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TITLE: DEMONSTRATION OF NEAR-REAL-TIME ACCOUNTING AT THE AGNS BARNWELL PLANT

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DEMONSTRATION OF NEAR-REAL-TIME ACCOUNTING AT THE AGNS BARNWELL PLANT*

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Abstract

Near-real-time nuclear materials accounting is being demonstrated in a series of experiments at the Allied-General Nuclear Services Barnwell Nuclear Fuels Plant. Each experiment consists of operating the second and third plutonium cycles continuously for 1 week using uranium solutions. Process data are collected in near-real time by the AGNS computerized nuclear materials control and accounting system, and the data are analyzed for diversion using decision analysis techniques developed and implemented by Los Alamos. Although the measurement system primarily consists of process control measurements that have not been optimized for near-real-time accounting, the results of a series of diversion tests show that diversion and unexpected losses from the process area can be detected.

1. Introduction

An essential step in the development of safeguards technologies is the demonstration that they are both cost effective and operationally acceptable in actual nuclear facilities. A promising safeguards technique that currently is being demonstrated in an existing reprocessing plant is near-real-time accounting (NRTA).[1-3] By this technique, an updated book inventory is maintained almost in real time by combining automated data-base and inventory-control methods with on-line measurements of the net transfers (inputs minus outputs) of nuclear material across selected unit-process accounting areas (UPAAs). Frequently, the inventory in process equipment, as indicated by the computerized book inventory, is verified using available process measurements and engineering estimates.

In principle, the advantages of the NRTA technique are that it provides sensitive, timely, and localized detection of diversion or unexpected and unmeasured losses of nuclear material from the process area of a nuclear facility. This is accomplished by collecting and analyzing data from existing or, in some cases, from upgraded process instrumentation. In many respects, the objectives and techniques of NRTA parallel those of improved process control, making it attractive to plant operators.

It is these advantages and the operational compatibility of NRTA that are currently being demonstrated in a series of experiments at the Allied-General Nuclear Services (AGNS) Barnwell Nuclear Fuels Reprocessing Plant. Beginning in 1976, the AGNS Barnwell plant was adopted by the Los Alamos Safeguards Systems Group as the baseline facility for a series of studies to develop NRTA systems for reprocessing plants.[1,2] These studies showed through the use of computerized modeling and simulation techniques that NRTA could provide sensitive and timely detection of diversion from the chemical separations area of a reprocessing plant.

In these studies the plutonium purification process (Fig. 1) received special attention because this is where decontaminated plutonium solutions would be processed to the final, concentrated plutonium-nitrate product. A reference NRTA strategy was developed that considered the plutonium purification process as a separate unit-process accounting area.

In 1977 AGNS, under the sponsorship of DOE, began the development and testing of a Computerized Nuclear Material Control and Accounting System (CNMCAS).[3] Initial work on CNMCAS involved the entire chemical separations line and focused on computerization of measurement, measurement control, and accounting procedures for "conventional" accounting. ("Conventional" accounting is the measurement of inputs and outputs for a materials balance area, coupled with periodic cleanout and physical inventory to close the materials balance.)

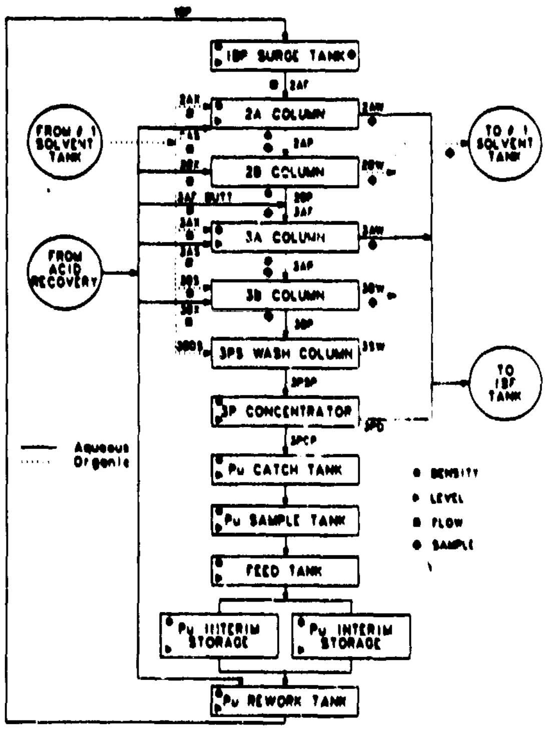

Fig. 1. AGNS minirun block diagram.

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As on-line measurement and computer capabilities improved, AGNS began to experiment using routine measurements of process variables to estimate the quantity of material in process. These experiments were initially conducted for the entire process, but by 1980 reduced funding required AGNS to find a less costly mode of testing. Because of the widespread and continuing interest in computerized nuclear materials control and near-real-time accounting methods, the minirun concept was devised. This concept involves cycling uranium solutions through the plutonium purification process in a closed loop, supported only by the solvent recycle system, the acid recovery/condensate recycle systems, and the process off-gas system.

2. Minirun Description*

The minirun cycle (Fig. 1) consists of four pulsed-column contactors (2A, 2B, 3A, and 3B); one packed column (3PS); a product evaporator (3P concentrator); and seven product, feed, and blending tanks. Support systems include aqueous waste tanks, a waste evaporator and acid fractionator, a solvent surge and recycle tank, an off-gas system, and associated process and chemical distribution systems. This represents a good cross section of routinely used plant equipment for development of improved materials control and accounting methods. A modified Purex solvent-extraction flowsheet is used with unirradiated natural uranium in place of plutonium for the tests.

The normal starting inventory for each run was 400-500 kg of uranium. After attaining equilibrium, a "process holdup" (pulsed columns, lines, product evaporator) of about 70-75 kg was observed, with the remaining material distributed among product tanks. Waste losses from the system varied from run to run, averaging something on the order of 100 kg for each run.

Five minirun experiments were performed at AGNS during 1980 with participation by Los Alamos National Laboratory to demonstrate NRTA and Oak Ridge National Laboratory to demonstrate process monitoring. Table I summarizes the purpose and activities of each of the five runs.

3. Demonstration of NRTA at AGNS

Demonstration of NRTA at AGNS required the development of a pulsed-column inventory estimator, formulation of materials accounting strategies, development of computer programs to acquire and analyze the measurement data, and implementation of these programs on the AGNS computer system.

Measurements.

Measurement data from the AGNS process-control instrumentation, including estimates of random and systematic measurement uncertainties, were received in a data file (ARANGE) every hour.

*See companion paper by J. M. Crawford, J. H. Ellis, and M. H. Ehinger (AGNS) entitled "Near Real-Time Accounting in a Reprocessing Facility Using In-Process Inventory Estimation."

TABLE I

1980 MINIRUN DESCRIPTION

No.	Purpose	Special Test Activities
1	Shakedown	Program debugging; Column inventory experiment
2	Shakedown/baseline run	Accumulation of steady-state data
3	Announced diversions (all parties informed of diversion timing)	17 abrupt (batch) diversions ranging from 5 to 0.25 kg of uranium; 4 protracted removals, each of 16-h duration with rates from 0.2 to 0.6 kg/h of uranium
4	Unannounced diversions (accounting personnel not informed of timing)	3 abrupt removals of 0.3, 0.5, and 1.2 kg of uranium; 2 protracted removals of 0.5 kg/h of uranium, each ∿12-h duration
5	DOE contractor demonstration	1 abrupt removal of 0.25 kg of uranium; 1 protracted removal of 0.85 kg/h of uranium for 16 h; Column inventory experiment

Sample data from the analytical laboratory were added to the ARANGE file as they became available.

The measurement data in the ARANGE file included volumes and concentrations for process tanks and flow rates and concentrations for process streams. The measurement types and locations are shown in Fig. 1. The level and density in each of the process tanks were measured using dip-tube manometers. The uranium concentrations in tanks were calculated from density, temperature, and free-acid measurements. The uranium concentrations of the "B" column organic product streams were also determined from on-line density measurements. Samples were taken periodically for chemical analysis from the 1BF surge tank, the column product streams, the waste streams, the solvent feed tank, and all feed and product batches. Flow rates of all organic and aqueous inlet streams were measured by flowmeters. The 2AF flow rate was measured using a metering headpot.

Analysis Programs.

Three computer programs (RADAR, FUNNEL, and DECANAL) were implemented at AGNS for analyzing minirun measurement data. RADAR is a utility code that reads the measured data from ARANGE and performs minimal formatting and data checking. RADAR then writes the input measurement data file for the FUNNEL program. FUNNEL is the executive program that forms materials balances. It was written specifically for the AGNS minirun process. The program allows the user to select for analysis data spanning particular time periods and to select any of several unit-process accounting areas (UPAAs) covering different areas of the process. FUNNEL calculates estimates of the pulsed-column in-process inventory, checks for uncovered dip tubes, flags unreasonable measured values, and tracks multiple batch transfers. The FUNNEL code also builds tables of the raw measurement data. These tables are used to identify and explain anomalies, such as plugged probes or other faulty measurements. For a specified UPAA and time period, the FUNNEL program combines the raw measured values to calculate net transfers, in-process inventories and their statistical uncertainties, and transmits them to the decision analysis (DECANAL) package for analysis using decision analysis methods.[4,5] These analysis methods are incorporated in the computer program DECANAL that calculates sufficient statistics containing all accounting information, sets decision thresholds, and compares these statistics to the thresholds in testing for diversion. The DECANAL output includes various graphical displays, such as alarm charts that indicate the likelihood and location of diversion, and plots of various statistics that estimate the amount of diversion.

Data from each UPAA were examined by DECANAL using a two-step scan-search procedure. In the scan mode, materials balance and cusum plots were produced for selected time intervals, along with tables and plots of measurement data from selected instruments. These data were scanned for evidence of probable outliers or trends. If significant losses of uranium were indicated, the data were searched and an alarm chart was generated. In the search mode, the most significant sequence of materials balances was identified, and the amount, time, and location of the apparent loss were determined.

Pulsed Column Inventory Estimation.

Under normal process conditions it is not possible (or at least not very convenient) to measure the in-process inventory of nuclear material in the pulsed columns. However, estimates of the in-process inventory can be obtained if flow-rate and concentration measurements are available on the column inlet and outlet streams.

The systems studies of near-real-time accounting[1,2] showed that estimates of the AGNS column inventories to 10% or better should be adequate for sensitive detection of losses. Under the sponsorship of Los Alamos, with participation by AGNS, General Atomic Company, Iowa State University, and Clemson University, techniques for estimating the inventory in the pulsed-column contactors were developed.[6,7]

Flow rates of all inlet streams are monitored to control the columns. For improved control and for NRTA, the concentrations of nuclear materials in the feed, product, and waste streams should also be measured. These measurements can be used to estimate the in-process inventory of nuclear materials in the columns. The form of the estimator is given by

$$H = H_f C_f + H_p C_p + H_w C_w \quad , \tag{1}$$

where H is the total column inventory and C_f, C_p, and C_w are measured concentrations in feed, product, and waste streams; H_f, H_p, and H_w are constants determined experimentally and through engineering models for each pulsed column.

Experiments at AGNS during run numbers 1 and 5 indicate that the column inventory estimates are good to 5 to 25% for individual columns and to about 10% for the total uranium inventory in all four pulsed columns. These column inventory experiments consisted of draining the columns into holding tanks at the end of the minirun. The contents of the holding tanks were sampled and analyzed for uranium, and the measured uranium inventory was compared with the estimated inventory for each of the columns.

Accounting Strategies.

The definition of several UPAAs with overlapping boundaries was desirable and possible because at certain points in the process there were redundant measurements; for example, the 1BP tank drop-out rate and the 2AF stream headpot flow meter both measure the 2AF stream flow rate. Likewise, product solutions can be measured in the product catch tank, the product sample tank, and the product storage tanks. Materials-balance data from overlapping UPAAs and redundant measurements were very useful in detecting and localizing losses and in maintaining continuity when there were measurement problems.

The major UPAAs are:

1. Full Process UPAA - includes the entire closed loop of the plutonium purification process, as operated for the miniruns;
2. Column UPAA - isolates the columns into a single accounting area bounded by the 1BP tank and the 3PS concentrator;
3. 1BP surge tank UPAA - isolates the 1BP surge tank with the plutonium rework tank and the 2AF stream;
4. PPP UPAA - includes the columns and the 3PS concentrator with boundaries at the 1BP surge tank and the Pu catch tank (alternatively, the catch tank can be included in the UPAA, and the sample tank can be used for the output transfer measurement); and
5. Tank UPAA - isolates any single tank in the process as a separate UPAA.

4. Results

Sample NRTA results obtained during miniruns 3, 4, and 5 are shown in Figs. 2-5. Two data analysis statistics are included in the examples, the materials balance and the cusum. Each figure shows plots of the test statistic and the corresponding alarm chart. Each test statistic is plotted sequentially in time with 1σ error bars. The alarm chart is a point plot of initial time vs final time for each sequence of materials balance data that caused an alarm. Thus, the position of each point on the chart indicates the time period when each alarm occurred. The significance of each alarm is indicated by the plotting symbol. The letters A-G are used to indicate increasing significance.

Figure 2 is a materials balance and alarm chart for a static storage tank during minirun

3. A series of abrupt diversion tests (5.2, 2.6, 1.3, 0.65, and 0.26 kg) were conducted. The first and second diversions generated highly significant alarms. The other three removals did not generate any single point alarms because they were not statistically significant. The estimated amount of material lost in the first two diversions is 6.0 and 3.1 kg, respectively. The difference between the estimated loss and the true loss results from a difference in the chemical analysis of the diverted material and the on-line concentration measurements for the tank. Note that the variability of the data in Fig. 2 is relatively small, indicating that we may be underestimating the precision of measuring the tank volume or the concentration.

Figure 3 shows materials balance, cusum, and cusum alarm charts for a storage tank (305). The cusum alarm chart shows numerous, highly significant alarms representing long sequences of materials balance data. This indicates that a protracted diversion test started between 1700 and 1800 on 7/17/80 and ended between 0800 and 0900 on 7/18/80. We estimated that 4.1 kg of material was removed during this time. GNS

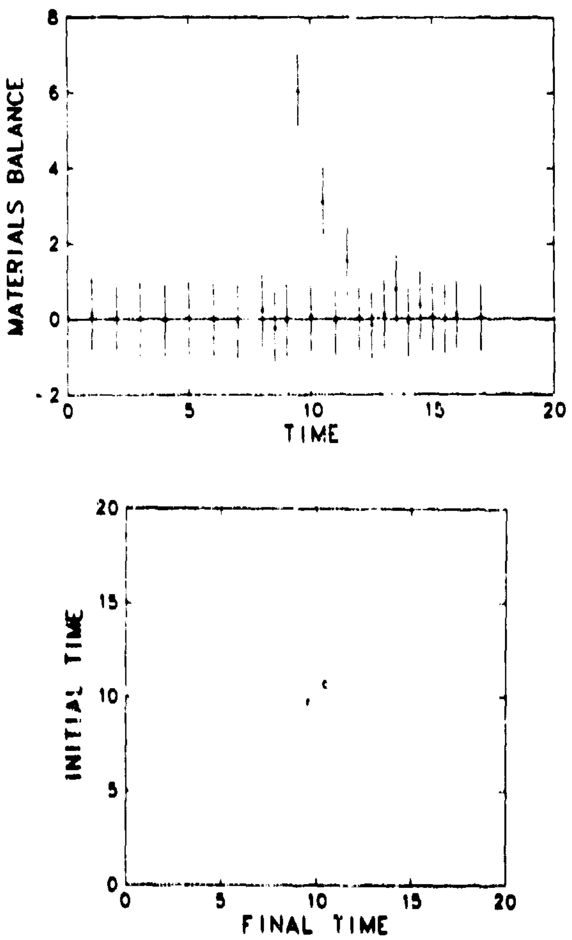

Fig. 2. Tank 305 (0000 7/17/80 - 1700 7/17/80): materials balance chart (upper), materials balance alarm chart (lower).

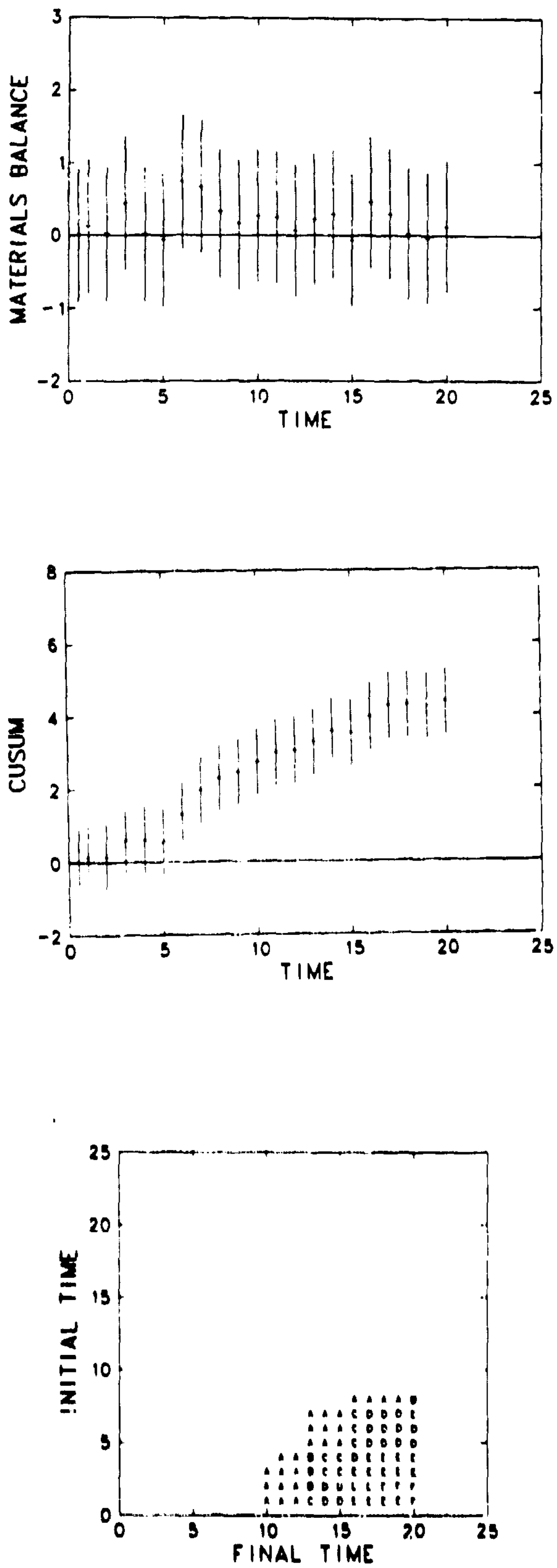

Fig. 3. Tank 305 (1500 7/17/80 - 1200 7/18/80): materials balance chart (upper), cusum chart (middle), and cusum alarm chart (lower).

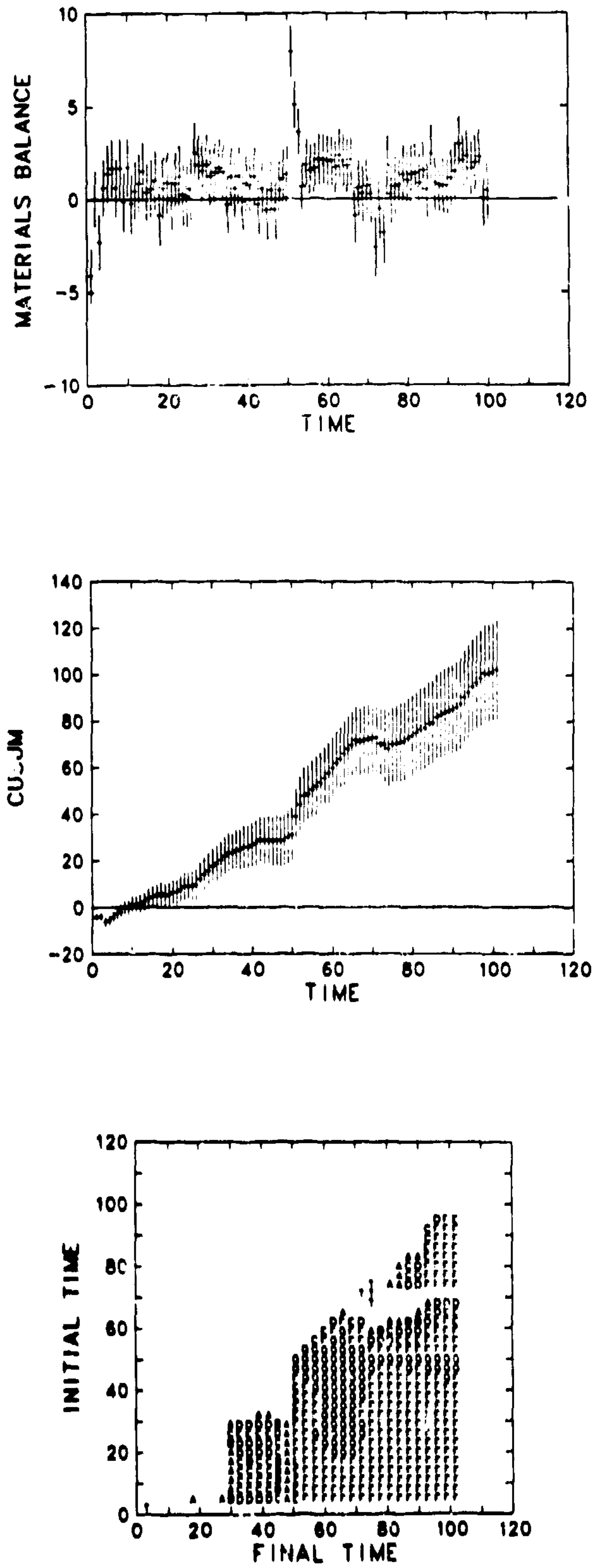

Fig. 4. Column UPAA (0700 9/4/80 - 1200 9/8/80): materials balance chart (upper), cusum chart (middle), and cusum alarm chart (lower).

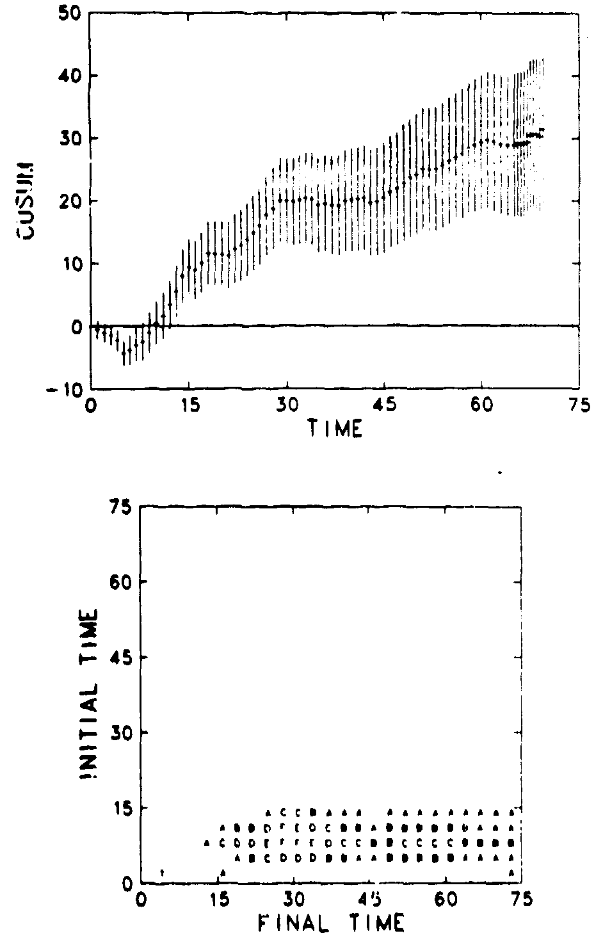

Fig. 5. Column UPAA (2000 11/18/80 - 1650 11/21/80): cusum chart (upper) and cusum alarm chart (lower).

records later showed that 3.6 kg of material was removed between 1750 on 7/17/80 and 1055 on 7/18/80.

Figure 4 shows materials balance, cusum, and cusum alarm charts for the column UPAA during a 100-hour time period of minirun 4. The only prominent feature in the materials balance chart comprises three balances ∿50 hours into the run. These balances result from an anomalously low uranium product concentration measurement. Three separate positive trends are apparent in the cusum, corresponding to two protracted diversion tests from intermediate column product streams (20 to 40 hours and 80 to 100 hours) and an unmeasured rapid loss of uranium to waste (50 to 70 hours). The alarm chart shows three clusters of alarms corresponding to the cusum trends. All three trends produced highly significant alarms.

Figure 5 shows a cusum and its corresponding alarm chart for the column UPAA during minirun 5. During this time period, some problems were experienced with the 2AF flowmeter; yet we were able to draw materials balances about this UPAA by using the 1BP tank dropout rate instead of the flowmeter to determine the input transfer to the UPAA. The cusum shows a significant trend, and the corresponding alarm chart indicates that the most significant sequence started at 5 hours (0000 11/19/80) and ends at 29 hours (0400 11/20/80). During that time we estimated that 24 kg of material was diverted. AGNS records later indicated that 21 kg of material was removed from the 1BP tank during the period 0000 on 11/19/81 through 0115 on 11/20/81.

5. Discussion

The results of the AGNS minirun experiments illustrate that NRTA can be implemented at a chemical reprocessing facility and can provide timely and sensitive information concerning the location and quantity of nuclear material within the process. The results also demonstrate the importance of having redundant measurements and overlapping UPAAs to isolate losses and circumvent measurement problems. These results are more striking if we remember that the majority of instruments used during these miniruns were originally installed for process control. Future minirun experiments at AGNS will incorporate on-line nondestructive measurements, such as x-ray fluorescence and absorption-edge densitometry, to measure concentrations in process and waste streams. Column inventory experiments are also planned for each run.

The following is a list of "lessons learned" from the 1980 miniruns.

- Near-real-time accounting for nuclear materials is sensitive to losses (both abrupt and protracted) from the process area of a large nuclear fuels reprocessing plant.
- Measurements of flow rates and concentrations are needed on process streams, including waste streams, that cross accounting area boundaries. Many of these measurements can be obtained from process control measurements made on adjacent process vessels. A few concentration measurements require the addition of NDA instruments on sample lines.
- In-process inventory measurements and estimates usually can be obtained from available process control data. These measurements in general need not be as accurate or precise and may be made less often than the stream measurements. Estimates that are satisfactory for NRTA can be made of the in-process inventory in pulsed columns. The presence of two-phase liquids must be considered for all process vessels.
- Overlapping UPAAs and redundant measurements are helpful for systems reliability and for localization and detection of losses.
- Analysis and display methods geared to ease of understanding and interpreting the data and the status of the process are necessary components of near-real-time accounting and process control systems.
- The reprocessing facility is an integrated whole, and the safeguards system must address the entire facility. Statements concerning the overall sensitivity of the safeguards system must recognize this fact.

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