effective method for measuring uranium and plutonium. However, its applicability to plutonium measurements in the presence of a 10-fold excess of uranium with the required precision and accuracy has not yet been demonstrated.

- Large neutron-detection systems for measuring the plutonium inventory in process vessels and in large, annular filters must be built and demonstrated.
- The relative sensitivity and reliability of volume and mass measurement systems for the feed-blend tanks should be evaluated.

The diversion-detection sensitivity of the dynamic materials accountability system developed for the reference Coprecal facility is demonstrably less than that attainable in the previously studied conversion facility (Ref. 2). This is not necessarily a consequence of the Coprecal process itself. It may be--at least in part--the result of the particular process design and equipment selected for the reference facility and the operating philosophy chosen. Greater quantities of material must be handled in a coprocessing facility to achieve a given throughput of fissile material, and this requirement can be expected to have a deleterious effect on accountability. It may be possible to design the process to reduce this effect. If, however, the problems cited in Sec. VI.C are inherent in the Coprecal process or in coconversion processes in general, the value of these processes as a safeguards and nonproliferation aid should be reexamined.

Much can be gained if safeguards system designers and process design engineers jointly examine alternative arrangements of the Coprecal process that could facilitate materials accountability without unduly disrupting process operation. Such a cooperative effort is recommended highly.

The reference Coprecal process[5] studied is based on a preliminary design. The entire Coprecal process design will be modified before it is finalized, and we expect these modifications to address the major recommendations in this report. We will evaluate the impact of these modifications on materials accountability in the Coprecal process as soon as these modifications are sufficiently well defined.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the helpful suggestions and criticism of D. F. Bowersox, LASL (CMB-11); E. F. Kurtz, R. J. Sloat, and I. N. Taylor of the General Electric Company; S. Baloga, DOE/OSS; and L. A. Heinrich, R. S. Swingle, and E. Lukosius, SRL. We wish to express our gratitude to S. L. Klein, M. S. Scott, D. P. Martinez, N. S. Han, and K. C. Eccleston, LASL (Q-4), for the preparation of the manuscript. Much of the information on NDA instrumentation was obtained from H. O. Menlove, LASL (Q-5), and P. Russo, J. Parker, M. Krick, and N. Ensslin, LASL (Q-1).

REFERENCES

1. 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 Labortory report LA-6881 (September 1977).

2. H. A. Dayem, D. D. Cobb, R. J. Dietz, E. A. Hakkila, E. A. Kern, J. P. Shipley, D. B. Smith, and D. F. Bowersox, "Coordinated Safeguards for Materials Management in a Nitrate-to-Oxide Conversion Facility," Los Alamos Scientific Laboratory report LA-7011 (April 1978).

3. J. P. Shipley, D. D. Cobb, R. J. Dietz, M. L. Evans, E. P. Schelonka, D. B. Smith, and R. B. Walton, "Coordinated Safeguards for Materials Management in a Mixed-Oxide Fuel Facility," Los Alamos Scientific Laboratory report LA-6536 (February 1977).

4. Code of Federal Regulations (Energy, Vol. 10), Office of the Federal Register, National Archives and Records Service, General Services Administration, Washington, DC, January 1, 1977, pp. 385-386.

5. Savannah River Laboratory document DPSTD-AFCT-77-10-TL (November 1977).

6. J. M. de Montmollin and R. B. Walton, "The Design of Integrated Safeguards Systems for Nuclear Facilities," Nucl. Mater. Manage. V(III), 317-332 (1976).

7. L. D. Chapman, J. M. de Montmollin, J. E. Deveney, W. C. Fienning, J. W. Hickman, L. D. Watkins, and A. E. Winblad, "Development of an Engineered Safeguards System Concept for a Mixed-Oxide Fuel Fabrication Facility," Sandia Laboratories report SAND76-0180 (August 1976).

8. W. C. Fienning, A. E. Winblad, and J. P. Shipley, "A Preliminary Concept Definition of a Combined Safeguards System for a Mixed-Oxide Fuel Fabrication Facility," Sandia Laboratories report SAND77-0224 (October 1977).

9. W. D. Chadwick, W. C. Fienning, B. G. Self, and G. E. Rochau, "A Concept Definition of an Engineered Safeguards System for a Spent Fuel Reprocessing Facility," Sandia Laboratories report, to be published.

10. J. P. Shipley, "The Structure of Safeguards Systems," Los Alamos Scientific Laboratory report LA-7337-MS (June 1978).

11. R. H. Augustson, "Development of In-Plant Real Time Materials Control: The DYMAC Program," Nucl. Mater. Manage. V(III), 302-316 (1976).

12. G. R. Keepin and W. J. Maraman, "Nondestructive Assay Technology and In-Plant Dynamic Materials Control--DYMAC," in Safeguarding Nuclear Materials, Proc. Symp., Vienna, 1975 (International Atomic Energy Agency, Vienna, 1976) paper IAEA-SM-201/32, pp. 305-320.

13. G. L. Booman and F. L. Cartan, Idaho Chemical Processing Plant, personal communication, March 1977.

14. E. P. Schelonka, "Availability Analysis for High-Reliability Computer Systems in Nuclear Facilities," Proc. 18th Ann. Mtg. Instit. Nucl. Mater. Mgmt., Washington, DC, June 29-July 1, 1977.

15. J. H. Fuchs, D. M. Kisner, R. K. McClean, H. R. Milder, K. Mizuba, and G. E. Short, "Unified Safeguards Information System Concept," TRW Defense and Space Systems Group report 32076-6001-RU-00 (May 20, 1977).

16. D. M. Kisner, H. R. Milder, and K. Mizuba, "Preliminary Hardware Configuration Recommendations for Safeguards Information System," TRW Defense and Space Systems Group report 31134-6001-RU-00 (May 24, 1977).

17. R. H. Augustson, "Dynamic Materials Control Development and Demonstration Program," Proc. 19th Annual Meeting, Cincinnati, J. INMM VII III (1978).

18. R. H. Augustson, "DYMAC Demonstration Program: Phase I Experience," Los Alamos Scientific Laboratory report LA-7126-MS (February 1978), Appendix A.

19. R. H. Augustson, N. Baron, R. F. Ford, W. Ford, J. Hagen, T. K. Li, R. S. Marshall, V. S. Reams, W. R. Severe, and D. G. Shirk, "A Development and Demonstration Program for Dynamic Nuclear Materials Control," paper presented at the IAEA Symp. Nucl. Mater. Safeguards, Vienna, Austria, October 2-6, 1978.

20. L. J. Hoffman, Modern Methods for Computer Security and Privacy, (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1977), p. 3.

21. Computer Security Bibliography, internal document, Los Alamos Scientific Laboratory, Group Q-4, (June, 1978).

22. J. Campbell, Sandia Laboratories, personal communication to E. P. Schelonka, (August 1978).

23. A. M. Fine, R. S. Reynolds, R. E. D. Stewart, and P. A. Stokes, "Feasibility Study of an Advanced Unattended Seismic Observatory", Sandia Laboratories report SLA-73-0008 (March 1973).

24. L. G. Roberts and B. D. Wessler, "Computer Network Development to Achieve Resource Sharing", AFIPS Conference Proceedings, Spring Joint Computer Conference (1970).

25. G. E. Reynolds, "Multics Security Evaluation: Exemplary Performance Under Demanding Workload", ESD-TR-74-193, Vol. IV, Electronics Systems Division, Hanscom Air Force Base, Massachusetts, (November, 1976).

26. M. Kert, "Advances in Minicomputer and Front-End Security", Proc. Computer Security and Privacy Symposium, Scottsdale, Arizona, (April 19-20, 1977).

27. E. P. Schelonka, "Event and Effectiveness Models for Simulating Computer Security", presented at the IEEE Computer Conference COMPCON FALL, Washington, DC, September 5-8, 1978 (to be published in the Conference Proceedings).

28. International Atomic Energy Agency, "The Structure and Contents of Agreements between the Agency and States Required in Connection with the Treaty on the Non-Proliferation of Nuclear Weapons," INFCIRC/153, IAEA, Vienna (1971).

29. J. L. Jaech, "Statistical Methods in Nuclear Material Control," TID-26298, Technical Information Center, Oak Ridge, Tennessee (1973).

30. J. P. Shipley, "Decision Analysis and Nuclear Safeguards," in *Nuclear Safeguards Analysis - Nondestructive and Analytical Chemical Techniques*, E. A. Hakkila, Editor, Am. Chem. Soc., Washington, DC (1978).

31. J. P. Shipley, "Decision Analysis for Dynamic Accounting of Nuclear Material," paper presented at the American Nuclear Society Topical Meeting, Williamsburg, Virginia, May 15-17, 1978.

32. J. P. Shipley, "Efficient Analysis of Materials Accounting Data," Nucl. Mater. Manag., VII, No. 3 (1978).

33. R. E. Kalman, "A New Approach to Linear Filtering and Prediction Problems," Trans. ASME J. Basic Eng. 82D, 34-45 (March 1960).

34. R. E. Kalman and R. S. Bucy, "New Results in Linear Filtering and Prediction Theory," Trans. ASME J. Basic Eng. 83D, 95-108 (March 1961).

35. D. D. Cobb, D. B. Smith, and J. P. Shipley, "Cumulative Sum Charts in Safeguarding Special Nuclear Materials," Los Alamos Scientific Laboratory internal document.

36. A. J. Duncan, *Quality Control and Industrial Statistics*, 3rd ed. (Richard D. Irwin, Inc., Homewood, Illinois, 1965).

37. D. H. Pike, G. W. Morrison, and C. W. Holland, "Linear Filtering Applied to Safeguards of Nuclear Material," Trans. Am. Nucl. Soc. 22, 143-144 (1975).

38. D. H. Pike, G. W. Morrison, and C. W. Holland, "A Comparison of Several Kalman Filter Models for Establishing MUF," Trans. Am. Nucl. Soc. 23, 267-268 (1976).

39. D. H. Pike and G. W. Morrison, "A New Approach to Safeguards Accounting," Oak Ridge National Laboratory report ORNL/CSD/TM-25 (March 1977).

40. D. H. Pike and G. W. Morrison, "A New Approach to Safeguards Accounting," Nucl. Mater. Manag. VI, No. 3, 641-658 (1977).

41. K. B. Stewart, "B-PID and Inventory Estimates with Minimum Variance," Hanford Laboratories report HW-56536 (July 1958).

42. E. A. Hakkila, "A Critical Review of Analytical Techniques for Safeguarding the Uranium-Thorium Fuel Cycle," Los Alamos Scientific Laboratory report LA-7372 (October 1978).

43. E. A. Hakkila, J. W. Barnes, H. A. Dayem, R. J. Dietz, and J. P. Shipley, "Preliminary Concepts: Coordinated Safeguards for Materials Management in a Thorium-Uranium Fuel Reprocessing Plant," Los Alamos Scientific Laboratory report LA-7411-MS (October 1978).

44. L. E. Shuler, L. W. Doher, and L. H. Morrow, "A Displacement Technique for Calibration of Special Nuclear Material Tankage Volumes," Nucl. Mat. Manag. VI, No. III, pp. 472-479 (1977).

APPENDIX A

THE REFERENCE COCONVERSION PROCESS

I. INTRODUCTION

The reference nitrate-to-oxide coconversion process for this study is based on the G.E. Coprecal design, which was released to SRL under the AFCT program. A general description of the process is given in Sec. I. The process description given in this appendix is based on an SRL technical data summary[1] (TDS) and is included here for completeness.

II. THE COCONVERSION AREA

Three parallel process lines are required to meet the design throughput of 117 kg plutonium/day. The reference design includes only the coconversion area; any recyclable waste or scrap will be transferred to other components of the integrated fuel cycle complex. A simplified block diagram of the process is shown in Fig. A-1. Table A-I lists the flow rates for each stream integrated over all three process lines for the continuous portion of the process, and Table A-II lists the amounts of material transferred per batch for the batch portion of the process.

The coconversion area is divided into five process steps: (1) receiving and blending, (2) precipitation, (3) calcination, (4) reduction and stabilization, and (5) screening. Each step is described in the following sections for a single process line.

A. Receiving and Blending

Three parallel feed-blend tanks are provided for each process line. Each tank

(1) receives uranium-plutonium nitrate solution (48 g plutonium/L and 352 g uranium/L in 2.7 $\underline{M}$ HNO_3) from the chemical separations facility,

(2) receives natural uranium nitrate solution (400 g uranium/L in 0.9 $\underline{M}$ HNO_3) from the batch make-up tank to adjust the plutonium content, relative to total heavy metal, from 12% to 10%, and

(3) feeds the process line continuously for 2 weeks at 0.67 L/min with a solution containing 40 g plutonium/L and 360 g uranium/L in 2.1 $\underline{M}$ HNO_3.

TABLE A-I

COPRECAL CONTINUOUS FLOW STREAMS

Stream No.[a]	Total Flow Rate[b] (g/min)	Plutonium Flow Rate[b] (g/min)
1	397 500[c]	12 000
2	397 500[c]	---
3	3 209	81.27
4	1 288	---
5	4 498	81.27
6	222	---
7	232	---
8	1 054	---
9	5 998	81.27
10	3[d]	---
11	5 031	0.04
12	0.3[d]	---
13	5 031	trace[e]
14	1 968	trace[f]
15	3 064	trace[g]
16	970	81.27
17	1	0.09
19	5 897	---
20	39[d]	---
21	5 981	0.04
22	39[d]	---
23	6 020	0.004

---

[a] See Fig. A-1 for stream identification.

[b] Flow rates may not balance exactly because of round-off error.

[c] Flow rate when material is received from chemical separations.

[d] Average flow rate--periodically pulsed to blowback filters.

[e] 0.60 g/day.

[f] 0.25 g/day.

[g] 0.35 g/day.

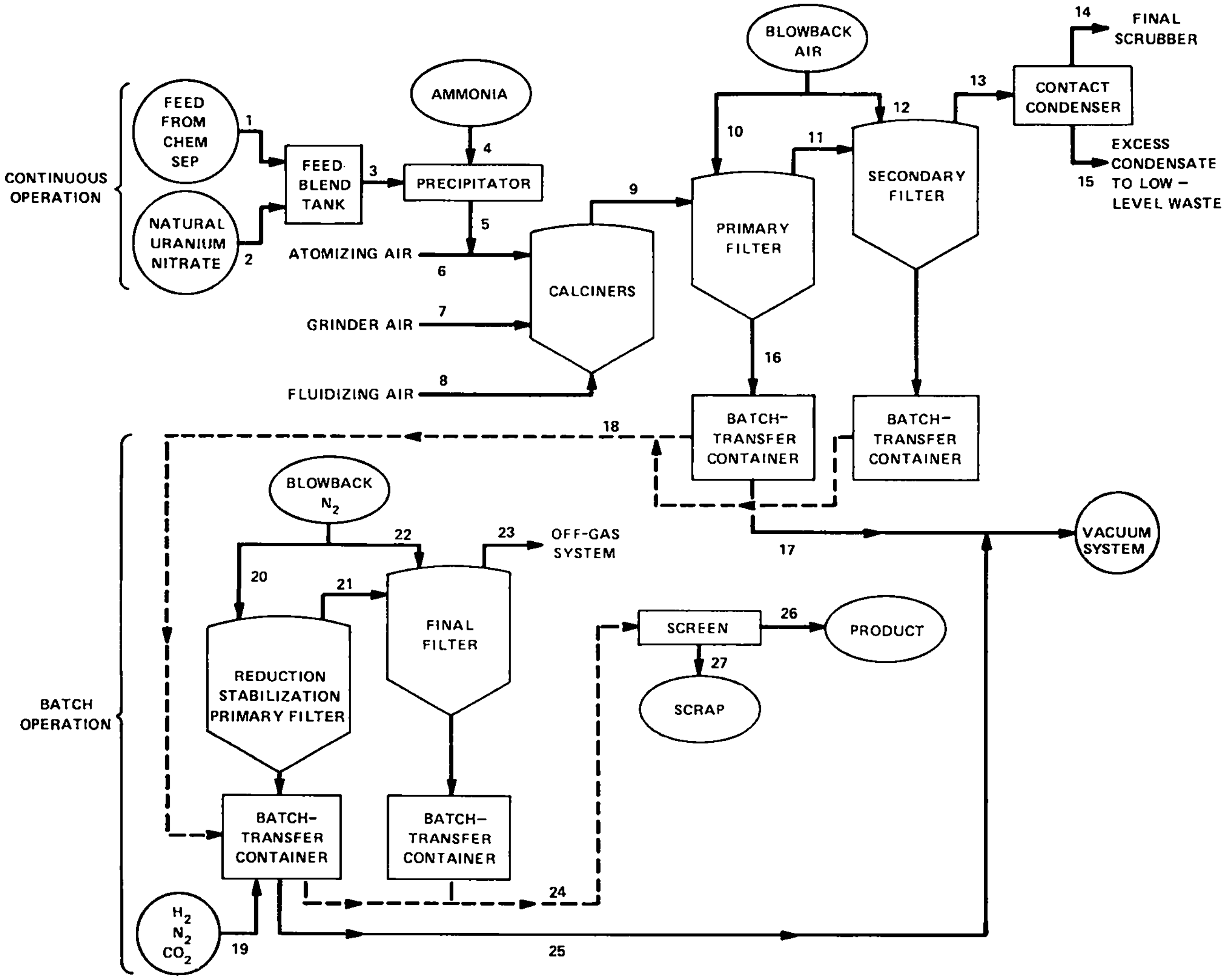


Fig. A-1.
Coprecal simplified flow diagram.

A large tank (18 000 L) with a working volume of 16 000 L is required to feed the process continuously for 2 weeks. Nuclear criticality control in such tanks is achieved with stainless steel tubes (2.54-cm o.d., on 4.08-cm centers) containing boron-bearing materials. A mixing header distributes material uniformly throughout the tank; high-capacity circulation pumps and spargers or air circulators are required for blending and to maintain uniformity in the feed batch over the 2-week period. A low-capacity pump feeds the precipitator at the required rate.

Each tank is equipped with cooling coils to remove heat from plutonium decay and pumping. The tanks are valved to limit feed to one tank at a time to preclude the possibility of tampering with a certified batch.

TABLE A-II

COPRECAL BATCH TRANSFERS

Stream No.[a]	Total[b,c] (kg/batch)	Plutonium[b,c] (kg/batch)
18	38.79	3.25
24	36.66	3.23
25	0.26	0.02
26	36.38	3.21
27	0.28	0.02

---

[a] See Fig. A-1 for stream identification.

[b] Flow rates may not balance exactly because of round-off error.

[c] A batch is processed every 40 min.

Each tank is provided with temperature, specific gravity, level (high- and low-level alarms), and pump pressure measurements for process control. Each tank is also provided with sample lines.

Operating parameters for the feed-blend tanks are summarized in Table A-III. A single tank is shown in Fig. A-2.

B. Precipitation

Uranium-plutonium-nitrate solution from a feed-blend tank is pumped at ∿0.67 L/min to a single precipitator. Ammonia (27-30% NH_3) is added at ∿0.5 L/min to maintain a pH >10 to assure coprecipitation; plutonium precipitates preferentially at a pH <3.5. The ammonia reacts with the nitrate solution to form an

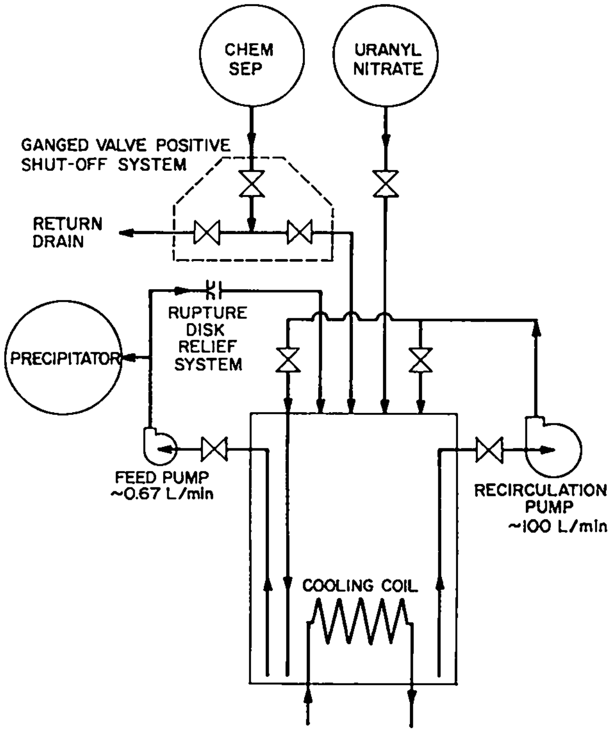


Fig. A-2.
Feed-blend tank.

TABLE A-III

FEED-BLEND TANK OPERATING PARAMETERS

Number of tanks/line	3
Tank capacity (L)	18 000
Normal working volume (L)	16 000
Pu concentration (g/L)	
Receipt	48
Product	40
Heavy metal concentration (g/L)	400
Pu tank content (full) (kg)	640
Feed rate (L/min)	0.67
Feeding time (weeks)	2
Analytical sample frequency	1 per batch

unfilterable precipitate of plutonium hydroxide, ammonium diuranate, and ammonium nitrate. The reactions occurring in the precipitator are

$$Pu(NO_3)_4 + 4NH_3 + 4H_2O \longrightarrow Pu(OH)_4 + 4NH_4NO_3 \quad , \tag{A-1}$$

$$2UO_2(NO_3)_2 + 5NH_3 + 5H_2O \longrightarrow 2UO_3 \cdot NH_3 \cdot 3H_2O + 4NH_4NO_3 \quad , \tag{A-2}$$

and

$$HNO_3 + NH_3 \longrightarrow NH_4NO_3 \quad . \tag{A-3}$$

The precipitator slurry is pump-circulated at 100 L/min to keep the slurry dispersed and to insure intimate mixing of the ammonia and the nitrate solutions. A portion of the circulation stream is fed through orifice plates to four parallel calciners. The calciner feed is orificed so that the slurry flow to all four units is equal.

The precipitator is a cylinder 15 cm in diameter, 2.5 m high, having a 35-L minimum working volume and a 10-L freeboard. The precipitator overflows to the sump system. The residence time in the precipitator is approximately 30 min, providing a surge volume and allowing variability in the reagent addition rates. The residence time is not required for digestion.

Each precipitator requires measurements of flow (nitrate and ammonia input), level (with high- and low-level alarms), pH, and pump discharge pressure for process control.

A block diagram of the precipitator is given in Fig. A-3. The precipitator operating parameters are summarized in Table A-IV.

C. Calcination

The calcination process involves calcination in fluidized-bed calciners and filtration of the calciner off gas. Four parallel fluidized-bed calciners are required for each process line. Each unit is a cylinder 15 cm in diameter and 1 m high. The calciners are manifolded through an off-gas cooler to a single primary filter connected to a secondary filter. The calciner system is shown in Fig. A-4.

Feed nozzles meter the precipitator slurry to each calciner where it is air injected into the fluidized bed. The fluidized bed is maintained at 400°C. The slurry contacts the fluidized bed, producing plutonium and uranium oxide (PuO_2 and UO_3). The reactions occurring in a calciner are

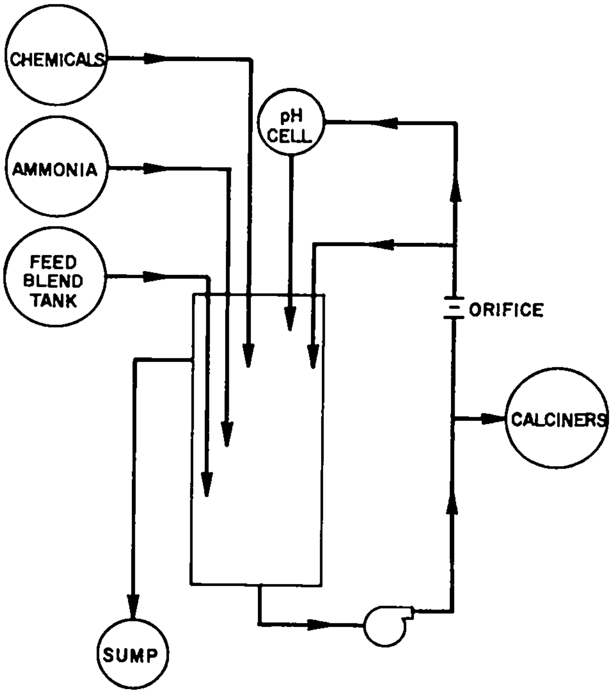


Fig. A-3.
Precipitator.

TABLE A-IV

PRECIPITATOR OPERATING PARAMETERS

Number/line	1
Capacity (L)	45
Minimum working volume (L)	35
Nominal Pu inventory (g)	800
Normal receipt flow (L/min)	
U-Pu	0.67
Ammonia	0.5

$$Pu(OH)_4 \longrightarrow PuO_2 + 2H_2O \quad , \qquad (A\text{-}4)$$

$$2UO_3 \cdot NH_3 \cdot 3H_2O \longrightarrow 2UO_3 + NH_3 + 3H_2O \quad , \qquad (A\text{-}5)$$

and

$$3NH_4NO_3 + 2NH_3 \longrightarrow 4N_2 + 9H_2O \quad . \qquad (A\text{-}6)$$

Most of the excess ammonia added at the precipitator will pass through the calciner, although the reaction

$$2NH_3 + 2O_2 \longrightarrow N_2O + 3H_2O \qquad (A\text{-}7)$$

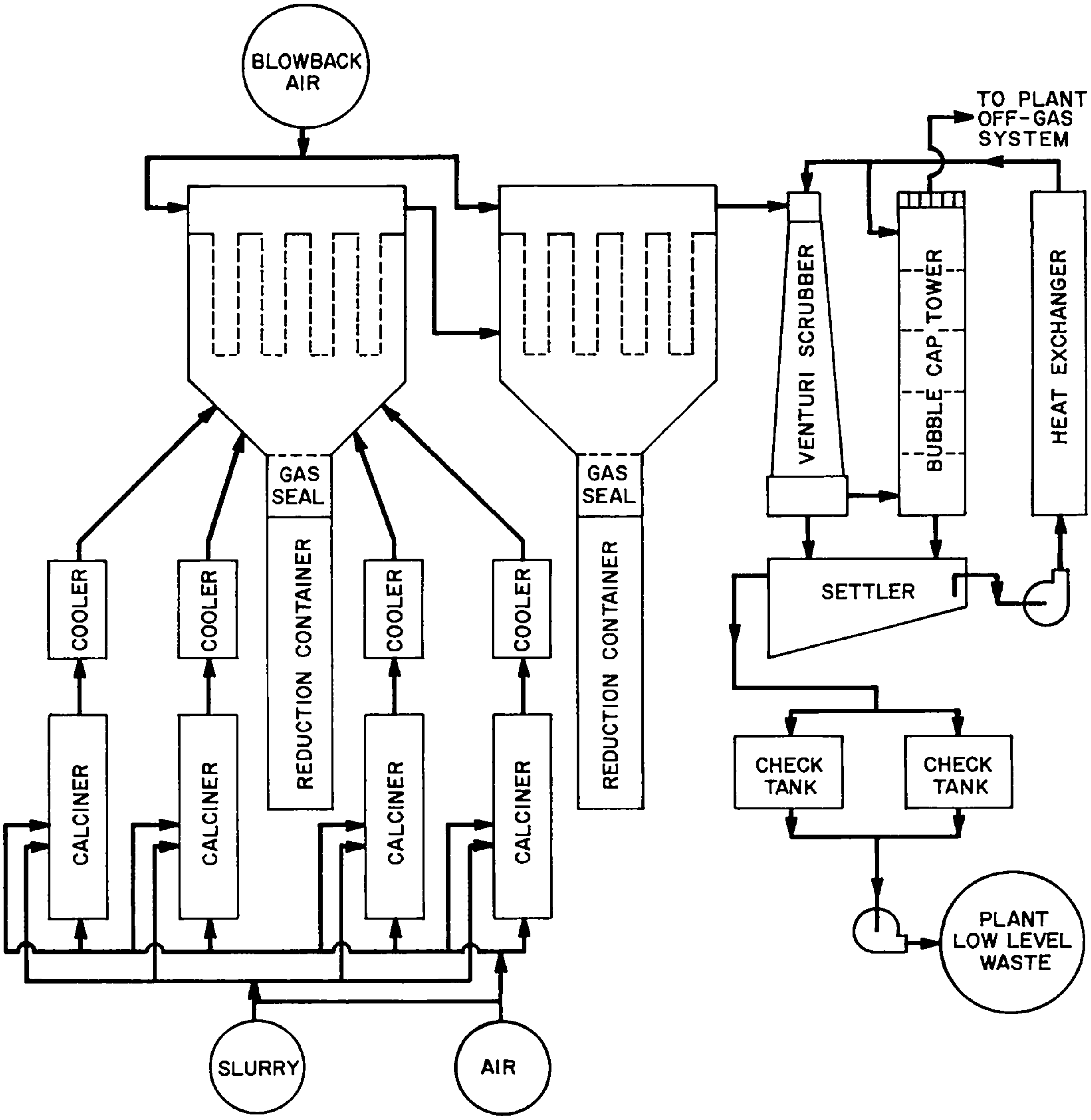


Fig. A-4.
Calciner system.

may take place to a limited extent. Reaction kinetics are thought to be unfavorable at 300-400°C. Hot ammonia (>1000°C) could oxidize on the metal filters in the primary filter, forming HNO_3. To preclude this possibility, a gas cooler is provided between the calciner and the primary filter to reduce the off-gas temperature to ∿300°C.

The uranium- and plutonium-oxide powders formed in the calciners are carried by the off-gas stream to the calciner filter system. The off-gas stream also contains process-generated steam, decomposition gases, and air. The gas passes through the gas cooler, the primary filter, which collects the product, the secondary filter, and the off-gas treatment system before being discharged to the plant off-gas system.

The primary and secondary filters are annular cylinders 7.6 cm thick, 3.5-m o.d., and 1.2 m tall. Each contains 43 equally spaced filter elements 5.1 cm in diameter and 91 cm long. The filter elements are porous Inconel metal with a 5-μm rating. The powder collected on the filter elements is blown back periodically and collects at the bottom of the filter. The bottom of the filter chamber is conical to aid in collection of the powder and its discharge to the reduction container.

The primary filter is unloaded approximately every 30 min through a powder valve into a reduction container. The filled container is replaced every 2 h and is transferred to a reduction-stabilization station.

The off gas passes through the primary to the secondary filter. The powder content of the filtered gas is very low, requiring that the reduction container be disconnected from the secondary filter only at the completion of an operating campaign (approximately every 2 weeks).

Gases leaving the secondary filter are cooled in a contact condenser and gas-scrubbing system (see Fig. A-4). Liquid waste from the condenser and scrubber are discharged to a recirculation settler slab tank (7.6 cm thick, 100 L). The settler overflows to one of two parallel condensate check tanks (3000 L). After sampling, the check-tank contents are discharged to the low-level waste treatment portion of the reprocessing complex.

Calciner process monitoring requires measurement of the fluidized-bed temperature, slurry feed rate, air flow, and air temperature. Differential pressures are measured across the primary and secondary filters. The off-gas treatment system requires measurement of scrubber vacuum, mist-eliminator differential pressure, heat-exchanger temperatures, and of check-tank level, specific gravity, and temperature. Monitors are also required to detect unusual plutonium concentrations in the scrubber, which would indicate failure of a calciner rupture disk.

D. Reduction and Stabilization

After the reduction container is disconnected from the calciner primary filter, it is transferred to the reduction-stabilization area. The process becomes a batch operation at this point. Four parallel reduction-stabilization stations are required for each process line because the operation takes 8 h per batch. A single station is shown in Fig. A-5.

The reduction container is connected to the filter station and gas supply. The system is evacuated, and the unit is heated (∿550°C primary filter, ∿150°C final filter). A heated (∿550°C) mixture of 6% hydrogen and 94% nitrogen is blown through the powder fast enough to turn the powder over, but not to blow it out of the container. The hot hydrogen reduces the UO_3 by the following reaction.

$$UO_3 + H_2 \longrightarrow UO_2 + H_2O. \tag{A-8}$$

After reduction, hot CO_2 (∿525°C) is introduced to stabilize the powder so that the UO_2 does not reoxidize to U_3O_8. The stabilization equation is

$$UO_2 + 0.07\ CO_2 \longrightarrow UO_{2.07} + 0.07\ CO. \tag{A-9}$$

At completion of the reduction-stabilization cycle the container is cooled by passing room-temperature CO_2 through the powder and by external air cooling.

The reduction-stabilization primary and final filters are cylindrical vessels 30.5 cm in diameter and 1.2 m tall. The bottom is conical to aid in powder collection and dispensation. Each vessel contains 11 Inconel elements similar to those in the calciner filters. The filters are blown back with nitrogen.

The powder loading in the gas discharged from the primary to the final filter is very light unless there is a break in the primary filter. Therefore the final filter will be unloaded infrequently (at physical inventory).

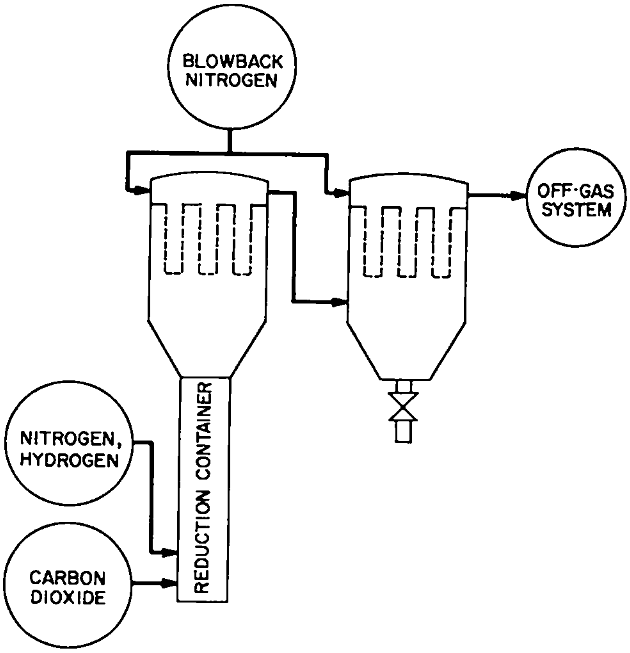


Fig. A-5.
Reduction-stabilization station.

Process control requires measurements of reduction-container weight before and after processing, filter differential pressure, and gas flow and temperature.

E. Screening

At the completion of the reduction-stabilization and cooling cycle, the reduction container is transferred to the screening station. Its contents are dumped onto a belt that feeds the powder to a vibrating screen (100 U.S. mesh size). The screen removes oversized particles and foreign matter and discharges the product into a tared storage can (a 20.3-cm-diam and 91.3-cm-tall cylinder). After the batch is processed, this storage container is removed, sampled, weighed, sealed, and transferred to the storage vault. Material collected on the screen is dumped into a tared scrap container, which is sent to scrap recovery when full.

Measurements of the reduction-container weight before and after filling and the storage-container weight are required for process control.

III. THE STORAGE VAULT

The vault receives certified batches from all three process lines. The storage containers are transferred by remote control into and out of the vault through a monitored access portal. Personnel do not have routine access to the vault.

The vault serves only as a buffer between the coconversion process and the fuel-fabrication process. It therefore has a capacity of only 500 cans (∿2 weeks of production).

IV. FACILITY LAYOUT

The facility layout was developed with the philosophy that all process equipment might fail in such a way that hands-on maintenance could not be accomplished quickly. The facility is therefore designed for remote equipment removal and replacement. Failed equipment is transferred to a centralized decontamination facility for cleanout and decontamination, then to gloveboxes for repair or disposal. Figures A-6 through A-9 show the facility layout.

The facility is contained in a single canyon having one process cell for each process line and one storage vault. As currently designed all material must be removed from the process lines before personnel can enter the canyon.

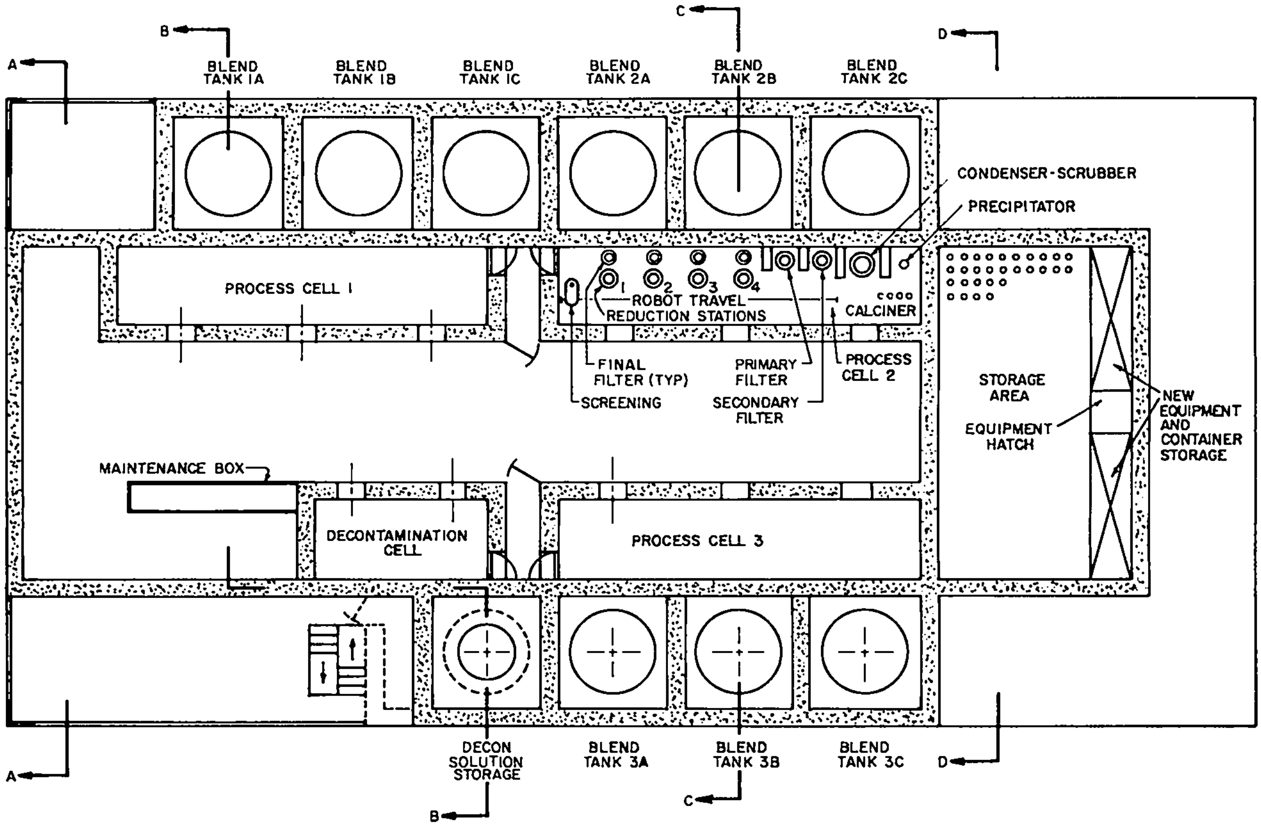


Fig. A-6.
Coprecal facility layout, grade level.

Process cells have shielded windows and manipulators in the maintenance gallery. The manipulators are used for simple, in-place repairs. An overhead crane performs all equipment transfers.

All material and equipment are transferred into and out of the canyon through hatches in the storage area and must pass through the controlled portals in the storage area.

Each process line is served by a remotely operated, programmable robot. The robot travels along ∿12-m-long tracks parallel to the process line (from calciners to screen). The robot performs the following functions.

- Obtains an empty reduction container, transfers it to the calciner primary or secondary filter, and connects the container to the filter.
- Disconnects and moves a filled container from the calciner primary filter to one of four parallel reduction-stabilization stations.

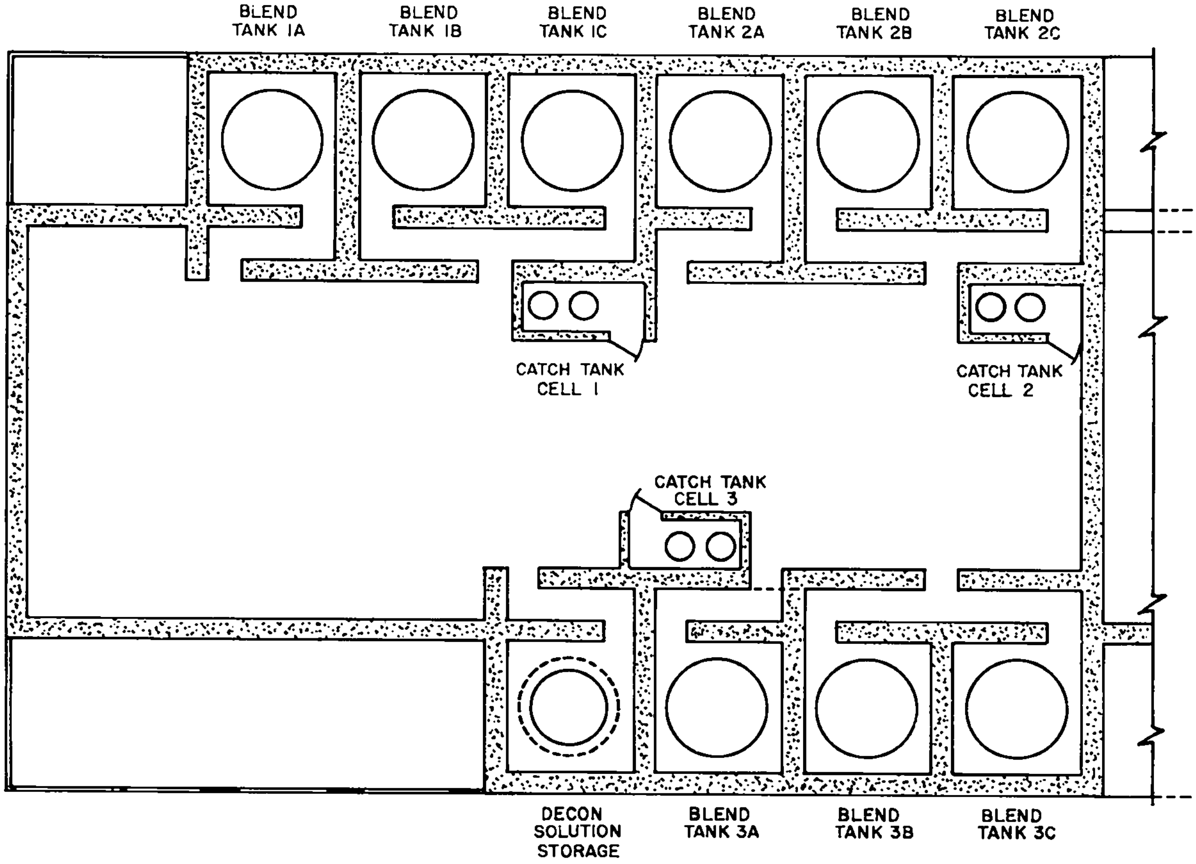
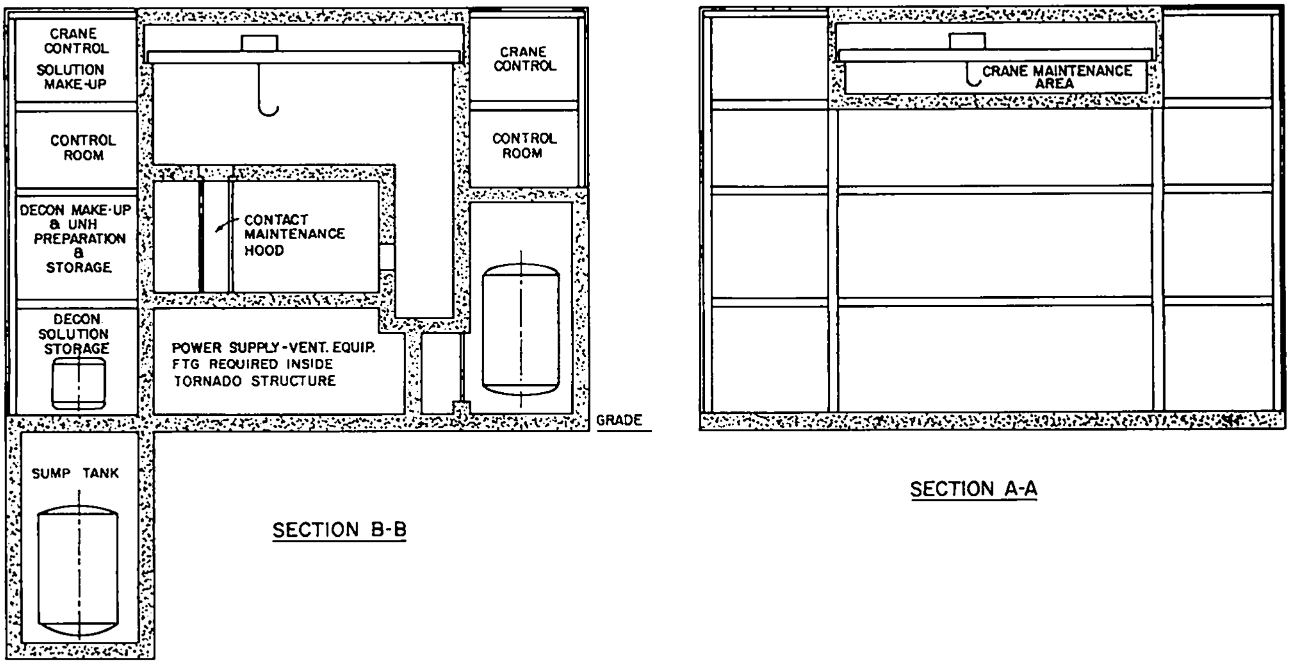


Fig. A-7.
Coprecal facility layout, 2nd level.

- Connects the container to a reduction-stabilization station and makes the proper gas line connections.
- Disconnects the container from a reduction-stabilization station and transfers it to the screening station.
- Inverts the container into the screening station feeder.
- Obtains an empty storage container and places it under the screen.
- Disconnects the container from the screen and transfers it to the storage vault.
- Disconnects the emptied container from screen and transfers it to rack for reuse.

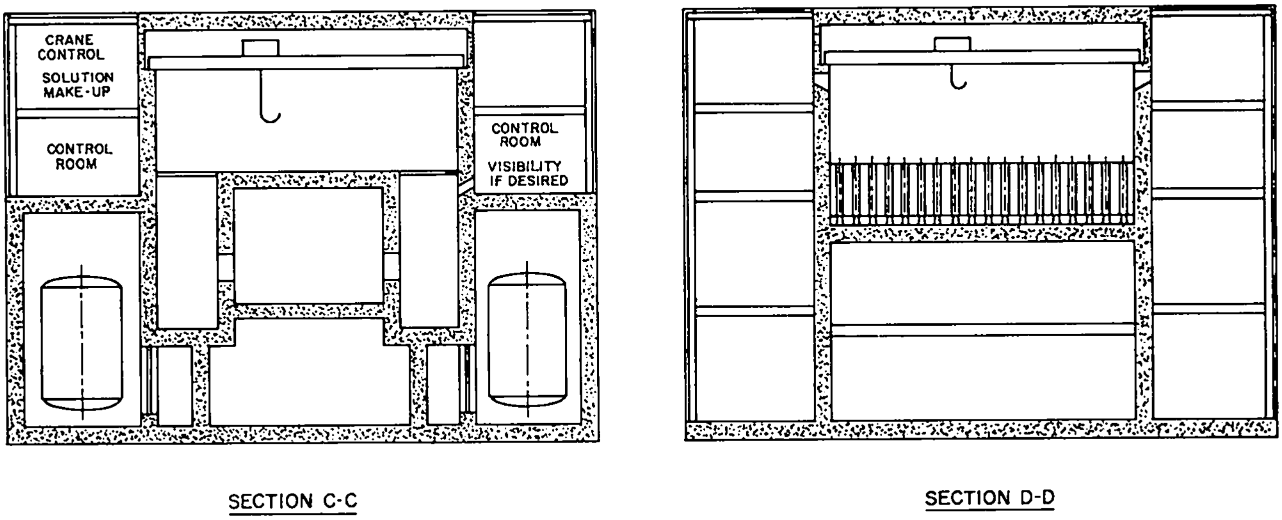
The robot is designed to transfer 100-kg cans. Each robot must process 12 batches through a line per day. Each batch requires eight major moves.



SECTION B-B

SECTION A-A

Fig. A-8.
Coprecal facility layout, sections A-A, B-B.



SECTION C-C

SECTION D-D

Fig. A-9.
Coprecal facility layout, sections C-C, D-D.

The process areas contain solid and liquid sumps. Each solids area in each process line is a sump with a vacuum system for cleaning spilled materials. Each liquids area has its own criticality-safe sump. A poisoned-sump tank is provided in the lowest level of the building to accept drainage from all the individual sumps.

REFERENCE

1. Savannah River Laboratory document DPSTD-AFCT-77-10-TL (November 1977).

APPENDIX B

DYNAMIC MODEL OF THE COCONVERSION PROCESS

I. INTRODUCTION

The design and evaluation of the MMAS is based on computer simulations of the reference coconversion facility. The use of simulation techniques permits prediction of the dynamic behavior of materials flows over the range of possible operating conditions and provides a means for rapid accumulation of data for relatively long operating periods. Alternative materials measurement strategies and safeguards data-analysis algorithms are readily compared. In principle, the necessary data could be obtained from experiments on test loops and mockups of plant operation, but this is both time consuming and expensive. Carefully selected test loops can be used more effectively to validate the computer models and to test portions of the final MMAS design.

Modeling and simulation have been used extensively in coordinated safeguards studies of chemical separations (see Ref. 1, Chap. VI and Apps. E and G), plutonium nitrate-to-oxide conversion facilities (see Ref. 2, Chap. IV and App. A), and mixed-oxide fuel fabrication (see Ref. 3, Chap. IV and App. D). This approach requires[4] (1) a detailed dynamic model of the process based on actual design data; (2) simulation of the model process on a digital computer; (3) a model for each measurement system; (4) simulation of accountability measurements applied to SNM flow and in-process inventory data generated using the model process; and (5) evaluation of simulated data from various materials accounting strategies.

This appendix describes COPSIM, a computer model written to simulate the dynamic behavior of the Coprecal process under anticipated normal operating conditions. Measurement models and data-evaluation techniques are discussed in Secs. II and III.

II. THE MODEL PROCESS

Coprecal, the model coconversion process, is based on a G.E. design (see Sec. I and App. A).[5] The process converts uranium-plutonium-nitrate solution received from a chemical separations facility to mixed uranium-plutonium-oxide powder that is to be transferred to a mixed-oxide fuel fabrication facility. Coprecipitation in ammonium hydroxide is followed by calcination of the entire precipitator product. The reference facility has a design basis throughput of 117 kg plutonium/day through three parallel process lines.

A single Coprecal process line is shown in Fig. B-1. Plutonium-uranium-nitrate solution (∿30 g plutonium/kg) from the chemical separations facility is transferred to one of three parallel feed-blend tanks. Approximately 21 000 kg of solution are transferred to the tank at ∿398 g/min (reduced to ∿50 g/min when ∿18 000 kg have been transferred). The plutonium concentration is adjusted to ∿25 g plutonium/kg by adding natural- uranium-nitrate solution at ∿398 g/min. The tank is then sampled and certified to feed the process line.

A feed-blend tank feeds the process line continuously at ∿1.07 kg/min until a heel of ∿258 kg is reached. The tank is mixed continuously to maintain a constant plutonium feed concentration.

Ammonium hydroxide is added to the precipitator at ∿0.4 kg/min. The precipitator in-process heavy-metal (HM) inventory is controlled at ∿10.6 kg HM and the total mass at ∿59 kg. The uranium-plutonium ratio of the precipitator product is assumed to be the same as that of the in-process inventory. The precipitator slurry is fed equally to four parallel calciners. The in-process HM inventory in each calciner is controlled at ∿9 kg. The calcined powder is discharged to the primary filter. Approximately 0.5% of the entering powder is discharged to the secondary filter and ∿0.7 g HM/min are lost to the vacuum system. Powder from the primary filter is discharged to a reduction container. The container is disconnected every 2 h. At this time the holdup in the primary filter is ∿6.5 kg HM. Approximately 1% of the powder entering the secondary filter is lost to the off-gas treatment system. The reduction container from the secondary filter is removed when the feed-blend tank operation is switched.

A reduction container is transferred from the primary filter to one of four parallel reduction-stabilization stations. The residence time in each station is 8 h. The holdup in each reduction-stabilization filter and in each final filter is ∿1.85 kg HM. Approximately 0.002 g HM/min pass from a reduction-stabilization primary filter to the final filter and ∿10% of that is lost from the final filter to the off-gas treatment system. Because of powder spills and leaks, ∿2 g HM/min are lost from the reduction-stabilization station to the vacuum system.

At the completion of the reduction-stabilization cycle, the reduction container is transferred to the screen where 27 g HM/batch of oversized particles are removed.

THE COPRECAL MODEL PROCESS

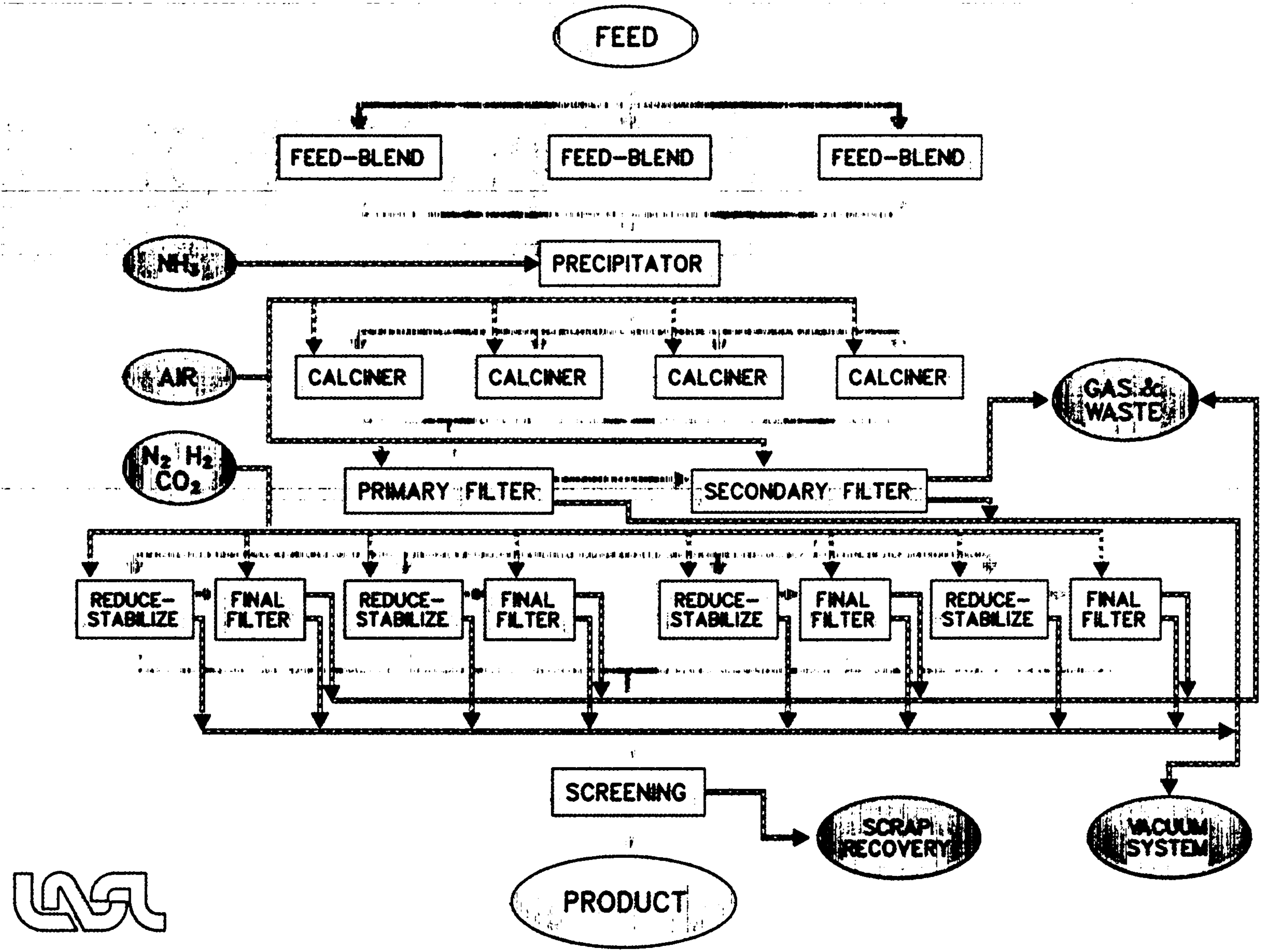


Fig. B-1.
The Coprecal model process.

III. THE DYNAMIC MODEL

Operation of the model process is simulated using standard Monte Carlo techniques developed for dynamic systems.[6,7] The model process can be divided into continuous and batch operations. The continuous portion includes the feed-blend tanks, the precipitator, the calciners, and the primary and secondary filters. Reduction-stabilization and screening are batch operations.

The dynamics of each process operation (that is, each process step shown in Fig. B-1) are described by the continuity equation written for the flows of bulk materials and SNM.

$$\frac{dM(t)}{dt} = \sum F_i(t) \quad , \tag{B-1}$$

and

$$\frac{dS(t)}{dt} = \sum C_i(t)\ F_i(t) \quad , \tag{B-2}$$

where $M(t)$ = bulk material in process at time t,
$S(t)$ = SNM in process at time t,
$F_i(t)$ = bulk mass flow rate at time t in stream i (input positive, output negative), and
$C_i(t)$ = SNM concentration at time t in stream i.

The solution of a complete set of differential equations for all process operations, subject to initial conditions and process constraints, determines the process dynamics. Random variation in the process is determined by the statistics selected for each independent process variable. The independent variables and their ranges for the Coprecal model process are given in Table B-1.

The time dependence of all flow rates is linear over sufficiently small time steps (Δt). The SNM concentration is either constant (for example, precipitator feed) or linear (for example, precipitator inventory). With these assumptions the quantity of bulk material transferred in Δt is

$$M(t_2) = \frac{F(t_1) + F(t_2)}{2} \Delta t \quad , \qquad \text{(B-3)}$$

where $\Delta t = t_2 - t_1$.

TABLE B-1

INDEPENDENT PROCESS VARIABLES IN THE MODEL COPRECAL PROCESS

Identification	Nominal Value	Range (%)
Feed-blend tank		
Feed (chemical separations)		
Flow rate	397.5 kg/min	5
Plutonium concentration	30.2 g/kg	10
Heavy-metal concentration	251.6 g/kg	5
Blend (natural U)		
Flow rate	397.5 kg/min	5
U concentration	251.6 g/kg	5
Product		
Flow rate	1.07 g/min	2
Precipitator in-process inventory		
Total mass	59.2 kg	13
Heavy metal	10.6 kg	7
Calciner in-process inventory		
Heavy metal	9.0 kg	10
Primary filter holdup		
Heavy metal	7.5 kg	20
Secondary filter holdup		
Heavy metal	7.5 kg	20
Reduction-stabilization		
Heavy-metal holdup	1.8 kg	10
Final filter		
Heavy-metal holdup	1.8 kg	10

The total amount of SNM transferred in Δt is

$$\bar{S}(t_2) = \bar{M}(t_2)\ C \tag{B-4}$$

for constant C, or

$$\bar{S}(t_2) = \left[C(t_1)\ F(t_1) + C(t_2)\ F(t_2)\right]/3 + \left[C(t_1)\ F(t_2) + C(t_2)\ F(t_1)\right]/6 \tag{B-5}$$

for C linear in time.

The SNM inventory after Δt is

$$S(t_2) = S(t_1) + \bar{S}(t_2) \quad . \tag{B-6}$$

The independent process variables execute a random walk in time. The variation of each variable is uniform over the interval given in Table B-1. To limit the process variability rate so that the model corresponds more closely to the expected process behavior, the random walk is constrained. A minimum of 1 h is required to make a maximum change from one boundary to the other, and the range of variation is proportional to the length of the time step.

For example, the mass flow rate out of the feed-blend tank is the only independent variable while the tank is feeding the process. At each event the flow rate (F_i) is chosen from a uniform distribution with a 1.05 to 1.09 kg/min range. Events are scheduled every 30 min; therefore, the flow rate is chosen so that it does not differ from the previous flow rate (F_{i-1}) by more than 0.02 kg/min. The total amount of material transferred in 30 min is

$$\bar{M}_i = 15\ (F_{i-1} + F_i) \quad . \qquad \text{(B-7)}$$

The total mass left in the tank becomes

$$W_i = W_{i-1} - \bar{M}_i \quad . \qquad \text{(B-8)}$$

The plutonium concentration in the tank is constant, the total amount of plutonium transferred out of the tank is

$$\bar{T}_i = C\ \bar{M}_i \quad , \qquad \text{(B-9)}$$

and the in-process plutonium inventory is

$$S_i = S_{i-1} - \bar{T}_i \quad . \qquad \text{(B-10)}$$

IV. THE COPSIM COMPUTER CODE

The computer code COPSIM has been developed to simulate the model Coprecal process. COPSIM was written in FORTRAN and uses the GASP IV simulation package to schedule events.[8] The program is run on a CDC 6600 and requires ∿50 000 words of core memory.

Input data include initial values for all process variables and the range of variability for each independent process variable. COPSIM is designed for ease of sequential running so that extended operating periods can be simulated. Each COPSIM simulation run generates a restart file to continue the simulation where it ended.

Each process step shown in Fig. B-1 is modeled separately; in most cases an individual subroutine is written for each step. Events are scheduled through the GASP attribute file. When an event is scheduled in a particular process step, the values of all independent process variables are sampled from the given range, the value of all dependent process variables are calculated, and then all variables are stored in a data vector containing 17 elements. The data vectors from each process step are stored sequentially in a separate data file. This format is useful for further processing and as input to the measurement simulation programs.

V. EXAMPLES

Figures B-2 through B-10 are examples from a 24-hour period of simulated process operation. In each graph the process variable is plotted at the event time (events can be 5 min, 30 min, 2 h, or 8 h apart depending on the process step). During this particular day, feed-blend tanks are switched at 11.5 h.

Figure B-2 shows the mass and plutonium concentration of the in-process inventory solution in the feed-blend tank that is feeding the process at the start of the day. The tank reaches the desired heel at 11.5 h and is removed from the process while another tank is switched to the process line. The removed tank then receives solution from chemical separations, and the solution is blended with natural uranium nitrate to adjust the plutonium concentration. Note that during the feeding process, events are 30 min apart because the flow rate is low ($\sim$1 kg/min), whereas events are 5 min apart when the tank is receiving solution because the flow rates are high ($\sim$400 kg/min).

The precipitator-feed flow rate and plutonium concentration are shown in Fig. B-3. The concentration changes at 11.5 h when the feed-blend tanks are switched; otherwise the concentration is constant. The flow rate varies between 1.05 and 1.09 kg/min. Note that adjacent flow rates do not differ by more than 0.02 kg/min.

The total mass and plutonium concentration of the precipitator in-process inventory are shown in Fig. B-4. The solution mass varies between $\sim$52 and $\sim$67 kg. The change of mass between adjacent events is not more than $\sim$7.5 kg because the time between events is 30 min and the random walk is constrained so that at least 1 h is required to make a change from one boundary to the other ($\sim$15 kg). The total amount of heavy metal varies between $\sim$9 and 11 kg.

Figure B-5 shows the plutonium in-process inventory for a single calciner (4 calciners per process line). The calciner inventory is controlled at 9 kg HM $\pm$ 10%.

The plutonium content of the reduction container, following removal from the primary filter, and the plutonium holdup in the filter (again when the container is removed) are shown in Fig. B-6. The reduction container is removed every 2 h. The

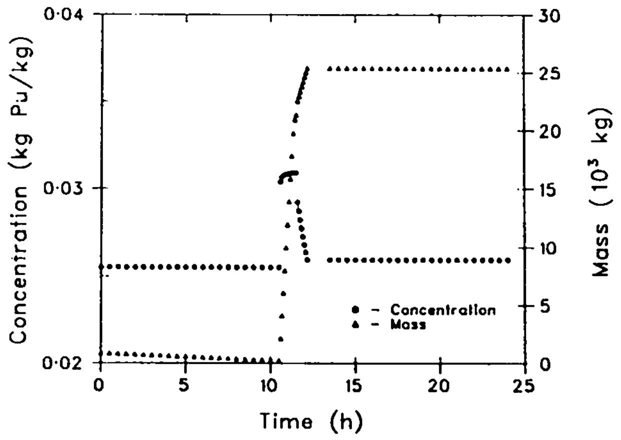


Fig. B-2.
Feed-blend tank in-process inventory: total mass and concentration.

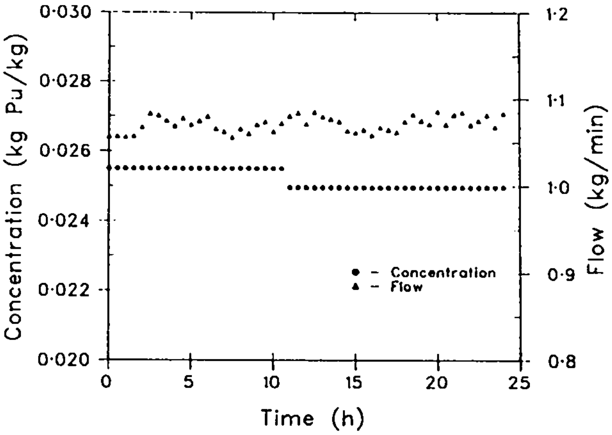


Fig. B-3.
Precipitator feed: flow rate and concentration.

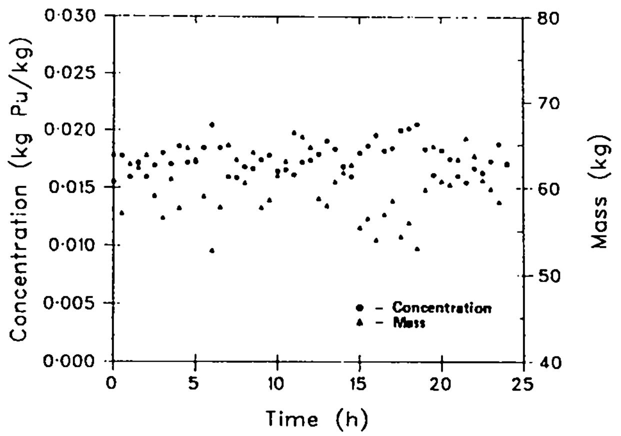


Fig. B-4.
Precipitator in-process inventory: total mass and concentration.

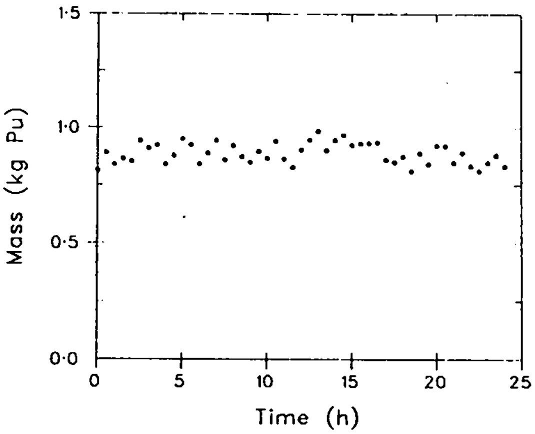


Fig. B-5.
Calciner in-process inventory.

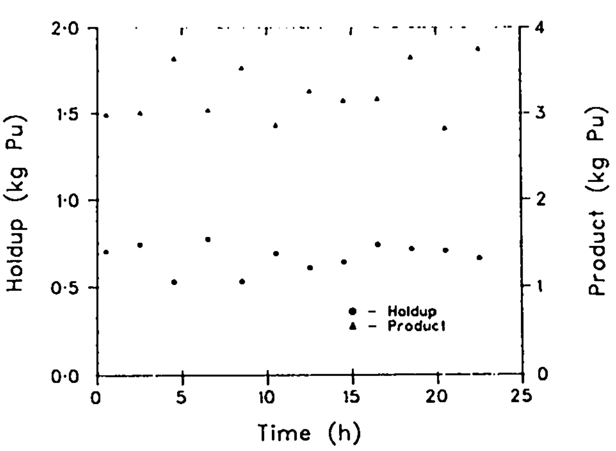


Fig. B-6.
Calciner primary filter: product and holdup.

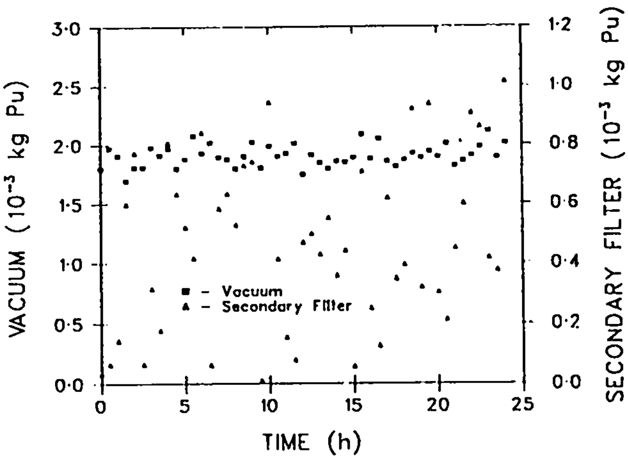


Fig. B-7.
Calciner primary filter losses: vacuum and secondary filter.

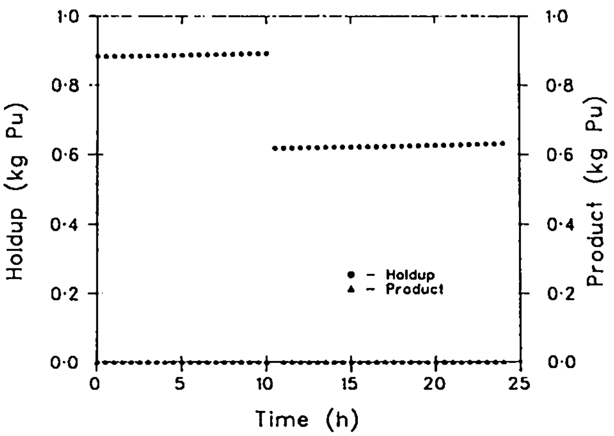


Fig. B-8.
Calciner secondary filter: product and holdup.

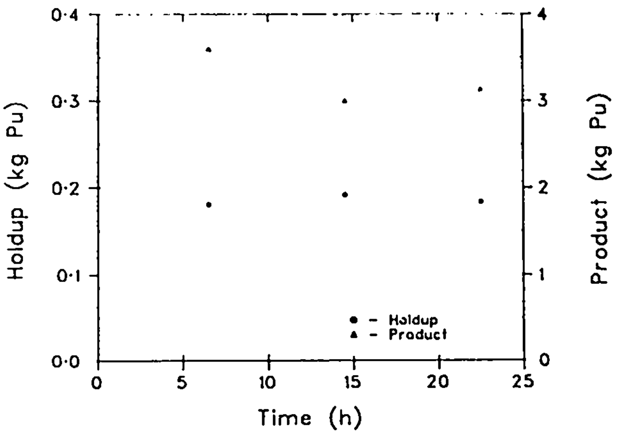


Fig. B-9.
Reduction-stabilization primary filter: product and holdup.

heavy-metal holdup in the filter is controlled to vary between 5 and 8 kg. The plutonium holdup is the product of heavy-metal holdup and the plutonium fraction of the input material since the last event.

Figure B-7 shows the plutonium losses from the primary filter to the vacuum system and to the secondary filter integrated over 30-min periods. Loss to the vacuum system is ∿2 g plutonium/30 min. Plutonium powder loss to the secondary filter varies between 0 and 0.1% of the primary filter input.

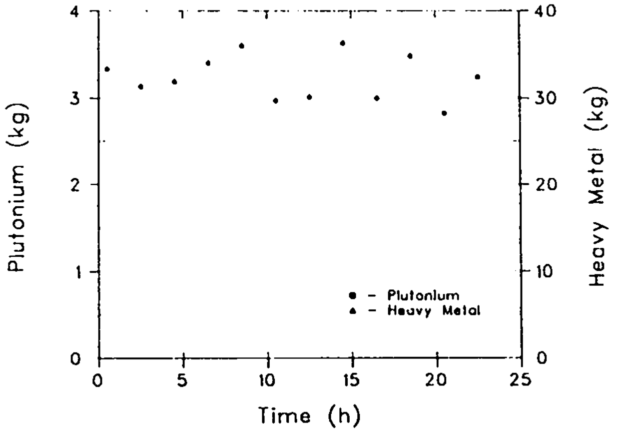


Fig. B-10.
Product canister contents.

The secondary filter plutonium holdup and product are shown in Fig. B-8. The plutonium holdup increases gradually until the filter is cleaned. The filter is blown back periodically, but the powder is removed only when feed-blend tanks are switched.

Figure B-9 shows the plutonium holdup and product for a single reduction-stabilization station (four parallel stations per process line). The heavy-metal holdup is controlled at 1.8 kg ± 10%.

The plutonium content in the product canisters for this single process line is shown in Fig. B-10. The heavy-metal content is almost 10 times the plutonium content. The product batches at 8, 16, and 24 h are the batches discharged from the reduction-stabilization station shown in Fig. B-9 minus any losses in the screening operation.

REFERENCES

1. 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 Labortory report LA-6881 (September 1977).

2. H. A. Dayem, D. D. Cobb, R. J. Dietz, E. A. Hakkila, E. A. Kern, J. P. Shipley, D. B. Smith, and D. F. Bowersox, "Coordinated Safeguards for Materials Management in a Nitrate-to-Oxide Conversion Facility," Los Alamos Scientific Laboratory report LA-7011 (April 1978).

3. J. P. Shipley, D. D. Cobb, R. J. Dietz, M. L. Evans, E. P. Schelonka, D. B. Smith, and R. B. Walton, "Coordinated Safeguards for Materials Management in a Mixed-Oxide Fuel Facility," Los Alamos Scientific Laboratory report LA-6536 (February 1977).

4. D. D. Cobb and D. B. Smith, "Modeling and Simulation in the Design and Evaluation of Conceptual Safeguards Systems," Proc. 18th Annual Meeting on the Institute of Nuclear Materials Management, Washington, D.C., June 29-July 1, 1977.

5. Savannah River Laboratory document DPSTD-AFCT-77-10-TL (November 1977).

6. J. Kleijnen, Statistical Techniques in Simulation (Marcel Dekker, New York, 1974).

7. T. Naylor, J. L. Balintfy, D. S. Burdick, and K. Chu, Computer Simulation Techniques (John Wiley & Sons, New York, 1966)..

8. A. Pritsker, The GASP IV Simulation Language (Wiley-Interscience, New York, 1976).

APPENDIX C

COPRECAL MATERIALS ACCOUNTING EQUATIONS

I. INTRODUCTION

The MB and cusum equations and their associated variance equations for the basic accounting strategy in the Coprecal process are given in this appendix. The basic materials accounting strategy contains three UPAAs: (1) feeding, (2) precipitation-calcination, and (3) reduction-stabilization-screening.

The measurement points and types of measurements used for dynamic materials accounting are given in Table III. Both additive and multiplicative measurement-error models are used (see Sec. IV). In this study the additive model is used for the weighing devices, and the multiplicative model is used for all other measurements.

A materials balance is drawn about each UPAA when a batch is discharged, approximately every 2 h. The form of a materials balance equation depends on whether the UPAA consists of a continuous or a batch process. The n^{th} materials balance for a continuous UPAA is given by

$$MBC_n = I_{n-1} - I_n + T_n \quad , \qquad (C\text{-}1)$$

where I is the measured materials inventory and T is the measured net materials transfer (input positive and output negative). The n^{th} materials balance for a batch UPAA is given by

$$MBB_n = HU_{n-1} - HU_n + T_n \quad , \qquad (C\text{-}2)$$

where HU_{n-1} and HU_n are the measured materials holdup in that UPAA before and after the batch is processed, and T_n is the measured net change of SNM in the batch (the measured input minus the measured output of that batch).

II. UPAA ONE

The first UPAA contains the feed-blend tanks.

A. Materials Balance

Dynamic materials balances are drawn about the feeding process by combining a single SNM concentration measurement (obtained when the tank is analyzed and certified) with a flow measurement of the tank product every 1/2 h and a measurement of the tank mass every 2 h.

The materials balance at time n is given by

$$MB1_n = C_n \left(W_{n-1} - W_n - \Delta t \sum_{4(n-1)}^{4n} A_i \, F_i \right) , \qquad (C\text{-}3)$$

where

C = solution plutonium concentration (kg Pu/kg),

W = tank total mass,

Δt = time between flow measurements (uniform time intervals, 30 min)

= $t_i - t_{i-1}$,

$$A_i = \begin{cases} 1/2 & i = 4n \text{ or } i = 4(n-1) \\ 1 & \text{otherwise, and} \end{cases}$$

F = mass flow rate (kg/min).

The materials balance equation can be rewritten as

$$MB1_n = \Delta I1_n + T1_n \quad , \qquad (C\text{-}4)$$

where

$$\Delta I1_n = I1_{n-1} - I1_n = C_n\left(W_{n-1} - W_n\right) \qquad \text{(C-5)}$$

= change in in-process inventory,

and

$$T1_n = C_n \ \Delta t \sum_{4(n-1)}^{4n} A_i \ F_i \quad . \qquad \text{(C-6)}$$

B. Materials Balance Variance

The variance of the n^{th} materials balance is

$$\sigma^2_{MB1_n} = \sigma^2_{I1_{n-1}} + \sigma^2_{I1_n} + \sigma^2_{T1_n} - 2\sigma_{I_{n-1,n}} - 2\sigma_{I_{n-1,n}T_n} \quad , \qquad \text{(C-7)}$$

where

$\sigma^2_{I1_n}$ = in-process inventory variance at n,

$\sigma^2_{T1_n}$ = n^{th} net transfer variance,

$\sigma_{I_{n-1,n}}$ = covariance between in-process inventories through the common concentration and common weight calibration, and

$\sigma_{I_{n-1,n}T_n}$ = covariance between in-process inventories and net transfer owing to the common concentration.

The variance $\sigma^2_{Il_n}$ is given by the expected value of the square of the deviation of the measured value from its expectation,

$$\sigma^2_{Il_n} = E\left\{\left[Il_n - E\left(Il_n\right)\right]^2\right\} \quad . \tag{C-8}$$

Substituting for the measured inventory and using the additive measurement-error model for the weight and the multiplicative measurement-error model for the concentration (see Sec. IV) yields

$$\sigma^2_{Il_n} = E\left\{\left[\left(C_n\ 1 + \varepsilon_c + \eta_c\right)\left(W_n + \varepsilon_w + \eta_w\right) - C_nW_n\right]^2\right\} \quad , \tag{C-9}$$

which reduces to

$$\begin{aligned}\sigma^2_{Il_n} = C^2_n \Big[& W^2_n\left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_c + \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_w \\ & + \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_c \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_w\Big] \quad ,\end{aligned} \tag{C-10}$$

where the measured value is substituted for the true value because the true value is unknown.

The second-order term, the last one in Eq. (C-10), contributes less than 1 part in 100 000 for values used in this analysis and can therefore be neglected. Thus, an estimate of the in-process inventory variance becomes

$$\sigma^2_{Il_n} = C^2_n\left[W^2_n\left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_c + \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_w\right] \quad , \tag{C-11}$$

which is identical to the form given in Sec. IV. This form is obtained by applying first-order propagation of errors and will be used for the remaining variance and covariance equations.

Following the form of Eq. (11), the total variance of the net transfer is estimated by

$$\sigma^2_{T1_n} = C_n^2 \, \Delta t^2 \left[\left(\sigma_\varepsilon^2 + \sigma_\eta^2\right)_c \left(\sum_{4(n-1)}^{4n} A_i \, F_i \right)^2 + \sigma^2_{\varepsilon_f} \sum_{4(n-1)}^{4n} A_i^2 \, F_i^2 + \sigma^2_{\eta_f} \sum_{4(n-1)}^{4n}\sum A_i \, A_j \, F_i \, F_j \right] \quad . \tag{C-12}$$

The covariance between in-process inventories is estimated by

$$\sigma_{I_{n-1,n}} = C_n^2 \left[W_n W_{n-1} \left(\sigma_\varepsilon^2 + \sigma_\eta^2\right)_c + \sigma^2_{\eta_w} \right] \quad . \tag{C-13}$$

The covariance between the net transfer and the in-process inventories is given by

$$\sigma_{I_{n-1,n} \, T_n} = C_n^2 \quad \Delta t \sum_{4(n-1)}^{4n} A_i F_i \left(W_{n-1} - W_n\right) \times \left(\sigma_\varepsilon^2 + \sigma_\eta^2\right)_c \quad . \tag{C-14}$$

Collecting terms, the materials balance variance becomes

$$\begin{aligned}\sigma^2_{MB1_n} = C_n^2 \Bigg\{ & \left[W_{n-1} - W_n - \Delta t \sum_{4(n-1)}^{4n} A_i \; F_i \right]^2 \left(\sigma_\varepsilon^2 + \sigma_\eta^2\right)_c \\ & + 2\sigma^2_{\varepsilon_w} + \Delta t^2 \left[\sigma^2_{\varepsilon_f} \sum_{4(n-1)}^{4n} A_i^2 \; F_i^2 \right. \\ & \left. + \sigma^2_{\eta_f} \sum\sum_{4(n-1)}^{4n} A_i A_j \; F_i F_j \right] \Bigg\} \; .\end{aligned} \tag{C-15}$$

Note that with similar measurement errors and no diversion

$$\begin{aligned}\sigma^2_{MB1_n} = C_n^2 \Bigg\{ & 2\sigma^2_{\varepsilon_w} + \Delta t^2 \left[\sigma^2_{\varepsilon_f} \sum_{4(n-1)}^{4n} A_i^2 \; F_i^2 \right. \\ & \left. + \sigma^2_{\eta_f} \sum\sum_{4(n-1)}^{4n} A_i A_j \; F_i F_j \right] \Bigg\} \; .\end{aligned} \tag{C-16}$$

C. Cusum

The cusum can extend over any sequence of materials balances. The cusum from the M^th^ to the N^th^ (N > M) materials balance is given by

$$CS1_N = C_N \left(W_M - W_N - \Delta t \sum_{M}^{N} A_i \; F_i \right) \; . \tag{C-17}$$

Note that the cusum equation and the materials balance equations [Eq. (C-3)] differ only in the indices.

D. Cusum Variance

The cusum variance is a linear combination of the variance of the initial in-process inventory, the final in-process inventory, and the net transfers, and the correlation between transfers, initial and final inventories, and inventories and transfers.

$$\sigma^2_{CS1_N} = \sigma^2_{I1_M} + \sigma^2_{I1_N} + \sigma^2_{T1_{N \to M}} - 2\sigma_{I1_{M,N}} + 2\sigma_{T1_{M,N}} - 2\sigma_{I1_{M,N}T1_{N \to M}} \quad . \tag{C-18}$$

Following the form of Eq. (C-15), the cusum variance is given by

$$\begin{aligned} \sigma^2_{CS1_N} = C_n^2 \Bigg\{ & \left[W_M - W_N - \Delta t \sum_{4(M-1)}^{4N} A_i \; F_i \right]^2 \left(\sigma_\varepsilon^2 + \sigma_\eta^2 \right)_c \\ & + 2\sigma^2_{\varepsilon_w} + \Delta t^2 \Bigg[\sigma^2_{\varepsilon_f} \sum_{4(M-1)}^{4N} A_i^2 \; F_i^2 \\ & + \sigma^2_{\eta_f} \sum\sum_{4(M-1)}^{4N} A_i A_j \; F_i F_j \Bigg] \Bigg\} \quad . \end{aligned} \tag{C-19}$$

Again, note that with similar measurement errors and no diversion

$$\sigma^2_{CS1_N} = C_n^2 \left\{ 2\sigma^2_{\varepsilon_w} + \Delta t^2 \left[\sigma^2_{\varepsilon_f} \sum_{4(M-1)}^{4N} A_i^2 F_i^2 \right.\right. \tag{C-20}$$

$$\left.\left. + \sigma^2_{\eta_f} \sum\sum_{4(M-1)}^{4N} A_i A_j F_i F_j \right] \right\} .$$

E. Volume vs Weight

If we substitute a volume measurement of the feed-blend tank contents for the weight measurements, the materials balance is given by

$$MB1_n = C_n \left[\rho \left(V_{n-1} - V_n \right) - \Delta t \sum_{4(n-1)}^{4n} A_i F_i \right] , \tag{C-21}$$

and the cusum is given by

$$CS1_N = C_N \left[\rho \left(V_M - V_N \right) - \Delta t \sum_{4(M-1)}^{4N} A_i F_i \right] , \tag{C-22}$$

where ρ is the solution density and V is the solution volume (all other terms, including units, retain the definitions given above). Equations (C-21) and (C-22) are for all practical purposes identical to Eqs. (C-3) and (C-17). However, the variance equations are not the same because a multiplicative measurement-error model is used for the volume measurements.

The materials balance variance using a volume measurement is given by

$$\begin{aligned}\sigma^2_{MB1_n} = C_n \Bigg\{ & \rho^2 \Bigg[\left(V_{n-1} - V_n\right)^2 \left(\sigma^2_{\varepsilon_c} + \sigma^2_{\eta_c} + \sigma^2_{\varepsilon_v} + \sigma^2_{\eta_v}\right) \\ & + 2V_{n-1} V_n \sigma^2_{\varepsilon_v} \Bigg] + \Delta t^2 \Bigg[\left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_c \left(\sum_{4(n-1)}^{4n} A_i F_i \right)^2 \\ & + \sigma^2_{\varepsilon_f} \sum_{4(n-1)}^{4n} A_i^2 F_i^2 + \sigma^2_{\eta_f} \sum\sum_{4(n-1)}^{4n} A_i A_j F_i F_j \Bigg] \\ & - 2\Delta t \rho \sum_{4(n-1)}^{4n} A_i F_i \left(V_{n-1} - V_n\right) \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_c \Bigg\} \ . \end{aligned} \qquad \text{(C-23)}$$

The cusum variance is given by Eq. (C-23) also, if the subscripts n - 1 and n are replaced by M - 1 and N, respectively.

III. UPAA TWO

The second UPAA contains the precipitator, the four parallel calciners, the primary filter, and the secondary filter.

A. Materials Balance

The transfer measurements from the first UPAA are combined with a measurement of the transfer container contents and the in-process inventory in all vessels to form a dynamic materials balance. The materials balance at time n is given by

$$MB2_n = I2_{n-1} - I2_n + T2_n \ . \qquad \text{(C-24)}$$

The in-process inventory at time n is given by

$$I2_n = IP_n + IC_n + IPF_n + ISF_n \quad , \qquad (C-25)$$

where IP = precipitator SNM in-process inventory,
IC = IC1 + IC2 + IC3 + IC4
= SNM in-process inventory in the four calciners,
IPF = primary filter in-process inventory, and
ISF = secondary filter in-process inventory.

Note that n is the time when the batch transfer container is removed from the primary filter.

The net materials transfer at n is given by

$$T2_n = T1_n - PPF_n - \delta_{PSF_n} PSF_n - \delta_{V2_n} V2_n \quad , \qquad (C-26)$$

where PPF = primary filter transfer container SNM content,
PSF = secondary filter transfer container SNM content,
V2 = vacuum system SNM content,

$$PSF_n = \begin{cases} 1 & \text{if the secondary filter transfer container is removed } (\sim \text{ once every 2 weeks}), \\ 0 & \text{otherwise, and} \end{cases}$$

$$V2_n = \begin{cases} 1 & \text{if vacuum system contents are measured,} \\ 0 & \text{otherwise.} \end{cases}$$

B. Materials Balance Variance

The materials balance variance is given by

$$\sigma^2_{MB2_n} = \sigma^2_{I2_{n-1}} + \sigma^2_{I2_n} + \sigma^2_{T2_n} - 2\sigma_{I2_{n-1,n}} \quad , \qquad (C-27)$$

where the in-process inventory variance is given by

$$\sigma^2_{I2_n} = IP^2_n \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_{ip} + IC^2_n \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_{ic}$$

$$+ IPF^2_n \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_{ipf} + ISF^2_n \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_{isf} \quad , \tag{C-28}$$

the transfer variance is given by

$$\sigma^2_{T2_n} = \sigma^2_{T1_n} + PPF^2_n \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_{ppf} + \delta_{PSF_n} \, PSF^2_n \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_{psf}$$

$$+ \delta_{V2_n} \, V2^2_n \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_{v} \quad , \tag{C-29}$$

and the covariance between in-process inventories is

$$\sigma_{I2_{n-1,n}} = IP_{n-1} \, IP_n \, \sigma^2_{\eta_{ip}} + IC_{n-1} \, IC_n \, \sigma^2_{\eta_{ic}}$$

$$+ IPF_{n-1} \, IPF_n \, \sigma^2_{\eta_{ipf}} + ISF_{n-1} \, ISF_n \, \sigma^2_{\eta_{isf}} \quad . \tag{C-30}$$

Combining terms and noting that the terms $(I_{n-1} - I_n)^2 \sigma^2_{\eta_I} \approx 0$, the materials balance variance becomes

$$\begin{aligned}
\sigma^2_{MB2_n} = {} & \left(IP^2_{n-1} + IP^2_n\right) \sigma^2_{\varepsilon_{ip}} + \left(IC^2_{n-1} + IC^2_n\right) \sigma^2_{\varepsilon_{ic}} \\
& + \left(IPF^2_{n-1} + IPF^2_n\right) \sigma^2_{\varepsilon_{ipf}} + \left(ISF^2_{n-1} + ISF^2_n\right) \sigma^2_{\varepsilon_{isf}} \\
& + \sigma^2_{T1_n} + PPF^2_n \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_{ppf} \qquad \text{(C-31)} \\
& + \delta_{PSF_n} \; PSF^2_n \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_{ppf} + \delta_{V2_n} \; V2^2_n \left(\sigma^2_\varepsilon + \sigma^2_\eta\right)_V \quad .
\end{aligned}$$

C. Cusum

The cusum of the M^th^ through the N^th^ materials balances is given by

$$\begin{aligned}
CS2_N = {} & IP_M + IC_M + IPF_M + ISF_M \\
& - IP_N - IC_N - IPF_N - ISF_N + C_N \, \Delta t \sum_{4(M-1)}^{4N} A_i \, F_i \qquad \text{(C-32)} \\
& - \sum_{N}^{N} \left(PPF_i + \delta_{PSF_i} \, PSF_i + \delta_{V2_i} \, V2_i \right) .
\end{aligned}$$

D. Cusum Variance

The corresponding cusum variance is given by

$$
\begin{aligned}
\sigma^2_{CS2_N} = {} & \left(IP_M^2 + IP_N^2\right)\sigma^2_{\varepsilon_{ip}} + \left(IC_M^2 + IC_N^2\right)\sigma^2_{\varepsilon_{ic}} \\
& + \left(IPF_M^2 + IPF_N^2\right)\sigma^2_{\varepsilon_{ipf}} + \left(ISF_M^2 + ISF_N^2\right)\sigma^2_{\varepsilon_{isf}} \qquad \text{(C-33)} \\
& + C_N^2\,\Delta t^2 \left[\sigma^2_{\varepsilon_f} \sum_{4(M-1)}^{4N} A_i^2\,F_i^2 + \sigma^2_{\eta_f} \sum\sum_{4(M-1)}^{4N} A_iA_j\,F_iF_j\right] \\
& + \sigma^2_{\varepsilon_{ppf}} \sum_M^N PPF_i^2 + \sigma^2_{\eta_{ppf}} \sum\sum_M^N PPF_i\,PPF_j \\
& + \sigma^2_{\varepsilon_{psf}} \sum_M^N PSF_i^2 + \sigma^2_{\eta_{psf}} \sum\sum_M^N PSF_i\,PSF_j \\
& + \sigma^2_{\varepsilon_v} \sum_M^N \delta_{V2_i}\,V2_i^2 + \sigma^2_{\eta_v} \sum\sum_M^N \delta_{V2_{ij}}\,V_i\,V_j \;.
\end{aligned}
$$

Note that the double sums give the correlation between materials transfers and are reinitialized every time an instrument is recalibrated.

IV. UPAA THREE

The third UPAA consists of four parallel and identical UPAAs each drawn about a reduction-stabilization station and the screening station. The four areas overlap at the screening station.

A. Materials Balance

A materials balance is drawn about each batch of material by combining the measurement of the transfer container from the second UPAA with measurements of the product canister and the holdup in each vessel. The materials balance after the n^{th} batch is processed is given by

$$MB3_n = HU3_{n-1} - HU3_n + T3_n \quad , \tag{C-34}$$

where

$HU3_n$ = $HURS_n + HUFF_n$

= reduction-stabilization and final filter holdups,

$T3_n$ = $PPF_{n-1} - P_n - \delta_{S_n} S_n - \delta_{V_{3n}} V3_n$,

P_n = storage container plutonium content,

$\delta_{S_n} = \begin{cases} 1 & \text{if scrap is measured,} \\ 0 & \text{otherwise,} \end{cases}$

S_n = scrap plutonium content,

$\delta_{V3_n} = \begin{cases} 1 & \text{if vacuum is measured,} \\ 0 & \text{otherwise, and} \end{cases}$

$V3_n$ = vacuum system plutonium content.

B. Materials Balance Variance

The corresponding materials balance variance is given by

$$\sigma^2_{MB3_n} = \left(HURS^2_{n-1} + HURS^2_n \right) \sigma^2_{\varepsilon_{hurs}}$$

$$+ \left(\mathrm{HUFF}_{n-1}^2 + \mathrm{HUFF}_n^2\right) \sigma^2_{\varepsilon_{\mathrm{huff}}}$$

$$+ \mathrm{PPF}_{n-1}^2 \left(\sigma_\varepsilon^2 + \sigma_\eta^2\right)_{\mathrm{ppf}} + P_n^2 \left(\sigma_\varepsilon^2 + \sigma_\eta^2\right)_p \tag{C-35}$$

$$+ \delta_{S_n} S_n^2 \left(\sigma_\varepsilon^2 + \sigma_\eta^2\right)_s + \delta_{V3_n} V3_n^2 \left(\sigma_\varepsilon^2 + \sigma_\eta^2\right)_v \quad .$$

C. Cusum

The cusum of the M^th^ through N^th^ materials balances is given by

$$\mathrm{CS3}_N = \mathrm{HURS}_M + \mathrm{HUFF}_M - \mathrm{HURS}_N - \mathrm{HUFF}_N$$

$$+ \sum_M^N \left(\mathrm{PPF}_{i-1} - P_i - \delta_{S_i} S_i - \delta_{V3_i} V3_i\right) \quad . \tag{C-36}$$

D. Cusum Variance

The corresponding cusum variance is given by

$$\sigma^2_{\mathrm{CS3}_N} = \left(\mathrm{HURS}_M^2 + \mathrm{HURS}_N^2\right) \sigma^2_{\varepsilon_{\mathrm{hurs}}} + \left(\mathrm{HUFF}_M^2 + \mathrm{HUFF}_N^2\right) \sigma^2_{\varepsilon_{\mathrm{huff}}}$$

$$+ \sum_M^N \Bigg(\mathrm{PPF}_{i-1}^2 \, \sigma^2_{\varepsilon_{\mathrm{ppf}}} + P_i^2 \, \sigma^2_{\varepsilon_p}$$

$$+ \delta_{S_i} S_i^2 \, \sigma^2_{\varepsilon_s} + \delta_{V3_i} V3_i^2 \, \sigma^2_{\varepsilon_v} \Bigg) \tag{C-37}$$

$$+ \sum_{M}^{N} \sum \left(PPF_{i-1} \, PPF_{j-1} \, \sigma^2_{\eta_{ppf}} + P_i \, P_j \, \sigma^2_{\eta_p} \right.$$

$$\left. + \delta_{S_{ij}} \, S_i \, S_j \, \sigma^2_{\eta_s} + \delta_{V_{ij}} \, V_i \, V_j \, \sigma^2_{\eta_v} \right) ,$$

where

$$\left.\begin{matrix} \delta_{S_{ij}} \\ \\ \delta_{V_{ij}} \end{matrix}\right\} = \begin{cases} 1 & \text{if i and j are within the same calibration period,} \\ \\ 0 & \text{otherwise.} \end{cases}$$

APPENDIX D

MEASUREMENT METHODOLOGY

Both conventional chemical and NDA techniques are required to implement a near-real-time measurement control and SNM accountability system for the Coprecal facility. The analysis scheme must consider such factors as standards, sampling, analysis time, and applicabiity to the measurements required. The applications of conventional analytical and NDA methods to measurements of uranium and plutonium have been treated extensively in previous LASL reports,[1-5] including that for a plutonium nitrate-to-oxide conversion facility.[3] Measurement methods applicable to determining plutonium in a plutonium conversion process generally are applicable to a coconversion facility, possibly with some loss in precision for methods such as absorption-edge densitometry where plutonium must be measured in the presence of a 10-fold excess of uranium. The techniques appropriate to the Coprecal process are reviewed here.

I. STANDARDS

All procedures and instruments used for nuclear safeguards materials accounting should be calibrated against approved standard reference materials (SRMs). All weight and volume measurements should be traceable to NBS-certified weight standards. NBS-certified Class S weights are used as reference standards for laboratory measurements. Volumes are calibrated using appropriate NBS-certified weights as reference standards.

The primary standards available for use as oxidation-reduction standards in the nuclear safeguards accountability program are SRM 136c (potassium dichromate), SRM 83c (arsenic trioxide), and SRM 40b (sodium oxalate). SRMs available for uranium and plutonium analyses are shown in Ref. 6, p. 42. In addition to the NBS standards, standards for the spectrographic determination of impurities in uranium are obtainable from the New Brunswick Laboratory and the Oak Ridge National Laboratory.[7]

Secondary or bench standards may be working standards obtained from a source such as NBS, from various DOE contractors, or from international sources.[8] Alternatively, they may be prepared from process solutions by characterization against NBS SRMs. The preparation and evaluation of secondary plutonium nitrate standards have been described[9] and should be incorporated into the analytical laboratory standard operating procedure. The same techniques are applicable to the preparation of uranium working standards from uranium process materials. These standards should be analyzed daily or by each shift to insure that the method is under control. Process samples should not be analyzed until satisfactory values are obtained on bench standards.

Primary standards for chemical analysis also can be used for NDA applications, but generally must be incorporated into a matrix to simulate process samples. This can introduce errors, and these secondary standards must be evaluated for homogeneity, etc. The New Brunswick Laboratory has initiated a program to develop low- and medium-density NDA standards for uranium analyses.[10] The same technique can be used for plutonium reference materials.

II. WEIGHT AND VOLUME MEASUREMENTS

A. Weight Measurements

Load-cell weighing systems are particularly well suited for measuring bulk quantities of material in nuclear facilities. The weighing platform can be separated physically from the associated electronics and read-out mechanism. Only the platform need be exposed to the environment of a glovebox or process area, and the electronics can be located elsewhere to provide ready access for calibration and maintenance.

Load cells having 10-, 100-, and 1000-kg capacities are available commercially, and they should be large enough for all anticipated applications in the conversion facility. These units have the following measurement errors (per cent of full scale) (see Ref. 1, p. C-23):

Zero balance	1%
Hysteresis	0.02-0.05%
Repeatability	0.01-0.03%
Linearity	0.05-0.25%

Determination by weighing the mass of solution in large process tanks and vessels would be the most direct method of obtaining this information, but successful use of load cells for such measurements has not yet been demonstrated. Idaho Chemical Processing Plant (ICPP) has evaluated the in-plant performance of a load-cell weighing system on an existing input accountability tank.[11] Because of large thermally induced forces in the tank and attached piping, it was impossible to attain the measurement accuracy for which the weighing system was designed. The study showed that to obtain useful accuracy, tanks installed on weighing systems must be specially designed for the application.

B. Volume Measurements

Materials accountability in the nitrate-to-oxide coconversion facility may require frequent measurements of the plutonium content of feed-blend tanks. We anticipate that these measurements will be based on measurement of the liquid volume in the tank combined with liquid-density and plutonium-concentration measurements to yield the required inventory. Volume-measurement technology for large process vessels has been reviewed recently.[12] The results are as follows.

Volume determinations usually are inferred from the measured liquid level in a calibrated tank. At present, the best developed liquid-level measuring system for use in nuclear facilities is the dip-tube manometer, or pneumatic bubbler. When combined with a commercially available electromanometer, such systems can have a detection sensitivity of 0.25 mm of water. Furthermore, the instrument output is directly computer-compatible. With careful calibration and an effective measurement-control program, calibration errors approaching 0.1% RSD and single volume-measurement errors of 0.5% or less should be attainable.

III. FLOW MEASUREMENT

Flow measurement instruments in nuclear fuel cycle plants are used principally for process control, where high precision is not a major requirement.[13] However, the dynamic materials-measurement system for safeguards accountability in the Coprecal process requires accurate and precise monitoring of flow as well as concentration at the feed-blend tank.

The most accurate means of measuring flow is to follow the progress of batch transfers. Batch volumes can be measured to high accuracies (∿0.025 cm or 0.1% of scale, depending upon the system). Measurement of the rate of change in tank inventory can give a highly accurate indication of flow rate and is the best technique available. Where materials accounting is handled by batch accumulation and analysis, this method is the most convenient and accurate for handling the accountability and can be used for the filling of the feed-blend tank. Continuous rather than batch transfer is used to feed the precipitator, and other means of flow measurement are required.

A bubble flowmeter is under evaluation at the ICPP for measurement of low flow rates, such as may be encountered in product concentrator lines.[14] This flowmeter measures the transit time of a small bubble injected into the flowing stream. Ultrasonic detectors mounted on the outside of the line detect the passage of the injected bubbles.

Magnetic flowmeters have been proposed for flow measurement in the EXXON fuel reprocessing plant.[15] They can measure flow rates to ∿1% accuracy.

The ultrasonic flowmeter[16-19] can measure flow with better than 1% accuracy. Transducers can be mounted either in the pipe or exterior to the pipe wall so that intrusion into the pipe is not necessary. Pipe diameter is a limiting factor: diameters >4 cm (1.5 in) are generally recommended. Interference by entrained air can be eliminated by transducer and detector-circuit design.[19] The use of ultrasonic flowmeters and extrinsic factors affecting measurement accuracy have been reviewed by Managan.[20]

IV. CHEMICAL ANALYSIS

Several chemical analysis methods can provide better than 0.1% (1 σ) precision with high accuracy. High sensitivity (<1 mg) can be attained. Methods have been developed and evaluated using natural uranium or weapons-grade plutonium and should be re-evaluated for high-burnup, reactor-grade materials.

A. Electrometric Titrations

Amperometry, potentiometry, and coulometry have been applied to the determination of uranium and plutonium with better than 0.1% RSDs and probably provide the most accurate and precise methods for determining these elements in high-purity materials.[21-40] The attainment of similar precision with reactor-grade samples must be demonstrated.

Electrometric methods for high-precision measurements of uranium (Ref. 22, pp. 74-77, 77-83, 86-88; Ref. 23) and plutonium have been summarized (Ref. 2, App. F; Ref. 21). The electrometric methods for determining uranium rely on the reaction $U^{4+} \rightarrow U^{6+}$ after quantitative reduction of the uranium to the tetravalent state.[22-27] Differences between methods result from the reductants and oxidants used. Reducing agents used to obtain U^{4+} include zinc amalgam, the Jones reductor, Ti^{3+}, the lead reductor, Fe^{2+} in phosphoric acid, and electrical reduction. The Jones reductor can generate a mixture of U^{3+} and U^{4+}, and the trivalent uranium must be air-oxidized to the tetravalent state before titration. The reduction with an excess of ferrous ion in concentrated phosphoric acid followed by titration with dichromate can be performed in the presence of Pu^{4+} without interference. Some of the standard electrometric techniques for determination of uranium are summarized in Ref. 21, Table XII. Of these methods for determining uranium, the Davies-Gray method is the most versatile and has been automated for rapid analysis of process samples.

For the electrometric determination of plutonium, the plutonium may be oxidized quantitatively to Pu^{6+}, then titrated to Pu^{4+} (Ref. 22, pp. 254-261, 261-268, 299-306; Refs. 28-34). This couple generally is preferred if uranium or iron is present. Oxidants for the first step include AgO and $HClO_4$. If AgO is used, the excess is destroyed by gentle heating. The oxidation capability of $HClO_4$ is destroyed by diluting the sample after the plutonium has been oxidized. Errors can be introduced into the plutonium measurement if conditions for the dilution are not followed properly. The reduction to Pu^{4+} usually is carried out with Fe^{2+}, and can be performed directly or, by using a potentiometric end point, can be sharpened by adding a measured excess of Fe^{2+} with the excess titrated with Ce^{4+}.

Alternatively, plutonium can be determined by quantitative reduction to Pu^{3+} and subsequent titration to Pu^{4+} (Ref. 22, pp. 274-285, 285-292, 268-274, 293-299, 385-388; Ref. 29, pp. 209-212; Refs. 35-40). Common reducing agents are liquid zinc amalgam, the Jones reductor, the lead reductor, Ti^{3+}, Cr^{2+}, and electrical methods. The liquid-zinc-amalgam method suffers in that an extra transfer is required to remove the amalgam before titration. Small amounts of titratable material can be leached from the Jones reductor. Fewer elements are reduced with $TiCl_3$ than with the Jones or lead reductors and, therefore, there may be fewer potential interferences present.[36] However, Ti^{3+} is destroyed readily by contact with air.

The oxidation titration to Pu^{4+} usually is performed electrolytically or with Ce^{4+} or $K_2Cr_2O_7$. Dichromate has the advantage of being available as a primary standard and it is more stable than Ce^{4+} over extended storage periods. Uranium and iron interfere with methods involving the Pu^{3+}-Pu^{4+} couple. An exception is the controlled-potential coulometric technique, which in the presence of uranium can be used to measure successively both elements in the same sample.

B. Mass Spectrometry

The isotopic compositions for uranium and plutonium in the feed-blend tank can be measured using thermal-ionization mass spectrometry. Isotope-dilution mass spectrometry also can be used to measure the uranium and plutonium concentrations of the tank.

For determination of plutonium, either ^{242}Pu or ^{244}Pu can be used as the spike, with the latter preferred if available, because it is not produced in significant quantities in the reactor. Uranium-233 is used as the spike for analyzing uranium LWR fuels.

For determination of major isotopes, RSDs of 0.01-0.02% (1 σ) can be attained. The precision for well-characterized materials such as NBS SRMs generally is significantly better than for process and product samples, probably reflecting problems in sampling and sample preparation. Overall precision for measuring dissolver samples has been 0.3-1%, but may be improved to 0.1-0.2% with more stringent quality control.[41]

V. NDA

NDA techniques are either active or passive. Active assay involves irradiation with neutrons or photons to induce nuclear or electron interactions in the sample. The resulting neutron, gamma-ray, or x-ray "signatures" are interpreted to determine quantitatively the amount of the designated element present.

Passive assay uses naturally occurring gamma-ray and neutron radiations as direct elemental or isotopic signatures.

A. Neutron Techniques

Application of neutron techniques to plutonium assay has been discussed elsewhere (for example, see Ref. 42), so only details important to measurements in a coconversion facility are reviewed here. Neutron yields from uranium are too low to enable uranium determination in the presence of plutonium.

Neutrons from plutonium samples arise from either (α,n) reactions or fission. The (α,n) neutrons result from reactions of alpha particles from the radioactive decay of plutonium with light elements in the matrix material. The neutron yield is a function of alpha-particle energy, the (α,n) cross sections of the matrix elements, and the matrix configuration. Furthermore, the alpha-particle intensity depends on the isotopic composition and is particularly sensitive to the ^{238}Pu content. As a consequence, (α,n) neutrons ordinarily are not useful as a quantitative signature for plutonium, and, in fact, they usually constitute a large background that must be eliminated from most measurements.

Fission neutrons result from spontaneous fission or from fissions induced by an external neutron source such as ^{252}Cf. Because the fission process produces more than one neutron per fission, fission neutrons can be differentiated from (α,n) neutrons by coincidence counting. Indeed, coincidence counting of fission neutrons will be a requisite feature of any neutron-based NDA system intended for quantitative assay of plutonium in the coconversion facility. Only the even isotopes of plutonium, ^{238}Pu, ^{240}Pu, and ^{242}Pu, have high enough spontaneous-fission rates to contribute to a passive neutron measurement. Hence, the isotopic composition must be known to infer total plutonium content.

Coincidence counting of ^{240}Pu and ^{242}Pu spontaneous-fission neutrons is accomplished with assemblies of ^{3}He (or BF_3) counters embedded in polyethylene moderators.[43-47] Table D-I lists estimates of the isotopic composition and the neutron yields from spontaneous fission and (α,n) reactions of PuO_2 recycle fuel.[48] High (α,n) rates can complicate the coincidence method through accidental (random) coincidences and by fissions induced in the fissile components of the material, that is, the multiplication effect.

If coincidence counting is to be used to analyze large amounts of PuO_2 or scrap containing light elements that have high (α,n) yields, such as boron and fluorine, coincidence detectors with short die-away times are required to enhance the contribution of real coincidence events relative to accidental events, thereby reducing statistical uncertainties. This requirement has led to the concept of varying the physical die-away time of a neutron coincidence detector by changing the number and geometrical configuration of polyethylene moderators and cadmium (or boron) neutron absorbers.[49]

Table D-II shows typical precisions and accuracies of neutron coincidence counters for the general categories of product, scrap, and waste[50] in the process environment. The data demonstrate the loss of precision and accuracy possible when the elemental and isotopic material composition is poorly characterized or nonuniform.

In the coconversion facility, neutron NDA methods will be used to measure $(U,Pu)O_2$ product and in-process inventories.

B. Gamma-Ray Techniques

Only the passive gamma-ray techniques appear to be appropriate for measuring uranium and plutonium in the coconversion facility. Use of passive gamma-ray spectroscopy in assaying plutonium-bearing solutions is evaluated in Refs. 51-53. The useful gamma-ray signatures from the plutonium isotopes and ^{241}Am are summarized in Table D-III. The ^{242}Pu isotope does not have a useful gamma ray, so it cannot be measured by this method.

Sodium iodide, lithium-drifted germanium, Ge(Li), and intrinsic germanium detectors generally are used for gamma-ray measurements. The Ge(Li) and intrinsic germanium detectors provide much better energy resolution for adjacent peaks, but they

TABLE D-I

EMISSION RATE FROM 1 g OF PLUTONIUM[a]

Isotope	Wt%	Spontaneous Fission (sf/s)	Spontaneous Fission (%)	Spontaneous Fission Prompt Neutrons (n/s)	Spontaneous Fission Prompt Neutrons (%)	(α,η) Neutrons from PuO_2 (n/s)	(α,η) Neutrons from PuO_2 (%)
^{238}Pu	1.9	20.9	14.8	47.2	15.3	266.0	76.1
^{239}Pu	63.0	0	0	0	0	28.4	8.1
^{240}Pu	19.0	89.5	63.6	194.2	63.2	32.3	9.2
^{241}Pu	12.0	0	0	0	0	1.2	0.3
^{242}Pu	3.8	30.4	21.6	65.7	21.4	0.4	0.1
^{241}Am	0.6	0	0	0	0	21.4	6.1
Total		140.8	100.0	307.1	100.0	349.7	100.0

---

[a] LWR fuel with 35 000-MWD/MTU burnup.

TABLE D-II

TYPICAL NEUTRON COINCIDENCE COUNTER UNCERTAINTIES

Material Category	Precision (%,1 σ)	Accuracy (%,1 σ)
Feed and product	1	1
Scrap	2-8	2
Waste	10-15	5-10

require liquid N_2 cooling. Nevertheless, one or the other is required if good quantitative data on isotopic abundances are to be obtained. For rugged on-line instruments, the NaI detector is preferred if overlapping spectra are not a problem. More recently, CdTe detectors, which have an energy resolution of 8% at 122 keV and are usable at room temperature, have been investigated as a substitute for NaI.[54] This detector shows promise, but further development is required to match detectors and preamplifiers and to reduce charge-collection problems.

Gamma-ray spectroscopy can be used to measure the relative isotopic abundances of the plutonium isotopes and to measure total plutonium concentration. The relative isotopic ratios are required for interpretation of neutron coincidence measurements of spontaneous fission of ^{240}Pu and ^{242}Pu. Relative isotopic measurements generally are obtained by using gamma-ray energies in a narrow energy region to minimize self-absorption effects and by applying peak-stripping and background subtraction corrections with a computer. The relative precision of this measurement can be better than 0.5%. Total plutonium concentrations between 0.1 and 400 g/L can be determined in a 30-min counting period with a better than 1% RSD and a 0.5% accuracy by measuring the ^{239}Pu isotope.[53]

The solid low-level waste from the coconversion plant may be packaged in 55-gal drums for disposal and should be analyzed for plutonium. A drum scanner that uses an NaI detector to measure the 414-keV gamma ray from ^{239}Pu can detect as little as 1 g of ^{239}Pu in a 5-min scan.[55] The relative accuracy for measuring >10 g of ^{239}Pu can be as good as 10% in matrices of low atomic number and ranges to 50% in unknown matrices.

TABLE D-III

MAJOR GAMMA-RAY SIGNATURES OF PLUTONIUM AND AMERICIUM

Isotope	Energy (keV)	Intensity (g-s)$^{-1}$	Comments
^{238}Pu	766.40	1.5×10^5	Most useful for quantitative assay. Ge(Li) or NaI. Useful for isotopic determinations with Ge(Li).
	152.77	6.5×10^6	
^{239}Pu	413.69	3.4×10^4	The 413.69 usually provides the basis for Ge(Li) assays. The 413.69 plus the 375.02 and its weak neighbors form a complex upon which NaI assays are based.
^{239}Pu	129.28	1.4×10^5	Useful for isotopic determinations with Ge(Li). Plutonium-239 has over 100 gamma rays, some of which are useful for careful work with Ge(Li).
^{240}Pu	--	--	Several weak gamma rays, but all suffer bad interference from gammas of other isotopes. Requires very careful work with high-resolution detector to make use of any of them.
^{241}Pu	207.98	2.0×10^7	Actually from ^{237}U daughter and requires about 25 days after chemical separation to come into equilibrium at stated value. May also have a few per cent interference from ^{241}Am, which emits same gamma. Nevertheless a good clean strong gamma useful with both NaI and Ge(Li).
	164.59	1.8×10^6	Useful with Ge(Li). Also from ^{237}U.
	148.60	7.5×10^6	Useful with Ge(Li). Direct from ^{241}Pu.
^{241}Am	59.54	4.6×10^{10}	Very strong gamma but attenuation problems. Useful with Ge(Li) or NaI. Has several other much less intense gammas sometimes useful for Ge(Li) work.
^{242}Pu	--	--	No useful gamma rays at all. Nature failed us at this point.

Hence, the measurement accuracy will depend largely on the administrative control exercised in sorting waste. This instrument is in advanced development and requires only additional field testing and evaluation.

C. X-Ray Techniques

Two active techniques, x-ray emission and x-ray absorption, are applicable to uranium and plutonium assay.[53] X-ray emission can be used to measure uranium and plutonium concentrations of one to several grams per liter with a 1% RSD. The K-, L-, and M-series x-ray lines have been used for this analysis, but the L-series lines generally are preferred for solution assay. Wide variations in matrix composition may require an internal standard such as yttrium or thorium. The procedure is rapid and amenable to automation;[56,57] however, it is not an on-line method, and samples must be removed from the process stream.

Absorption-edge densitometry[53] is an element-specific analytical method that can be applied in-line or on-line to most measurement situations amenable to gross-absorption techniques. With proper choice of cell path length and K or L_{III}-absorption edges, plutonium (or uranium) concentrations between ∿5 and 400 g/L can be measured with better than 1% RSD. For example, plutonium concentrations of 10-25 g/L were measured using the L_{III} edge with a 0.6% RSD,[58] and concentrations of 140-400 g/L were measured using the K edge with a 0.3% RSD.[59] For in-line safeguards accounting measurements, absorption-edge densitometry, rather than gross absorptiometry, should be used.

Absorption-edge densitometry offers an added advantage in that if both uranium and plutonium are present, as they would be in coprocessing, both can be measured simultaneously.

REFERENCES

1. J. P. Shipley, D. D. Cobb, R. J. Dietz, M. L. Evans, E. P. Schelonka, D. B. Smith, and R. B. Walton, "Coordinated Safe guards for Materials Management in a Mixed-Oxide Fuel Facility," Los Alamos Scientific Laboratory report LA-6536 (February 1977).

2. E. A. Hakkila, D. D. Cobb, H. A. Dayem, R. J. Dietz, E. A. Kern, E. P. Schelonka, J. P. Shipley, D. B. Smith, R. H. Augustson, and J. W. Barnes, "Coordinated Safeguards for Materials Management in a Fuel Reprocessing Plant," Los Alamos Scientific Laboratory report LA-6881 (September 1977).

3. H. A. Dayem, D. D. Cobb, R. J. Dietz, E. A. Hakkila, E. A. Kern, J. P. Shipley, D. B. Smith, and D. F. Bowersox, "Coordinated Safeguards for Materials Management in a Nitrate-to-Oxide Conversion Facility," Los Alamos Scientific Laboratory report LA-7011 (April 1978).

4. E. A. Hakkila, "A Critical Review of Analytical Techniques for Safeguarding the Thorium-Uranium Fuel Cycle," Los Alamos Scientific Laboratory report LA-7372 (October 1978).

5. E. A. Hakkila, J. W. Barnes, H. A. Dayem, R. J. Dietz, and J. P. Shipley, "Preliminary Concepts: Coordinated Safeguards for Materials Management in a Thorium-Uranium Fuel Reprocessing Plant," Los Alamos Scientific Laboratory report LA-7411-MS (October 1978).

6. National Bureau of Standards, "Catalog of NBS Standard Reference Materials," NBS Special Publ. 260 (1975).

7. F. B. Stephens, R. G. Gutmacher, K. Ernst, J. E. Harrar, and S. P. Turel, "Methods for the Accountability of Uranium Dioxide," US Nuclear Regulatory Commission report NUREG-75/010 (June 1975), p. 5-17.

8. K. F. Lauer and Y. LeDuigou, "Proper Use of Reference Materials for the Accurate Determination of Uranium, Plutonium, and Thorium in the Nuclear Fuel Cycle," in Analytical Methods in the Nuclear Fuel Cycle, Proc. Symp., Vienna, November 29-December 3, 1971 (International Atomic Energy Agency, Vienna, 1972), pp. 145-152.

9. J. E. Rein, S. F. Marsh, G. C. Swanson, G. R. Waterbury, G. L. Tietjen, and R. K. Ziegler, "Preparation of Working Calibration and Test Materials: Plutonium Nitrate Solution," Los Alamos Scientific Laboratory and US Nuclear Regulatory Commission report LA-NUREG-6348 (January 1977).

10. A. M. Voeks, M. M. Trahey, and J. M. Scarborough, "Preparation of Test Materials for an Interlaboratory Comparison Program on NDA Physical Standards," in Analytical Chemistry in Nuclear Fuel Reprocessing, W. S. Lyon, Ed., (Science Press, Princeton, 1978) pp. 338-344.

11. F. M. Groth and F. O. Cartan, "Evaluation of Instrumentation for Nuclear Fuels Reprocessing Plant Input Weight Measurements," Allied Chemical Corp. report ICP-1014 (July 1972).

12. D. B. Smith, "A Critical Review of Volume Measurement Technology," in "Coordinated Safeguards for Materials Management in a Fuel Reprocessing Plant, Vol. II," Los Alamos Scientific Laboratory report LA-6881 (September 1977), App. C.

13. J. W. Barnes, "Flow Measurement Technology," in "Coordinated Safeguards for Materials Management in a Fuel Reprocessing Plant, Vol. II," Los Alamos Scientific Laboratory report LA-6881 (September 1977), App. B.

14. H. R. Deveraux and W. J. Harris, "Transit Time Flowmeter," in "Technical Division Quarterly Progress Report October 1-December 31, 1976," C. M. Slansky, Ed., Allied Chemical Corporation report ICP-1111 (March 1977).

15. "Nuclear Fuel Recovery and Recycling Center, Preliminary Safety Analysis Report," Docket 50-564, EXXON Nuclear Company.

16. J. L. McShane, "Ultrasonic Flowmeters," in Flow, Its Measurement and Control in Science and Industry, R. B. Dowdell, Ed.-in-Chief (Instrument Society of America, Pittsburgh, 1974) Vol. I, Part 2, pp. 897-915.

17. L. C. Lynnworth, N. E. Pedersen, E. P. Papadakis, and J. H. Bradshaw, "Nonintrusive Ultrasonic Measurement of Flow Velocity and Mass Flow Rate," in Flow, Its Measurement and Control in Science and Industry, R. B. Dowdell, Ed.-in-Chief (Instrument Society of America, Pittsburgh, 1974) Vol. I, Part 2, pp. 917-924.

18. W. H. Lynch and A. E. Brown, "Flow Measurement with a New Ultrasonic Flowmeter," in Flow, Its Measurement and Control in Science and Industry, R. B. Dowdell, Ed.-in-Chief (Instrument Society of America, Pittsburgh, 1974) Vol. I, Part 2, pp. 925-931.

19. Envirotech, Sparling Division, Series 500 Flowmeters, Technical Data Sheet (1975).

20. W. W. Managan, "Extrinsic Factors Affecting Accuracy of Ultrasonic Flowmeters for LMFBRs," Argonne National Laboratory report ANL-CT-76-40 (1976).

21. G. Phillips and D. Crossley, "The Determination of Plutonium/ Uranium Ratios by Controlled-Potential Coulometry at a Solid Electrode," United Kingdom Atomic Energy Authority report AERE-R-8794 (May 1977).

22. C. J. Rodden, Ed., Selected Measurement Methods for Plutonium and Uranium in the Nuclear Fuel Cycle (Office of Information Services, US Atomic Energy Commission, Virginia, 1972), pp. 70-74.

23. W. Davies and W. Gray, "A Rapid and Specific Titrimetric Method for the Precise Determination of Uranium Using Iron (II) Sulphate as Reductant," Talanta 11, 1203-1211 (1964).

24. A. R. Eberle, M. W. Lerner, C. G. Goldbeck, and C. J. Rodden, "Titrimetric Determination of Uranium in Product, Fuel, and Scrap Materials After Ferrous Ion Reduction in Phosphoric Acid," New Brunswick Laboratory report NBL-252 (July 1970).

25. G. C. Goode, J. Herrington, and G. Hall, "The Determination of Uranium in the Presence of Plutonium by Controlled Potential Coulometry, Application to the Analysis of Fuel Element Processing Solutions," Anal. Chim. Acta 30, 109-113 (1964).

26. J. T. Byrne, M. K. Larsen, and J. L. Pflug, "Accurate Determination of Uranium in the Presence of Small Amounts of Molybdenum," Anal. Chem. 31, 942-945 (1959).

27. J. E. Harrar, W. G. Boyle, J. D. Breshears, C. L. Pomernacki, H. R. Brand, A. M. Kray, R. J. Sherry, and J. A. Pastrone, "An Automated Sample-Processing and Titration System for the Determination of Uranium," Nucl. Mater. Manage. 5, 199-210 (1976).

28. W. D. Shults, "Applications of Controlled-Potential Coulometry to the Determination of Plutonium," Talanta 10, 833-849 (1963).

29. American Society for Testing Materials, 1974 Annual Book of ASTM Standards, Part 45, Nuclear Standards (ASTM, Philadelphia, 1974), pp. 709-714.

30. C. A. Seils, R. J. Meyer, and R. P. Larsen, "Amperometric Titration of Plutonium (VI) with Iron (II)," Anal. Chem. 35, 1673-1675 (1963).

31. K. S. Bergstresser and G. R. Waterbury, "An Evaluation of the Amperometric Titration of Plutonium," Los Alamos Scientific Laboratory report LA-3091 (May 1964).

32. J. L. Drummond and R. A. Grant, "Potentiometric Determination of Plutonium by Argentic Oxidation, Ferrous Reduction, and Dichromate Titration," Talanta 13, 477-488 (1966).

33. G. R. Waterbury and C. F. Metz, "Precise Determination of Plutonium by Potentiometric Titration," Anal. Chem. 31, 1144-1148 (1959).

34. W. N. Carson, Jr., J. W. Vanderwater, and H. S. Gile, "Coulometric Determination of Plutonium," Anal. Chem. 29, 1417-1422 (1957).

35. C. E. Pietri and J. A. Baglio, "The Determination of Plutonium Based on National Bureau of Standards Potassium Dichromate," Talanta 6, 159-166 (1960).

36. R. J. Kofoed, Jr., "Plutonium Oxidation-Reduction Titrations," Hanford Atomic Products Operation report HW-53368 Suppl. (December 1963).

37. Technical Manager Chemical Services, "The Titrimetric Determination of Plutonium, Chromous Reduction-Ceric Sulphate Titration," United Kingdom Atomic Energy Authority report PG309(w) (1962).

38. D. D. Jackson, R. M. Hollen, F. R. Roensch, and J. E. Rein, "Highly Selective Coulometric Method and Equipment for the Automated Determination of Plutonium," in Analytical Chemistry in Nuclear Fuel Reprocessing, W. S. Lyon, Ed., (Science Press, Princeton, 1978) pp. 151-158.

39. G. S. Barney, "Coulometric Determination of Plutonium in Solutions of Dissolved Reactor Fuel Elements," Atlantic Richfield Hanford Co. report ARH-SA-55 (January 1970).

40. F. A. Scott and R. M. Peekema, "Analysis for Plutonium by Controlled-Potential Coulometry," in Proc. U.N. International Conf. on Peaceful Uses of Atomic Energy, 2nd, Geneva (United Nations, New York, 1958), Vol. 28, 573-578.

41. R. E. Perrin, Los Alamos Scientific Laboratory, personal communication, January 1977.

42. R. H. Augustson and T. D. Reilly, "Fundamentals of Passive Nondestructive Assay of Fissionable Material," Los Alamos Scientific Laboratory report LA-5651-M (September 1974).

43. R. Berg, R. Swennen, G. Birkhoff, L. Bondar, J. Ley, and G. Busca, "Determination of the Pu-240 in Solid Waste Containers by Spontaneous Fission Neutron Measurements. Application to Reprocessing Plant Waste," EUR 5158e (1974).

44. K. Bohnel, "Die Plutoniumbestimmung in Kernbrennstoffen mit der Neutronenkoinzenmethode," Kernforschungszentrum Karlsruhe report KFK 2203 (1975).

45. K. P. Lambert and J. W. Leake, "A Comparison of the Ispra and Harwell V.D.C. Systems for the Assay of Pu-240 by Spontaneous Fission Neutron Measurements," United Kingdom Atomic Energy Authority report AERE-R 8300 (April 1976).

46. M. M. Stephens, L. V. East, T. L. Atwell, and T. D. Reilly, "Automated Neutron Assay for Richland Operations Office," in Los Alamos Scientific Laboratory report LA-6142-PR (May-August 1975), p. 13.

47. J. E. Foley, "Neutron Coincidence Counters for Nuclear Safeguards Applications," IEEE Trans. Nucl. Sci. NS-19, No. 3, p. 453 (1972).

48. E. V. Weinstock, "The LMFBR Fuel Cycle: Part I, Description and Schedules," report prepared by BNL-TSO for the Division of Nuclear Materials Security of the AEC (November 15, 1973), p. 39.

49. J. E. Foley, "Analytical Study on Design of Neutron Coincidence Counters," in "Nuclear Safeguards Research and Development Program Status Report, September- December 1970," Los Alamos Scientific Laboratory report LA-4605-MS (January 1971).

50. T. D. Reilly and M. L. Evans, "Measurement Reliability for Nuclear Material Assay," Los Alamos Scientific Laboratory report LA-6574 (January 1977).

51. T. D. Reilly and J. L. Parker, "A Guide to Gamma-Ray Assay for Nuclear Material Accountability," Los Alamos Scientific Laboratory report LA-5794-M (March 1975).

52. R. Gunnink and J. E. Evans, "In-Line Measurement of Total and Isotopic Plutonium Concentrations by Gamma-Ray Spectrometry," Lawrence Livermore Laboratory report UCRL-52220 (February 1977).

53. T. R. Canada, D. G. Langner, J. L. Parker, and E. A. Hakkila, "Gamma and X-Ray Techniques for the Nondestructive Assay of Special Nuclear Material in Solution," in "Coordinated Safeguards for Materials Management in a Fuel Reprocessing Plant, Vol. II," Los Alamos Scientific Laboratory report LA-6881 (September 1977), App. A.

54. J. W. Tape, "CdTe Gamma-Ray Detector Investigations," in "Nuclear Safeguards Research and Development Program Status Report January-April 1977," Los Alamos Scientific Laboratory report LA-6849-PR (August 1977), p. 27-28.

55. R. B. Walton, Los Alamos Scientific Laboratory, personal communication, September 1977.

56. A. von Baeckman, D. Ertel, and J. Neuber, "Determination of Actinide Elements in Nuclear Fuels by X-Ray Analysis," Advan. X-Ray Anal. 18, 62-74 (1975).

57. P. A. Pella and A. von Baeckmann, "The X-Ray Spectrometric Determination of Uranium and Plutonium in Solutions of Spent Nuclear Fuels," Anal. Chim. Acta 47, 431-438 (1969).

58. E. A. Hakkila, "Three-Wavelength X-Ray Absorption Edge Method for Determination of Plutonium in Nitrate Media," Anal. Chem. 38, 425-427 (1966).

59. K. J. Hofstetter, G. A. Huff, R. Gunnink, J. E. Evans, and A. L. Prindle, "On-Line Measurement of Total and Isotopic Plutonium Concentrations by Gamma-Ray Spectrometry," in Analytical Chemistry in Nuclear Fuel Reprocessing, W. S. Lyon, Ed., (Science Press, Princeton, 1978), pp. 266-274.

* U.S. GOVERNMENT PRINTING OFFICE: 1979-678-850

Printed in the United States of America. Available from
National Technical Information Service
US Department of Commerce
5285 Port Royal Road
Springfield, VA 22161

Microfiche $3.00

001-025	4.00	126-150	7.25	251-275	10.75	376-400	13.00	501-525	15.25
026-050	4.50	151-175	8.00	276-300	11.00	401-425	13.25	526-550	15.50
051-075	5.25	176-200	9.00	301-325	11.75	426-450	14.00	551-575	16.25
076-100	6.00	201-225	9.25	326-350	12.00	451-475	14.50	576-600	16.50
101-125	6.50	226-250	9.50	351-375	12.50	476-500	15.00	601-up	

Note: Add $2.50 for each additional 100-page increment from 601 pages up.