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IN-PROCESS INVENTORY ESTIMATION FOR PULSED COLUMNS AND MIXER-SETTLERS*

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Abstract

Nuclear materials accounting and control in fuels reprocessing plants can be improved by near-real-time estimation of the nuclear materials inventory in solvent-extraction contactors. Techniques are being developed for the estimation of the in-process inventory in contactors. These techniques are derived from recent developments in chemical modeling of contactor systems, on-line measurements for materials accounting and control of the Purex process, and computer-based data acquisition and analysis methods.

1. Introduction

Reprocessing plants use the Purex process for the separation and purification of uranium and plutonium from spent nuclear fuels. Separation and purification are achieved with a series of solvent extraction contactors in which uranium and plutonium are selectively transferred between relatively immiscible countercurrent aqueous and organic streams. The three types of contactors that are generally in use are mixer-settlers, pulsed columns, and centrifugal contactors. For equal process throughput, mixer-settlers and pulsed columns generally contain a substantially larger inventory of nuclear materials than do centrifugal contactors.

Safeguards studies[1-6] indicate that the capability of proposed near-real-time ("dynamic") accounting methods to detect short-term losses of nuclear material in reprocessing plants can be limited by normal inventory variations in the contactors. Techniques for direct on-line measurement of contactor inventory are currently not available, and their development likely would entail substantial effort and cost. However, techniques are being developed to estimate the contactor inventory using currently available process monitoring, accounting, and control measurements.

The goal of this investigation is to develop a technique for estimating contactor inventory that is good to 5% or better. The safeguards studies show that this level of certainty will provide good short-term detection sensitivity. A promising technique that is now being developed[7,8] uses (1) detailed chemical models to predict the inventory for expected process conditions, (2) on-line process measurements and computer-based data acquisition systems to monitor fluctuations in the inventory during actual process operation, and (3) simplified linear models and computerized data-analysis methods to provide estimates of the inventory that can then be used to close materials balances in near-real time.

2. Contactor Models

Several mathematical models of solvent-extraction contactors have been developed in the U.S.[9-15] The most widely used model for solvent-extraction process development, SEPHIS, was originally developed to predict the transient and steady-state behavior of solvent-extraction contactors operating with a dilute Purex (15% TBP) LMFBR flow sheet; SEPHIS has since been modified for standard Purex (30% TBP) flow sheets.

The SEPHIS computer model performs a stage-wise, iterative calculation of the approach to steady state of the uranium, plutonium, and HNO_3 concentrations in a multistage contactor. The contactor is modeled as a series of ideal mixer-settler stages. Mass-transfer equilibrium is maintained at each stage, and the number of ideal stages is selected to achieve the desired overall separation efficiency.

Two contactor models are being developed under the current LASL safeguards study of contactor inventory. The first, being developed at Clemson University, is designed specifically for simulating mixer-settlers.[16] The second, previously developed at Iowa State University,[17,18] is designed specifically for simulating pulsed columns. This pulsed-column model is currently being updated and validated with data obtained under LASL sponsorship at Allied-General Nuclear Services (AGNS) and General Atomic (GA) Corporation.[19] Data suitable for model validation, especially for mixed uranium-plutonium recovery processes, are virtually nonexistent.

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Clemson Mixer-Settler Model

The mathematical basis for the Clemson mixer-settler model, called PUBG, is given in Refs. 16 and 20. This new model accounts for deviations from mass-transfer equilibrium, referred to as the "SEPHIS limit", that reduce the mass-transfer efficiency of the contactor and generally increase the nuclear materials inventory.

The mass-transfer rate R_i for chemical species i can be expressed in terms of the deviation of the species concentration in the mixer from its equilibrium value. The simplest such expression is given by

$$R_i = K_i A\left[M_i^o - M_i^o(eq)\right] \quad , \quad M_i^o(eq) = D_i M_i^a \quad . \qquad (1)$$

The superscripts "o" and "a" refer to the organic and aqueous phases, K_i is the mass-transfer coefficient for species i, A is the interfacial area between the aqueous and organic phases, $M_i(eq)$ is the concentration that would be obtained in the mixer if mass-transfer equilibrium were obtained, and D_i is the distribution coefficient for species i.

The PUBG model currently uses the equilibrium correlations of G. L. Richardson,[11] as modified by Watson and Rainey,[10] to calculate D_i. Contactor models in general are limited by the quality and availability of the equilibrium chemical data needed to calculate suitable distribution coefficients. Work is current v in progress to review the equilibrium correlations of Richardson and to revise them based on new chemical data.

Mass-transfer equilibrium requires that $M_i = M_i(eq)$. This condition can be approached by letting K_i or A approach large values forcing $(M_i - M_i(eq))$ to have small values. Few measurements of K_i have been made. A reasonable method of estimating both K_i and A is a variational method that matches model calculations with selected data from real mixer-settler contactors.

A detailed comparison of PUBG model calculations with measured data[9,10] on the coextraction of uranium and plutonium shows that the laboratory mixer-settler used to obtain the data was operated close to mass-transfer equilibrium. Table I shows the calculated and measured uranium and plutonium inventories.

More recently, Thompson and Shankle[13] reported reasonably complete sets of uranium recovery data for a D-type mixer-settler (uranium extraction from the aqueous to the organic phase) and an E-type mixer-settler (uranium stripping from the organic to the aqueous phase). In the case of the D mixer-settler, the experimental uranium inventory of 4990 g differed from the SEPHIS-calculated value of 4479 g by about 11%. Agreement between the PUBG calculated inventory and the measured inventory was obtained by adjusting the mass-transfer area A to a value of about 2, indicating significant deviations from mass-transfer equilibrium. This adjustment of the parameter A also markedly improved the agreement between the calculated and measured concentration profile.

Detailed comparison of measured uranium concentrations and inventories with calculations using PUBG are shown in Table II for the E mixer-settler. Table II also contains calculations using SEPHIS-MOD4 and TRANSIENTS.[13] The latter model accounts for deviations from mass-transfer equilibrium by adjusting the mass-transfer efficiency. The results again indicate a significant deviation from mass-transfer equilibrium.

Figure 1 shows the dependence of the inventory in the E mixer-settler on the mass-transfer area A. In practice, it might be possible to "calibrate" the model predictions for a mixer-settler by choosing an appropriate value for the A parameter.

Calculations indicate that the plutonium concentration in the waste stream is sensitive to the deviation from mass-transfer equilibrium. Table III shows the predicted dependence on the A parameter of the plutonium inventory and the aqueous waste stream concentration in a 15-stage extraction/scrub mixer-settler. The following procedure for mixer-settler inventory estimation might be feasible. First the inventory, and hence the value of the A parameter, would be determined experimentally under the expected

TABLE I

EXPERIMENTAL PLUTONIUM AND URANIUM INVENTORIES VERSUS SEPHIS-MOD4 AND PUBG (11-STAGE EXTRACTION/SCRUB MIXER-SETTLER)

	Plutonium Inventory Aqueous	Plutonium Inventory Organic	Uranium Inventory Aqueous	Uranium Inventory Organic
Experiment[9,10]	18.9	14.9	81.6	181.5
SEPHIS-MOD4[17]	21.3	16.6	80.5	171.1
PUBG[16] A = [illegible]	20.0	15.7	78.9	171.1

TABLE II

EXPERIMENTAL URANIUM CONCENTRATIONS AND INVENTORIES VERSUS PUBG, SEPHIS MOD4, AND TRANSIENTS (12-STAGE STRIPPING MIXER SETTLER)

Stage No.	Experimental Data[13]	PUBG[16] (A = 4.8)	PUBG (A = ∞)	SEPHIS-MOD4[17]	TRANSIENTS[13] (Eff = [illegible])
	Aqueous Phase Concentrations (g/l)[a]				
1	0.005	0	0	0	0
7	0.005	0.021	0	0	0.0[illegible]
8	0.005	0.110	0	0	0.2[illegible]
9	0.065	0.501	0.019	0.017	0.916
10	2.020	2.811	0.819	0.750	2.[illegible]
11	11.570	10.181	8.615	2.[illegible]	8.47[illegible]
12	21.500	21.360	21.360	21.115	21.[illegible]
	Organic Phase Concentrations (g/l)[a]				
1	0.001	0	0	0	0
7	0.033	0.001	0	0	0.011
8	0.018	0.00[illegible]	0	0	0.0[illegible]
9	0.180	0.037	0	0	0.100
10	0.185	0.194	0.006	0.0055	0.30[illegible]
11	0.731	0.944	0.271	0.354	0.934
12	Not Analyzed	3.394	2.879	3.015	
Total Uranium (g)	3532	3542	2060	2914	3680

[a]Concentrations less than 0.001 are given as 0

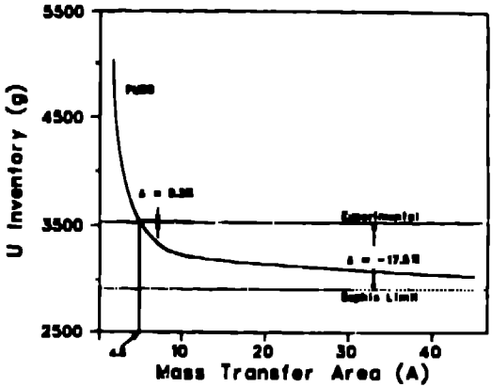

Fig. 1. The dependence of the E mixer-settler inventory on the mass-transfer area.

steady-state run conditions. Next a data table similar to Table III would be calculated and stored in a computerized data base. Then, during actual process operation, fluctuations in the mixer-settler inventory would be estimated from waste-stream concentration measurements. Such measurements of plutonium can be made with on-line α monitors. Mass balance correlations, including residence-time delays, could be used to improve the quality of the inventory estimates (see Sec. 4).

Iowa State Pulsed-Column Model

A computer model developed at Iowa State University for simulating pulsed column operation incorporates several improvements over SEPHIS.[17,18] Development of this model followed a critical review of the literature and an extensive survey of design and operating features of full-scale pulsed columns. Calculations are performed stage-wise using finite-difference equations that include the effects of reaction kinetics, nonequilibrium mass transfer, back-mixing, and new correlations of phase volumes with phase flow rates.

The mathematical basis for the model and the important chemical and hydrodynamic effects have been reviewed by Burkhart.[16] The relative importance of each effect depends on the specific column design and operating conditions, and the current experimental and theoretical descriptions of these effects generally is not adequate for accurate modeling of pulsed columns. For example, a recent experimental study at GA[19] of dispersed-phase holdup concluded that correlations given in the literature between holdup and observable process parameters did not agree with experiment.

Nevertheless, reasonably accurate model predictions of pulsed-column inventory can be obtained over the limited ranges of operating conditions encountered in industrial reprocessing plants, if semi-empirical correlations are used to describe the important effects. The exact form of these correlations for specific contactor systems must be determined experimentally, and work is continuing at GA and AGNS to provide the necessary data.

TABLE III

PLUTONIUM INVENTORIES AND AQUEOUS WASTE STREAM CONCENTRATIONS CALCULATED FOR DIFFERENT MASS-TRANSFER AREAS (15-STAGE EXTRACTION/SCRUB MIXER-SETTLER)

Model	Plutonium Inventory (g) Aqueous	Organic	Total	Plutonium Concentration (g/L) Aqueous Waste Stream
SEPHIS-MOD4	94.9	521.4	616.3	8.31×10^{-6}
PUREX: A = ∞	89.1	523.5	612.6	6.44×10^{-6}
A = 100	96.2	530.7	626.9	3.54×10^{-5}
A = 40	114.0	548.6	662.7	2.58×10^{-4}
A = 20	150.9	585.5	736.5	1.40×10^{-3}
A = 10	214.6	648.3	862.8	5.61×10^{-3}

Accurate column models are important for Purex process design, especially for the design of criticality controls in the reprocessing of FBR fuels. This is illustrated by the results reported in Ref. 17 in which three versions of the Iowa State model, ranging from a SEPHIS-type version to the most comprehensive version (one "real stage" per sieve plate with back-mixing and holdup correlations), were compared using input-output data from a Hanford partitioning column. Even though all three versions of the model used the same input-output data, the plutonium inventory calculated by the most sophisticated version was more than double that given by the SEPHIS-type version. Plutonium concentration profiles calculated by the comprehensive "real stage" model and the SEPHIS-type model are compared in Fig. 2.

Recently, Germak at AGNS obtained excellent pulsed-column data for uranium recovery using a 2-inch diameter extraction/scrub pulsed column. The AGNS pilot-scale columns are instrumented with a series of sample taps so that profiles of aqueous and organic uranium concentrations and the corresponding dispersed-phase holdup profile can be obtained. Table IV compares the AGNS concentration-profile data with the Iowa State (Burkhart) model and a SEPHIS-type model. The Iowa State model agrees better with the profile data than does the SEPHIS-type model. Table IV also contains total uranium inventories calculated with the SEPHIS and Burkhart models.

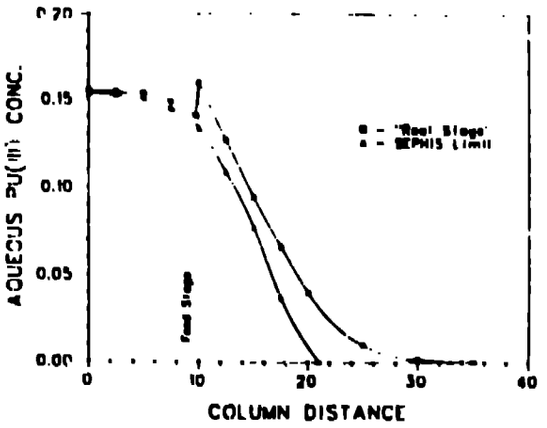

Fig. 2. Plutonium concentration profiles: "real-stage" vs SEPHIS-limit models.

TABLE IV

EXPERIMENTAL URANIUM CONCENTRATIONS VERSUS SEPHIS AND THE BURKHART MODEL (3-IN.-I.D. EXTRACTION/SCRUB PULSED COLUMN)

Distance (ft) From Column Bottom	Uranium Concentration (g/L)[a] SEPHIS Aq	SEPHIS Org	Experiment Aq	Experiment Org	Burkhart Model Aq	Burkhart Model Org
24 (scrub)	0	52.5	0	54.0	0	54.0
21	17.2	65.0	15.0	62.7	15.0	62.7
18	25.0	71.0	19.7	67.5	19.8	67.5
15 (feed)	25.0	71.0	20.0	68.5	21.4	69.0
12.8	10.0	32.5	9.0	38.0	9.1	39.2
12.2	1.5	6.2	1.5	20.1	1.5	17.6
11.2	0.4	1.0	0.4	8.0	0.4	7.0
9	0	0	0	1.0	0	1.0
7	0	0	0	0	0	0
Total U Inventory (g)	43.0	299.3	---	---	38.8	531.3

[b] Concentrations <1 g/L are given as 0.

The difference is substantial and is caused primarily by the dispersed-phase holdup and back-mixing effects that are included in the Burkhart model.

3. Contactor Measurements

Figure 3 is a schematic diagram of an extraction/scrub (A-type) pulsed column. A variety of process measurements are made in a modern reprocessing plant to monitor and control the solvent-extraction systems. The following list indicates process-control measurements that might be expected for an A-type column:

1. Pulse amplitude and frequency.
2. Primary aqueous-organic interface position (bottom).
3. Organic product liquid level and density (top).
4. Column hydrostatic pressure ("weight").
5. Extractant flow rate, temperature, and TBP concentration.
6. Scrub flow rate, temperature, and acid molarity.
7. Feed flow rate, temperature, density, and acid molarity.
8. Waste stream concentration.

These measurements would be supplemented by periodic visual checks and chemical analyses of samples collected from sample lines placed on selected streams and tanks.

In a modern industrial plant, this process information could be collected and made available in near-real time using computer-based data acquisition, storage, and retrieval methods. The AGNS computerized nuclear material control and accounting system (CNMCAS) is an example of the application of these techniques in a large-scale reprocessing plant.[21]

Additional stream concentration and flow rate measurements would be required for accurate near-real-time materials accounting. Some of these measurements could consist of upgrading existing process control measurements, such as using α monitors to measure plutonium concentrations in the waste streams.

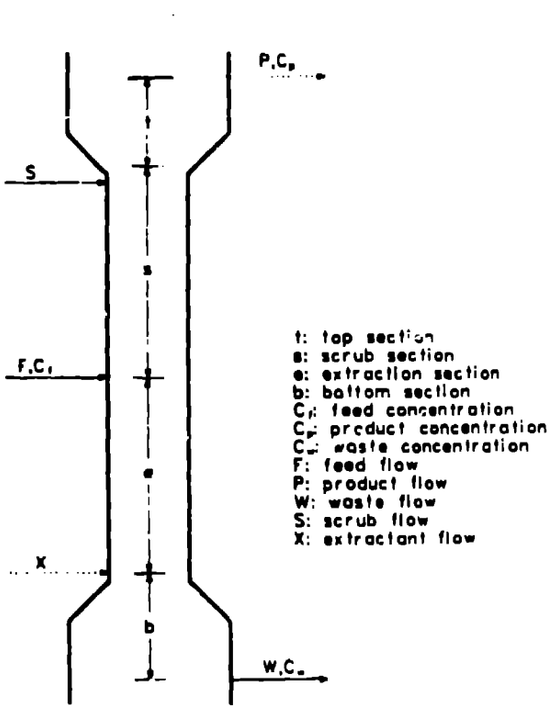

Fig. 3. Extraction/scrub pulsed column.

In-line flow measurements can be obtained using calibrated, metering headpots in place of the unmetered headpots normally used in liquid transfer systems. Various types of on-line flow meters have also been developed and are currently being evaluated by Oak Ridge for use in reprocessing plants.

For single-solute systems, i.e., uranium or plutonium, on-line liquid density and free-acid measurements can provide accurate estimates of the heavy-metal concentrations in concentrated product streams. For mixed uranium-plutonium streams, nondestructive measurement techniques supported by conventional wet chemistry methods can provide rapid, accurate analyses of process samples. Applicable NDA techniques include gamma-ray spectroscopy, x-ray absorption, and x-ray fluorescence.

A review of various on-line and at-line measurement techniques for potential application in reprocessing plants is given in App. L of Ref. 4, Vol. III. Wet chemistry methods are also reviewed in App. K of that reference.

4. Contactor Inventory Estimation

Modern systems identification and state estimation techniques potentially can provide the best possible estimates of contactor inventory. Ideally, a sequential analysis framework, such as the Kalman filter, would be used to combine the following elements: (1) a dynamic

contactor model that is valid over a wide range of operating conditions, including startup, shutdown, and upset transients; (2) a sufficient set of process observables and corresponding measurements, including high-quality real-time measurements of contactor inputs and outputs; (3) the statistics of both the process and the measurements; and (4) on-line computer data acquisition and analysis techniques.[20-22]

The full development of dynamic state estimation techniques is a future goal for on-line materials accounting. A more restricted approach is considered here for near-term applicability. We refer to this restricted state estimation scheme as "reduced-order" dynamic state estimation because, although the same three elements (models, measurements, and statistics) are required for its implementation, the scope of the required input information is restricted to a currently practicable level. The penalty is that the estimation model only applies over a limited range of near-steady-state run conditions.

An example of this approach is given in Figs. 4 and 5 for the A-type pulsed column of Fig. 3. Figure 4 lists the assumptions and the basic equations. The assumption that phase volumes are proportional to phase flow rates is probably the poorest and therefore the most restrictive. This assumption can be relaxed if better phase volume-flow rate correlations are available.[18]

The linear inventory estimator is based on first-order perturbations about an expected steady-state value. The steady-state inventory value is calculated for the expected run conditions using a detailed chemical model that has been validated experimentally for the particular contactor system. Alternatively, experiments can be performed directly to determine the expected inventory by bringing the contactor to steady state and then draining the contents to holding tanks for measurement.

The linear state equations for the estimator are also given in Fig. 4. The first equation is the materials balance relating the column inventory (H) at time k + 1 to the inventory at time k. The remaining terms in the materials balance equation are the measured transfers of material across the column in the feed, product, and waste streams and any unmeasured losses (M). The second state equation describes any postulated loss mechanism by choosing the specific form of the function ϕ.

If the transfers and inventories are measured and the measurement error statistics are known, a Kalman filter algorithm can be used to provide estimates of H at each time k. This approach has been developed and shown to be effective in the analysis of near-real-time accounting data.[23,24]

Because we assume that the transfers of nuclear material across the column are measured and the measurement error statistics (including random and systematic error components) are known, it only remains to find measured values and error statistics for the column inventory. Possible inventory measurement equations are given in Figs. 4 and 5. They are linear equations for the inventories in the organic and

Pulsed Column Inventory Linear Estimator

Assumptions

1. The column is operating near a steady-state operating point.
2. The column inventory near the operating point is linear in the concentrations and flow rates.
3. Phase volumes are proportional to phase flow rates near the operating point.
4. Concentration and flow rate measurements are available in near-real time.
5. The column inventory at the nominal operating point has been previously determined from chemical model calculations and calibration experiments.

State Equations

Mass Balance:

$$H(k+1)=H(k) + (\bar{C}_f F - \bar{C}_p P - \bar{C}_w W)\Delta t(k,k+1) - M(k,k+1)$$

Loss: $M(k+1) = \phi\ (k+1/k)\ M(k)$

Measurement Equations

Total Inventory: $h(k) = h^o(k) + h^a(k)$

Organic Phase:

$$h^o(k) = h^o_T(k) + h^o_S(k) + h^o_E(k) + h^o_B(k)$$

Aqueous Phase:

$$h^a(k) = h^a_T(k) + h^a_S(k) + h^a_E(k) + h^a_B(k)$$

Fig. 4.

"A" Column Measurement Equations*

Top Section

$$h^o_T(k) = h^o_{To}\left[\frac{C_o(k)}{C_{oo}} + \frac{V_T(k)}{V_{To}} - 1\right]$$

$$h^a_T(k) = h^a_{To}$$

Bottom Section

$$h^o_B(k) = h^o_{Bo}$$

$$h^a_B(k) = h^a_{Bo}\left[\frac{C_a(k)}{C_{ao}} + \frac{V_B(k)}{V_{Bo}} - 1\right]$$

Scrub Section

$$h^o_S(k) = h^o_{So}\left[\frac{C_o(k)}{C_{oo}} + \left(\frac{S}{X+S}\right)_o\left(\frac{X(k)}{X_o} - \frac{S(k)}{S_o}\right)\right]$$

$$h^a_S(k) = h^a_{So}\left[\frac{C_a(k)}{C_{ao}} + \left(\frac{X}{X+S}\right)_o\left(\frac{S(k)}{S_o} - \frac{X(k)}{X_o}\right)\right]$$

Extraction Section

$$h^o_E(k) = h^o_{Eo}\left[\frac{C_o(k)}{C_{oo}} + \left(\frac{S+F}{X+S+F}\right)_o\left(\frac{X(k)}{X_o} - \frac{S(k)+F(k)}{S_o+F_o}\right)\right]$$

$$h^a_E(k) = h^a_{Eo}\left[\frac{C_a(k)}{C_{ao}} + \left(\frac{X}{X+S+F}\right)_o\left(\frac{S(k)+F(k)}{S_o+F_o} - \frac{X(k)}{X_o}\right)\right]$$

*The subscript o denotes nominal values at the operating point; V_T and V_B denote phase disengagement volumes at the top and bottom ends of the column, respectively.

Fig. 5.

aqueous phases of the four sections of the column: top, bottom, scrub, and extraction. The inventory in each section is given by a first-order Taylor series expansion about the steady-state inventory. The first-order terms depend on ratios of the measured concentrations, flows, and volumes. Because the error statistics of these measurements are known, the statistics of the inventory measurements can be calculated.

Thus, a Kalman filter algorithm for estimating the column inventory is completely specified, but there are certain complications. Systematic errors will correlate all measurements of a particular type in time, and the column inventory and transfer measurements will be strongly correlated. These correlations must be included in the filter for good results. Furthermore, the steady-state inventory values appear in the inventory measurement equations as "calibration constants," and uncertainties in these calibration constants must be included as additional sources of systematic error. The decision analysis techniques[24] previously developed for analyzing near-real-time accounting data can treat the systematic errors and measurement correlations described above.

Computer simulations of the contactor inventory estimation technique are currently in progress, and the technique will be evaluated at AGNS during future cold-test uranium runs.

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