AN ANALYSIS OF PLUTONIUM ACCOUNTABILITY IN THE COPRECAL PROCESS

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(ABSTRACT)

In the late 1970's, emphasis on non-proliferation forced suspension of all commercial spent-fuel reprocessing. The spent-fuel storage problem plaguing the nuclear industry can be alleviated by reprocessing. For commercial spent-fuel reprocessing to again become a reality, a process is needed to reform reprocessing operations such that non-proliferation goals are satisfied. To satisfy these goals, the existing process which generates plutonium-nitrate solution must be altered to generate plutonium-uranium oxide powder. The COPRECAL process is designed to produce this solid. The COPRECAL process allows uranium and plutonium to be extracted from spent-fuel for reuse in commercial light-water reactors. The COPRECAL process is unique in that no pure plutonium is ever present throughout the process. Whether the COPRECAL process is intrinsically vulnerable to plutonium diversion is the object of this work.

A simulation model of the COPRECAL process is presented which employs state-of-the-art instrumentation to measure in-process plutonium through the simulated passage of time. Plutonium diversion schemes are incorporated into the model. After simulated thefts, model output statistics are plotted on control charts and analyzed. Results show need for major design changes in the COPRECAL process.

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COPRECAL - Co-precipitation and calcination

UPAA - Unit Process Accounting Area

DOE - Department of Energy

NDA - Non-destructive assay

LASL - Los Alamos Scientific Laboratory

CHAPTER ONE - INTRODUCTION

The development of domestic nuclear power systems has occurred with the assumption that thermal light-water reactors (the current type of reactors) fueled by low enriched uranium would be followed by thermal light-water reactors using mixed-oxide fuel (PuO_2 and UO_2), and finally by fast reactors using mixed-oxide fuel and functioning as breeders.[1] Nuclear fuel reprocessing was a vital part of this plan which provided an economical way to convert spent fuel into usable mixed-oxide fuel. This planned evolution is presently stalemated due to the potential of these advanced nuclear technologies spreading nuclear weapons material and technology.

The reprocessing link of the nuclear fuel cycle chain came under particular scrutiny because of the high-purity plutonium available in concentrated form thought to be attractive to potential divertors. As a result, in April of 1977, the Carter Administration mandated the halt of all commercial nuclear fuel reprocessing in order to comply with non-proliferation objectives. Research and development is now geared toward identifying fuel recycling technologies which offer less proliferation risk if deployed. The COPRECAL process (co-precipitation and calcination), where no chemically pure plutonium is ever present throughout the operation, is one such technological advance.

The COPRECAL process could be used at the product end of a reprocessing plant or the head of a fuel fabrication facility. In the process, a uranium-plutonium nitrate solution is injected continuously into a concentrated ammonium-hydroxide solution and immediately precipitated as an unfilterable slurry. The entire slurry is pumped to a calciner bed and is converted to mixed UO_3 and PuO_2. These dry powders are then batch-fed to a reduction-stabilization unit that reduces the UO_3 to UO_2. The result is a mixed-oxide powder suitable for fuel fabrication. The solid powder product afford some intrinsic protection against diversion that pure plutonium products do not.

Presently, reactor by-product plutonium, formed in the reactor core by transmutation of uranium in fission events, is left in stored spent-fuel assemblies are the assemblies have been removed from the reactor core. Most of the existing spent-fuel assemblies have cooled for only a relatively short time. Due to high radioactivity and heat production, they require considerable technical effort to process. This creates a technical barrier to the separation of plutonium that is difficult for individuals or sub-national groups to overcome. Should plutonium be reprocessed and used on a larger scale, these technical barriers would, to a large extent, disappear. The concern for the future is that the best technical and safeguards measures should be adopted to increase the protection of such material against diversion.

Nuclear safeguards consist of an integration of physical protective measures, materials control, and materials accountability.[2] This work addresses the materials accountability aspect of the COPRECAL process. Its purpose is to evaluate the COPRECAL safeguards effectiveness. Specifically, can plutonium be diverted from the process without detection using state-of-the-art instrumentation?

The analysis of the complex, high-throughput fuel-cycle facility requires in-depth knowledge of the facility, its processes, and its operational details. In addition to these requirements, there are other unique characteristics introduced by the decision analysis method which subdivides the facility into individual unit process accounting areas (UPAAs). To satisfy the material accounting demands, we need to use the computer to analyze large quantities of in-process plutonium data. Such an analysis is implemented in this work by modeling and simulating COPRECAL facility operations and the functions of the plutonium measurement instrumentation. The accuracy of the measurements and precision with which measurement are performed on in-process plutonium become the bases for the uncertainty attached to the safeguards question.

Although many different statistical tests are suitable for use in materials accountability, they all have several characteristics in common. Each operates on the estimation results to decide whether diversion has or has not occurred, and each requires some indication of detection probability.[3] This work uses control charts in its statistical treatment of model output data. After simulated diversion, each successive measurement of in-process plutonium is plotted as a function of time and a determination is made whether or not each falls within a predetermined critical region. The control limits and mean amounts of plutonium are established by assembling data from multiple computer runs without plutonium diversion. If simulated thefts cannot be successfully implemented without detection, the COPRECAL facility meets materials accountability goals and passes the safeguards test. Note, however, statistical treatments are always based on simplified models derived from assumptions that may or may not be valid.

The test proceeds as follows: Chapter Two presents a short history of nuclear fuel reprocessing and materials accountability and ends with a detailed description of the COPRECAL process. Chapter Three presents a formal literature search; Chapter Four outlines the simulation language; Chapter Five describes the simulation model; Chapter Six discusses model output and its statistical treatment; Chapter Seven lists conclusions; and Chapter Eight makes recommendations.

CHAPTER TWO - BACKGROUND

The Spent-Fuel Dilemma

After President Carter's non-proliferation policy statement in April of 1977, commercial spent-fuel reprocessing was prohibited on the grounds that for safeguards reasons it presented an unacceptable risk. The increase in international terrorism no doubt influenced the shift in United States policy. This policy statement created the problem of what to do with the spent fuel generated by commercial reactors already in operation.

In 1977, the Department of Energy (DOE) addressed this problem by creating a program to save any commercial reactor from shutdown due to lack of storage space for its spent-fuel. Repositories for spent-fuel storage would be built under the auspices of the DOE and the commercial utilities' discharged fuel would be ultimately stored there. In 1981, this concept was dropped in favor of augmenting existing utility spent-fuel storage pools.[4] Hopefully, this program can be successful. If not, the re-introduction of spent-fuel reprocessing into the present-day nuclear fuel cycle would certainly mitigate the consequences of accumulated spent-fuel.

The Nuclear Fuel Cycle

Figure 1 illustrates the pre-1977 nuclear fuel cycle with plutonium recycling. Uranium ore is mined and milled. The product, U_3O_8, is then refined into uranium hexafluoride (UF_6). The uranium must then be enriched in the U-235 isotope in order to satisfy criticality specifications. After enrichment, UO_2 is extracted by fluoride conversion. Fuel fabrication, the construction of uranium fuel assemblies precedes the use of UO_2 in the reactor. The lifetime of a fuel assembly in a commercial nuclear reactor is approximately three years, although about one-third of the fuel assemblies are replaced each year. Afterwards, a spent-fuel pool cools the discharged fuel before transport to a reprocessing center. After reprocessing, the uranium-nitrate and plutonium-nitrate solutions undergo conversion into solid fuels. The uranium is reflourinated into UF_6, enriched, then converted to UO_2 before being refabricated into fuel. Any highly radioactive raffinate generated in the cycle is sent to a storage facility for disposal.

The Reprocessing Dilemma

There is today only a single reprocessing facility available to handle commercial spent-fuel. It is not operating. Current limits to its use are largely sociopolitical rather than technological. A synopsis of the

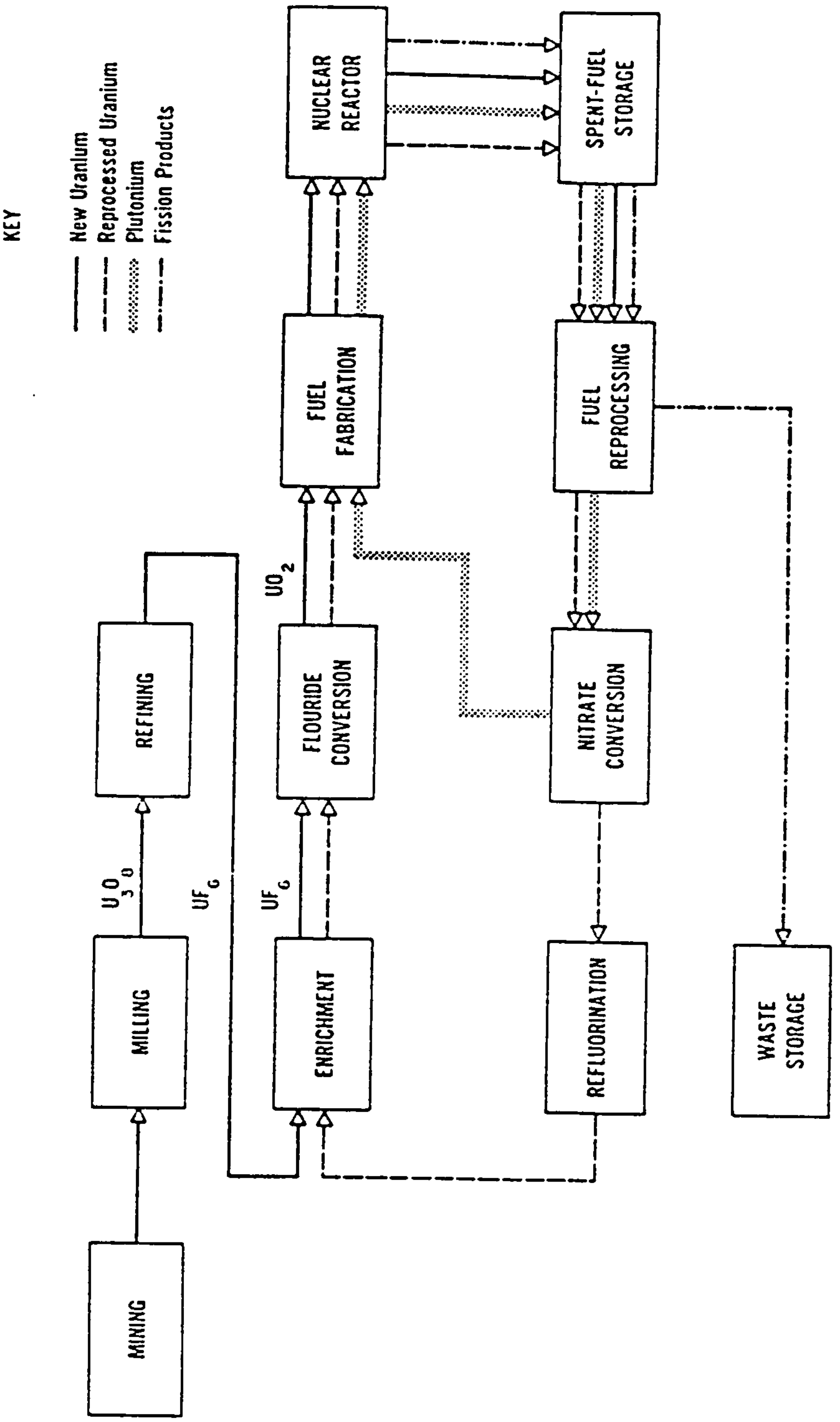

Figure 1. The pre-1977 Nuclear Fuel Cycle

United States commercial and military reprocessing centers is presented in Table 1 and is taken from Reference 5.

The commercial reprocessing industry had its birth in the late 1950's when the Atomic Energy Commission embarked on a program to encourage private industry to enter the commercial reprocessing field.[6] Previous experience was for military application only.

The Nuclear Fuel Services plant located in West Valley, New York entered the picture first. It recovered plutonium via the Purex process. Prior to its shutdown in 1972, it processed more than 630 metric tons of spent-fuel.[6] Newly imposed health and safety regulations prevented its returning to service.

The Midwest Fuel Recovery Plant in Morris, Illinois was General Electric's venture into spent-fuel reprocessing. This plant was based on an unproven process that was projected to possess the benefit of process simplicity to the extent a small reprocessing plant located near a nuclear reactor complex could operate economically. In 1974, General Electric announced the plant was not operable in its present form, that there were fundamental plant technical problems, and that substantial modification would be required if the plant were to have an opportunity for commercial operation.[6]

Table 1. Summary of United States Reprocessing Experience

FACILITY	DATE(S) OPERATIONAL	USE	PROCESS	STATUS
Hanford (WA)	1944-1972	Military	Various	May be Reactivated in 1980's
Savannah River (SC)	1954	Military	Purex	Operating
Idaho	1953	Military	Purex	Operating
Nuclear Fuel Services (NY)	1966-1972	Commercial	Purex	Shutdown for expansion; will not reopen due to inability for backfit to new regulations
Midwest (IL)			Hydrofluor	Process will not work on production scale; plant modification not planned at present
Barnwell (SC)	?	Military Commercial	Purex	Ready to operate pending federal decision for commercial reprocessing

The Barnwell facility, located in South Carolina, is the only plant currently available to restart United States commercial reprocessing. Its fate remains entrenched in a quagmire of political controversy. If spent-fuel storage requirements force a change in United States policy and allow the Barnwell facility to start operations, the technical question of how to comply with safeguards regulations remains. The relations stipulate plutonium-nitrate solutions must not be transported in liquid form; moreover, the plutonium cannot appear as a single pure product anywhere in the nuclear fuel cycle. The Purex processing, utilized by the Barnwell plant to covert the spent-fuel, produces a pure plutonium-nitrate solution and thus violates regulations. The COPRECAL process was designed to correct these faults.

The COPRECAL Solution

The COPRECAL process can be used to couple continuous fuel reprocessing and fabrications facilities. In this mode, pure plutonium would never be isolated and the uranium and plutonium-nitrate products from the Purex reprocessing facility would be feed material for the COPRECAL plant.[7] The COPRECAL product, a mixed-oxide powder, would satisfy solids transportation requirements.

The COPRECAL fuel product dissolves readily in nitric acid (HNO_3); however, the uranium cannot be separated from the plutonium solely by this technique. Uranium and plutonium are chemically very similar. Recovery of the plutonium from the mixed product is much more complicated because of the uranium dilution.[8]

Ostensibly, the absence of purified plutonium in the nuclear fuel cycle precludes the use of this material in surreptitous nuclear weaponry and meets with non-proliferation standards. Whether the COPRECAL process instrumentation can satisfy the materials accountability portion of nuclear safeguards is a different matter.

Materials Accountability

Reliable materials accountability includes the need for a quantitative determination of what, where, and how much material is being protected and a rapid detection and localization mechanism to account for a loss.[9]

Materials accountability in the nuclear facilities is not new. The nuclear materials produced at Oak Ridge and Hanford in the early 1940's were guarded carefully because of their extremely limited quantities and very sensitive potential military applications. Wet analytical chemistry was the predominant method of materials measurement.

In subsequent years, technical and political priorities related to nuclear energy changed. Federal regulations were adopted which placed a specific obligation on the private industrial sector to safeguard nuclear materials. It is now recognized that a truly effective measurement system must make use of both wet and non-destructive assay (NDA) methods.[9] Materials measurement in the COPRECAL facility will rely primarily on NDA techniques, though wet chemistry is also to be used.

Chapter Five elaborates on NDA instrumentation.

The COPRECAL Process

The COPRECAL process discussed in this work is assumed to be a combination of continuous and batch operations designed to convert 117 kg of plutonium per day. Three parallel process lines are required to meet this design-basis throughput. The author's computer model simulates the operation of a single process line as illustrated in Figure 2. Each line has as its main components three feed-blend tanks, a precipitator, four calciners, a primary filter, a secondary filter, four parallel reduction-stabilization stations each containing a primary and final filter, and a screening station.

Each feed-blend tank combines plutonium-nitrate solution from a Purex reprocessing facility and natural uranium-nitrate solution from a make-up tank to adjust the plutonium content relative to the heavy metal content to

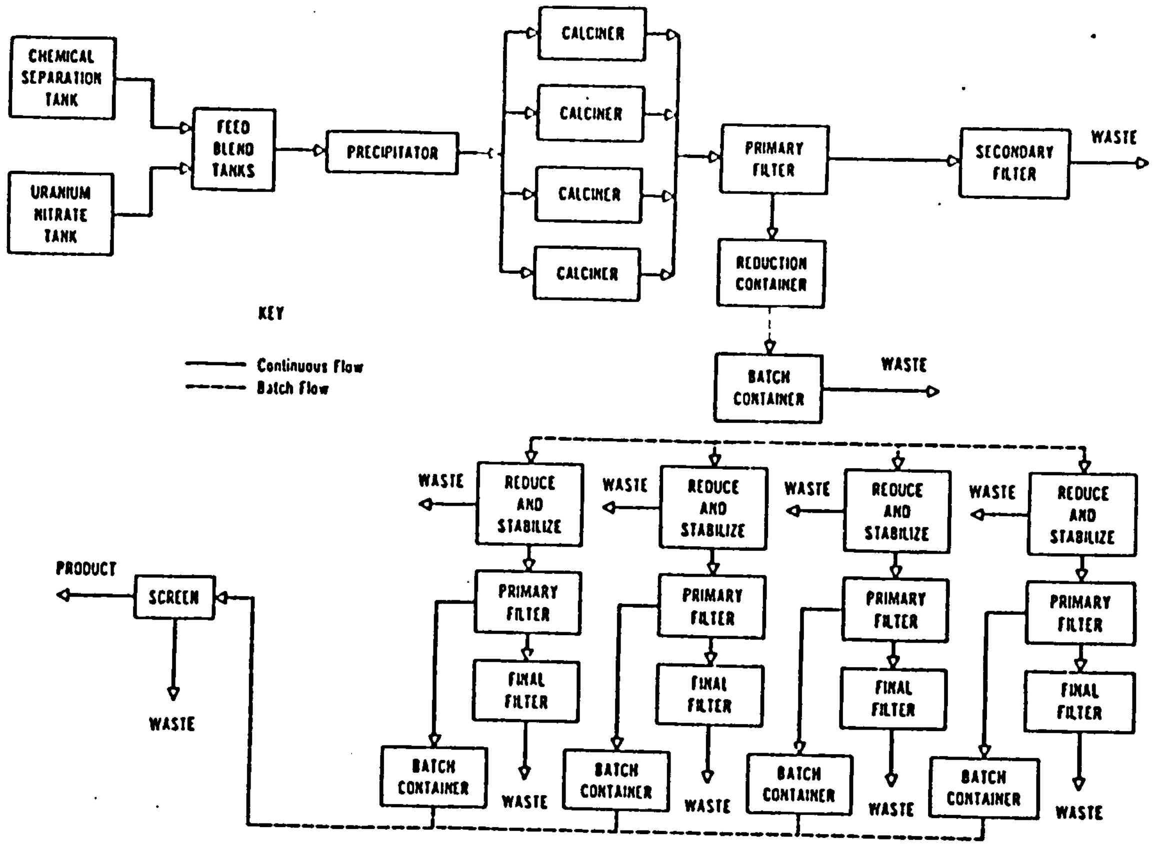

Figure 2. COPRECAL Flow Diagram

approximately ten percent. (This ratio remains constant throughout the process, thus allowing pure plutonium flow rates for the author's computer model to be established.) The feed-blend tanks feed the process line, in series, for about six weeks.

After testing and certification, the plutonium-uranium-nitrate solution from the feed-blend tank is pumped to the precipitator where ammonia is added to the solution to enhance plutonium precipitation. The ammonia reacts with the nitrate solution to form an unfilterable slurry of plutonium hydroxide, ammonium diruanate, and ammonium nitrate. The slurry is constantly circulated to ensure uniform mixing.

From this stream, a small amount of slurry is forced through orifices into four parallel calciners. The slurry metered into each bed is calcined to uranium and plutonium-oxide powder. The powder is raised from the bed by steam and decomposition gases resulting from the high temperatures (400°C) environment. Most of the ammonia added at the precipitator will pass through the calciner in the form of gas, though some will combine with oxygen to form water vapor.

A vacuum system helps the primary filter to collect the powder from the gas stream; the secondary filter collects the residue. Steam and decomposition gases are ejected through the secondary filter as waste. Every 30 minutes, powder caked on the primary filter is blown in to a reduction container located at the bottom of the primary filter. This blow-back procedure takes one minute. The process now becomes a batch operation.

The reduction container is emptied into the batch container at 120-minute intervals and is sent to one of four reduction-stabilization areas. Residence time there is 7.5 hours. A heated mixture of hydrogen and nitrogen is blown through the powder fast enough to turn the powder over, but not to blow it out of the container. The gas converts the UO_3 powder into UO_2 powder. The PuO_2 is unaffected. A vacuum system collects any spilled powder and expels it as waste.

At the completion of the reducing and stabilizing phase, the mixed-oxide powder is sucked through a primary filter. The powder entrained in the gas is caught and removed. This procedure takes seven minutes. A final filter acts as a back-up in case of a primary filter rupture. Waste gas is drawn through the final filter. The powder caught by the primary filter is dumped into another batch container, then transferred to a screening station. Transportation to the screening station takes nine minutes. Screening the powder takes one minute. Waste products are separated by the screen. The final product, PuO_2 and UO_2 powder, is transferred to a storage vault.

The interested reader is referred to Reference 10 for further details.

CHAPTER THREE - LITERATURE REVIEW

The Los Alamos Scientific Laboratory (LASL) has compiled a series of studies applying advanced nuclear materials accountability techniques to nuclear fuel-cycle facilities. The studies develop methods for evaluating safeguard systems and the data they produce, and stimulate further development of the facilities, processes, and instrumentation needed to improve nuclear safeguards. Included in the series is a preliminary report[7] on materials accountability in the COPRECAL process. LASL uses a continuous mode GASP IV model to simulate COPRECAL plant operations, to simulate the measurement process, and to generate statistical data for the evaluation of the safeguards effectiveness of the COPRECAL process.

The required operating parameters for the LASL study are obtained from a LASL computerized model of the COPRECAL process based on actual design data. The LASL model has three codes. The first code yields data representing material flows and inventories under expected normal conditions when the process is operating at the steady state. The first code is written in FORTRAN and uses GASP IV continuous simulation package to schedule events.[10]

Simulated accountability measurements and associated measurement errors are applied to the simulated process flow and in-process inventory data using the second code. The second code simulates measurements of the true materials in process generated by the first code and transmits appropriate measured values and their computed uncertainties to the third code, the safeguards data-analysis code.

The safeguards data-analysis code incorporates graphic-display techniques developed to analyze the data assembled by the previous codes. The four types of output charts generated are the cusum, Shewhart, Wilcoxon rank sum, and the Kalman filter.

The cusum or cumulative sum chart is a sequential plot of cusum values. A cusum value is defined as the difference between a mean value and an expected value.[11] The cusum analysis is most sensitive to small persistent shifts in process parameters.

The Shewhart chart is a sequential plot of materials balance data with error bars included.[11] Materials balance is a measure of the materials in a system or group of systems at set intervals of time. Unexpected deviations in material balance data signal a loss of material or a measurement error.

The Wilcoxon rank sum chart displays results of non-parametric statistics. This treatment assumes measurement error statistics are unknown or non-Gaussian.[12]

The Kalman filter approach arises because accountability systems rapidly generate large quantities of data that may contain weak signals caused by repeated small deviations in the noise produced by measurement errors.[13]

Two types of diversion were employed in the LASL work--abrupt and prolonged. The results were recorded by the aforementioned methods, evaluated, and the following conclusions formulated on the COPRECAL process:

1. The large 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.
2. Since none of these systems have been built or tested, the large NDA detection systems for measuring the plutonium inventory must demonstrate the degree of accuracy and precision simulated.
3. The COPRECAL process will need design changes to achieve effective materials accountability.

CHAPTER FOUR - THE SIMULATION CHOICES

The Decision to Simulate

Data is needed for the author's statistical analysis. The logical alternatives for generating the data are by either computer simulation of the process, conducting real-world experiments on the process, or by mathematical analysis.

The COPRECAL facility is in the design stages; therefore, obtaining all necessary data from direct experimentation is not possible. Hands-on experimentation on the NDA detection systems prior to plant construction will facilitate the decision-making process, but it cannot contribute enough significant input covering the range of materials accountability questions.

Mathematical analysis alone is not a sufficient substitute for simulation. The task of tracking plutonium through the complex is a long, tedious job tailored to computer techniques. Experimental design in conjunction with simulation is a more realistic venture in that the random elements of plant operations can be readily incorporated.

The Language Choice

The computer facility at VPI & SU supports several simulation languages; among them are CSMP, DYNAMO, GPSS, SIMSCRIPT, and GASP IV. Most share typical simulation traits.[14] They include:

1. random deviate generation
2. advancement of time
3. recording of data for output
4. performing statistical analysis
5. arranging outputs
6. error reporting

The decision to choose GASP IV as the simulation language was not predicated on its superiority in any one of these noted features. The prime motive behind the selection was the availability of Dr. J. Turek, whose technical expertise in GASP IV methodology greatly accelerated the model's successful completion. To a lesser degree, other factors influencing the decision were that GASP IV is FORTRAN based, a language not unfamiliar to the author. Also, the implementation of GASP IV simulation concepts is relatively easy. And, the language is very flexible and well-suited to modifications by the user. Further reasons for the choice can be found in the following short synopsis of the other languages.

CSMP is useful in solving non-linear differential equations. DYNAMO is advantageous when solving finite-difference equations.[15] Neither has any application in the author's model. GPSS is easily mastered, but lacks sophistication. SIMSCRIPT is too difficult to learn, especially due to the lack of self-diagnostics.[16]

The Discrete Mode Choice

GASP IV uses either a discrete, continuous, or combined simulation mode. Time is usually the independent variable and the dependent variables are functions thereof. Discrete simulation changes the dependent variables of the model at known intervals in simulated time, whereas continuous simulation allows the model's dependent variables to fluctuate depending on user-prescribed conditions. Combined simulation accommodates both discretely and continuously changing variables.

Accuracy pertaining to the author's model output is of the utmost importance in that the statistical inferences drawn are only as good as the model data output. Detecting the loss of several grams of plutonium in a process containing thousands of grams of the metal requires a significant effort in attaining model accuracy. Combined simulation seemed the ideal candidate for use in the model as the COPRECAL process contains both continuous and batch processes. But initial experimentation with the combined simulation mode yielded dubious results. The crucial criteria of accuracy was not evidenced.

The lack of accuracy in model data output can be ascribed to the author's programming inefficiency or if, indeed, the GASP IV combined simulation mode is not geared to pinpoint accuracy. Consultation with programmers experienced in GASP IV combined simulation methods yielded some insight. While they could cite no technical reason as to why combined simulation could not provide exactness, none had ever witnessed a great deal of accuracy in model output based on it. The LASL work, which used the combined simulation mode, attributed their model output inaccuracy to round-off error.

The inadequacy of the combined simulation mode dictated, by default, the discrete mode as the logical choice for the author's simulation. Apparently, continuous simulation introduces round-off error. The discrete mode meets the model data output accuracy requirements and lends itself well to model verification. The author's discrete model model output contained no round-off error.

GASP IV Overview

The author's GASP IV program consists of a structured arrangement of subroutines interrelated by two forms of program control. They are the executive function and the event-selection function.

The executive function decides how the program is to proceed in simulated time and directs control to the proper place to accomplish the given task. Examples of these tasks include record keeping, updating system status, producing intermediate reports, and advancing time.

The event-selection function operates under the auspices of the executive function. An event can be defined as an occurrence changing the status of the system. This type of program control provides instructions on a time-basis scheme. In other words, at a given point in simulated time, system status may be changed. Virtually all events are executed by user-written subprograms.

Consider the filling of the precipitator tank with slurry. When time has advanced to the point at which the event should occur (i.e., to start the filling of the tank), GASP IV recognizes this and program control is directed to the proper subroutine. After its execution, control is returned to GASP IV, which continues according to the logic of the model. At the end of simulation, GASP IV collects the model output data--the measured plutonium amount in the precipitator tank--and passes the data on to a user-written output subroutine.

For a more expert treatment concerning general GASP IV programming, the reader should refer to Reference 17.

The author's GASP IV simulation of the COPRECAL process predicts measured amounts of in-process plutonium before, during, and after diversion. A discussion on the author's model is found in Chapter Five.

CHAPTER FIVE - THE AUTHOR'S MODEL

The author's model is a user-friendly, uncomplicated model honed to yield strategic results. This model uses COPRECAL design-based data and projected COPRECAL instrumentation efficiencies to generate data for the construction of control charts. The control charts show the likelihood of detecting plutonium thefts. This model is unique in that it can bypass the operational phase of decision-making and be used directly by a strategic-level manager to probe COPRECAL safeguards issues.

The LASL model is extremely complicated and would require a user to have considerable programming knowledge. The author's model is much easier to use, especially for those without computer programming experience. The strengths of the author's model are that it allows the input variables, that exert the major influence over model output (the flow rates and instrument efficiencies), to be easily changed, it can incorporate future design changes with minimal effort, and the output data contains no round-off error. The major weakness of the author's model is its lack of sophistication in analyzing the output data. The LASL model uses four types of advanced statistical techniques to analyze its output data. The results of both the author's and LASL's analyses are strikingly similar. Thus the simplified analysis of the author did not sacrifice a great degree of accuracy.

This chapter explains the author's model logic, discusses the model variables and their roles, then comments on the end of simulation. The two types of diversion schemes, abrupt and prolonged, are examined. Model verification procedures are outlined; Appendix A provides the verification details.

Model Logic

Figure 3 illustrates the model logic flow. There are seven event codes, each responsible for directing the simulation to the appropriate task it must perform at the appropriate time. The discrete model advances in time on a minute-by-minute basis and updates the plutonium inventories after filling the plant components. The continuous flow of material is discretized by invoking material-balance type equations throughout the components of the COPRECAL process:

$$\text{Pu (TNOW)} = \text{FI} - \text{FO} + \text{Pu (TLAST)} \qquad (1)$$

where,

FI = plutonium flow into component (gm/min),

FO = plutonium flow out of component (gm/min),

Pu (TLAST) = plutonium in component at the last event (gm).

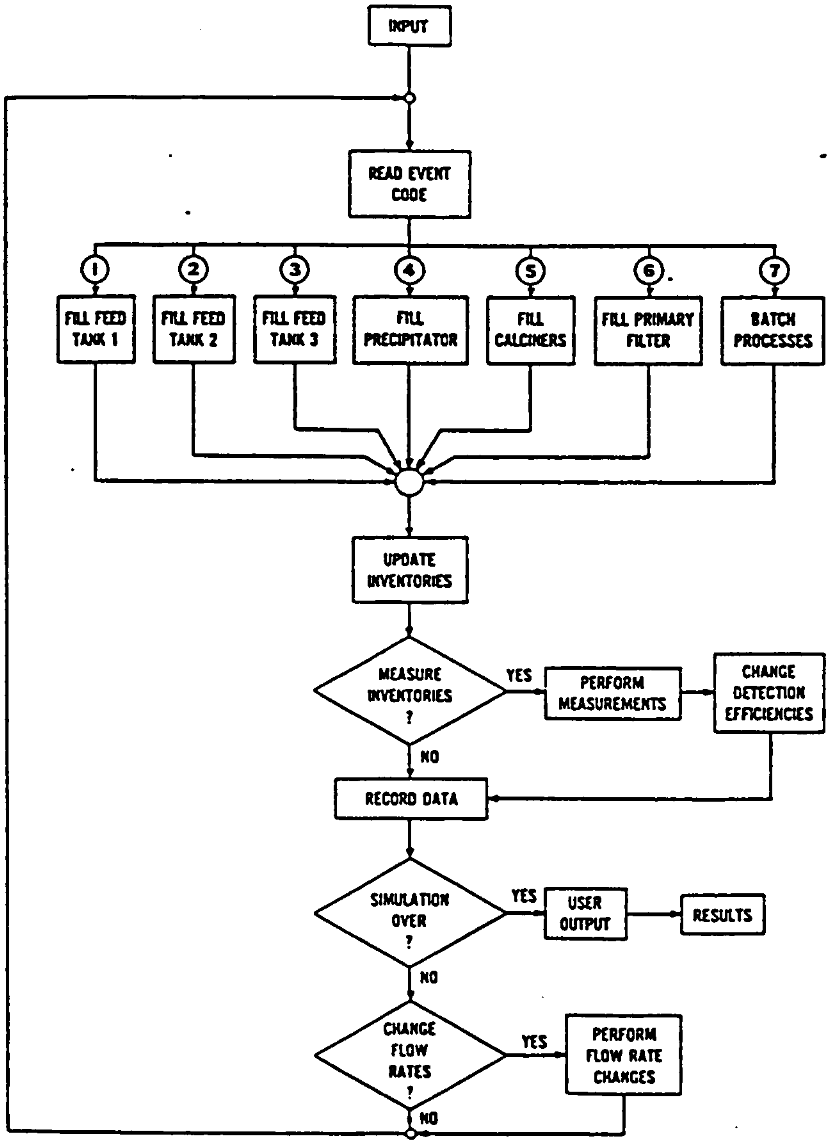

Figure 3. Model Logic Flow

This type of equation is applied to each plant component to determine the amount of plutonium stored there. Every 120 minutes, a measurement subroutine incorporates projected instrument calibration errors and precision errors to generate measured amounts of plutonium in the components. The measured amounts of plutonium are recorded for output and will be used in the statistical analysis. New efficiency values are generated for each subsequent measurement routine in order to take a conservative approach to measurement analysis.

Flow rates are adjusted hourly to force randomness into the process operations. Batch operations, the type of simulation more suited to the discrete mode, are scheduled and implemented at their allotted times. At the end of simulation, the output is user-assembled into tabular forms.

Flow Rate Variables

Realistic plant operations are simulated by incorporating random fluctuations into the model flow rate variables. Hopefully, model output data will represent an accurate basis from which safeguards predictions can be formulated. If not, statistical analysis of model output data and the corresponding conclusions will be dubious at best. Equipment malfunction, tank overflows, instrument failure, and human error are several abnormalities expected to interrupt routine operations in real-time operations.

These abnormalities are ignored in the author's simulation model.

Knowing the amount of plutonium flow per minute between plant components permits the model to discretely inventory, per minute, the in-process plutonium. Table 2 lists the mean flow-rate variables. Feed-blend, precipitator, and calciner flow variables are initially calculated by consideration of the design throughput of 117 kg/d of plutonium and of the physical dimensions of the component. When sufficient background information is not available, the mean flow-rate variables are assumed.

The simulation model updates the pertinent flow variables at hourly intervals by generating random deviations from assumed normal distributions. The new flow variables are then implemented. The random deviations are prohibited from exceeding a minimum or maximum value. The limits are prescribed as ± 5% of the mean values listed in Table 2 and lie within 3 standard deviations from the mean. If the value of the deviation generated is less than the prescribed minimum value, the minimum value is chosen by default. If the deviation generated is greater than the prescribed maximum value, the maximum value is chosen as the deviation. This is not sampling from a truncated distribution, but sampling from a distribution having a discrete probability of obtaining extreme values.[17]

Table 2. Mean Flow-Rate Variables

PUMP INDENTITY	MATERIAL	MEAN FLOW RATE
Chemical-Separations Tank	Pu in HNO_3	319.44444 l/m
Uranium-Nitrate Tank	U in HNO_3	63.888888 l/m
Feed-Blend Tank	U-Pu in HNO_3	0.67760942 l/m
Precipitator	PuO_2	27.104377 gm/m
Calciners	PuO_2	27.104377 gm/m
Primary-to-Secondary Filter	PuO_2	0.27104377 gm/m
Secondary Filter to Waste	PuO_2	0.0027104377 gm/m
Batch Container Waste	PuO_2	1.0 gm/batch
Reduction-Stabilization Waste	PuO_2	1.0 gm/batch
Reduction-Stabilization to Primary Filter	PuO_2	200.0 gm/m
Primary-to-Final Filter	PuO_2	2.0 gm/m
Final Filter to Waste	PuO_2	0.02 gm/m
Screen to Waste	PuO_2	32.0 gm/batch

The initial input flows to the feed-blend tanks are assumed values. They are updated accordingly. The feed-blend tank plutonium concentration is randomly chosen using the design-basis concentration of 40 gm/l as the mean. The sampling distribution area lies within ± 1% of that figure. These limits are justifiably small because wet analytical chemistry techniques test for the concentration criteria.[10] The mean primary-to-secondary filter flow rate is assumed to be 1% of the calciner feed rate. The mean secondary-filter-to-waste flow rate is assumed to be 1% of the primary-to-secondary filter flow. Remaining model flow variables are chosen solely by user discretion. Waste loss and screen loss are constants not updated hourly by the author's simulation model.

Instrumentation

Safeguards effectiveness relies heavily on the detection capabilities of the process instrumentation. All facets of real-time plant activities involving plutonium should be monitored to determine whether material accountability standards are met. The simulation model complies with this intent by providing a routine to simulate process monitoring. The routine yields output representative of real-time NDA measurements of the in-process plutonium. Inventory assessment is done in all major COPRECAL components involved with plutonium. The components are assumed to include any respective piping volumes.

The LASL work[10] stipulated the use of NDA techniques for process monitoring. NDA instrumentation is based on the nuclear properties of plutonium; these properties are used to measure the content of material which cannot be sampled in representative fashion or which does not easily yield to dissolution. The detection system utilizes the intrinsic radiation of the plutonium by neutron coincidence counting of Pu-240 spontaneous fissions.[9] The monitoring does not disrupt normal process operations unnecessarily and gives relatively quick and accurate results. The precision attainable by NDA techniques ranges from 20% to 1%.[9] The major drawback of the method is its inability to measure uranium.[18]

Instrumentation Error

The essence of the detection capabilities lies within the projected instrument error. Foreseen factors adversely affecting NDA instrumentation applications are background and shielding effects, backscatter, geometry effects, high temperature environments, and electronic interference.[19] The magnitude of the error introduced by these combined effects will have to be ascertained through field testing. This type of error is neglected by the author's model.

Table 3 describes measurement points, measurement types, and their estimated uncertainties for plutonium materials accountability. The data in Table 3 was taken from the LASL work.[10] There are two categories of instrument error. Calibration error represents the uncertainty in the the instrument's ability to convert raw measurements to plutonium mass. Precision error represents the estimated scatter in the individual raw measurements. Both types of error are incorporated into the model by the following method.

A random deviation from a normal distribution is generated utilizing the Table 3 calibration error parameter. Then, the same procedure is repeated using the calibration error output value as the mean and the precision error parameter as input. The resultant random deviation equals the simulated measurement of the in-process plutonium. Except for inventory measurements of the feed-blend tank, all measurements are produced in the aforementioned manner.

Because of its large size, the feed-blend tank plutonium inventory measurement combines a volume measurement with a concentration measurement. Their product equals the simulated measurement of the feed-blend tank plutonium amount.

End of Simulation

The simulation is terminated prior to the first

Table 3. Instrument Error Parameters

MEASUREMENT POINT	MEASUREMENT TYPE	PRECISION ERROR % (1σ)	CALIBRATION ERROR % (1σ)
Feed-Blend Tank	Volume	1	1
	Concentration	0.2	0.2
Precipitator	NDA	10	1
Calciners	NDA	10	1
Primary Filter	NDA	10	1
Secondary Filter	NDA	10	1
Reduction Container	NDA	2	1
Reduction-Stabilization Area	NDA	10	1
Final Filter	NDA	10	1
Screen Waste	NDA	2	10
Other Waste	NDA	2	5
Product	NDA	1	0.5

feed-blend tank emptying. Each of three feed-blend tanks are designed to continuously feed the process line for two weeks; as a tank empties, another replaces it. The total real-time operation of one COPRECAL process line is approximately six weeks. But it is not necessary, or prudent, to stimulate over the entire time span. The decision to halt the model after it executes about 32 hours of simulated operations is based on considerations given below.

The primary objective of the model is to generate output for statistics-gathering on plutonium accountability. Material accountability presents the greatest problem at a time when the greatest amount of material exists. This situation occurs at the start of COPRECAL operations when the feed-blend tank is full. Thus, it is not necessary to extend the simulation beyond a reasonable time frame. Moreover, the estimated cost at the VPI&SU computer facility for only one model run simulating the entire six-week operation of a COPRECAL facility is $200.00. This amount is far too excessive, especially since the output data following several days of simulated operations is not enlightening.

Diversion schemes are programmed to occur when the process line contains the largest amount of plutonium and detection capabilities correspondingly are diminished.

Diversion Schemes

Two methods of theft are devised for model implementation. They are abrupt diversion; the removal of approximately 1 to 3 kg of plutonium from the process line components within an allotted time of one minute, and prolonged diversion; the removal of .75 gm of plutonium per minute. To the author's knowledge, no literature exists which can validate said amounts as logistically viable; hence, they are assumed to represent realistic figures.

The feed-blend tank, the precipitator, and the reduction-stabilization area are the targets for the simulated plutonium thefts. They are chosen because of their large plutonium inventories in relation to their positions in the sequence of plant operations. The large inventories severely inhibit instrument detection capabilities. The degree of ease of separations of the plutonium from the uranium was not considered when choosing plant components. If this strategy were to be adopted, the solid powdered-oxide product would probably be the most desirable target to a potential divertor. Furthermore, solids are physically more manageable than liquids.

The bulk removal of plutonium from the process line categorizes abrupt diversion. After the act is perpetrated, the instrumentation system is queried to determine if it can detect the loss. If the instrument outputs can indicate the theft, then material accountability standards are not compromised. The process instruments are read and the data correlated every two hours in case random fluctuations in the flow variables or instrument output precluded immediate theft detection.

Prolonged diversion is the continuous bleeding of small amounts of plutonium from the process line. This is a more insidious type of tampering. Since the instrumentation is not expected to detect such small amounts quickly, the issue is how much time elapses and how much material is stolen before the diversion attempt is discovered. Comparing the time actual diversion begins to the time the diversion attempt is discovered can yield the stolen amount of plutonium.

One computer run produces output for the three distinct abrupt diversion cases. Model output mechanics allow this efficient approach. Similarly, only one computer run is necessary for the prolonged diversion cases. Chapter Six elaborates on the mechanics of model output.

Model Verification

The verification of the model is approached in two ways. One compares the LASL model output to the output of this work to show concurrence. The second, a more rigorous endeavor, entails suppressing the instrument error subroutines to yield data amenable to hand calculations. The detailed hand calculations corroborate the author's model output. These methods are presented in Appendix A.

CHAPTER SIX - RESULTS

This chapter covers the results of the simulation. First, the need for grouping model output data into unit process accounting areas (UPAAs) is explained. The following section discusses the diversion testing procedure via control charts. Control chart construction for the UPAAs is then outlined. Prior to presenting the outcome of the simulated abrupt and prolonged diversion attempts, a reference case is tested and the results are examined. Finally, the sensitivity of the model to instrument error is examined.

Unit Process Accounting Areas

To determine the safeguards status of the facility from the large body of measured information is a formidable task. The many measurements certainly contain useful information when viewed collectively, but an isolated set of measurements will probably not shed any light on the accountability matter. A method is needed to treat the large amounts of output safeguards data in a simple yet effective framework.

The strategy used involves partitioning the facility into discrete accounting envelopes, i.e.,(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 material balance can be drawn around it. Figure 4 illustrates the three UPAAs used in the model. The LASL work[10] also used three UPAAs in a similar manner. By dividing the facility into unit processes and measuring all material transfers, quantities of material much smaller than the total plant inventory can be controlled on a timely basis. Also, any discrepancies are localized to that portion of the process contained in the individual UPAA.

UPAA 1 contains the feed-blend tank only. The very large plutonium inventory in the tank necessitates its separation from the remaining plant processes for accountability purposes. UPAA 2 includes most of the remainder of the components used in the continuous operations phase while UPAA 3 includes most of the batch operations components. The product is not included in UPAA 3 as its constant accumulation would serve to weaken the accountability aspects of the UPAA strategy. With the exception of UPAA 1, the balance of material in the areas can be assumed to be constant after the initial filling of the process components. Accumulations of waste are small enough to be neglected. This constant balance greatly simplified UPAA 2 and UPAA 3 control chart construction.

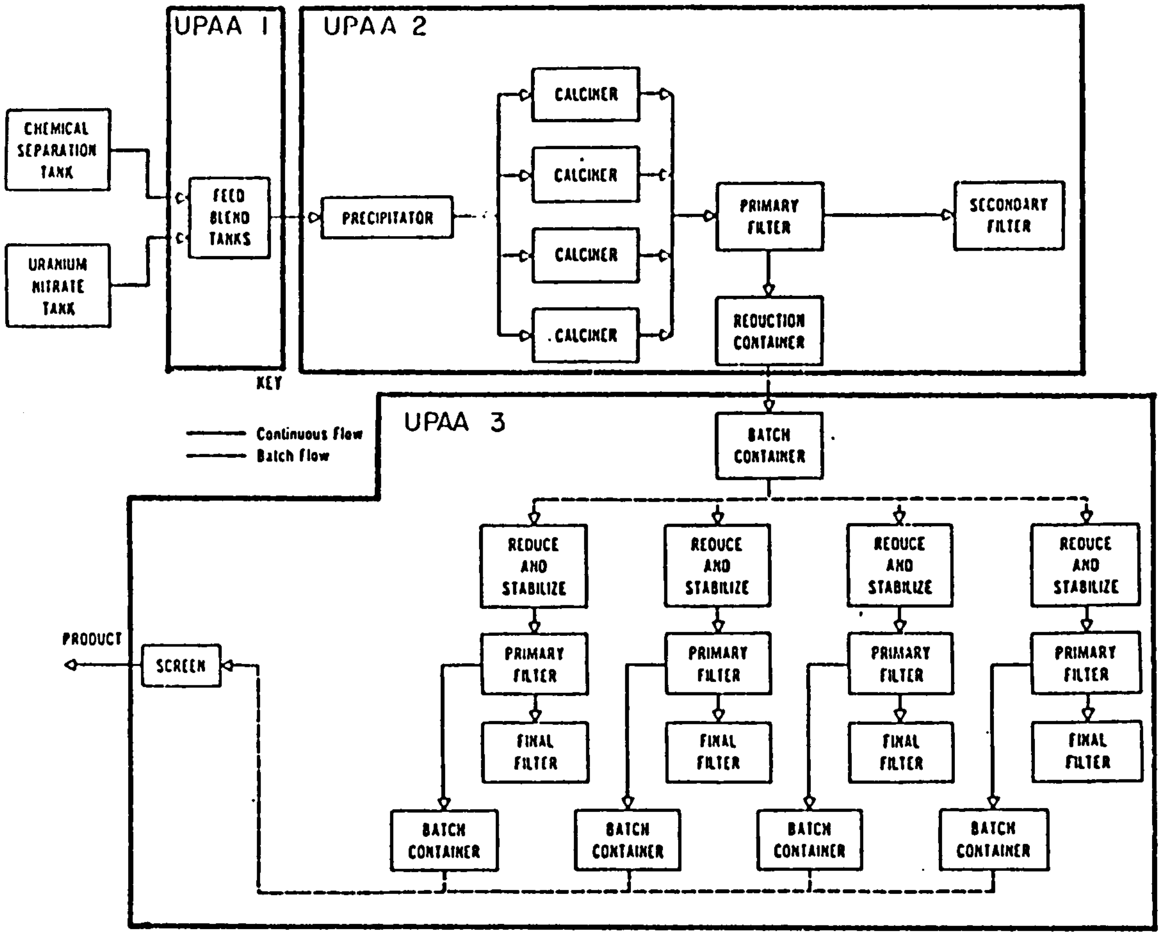

Figure 4. Model Unit Process Accounting Areas

A summary of model output data for one computer run simulating 32 hours of one process line's operation is given in Table 4. Measurements of the UPAA in-process plutonium were simulated at two-hour intervals. The measurement locations were given in Table 3. The values listed in Table 4 are the sums of those measurements performed per respective UPAA. No values are listed when the UPAA has not been completely filled.

Control Chart Testing

Hypothesis testing provides a logical method for analyzing possible diversion attempts.[12] The two hypotheses are:

1. H_0 -- diversion has not occurred ($u = u_0$)
2. H_1 -- diversion has occurred ($u > u_0$)

where,

u = the mean amount of UPAA plutonium (gm) and

u_0 = the measured amount of UPAA plutonium (gm).

A control chart and a test of hypotheses are equivalent.[12] The control chart construction involves determining the null hypothesis value, u, and the critical region defined by the control limit. Only a lower control limit is necessary as diversion constitutes removal of material.

The null hypothesis parameter, u, is estimated by

Table 4. Sample of Model Output Data

TIME (hr)	MEASURED PLUTONIUM AMOUNTS (gm)		
	UPAA 1	UPAA 2	UPAA 3
	X_1	X_2	X_3
2	0.645308943 D + 06	0.0	0.0
4	0.620167438 D + 06	0.295829997 D + 04	0.0
6	0.628798378 D + 06	0.306566056 D + 04	0.0
8	0.623554236 D + 06	0.305054273 D + 04	0.0
10	0.614384754 D + 06	0.299439493 D + 04	0.0
12	0.616389472 D + 06	0.289106260 D + 04	0.132795231 D + 05
14	0.621372737 D + 06	0.291550594 D + 04	0.119447949 D + 05
16	0.622060098 D + 06	0.290626151 D + 04	0.130204449 D + 05
18	0.616935829 D + 06	0.296674320 D + 04	0.126045732 D + 05
20	0.623748402 D + 06	0.299797916 D + 04	0.136432010 D + 05
22	0.600938070 D + 06	0.294371308 D + 04	0.139206526 D + 05
24	0.606088466 D + 06	0.297514394 D + 04	0.131485860 D + 05
26	0.590888112 D + 06	0.304241022 D + 04	0.142067548 D + 05
28	0.600703652 D + 06	0.299014614 D + 04	0.130546260 D + 05
30	0.580748535 D + 06	0.290965642 D + 04	0.147859610 D + 05
32	0.588219691 D + 06	0.293228207 D + 04	0.128522256 D + 05

averaging values output from five computer runs. Expense is the limiting factor here. The decision to accept or reject H_0 relies on a 95 percent confidence level chosen for the lower control limit.

After a diversion attempt, the sample statistics (the measurement figures) are plotted on the control chart as a function of time and each statistic is noted to see if it falls within the critical region. Those falling outside the region could indicate a diversion attempt and be used to estimate the amount of plutonium stolen. Of course, the random fluctuations programmed into the model can give false alarms due to shifting means and randomly generated poor measurements.

Control Chart Construction

For the case of UPAA 1, the plutonium inventory decreases in time as the feed-blend tank empties into the process line. Thus, the same size cannot be assumed to remain constant. This is remedied in the following manner.

A control limit value, $\mu - 2\sigma$, is calculated for each measurement time using the equation:

$$\mu - 2\sigma = \bar{x} - k\left(\frac{s}{\sqrt{n}}\right) \tag{2}$$

where, $\bar{x}$ = the mean amount of plutonium at the corresponding measurement time, i, calculated by averaging single values, x_i, from the output of five computer runs,

$k = 2.132$, the one-sided 95 percent confidence level constant,

$$s = \sum_{i=1}^{5}\left(\frac{x_i - \bar{x}^{\frac{1}{2}}}{n}\right)$$, the estimated standard deviation,

$n = 4$, the number of degrees of freedom minus one and

$2\sigma =$ the 95 percent confidence level.

The calculated mean amounts of plutonium, x, and corresponding lower control limits are fitted using the method of least squares to yield the UPAA 1 control chart presented in Figure 5.

The UPAA 2 and UPAA 3 plutonium inventories can be assumed to remain constant for the 32 hours of simulated operations; thus, the mean amount of plutonium expected, x, and the estimated standard deviation, s, will not change drastically for each measurement time. The sample size is increased, so the number of degrees of freedom is increased. This adds to the likelihood of the sample statistics estimating their respective parameters more accurately.

The result of the application of Equation 2 to UPAA 2 statistics to yield a control chart is displayed in Figure 6. The result of the application of Equation 2 to UPAA 3 statistics to yield a control chart is displayed in

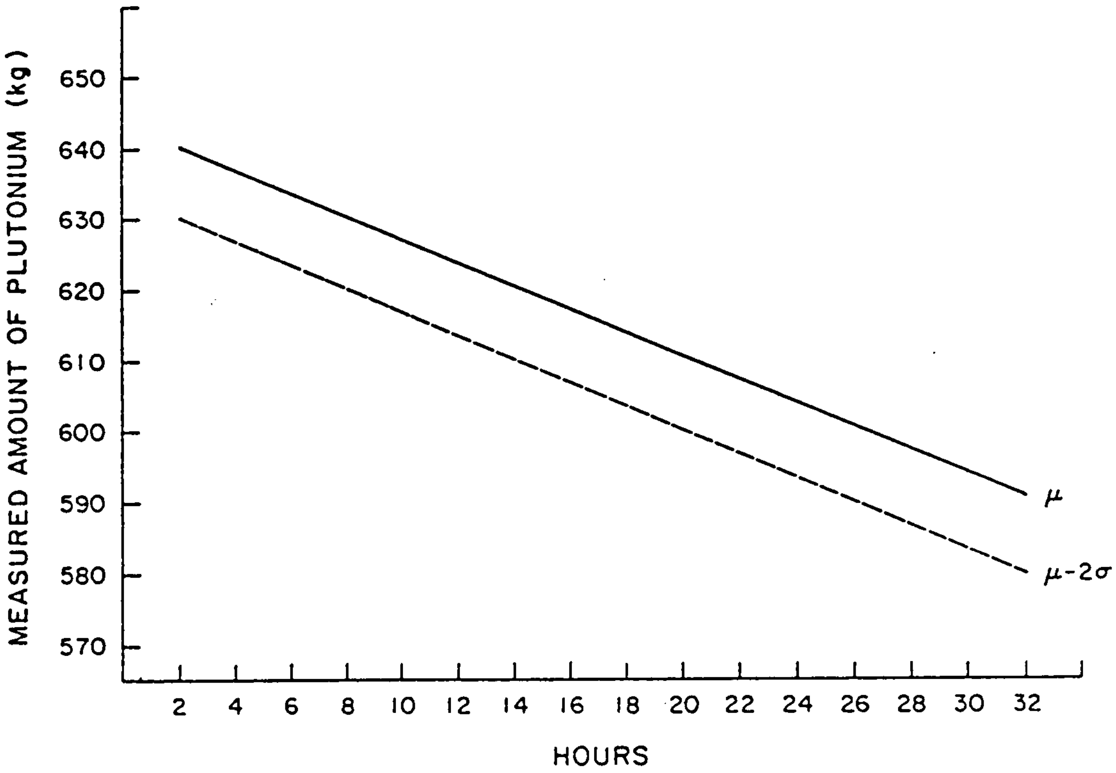

Figure 5. UPAA 1 Control Chart

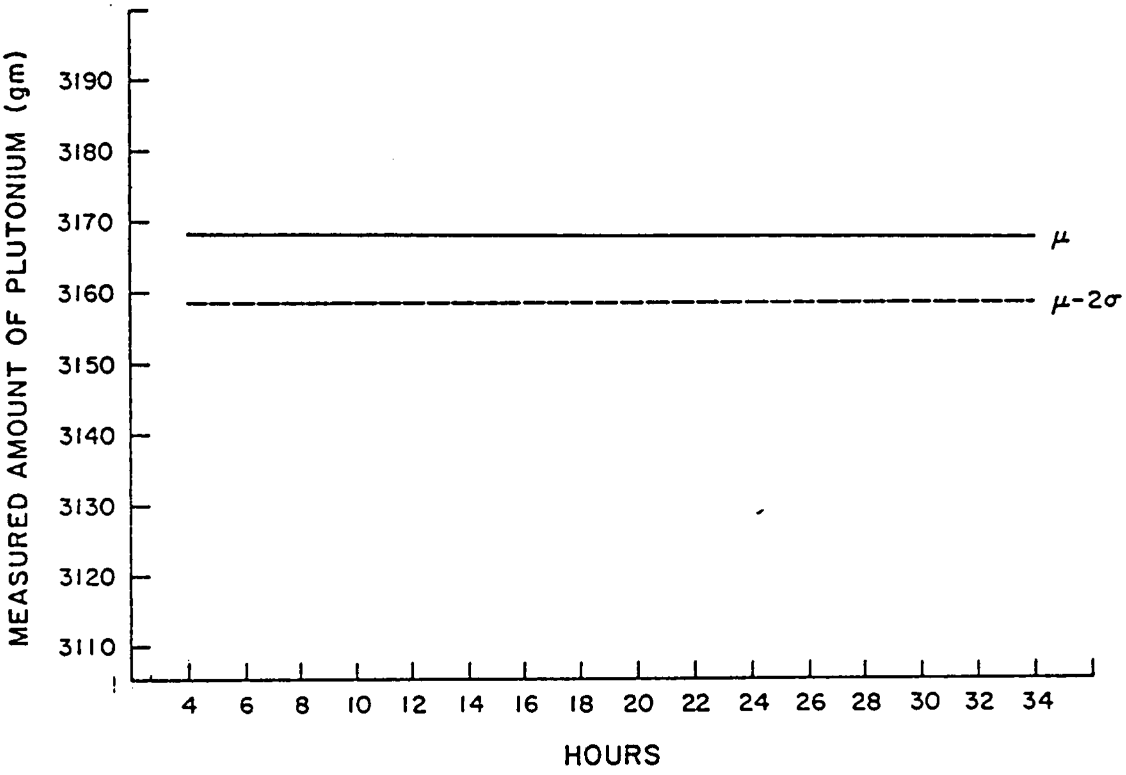

Figure 6. UPAA 2 Control Chart

Figure 7.

Reference Case Analysis

A reference case was tested to determine the number of false alarms on the control charts generated by the simulated measurements. No diversion schemes were implemented in this reference case simulation. Figure 8 shows the results plotted on the UPAA control charts.

There are four instances in the UPAA 1 control chart that could be interpreted as signaling a diversion. These are marked by the star symbols (*). The remainder of the measurements fall within the accepted critical region. Since no two stars fall together in consecutive measurement intervals, it is probably safe to assume no diversion has taken place and the four readings represent erroneous measurements produced by large calibration and/or precision errors in the instrumentation.

The control chart of UPAA 2 indicates five possible diversion attempts from its constituent components. Most of the measurement values falling under the control limit do not do so consecutively; thus inadequate measurement instrumentation can be blamed.

In the UPAA 3 control chart, the sporadic nature of the outliers again suggests instrumentation errors rather than an actual loss of plutonium.

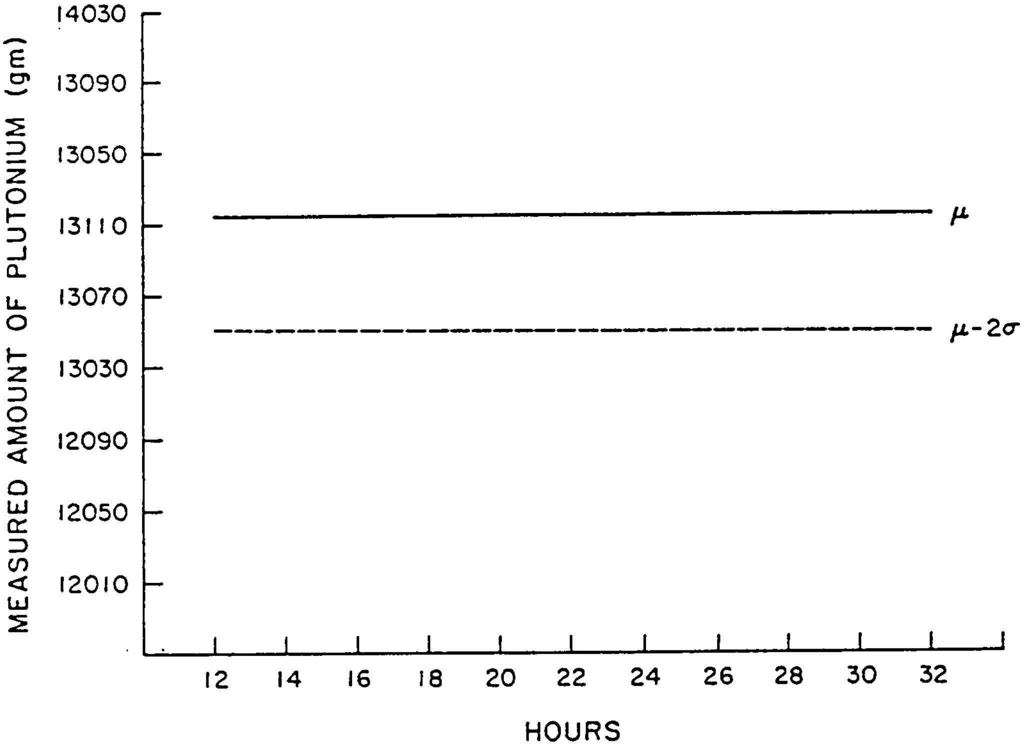

Figure 7. UPAA 3 Control Chart

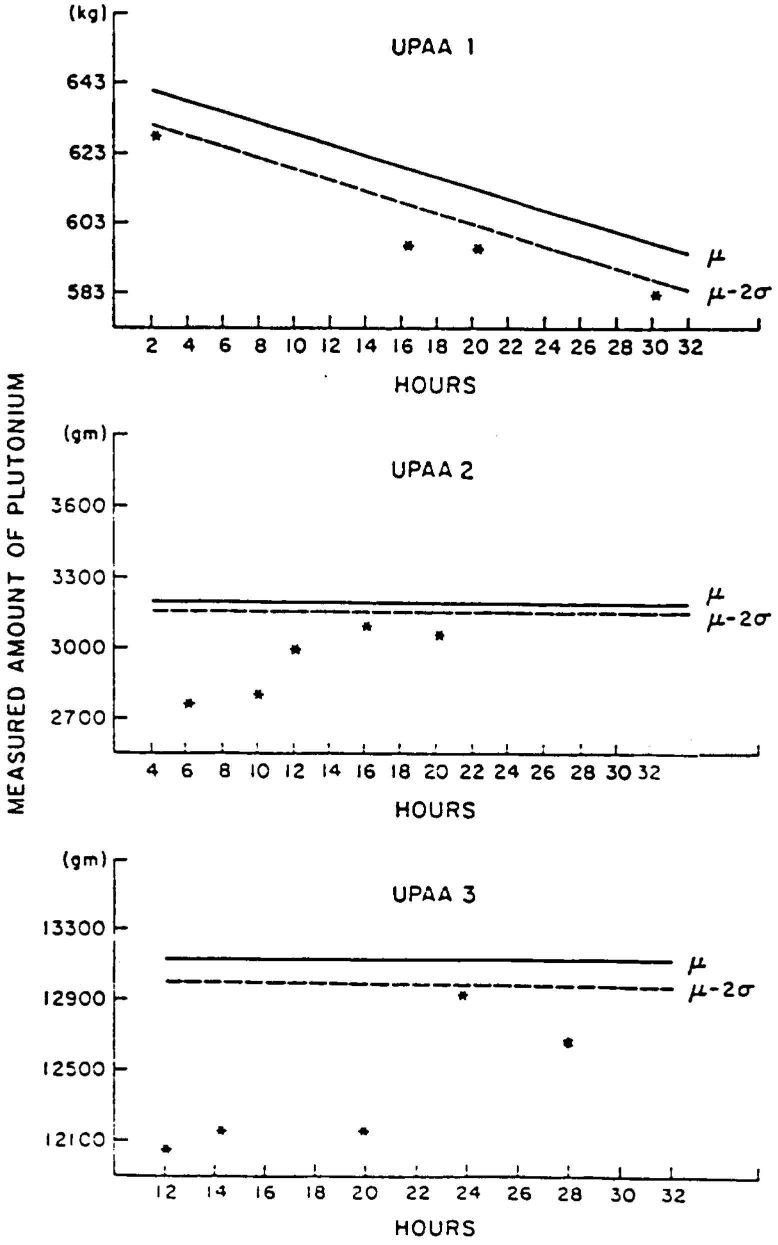

Figure 8. Reference Case Control Charts

Abrupt Diversion Case Analysis

The only difference between the abrupt diversion simulation and the reference case simulation was that at predetermined times, approximately 1 kg to 3 kg amounts of plutonium were diverted from each UPAA within a time span of one minute. The flow variable and instrument error subprograms, discussed in Chapter Five, maintained uniqueness of operations by generating and implementing the exact same random deviations for both simulation cases. This is true for all of the simulation cases analyzed.

The abrupt diversion of plutonium in the UPAA 1 feed-blend tank occurred at approximately 1.5 hours after the start of the simulation time. The abrupt diversion in the UPAA 2 precipitator occurred at approximately 16.5 hours. The abrupt diversion in the UPAA 3 reduction-stabilization area was programmed to occur at approximately 18.5 hours.

Measurements taken after these times should reflect the diversions by falling below their respective control limits. Figure 9 displays the control charts assembled for the abrupt diversion cases.

The UPAA 1 control chart indicates no diversion has taken place. But the failure of the instrumentation to detect the removal of an estimated .001 percent of the total UPAA 1 plutonium inventory is not surprising.

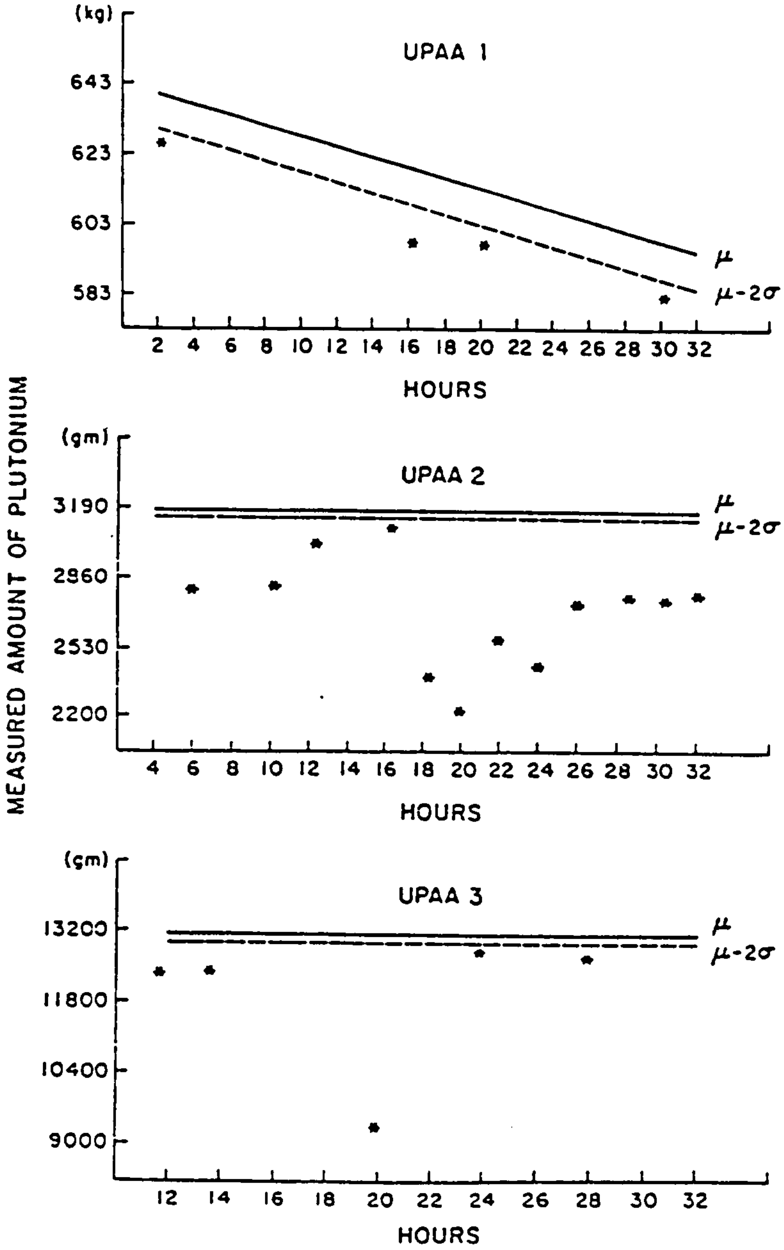

Figure 9. Abrupt Diversion Control Charts

The UPAA 2 control chart indicates a large diversion of plutonium occurred at the 18-hour point. Each successive measurement corroborates the evidence. The removal of approximately 1 kg of material from UPAA 2 is an estimated 30 percent reduction in its inventory. The maximum instrument error cited in Table 3 was ten percent; hence, it is within reason to expect the UPAA 2 control chart to function efficiently for this case.

The time at which the diversion occurred, somewhere between the 16- and 18-hour measurement points, is predicted by the control chart. But, the amount of diverted plutonium is predicted by the measurements' averages to be only 620 gm. This is in error by about 30 percent.

The UPAA 3 control chart fails to show any diversion trend. The seven percent reduction in its in-process plutonium by abrupt diversion is not a substantial enough amount for the UPAA 3 instrumentation and control chart analysis to effectively perform the task of accountability.

Prolonged Diversion Case Analysis

Prolonged diversion is categorized by the removal of .75 gm of plutonium per minute from the UPAAs. The diversion locations and starting times were the same as in the abrupt diversion case. For this case analysis, besides detecting losses, it was hoped that something could be learned about the sensitivity of the instruments to identifying quantities of material removed.

The UPAA 1 and UPAA 3 control charts shown in Figure 10 fail to predict any downward shifts in measurements. The total amount of plutonium removed from each UPAA after 32 hours of simulated process operations corresponds to the total amount of material removed for each UPAA in the abrupt diversion case. Both UPAA control charts are incapable of detecting either bulk or small, continuous diversion.

The UPAA 2 control chart, at the end of simulation, tends to show a measurement trend falling below the critical region. A longer simulation time is needed for confirmation. The average amount of diverted material predicted by the last two measurements is 175 gm. In actuality about 700 gm of plutonium had been programmed out of the process area at that point in simulated time.

Sensitivity Case Analysis

The factors exerting the major influence over the model output are the flow rate and the instrument error parameters. The instrument precision and calibration errors were chosen for the sensitivity analysis as they are deemed as having the greatest control over the instrumentation output.

The Table 3 values were improved by 25 percent, then applied to the abrupt diversion case to determine whether

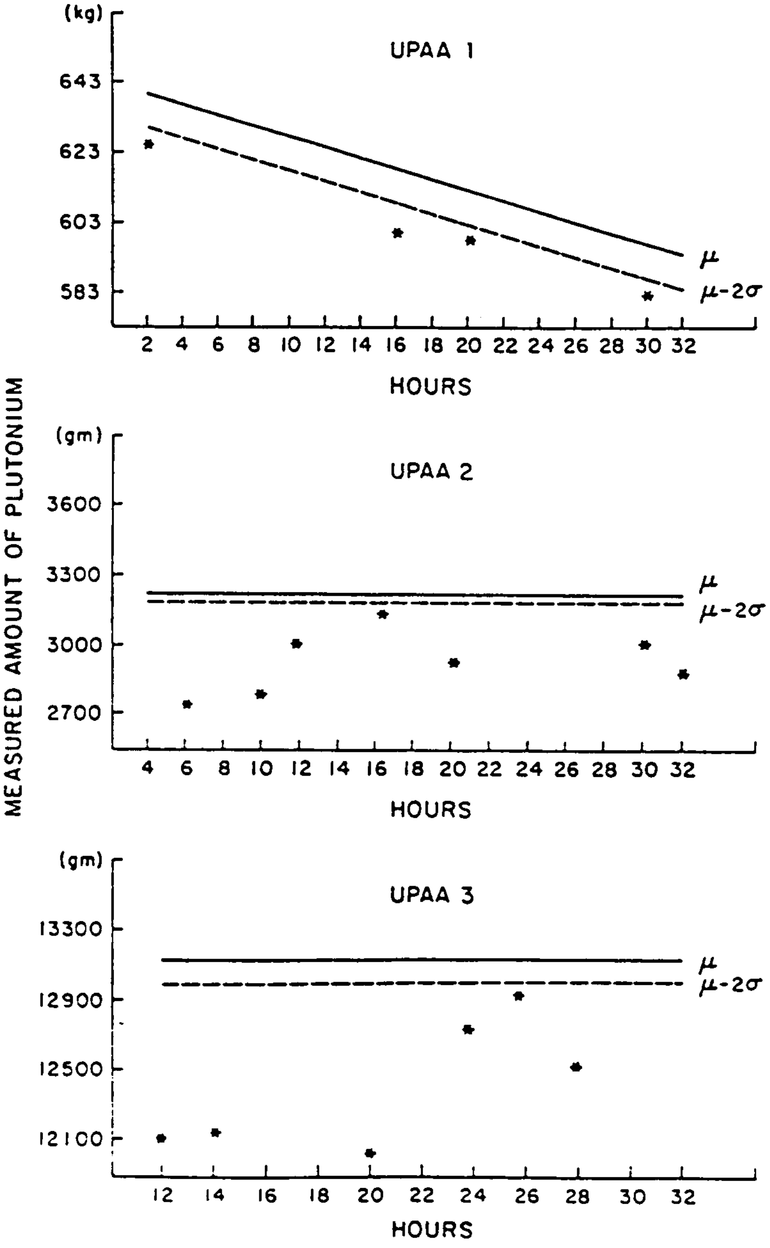

Figure 10. Prolonged Diversion Control Charts

the instrument accuracy improvement contributed significantly in the control chart analyses. Figure 11 presents the sensitivity case control chart results.

In the UPAA 1 control chart, the abrupt diversion remains undetected. Only one measurement exceeded the control limit. But this is an improvement in that false alarm incidents can be projected to be much lower for decreasing instrumentation error ranges.

The UPAA 2 and UPAA 3 control charts show less dramatic increases in measurement accuracy. The Figure 11 star symbols have shifted slightly higher in comparison to the Figure 9 star symbols before the diversions and slightly lower after the diversions.

An improvement of 25 percent in instrumentation error parameters represents a huge technological advance. To conduct another sensitivity analysis using, perhaps 50 percent improvements, seems to the author to be excessively unrealistic in that process instrumentation does have an upper limit on its efficiency.

Adjusting the flow variable spectrum to a smaller range would affect accountability results also. But since a default minimum and maximum flow-rate scheme was employed, a sensitivity study on the flow variables does not seem justified.

Expanding the number of UPAAs could be another factor

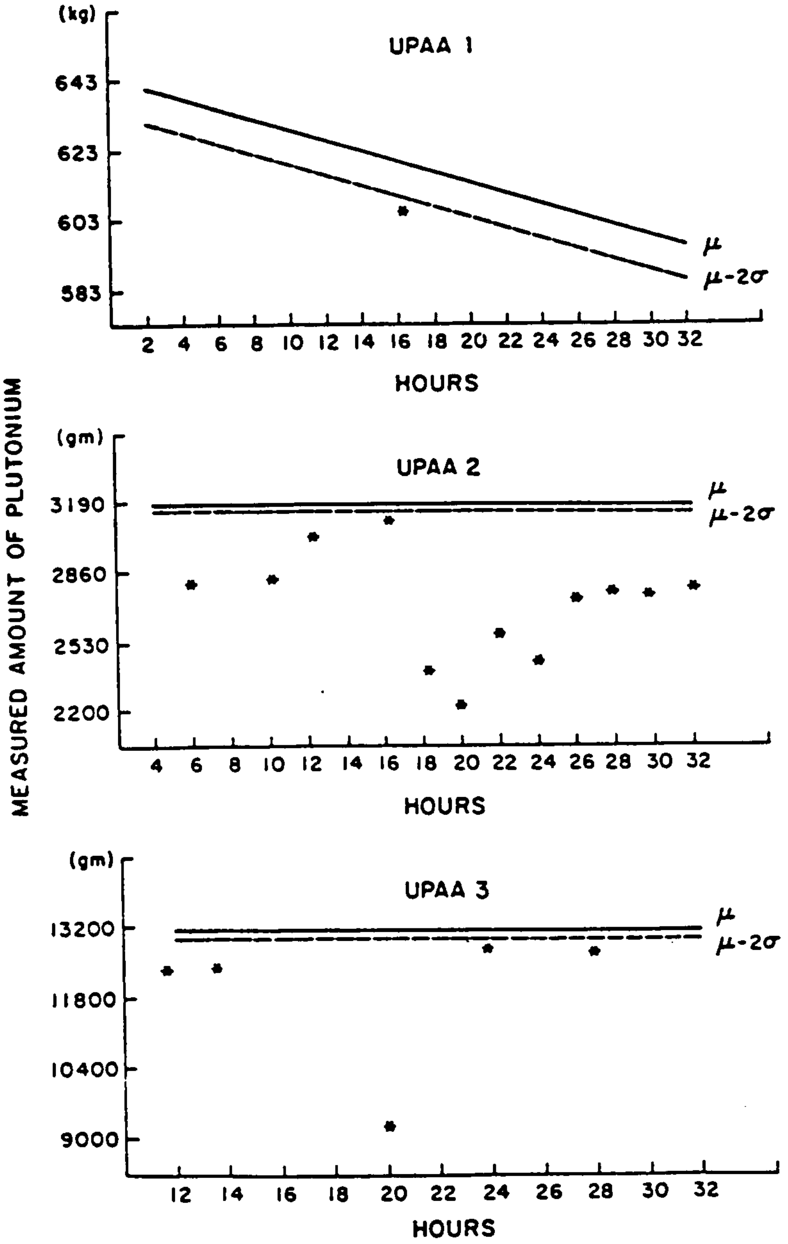

Figure 11. Sensitivity Case Control Charts

for sensitivity analysis. Specifically, splitting UPAA 3 into two processing areas would probably produce very favorable control chart results. This type of analysis was not undertaken because the LASL work[10] used only three UPAAs and projected that the use of more than three UPAAs would generate too much information for timely decisions to be made.

The next chapter offers some conclusions based on the results derived from the control charts.

CHAPTER SEVEN - CONCLUSIONS

Adequate material accountability standards cannot be met by the present COPRECAL facility design and UPAA strategy. The results of the control charts in all cases point to the need for a substantial reduction, perhaps by as much as a factor of one thousand, in the size of the feed-blend tanks. Furthermore, the facility is designed to operate with a plutonium throughput which is too high considering that only three UPAAs are used. Even drastic improvements in the efficiency of the process instrumentation do not yield the level of accuracy needed to effectively monitor for plutonium theft.

If the COPRECAL process is to become part of the back end of the nuclear fuel cycle and abide by present non-proliferation regulations, it either must undergo design changes or be scrutinized by a more suitable safeguards philosophy. A combination of the two is the most likely alternative.

CHAPTER EIGHT - RECOMMENDATIONS

Should the Reagan Administration adopt nuclear fuel reprocessing as an alterantive to the spent-fuel storage problem, the COPRECAL process can become a reality. If so, design changes to the process will be imminent.

The model's use of event codes in filling the process systems makes it very amenable to alterations in the capacities of the process components. Output mechanics can accommodate any number of different material accountability strategies, including the use of more unit process areas. Flow and instrument variables are treated in separate subprograms by the model and can handle a wide variety of input readily.

As more technical information is gathered on the plant specifics, the model should be updated to assimilate the changes. Output extracted from the model can then project new accountability prospects.

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APPENDIX A. VERIFICATION PROCEDURES

Many models use results generated by previous models for a verification procedure. The LASL GASP IV continuous model simulating the COPRECAL process produced output which can be compared to the author's model output to validate it. Table 5 summarizes the comparison of the models' batch outputs. The values represent actual amounts, not measured amounts, of in-process material. The result of the comparison shows good agreement between both models.

The second type of verification analysis involves proving that all plutonium input into the process line is accounted for at the end of simulation. The author's model terminates after 32 hours of simulating plant operations. Table 6 lists the model output data and results of hand calculations. Again, the values represent actual amounts of in-process material. The total amount of plutonium at the beginning of simulation is held in the feed-blend tank. This is compared to the total amount of plutonium in the process line at the end of simulation. The perfect agreement between the figures verifies the model logic integrity and accuracy. Model output data is good to nine significant figures.

Table 5. Batch Output Comparison

LOCATION	LASL MODEL OUTPUT (kg/batch)	AUTHOR'S MODEL OUTPUT (kg/batch)
Primary Filter Batch-Transfer Container	3.25	3.28
Reduction-Stabilization Batch-Transfer Container	3.23	3.25
Product	3.21	3.22

Table 6. Summary of Verification Output

LOCATION	PLUTONIUM AMOUNT (gm)	COMMENTS
Feed-blend Tank	0.645643031 D + 06	Beginning of Simulation Total
Feed-blend Tank	0.594685866 D + 06	End of Simulation Component Totals
Precipitator	0.939175924 D + 03	
Calciners	0.137155754 D + 04	
Primary Filter	0.506400284 D + 03	
Secondary Filter	0.479995028 D + 03	
Reduction Container	0.248013347 D + 04	
Batch Container	0.000000000	
Red.-Stbl. Area 1	0.320328212 D + 04	
Red.-Stbl. Area 1 Primary Filter	0.000000000	
Final Filter	0.942828422 D + 02	
Red.-Stbl. Area 2	0.326345915 D + 04	
Red.-Stbl. Area 2 Primary Filter	0.000000000	
Final Filter	0.957137283 D + 02	
Red.-Stbl. Area 3	0.606106052 D + 03	
Red.-Stbl. Area 3 Primary Filter	0.259772275 D + 04	
Final Filter	0.846698074 D + 02	
Red.-Stbl. Area 4	0.318056838 D + 04	
Red.-Stbl. Area 4 Primary Filter	0.000000000 D + 04	
Final Filter	0.611364855 D + 02	
Red.-Stbl. Batch Container 1	0.000000000	
Red.-Stbl. Batch Container 2	0.000000000	
Red.-Stbl. Batch Container 3	0.000000000	
Red.-Stbl. Batch Container 4	0.000000000	
Screen	0.000000000	
Screen Waste	0.320000000 D + 03	
Product	0.316299507 D + 05	
Waste Streams	0.466002106 D + 01	
	0.868626753 D + 00	
	0.899512908 D + 00	
	0.790592035 D + 00	
	0.592546299 D + 00	
	0.140000000 D + 02	
	0.140000000 D + 02	
	0.140000000 D + 02	
	0.645643031 D + 06	End of Simulation Sum Total