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SIMULATION OF NUCLEAR FUEL REPROCESSING FOR SAFEGUARDS

by

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Abstract

For safeguarding the chemical process area of future reprocessing plants the near-real-time material accountancy (NRTMA) method might be applied. Experimental data are not yet available for testing the capability of the NRTMA method but can be simulated using a digital computer. This report describes the mathematical modeling of the Pu-bearing components of reprocessing plants and presents first results obtained by simulation models.

Simulation der Wiederaufarbeitung von Kernbrennstoff für Zwecke der Kernmaterialüberwachung

Zusammenfassung

Im chemischen Prozeßbereich künftiger Wiederaufarbeitungsanlagen könnte zur Kernmaterialüberwachung die Methode der sogenannten "Quasi-Echtzeit-Bilanzierung" eingesetzt werden. Derzeitig sind zum Testen der Leistungsfähigkeit dieser Methode keine experimentellen Daten verfügbar; jedoch können solche Daten mittels digitaler Rechner simuliert werden. Dieser Bericht beschreibt die mathematische Modellbildung der Pu-führenden Komponenten von Wiederaufarbeitungsanlagen und stellt erste Ergebnisse vor, die über Simulationsmodelle gewonnen wurden.

C O N T E N T S

+ The authors were deeply saddened by the unexpected death of their colleague Andreas Berliner.

1. Introduction

In nuclear fuel reprocessing facilities currently under IAEA safeguards, conventional nuclear material accountancy (CMA) complemented by containment and surveillance constitutes the fundamental measure for verifying the non-diversion of nuclear material. The accountancy measures are conventional in the sense that the safeguards-relevant information provided by closure of the material balance is only available after a clean-out physical inventory of the chemical process area, usually coinciding with a plant shutdown. The frequency of inventory taking is specified in the Facility Attachment, and is to be not less than once per year. In the future it is expected that increasingly larger commercial scale reprocessing facilities will become subject to IAEA safeguards. Therefore the Agency will have to consider intensifying its accountancy and C/S measures in an attempt to continue to meet its internal goals for timely detection of significant diversions.

In recent years the concept of near real time material accountancy (NRTMA) has emerged in international safeguards and refers to an extention of CMA in which material balances are struck at a frequency comparable to or exceeding that required to meet Agency detection time criteria for direct use material (currently detection of the diversion of one significant quantity within one to three weeks). Moreover, whereas material balance data derived from CMA are generally evaluated using a single period statistical model, implicit in the NRTMA approach are evaluation procedures which take specifically into account the time-sequential nature of the incoming data.

Discussion of the possible role of NRTMA for safeguarding large scale reprocessing plants has focussed on the chemical separation and purification process where, after dissolution of the spent fuel, Pu in bulk form is processed to high purity and concentration preparatory to conversion and refabrication into new fuel elements. It is generally agreed that other areas of the reprocessing plant containing nuclear material (spent fuel pond, head-end, product storage) can be adequately controlled with conventional accountancy and C/S measures.

Implicit in any practical application of NRTMA to a commercial reprocessing plant is the need to establish in-process inventories during plant operation; partial or complete clean-outs and/or plant shutdowns are out of the question at the material balance frequencies implied by the safeguards detection goals. This need imposes one of the more fundamental limitations on the application of NRTMA. The problem can be a purely technical one - the difficulty of determining or estimating the inventory in specific components to the required precision. It can also be of a non-technical nature - e.g. the unwillingness of a plant operator, for reasons of commercial sensitivity, to provide the detailed process information which may be necessary.

As mentioned above, another characteristic feature of NRTMA is the collection of materials-balance data sequentially in time at a frequency commensurate with goals for timely detection without disrupting normal process operation. Therefore, statistical techniques developed for sequential decision making can be invoked. The capability and robustness of the proposed techniques to detect abrupt and protracted diversion are an important consideration in the application of NRTMA.

The many free parameters and still open questions involved in developing and optimizing NRTMA safeguards approaches preclude to a large extent experimentation with existing facilities. Moreover the high throughput plants of the future might well be designed with NRTMA considerations in mind and would not necessarily be comparable with the small-to-medium scale facilities currently under international safeguards. For these reasons a modeling and simulation approach was developed to design and evaluate theNRTMA system: a mathematical and logical representation of the process was built up and the process operation simulated using a digital computer. In this report the mathematical modelling and simulation of critical stages of Pu handling in fuel reprocessing facilities will be discussed in relation to NRTMA and IAEA safeguards. The discussion includes models designed to enable an independent and verifiable determination of Pu inventory in operating solvent extraction contactors, as well as full scale simulation of Pu distributions and flows in the extraction partitioning and purification process of a high throughput reference reprocessing plant.

2. Computer simulation of a reference flow sheet

2.1 General description

As a contribution to an International Workshop on Near Real Time Accountancy in Large Reprocessing Facilities /1/, KfK and KFA have collaborated in the development of a simulation program for a 1000 t/a reference commercial reprocessing plant operating with a conventional PUREX flowsheet. The simulation focusses on plutonium flows and distributions in the co-extraction, partitioning and purification stages of the process, beginning at the input accountability tanks and ending at product analysis and loadout. It thus corresponds to that part of the so-called "process material balance area" to which NRTMA might be applied for safeguarding purposes. The flowsheet, shown schematically in Figs. 1 through 5, was provided by KfK-IHCh /2/; the relevant operational data were obtained in discussions with Deutsche Gesellschaft für Wiederaufarbeitung von Kernbrennstoffen (DWK) /3/. Data generated in the simulation runs are being processed by a measurement simulation program developed by KfK-EKS /4/. The simulated measurement data are, in turn, being evaluated statistically /5/ with a view to determining the overall capability of NRTMA with respect to the timely detection of plutonium losses.

The various components of the simulation program are shown schematically in Fig. 6, as well as the relationship to measurement simulation and statistical evaluation programs.

2.2 Mathematical models

2.2.1 Independent process variables

Input flow rates and Pu-concentrations of external streams entering the process constitute the independent or "exogenous" variables of the simulation model. These variables have been treated as stochastic quantities as in Ref. /6/. If $x(t)$ is the value of an exogenous variable at time t (for example, the extractant flow rate to a pulse column) then its value at time $t + \Delta t$ is given by the algorithm

$$x(t + \Delta t) := x(t) + \bar{x}\, r\, F\, \Delta t$$

$$\text{If } \mathrm{abs}(x(t + \Delta t) - \bar{x}) \geq F\, \bar{x} \text{ then } x(t + \Delta t) := \bar{x}$$

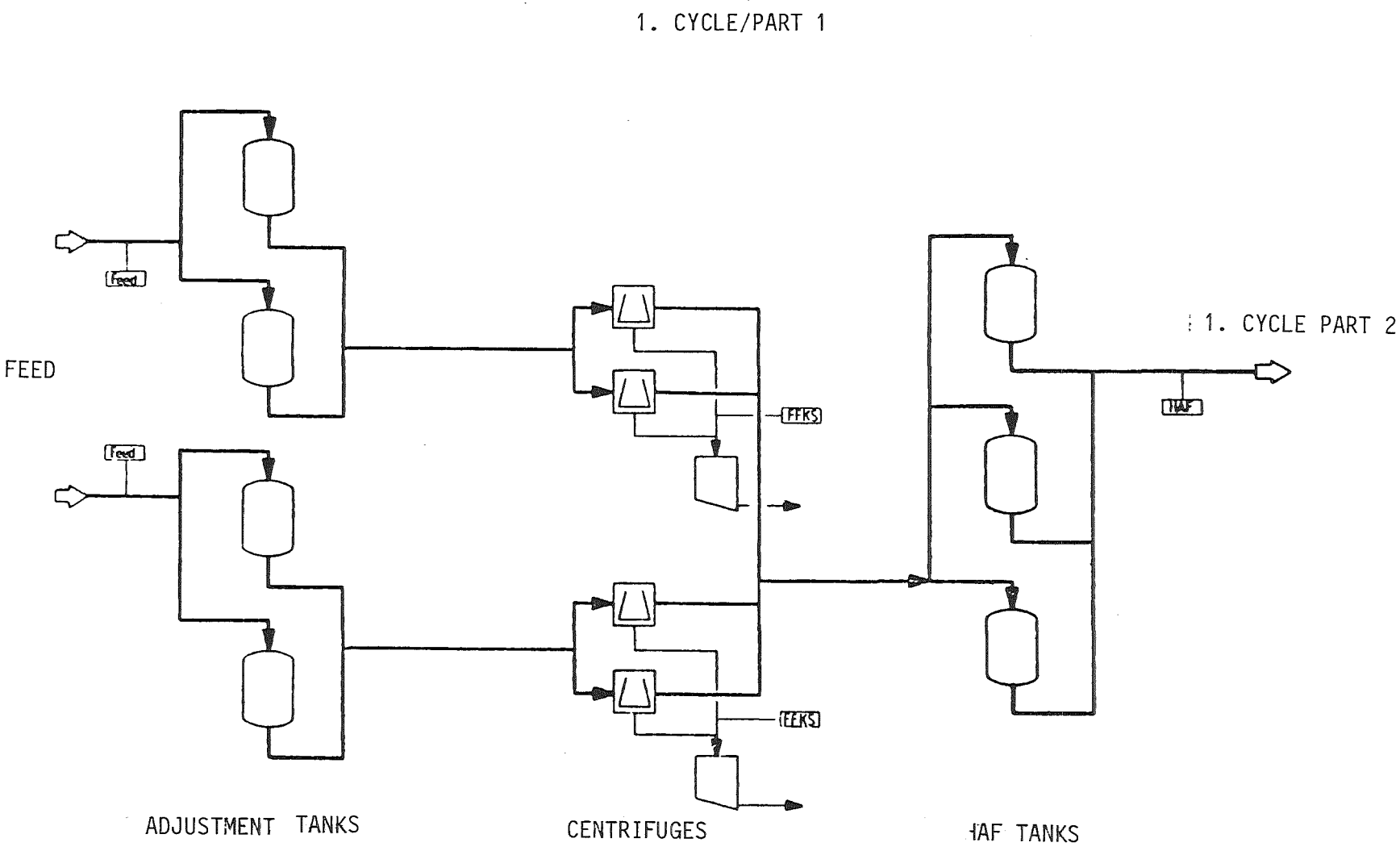

Fig. 1: 1. Cycle/part 1 of reference reprocessing plant.

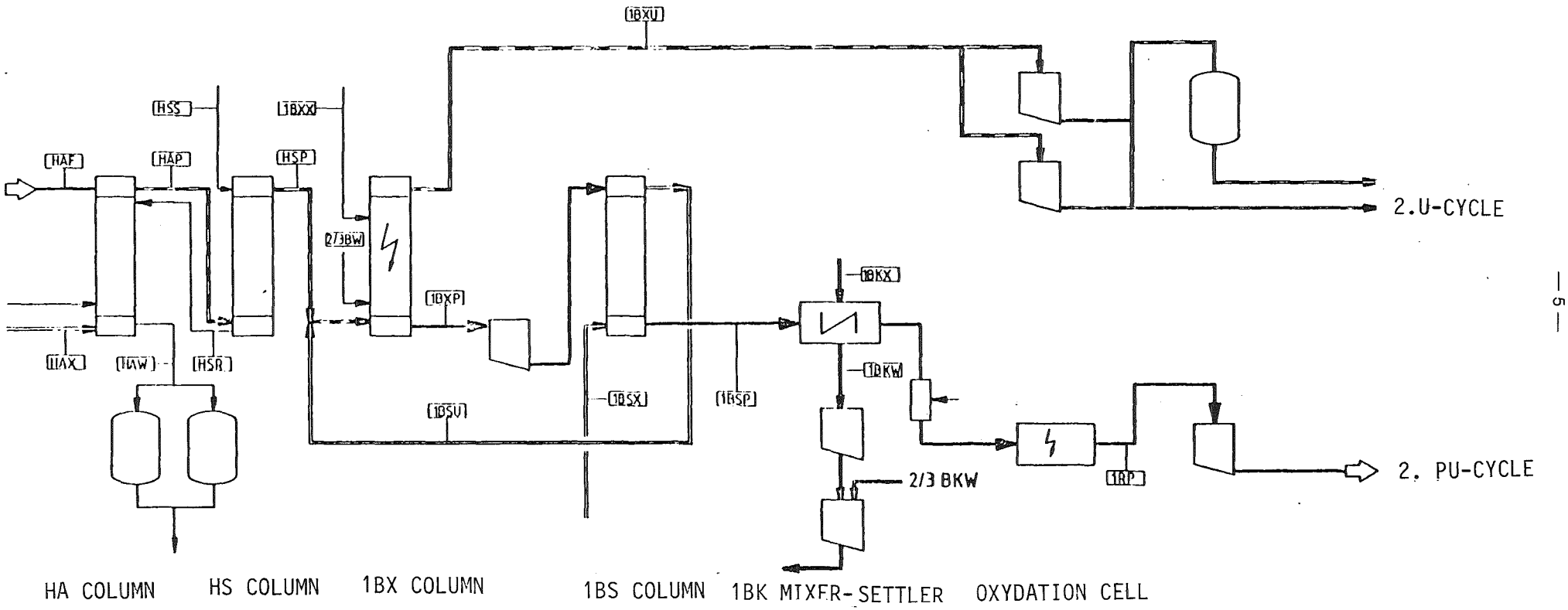

Fig. 2: 1. Cycle/part 2 of reference reprocessing plant.

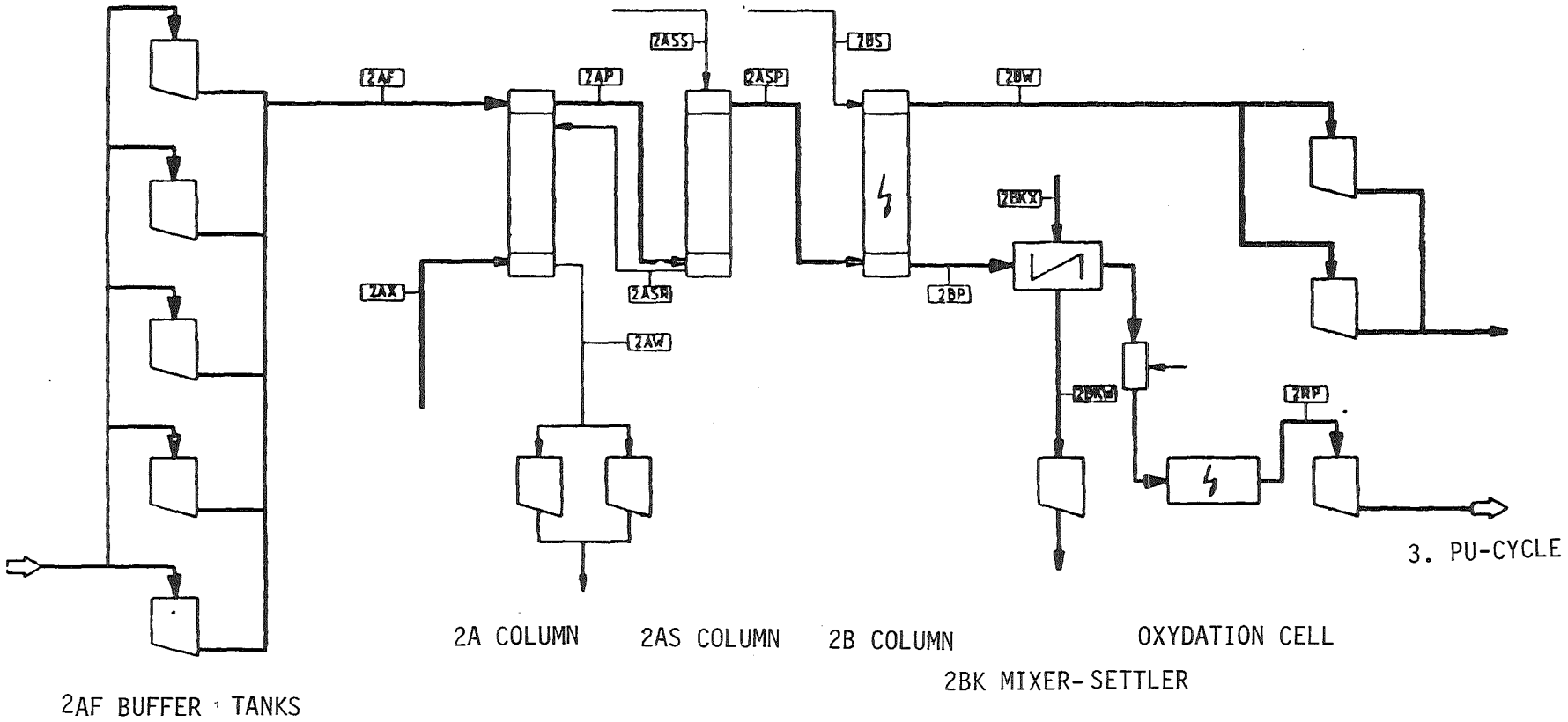

Fig. 3: 2. Pu - cycle of reference reprocessing plant.

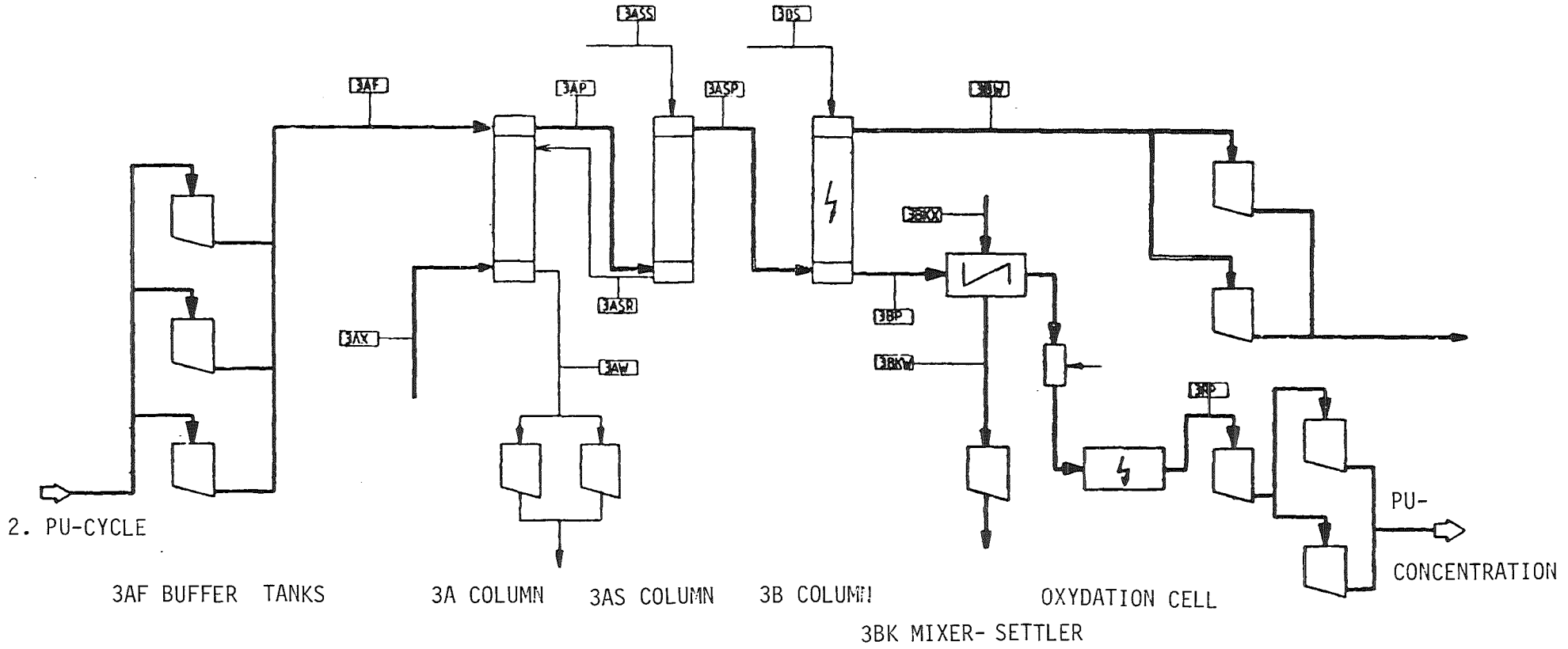

Fig. 4: 3. Pu - cycle of reference reprocessing plant.

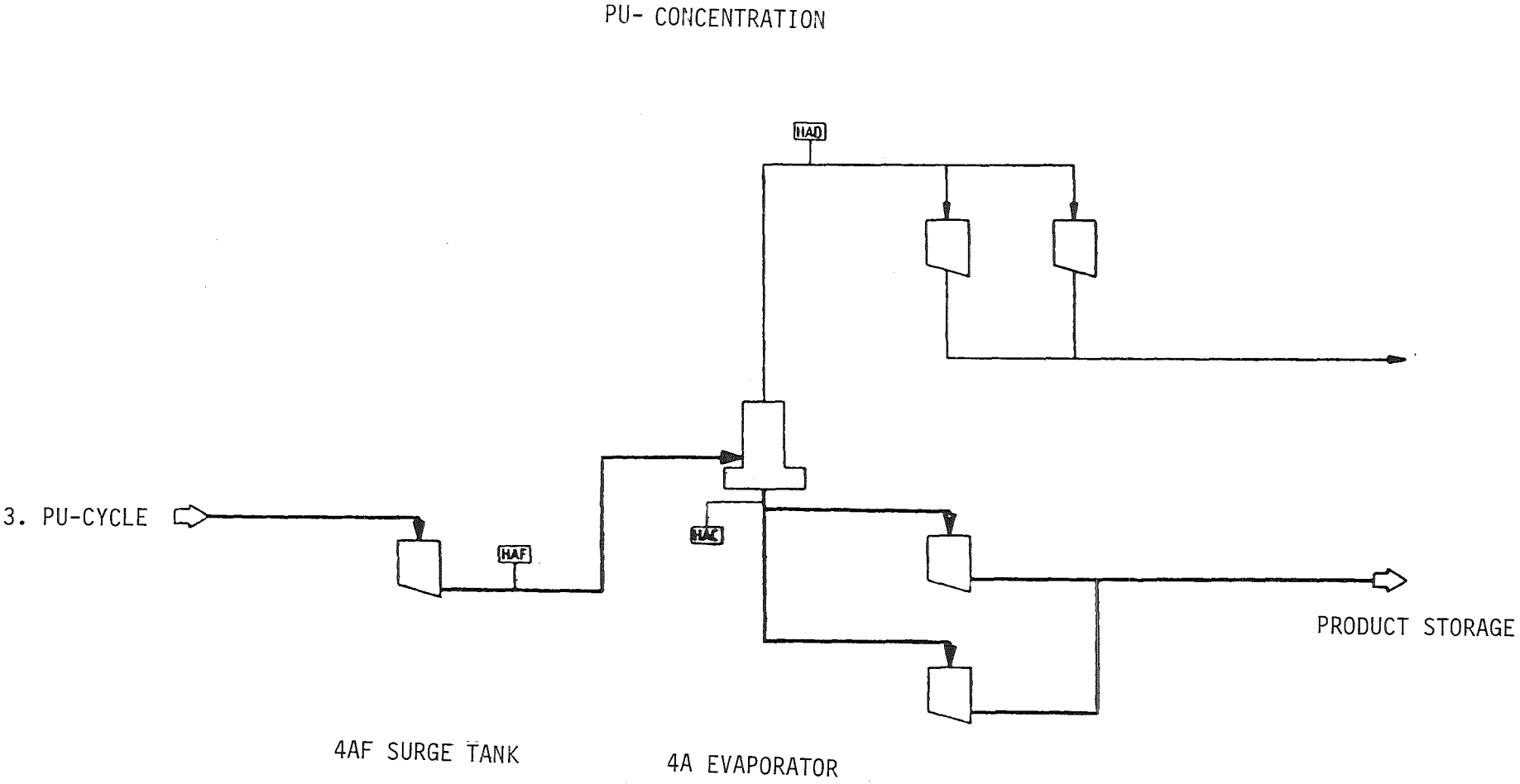

Fig. 5: Pu - concentration of reference reprocessing plant.

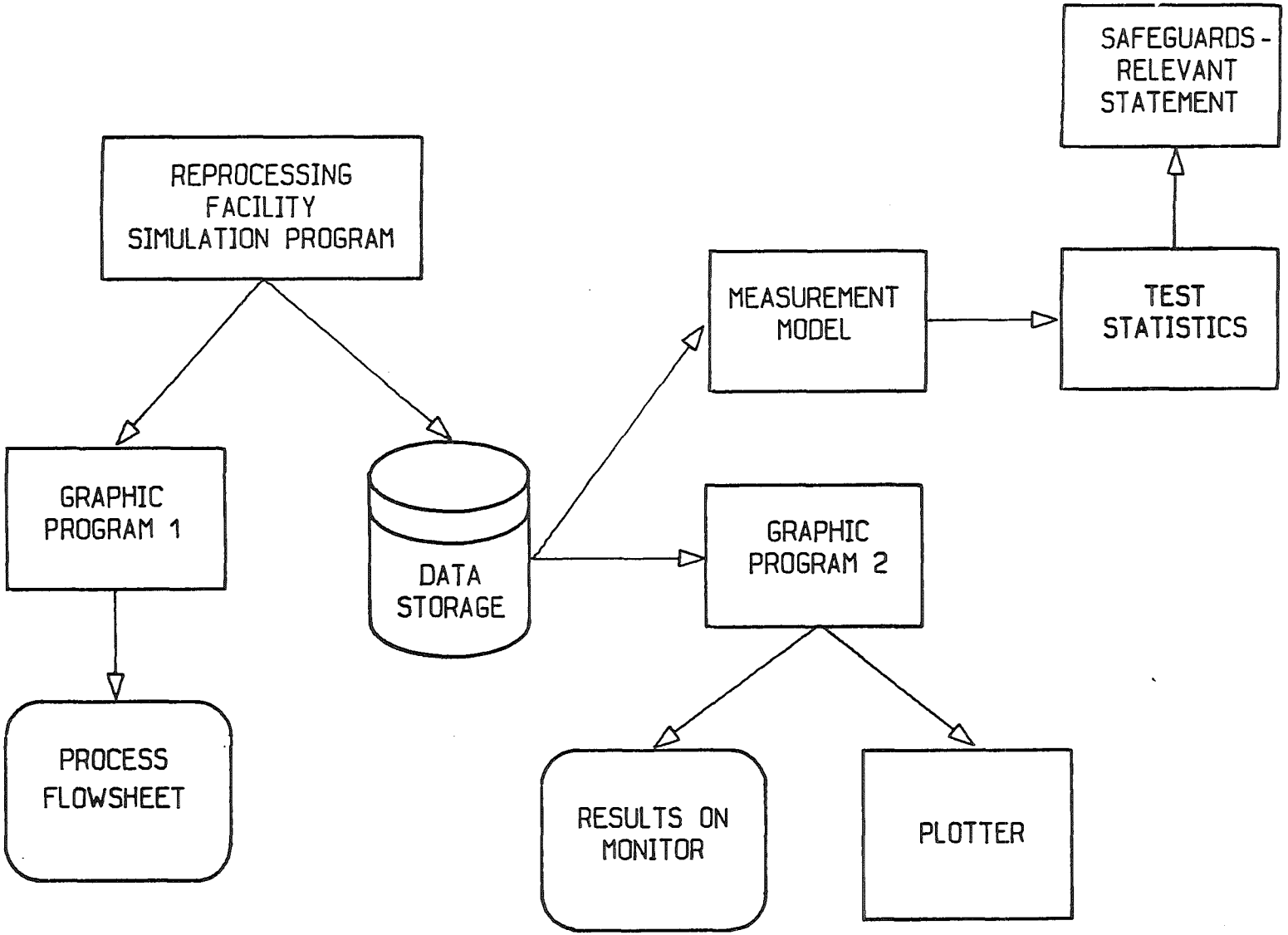

Fig. 6: Relation between process simulation and overall investigation of the near-real-time accountancy method.

where $\bar{x}$ is the flowsheet value, r is a random number distributed uniformly over the interval (- 1,1) and F is a fraction determining the limits within which x(t) can vary.

2.2.2 Tanks

All tanks in the reference flowsheet are assumed to be equipped with capabilities for volume measurement, homogenization and sample taking. Preliminary values have been established for those parameters which determine the working cycle of tanks but which cannot be derived from the flow sheet.

For a perfectly mixing tank with single input and output, the conditions of mass and volume conservation over the iteration time increment Δt are

$$C_T(t + \Delta t)\, V(t + \Delta t) - C_T(t)\, V(t) = \Delta t\ \overline{C_1 F_1} - \Delta t\ \overline{C_T F_2}$$

$$V(t + \Delta t) - V(t) = \Delta t\ \bar{F}_1 - \Delta t \bar{F}_2$$

where:

F_1 = input flow rate (l/hr)

C_1 = input concentration of Pu (g/l)

C_T = Pu conc. in tank (g/l)

F_2 = output flow rate

V = liquid volume in the tank

$\overline{CF}$ = average mass transfer during Δt

$\bar{F}$ = average flow rate during Δt

Following /6/ one can write, for small Δt,

$$\overline{CF} = \{C(t + \Delta t)\, F(t + \Delta t) + C(t)\, F(t)\}/3 + \{C(t)\, F(t + \Delta t) + C(t + \Delta t)\, F(t)\}/6$$

and $$F = \{F(t + \Delta t) + F(t)\}/2\ .$$

Solving for the output concentration at t + Δt:

$$C_T(t + \Delta t) = \{C_T(t)\ V(t)/\Delta t + \overline{C_1 F_1} - C_T(t)\ F_2(t)/3 - C_T(t)\ F_2(t + \Delta t)/6\} \,/\, \{V(t)/\Delta t + \bar{F}_1 - \bar{F}_2 + F_2(t + \Delta t)/3 + F_1(t)/6\}$$

where all quantities on the right hand side are known from the preceding time step or from the output of the preceding process element (F_2 is treated as a stochastic variable as described in 2.2.1).

2.2.3 Solvent extraction contactors

The solvent extraction contactors are the most complex elements in the simulation model. It is necessary to seek a compromise between a realistic mathematical description of their dynamic behavior and subroutines which do not lead to prohibitively long computation times. Three simple contactor models for process simulation and inventory estimation are discussed in section 3. In particular, the model used for the simulation of pulse columns and mixer-settlers in the reference process can be found in subsection 3.1.2 .

2.2.4 Miscellaneous components

All other Pu-bearing components of the flow sheet were modeled as variations of the tank model already described. The evaporator, for example, is treated as an overflowing (constant volume) tank. The input stream is reduced in volume by a "concentration factor", the excess (vapour) leaving via a second output. The concentrated liquor is assumed to mix perfectly with the evaporator inventory. In the case of the centrifuges, similarly, a fraction of liquid with a small Pu-concentration (representing sludge) is separated from the input stream. Perfect mixing within the volume of the centrifuge is again assumed.

2.3 Development of the process simulator

The process simulator is a computer program which incorporates and combines the simulation algorithms for the various components into a logical whole intended to represent the functioning of the entire chemical process. The

computer language SIMULA provides an efficient, flexible and transparent tool for achieving such an integration and was chosen in the present instance. A description of the SIMULA program is given in Appendix I. Broadly speaking, the process simulator can be thought of as a (large) number of parallel, autonomous co-routines interacting and communicating with one another in time. Co-routines are set up not only for all process components, but also for the "operators" responsible for coordinating the various cycles and keeping the process running in a controlled way.

3. Solvent extraction contactor modelling

3.1 Simple linear models

Simulation of solvent extraction contactors generally demands sophisticated mathematical models as well as extensive computer programs.

Since in the present application the associated algorithms are used a very large number of times during a simulation run the contactor models should not consume too much computer time while reproducing in a reasonable way the quantities of interest. In the following we describe 3 such models for the simulation of the time dependence of the Pu inventory.

3.1.1 Exponential model

The exponential model provides a simplified method for approximating solvent extraction contactor inventories. This model does not consider the many complex variables that affect contactor performance and is intended for steady-state operation only. It has been used for the simulation of the pulse columns in the Plutonium Purification Process of the Barnwell reprocessing facility /6/.

A schematic diagram of a pulse column is shown in Fig. 7. The input streams consist of Pu bearing feed, extractant (both organic or aqueous) or scrub. The extractant stream flows countercurrently to the feed stream and scrub stream (if any). The column is pulsed through either the aqueous or organic stream. Typically, the feed stream is the dispersed phase whereas the extractant is the continuous phase. The aqueous-organic interface would be controlled in the disengaging section at the waste end of the column. The column will have an extraction section and may or may not have a scrub section.

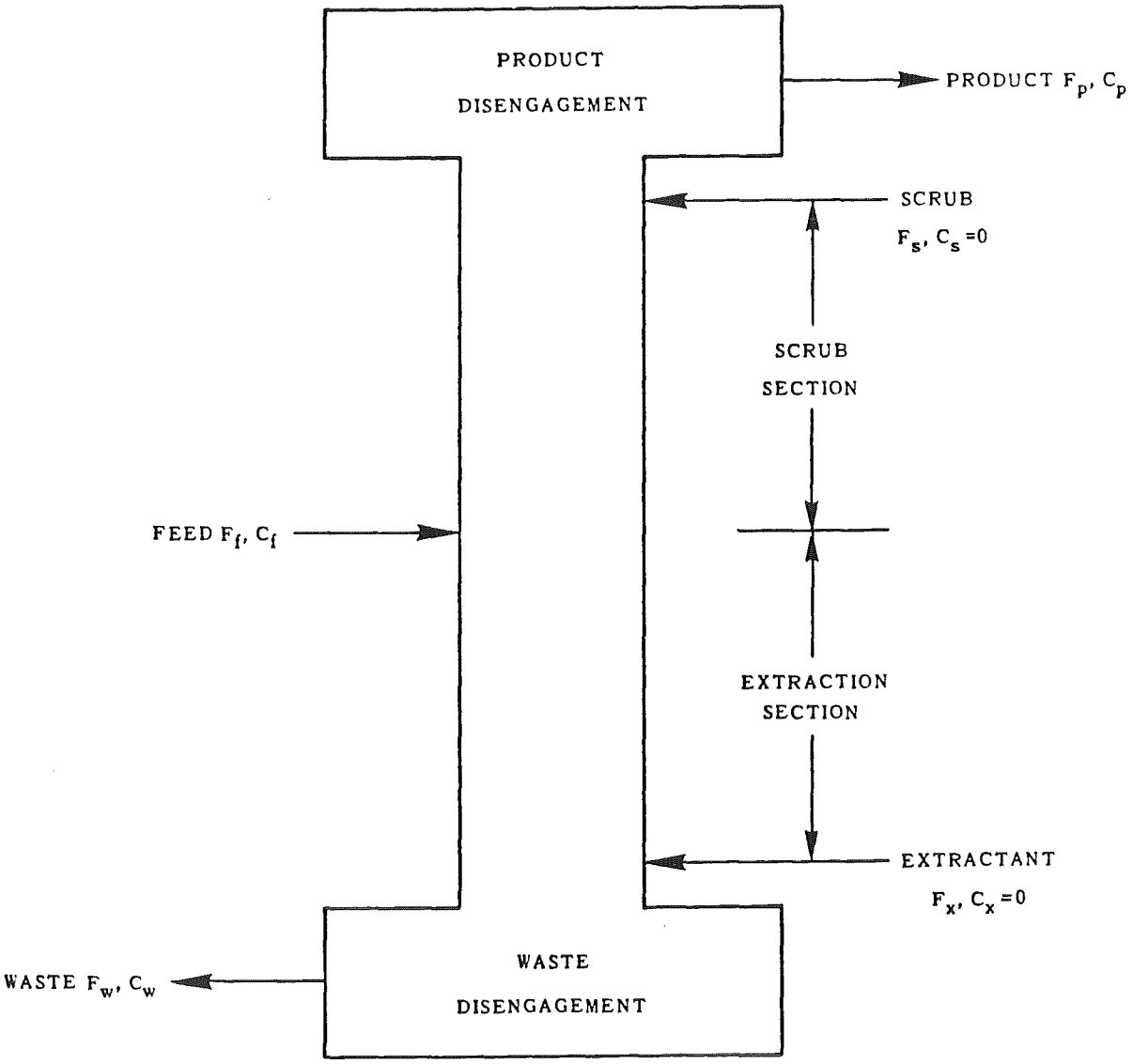

Fig. 7: Pulse column schematic diagram of the exponential model.

For this model we assume that the concentration profiles are exponential within the extraction section. If C_f and C_w are the Pu concentrations in the feed and waste streams, and L is the length of the extraction section, then

$$C_w = C_f \exp(-\alpha L)$$

so that

$$\alpha = \frac{1}{L} \ln (C_f/C_w) \ .$$

The concentration in the feed phase, $c_f(y)$, at any point y in the extraction section is

$$c_f(y) = C_f \exp(-\alpha y) \ , \qquad 0 \leq y \leq L \ .$$

Similarly, if the concentrations in the extractant and product streams are zero and C_p, respectively, the concentration of Pu in the extractant phase is given by

$$c_x(y) = C_p \{\exp(-\alpha y) - \exp(-\alpha L)\} \ / \ \{1 - \exp(-\alpha L)\} \ .$$

The Pu holdups in the aqueous and organic phases of the extraction section are obtained by integrating the concentrations over the phase volumes:

$$H_f = \int_0^L c_f(y) dV_f(y) \ ,$$

$$H_x = \int_0^L c_x(y) dV_x(y) \ .$$

The assumption of exponential concentrations underestimates the column holdup because refluxing of solute from the extractant to the feed stream can produce nonuniformities (bulges) in the concentration profiles.

The above equations are evaluated under the added assumption that the phase volumes are constant fractions of the free volume in the extraction section, V_e:

$$V_f = k_f V_e \quad ,$$

$$V_x = k_x V_e \quad ,$$

where $k_f + k_x = 1$. If the extraction section has a constant cross section, the phase holdups are given by

$$H_f = V_f(C_f - C_w)/\hat{\alpha} \quad ,$$

$$H_x = V_x C_p \{1/\hat{\alpha} - C_w/(C_f - C_w)\} \quad ,$$

where

$$\hat{\alpha} = \ln (C_f/C_w) \quad .$$

The total Pu holdup in the column is the sum of the holdups in the feed and extractant phases, the holdup in the scrub section (if any), and the holdups in the waste and product disengagement volumes (V_w and V_p). The total holdup is given by

$$H = H_f + H_x + C_w V_w + C_p V_p + H_s \quad ,$$

where H_s is the holdup in the scrub section.

For the Barnwell process simulation specific consideration for columns in the Plutonium Purification Process included the following: in this process columns 2A, 3A, and 3B have scrub sections as well as extraction sections. We assume that no refluxing of Pu occurs from the product stream into the scrub stream. If the flow rate in the scrub stream is F_s, the concentration in the stream feeding the extraction section is taken to be $C_f F_f/(F_f + F_s)$; i.e., it is assumed that the feed and scrub streams mix perfectly. Pu holdup in the scrub section, H_s, is calculated from

$$H_s = k_p C_p V_s \quad ,$$

where V_s is the free volume in the scrub section, and k_p is the volume fraction in the product phase.

The dynamics of pulse column operation are determined by integrating the continuity equations :

$$\frac{\Delta V}{\Delta t} = \bar{F}_f + \bar{F}_s + \bar{F}_x - \bar{F}_p - \bar{F}_w \quad ; \quad \bar{F} = \frac{1}{\Delta t} \int_{\Delta t} F(t)dt;$$

$$\frac{\Delta H}{\Delta t} = \bar{S}_f - \bar{S}_p - \bar{S}_w \quad ; \quad \bar{S} = \frac{1}{\Delta t} \int_{\Delta t} F(t)\, C(t)dt.$$

The equation for $\Delta V/\Delta t$ is solved under the subsidiary constraint that plug flow is maintained throughout the column:

$$F_w = F_f + F_s \quad ,$$

$$F_p = F_x \quad .$$

With this constraint, $\Delta V/\Delta t = 0$, and the total volume is constant; in addition, the values of all flow-rate variables are determined.

In the $\Delta H/\Delta t$ equation, the final concentrations in the product and waste streams ($C_p(t_2)$ and $C_w(t_2)$) must still be determined. The concentration in the waste stream is obtained from another subsidiary constraint: the Pu flow rate in the waste stream is assumed to be proportional to the Pu flow rate in the feed stream:

$$C_w F_w = k C_f F_f \quad .$$

The constant of proportionality, k, is taken to be the ratio of the average Pu flow rates:

$$k = \frac{\overline{C_w F_w}}{\overline{C_f f_f}} \, .$$

With the plug flow and waste stream constraints and the contactor holdup model, the above equation for $\Delta H/\Delta t$ can be solved explicitly for the final product concentration, $C_p(t_2)$, in terms of known quantities. This solution is not consistent with our original assumption that the concentration and flow-rate variables are linear in time; however, it is a valid solution if the time step is taken sufficiently small that the trajectories may be approximated by straight lines. Fortunately, the column holdups vary slowly in time so that a good approximation is obtained with a reasonable time step (0.25 h).

3.1.2 Ideal stage model

An ideal stage model, shown schematically in Fig. 8, was chosen to represent the behavior of pulse columns and mixer settlers in the reference flow sheet of Figs. 1 to 5. The C_i represent Pu-concentrations (in g/l) and the F_i flow rates (in l/hr).

V_4 and V_5 are disengagement volumes, V is the total volume of liquid in the mixing section and N is the number of ideal stages. Plug flow is assumed with organic and aqueous phase volumes proportional to the respective flow rates. Streams 1 and 2 are assumed to mix perfectly and all stages are assumed to be 100 % efficient (organic and aqueous concentration within a stage are at equilibrium). As an example, if the model is applied to the 2A column of Fig. 3, the C_i and F_i are interpreted as follows:

C_1, F_1	aqueous feed (2AF stream)
C_2, F_2	aqueous reflux from 2AS scrub column (2ASR stream)
C_3, F_3	organic extractant (2AX stream)
C_4, F_4	organic product (2AP stream)
C_5, F_5	aqueous waste (2AW stream)

A simple equation for the dynamic behavior of the extractor is obtained by requiring mass conservation within the i^{th} stage over a small time increment Δt. If $x_i(t)$ is the i^{th} stage Pu-concentration at time t in the

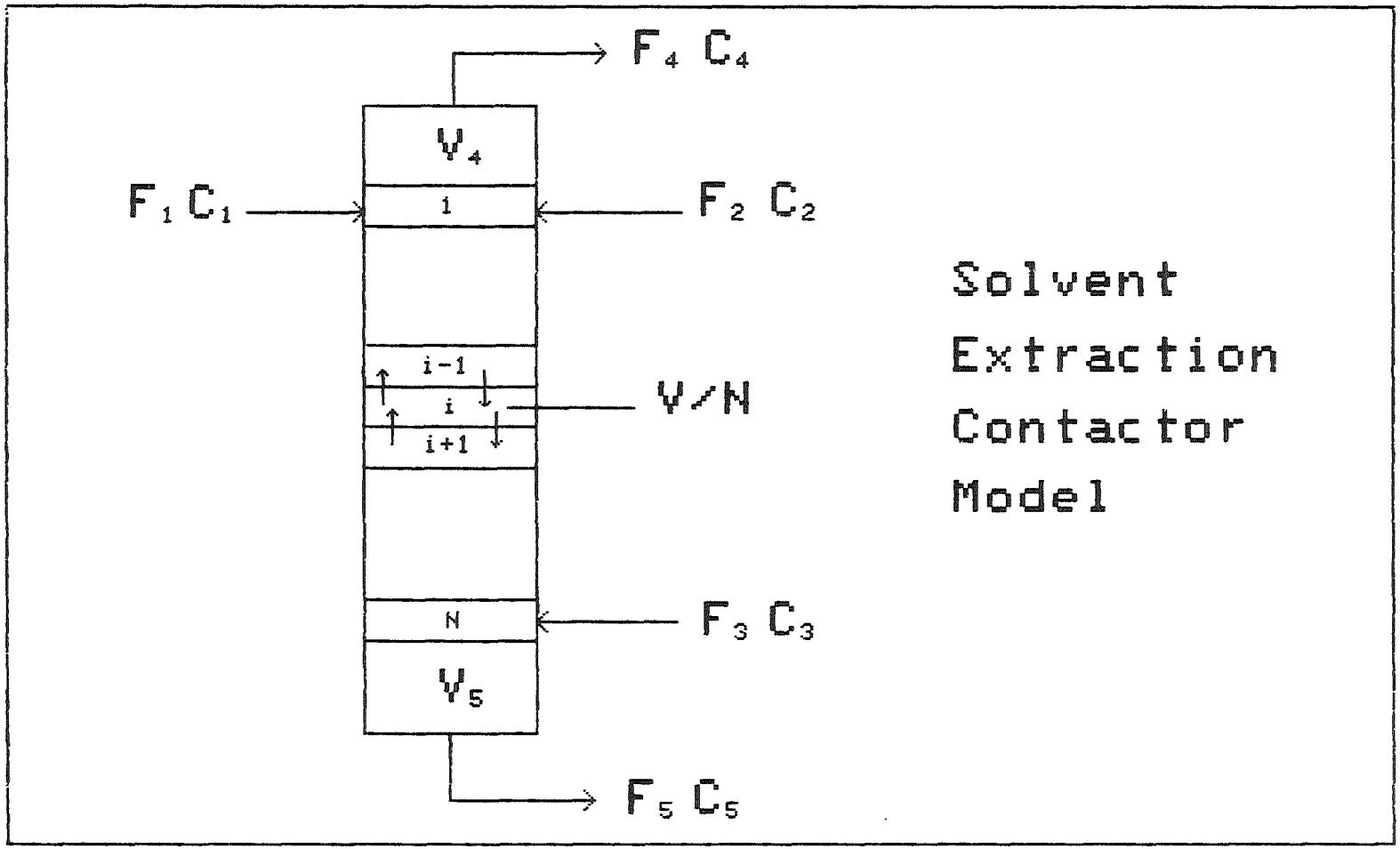

Fig. 8: Pulse column schematic diagram of the ideal stage model.

(1,2)-phase (mixing at the top in Fig. 8), then the following approximate relation holds:

$$x_i(t + \Delta t) = x_i + \{(F_1 + F_2)\,\Delta t\,(x_{i-1} - x_i) + F_3\,\Delta t\,(D_{i+1}\,x_{i+1} - D_i\,x_i)\} \,/$$

$$\{(F_1 + F_2 + D_i\,F_3)\left(\frac{V}{N}\,\frac{1}{F_1+F_2+F_3} + \frac{\Delta t}{2}\right)\}$$

all quantities on the right hand side being evaluated at time t. D_i, the distribution coefficient in the i^{th} stage, is assumed to be time independent. For each increment Δt in simulated time, the above equation is evaluated across the column (i = 1 ... N) subject to the boundary conditions

$$x_0 = (F_1\,C_1 + F_2\,C_2)/(F_1 + F_2)$$

$$x_{N+1} = C_3 / D_N$$

The concentrations in the streams leaving the disengagement sections (C_4 and C_5) are evaluated in a similar way, with the assumption that V_4 and V_5 are constant. The total column holdup is then simply

$$H = \frac{V}{N}\,\frac{1}{F_1+F_2+F_3}\sum_{i=1}^{N} x_i(F_1 + F_2 + D_i\,F_3) + C_4V_4 + C_5V_5$$

In this very simple geometrical model for solvent extraction, physical and chemical effects (such as backmixing, temperature and molarity dependence of distribution coefficients, etc) are fully neglected. The parameters D_i and N were chosen in order to ensure that the separation factors implied by the reference flowsheet were achieved. If the distribution coefficient is assumed to be roughly stage independent, D_i = D = constant, then N and D can be related by drawing a mass balance around the bottom end of the mixing volume V in Fig. 8. The result is

$$N = \frac{\ln\left\{\frac{C_1\,F_1 + C_2\,F_2}{C_5\,(f_1 + F_2)}\left(1 - \frac{F_1 + F_2}{DF_3}\right) + \frac{F_1 + F_2}{DF_3}\right\}}{\ln\frac{DF_3}{F_1 + F_2}}$$

For a given extractor, the maximum allowable iteration time interval is essentially determined by the liquid volume displacement time in one ideal stage. This restriction was used to choose a convenient N value for each extractor. The above equation was then solved for the average D-value giving the desired output concentration C_5. As an example, for the 2A column of Fig. 3, the values D = 5.5 and N = 7 were used. Possible variations in D from stage to stage (D_i) can be modelled by expanding D_i as a polynomial in x_i.

3.1.3 Reduced-order linear model

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 /7/ 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 column inventory calculations are based on the following 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.
3. Concentration and flow rate measurements are available in near-real time.
4. The column inventory at the nominal operating point has been previously determined from chemical model calculations and calibration experiments.

In calculating the inventory, the column is divided into four sections: top, bottom, scrub and extraction. In a type-A column, for example, the inventories in each of these sections are computed as shown in Table 1. The product concentration, C_p, is calculated from the standard mass balance equation for the column.

T A B L E 1 Equations used for inventory calculations in a type-A column

Section	Inventory
top	$H_T = H_T^o \left(\frac{C_P}{C_P^o} + \frac{V_T}{V_T^o} - 1 \right)$
bottom	$H_B = H_B^o \left(\frac{C_W}{C_W^o} + \frac{V_B}{V_B^o} - 1 \right)$
scrub	$H_S = H_S^o \frac{C_I}{C_I^o}$
extraction	$H_E = H_E^o \frac{C_I}{C_I^o}$

Notation: H = inventory, C = concentration, V = volume

Subscripts: T = top, B = bottom, S = scrub, E = extractant,
P = product, W = waste, I = input,

Superscript: o = nominal value at the operating point

T A B L E 2 Nominal flowsheet values and calculated inventories for 2A columns

Model	F_f (l/hr)	F_s (l/hr)	F_x (l/hr)	C_f (g/l)	H (kg)
1*	106	57	119	54.2	16.2
2	363	81.6+	163	5.66	0.599
3	363	81.6+	163	5.66	0.559

* related to U (Barnwell)
\+ reflux from 2AS column

T A B L E 3 Nominal flowsheet values and calculated inventories for 2 B columns

Model	F_f (l/hr)	F_x (l/hr)	C_f (g/l)	H (kg)
1*	119	160	48.4	4.94
2	163	75	12.5	1.08
3	163	75	12.5	1.13

* related to U (Barnwell)

3.2 Model intercomparison

A realistic simulation of solvent extraction contactors, at least near equilibrium operation, is essential if the respective models are to be of use for in-process holdup estimation or as elements of an overall process simulator. In the following the three contactor models are compared with one another. Model 1 is the 'reduced order' linear estimator discussed in section 3.1.3, model 2 is the ideal stage model of subsection 3.1.2 and model 3 is the exponential model of subsection 3.1.1.

As a basis of comparison, the 2A and 2B pulse columns of the Barnwell facility (model 1) and of the reference flowsheet (Fig.3) were chosen. The respective columns differ considerably from each other (the Barnwell 2A column for example has a scrub section, while the 2A column of Fig. 3 is in series with a separate scrub column 2AS). For this reason only the responses to equal relative changes in the feed and extractant streams were compared.

Tables 2 and 3 show the nominal flow sheet flow rates and concentrations for feed, scrub and extractant streams for the 2A and 2B columns. Also shown are the calculated holdups at equilibrium.

The responses (inventory changes) of the models to 10 % increases in feed and extractant flow rates and feed concentration are shown in Tables 4 and 5. In all 3 models, holdup variations are seen to be roughly proportional to the variations in input flows and concentrations.

T A B L E 4 Inventory change caused by 10 % changes in either F_f, F_x or C_f (2A-column)

10 % change in	Change in inventory (%) Model 1	Model 2	Model 3
F_f	9	10	6
F_x	- 9	- 11	- 6
C_f	10	9	8

T A B L E 5 Inventory change caused by 10 % changes in either F_f, F_x or C_f (2B-column)

10 % change in	Change in inventory (%) Model 1	Model 2	Model 3
F_f	7	9	8
F_x	- 7	- 10	- 6
C_f	10	9	10

3.3 Model validation

Clearly experimental validations of the contactor simulation models would be highly desirable. Recently, experimental data have been obtained at the Barnwell plant for large-scale commercial pulse column contactors. Cold runs were performed operating the 2^{nd} and 3^{rd} Pu-purification cycles in a closed loop (so-called miniruns). After each run, actual column inventories were determined in a column dump. In addition input and output flow rates and concentrations were recorded.

Calculated and experimental results for the 2A and 2B columns are compared in Table 6. It should be emphasized that the inventory estimates were obtained without any usage of the product concentration; the latter quantity was estimated along with the inventory. It is then used as the estimated feed concentration of the 2B column, increasing the uncertainty in the 2B column inventory estimate.

T A B L E 6 Comparison of measured (Barnwell minirun No. 5) and calculated inventories for the 2A and 2B columns

	Deviation between measured and estimated inventory (%)	
Model	2A	2B
1	- 27	- 3
2	- 24	19
3	- 26	15

4. Preliminary results for the reference process

4.1 Individual components

The simulated startup of the pulse columns in the first cycle (co-extraction and partitioning steps) is shown in Figs. 9, 10 and 11. Startup occurs approximately 100 hours after the first transfer of material to the input accountability tanks. Fig. 9 shows the build-up of column inventory and product and waste concentrations for the HA column during the first hours of operation. Inventory build-up for all four pulse columns in the cycle is shown in Fig. 10. The Pu-concentration profile in the feed (organic) phase in the extraction section of the 1BX column is displayed in Fig. 11 as a function of time. Equilibrium is reached after about 3 hours.

Typical buffer tank behavior is shown in Fig. 12. The three tanks indicated supply feed to the 3rd Pu purification cycle and are operated in such a way that, on average about two thirds of buffer capacity is in use at any given time.

4.2 Global behavior

The overall time dependence of Pu inventory for the three extraction and purification cycles and for the entire MBA is shown in Figs. 13 - 16. In all four figures, material in process components (i.e. pulse columns, mixer settlers, centrifuges, evaporator, etc.) is plotted separately from the inventory in feed, analysis and buffer tanks.

As can be seen from these figures, process component inventory accounts for only about 5 % of the total Pu inventory. This relatively small value and its still smaller fluctuation are clearly of relevance to the application of NRTMA to the reference facility. That is, for reasons already mentioned in section 1, frequent redetermination of process component inventories may be difficult or impractical. It is then of particular interest to examine by simulation the statistics of any hypothetical "hidden inventory" - i.e. material not measured in a particular NRTMA implementation. The variance of the hidden inventory would represent an additional contribution to the uncertainty of the total MBA inventory and can be compared directly with measurement variances for material contained in measured vessels. Covariance terms arising from long term fluctuations may also be of relevance, since sequential statistical tests for both abrupt and protracted diversions are sensitive

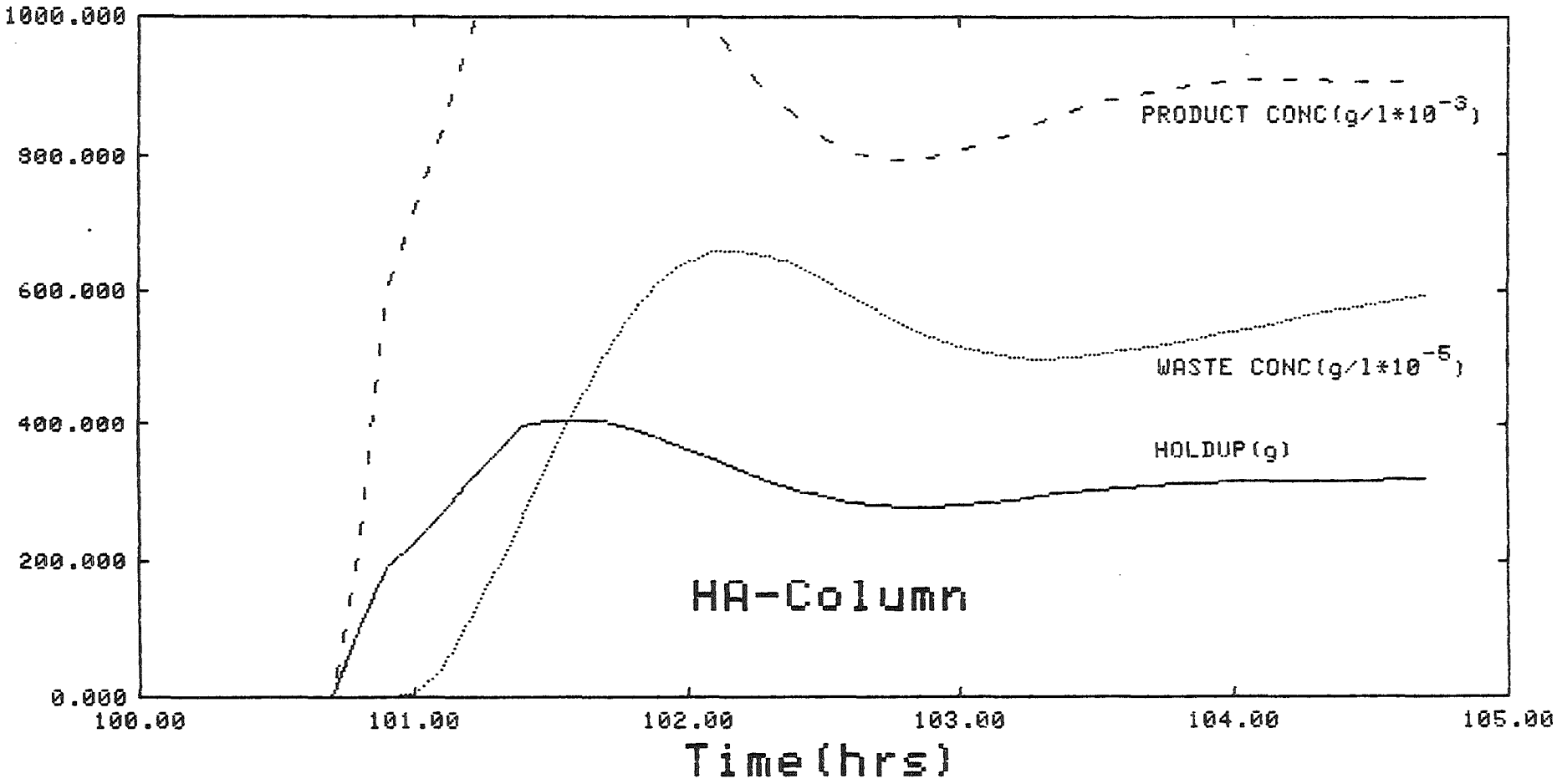

Fig. 9: Start-up of the HA pulse column. Equilibrium in column holdup and output concentrations is reached within 3 hours.

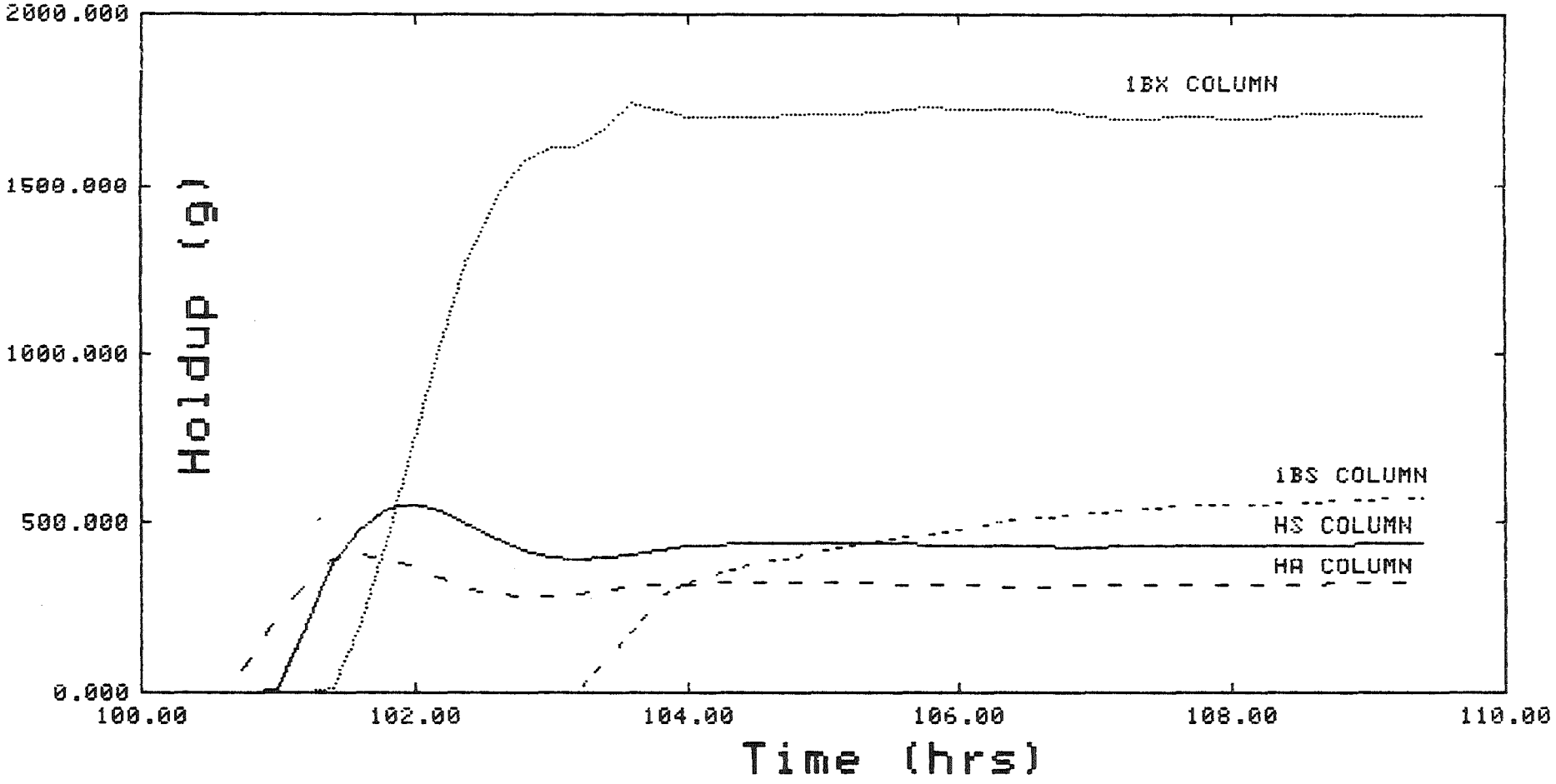

Fig. 10 : Start-up of the four pulse columns in the first (coextraction and partitioning) cycle.

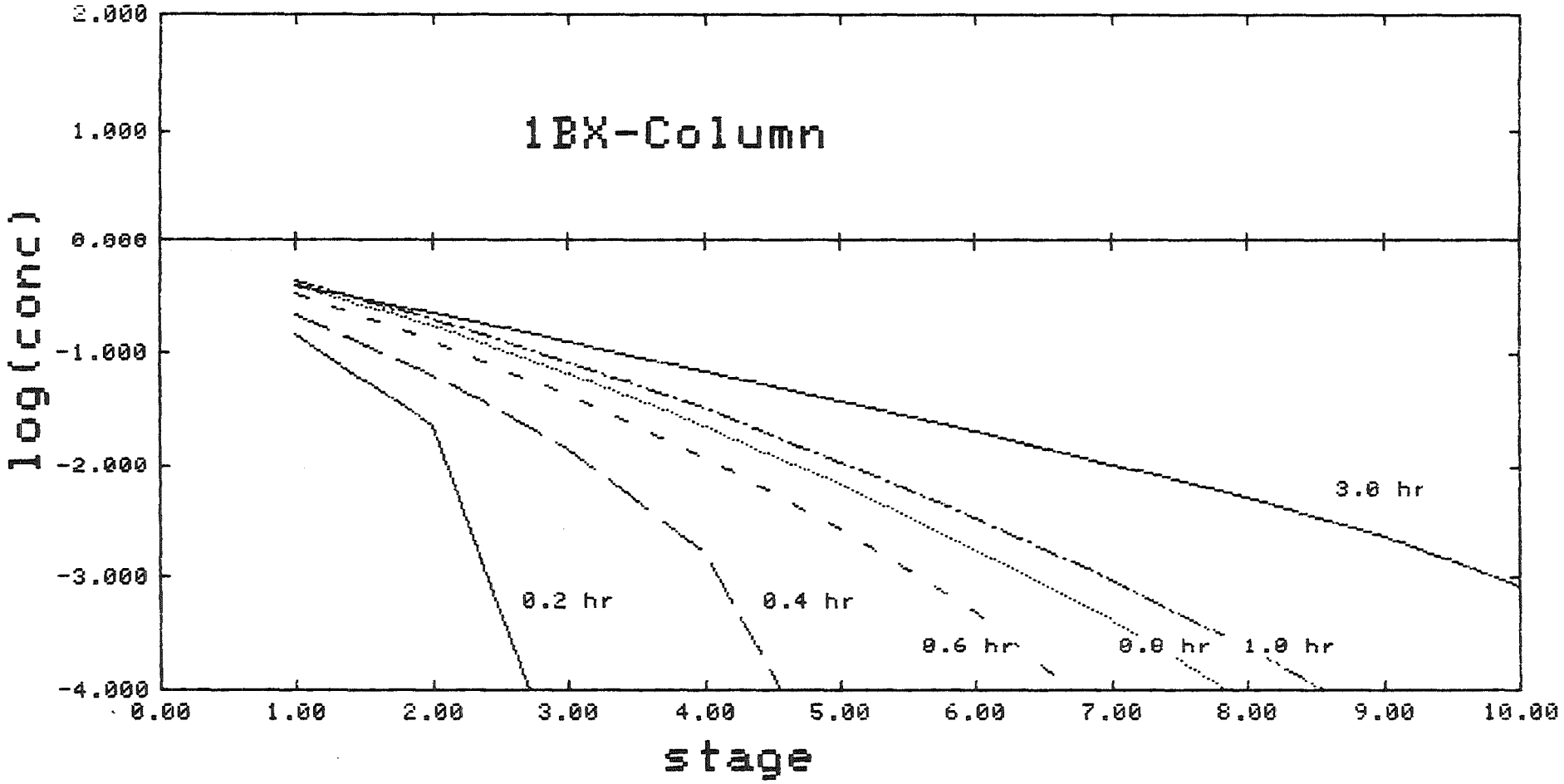

Fig. 11: Calculated Pu-concentration profiles of the 1BX column.

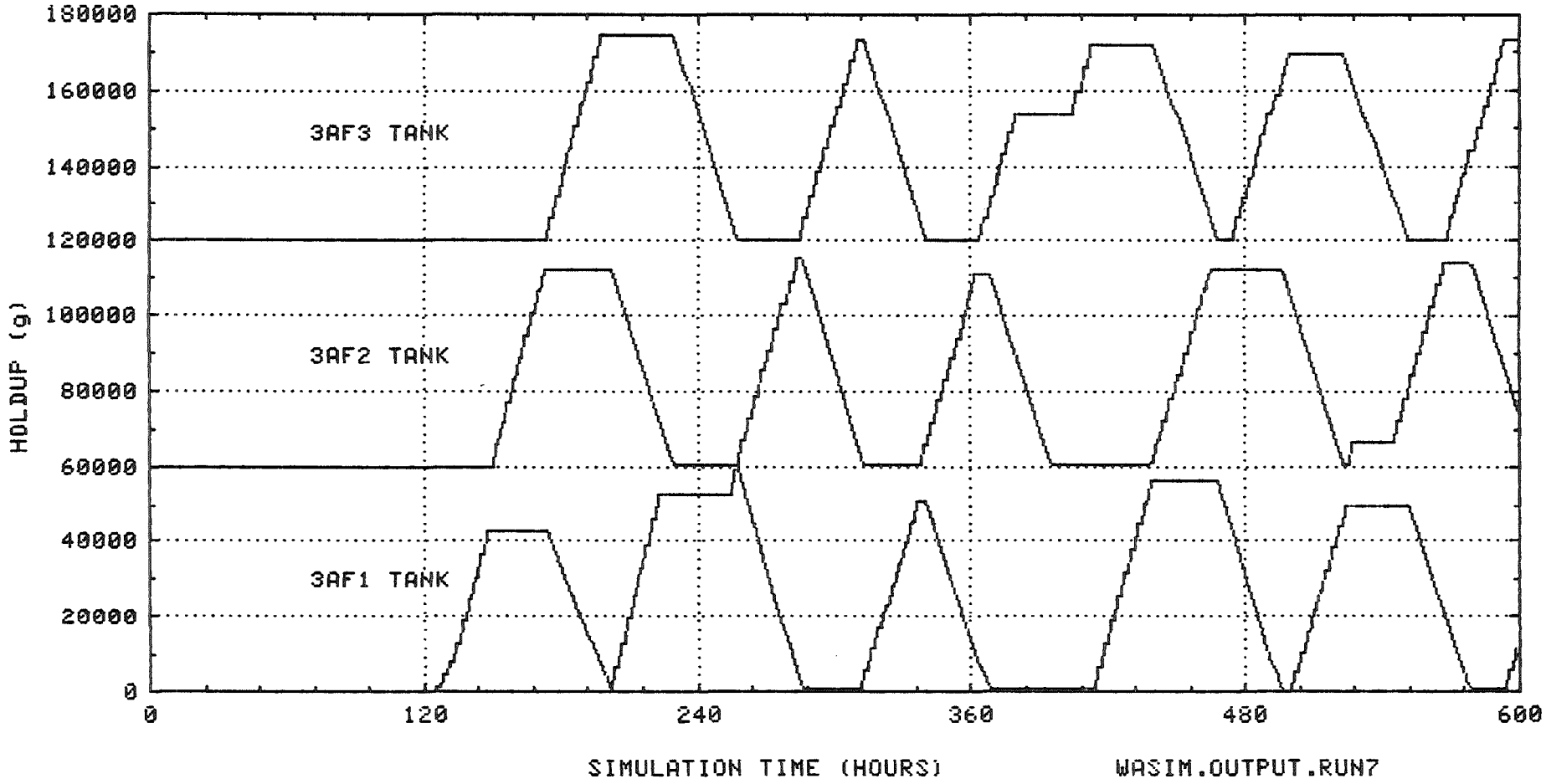

Fig. 12: Time dependence of inventory in the 3AF buffer tanks.

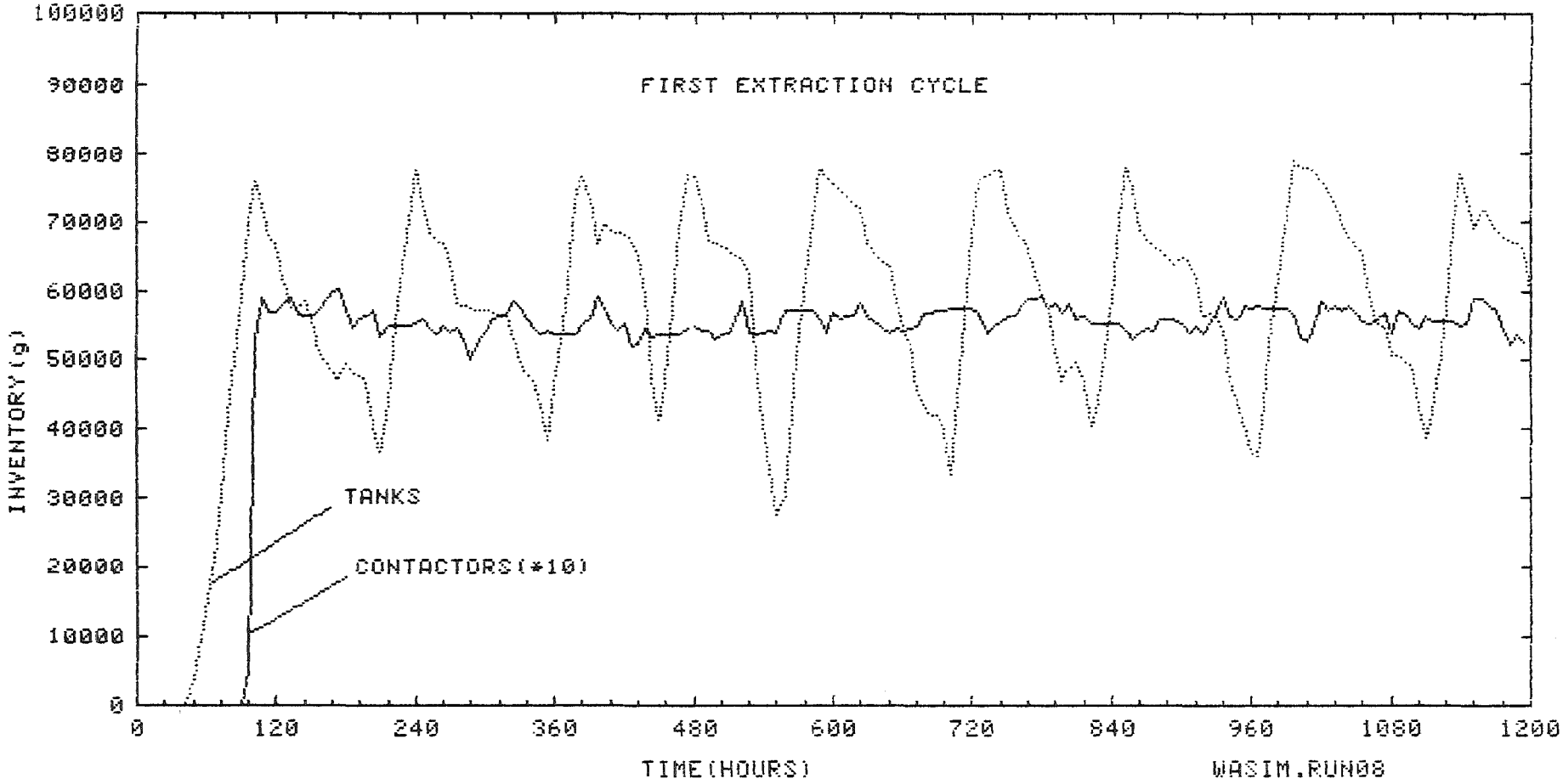

Fig. 13: In-process inventory of the 1. cycle, simulated for 50 days.

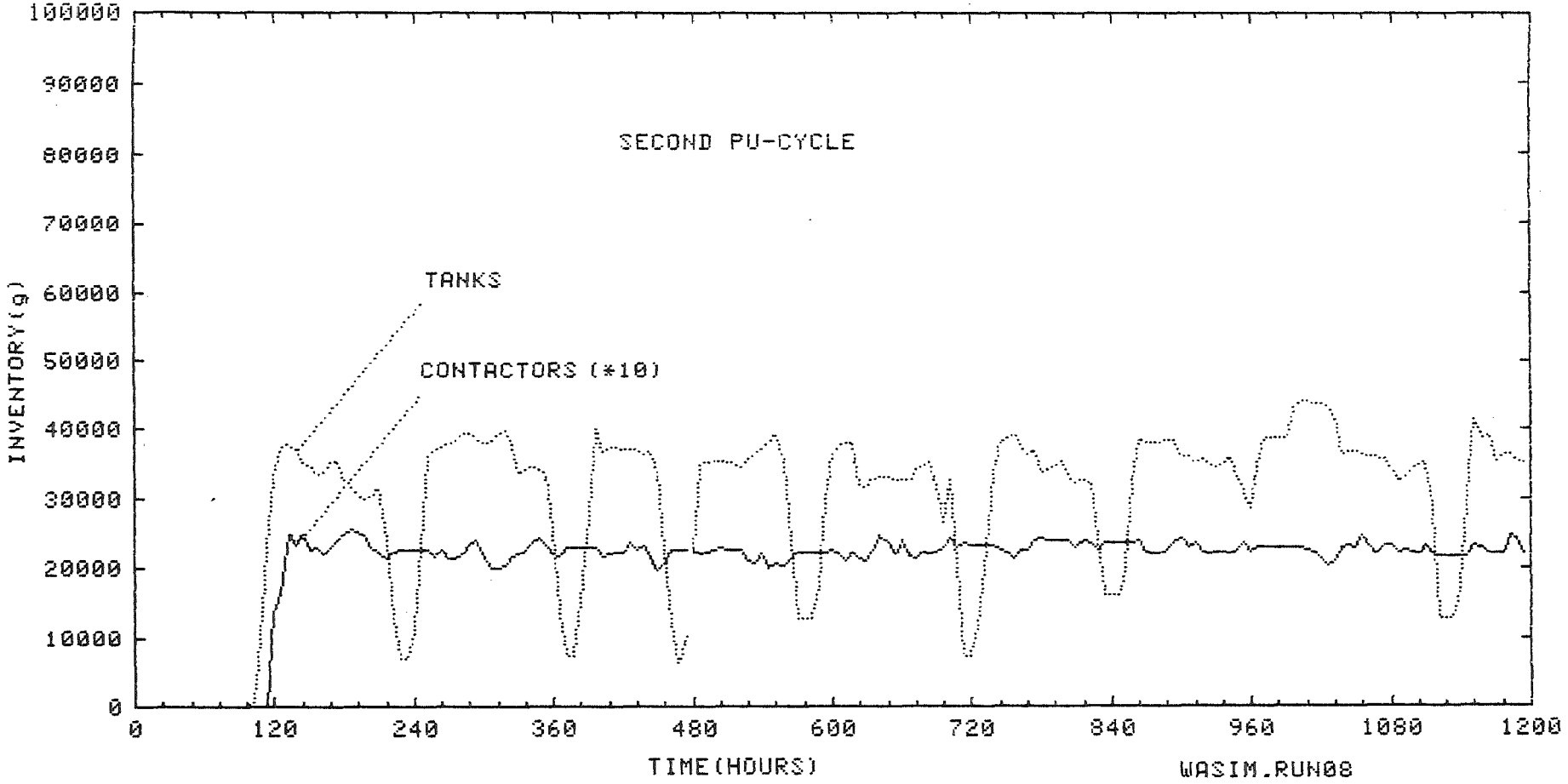

Fig. 14: As in Fig. 13, but 2. Pu - cycle only.

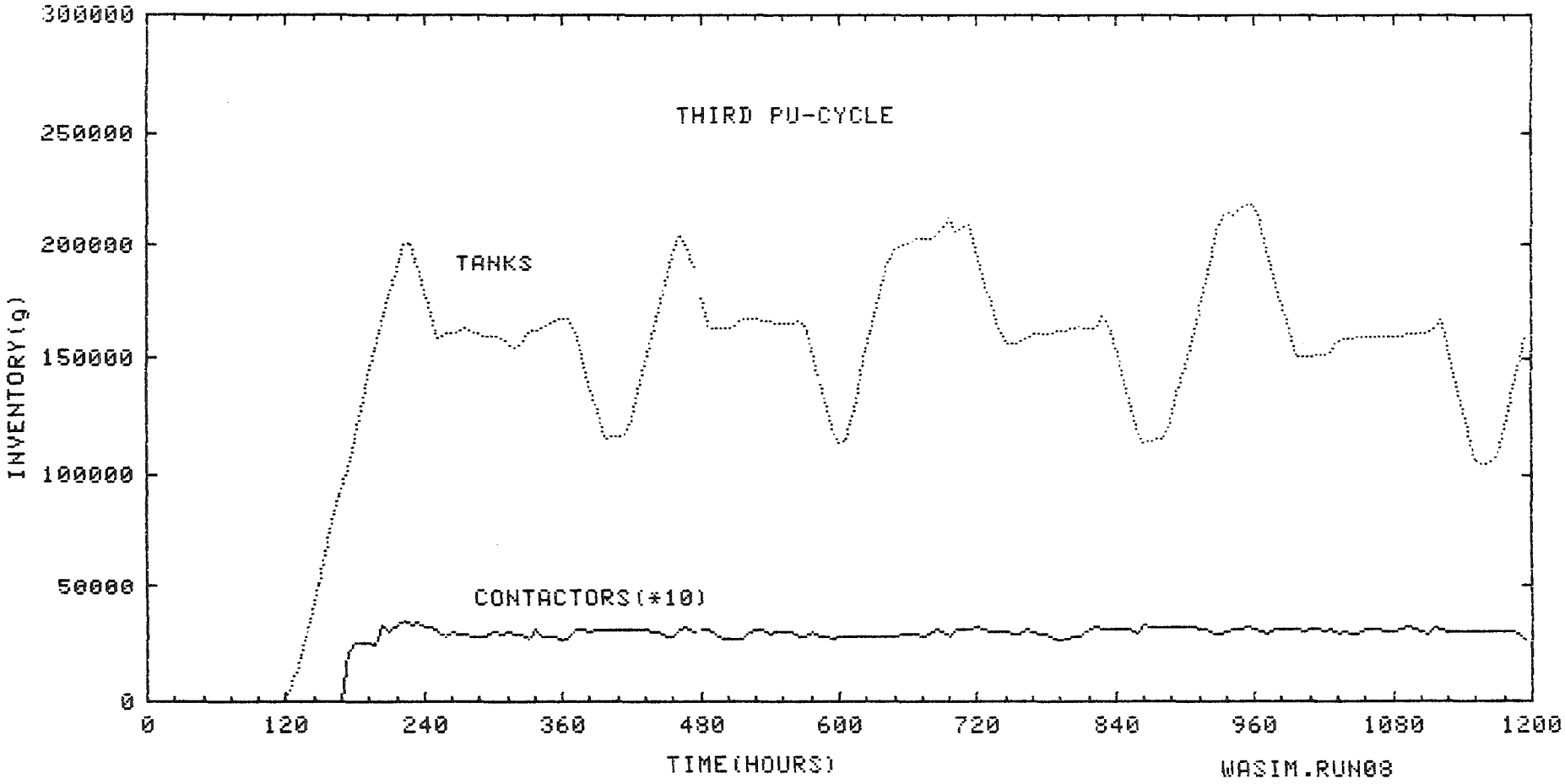

Fig. 15: As in Fig. 13, but 3. Pu - cycle only.

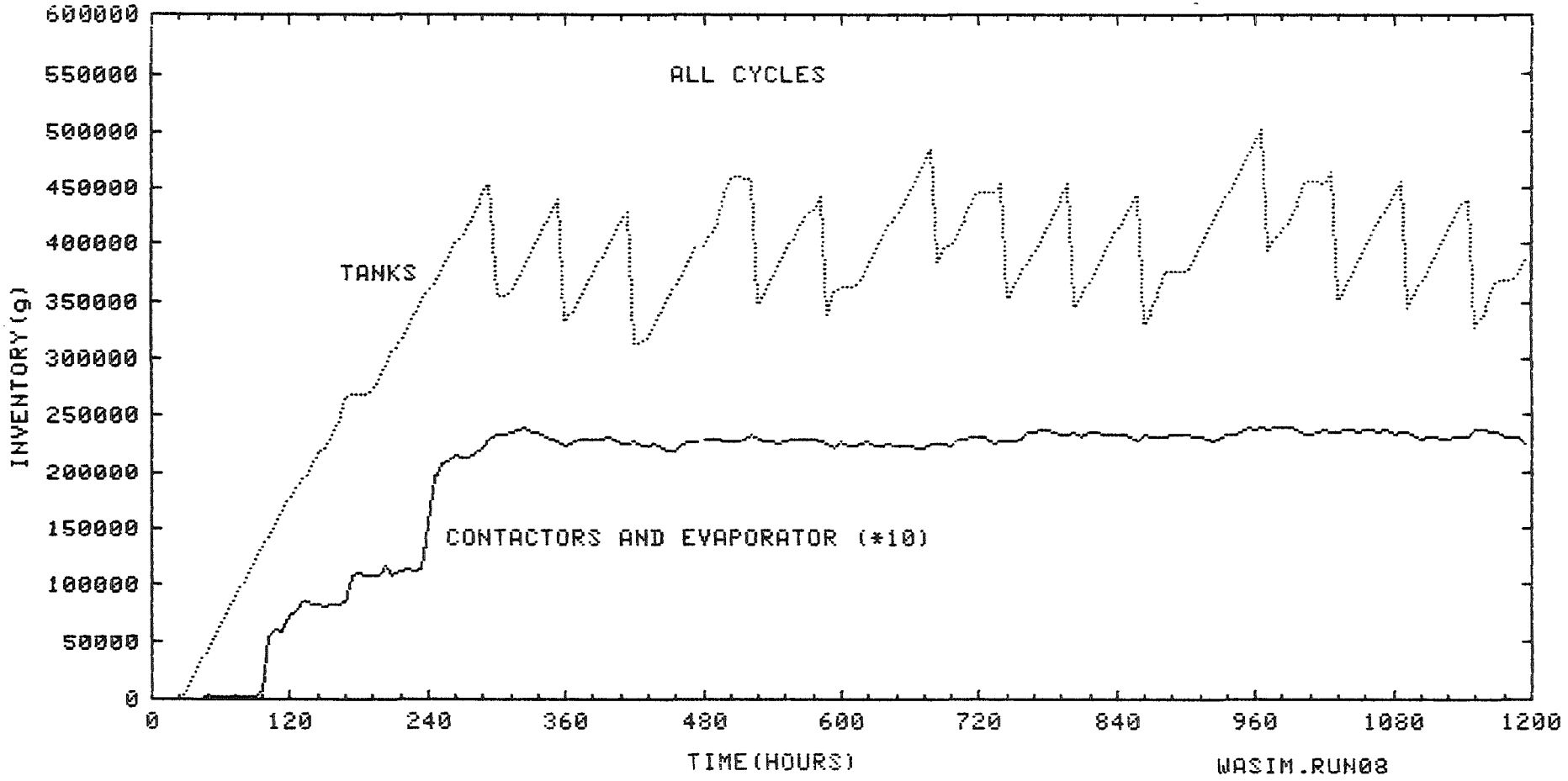

Fig. 16: As in Fig. 13, but entire in-process inventory.

to the covariance structure of the MUF time series as a whole.

After equilibrium was attained in the simulated plant operation, the Pu-inventory in centrifuges, solvent extraction contactors and evaporator was treated as a stationary time series. Its mean, variance and auto-covariance were determined by sampling at regular time intervals. Results for a 50 day simulation and 6 hour sampling period are shown in Table 7.

The stability of the process component inventory is evident from the table. While Pu amounts in individual solvent extraction contactors can vary fairly widely even in the simple linear model used in the simulator, on the average such variations tend to cancel out over the whole MBA.

T A B L E 7 Process Component Inventory Statistics for Observations

Mean	23.1	kg
Variance	0.28	kg^2
S-Dev	0.53	kg
Covariance:		
Lag = 24 hrs	0.19	kg^2
Lag = 48 hrs	0.15	kg^2
Lag = 72 hrs	0.11	kg^2
Lag = 96 hrs	0.07	kg^2
Lag = 120 hrs	0.05	kg^2

5. Recommendation for future activities

Simulation experiments using the models described in this report are currently underway to provide raw data for investigation of NRTMA procedures. In combination with detailed measurement simulation and sequential statistical analysis, the sensitivity of NRTMA to different loss patterns and its robustness under more realistic operating conditions will be tested. Of particular interest are effects of process noise (inventory fluctuations), hidden inventory, changes in burnup of processed fuel and deviations from equilibrium plant operation. This work is being carried out under the Support Program to the IAEA of the Federal Republic of Germany.

In parallel to the above investigations validity checks of the simulation model will continue. Unfortunately, the reference process exists on paper only and realistic operation data of a commercial reprocessing plant are not available. Therefore, model validation will be limited to information obtained at small non-commercial scale plants. In accordance with the preliminary results the emphasis should be laid more on validation of realistic operating strategies than of models of individual components (e.g. pulse columns). The latter could be done on the basis of experimental results from laboratory-scale facilities and of the outcome of more sophisticated model codes.

In connection with the development of safeguards system for a commercial scale reprocessing plant in the Federal Republic of Germany it is intended to accommodate the simulation program to the flow sheet of an actual planned facility. This work will be carried out in close cooperation with the future operator of the facility (DWK).

Acknowledgement

Two authors (M.C. and G.S.) would like to thank R. Weh/DWK for his cooperation in providing essential information on reprocessing plant operation.

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Appendix Process Simulation Program SPFK

(A. Berliner /KfK)

1. General

The program described in this appendix was designed to simulate Pu inventory and flow within the process material balance area of a reference spent fuel reprocessing facility. It is intended as a tool to assist in the development and evaluation of measures for near real time material accountancy.

2. Principles of operation

2.1 Material transport

The following conventions were established for representation of flows and concentrations:

Material can be transported among the various process components through piping. This movement is specified in terms of flow rate F (l/hr) and plutonium concentration C (g/l). Only linear changes in F and C are permitted within one simulation time increment dt. Thus at time t_o a given material flow is specified by F_o, C_o, dF and dC with

$$F(t) = F_o + dF\,dt, \quad C(t) = C_o + dC\,dt \quad t = t_o + dt$$

All material movements are described in the simulation program in terms of these basic equations.

2.2 Information exchange

The behavior of an individual component can be calculated on the basis of its own current state and information provided by surrounding components. Components can communicate with their environment in two ways: by specifying an output flow to neighbouring components or by sending messages to an operator. All components are required

to accept material from their predecessors or to issue an error message which causes the program to abort (guarantee against 'disappearance' of material). The operators control the process in the sense that they open and close valves, take samples, activate switches, open and close system boundaries etc. A given operator can only influence those components located in "his" part of the facility, or open and close the valves connecting to neighbouring parts controlled by other operators. Other operators affected by such actions are informed via the system boundaries only.

3. Realisation in SIMULA*

3.1 Representation of flows

The class MSG is defined as a simple vehicle for exchange of information regarding material flows among the process components. This class contains all essential specifications concerning material flow:

ABSENDER (Sender):	origin of flow
EMPFÄNGER (Receiver):	destination of flow
F:	Flow rate at time t_o = ZEIT (time)
DF:	Time rate of change of F at time t_o
C:	Pu-concentration at time t_o
DC:	Time rate of change of C at time t_o
ZEIT:	Current simulation time
NEXT:	Pointer to allow chaining of messages

---

* English translations of program variables, procedures etc. in parentheses

In addition, MSG contains the procedure INTO which deposits the above information in the "mail box" of EMPFÄNGER. Senders and receivers of MSG's must be subclasses of the class KOMPONENTE (component), discussed below.

3.2 Information to an operator

In analogy to transmission of flow information, the class OP-MSG transmits messages to the operators. The message is in the form of an EVENT, coded as an integer. Receivers must belong to the class OPERATEUR.

3.3 Representation of components

In addition to the general functions of information exchange, a class has to be defined containing all attributes shared by the various components of the simulation model, including MSG defined above. The actual process components are then designated as subclasses of this class. This enables complete freedom in subsequent interconnection of the individual components to form the complete process.

The general component class was named KOMPONENTE and was given the (SIMULA internal) prefix PROCESS enabling active participation of all of its members in the simulation.

The first common attribute of all components is ID, a text object which is assigned the component name when the component is generated. Furthermore, virtual declarations for procedures OPEN-E, ..., PRINT-STAT are provided. The corresponding actual declarations can (but needn't necessarily) be included in the subclasses of KOMPONENTE. The variable MAIL serves as a "mail box" to receive MSG'S. The following chaining procedure is used for the MSG's:

KOMPONENTE. MAIL	first MSG or NONE
First MSG. NEXT	second MSG or NONE, etc.

The variable OPERATOR specifies which operator is responsible for a specific component.

The variables LASTEV, DT, DT-2, DT-3 are used to determine the time interval elapsed since the last activation of the component and are calculated with procedure DELTA-T.

Procedures DELTA-H and DELTA-V calculate mass or volume input/output during DT.

Transmission of flow information or operator messages is further simplified by the procedures SEND and ALARM. For instance, in order to inform a component XYZ that it is currently receiving a flow of 300 l/hr, changing at a rate of 25 l/hr^2 and a concentration of 5 g/l, changing at a rate of 0.1 g /l/hr, the procedure call

```
SEND ( XYZ, 300, 25, 5, 0.1);
```

suffices.

Similarly, the call ALARM (VOLL); (Full) initiated by a TANK component is sufficient to inform an operator that the maximum capacity has been reached.

The procedures OUTHEAD ... PRINT-STATUS are intended to transfer output information to a terminal or a protocol file. They find use in the procedures PROTOKOLL and PRINT-STATUS defined in the various sub-classes of KOMPONENTE.

Every process component capable of containing material includes among its attributes procedures H and V which calculate, respectively, current inventory and liquid volume, as well as procedures PRINT-H and PRINT-V for output of these quantities.

3.4 Description of individual process components

3.4.1 ROHR (Pipe)

In the present version of the program, this class is not used. It passes material on to the next component instantaneously and unchanged.

3.4.2 SYSTEMGRENZE (System boundary)

This component behaves as ROHR, with the addition that it can be closed and opened by an operator. Both occurences are reported immediately to other operators affected.

3.4.3 VERTEILER (Distributor)

A VERTEILER component receives material from exactly one input and passes it on to one of NAUS (Nout) possible outputs. Switching among outputs is effected by a call (by an operator) to the procedure SCHALTE.

3.4.4 SAMMLER (Collector)

A SAMMLER component receives material from one of NEIN (Nin) possible inputs, and passes it on immediately to a single output. Switching among inputs is effected by a call to procedure SCHALTE.

3.4.5 TANK

A TANK has the following attributes: It is filled until its KAPAZITAET (Capacity) is reached (or until its input is closed). This event is reported via ALARM (VOLL) to the operator responsible. There then follows a quiescent period PR-ZEIT (sample time). Expiry of this time is reported by ALARM (ANALYSE) and the tank can be emptied. This occurs, once the operator has opened the output (OPEN-A), with flow rate F-SOLL $\pm$ 5 % and constant concentration C-AUS = C_{tank}. After complete emptying (to less than 1 % of capacity) the tank can be immediately refilled.

3.4.6 VERSORGUNG (Supply)

Components belonging, to this class feed the process with material. When VERSORGUNG is not blocked by an operator (procedure CLOSE) it issues material at a rate

F-AUS in the range F-SOLL $\pm$ F-SCHRANKE

and with Pu concentration

C-AUS in the range C-SOLL $\pm$ C-SCHRANKE

("Schranke" = limit).

3.4.7 ENTSORGUNG (Removal)

These components collect material leaving the process.

3.4.8 EXTRAKTOR

This class is used in the current version of the program for representation of both pulse columns and mixer settlers. The model equations are described in detail in section 3.1.2.

3.4.9 C-TANK

Components of this class are used to simulate tanks which are in continuous operation (levels regulated about a reference point). They also serve to decouple components which are coupled by feed-back (for example the HA and HS columns). A C-TANK component behaves as follows: An input flow mixes with the content of the tank for DT-AEND time units. Only then is the output flow and/or concentration influenced by changes at the input. Output flow is so calculated that, up to the next time interval, the current deviation of the volume from the present reference value is reduced by half. Any changes in input which might occur in the meantime are ignored. class C-TANK is used to simulate oxidation cells, decoupling tanks and the 1BXP-Tank.

3.4.10 B-TANK

Tanks of this class are filled until KAPAZITAET is reached, upon which the message ALARM (VOLL) is sent to the operator. The operator then opens the output (assuming the process state permits this) and the tank is emptied "suddenly" - ie output flow is very much larger than input flow. In the meantime input of liquid to the tank can continue. After emptying, the tank's output is closed again. This class is used to represent the RP-Tanks in the process.

3.4.11 T-STUECK (T-piece)

This class enables representation of the combination of two flows into one. The output is the sum of the inputs. This can lead to a quadratic time dependence of output concentration during one time step, a situation not permitted in the simulation concept. In order to counter this effect, a linear change in output concentration is chosen in such a way as to produce the same output after time interval DT-AEND as would have been produced in the more exact non-linear case. If the T-STUECK happens to be reactivated prematurely, this can lead to a minimal material balance error which is corrected for in the next time step.

3.4.12 ZENTRIFUGE (Centrifuge)

This class is used to represent centrifuges present in the head-end of the reference flowsheet. It operates by separating a small amount of liquid from the input flow, SCHLAMM * F-EIN, with a likewise small Pu-concentration, EXTR-K* $C_{centrifuge}$. Uniform Pu-concentration in the entire volume of the centrifuge and perfect mixing with the input stream are assumed.

3.4.13 VERDAMPFER (Evaporator)

This class is essentially a constant volume tank. The input stream is reduced by the factor KONZ-FAKTOR, the excess (vapour) leaving via output A2. The concentrated liquor mixes perfectly with the evaporator inventory and exits via A1.

3.4.14 EINGRIFF (Intervention)

This component class permits external interference in the operation of the process, for example the removal (diversion) of material. An EINGRIFF component can be inserted anywhere in the process and is serviced by the MONITOR (see below).

3.5 Operators

The operators for the various cycles of the separation and purification process are subclasses of PROCESS CLASS OPERATEUR. Every operator has essentially the same attributes: procedures defining his sphere of action, permitting him to start and stop parts of the process, perform perodic material balances, identify the origin of messages and alarms and react to them, etc.

3.6 The MONITOR process

This component runs in parallel to, but independently of, the process simulation, and permits dialogue and intervention by the user.

3.7 The main program

In the main program, all process components are declared, general procedures set up, components created and their parameters read in. The components are connected together and, finally, the process is started.

4. Using the program

4.1 Starting the program

To start the program on the KfK IBM computer under TSO, the initialization command

```
NEWPROC    SIM
```

must be given. The program can then be started with

GO input member output file

where "input member" is the member of the data set TSO255.SIM.DATA containing input parameters. "Output file" in the name of the desired protocol file.

Example: GO T1000 * simulates with data from TSO255.SIM.DATA (T1000) and writes the protocol to the terminal.

4.2 Parameter file

At startup the program reads in parameters for the process components via file PARM. The corresponding file containing input parameters must therefore be allocated before program start.

4.3 The monitor

After the user initiates the program and answers the prompt for total simulation duration, the monitor reports with

** MONITOR ** TIME = 0.0

The following commands will then be accepted:

- ENTER - key — The simulation runs DT hours, and then the monitor reappears
- DT real number — Defines DT (default 10 hours)
- Component-ID — Lists the present status of the component to the terminal (inventory, volume, flows to and from the component)
- Component-ID LIST/NOLIST — Switches the protcol option for the component on/off
- Distributor-ID SCHALTE — Switches the distributor to its next output

- Intervention-ID HOLD/GIVE f df c dc — Sets flow and concentration values to be retained (removal) or added to the process (simulation of diversion stategies).

- GRAPH ON/OFF — Switches graphics on/off