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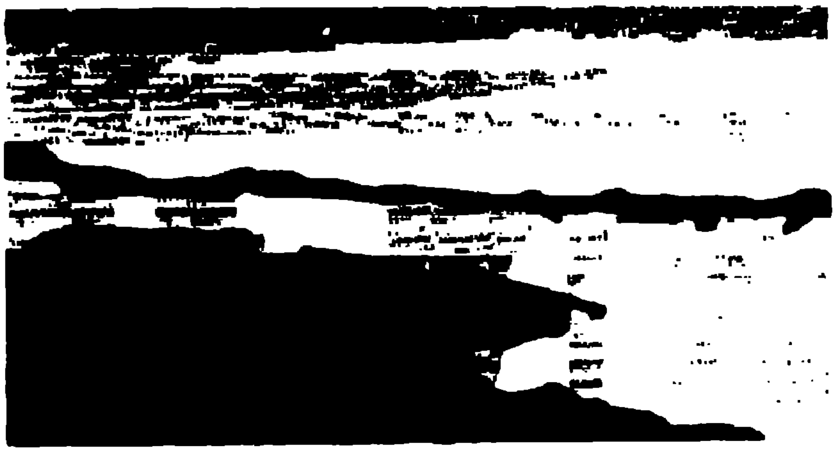

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Simulation and Analysis of Plutonium Reprocessing Plant Data*

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Abstract

It will be difficult for large throughput reprocessing plants to meet International Atomic Energy Agency (IAEA) detection goals for protracted diversion of plutonium by materials accounting alone. Therefore, the IAEA is considering supplementing traditional material balance analysis with analysis of solution monitoring data (frequent snapshots of such solution parameters as level, density, and temperature for all major process vessels). Analysis of solution monitoring data will enhance safeguards by improving anomaly detection and resolution, maintaining continuity of knowledge, and validating and improving measurement error models. However, there are costs associated with accessing and analyzing the data. To minimize these costs, analysis methods should be as complete as possible, simple to implement, and require little human effort. As a step toward that goal, we have implemented simple analysis methods for use in an "off-line" situation. These methods use solution level to recognize major tank activities, such as tank-to-tank transfers and sampling. In this paper, we describe their application to realistic simulated data (the methods were developed by using both real and simulated data), and we present some quantifiable benefits of solution monitoring.

1. Introduction

The idea of using process data from the facility operator for safeguards purposes has been advocated by some for a number of years.[1] For safeguards purposes, we define solution monitoring as the *essentially continuous monitoring of the level, density, and temperature of solutions in all tanks in the process that contain, or could contain, safeguards significant quantities of nuclear material.* These measurements should be authenticated and independently verified.

We have developed and evaluated methods for analyzing solution monitoring data under the following assumptions (Fig. 1):

- each tank is equipped with pressure-measurement dip tubes that provide a density (D) and level (L) measurement,
- temperature (T) is also measured;
- (L,D,T) *data are recorded frequently* (every 1-5 minutes) for all key tanks that contain or could contain a significant amount of Pu;
- the volume (V) *is obtained* from the L measurements via a tank calibration procedure; and
- the Pu concentration is measured, either in-line or periodically (every week or so) off-line.

This paper is organized as follows: Section 2 describes our approach to the simulation and analysis of solution monitoring data. Section 3 gives a brief description of our simulation and analysis software. Section 4 explains the quantifiable benefits of solution monitoring, and Section 5 is a summary.

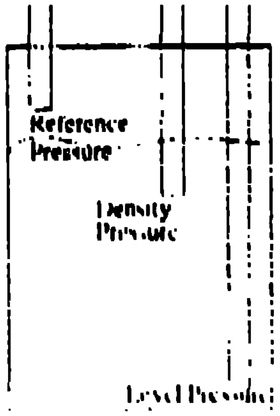

Fig. 1.
Fully instrumented tank with three dip tubes each of which measures pressure at a different height inside the tank. If solution level falls below the Density Pressure dip tube, no density measurement is available. The separation distance between the Density Pressure dip tube and the Level Pressure dip tube is known.

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2. Solution Monitoring

2.1 Data Analyzed

We simulated 18 days of continuous monitoring in a hypothetical PUREX reprocessing plant (simulation details are in Section 3). We consider here only the first three tanks, to simplify the presentation. The first 25 hours of data for these tanks is plotted in Fig. 2, and a simplified process diagram is given in Fig. 3.

2.2 Approach

We have developed algorithms to analyze solution monitoring data and algorithms to implement computer-aided manual investigation to confirm some of our results. We have concentrated on finding and classifying all tank events that involve the movement of nuclear material, e.g., sampling and transfer events. The (L,D,T) data were recorded in every vessel at approximately 5-minute intervals. Our analysis assumes that the basic pressure readings have been collected and transformed to (L,D) and volume (V).

The main three algorithms, which are designed to (1) find all tank events, (2) classify all tank events, and (3) perform consistency checks on all tank events, are as follows:

- The **Events Finder** module identifies a significant change in an L measurement as an unspecified "event" and finds the approximate start and stop point of that "event."
- The **Events Classifier** module classifies the events found by Events Finder by comparing the L behavior to a small library of recognized L behaviors. For example, sampling events should exhibit a modest drop in L, followed within about 30 minutes by a return to nearly the original L.
- The **Events Reconciler** module tries to reconcile each event with an accompanying event, either in the same tank (for sampling events) or in another tank (for transfers). For example, the reconciler will attempt to find the receiver tank for each "ship-to-tank" event. The volume or mass (mass = V • D) shipper receiver difference should not exceed some tunable threshold.

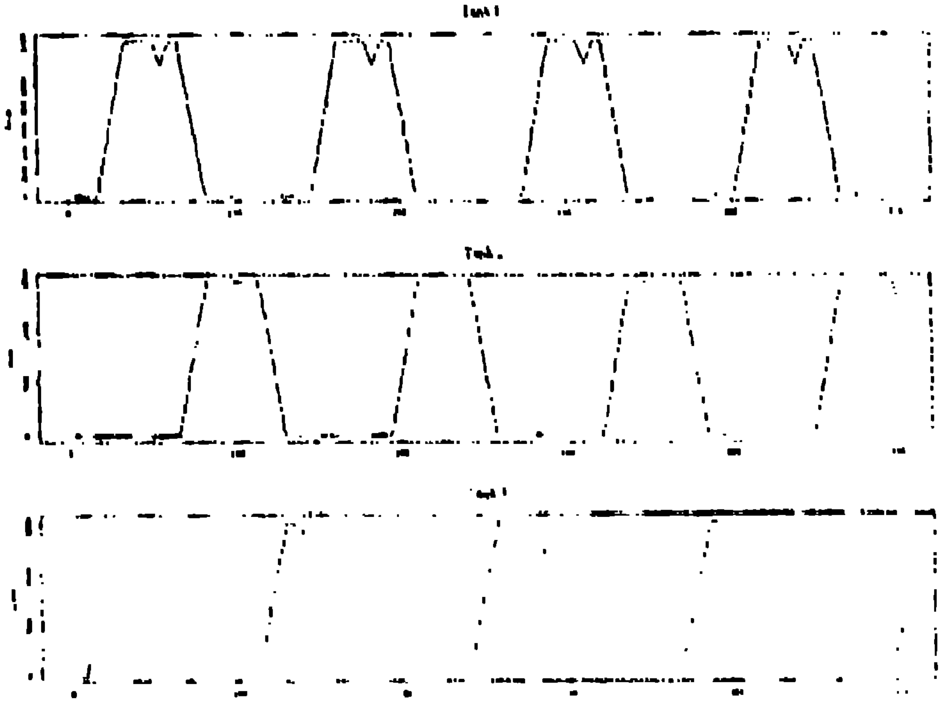

Fig. 2 The first 25 hours of L measurements from tanks 1, 2 and 3

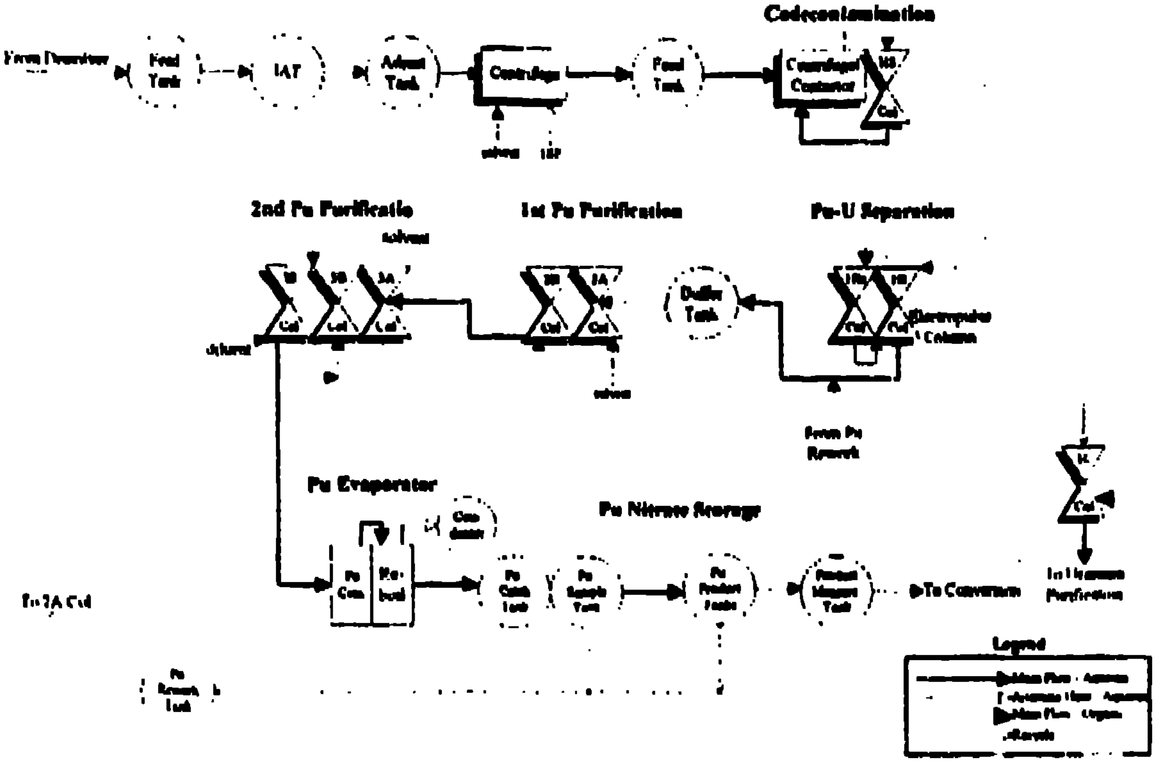

Fig. 3. Flow diagram of main process area at reprocessing plant.

We have written these modules to have both generic and facility-specific features. We use both generic "change-detection" methods and facility-specific threshold values for event durations and volume or mass losses.

2.2.1 The Events Finder Module

The Events Finder module is designed to find short term changes in level, such as would occur during a sampling or transfer event. The main steps followed by the current module are the following:

1. On the basis of previous value(s), forecast the current L value (the difference between the current value and the forecast is the current ERROR).
2. Compare the ERROR with user defined thresholds to determine whether the change is sufficient to signal an "event."
3. If an event occurs, determine the start time (and data point index) of the event.
4. Determine the stop time (and data point index) of the event.

We have other versions of the Events Finder module that calculate, in addition, a cumulative sum of ERRORS for events that occur more slowly. But because all of our legitimate events currently involve only abrupt changes, we do not present here any detail about these other versions. Instead, during each non-event data section, we apply a single final-initial check for any slow changes during that section. Both the start and stop times of an event may be determined by different criteria, e.g., successive L changes that are small or a significant change in slope. The primary output of Events Finder is an "events file" for each tank, which contains information about events (Table 1 shows an example for the first few Tank 1 events). Given a higher resolution plot, the output also includes well identified start and stop indices for each event.

2.2.2 The Events Classifier Module

The Events Classifier module attempts to recognize (classify) each event found by Event Finder by comparing the duration of the event and the change in level with what would be expected on the basis of historically observed changes or on process flow sheets for each kind of event for the same tank. For a given event to be correctly classified, there must be enough experience with (or data on) similar events for rules to be constructed to characterize the event. For example, transfers from Tank 1 to Tank 2 might be known to always involve approximately 6500 L, but the expected transfer-volume range around 6500 L should also be used in the rules. This module, then, is essentially a pattern classifier that uses an "expert system" approach based on historically observed events. Events currently recognized by Events Classifier are shipments, receipts, shipments to sample, receipts from sample, and unknown. "Shipment" and "receipt" refer to transfers to and from other tanks or to and from the plant. Events Classifier adds an "Event Class" column to the data set produced, as shown in Table 1. As our understanding evolves and a historical database of solution monitoring information becomes available, other events can be added to the Find and Classify modules,[5] such as mixing, sparging, evaporation, and chemical adjustments.

2.2.3 The Events Reconciler Module

The Events Reconciler module processes each event found by Events Finder and classified by Events Classifier and attempts to find another event with which it will be consistent, or reconciled. Reconciliation is based on user-defined allowed errors in masses (or volumes) shipped as compared to those received. It is assumed that these errors are related to how well we measure the levels, densities, and volumes. Transfers and sampling events can be reconciled on the basis of either mass or volume. To reconcile one event with another, the module must find that both took place within a specified time interval. Given appropriate assumptions and history, levels could also be used as a basis for reconciliation. If an event cannot be reconciled, it is flagged as such for investigation by the inspector. The return value from the Events Reconciler module for a given event may be True, False, or Possible. In Table 1, all events are reconciled, so the entry from the Events Reconciler module is True. ("Possible" would indicate that another event occurred within the specified time window but the masses or volumes of the two events did not agree within the specified limits—although they did agree within somewhat larger limits.) Whenever an event is reconciled, Events Reconciler also identifies the other event (and the other tank; see Other.tank column in Table 1) with which this event was reconciled and provides information on the volumes and/or masses transferred, so that records of differences, including cumulative differences, can be kept. Such information can be used for evaluation of tank calibrations and pipe holdup (pipe holdup is material that remains in the pipes that connect the tanks).

To check our results, we use interactive graphics to "manually" find and classify all tank events; we then compare those results with the results of Event.Find, Event.Classify, and Event.Reconcile.

3. Software Implementation and Functionality

Simulated L (or V) data are available from functions within our toolkit. More detailed simulated data that accurately depict D and T are available from the simulation code FacSim.

3.1 Program FacSim for Simulation

Our work was based on a design for a reprocessing plant having a throughput capacity of 5 metric tons of uranium per day. We considered only the part of the facility from the feed tank for the input accountability tank

TABLE 1. TABLE 1. The First 8 Events Found by Events Finder Module for Tank 1

Event. Number	Index. Start	Index. Stop	Time. Start	Time. Stop	Event. Class	Event. Reconcile	Status	Other. Tank
1	17	36	480.00	551.25	Receipt	True	3	0
2	50	56	603.75	626.25	Shipment	True	2	Sample
3	55	62	622.50	648.75	Rec from Sample	True	3	Sample
4	65	82	660.00	723.75	Shipment	True	2	2
5	145	164	960.00	1031.25	Receipt	True	1	0
6	178	184	1083.75	1106.25	Shipment	True	2	Sample
7	183	190	1102.50	1128.75	Rec from Sample	True	1	Sample
8	193	210	1140.00	1203.75	Shipment	True	2	2

(IAT) through the output accountability tank. This process area comprises several tanks, a codecontamination/separation cycle, two purification cycles, and an evaporator (see Fig. 3). Because the design has relatively few tanks and the output accountability tank is emptied frequently, the process area has a rather small in-process plutonium inventory for a facility having this throughput.

Operation of the design facility was simulated by means of the Safeguards Systems Group's program FacSim. FacSim is a continuous/discrete-event simulation program developed for evaluating process- and materials-accounting operations at facilities that handle nuclear material. It is facility-independent, written in C++, and operates on IBM PC compatible computers. We have previously reported on other FacSim applications (for example, see Refs. 2-4).

The operation of the facility is determined by a series of discrete events—namely, the initiation and termination of flows through pipes. Between these discrete events, the system evolves continuously in a way determined by a complex set of hydrodynamic and chemical processes. In the simulation it is assumed that mixing in tanks is efficient, so that a tank's output concentrations of solution constituents are the same as the tank's instantaneous average concentrations of these constituents. In addition, it is assumed that the output concentrations for pulsed columns are proportional to their instantaneous average concentrations, with proportionality constants that depend on a column's function. This assumption allows the evolution of the contents of process vessels to be described by systems of ordinary differential equations. Finally, it is assumed that a pipe's output concentrations are equal to its input concentrations at the time the material entered the pipe. This assumption introduces a time non-locality into the equations describing the system, but one that can be treated fairly simply by saving histories of differential equation solutions.

Anomaly detection in PUREX processes is complicated by the fact that fluid volumes often are not conserved in flows through pipes and through vessels. This is because steam/air jets used for transfers can increase/decrease the volume of fluid transferred, and because solution volumes are nonlinear functions of the quantity of solute. These effects are taken into account, to an adequate extent, in the simulation.

Operation of the design facility was simulated for a period of 18 days, beginning with preparation of the pulsed columns but no nuclear materials in the process. The evaporator inventory reached approximate equilibrium in less than 4 days. The last 10 days of the simulated operation were used for the solution-monitoring studies; during this period of "steady-state" operation, the total in-process plutonium inventory varies between about 100 and 160 kg of plutonium (depending on the phase of the input/output cycle), about 7 to 12 kg of which is in pipes.

3.2 S-PLUS Toolkit for Analysis

Our toolkit is written in S-PLUS, an object-oriented statistical and graphical programming language. We have working versions for both UNIX work stations and for PCs with Windows 3.1, 3.11, 95, or NT. Only the UNIX and Windows 3.11 versions have been tested. Because we originally planned to implement the toolkit in C++, we do have some C++ classes designed that could be used at some later stage. Our toolkit is "better than we need for in-house use" but not yet available for general use.[5] The "driver" menu, called roadmap(), is shown in Fig. 4 (the () notation indicates that roadmap() is an S-PLUS function). The roadmap() submenu item "specialized analyses" is shown in Fig. 5. We use the "srd" choice to estimate shipper-receiver differences (SRD) for each tank-to-tank transfer for the analyses given in Section 4. The three modules described in Section 2 are under the roadmap() submenu "preliminary analyses."

4. Quantifiable Benefits of Solution Monitoring

There are many proposed benefits of solution monitoring from a safeguards perspective,[1-6] but here we discuss only some of the **quantifiable benefits**.

1. partially validate measurement error models: if a large materials balance (MB) occurs, we do not want to question whether σ_{MB}—the theoretical standard deviation of the MB that includes all sources of measurement error—is understated. We show in Ref. 6 that solution monitoring can provide considerable assurance that measurement error models are acceptable; therefore, provided the variance propagation is done correctly, the estimate of σ_{MB} should be quite good;
2. monitor the quality of measurements;
3. identify all normal events affecting the solution occurring in a tank, such as mixing or sparging;
4. estimate and attempt to reduce bias in tank calibrations, thereby reducing σ_{MB}, which improves loss detection probability; and
5. estimate holdup in pipes between tanks.

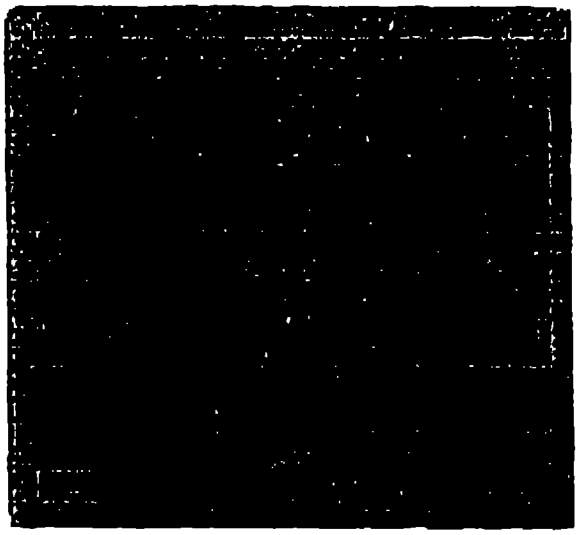

Fig. 4. Roadmap() function for menu access to S-PLUS functions.

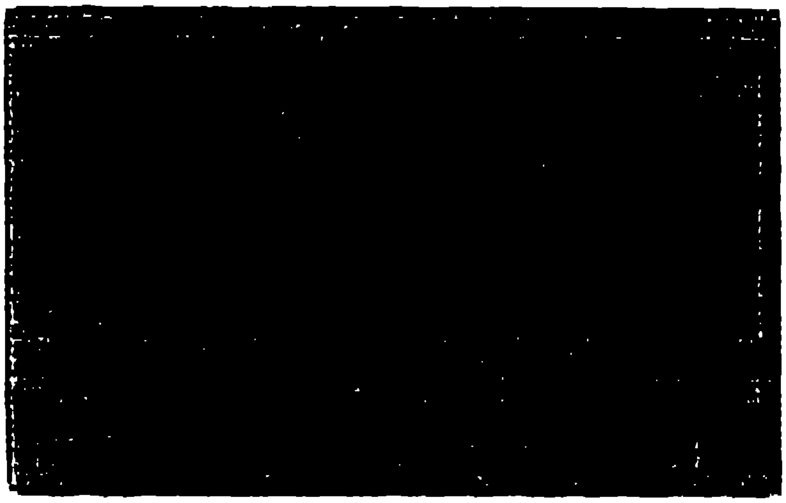

Fig. 5. The "specialized analyses" submenu of roadmap().

One benefit of identifying each tank-to-tank transfer is that each tank can be treated as a sub MBA (material balance area). That allows us to do several things, including (1) distinguish between a malfunction in the measurement equipment and an actual material loss and (2) do bias corrections.[2]

Loss-detection probability is higher if a loss occurs during a tank's wait mode than if it occurs during a tank's transfer mode, because during wait modes, we need detect a change only in volume or mass (systematic errors of the volume or mass measurement will cancel). We will describe loss detection during Tank 1 to Tank 2 transfers. For simplicity, we will consider monitoring only for volume loss (see Ref. 6 for further details).

Here is a typical error model for the Volume SRD during a Tank-1-to-Tank-2 transfer

$SRD_t = V_2 - V_1 =$

$$V_{1t} + [illegible]$$

where $V_{1t} = V_{2t}$ is the true volume shipped by Tank 1 and received by Tank 2, $V_{1t}\varepsilon_{S1}$ is the systematic volume error for Tank 1, $V_{1t}\varepsilon_{R1}$ is the random volume error) for Tank 2, and similarly for the Tank 2 errors. Note that we use a simplified model that assumes the absolute errors are proportional to the true volumes and that V_{1T} is the same for all shipments. We also assume that all measurement errors are approximately normally distributed.

The situation we consider is as follows: assume we have $n_{previous}$ shipments (at least 1) from Tank 1 to Tank 2 that are known to have zero true volume loss. Those previous shipments can be used to bias-correct all future shipments during the calibration period because ε_{VS1} and ε_{VS2} are assumed to be constant during the calibration period.

We denote the bias-corrected volume difference between $\tilde{V}_{1,i}$ and $\tilde{V}_{2,i}$ as $\tilde{SRD}_i$. The obvious bias correction is the average SRD during the $n_{previous}$ shipments, say $\overline{SRD}_{prev}$. If we write an error model for $\overline{SRD}_{prev}$,

$$\overline{SRD}_{prev} = V_{1T} \times ((\varepsilon_{VS1} - \varepsilon_{VS2}) + (\bar{\varepsilon}_{VR1} - \bar{\varepsilon}_{VR2})) \quad (2)$$

we see that if we let $\tilde{SRD}_i = SRD_i - \overline{SRD}_{prev}$, then the systematic errors would exactly cancel if $\bar{\varepsilon}_{VR1} - \bar{\varepsilon}_{VR2} = 0$. Unfortunately, we can expect only that $\bar{\varepsilon}_{VR1} - \bar{\varepsilon}_{VR2} \approx 0$ because $\bar{\varepsilon}_{VR1} - \bar{\varepsilon}_{VR2} \sim N(0, (\sigma^2_{VR1} + \sigma^2_{VR2})/n_{prev})$ (normal with mean 0 and specified variance). Therefore, the systematic errors only approximately cancel, leaving us with a new estimate of the systematic error variance equal to $(\sigma^2_{VR1} + \sigma^2_{VR2})/n_{prev}$. In conclusion, if we let $\tilde{SRD}_i = SRD_i - \overline{SRD}_{prev}$, then the random error variance will be the same as that for the non-bias-corrected SRD_i, and the systematic error variance will (perhaps surprisingly!) be determined by the random error variance and the number of previous shipments that are known to have zero volume change. Reference 7 has examples of bias corrections such as those given here (and our example is slightly different from that in Ref. 7).

Next, we will present a non-bias-corrected and a bias-corrected approach. Assume that we monitor the Tank-1-to-Tank-2 shipments for cumulative (over 1 year) volumes.

Case 1: No Bias Correction

It is simple to show that the cumulative (annual) measurement-error standard deviation is

$$\sigma_{\Delta V,cumulative} = V_{Total} \times \sqrt{(\sigma_S^2 + \sigma_R^2/n_{shipments})}\,, \quad (3)$$

where V_{Total} is the total true volume shipped during the year, σ_S^2 is the total systematic error variance (sum of volume measurement systematic error variances for both Tank 1 and Tank 2), and similarly for σ_R^2. For the Tank-1-to-Tank-2 transfers (450 transfers of 6500 L/yr having a Pu concentration of 3 g/L, for an annual throughput of approximately 8800 kg of Pu), Eq. (3) gives $\sigma_{\Delta V,cumulative} = 4141.0$ L. Given the Pu concentration of 3 g/L, the volume needed to accumulate 1 *SQ* is $V_{needed} = (SQ)/(conc) = (8)/(0.003) = 2667$ L. Therefore, the needed volume is only about 1.55 × $\sigma_{\Delta V,cumulative}$, which means we cannot detect a loss of this volume with sufficiently large probability—because to detect a loss of 1 SQ with a 0.95 detection probability and 0.05 false alarm probability, the needed volume is $5.15 \times \sigma_{\Delta V,cumulative}$ (we have applied the conservative Bonferroni correction to take multiple testing into account; we need a per-SRD false alarm probability of 0.05/450). Here, the detection probability is only 0.03.

Case 2: Bias Correction

To do a bias correction, the "catch" is that we must have at least one shipment from Tank 1 to Tank 2 for which we know there was no true volume change. For the method to be effective in reducing the cumulative measurement-error standard deviation, we assume 90 shipments (2[illegible] days of shipments and one shipment per 6.25 hours) from Tank 1 to Tank 2 that are known to have no true volume loss. Then Eq. (3) still applies, but with the systematic error equal to [illegible] rather than 2 × 0.01². Eq. (3) gives $\sigma_{\Delta V,cumulative}$ = [illegible] L. In this way, V_{needed} = 5.58 × $\sigma_{\Delta V,cumulative}$, and the detection probability is better than 0.95 if our MBA is only

Tank 1 and Tank 2. If the MBA includes Tank 3 as well, it is slightly more difficult to predict whether bias correction is a good idea, as we explain below.

More loss-detection results, for several loss scenarios, are provided in Ref. 5. As always, abrupt losses are easier to detect than protracted losses—and are easier still if we use solution monitoring data. One important issue that we will not address here is the role of material (holdup) in the pipes that connect the tanks. It is best for loss detection if holdup quickly increases to a quasi-equilibrium value and then fluctuates randomly around that value with some modest (10% relative) standard deviation. Another important issue is that tank transfers from Tank 2 to Tank 3 are more challenging because Tank 3 operates in batch receipt, continuous ship mode (B/C). For example, the measurement uncertainty for the amount received by Tank 3 must include the uncertainty in our estimate of the amount that Tank 3 shipped during its receipt from Tank 2. For our 18 days of simulated data, the standard deviation of the Tank-1-to-Tank-2 shipments is 7.9 (theoretical is 9.2), and for the Tank 2-to-Tank 3 shipments it is 10.4 (theoretical is 9.2 plus a hard-to-quantify amount to allow for our method of estimating the amount shipped by Tank 3 during its receipts). We would therefore use 3.5 × 7.9 = 27.7 L as our threshold volume SRD for transfers from Tank 1 to Tank 2 and 36.4 L for transfer from Tank 2 to Tank 3 to monitor future transfers (using 0.05/450 false alarm probability) for loss. These volume thresholds correspond to about 83 g and 109 g, respectively, of Pu. Using these thresholds, we can detect abrupt losses of 27.7, 55.4, and 83.1 L (one, two, and three times the threshold value) with probabilities of 0.50, 0.84, and 0.97, respectively.

5. Summary

We are strongly in favor of using solution monitoring data to enhance safeguards. We have begun to assemble analysis methods into a toolkit (intended for in-house use at this stage). Our simulation and analysis approach provided one bias-correction example that explained to what extent bias corrections might reduce σ_{MB} for each tank-to-tank transfer, and ultimately, for σ_{MB} for the entire MBA. If the MBA is Tank 1 and Tank 2, doing bias-corrected MBs is very attractive. If the MBA includes Tank 3, it is difficult to predict how well we can bias-correct the Tank-2-to-Tank-3 shipments because Tank 3 operates in B/C mode; the random error of our estimated volume SRD therefore increases, as does the systematic error of our bias-corrected SRD. Further, bias corrections are possible only if we know that some reasonable number of transfers have zero volume loss. There is at least one other barrier for reducing σ_{MB}: pipe holdup between tanks. Finally, our work does not contradict Ref. 8, which concludes that there is no advantage in terms of protracted loss detection in closing MBs frequently or around individual tanks. Our Tank-1-to-Tank-2 example in the "no-bias-correction" case could have reiterated the Ref. 8 result. The "bias-correction" case was not considered in Ref. 8.

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