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Determination of In-Process Inventory in a Reprocessing Plant by Means of Isotope Analysis

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by

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Introduction

D. Gupta

The importance of independent in-process inventory determination from the point of view of safeguards, has been emphasized in a number of recent publications /‾1, 2, 3_7. The method should be nonintrusive, should enable the safeguards authority to close the material balance over a given time period without appreciable time lag and should be as accurate as practicable. In Ref. /‾3_7, a method for independent determination of in-process inventory was discussed which was based on an intimate knowledge of the characteristics of the various process units of a fabrication plant. This method was found to be less effective in connection with a modern safeguards system /‾2_7 mainly because of the fact that the holdup functions can varry over a fairly wide range for the same throughput in an industrial scale plant.

A different method of independent determination of in-process inventory in a reprocessing plant has been discussed in this paper. The method is based on the measurement of different fissile isotope concentrations in the input and output batches of a reprocessing plant. The first chapter of this paper deals with the principle of this method and the mathematical considerations which form the basis for the determination of the process inventory. Some numerical examples have also been included in this chapter to illustrate the application of this method.

The second chapter describes a typical reprocessing plant (the NFS plant, USA), analyses the internal mixing mechanisms of different processstreams which may influence the inventory determination by this method and establishes mathematical equations to describe these mixing steps. The results of a simulation of these mathematical relations have also been presented in this chapter. The third chapter describes the simulation procedure in an analog computer to simulate the different mixing mechanisms in the NFS reprocessing plant.

The fourth chapter deals with the type of statements which can be made on the basis of the measured data obtained from this method, after considering the error propagation in the measurements.

On the basis of the analyses and the tests carried out in this paper, the method of estimating in-process inventory with the help of isotope analysis, appears to be highly interesting for safeguards purposes. This method will also be of great interest for the plant operators. The analog simulation indicates that such process inventory determinations can be carried out in industrial scale reprocessing plants. However, a large number of inplant experiments have to be carried out to prove the feasibility of this method under operating conditions in such plants.

Chapter I

Principle of Process Inventory Determination by Means of Isotope Analyses

H. Winter

1. Introduction

In most of the presently known reprocessing plants, the input and output operations for nuclear fuels are carried out batchwise. Fig. I-1 shows the schematic representation of such a plant. The input and output transfer can be measured by an inspector at the strategic points I and II. In Fig. I-2 the batchwise input and output operations are indicated.

In the present chapter the mathematical process has been developed, which can be used to determine in-process inventory. The principle of this method lies in the fact that the fissile material process inventory, which is pushed out by the incoming new batches, is measured quantitatively at the exit of the plant by identifying its isotopic composition. This is possible if the concentrations of a limited number of isotopes in the inventory differ sufficiently from those in the input batches which push it out.

The differences in the weight fractions of the isotopes, which are used as tracers may have their origin in different irradiation histories of the fuel elements in subsequent dissolver batches (passive tracing) or may be introduced artificially by adding for example Pu with an appropriate isotopic composition to a dissolver batch (active tracing).

Let $H(t_o)$ /‾kg Pu_7 be the inventory of the plant at the time t_o. The main part of $H(t_o)$ - more than 98 % - leaves the plant as final product, and a small part (less than 2 %) as measured losses. In principle, this part of the inventory which leaves the plant as measured losses can be determined in the same way as the main part by means of an isotope analysis. But in practice it will be sufficient to take the amount of losses measured during a period, which corresponds to the mean residence time of a batch in the plant, as an estimate of this part of the inventory.

2. Preliminary Considerations

The following notations have been introduced:

The input batches in Fig. I-2 are numbered by capital letters
$A, B = 1,2,3,4, \ldots$

the output batches in Fig. I-2 are numbered by small letters
$a, b = 1,2,3,4, \ldots$

the fuel isotopes are numbered by greek letters
$\mu, \rho = 1,2,3$

At the strategic point I of Fig. I-1 the total masses M_A [kg Pu] of the input batches and the isotopic composition of the fuel are measured (c_B^ρ is the weight fraction of the isotope ρ in the input batch B [kg isotope ρ/kg Pu]).

At the strategic point II the corresponding quantities M_a [kg Pu] and c_a^ρ [kg isotope ρ/kg Pu] are measured.

The nominal value of the inventory (book inventory), $J(t_o)$, at a certain time t_o, is

$$J(t_o) = \sum_{A \text{ before } t_o} M_A - \sum_{a \text{ before } t_o} M_a + J_o$$

with J_o as the inventory at the beginning of the campaign.

In Fig. I-2

$$J(t_o) = \sum_{A \leq 2} M_A - \sum_{a \leq 1} M_a + J_o .$$

Statements about a diversion of fuel inside the plant arise from a comparison of the book inventory, $J(t_o)$, which is not influenced by a diversion, with the actual inventory, $H(t_o)$, which is smaller than the book inventory in the case of a diversion.

$H(t_o)$ can be determined in the output of the plant, by measuring after t_o the outcoming amount of fuel which entered the plant before t_o. Therefore, it is necessary to identify the input batches in the output stream. This can be done by means of their isotope characteristic.

3. Identification of the Input Batches in the Output Stream

Let $m_{a,B}$ /‾kg Pu_7 denote the amount of fuel of the input batch B in the output batch a. Then the following equations hold:

$$\sum_{B} m_{a,B} = M_a \tag{I-1}$$

$$\sum_{B} m_{a,B}\, c_B^{\mu} = M_a c_a^{\mu} \quad ; \mu = 1,2, \ldots .$$

The first equation indicates that the sum of all amounts $m_{a,B}$ which are contained in the output batch a, must be equal to the total amount of fuel of this batch. The second one indicates the same fact for the isotope μ. M_a, c_a^{μ} and c_B^{μ} are quantities measured at the strategic points I and II of Fig. I-1.

$m_{a,B}$ are the unknown quantities. (I-1) is a linear system of algebraic equations. This system has a unique solution, if

(a) the number of equations is equal to the number of the unknowns

(b) the coefficient determinant is (sufficiently) different from zero.

The restriction (a) means, that the number of input batches which mix into one output batch must be smaller than n+1, where n is the number of fuel isotopes which are available.

The restriction (b) means, that the row and column vectors of the coefficient matrix must be linear independent. The coefficient matrix of (I-1) has the form

$$\begin{pmatrix} 1 & 1 & \ldots & 1 \\ C_{B=1}^{\mu=1} & C_{B=2}^{\mu=1} & \ldots & C_{B=N}^{\mu=1} \\ \cdot & \cdot & & \\ \cdot & \cdot & & \\ \cdot & \cdot & & \\ C_{B=1}^{\mu=n} & C_{B=2}^{\mu=n} & \ldots & C_{B=N}^{\mu=n} \end{pmatrix}$$

In this case there are n isotopes and N unknowns $m_{a,B}$. Restriction (a) requires $N = n+1$.

The requirement of linear independence of the row and column vectors of the matrix (I-3) restricts the number of fuel isotopes, which can be used for the process inventory determination. This fact will be discussed in a later section for the simplest cases N = 2 and N = 3.

If the requirements (a) and (b) are fulfilled, the linear system (I-1) has a unique solution, which means, that for each output batch the quantities $m_{a,B}$ can be calculated, and the problem of the identification of the input batches in the output stream is solved.

If the process losses of the input batch A are denoted by L_A, then

$$L_A = M_A - \sum_b m_{b,A} \qquad \text{(I-2)}$$

must hold. M_A is the total amount of fuel of the considered batch. By means of the equation (I-2), the process losses of each input batch can be estimated.

If the process losses (less than 2 %) are neglected, then the equation (I-2) (with $L_A=0$) can be used as a rough control of the solutions of the system (I-1).

4. Special Cases

It has been shown in chapter II that the successive input batches may get mixed internally while getting processed in the plant. This mixing will naturally reduce the differences in the isotope concentrations in these batches and make the inventory estimate less accurate. It is therefore necessary to consider some special cases in which typical mixing mechanisms have been included.

In this section two special cases namely, two and three input batches respectively mixing into one output batch have been analysed. An analysis of the operational procedures of a typical reprocessing plant has shown that these cases are fairly realistic and can be easily attained by either minimizing the mixing mechanisms at the product end of the plant or by grouping the input batches in a way to create superbatches (A superbatch consists of several dissolver batches with identical isotopic composition.).

4.1 Mixing of Two Consecutive Input Batches

When only fractions of two input batches (super batches) are contained in one output batch, a, the system (I-1) reduces to

$$m_1^a + m_2^a = M_a \tag{I-3}$$

$$m_1^a c_1 + m_2^a c_2 = M_a x_a$$

where

$$m_1^a = m_{a,B=1}$$

$$m_2^a = m_{a,B=2}$$

$$c_1 = c_{B=1}^{\mu=1}$$

$$c_2 = c_{B=2}^{\mu=1}$$

$$x_a = c_a^{\mu=1}$$

The coefficient matrix of the system (I-3) is

$$\begin{pmatrix} 1 & 1 \\ c_1 & c_2 \end{pmatrix}.$$

The two conditions for a unique solution of the system (I-3), which have been discussed in the third section, require that

(a) one fuel isotope with the weight fractions c_1 and c_2 must be present in the input batches

(b) the coefficient determinant D

$$D = \begin{vmatrix} 1 & 1 \\ c_1 & c_2 \end{vmatrix} = c_2 - c_1$$

is different from zero, which means

$$c_2 \neq c_1.$$

The unique solution of the system (I-3) is

$$m_1^a = \frac{x_a - c_2}{c_1 - c_2} \; M_a \qquad \text{(I-4)}$$

$$m_2^a = \frac{c_1 - x_a}{c_1 - c_2} \; M_a .$$

The weight fractions c_1 and c_2 are measured in the input stream and x_a and M_a in the output stream.

4.1.1 Process Inventory Calculation

Fig. I-3 shows the input and output of a reprocessing plant. The input consists of two superbatches, with 2 wt % of Pu 241 before t_o and 4 wt % after t_o. (As shown in App. 1, it is possible to get such differences in fuel subassemblies for the same reactor.)

Process inventory is equal to the amount of 2 % plutonium, leaving the plant after t_o in this case. In the example (Fig. I-3), the first output batch after t_o consists only of 2 % - Pu. Between t_1 and t_2 three batches with a mixture of 2 and 4 percent plutonium appear in the output stream. After t_2 only Pu with 4 % Pu-241 leaves the plant. For each output batch, the fractions m_1 of plutonium with 2 % Pu-241 and m_2 of 4 % plutonium are given by the equation (I-4). x_a is the actual weight fraction of Pu-241 in an output batch a. x_a is equal to 2 % before t_1 and equal to 4 % after t_2. For $t_1 \leqq t \leqq t_2$

$$2\,\% \leqq x_a \leqq 4\,\%$$

holds.

The process inventory, H (t_o), of the plant at the time t_o - see Fig. I-3 - is then

$$H\,(t_o) = \sum_a M_a \frac{x_a - c_2}{c_1 - c_2} . \qquad \text{(I-5)}$$

On the right hand, this formula contains only such quantities which can be measured at strategic points at the input and output of the plant.

4.1.2 Error Propagation[+)]

The quantities M_a, x_a, c_1 and c_2 can only be measured with a certain inaccuracy. We assume, that the relative mass measurement error, δ_M, and the relative weight-fraction measurement error, δ_c, are constant. The propagation of these measurement errors to the process inventory can be estimated in the following way:

From (I-5) by differentiation:

$$dH = \sum_a \left\{ \frac{x_a - c_2}{c_1 - c_2} dM_a + \frac{M_a}{(c_1-c_2)^2} \left[(c_1-c_2)\, dx_a + \right.\right.$$

$$\left.\left. + (c_2-x_a) dc_1 + (x_a-c_1) dc_2 \right] \right\}.$$

Hence

$$|dH| \leq \sum_a \left\{ |dM_a| + \frac{M_a}{|c_1-c_2|} \left[|dx_a| + |dc_1| + |dc_2| \right] \right\}.$$

With the relative measurement errors

$$\delta_M = \frac{|dM_a|}{M_a} \; ; \; \delta_c = \frac{|dx_a|}{x_a} = \frac{|dc_1|}{c_1} = \frac{|dc_2|}{c_2}$$

we have

$$|dH| \leq \left\{ \delta_M + \frac{2|c_1| + |c_2|}{|c_1-c_2|} \delta_c \right\} \cdot \sum_a M_a \quad \text{for } c_1 > c_2 \qquad \text{(I-6a)}$$

$$|dH| \leq \left\{ \delta_M + \frac{|c_1| + 2|c_2|}{|c_1-c_2|} \delta_c \right\} \cdot \sum_a M_a \quad \text{for } c_2 > c_1. \qquad \text{(I-6b)}$$

In these formulae, the summation is extended over all output batches, which leave the plant after the time t_o and which contain plutonium with the weight fraction c_1 of the isotope in consideration. (In Fig. I-3 the summation goes from t_o to t_2).

+) N.B. In these considerations, the influence of covariances has been neglected. An exact treatment of the propagation of error has been given in chapter IV. However, the following treatment makes the relation between the important measurement errors clear, without reducing too much the correctness of the statement.

In the case where

$$H(t_o) \approx \sum_a M_a$$

which corresponds to a very small mixing of traced and untraced material, equations (I-6a) and (I-6b) reduce to

$$\delta_H = \delta_M + \frac{2\kappa+1}{\kappa-1}\,\delta_c \qquad \text{(I-7)}$$

where δ_H is the relative error of inventory, and $\kappa = c_1/c_2$, with $c_1 > c_2$.

Equation (I-7) shows quite clearly the influence of the mass spectrometric measurement of the weight fractions. The ratio $\kappa = c_1/c_2$ is fixed for a given input signal. The function

$$f(\kappa) = \frac{2\kappa+1}{\kappa-1}$$

which appears as an importance factor for the weight fraction measurement error in (I-7) is shown in Fig. I-4. For $\kappa \to 1$ this factor goes to infinity, which corresponds to the fact, that a distinction of the two types of plutonium is not possible if $c_1 = c_2$. For $\kappa \to \infty$, which corresponds to a very significant step ($c_1 \gg c_2$), the importance factor tends to 2.

Realistic values of κ are in the range of 2 to 10 for the minor Pu isotopes and 1.2 to 2 for Pu-239, Pu-240 and Pu-241 (see App. 1). Thus the measurement of the normal Pu isotopes must be much more accurate than the measurement of the minor isotopes in order to have the same accuracy for the process inventory.

4.2 Mixing of Three Consecutive Input Batches

When three input batches are contained in one output batch, a, the system of equations (I-1) can be written as follows:

$$m_1^a + m_2^a + m_3^a = M_a \qquad \text{(I-8)}$$

$$m_1^a c_1 + m_2^a c_2 + m_3^a c_3 = M_a x_a$$

$$m_1^a d_1 + m_2^a d_2 + m_3^a d_3 = M_a y_a$$

where

$$m_1^a = m_{a,B=1};\ c_1 = c_{B=1}^{\mu=1};\ d_1 = d_{B=1}^{\mu=2}$$

$$m_2^a = m_{a,B=2};\ c_2 = c_{B=2}^{\mu=1};\ d_2 = d_{B=2}^{\mu=2}$$

$$m_3^a = m_{a,B=3};\ c_3 = c_{B=3}^{\mu=1};\ d_3 = d_{B=3}^{\mu=2}$$

$$x_a = c_a^{\mu=1};\ y_a = c_a^{\mu=2}$$

The coefficient matrix of the system (I-8) is

$$\begin{pmatrix} 1 & 1 & 1 \\ c_1 & c_2 & c_3 \\ d_1 & d_2 & d_3 \end{pmatrix},$$

The two conditions for a unique solution of the system (I-8) which have been discussed in the third section, require that

(a) two isotopes with weight fractions c_1, c_2, c_3 and d_1, d_2, d_3 in the three batches must be available,

(b) the coefficient determinant D

$$D = \begin{vmatrix} 1 & 1 & 1 \\ c_1 & c_2 & c_3 \\ d_1 & d_2 & d_3 \end{vmatrix} = c_1(d_2-d_3) + c_2(d_3-d_1) + c_3(d_1-d_2) \qquad \text{(I-9)}$$

must be different from zero. This is fulfilled if

1. all the c_i are not equal, and
2. all the d_i are not equal, and
3. no relation of the type

$$c_i = \alpha d_i + \beta;\ i = 1, 2, 3$$

with α and β constant, holds for the weight fractions c_i and d_i in the three input batches in consideration.

The first and second condition require that there must be differences in the weight fractions of the isotopes in consideration in the three input batches. The third one is a more sophisticated condition. It requires that there is no linear dependence of the isotopic vectors in the input batches. Physically such a linear dependence could be produced for example if the isotopic vector would be a linear function of the burn-up of the fuel elements and if the fuel elements in the input batches have the same initial composition and the same irradiation history with the exception of different burn-ups.

But in general this third condition will be fulfilled because of the different irradiation histories in subsequent dissolver batches.

The solution of (I-8) is

$$m_1^a = \frac{M_a}{D} \left\{ x_a(d_2-d_3) + c_2\ (d_3-y_a) + c_3\ (y_a-d_2) \right\}$$

$$m_2^a = \frac{M_a}{D} \left\{ c_1\ (y_a-d_3) + x_a(d_3-d_1) + c_3(d_1-y_a) \right\} \qquad \text{(I-10)}$$

$$m_3^a = \frac{M_a}{D} \left\{ c_1\ (d_2-y_a) + c_2\ (y_a-d_1) + x_a\ (d_1-d_2) \right\}$$

D is the coefficient determinant, given by (I-9).

All quantities, which appear on the right hand of the equations (I-10) and (I-9) are measured at strategic points at the plant's entrance and exit.

4.2.1 Process Inventory Calculation

Fig. I-5 shows the input and output of a reprocessing plant. The input consists of three superbatches with the isotopic vectors as indicated in this figure.

The process inventory at the time t_o is equal to the amount of plutonium with the isotope vector $c_{Pu\ 41} = 2\ \%$ and $c_{Pu\ 42} = 0{,}5\ \%$, leaving the plant after t_o in this case. It corresponds to the indicated area in the output stream. For each output batch, a, an analysis as desbribed in the preceding section has to be carried out, to determine the quantities m_1^a, m_2^a and m_3^a. Then the process inventory, H (t_o), of the plant at the time t_o is

$$H\ (t_o) = \sum_a M_a \frac{x_a(d_2-d_3) + c_2(d_3-y_a) + c_3(y_a-d_2)}{c_1(d_2-d_3)+c_2(d_3-d_1)+c_3(d_1-d_2)} \qquad \text{(I-11)}$$

The transfer-masses M_a and the weight fractions x_a and y_a are measured at the plant's exit and c_i and d_i are measured at the entrance of the plant.

5. Process Inventory of the Model Reprocessing Plant

The results of the theoretical considerations of the preceding sections have been used to determine the process inventory of the reprocessing plant which has been analysed and simulated in chapters II and III.

5.1 Step Function Input Signal

The simulated reprocessing plant is described in detail in chapter II. In Fig. I-6 the input signal (weight fraction of Pu-241) and the response of the simulated plant with minimum hold-up is shown.

In this case, the initial weight fraction of Pu-241 ($c_{Pu-241} = 4\ \%$) is raised to 8 % in all batches which enter the plant after t = 3d. This signal, together with the system response allows the calculation of the process inventory at the time t = 3 d. For this purpose, the masses m_1^a and m_2^a of both types of Pu in the input stream have to be calculated for each output batch a, by means of (I-4). The result of this calculation is shown in the following table. (M_a = 0.6 kg Pu, which corresponds to a throughput of 1 kg Pu/d).

a	0	1	2	3	4	5	6	7	8	9
m_1^a /‾kg Pu_7	0.600	0.570	0.368	0.188	0.113	0.053	0.030	0.015	0.008	0.
m_2^a /‾kg Pu_7	0.	0.030	0.232	0.412	0.487	0.547	0.570	0.585	0.592	0.600

The process inventory is then calculated by means of (I-5)

$$H\ (t = 3d) = 1.345 + 0.400\ /\overline{\ }\text{kg Pu}\_7.$$

The first term in this equation corresponds to the amount of Pu with $c_{Pu-241} = 4\ \%$ in the output batches 1 to 8. The second term corresponds to the fraction of the output batch 0, which leaves the plant after t = 3d. Thus

$$H\ (t = 3d) = 1.745 \text{ kg Pu} \qquad \text{(I-12)}$$

In order to show, that this calculated value of the process inventory within the error limits is equal to the real value known by the simulation, an error estimation has been carried out, assuming that

(a) the input signal is known exactly (The input signal could be adjusted with an accuracy better than $1^{o}/oo$ in the analog computer.)

(b) the frequency of the output batches are known exactly (adjusted in the analog computer)

(c) the system response, given by the plotter of the analog computer is given with a maximum error corresponding to ± 0.05 wt %.

The maximum error of H (t = 3d) is then

$$\Delta H \leq 0.06 \text{ kg Pu}. \qquad \text{(I-13)}$$

This corresponds to a maximum error

$$\frac{\Delta H}{H} \leq 3.4\ \%. \qquad \text{(I-14)}$$

The real inventory of the simulated plant was known by the simulation to be 1.695 kg, which is in agreement with the process inventory as calculated by an isotope analysis and with the error limit given above.

5.2 Double Step Function Input Signal

In Fig. I-7 the input signals (weight fractions of Pu-241 and Pu-242) and the corresponding responses of the simulated plant (with maximum hold-up) are shown.

In this case, the initial weight fractions of Pu-241 and Pu-242 ($c_{Pu-241} = 0$ %, $c_{Pu-242} = 0.18$ %) are raised to 1.8 % and 0.4 % respectively at the time t = 1d. At t = 2d these weight fractions change once more ($c_{Pu-241} = 4$ %, $c_{Pu-242} = 0$ %).

These signals, together with the system responses allow the calculation of the process inventory at the times t = 1 d and t = 2 d. The calculations have been performed for t = 1d.

For this purpose, the masses m_1^a, m_2^a and m_3^a have to be determined for each output batch, a, corresponding to the equations (I-10).

In this particular case, the weight fractions of the input signal are

$c_1 = 0$ % ; $d_1 = 0.18$ %

$c_2 = 1.8$ % ; $d_2 = 0.40$ %

$c_3 = 4.0$ % ; $d_3 = 0.$ %

The weight fractions of Pu-241 and Pu-242 in the output are read from Fig. I-7. From (I-9) one obtaines D = -1.204 and M_a = 3 kg Pu for a throughput of 1 kg Pu/d. The result of the calculation is given in the following table

a	0	1	2	3	4	5	6
m_1^a /¯kg Pu_7	3.000	2.741	0.732	0.239	0.045	0.022	0.
m_2^a /¯kg Pu_7	0.	0.267	0.645	0.042	0.055	0.027	0.
m_3^a /¯kg Pu_7	0.	0.	1.622	2.718	2.900	2.950	3.000

The process inventory is then calculated by means of (I-11)

$$H\ (t = 1d) = 3.779 + 1.0 \quad [\text{kg Pu}]$$

The first term gives the amount of Pu with $c_{Pu-241} = 0\ \%$ and $c_{Pu-242} = 0.18\ \%$, which is contained in the output batches 1 to 5. The second term corresponds to the fraction of the batch No. 0, which leaves the plant after t = 1d.

Thus:

$$H\ (t = 1d) = 4.779 \pm 0.386 \quad [\text{kg Pu}]. \qquad \text{(I-15)}$$

The error analysis is based on the same assumptions as in the preceding section, with errors of ± 0.05 wt % and 0.005 wt % respectively for the Pu-241 and Pu-242 weight fractions given by the plotter of the analog computer.

This is in a good agreement to the real value of the process inventory of 4.658 kg Pu, known from the simulation data.

5.3 Influence of Measuring Errors on Process Inventory Determination

The above numerical examples have shown that there is fairly good agreement between the values of process inventory determined by an isotope analysis and simulated by the analog computer. The accuracies with which such process inventories can be expected to be estimated, when different measuring errors have been taken into consideration are shown in the tables A and B. The influence of neglecting the covariance terms in error propagation has also been estimated.

As a numerical example the case described in the section 5.1 has been taken. The propagation of measurement errors has been calculated

(a) with the help of equation (I-7) of this chapter

(b) with the help of equation (IV-35) of chapter IV and neglecting the covariance terms

(c) with the help of equation (IV-35) of chapter IV with the covariance term.

Table A: Assumptions about measuring accuracy

Type of measurement	Relative accuracy = σ /expectation value	
	Symbol	Adopted values /⁻%_7
Transfer-mass determination	δ_M	0.1
Wt % determination	δ_c for Pu 241	0.1; 0.5; 1; 5

Table B: Accuracy of process inventory for the cases a, b and c (numerical example of section 5.1)

Relative accuracy of measurement	Equation used for error propagation - case -	Relative accuracy of process inventory
$\delta_M = 0.1$ % $\delta_c = 0.1$ %	a b c	0.60 % 0.24 % 0.44 %
$\delta_M = 0.1$ % $\delta_c = 0.5$ %	a b c	2.60 % 0.52 % 1.91 %
$\delta_M = 0.1$ % $\delta_c = 1$ %	a b c	5.10 % 0.73 % 3.74 %
$\delta_M = 0.1$ % $\delta_c = 5$ %	a b c	25.10 % 1.64 % 18.40 %

The last table shows the importance of the covariance term in the equation (IV-35) of chapter IV.

The results in the table B have been represented in Fig. I-8. This figure shows that

1. the relative accuracies of the process inventory calculated by the relatively simple method described in this chapter (I-7) and by a statistical method, which has been described in chapter IV (IV-35) are in good accordance. The values calculated with the equation (IV-35) are 27 % smaller than those calculated with the equation (I-7), which corresponds to the fact, that the latter equation gives a very pessimistic estimate of the inventory accuracy.
2. in a statistical treatment of the error propagation, the covariance term plays a key role. The importance of this term increases with increasing inaccuracy of the weight fraction measurement.

It has been shown in appendix 1, that differences in the weight fractions of Pu isotopes in subsequent dissolver batches in the range of 20 % to 900 % can be expected. In this range of variation similar conclusions as in table B and Fig. I-8 can be drawn.

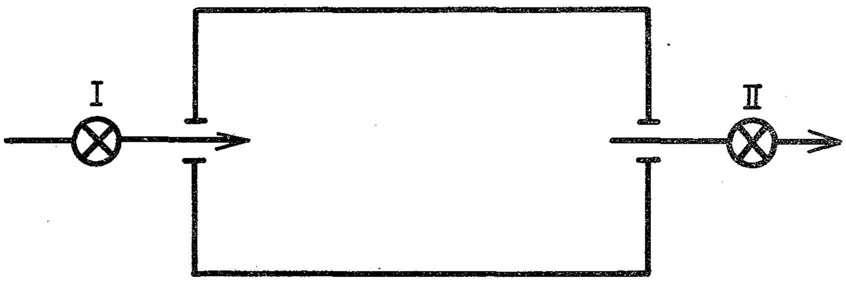


Fig.I-1 Schematic Representation of a Reprocessing Plant with Strategic Points I and II

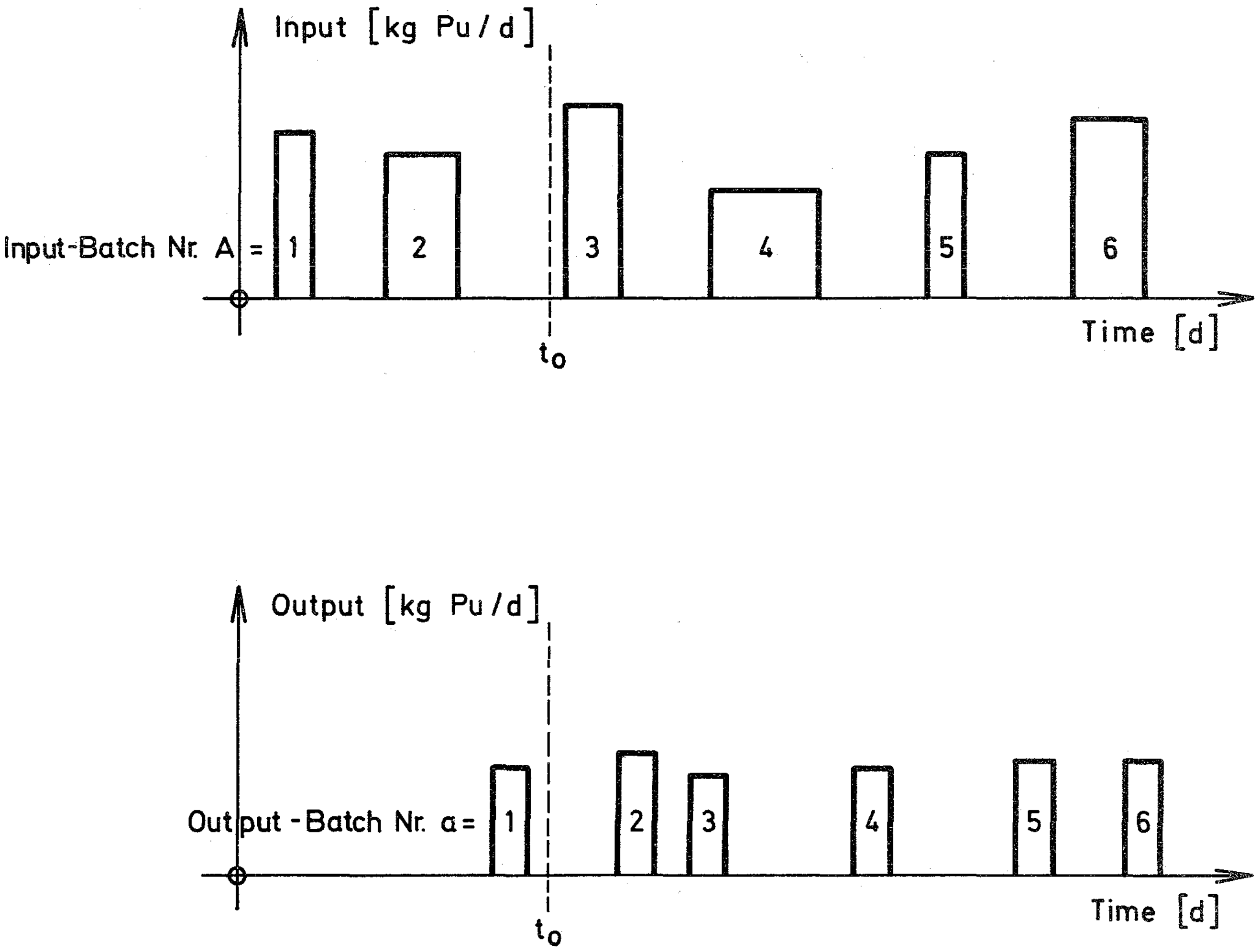


Fig.I-2 Batchwise Input and Output of a Reprocessing Plant

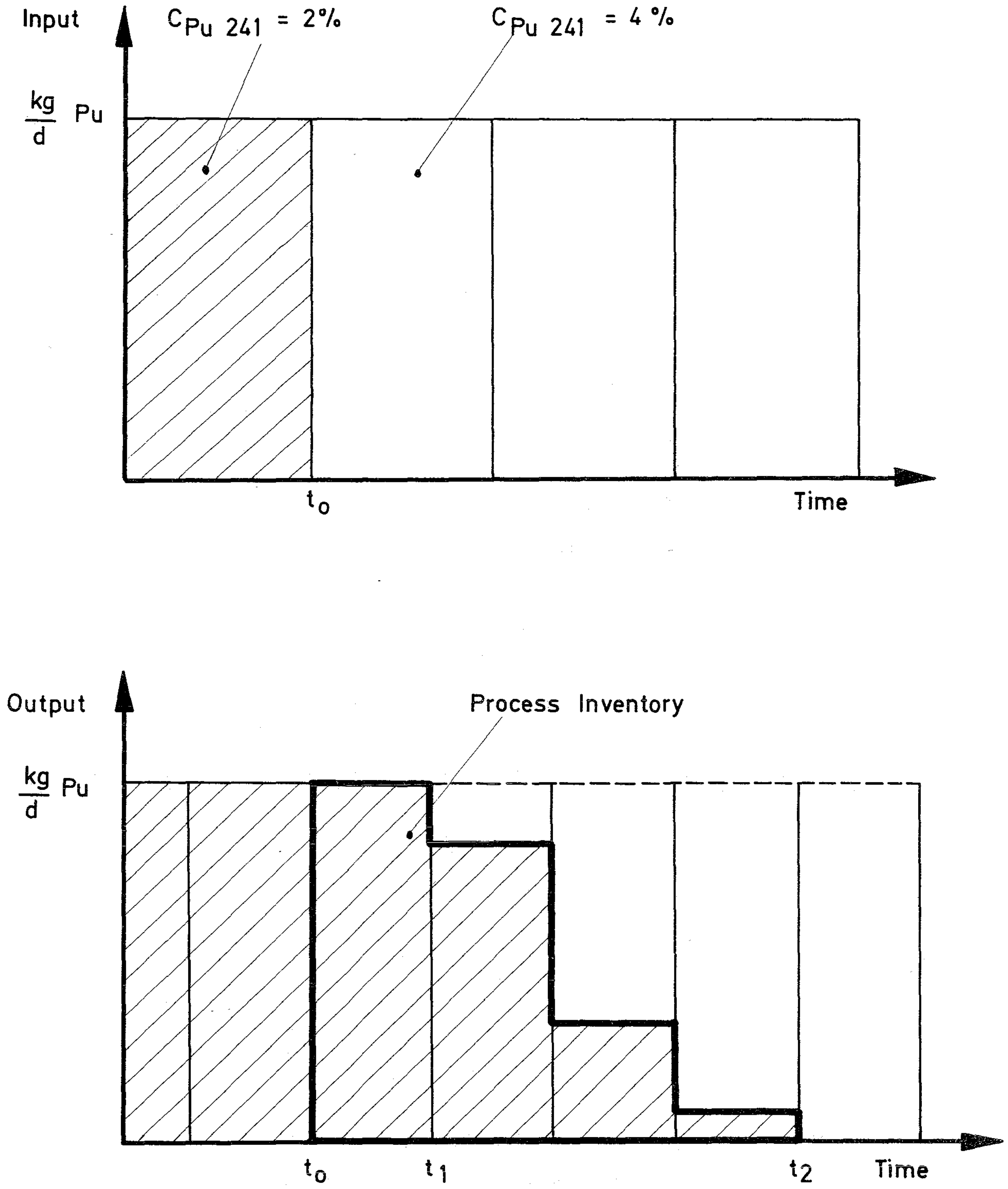


Fig. I-3 Process Inventory Calculation

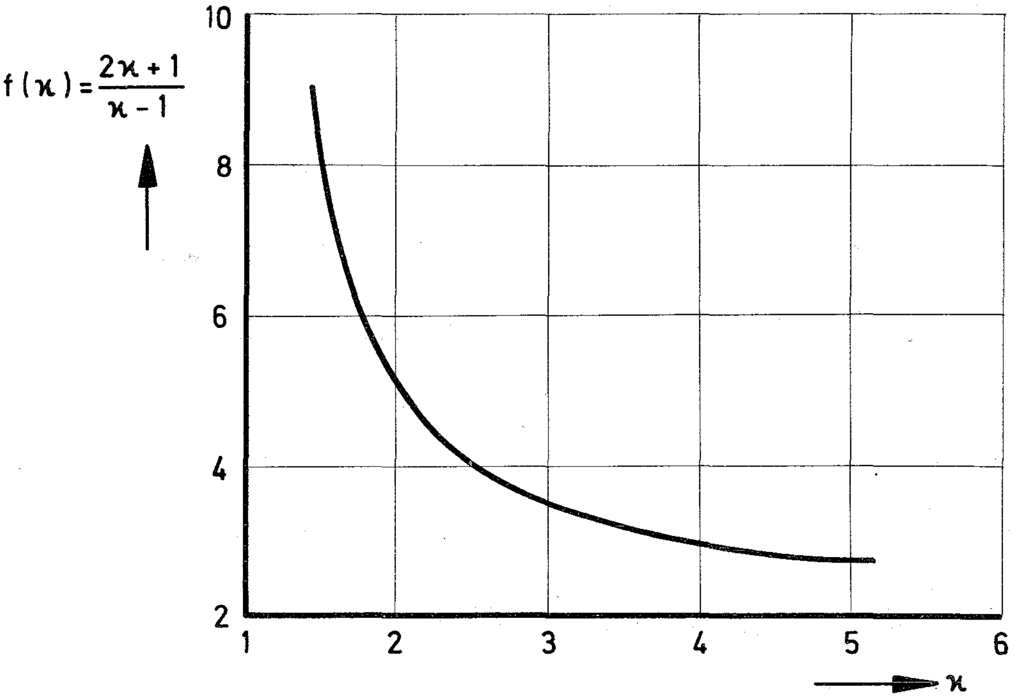


Fig.I-4 Importance Factor f (x) of the Weight Fraction Measurement Error.

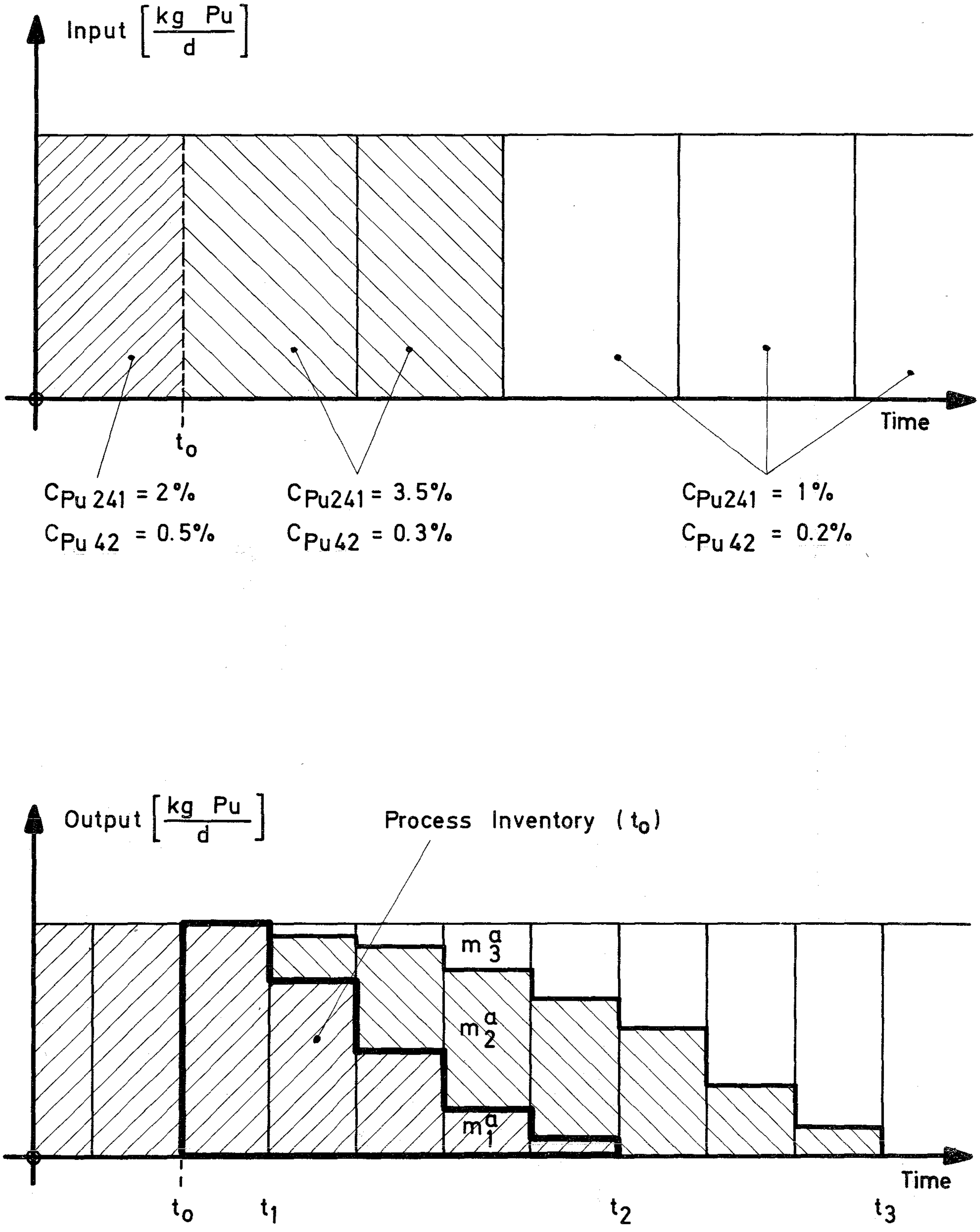


Fig. I-5 Process Inventory Calculation for three Types of Fuel

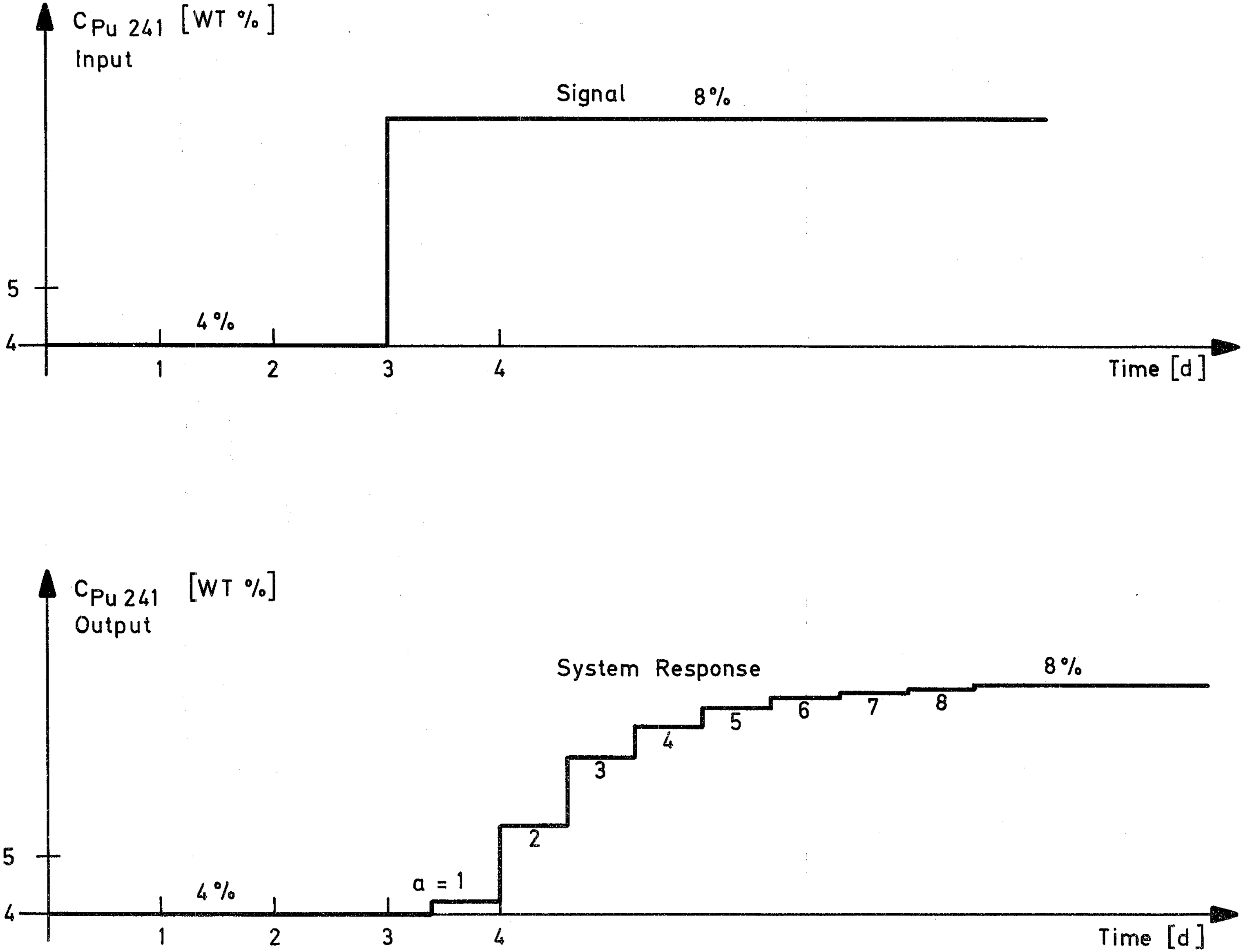


Fig. I-6 Variation of the Pu 241 Weight Fraction in the Input and Output Streams of the Simulated Reprocessing Plant, with Minimum Hold-ups.

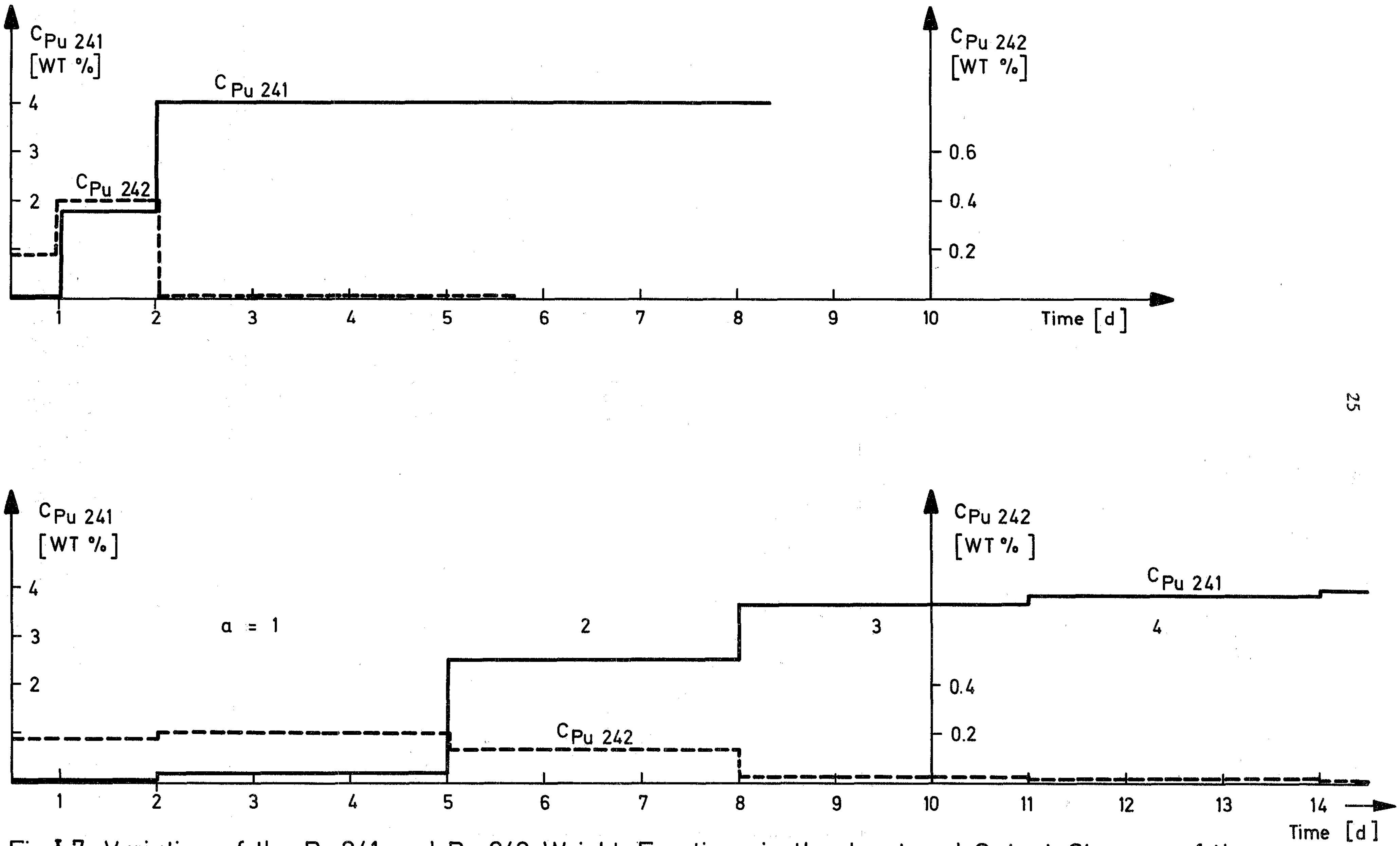


Fig.I-7 Variation of the Pu 241 and Pu 242 Weight Fractions in the Input and Output Streams of the Simulated Reprocessing Plant with Maximum Hold-ups.

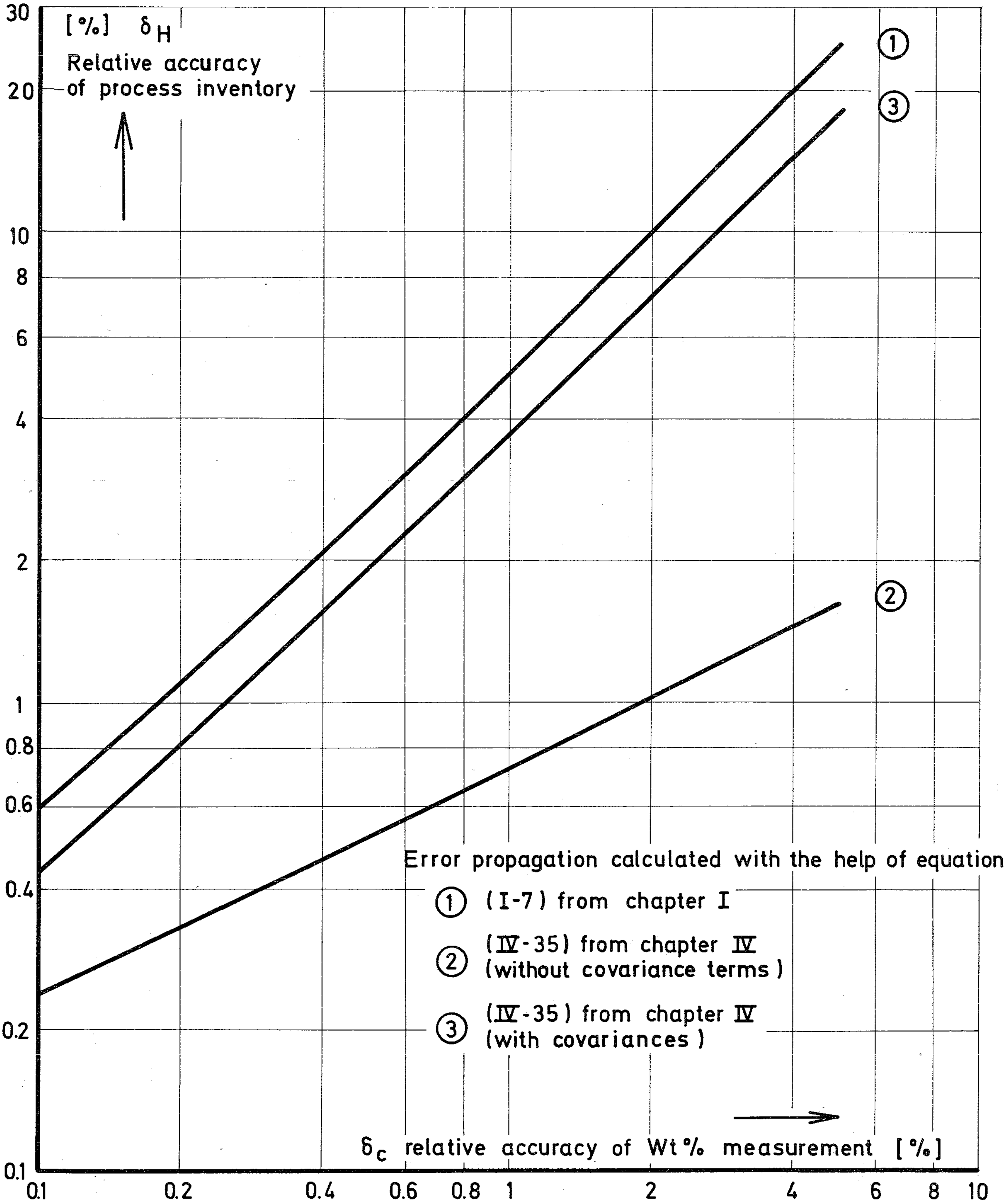


Fig. I-8 Accuracy of Process Inventory Determination as a Function of Wt % Measurement Accuracy

Chapter II

Analysis of a Typical Reprocessing Plant

R. Kraemer

The NFS plant, which has been under comercial operation for a number of years was chosen to test the theory of inventory determination by means of isotopic analysis, as developed in chapter I.

1. Description of the NFS Plant

The major process steps in this plant can be devided into the following five categories:

a) Separation: Canning material - fuel

b) Separation: Fission products - uranium and plutonium

c) Separation: Uranium - plutonium

d) End purification of uranium

e) End purification of plutonium

Before the first step, the fuel is stored in a water storage basin. A short analysis of all the steps is given in the following paragraphs. These steps are shown symbolically in Fig. II-12.

1.1 Fuel Reception and Storage

The irradiated fuel elements are unloaded from the transport casks under water and stored in water basins.

1.2 Mechanical Processing

The head and the bottom ends of the subassemblies are separated in a special cell and the rest of the subassemblies are chopped with a special machine. The chopped pieces are collected in a perforated basket which can be loaded directly into the dissolver tank for dissolution purposes.

1.3 Dissolution and Feed Adjustment

The fuel including the fission products is dossolved in the dissolver tank with the help of 4.5 Moler nitric acid. The dissolution is carried out batchwise and takes about 8 - 14 hours. The leached hulls in the basket are removed from the dissolver. The solution after dissolution is pumped to the accountability tank in which an exact measurement of the volume and the density is carried out. There is also possibility of sample-taking in this tank. After determination of the chemical composition and characteristics of the solution in the accountability tank, the solution is transferred to a feed tank in which it is finally adjusted to the process conditions. From this feed tank the solution is fed continuously to the extraction cycle.

1.4 Partition Cycle

The adjusted high-active feed solution (HAF) is sent to the so called HA column in which it is brought in intimate contact with the organic phase (HAX). The uranium and plutonium are separated from the fission products, which leave the partition cycle in the form of waste stream (HAW). Traces of uranium and plutonium are present in this stream and form a part of the losses of the fissionable material in the whole process. The organic phase containing uranium and plutonium (HAP) are sent to a second column called HB column in which plutonium is separated from uranium and the two streams are sent to the two end purification stages respectively.

1.5 First and Second Uranium Cycle

The uranium stream (HBU) is sent over a number of columns (HC, IA, IC, ID, IE) to decontaminate it to the required degree and finally, sent to a silica-gel column, in which a further decontamination is attained. After the silica-gel stage, purified and decontaminated uranium in the form of uranium nitrate with a concentration of about 350 g U/l is stored in the final storage area.

1.6 Plutonium Cycle

The aqueous phase containing the plutonium (HBP) is purified in two further columns and finally passed through an ion exchange column to attain the required decontamination factor. After the ion exchange column the solution is evaporated and concentrated to about 250 g Pu/l and stored in special containers.

1.7 Solvent and Acid Recovery

As in most of the reprocessing plants based on aqueous reprocessing, the acid and the organic solvent used in the process are also recovered according to standard methods. Because of the presence of traces of fissile material in these two streams, whenever acid or solvent is used back into the process a certain amount of fissile material is also recycled into the system.

2. Analysis of Mixing Mechanism

It was indicated in chapter I that the concentrations of a number of isotopes in the input batches have to be sufficiently different (the difference is determined mainly by the accuracies with which these isotopes can be measured) from those in the process inventory which is to be estimated. As shown in App. I, fairly large differences in isotopic concentrations in fuel elements coming from the same reactor can be obtained. In case the reprocessing plant did not have any internal mixing of process streams, the concentration step signal introduced with the input batch at the entrance of a plant, could be obtained unaltered at the exit of the plant. In practice however, a number of mixing mechanisms occur inside a plant. The net result of these internal mixing mechanisms is a degradation of the step signal introduced at the entrance which results in a reduction of the accuracy with which the process inventory can be determined in the extreme case, if the internal mixing mechanisms reduce the concentration difference of a given isotope below the accuracy with which the isotope can be measured, the process inventory cannot be determined. It is therefore, of importance to analyse all possible mixing mechanisms which may degrade an input signal and find out means of reducing their influence.

For this purpose the different possible mixing mechanisms were investigated for the NFS plant and mathematical expressions were developed for each of the mixing mechanisms. These mechanisms were then simulated for the NFS plant in an analog computer and the type of degradations of the input signals were analysed in the simulated plant. This study was carried out with the following objectives in view:

a) to check the use of process-inventory determination under realistic conditions

b) to determine the spread of the tracer-signal-system-response

c) to find out the process parameters, which cause the largest spread of the signal

d) to find the optimum input signal for the use of process inventory-determination.

A detailed analysis of the NFS plant operation indicated that there are four basic units with different mixing mechanisms:

1. Batchwise operation tanks (accountability tank)
2. Buffer tanks in which feed-in and feed-out may take place continuously
3. Continuously operating units
4. Integrating tanks (for example plutonium product storage tank)

2.1 Batchwise Operating Unit

This unit represents a batchwise operating tank such as an accountability tank for transfer measurements or a feed tank, which is filled batchwise and which feeds continuously or batchwise to a subsequent process step.

Fig. II-1 represents such a batchwise operating unit. The amount M_i [kg U or Pu] of U or Pu with the weight fraction c_i^{μ} [%] of the isotope μ is filled batchwise in the tank, in which a certain amount of heel volume from the previous filling still remains. This contains R kg U or Pu normally of a different isotopic weight fraction $(c^{\mu}_{i+1})_{n-1}$ which is identical with the isotopic vector of the former filling.

This heel volume is caused by the steamjet- or airlift-system, which can not empty the tank completely. The emptying of the tank may be carried out batchwise with an amount M_{i+1}, which is mostly of the same size as M_i, or continuously with a constant flowrate k_{i+1} [kg U or Pu/d].

The type of mixing, which takes place in this unit is described by a linear mixing equation of the following type giving the weight fraction of the isotope in the outgoing material after the n^th input batch.

$$(c^{\mu}_{i+1})_n = \frac{M_i\,(c^{\mu}_i)_n + R\,(c^{\mu}_{i+1})_{n-1}}{M_i + R} \quad [\%] \qquad \text{(II-1)}$$

2.2 Buffer Unit

The second reprocessing unit represents feed- or buffer-tanks between two columns or continuously operating evaporators.

Such a buffer tank is shown in Fig. II-2. The nuclear material is filled into the tank with a constant flowrate k_i /‾kg/d_7 but in general with a time variable weight fraction $c_i^\mu(t)$ of the isotope μ. At the same time the nuclear material leaves the tank at a flowrate of k_{i+1} /‾kg/d_7 with the time variable isotopic weight fraction $c_{i+1}^\mu(t)$. As k_i should be equal to k_{i+1} in routine processing, the level of the buffer volume in the tank stays constant and so does the amount of nuclear material R /‾kg U or Pu_7, which is dissolved in this volume.

The resulting weight fraction of the isotope μ in the outgoing flow may be described by a linear differential equation.

$$\frac{d\, c_{i+1}^\mu}{dt} = \frac{k}{R} \left(c_i^\mu - c_{i+1}^\mu\right) \qquad \text{(II-2)}$$

This equation holds, provided the input stream mixes immediately and homogeneously with R. If this is not the case a certain value of $R' < R$ has to be worked out, which is a fictitious amount of nuclear material meeting the mixing conditions mentioned above.

If $R \rightarrow \infty$, the differential quotient goes to zero, meaning that there is no change in c_{i+1} and if $R \rightarrow 0$, $c_{i+1} \rightarrow c_i$. In case of a step function of $c_i(t)$ system-responses $c_{i+1}(t)$ are observed as they are shown in Fig. II-3.

Feed tanks which feed batchwise operating units (for example: an evaporator,) have a time variable buffer volume (Fig. II-4).

Equ.(II-2) still holds with sufficient accuracy if R is replaced by the mean value $\overline{R}$.

2.3 Continuous Working Unit

The third type of mixing takes place in continuously operating units such as a column or a mixer settler. In these units only small amounts of nuclear material are mixed inside a continuous flow by diffusion (Fig. II-5).

The column is fed continuously at a flowrate k_i [kg/d] of nuclear material with the time variable weight fraction c^μ_i of the isotope μ. After a certain residence time the input signal c^μ_i appears at the exit of this unit c^μ_{i+1} deformed by internal mixing mechanisms such as diffusion. The deformation of a step function in c^μ_i is qualitatively shown in Fig. II-6.

This type of mixing may be approached by a second order differential equation:

$$T_2^2 \ddot{c}^\mu_{i+1} + T_1 \dot{c}^\mu_{i+1} + c^\mu_{i+1} = c^\mu_i \qquad \text{(II-3)}$$

The time constants indicate the magnitude of the diffusion and the mean residence time of the nuclear material in the column. $T_1/2T_2 > 0.7$ because there is physically no overswinging of c^μ_{i+1} over c^μ_i possible. Preliminary analog simulations indicated that the mixing effect from this mechanism is very small compared to the mixing mechanisms in other units.

2.4 Integrating Unit (Fig. II-7)

The fourth type of mixing is caused in a collecting tank at the product end. In such tanks, a continuous flow k_i [kg/d] of nuclear material is collected during a certain filling time T and then switched over to a product tank in parallel, which is generally of the same size. The same mechanism takes place in the ion exchange unit of the Pu-cycle or in batchwise operating evaporators. Since such product tanks are normally designed to store an amount of fissile material (for example Pu) which is considerably larger than that present in a single input batch, a very large degradation of the input signal $c^\mu_i(t)$ may occur through mixing in such tanks. The influence of such mixing can be reduced by either reducing the number of input batches to be stored in such tanks, or by increasing the size of the input batch by creating super batches.

The resulting weight fraction of the isotope at the exit of this unit after the n^{th} filling time T is givenby the following equation

$$(c^\mu_{i+1})_n = \frac{k_i \int_{nT}^{(n+1)T} c^\mu_i \, dt + R \, (c^\mu_{i+1})_{n-2}}{k_i T + R} \quad [\%] \qquad \text{(II-4)}$$

The remaining heel, which has to be considered in this mixing-equation, has a weight fraction of the isotope μ, which is identical to the value of two batches before because there are two units of the same size operating in parallel.

Some other mixing operations, such as acid recycle or recycle of a part of the process stream to a previous step, have not been considered here. Their influence is either negligible on the total mixing effect or they do not occur under normal operating conditions.

2.5 Mathematical Model of a Reprocessing Plant

With the help of the four basic units discussed under 2.1-2.4. the mixing mechanisms in any reprocessing plant based on aqueous processing, can be simulated in an analog (or a digital) computer. The change in the input step signal (represented by two different weight fractions of a given isotope in two consecutive input batches) can then be traced through each of these units and the total degradation in this signal at the output of the plant can be estimated.

This type of simulation has been carried out for the Pu-main flow through the NFS Reprocessing Plant (Fig. II-8). The units in Fig. II-8 which are identified by the NFS-code, are described in Table II-1.

Based on these data, the following set of equations has been established in order to describe the time-variable isotopic weight fraction (c) in the Pu-mainflow (k) running through the plant under equilibrium-conditions (k = const = 5 kg Pu/d). The subscripts refer to Fig. II-8

$$c_1 = \text{input signal}$$

$$c_2 = \frac{R_1 c_1^n + R_2 c_2^{n-1}}{R_1 + R_2}$$

$$c_3 \approx c_2$$

$$c_4 \approx c_3 \approx c_2$$

$$c_5 \rightarrow \frac{dc_5}{dt} = \frac{k}{R_5} \quad (c_4 - c_5)$$

$$c_6 \approx c_5$$

$$c_7 \approx c_6 \approx c_5$$

$$c_8 \rightarrow \frac{dc_8}{dt} = \frac{k_8}{R_8} \; (c_7 - c_8)$$

$$c_9 = \frac{1}{T_9} \int_{nT_9}^{(n+1)T_9} c_8 \, dt$$

T_9 = filling time of the ion-exchangers

$$c_{10} \rightarrow \frac{dc_{10}}{dt} = \frac{k}{R_{10}} \; (c_9 - c_{10})$$

$$c_{11} \rightarrow \frac{dc_{11}}{dt} = \frac{k}{R_{11}} \; (c_{10} - c_{11})$$

$$c_{12}^{n} = \frac{k \int_{nT_{12}}^{(n+1)T_{12}} c_{11} dt + R_{12} c_{12}^{n-2}}{k \; T_{12} + R_{12}}$$

T_{12} = filling time of the product storage tanks

This set of equations has been simulated by an analog computer in chapter III.

2.6 Results of the Simulation

In Figs. II-9, II-10 and II-11 typical simulation results have been shown. Initial analyses indicated that the deformation of the input signals is influenced mainly by the mode of operation of the partition cycle feed tank and the product storage tank. The following modes of operation of the two tanks were assumed for the simulation:

1) Minimum hold-up: This means, that the level of the partition cycle feed tank is reduced to 10 % before it is filled with a new batch and that the product tanks are only filled up to 20 % of their maximum capacity.
2) A routine hold-up: This means, that both the tanks are operated according to the routine procedures laid down for the operation, namely:

a) Minimum level of the partition cycle feed tank = 40 %

b) Maximum level of the product tank which corresponds to 3 days operation.

There are always three curves for each parameter variation, which represent the system responses at different points in the plant (the indices refer to Fig. II-8).

In Fig. II-9 the simulation was carried out by introducing two different isotopic weight fractions of one Pu-isotope in the form of a step input function in two consecutive super input batches. In Figs. II-10 and II-11 a double step function was introduced which represents one different input batch between two super batches. In this case one requires two minor isotopes for process inventory determination as three different materials may get mixed in one product batch.

In order to compare the mixing effects of the different units in the plant, a rate of mixing was defined and the results of the simulation were worked out and have been summarized in Table II-2. The figures in this table show clearly that the largest reduction in mixing is effected by changing the routine operation of the product tank. In addition to these figures the range of possible numbers (n) of input batch-fractions in one product batch are also given in Table II-2. It has been shown in Chapt. I that the following basic condition has to be fulfilled for the determination of a process inventory:

$n - 1 = K$ = number of minor plutonium isotopes which
are available in the solution.

The number n as shown in Table II-2 indicates clearly that a process inventory determination by means of isotope analysis for routine operation is only possible, if superbatches can be created, as the above mentioned condition is not fulfilled in the case of $n = 12$. But if one superbatch contains $m > \frac{n}{K+1}$ input batches, this requirement can easily be fulfilled.

Table II-1 :Reprocessing Units of the Pu-main flow in the NFS Reprocessing Plant

NFS-Code	Description	Type of basic unit given in chap.	Mode of flow		Vessel capacity [l]	Pu-concentration [g/l]	Vessel residue [l] Routine/Min.	Residue Identification [gr Pu]
			in	out				
3D-1	Accountability and Feed adjustment tank	2.1	batch	batch	9500	1.5-2	4000/2	R_1
4D-1	Partition cycle feed tank	2.1	batch	cont.	8700	1.5-2	4000/450	R_2
4C-1	HA/Partition cycle extraction column	2.3	cont.	cont.	neglected	-	-	-
4C-2	HBX/Partition cycle partition column	2.3	cont.	cont.	neglected	-	-	-
4C-3	HBS/Partition cycle Pu-scrub column	2.3	cont.	cont.	neglected	-	-	-
4D-6	Pu-cycle feed conditioner	2.2	cont.	cont.	570	1.5-2	220/60	R_5
4C-7	IIA/Pu-cycle extraction column	2.3	cont.	cont.	neglected	-	-	-
4C-8	IIB/Pu-cycle strip column	2.3	cont.	cont.	neglected	-	-	-
5D-1	Pu I-X Feed conditioner	2.2	cont.	cont.	80	8	40/8	R_8
5C-1A/B/C	Pu-ion-exchangers	2.4	cont.	batch	-	-	2000 gr.	-
5D-4	Pu-product evaporator feed tank	2.2	batch	cont.	140	25	60/5	R_{10}
5C-2	Pu-product evaporator	2.4	batch	batch	22	25/220	10/-	R_{11}
5D-5A/B	Pu-product storage tank	2.4	batch	batch	140	220	120/5	R_{12}

Table II-2 : Rate of Mixing[1] Caused by Different Units in the NFS-Plant

	Feed tank 4D-1	Ion Exchange 5C-1A/B/C	Product Tank 5D-5A/B	Total[3]
Min.Hold-up	2,2 %	8,8 %	19,2%	30,2 % $2 \leq n \leq 4$[2]
Routine Hold-up	13,2 %	8,8 %	78 %	100 % $5 \leq n \leq 12$[2]

1) Rate of Mixing = Amount of traced material mixed with untraced material divided by the amount of traced material mixed with untraced material for routine hold-up after its passage through the whole plant.

2) n = Number of the fractions of input-batches, which may be contained in one product batch.

3) Rate of mixing after a passage of traced material through the whole plant.

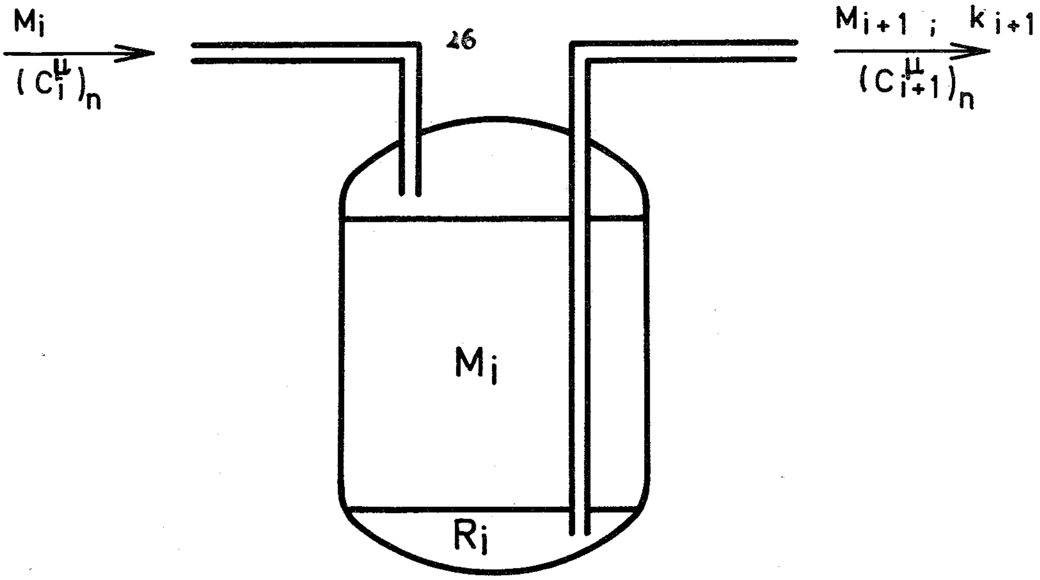


Fig. II-1 Batchwise Operating Unit

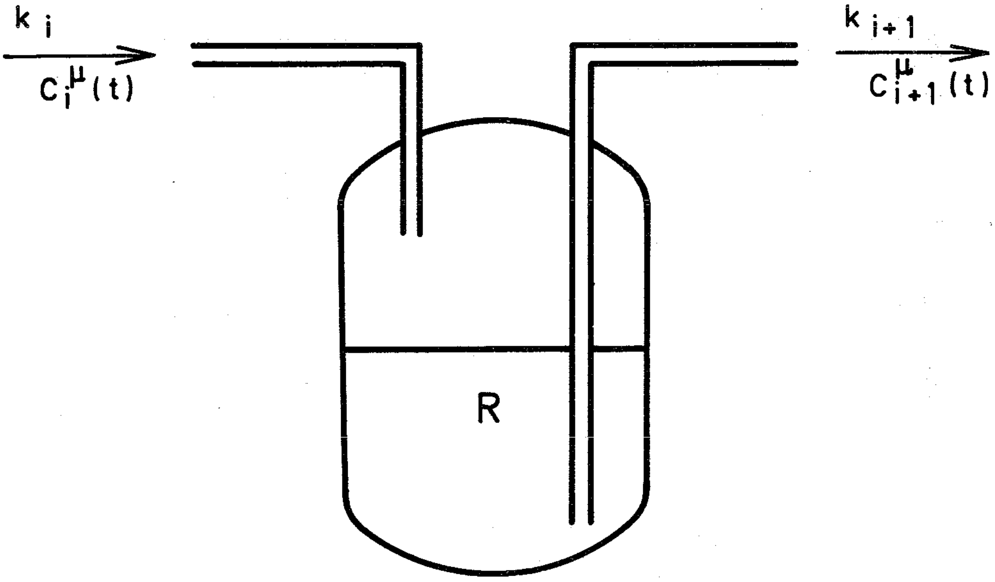


Fig. II-2 Buffer Unit

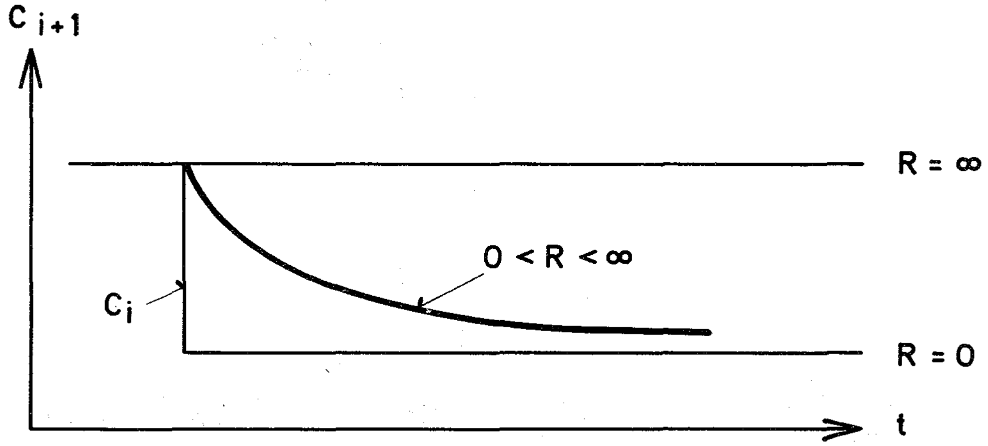


Fig. II-3 System Response to a Step-function in a Buffer Unit

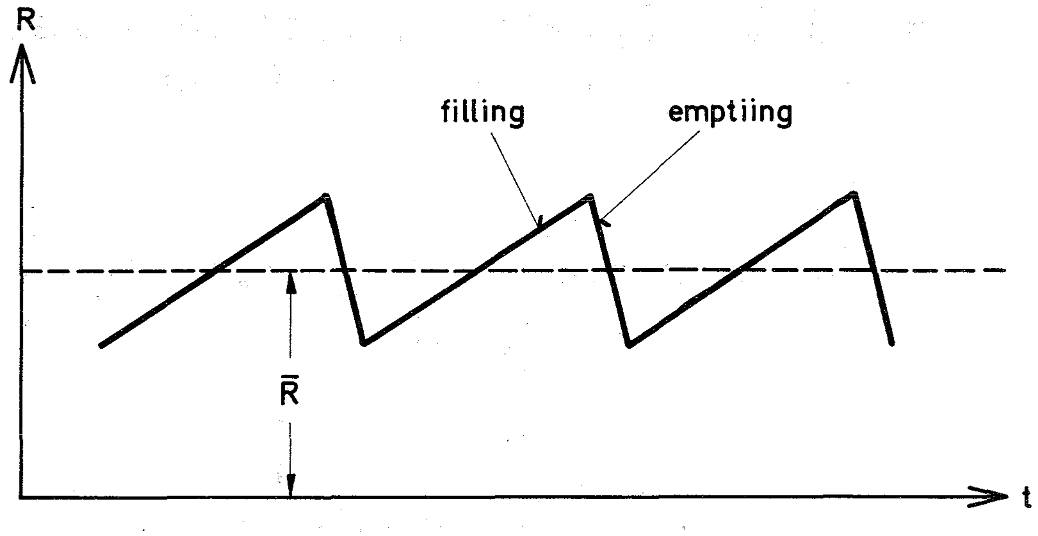


Fig. II-4 Characteristic of the Residue in a Batchwise Emptied Feed tank

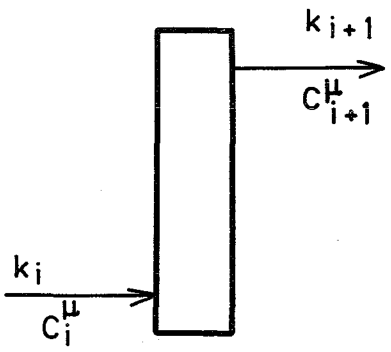


Fig. II-5 Continuously Working Unit

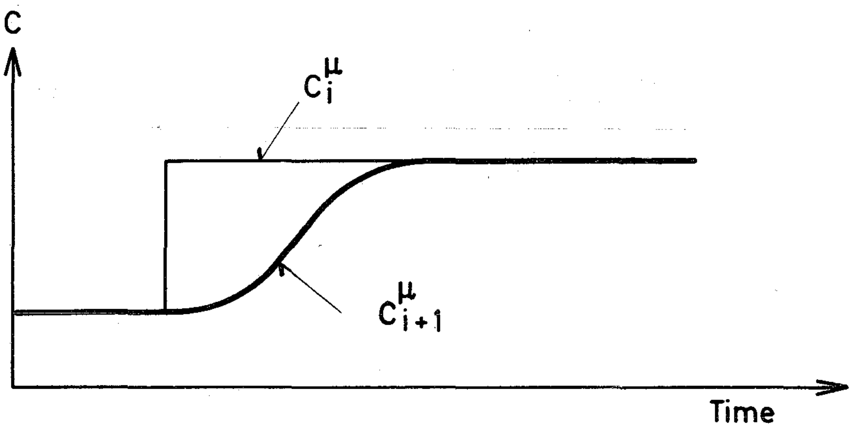


Fig. II-6 System Response to a Step-function in a columm

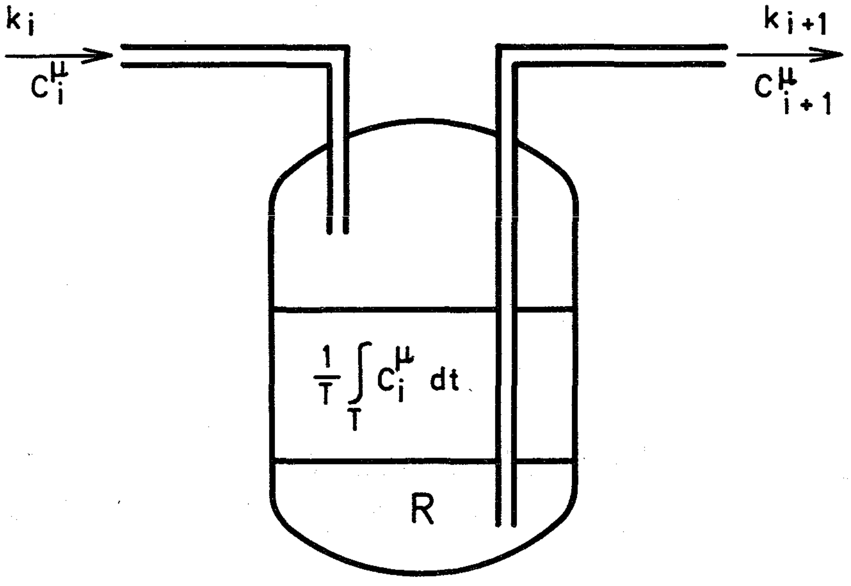
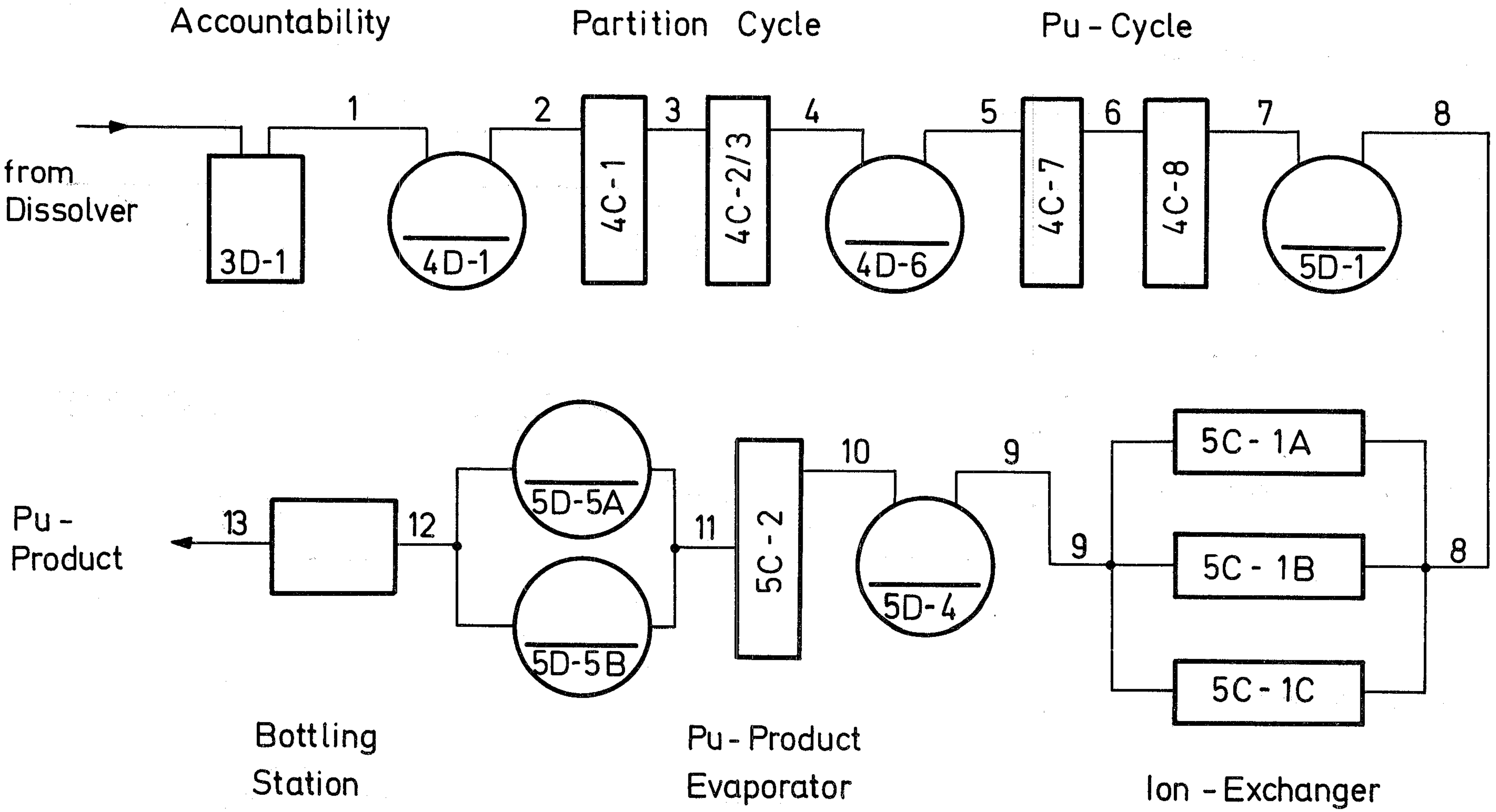
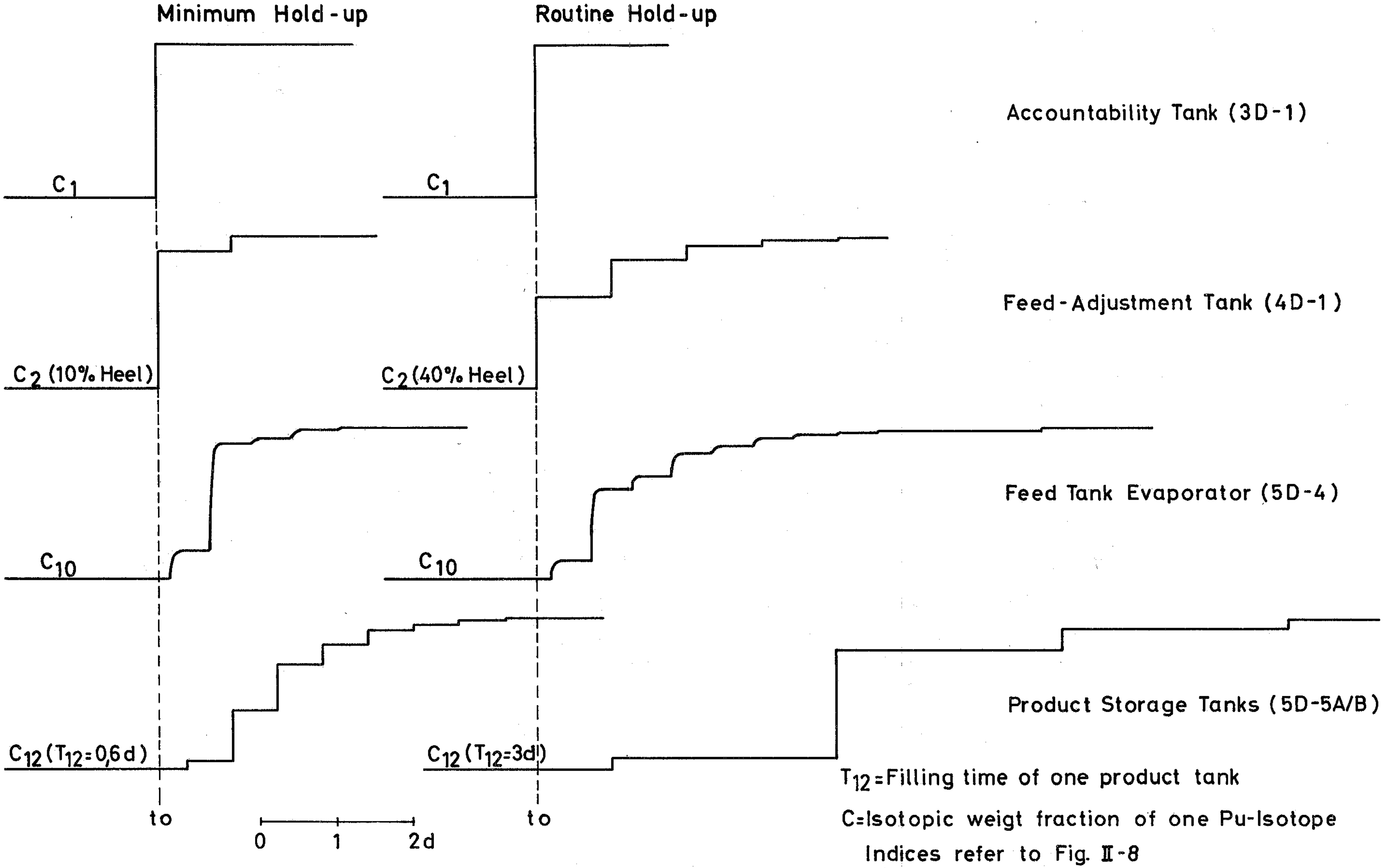


Fig. II-7 Integrating Unit

Fig. II-8 Flowsheet of Pu-Mainflow through the NFS-Plant

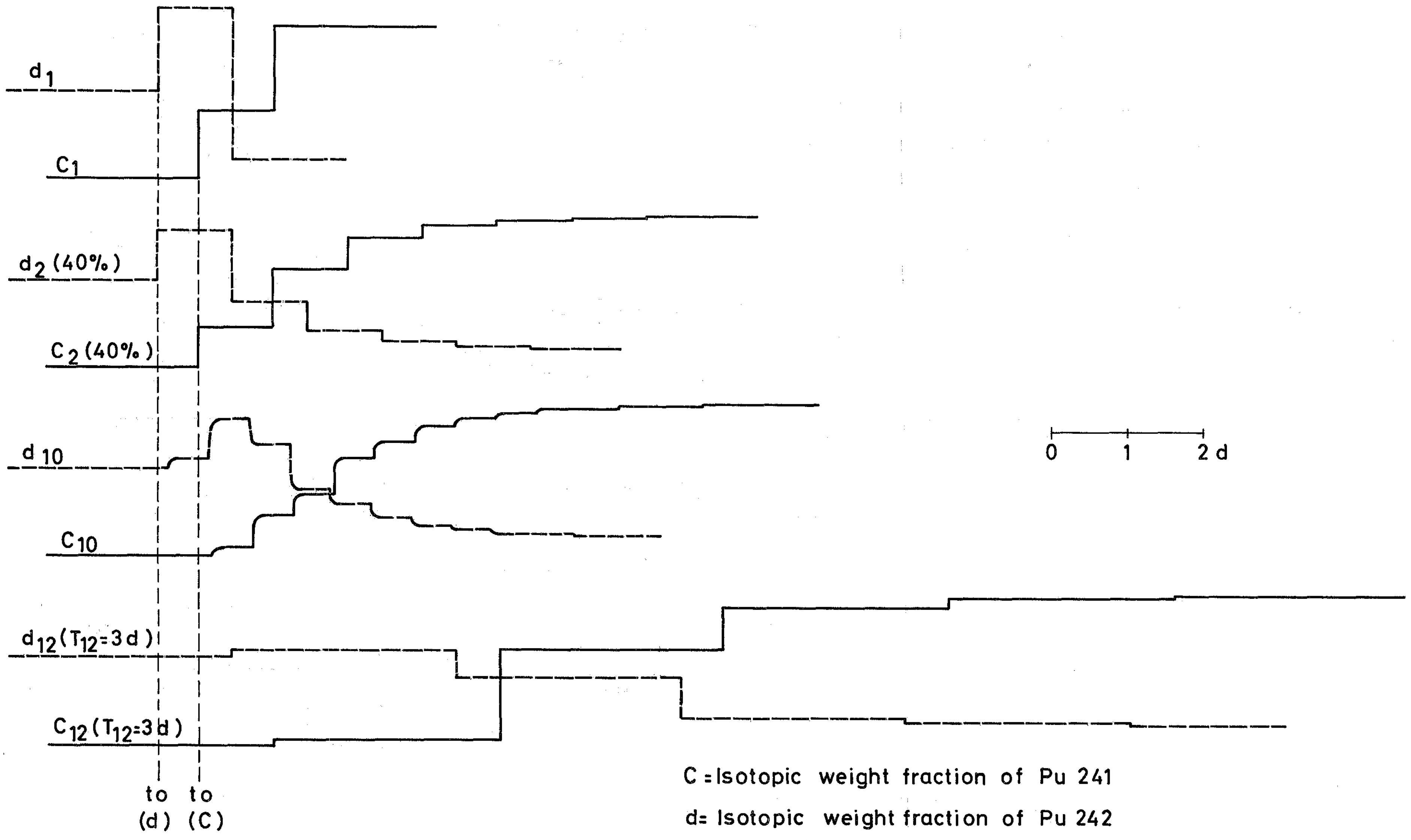




T_{12} = Filling time of one product tank

C = Isotopic weigt fraction of one Pu-Isotope
Indices refer to Fig. II-8

Fig. II-9 Deformation of a Step Input Function (C_1) by Internal Mixing Mechanisms in the NFS-Plant for Minimum Hold-up and Routine Hold-up.

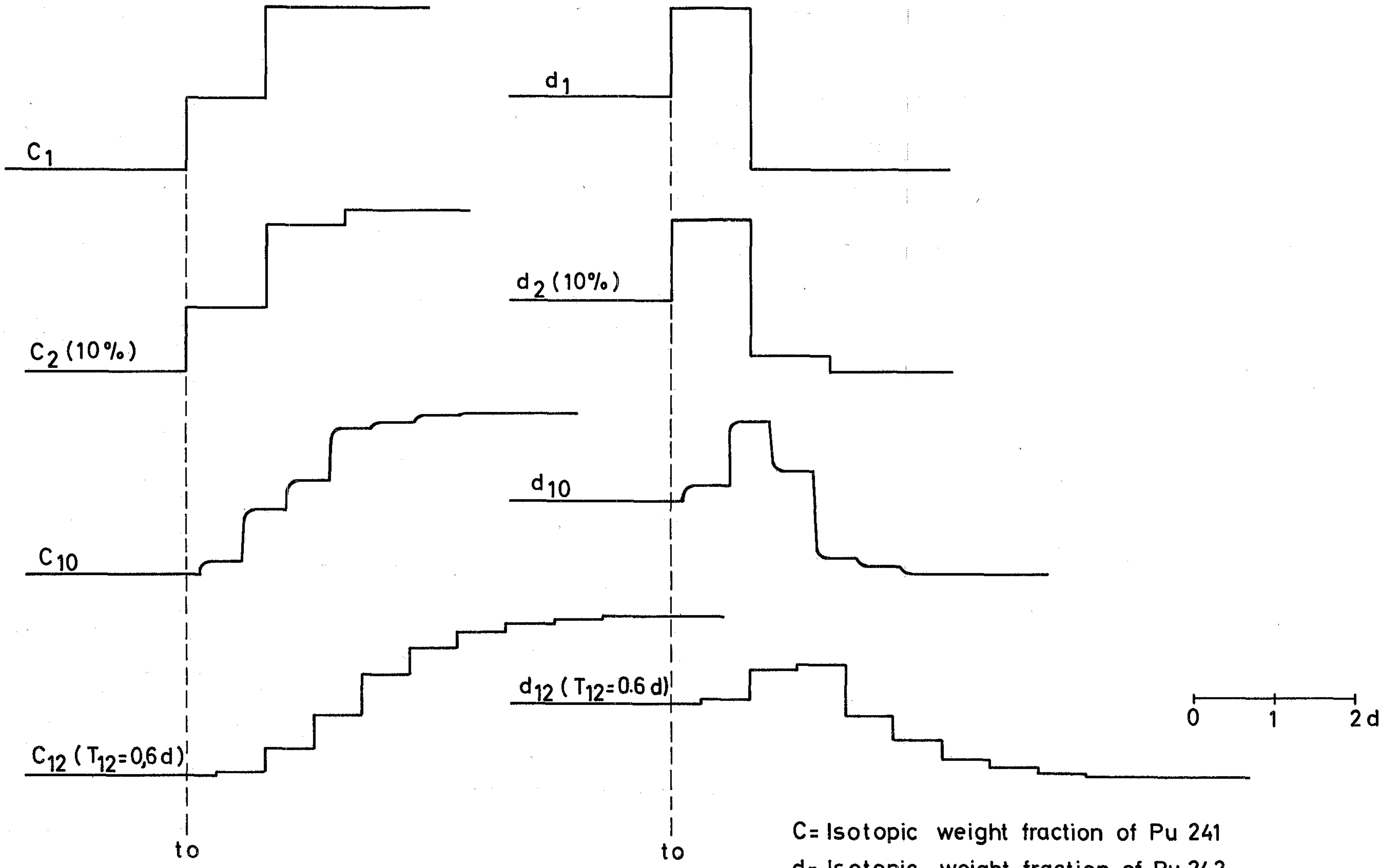


C = Isotopic weight fraction of Pu 241

d = Isotopic weight fraction of Pu 242

Indices refer to Fig. II-8

Fig. II-10 Deformation of a Double-Step-Function ($C_1 d_1$) by Internal Mixing Mechanisms in the NFS-Plant for Routine Hold-up.

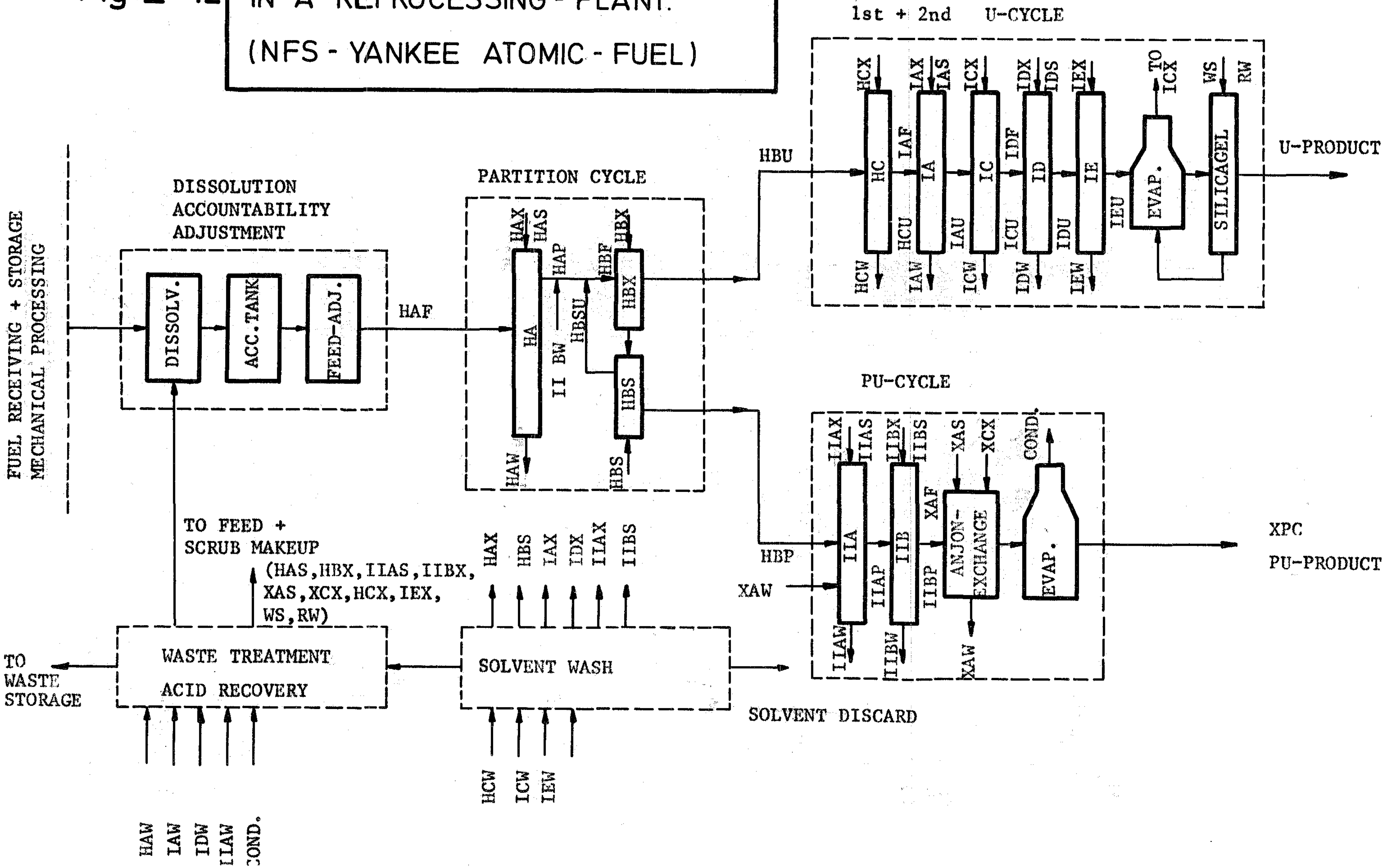


C= Isotopic weight fraction of Pu 241
d= Isotopic weight fraction of Pu 242
Indices refer to Fig. II-8

Fig. II-11 Deformation of a Double-Step-Function (C_1 d_1) by Internal Mixing Mechanisms in the NFS-Plant for Minimum Hold-up

Fig. II -12

NUCLEAR MATERIAL FLOWSHEET
IN A REPROCESSING - PLANT.
(NFS - YANKEE ATOMIC - FUEL)



Chapter III

Simulation of a Reprocessing Plant in an Analog Computer

F. Katz

The reprocessing plant has been simulated in the hybrid analog computer HYDAC 2000 (EAJ). The system HYDAC 2000 consists of a simple analog computer combined with a digital operating system (DOS). The DOS makes it possible to store functions and to analyse discontinuous operating systems.

1. The Block Diagram of the Reprocessing Plant

In Fig. III-1 the block diagram of the reprocessing plant is shown. The corresponding mathematical equations have been discussed in chapter II.

Fig. III-2 shows the meaning of the different components of this block diagram.

For the simulation of the discontinuously operating parts of a reprocessing plant, the following components are required (Fig. III-2):

1. The digitally controlled analog switch (DAS)
2. The digitally controlled integrator and
3. The track and store unit (TSU)

The digitally controlled analog switch has an analog input, an analog output, and a control input for binary signals. In case that a logical-1-condition exists at the control input, the analog input is connected to the output and a tension can be switched through. However, if there is a logical-0-condition at the control input, the analog input is connected to ground via a resistance.

For the digitally controlled integrator the control inputs of the relay circuits which are responsible for the mode control (initial condition, computing, stop) are arranged on the programming panel. From this panel they can be connected to the output of digitally controlled relay driver stages, so that each integrator can be individually controlled. This means that an integrator controlled by a relay driver, can change from the "initial condition" to the "computing" condition and vice versa, whilst the mode for the whole console is "computing".

The track- and store unit is an analog point storage, functioning on the principle of an integrator under the "initial condition" and "hold condition". If a logical-1-condition exists at the control input, the output voltage of this unit follows an analog input voltage. However, as soon as a logical-0-condition occurs at the control input, the input voltage is disconnected, whilst the output voltage keeps the value reached at the moment of switching.

The simulation of the filling periods of tanks and of the heels remaining in the tanks after discharge is explained in the Fig. III-3, III-4 and III-5.

The periods are simulated by periodically changing binary signals which control the analog computing elements.

The computing pulses are produced by an impulse generator which is composed of analog and digital computing elements (Fig. III-3). At the input of the integrator a positive voltage of $+P \cdot 50V$ initially prevails. The voltage ranges from + 50 V to -50 V at the output of the integrator, where it switches the comparator, since its biased input of +50 V takes a negative value. Via a digital analog switch (DAS) the input to the comparator is subsequently biased to -50V; simultaneously, the integrator receives $-P \cdot 50V$. Thus, a saw-tooth voltage is produced at the output of the integrator, whilst a binary pulse sequence exists at the exit of the comparator. The length of the period is set by the potentiometer P.

$$T = \frac{2}{P}$$

The heels, remaining in a tank after its discharge, have to be stored in the analog computer for one or two time periods. This storage is effected with the help of the TSU described above. Fig. III-4 shows the storage of the concentration from period n-1 to period n. In channel 1 a logical 1 exists in period n-1 at the control input of TSU 1, the output of which follows the value of the input voltage which corresponds to the concentration c^{n-1}. In the next period

TSU 1 is switched in "store". In this period DAS 1 has a logical 1 at the control input, i.e., its output voltage is switched through, so that the stored signal appears now at the exit of channel 1. Since channel 1 can only store each second period, channel 2 which operates in push-pull to channel 1, takes over the storage for the intermediate periods. In the case of storing the signal over two periods in each channel 2 TSU are connected in series (Fig.III-5).

A continuous input is collected in the ion-exchange feed tank. After a filling period T, the resulting concentration is given by

$$c = \frac{1}{T} \int_{nT}^{(n+1)T} c_i \, dt,$$

where T is the filling period

c the resulting concentration

$c_i(t)$ the concentration in the input stream.

Two tanks are represented which are alternatively fed from the preceding column (Fig. III-6). This means that while in channel 1 the integrator J1 corresponding to the first tank is integrating, the integrator J2 reaches the "initial condition", it is so to speak prepared for the next filling. The mean value calculated in the integrator J1 is stored during the following period in the TS unit TSU1. At the same time, the switch DAS1 is connected through, so that via channel 1 the mean value c_{i+1} is available. Since channel 2 operates in push-pull, the integrator J2 now computes the mean value of this period.

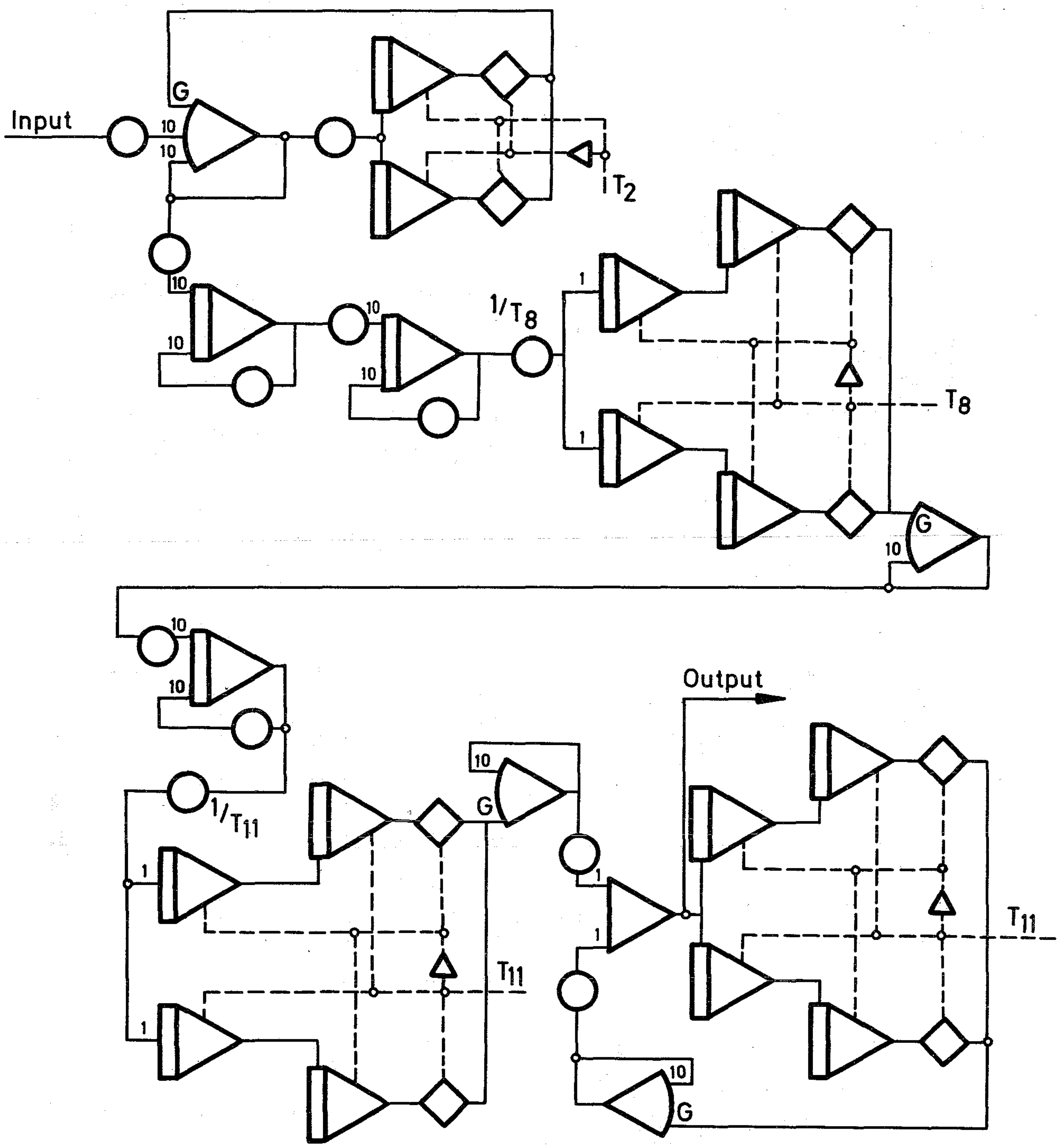


Fig. III-1 Block Diagram of the Reprocessing Plant

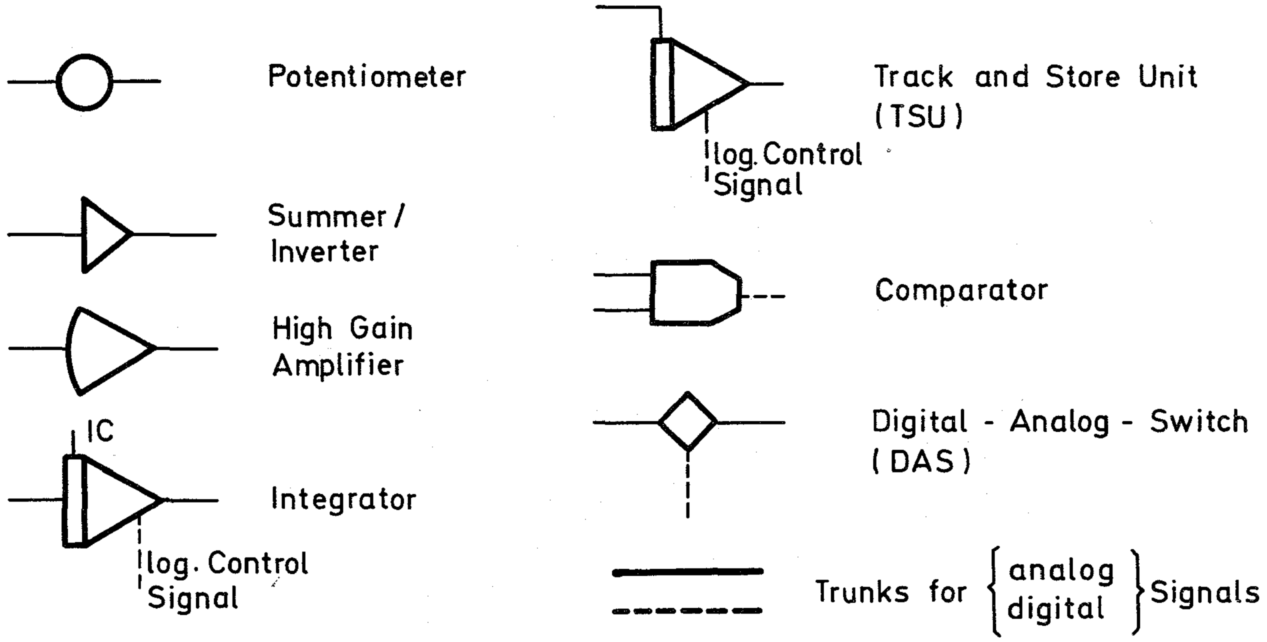


Fig. III-2 Components of the Analog Computer Used for the Simulation

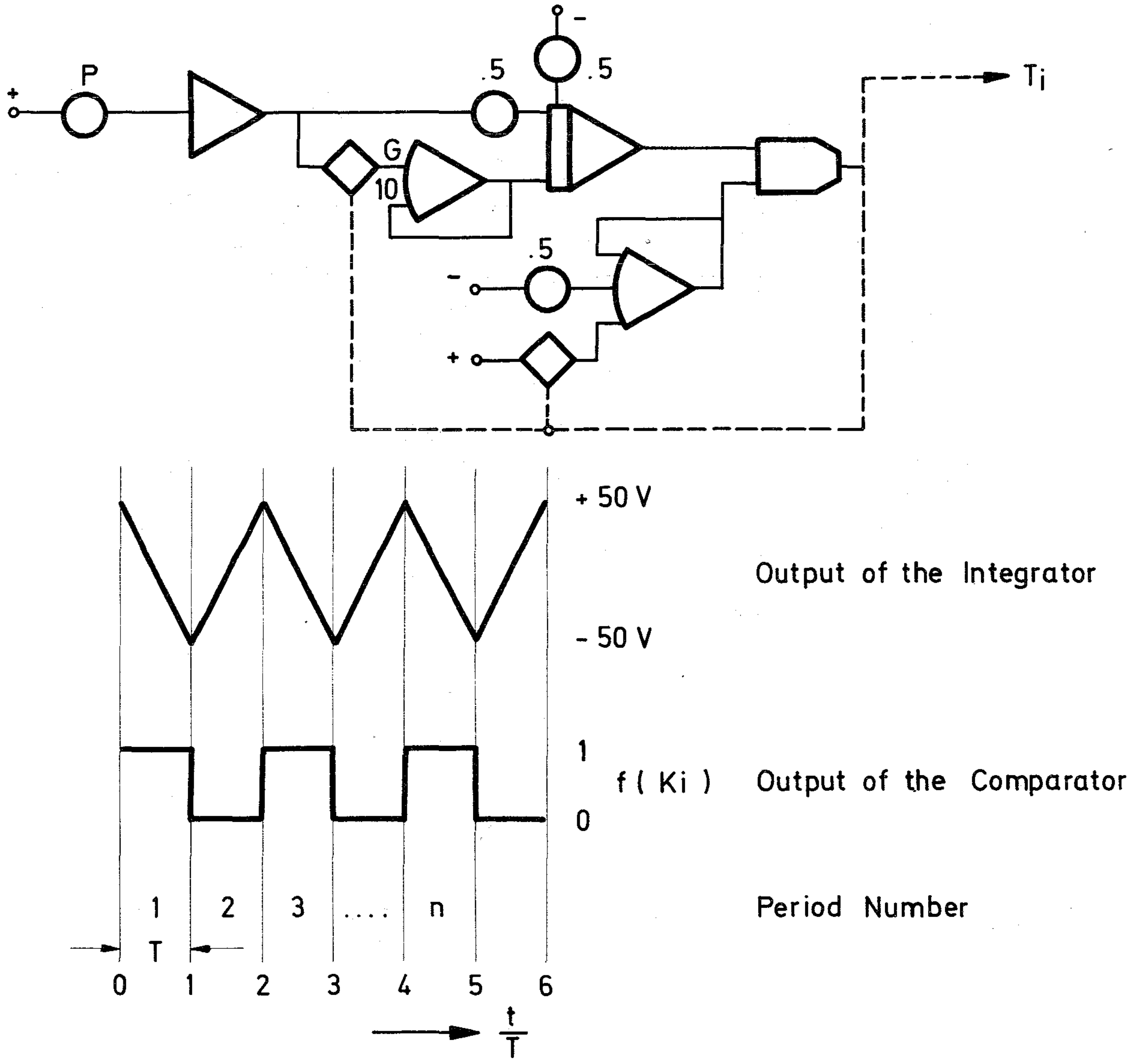


Fig. III-3 Impuls Generator

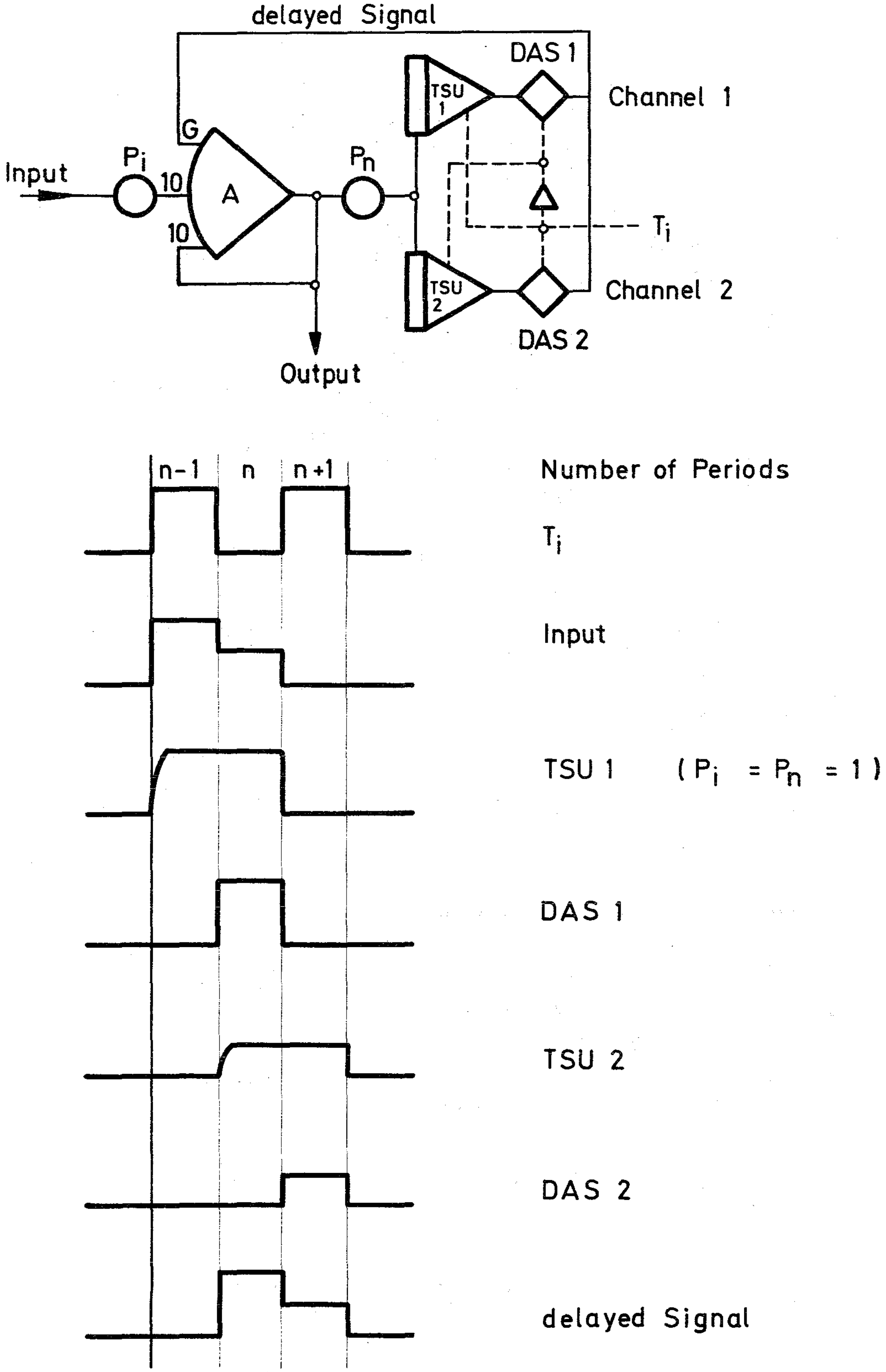


Fig. III-4 Simulation of the Heels Stored for one Time Period

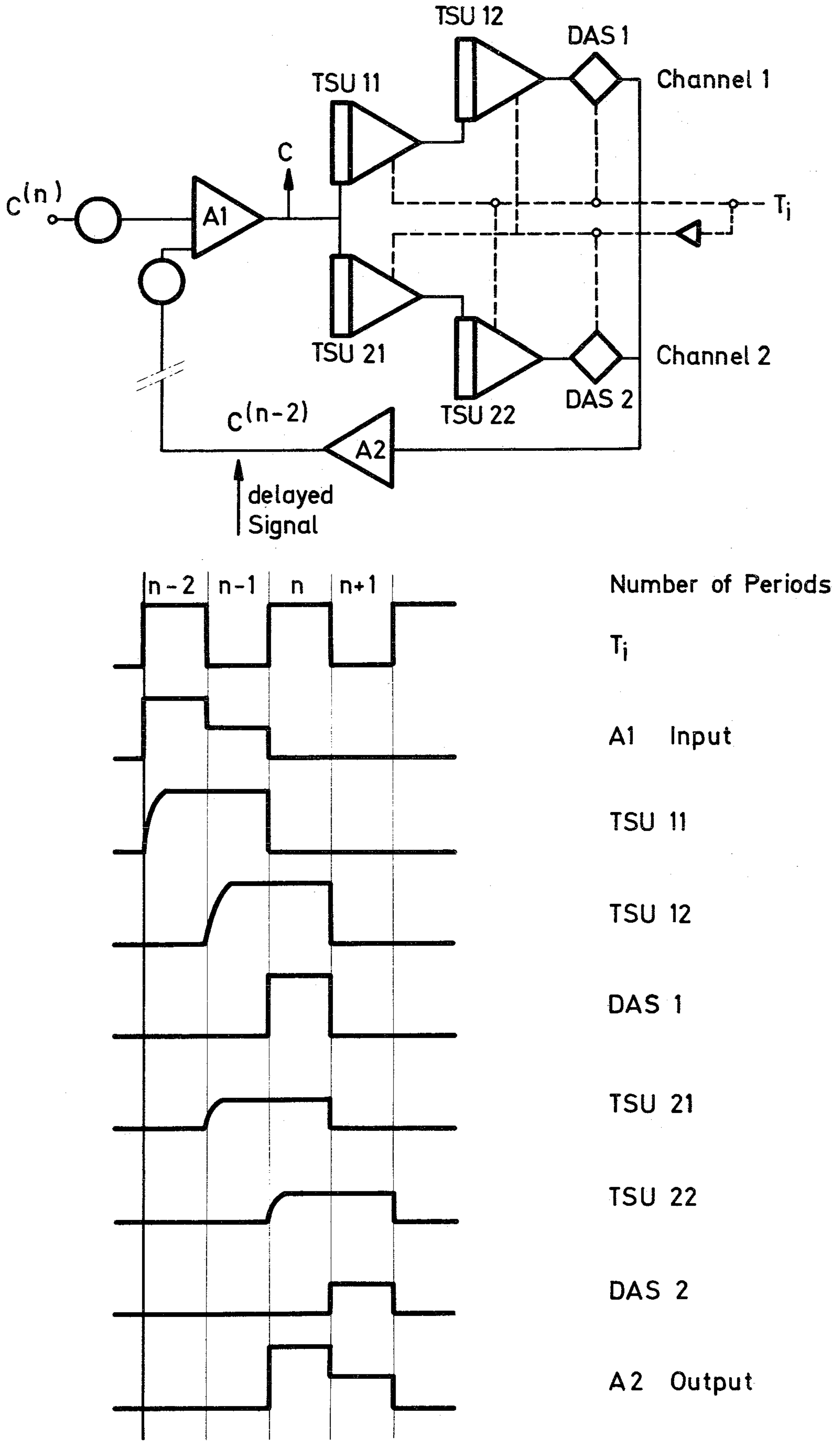


Fig. III-5 Simulation of the Heels Stored for two Time Periods

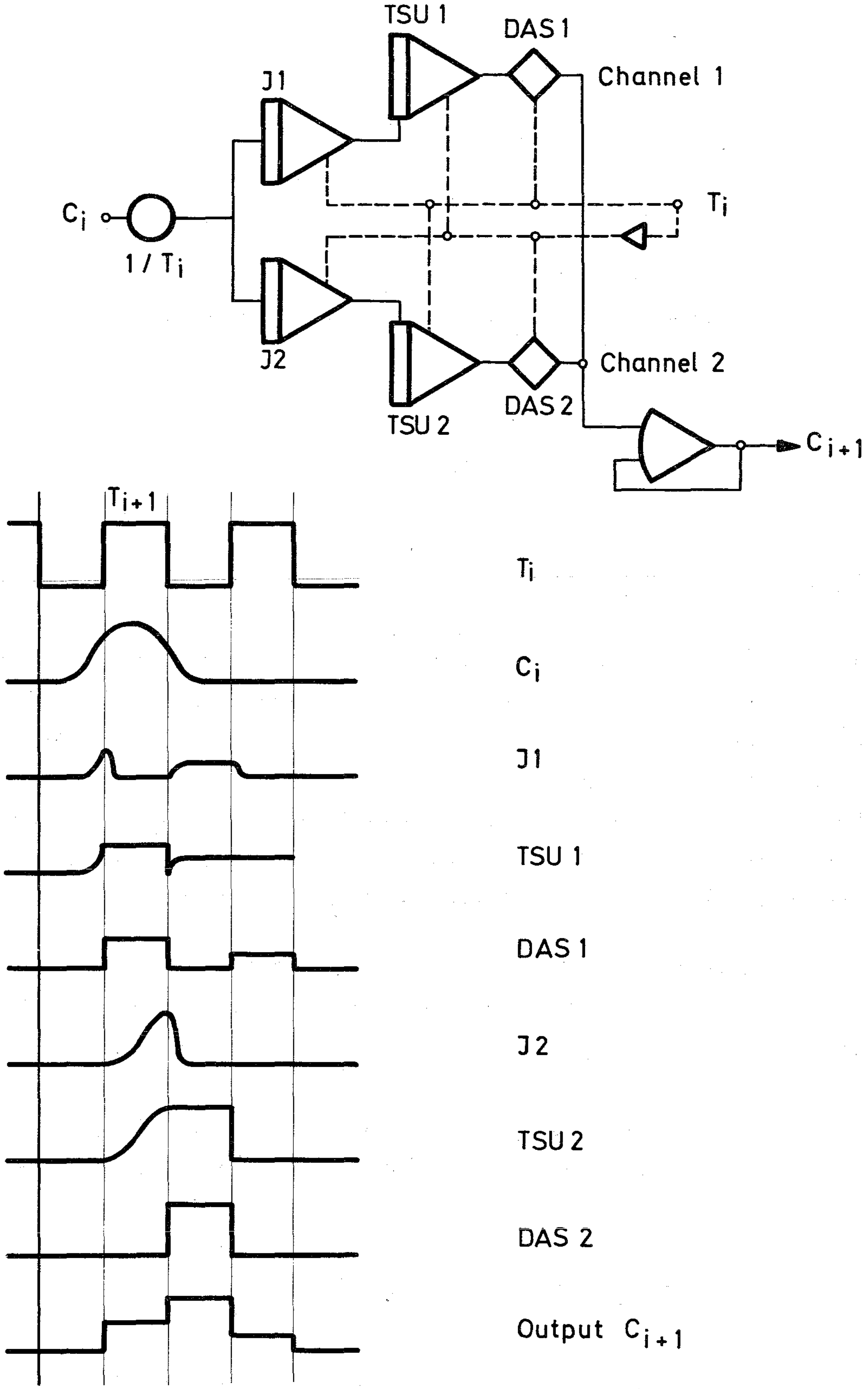


Fig. III-6 Simulation of the Continous Filling of a Tank and the Batchwise Discharge

Chapter IV

Statistical Statements about a Diversion in the Case of Inventory Determination by Tracer Methods

R. Avenhaus

1. Formulation of the Problem

In this chapter the propagation of measurement errors have been treated on a statistical basis. For this purpose the particular case of two consecutive input batches mixing into one output batch has been considered (chapter I, 4.1).
A similar treatment is possible in the general case, but these calculations have not been carried out here. Besides the propagation of error, some consideration has been given to the statistical statements, which can be made on the basis of data collected by the method of inventory determination.

The flow measurement gives at the time t the book inventory J. J is a stochastic variable because of random measurement errors. It is assumed, that J is normal distributed, with the expectation value EJ and variance σ_J^2.

The inventory measurement with the help of isotope analysis gives the inventory at the time t. In the case of two isotopes one has according to equation (I-5)

$$H = \sum_i h_i = \sum_i M_i b_i ; \; b_i = \frac{x_1 - c_2}{c_1 - c_2} \qquad \text{(IV-1)}$$

According to the central limit theorem the stochastic variable

$$\frac{\sum_i (M_i b_i - EM_i Eb_i)}{\sigma_H} \qquad \text{(IV-2)}$$

is approximately normal distributed, with the expectation value 0 and the variance 1. This means, H is approximately normal distributed, with the expectation value EH and the variance σ_H^2:

$$H \sim n\ (EH,\ \sigma_H^2) \qquad \text{(IV-3)}$$

Because of these assumptions, the difference J-H is normal distributed with the expectation value EJ-EH and the variance σ_{tot}^2:

$$J-H \sim n\ (EJ-EH;\ \sigma_{tot}^2);\ \sigma_{tot}^2 = \sigma_J^2 + \sigma_H^2 \qquad \text{(IV-4)}$$

With the help of eq. (IV-4) the operator of the plant and the inspector can give their statistical statements.

2. Statistical Statements of the Operator and the Inspector

2.1 Inspector's Statement

It is assumed that before obtaining the measurements, the inspector can make the statement that, with a probability of 1-α, the difference J-H will fall in the interval $(-\infty,\ c\cdot\sigma_{tot})$

$$p\ (-\infty \leq \frac{J-H-(EJ-EH)}{\sigma_{tot}} \leq c\cdot\sigma_{tot}) \qquad \text{(IV-5)}$$

$$= \phi(c) = 1-\alpha$$

Here, $\phi(x)$ is given by

$$\phi(x) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{x} dt\ \exp\ (-\frac{t^2}{2})$$

The inspector can choose the value of α(for example 5 %) on the basis of his experience, economics, or some other considerations. After fixing this value of α(which indicates the probability with which the safeguards system may err) and making the measurements which give him the values $\hat{J} - \hat{H}$ in such a way that

$$\hat{J} - \hat{H} > c\ \sigma_{tot}$$

then he can make the statement that something has been diverted. If required he can also make some more specific statement (for example he can say that the amount $\hat{J}-\hat{H}-c\,\sigma_{tot}$ has been diverted) depending on the statistical model he proposes to use. In case he realizes the values of $\hat{J}-\hat{H}$ which lie in such a manner that

$$\hat{J}-\hat{H} \leq c\,\sigma_{tot}$$

then he makes the statement that nothing has been diverted. Here c and α are connected by equation (IV-5).

2.2 Operator's Statement

The operator who intends to divert the amount of fissile material

$$m_o = EJ - EH \qquad \text{(IV-6)}$$

can calculate the probability $p(E/m_o)$ with which the inspector will make the statement that something has been diverted. One has

$$p\,(E/m_o) = p\,(J-H-c\cdot\sigma_{tot} > 0/m_o) \qquad \text{(IV-7)}$$

From eq. (IV-4)

$$p\,(E/m_o) = \frac{1}{\sqrt{2\pi}\sigma_{tot}} \int_{-\infty}^{c\cdot\sigma_{tot}} dt\, \exp\left(-\frac{(t-m_o)^2}{2\sigma_{tot}^2}\right) = \phi\left(\frac{m_o - c\cdot\sigma_{tot}}{\sigma_{tot}}\right)$$

$$\text{(IV-8)}$$

Here, $\phi(x)$ is given by

$$\phi(x) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{x} dt\, \exp\left(-\frac{t^2}{2}\right) \qquad \text{(IV-9)}$$

3. Calculation of the Variance of the Inventory Measurement

In order to be able to give statements in the form described above, the inspector has to know the variances of the flow measurement and the inventory measurement. Therefore, one has to calculate the variance of H according to eq.(IV-1) with the help of the known distributions of M_i, x_i, c_1, c_2.

h_i and h_j are not independent. Therefore in general one has

$$\text{var } H = \text{var} \sum_i h_i = \sum_i \text{var } h_i + 2 \sum_{i<j} \text{cov } h_i h_j \qquad \text{(IV-10)}$$

Furthermore

$$\text{cov } h_i h_j = E\,(h_i h_j) - Eh_i Eh_j \qquad \text{(IV-11)}$$

Therefore, because M_i and b_i are independent

$$\text{cov } h_i h_j = E(M_i M_j b_i b_j) - EM_i b_i \; EM_j b_j =$$

$$= EM_i \; EM_j (E(b_i b_j) - Eb_i Eb_j) =$$

$$= EM_i \; EM_j \text{ cov } b_i b_j \qquad \text{(IV-12)}$$

Furthermore,

$$\text{var } h_i = \text{var } M_i b_i = E^2 M_i \text{ var } b_i + E^2 b_i \text{ var } M_i + \text{var } M_i \text{ var } b_i$$

(IV-13)

With the assumption

$$E^2 M_i >> \text{var } M_i \qquad \text{(IV-14)}$$

one obtains from eq. (IV-13)

$$\text{var } h_i = E^2 M_i \text{ var } b_i + E^2 b_i \text{ var } M_i \qquad \text{(IV-15)}$$

Therefore one obtains from eq. (IV-10) with the help of eq. (IV-12, IV-15)

$$\text{var } H = \sum_i (E^2 M_i \text{ var } b_i + E^2 b_i \text{ var } M_i) + 2 \sum_{i<j} EM_i \; EM_m \text{ cov } b_i b_j$$

(IV-16)

In the following one has to use the special form of the distributions of x_i, c_1, c_2. One assumes that these variables are equally distributed, with the lengths of allowed intervals $2\alpha_i$, $2\alpha_1$, $2\alpha_c$:

$$f_{x_i}(x_1) = \begin{cases} \dfrac{1}{2\alpha_i} & \text{for } Ex_i - \alpha_i \leq x_1 \leq Ex_i + \alpha_i \\ 0 & \text{otherwise} \end{cases} \qquad \text{(IV-17)}$$

$$f_{c_{1,2}}(x_{2,3}) = \begin{cases} \dfrac{1}{2\alpha_{1,2}} & \text{for } Ec_{1,2} - \alpha_{1,2} \leq x_{2,3} \leq Ec_{1,2} + \alpha_{1,2} \\ 0 & \text{otherwise} \end{cases}$$

Note: In general eq. (IV-17) is not fulfilled. One assumes that the true distribution is approximated by an equal distribution with the true expectation value and the true variance.

(a) <u>Calculation of var b_i:</u>

According to

$$\text{var } b_i = Eb_i^2 - E^2 b_i \qquad \text{(IV-18)}$$

one has to calculate Eb_i and Eb_i^2 .

From eq. (IV-1) one has

$$Eb_i = \frac{1}{8\alpha_i\alpha_1\alpha_2} \int_{Ex_i-\alpha_i}^{Ex_i+\alpha_i} dx_1 \int_{Ec_1-\alpha_1}^{Ec_1+\alpha_1} dx_2 \int_{Ec_2-\alpha_2}^{Ec_2+\alpha_2} dx_3 \, \frac{x_1-x_3}{x_2-x_3} \qquad \text{(IV-19)}$$

With the assumption

$$\sigma^2_{c_{1,2}} \ll (Ec_1-Ec_2)^2$$

(which must be fulfilled in order that the signal can be measured) the following relation holds:

$$\frac{\alpha_{1,2}}{Ec_1-Ec_2-\alpha_{1,2}} \ll 1 \qquad \text{(IV-20)}$$

Thus from (IV-19), if one assumes $Ec_1 > Ec_2$

$$Eb_i = \frac{Ex_i-Ec_2}{Ec_1-Ec_2} + \frac{\text{var } c_1}{(Ec_1-Ec_2)^3}(Ex_i-Ec_2) + \frac{\text{var } c_2}{(Ec_1-Ec_2)^3}(Ex_i-Ec_1) \qquad \text{(IV-21)}$$

Here, terms of higher order in var c are neglected.

Furthermore

$$Eb_i^2 = \frac{1}{8\alpha_i\alpha_1\alpha_2}\int_{Ex_i-\alpha_i}^{Ex_i+\alpha_i} dx_1 \int_{Ec_1-\alpha_1}^{Ec_1+\alpha_1} dx_2 \int_{Ec_2-\alpha_2}^{Ec_2+\alpha_2} dx_3 \left(\frac{x_1-x_3}{x_2-x_3}\right)^2 \qquad \text{(IV-22)}$$

The calculation gives

$$Eb_i^2 = \left(\frac{Ex_i-Ec_2}{Ec_1-Ec_2}\right)^2 + \frac{3 \text{ var } c_1}{(Ec_1-Ec_2)^2}\left(\frac{Ex_i-Ec_2}{Ec_1-Ec_2}\right)^2$$

$$+ \frac{\text{var } c_2}{(Ec_1-Ec_2)^2}\cdot\left(3\left(\frac{Ec_1-Ex_i}{Ec_1-Ec_2}\right)^2 - \frac{2(Ec_1-Ex_i)}{Ec_1-Ec_2}\right) + \frac{\text{var } x_i}{(Ec_2-Ex_1)^2} \qquad \text{(IV-23)}$$

Therefore one has from (IV-18,IV-21, VI-23)

$$\text{var } b_i = \frac{1}{(Ec_1-Ec_2)^4}\left(\text{var } c_1\cdot(Ex_i-Ec_2)^2 + \text{var } c_2\cdot(Ec_1-Ex_i)^2 \right.$$

$$\left. + \text{ var } x_i\ (Ec_1-Ec_2)^2\right) \qquad \text{(IV-24)}$$

If one introduces the relative variances

$$\delta c_1 = E^2c_1\cdot\text{var } c_1 \ ; \ \delta c_2 = E^2c_2\cdot\text{var } c_2; \ \delta x_i = E^2x_i \cdot \text{var } x_i \qquad \text{(IV-25)}$$

and assumes, that these are equal

$$\delta c_1^2 = \delta c_2^2 = \delta x_i^2 = \delta^2 \qquad \text{(IV-26)}$$

one obtains from (IV-24)

$$\text{var } b_i = \frac{\delta^2}{(Ec_1-Ec_2)^4} \left(E^2 c_1 \, (Ex_i-Ec_2)^2 + E^2 c_2 (Ec_1-Ex_i)^2 + E^2 x_i (Ec_1-Ec_2)^2 \right) \qquad \text{(IV-27)}$$

(with some simplifying assumption this form can be reduced to an equivalant form as given by eq.(I-7))

(b) Calculation of cov $b_i b_j$

According to eq. (IV-16) one has to calculate $Eb_i b_j$. Now

$$Eb_i b_j = \frac{1}{8\alpha_i \alpha_1 \alpha_2} \int_{Ex_i-\alpha_i}^{Ex_i+\alpha_i} dx_1 \int_{Ex_j-\alpha_j}^{Ex_j+\alpha_j} dx_2 \int_{Ec_2-\alpha_2}^{Ec_2+\alpha_2} dx_3 \int_{Ec_1-\alpha_1}^{Ec_1+\alpha_1} dx_4 \; \frac{(x_1-x_3)(x_2-x_3)}{(x_3-x_4)^2} \qquad \text{(IV-28)}$$

Again with the assumption (IV-20) one obtains

$$Eb_i b_j = \frac{(Ex_i-Ec_2)(Ex_j-Ec_2)}{(Ec_1-Ec_2)^2} + \frac{3 \text{ var } c_1}{(Ec_1-Ec_2)^4} (Ex_j- Ec_2) \; (Ex_i-Ec_2)$$

$$+ \frac{\text{var } c_2}{(Ec_1-Ec_2)^4} \left((2\, Ec_1-Ex_i-Ex_j)(Ec_2-Ec_1)+3(Ec_1-Ex_i)(Ec_1-Ex_j) \right) \qquad \text{(IV-29)}$$

Therefore, from eq. IV-11, IV-21, IV-29) one obtains

$$\text{cov } b_i b_j = \frac{1}{(Ec_1-Ec_2)^4} \left[\text{var } c_1 (Ex_i-Ec_2)(Ex_j-Ec_2) + \text{var } c_2 (Ec_1-Ex_i) \cdot (Ec_1-Ex_j) \right] \qquad \text{(IV-30)}$$

or with eq. (IV-26)

$$\text{cov } b_i b_j = \frac{\delta^2}{(Ec_1-Ec_2)^4} \left[E^2 c_1 (Ex_i-Ec_2)(Ex_j-Ec_2)+E^2 c_2 (Ec_1-Ex_i)(Ec_1-Ex_j) \right] \qquad \text{(IV-31)}$$

As can be seen easily, the formulae (IV-27) and (IV-31) satisfy the general condition

$$\mathrm{cov}\ b_i b_j \leq +\sqrt{\mathrm{var}\ b_i} \cdot +\sqrt{\mathrm{var}\ b_j} \qquad \text{(IV-32)}$$

Now one obtains the variance of H as a function of the expectation values and the variances of M_i, x_i, c_1, c_2 by inserting the expressions (IV-19, IV-27, IV-31) into the formula (IV-16).

The following difficulty arises if the inspector wants to fix his critical value of $\hat{J}-\hat{H}$, which was given by $c \cdot \sigma_{tot}$. In the formula for the variance of H there occur the expectation values of M_i etc. which are not known. One can solve this problem approximately by replacing the expectation values by the measured values. This will be a good approximation for the following reasons: Firstly, the measured values are unbiased estimates of the expectation values. Secondly, the variance of H is a very complicated function of the expectation values, therefore one can assume that the errors which are introduced by this estimate, will approximately compensate.

4. Summary

In this chapter it has been indicated how the operator and the inspector can calculate their statistical statements with the help of their measurements

(a) The inspector has obtained the results of the measurements $\hat{J}$, $\hat{H}$. He states, if $\hat{J}-\hat{H} > c \cdot \sigma_{tot}$, fissile material has been diverted. His error α is connected to c by the relation

$$1-\alpha = \phi(c) \qquad \text{(IV-33)}$$

σ_{tot} is given by

$$\sigma_{tot}^2 = \sigma_J^2 + \sigma_H^2 \qquad \text{(IV-34)}$$

where

$$\sigma_H^2 = \sum_i \left(E^2 M_i \text{ var } b_i + E^2 b_i \text{ var } M_i\right) + 2 \sum_{i<j} EM_i \, EM_j \text{ cov } b_i b_j \qquad \text{(IV-35)}$$

$$Eb_i = \frac{Ex_i - Ec_2}{Ec_1 - Ec_2} + \frac{\delta^2}{(Ec_1 - Ec_2)^3} \left[E^2 c_1 \, (Ex_i - Ec_2) + E^2 c_2 \, (Ex_i - Ec_1) \right]$$

$$\text{var } b_i = \frac{\delta^2}{(Ec_1 - Ec_2)^4} \left(E^2 c_1 \, (Ex_i - Ec_2)^2 + E^2 c_2 \, (Ec_1 - Ex_i)^2 + E^2 x_i \, (Ec_1 - Ec_2)^2 \right)$$

$$\text{cov } b_i b_j = \frac{\delta^2}{(Ec_1 - Ec_2)^4} \left(E^2 c_1 (Ex_i - Ec_2)(Ex_j - Ec_2) + E^2 c_2 (Ec_1 - Ex_i)(Ec_1 - Ex_j) \right)$$

(b) The operator who intends to divert the amount m_o of fissile material can calculate the probability p (E/m_o), with which the inspector will make the statement that something has been diverted.

$$p(E/m_o) = \phi\left(\frac{m_o - c \cdot \sigma_{tot}}{\sigma_{tot}} \right)$$

Here, c and σ_{tot} are given by eq. IV-33, IV-34).

Conclusion

D. Gupta, H. Winter

It was indicated at the beginning that a new method for the estimation of process inventory in a reprocessing plant, has been treated in the present paper. The method was analysed mathematically and then tested in an analog computer simulated reprocessing plant. These analysis and testing permit a number of conclusions to be drawn.

1. An independent determination of process inventory in a reprocessing plant based on isotope analysis can be carried out if the following conditions are fulfilled:

   1.1 The concentrations of some isotopes should differ significantly in two consecutive input batches. It has been shown in the App. I, that such significant differences do exists in irradiated fuel elements coming out of commercial reactors.

   1.2 The number of input batches, mixed into one output batch must not exceed n+1, where n is the number of fissile isotopes which can be used for this purpose.

   1.3 The weight fractions of the relevant isotopes must be linear independent. This requirement is fulfilled in general because of the different irradiation history of the fuel elements in different dissolver batches of a reprocessing plant.

2. The four internal mixing mechanisms in four basic units of a reprocessing plant, which normally tend to degrade the input step signal, can be simulated in a highly realistic manner in an analog computer. They can be defined in such a general manner that it is possible to simulate any aqueous reprocessing plant with the help of these four basic units.

3. Although the internal mixing mechanisms tend to degrade the input step signal inside a reprocessing plant, the influence of this mixing can be reduced by combining different input batches with identical isotopic composition into super batches or by reducing the holdup in a number of process tanks. The reduction of these holdups does not cause any change in the normal process operation of a reprocessing plant.

4. The mathematical relations developed in this paper are applicable to a reprocessing plant operating batchwise at the input and the output stage. Similar relations can be developed for plants operating continuously at these states. This means that process inventory can be determined in such plants also provided the conditions in conclusion 1 are fulfilled.

5. A number of valuable statements in the line of those given in Ref. /‾1_7 can be made on the basis of measured data obtained from this method.

6. Since the analog simulation as carried out in this paper, cannot take care of all possible degradation mechanisms occuring inside an industrial scale plant, it is absolutely essential to test this method in actual plants under industrial operating conditions+). Such experiments can be carried out either by utilizing the naturally occuring differences in the isotopic composition of the dissolver batches of a reprocessing campaign or by utilizing the differences of the isotopic composition of different campaigns, which are carried out without an intermediate washout.

Acknowledgement

The authors would like to thank Prof. Häfele for his interest and Dr. Lorin Stieff of ACDA for valuable discussions.

+) Two such experiments are in progress at the time of writing this report; one at the NFS plant in collaboration with the ACDA (Arms Control and Disarmament Agency, USA) and the USAEC, and the other at EUROCHEMIC Mol/Belgium, in collaboration with the CEN, Mol.

Appendix 1

Differences in the Isotopic Composition of Fuel from one Reactor, Processed During one Campaign.

In the following table the minimum and the maximum weight fractions of Pu isotopes are shown that have been measured in the dissolver during a reprocessing campaign. The first example is the reprocessing campaign of the Yankee reactor (core 1-4) /‾4_7. The second campaign has been safeguarded in 1968 at Eurochemic, Mol by the Karlsruhe safeguards group /‾5_7.

Isotopes	Yankee campaign		Eurochemic campaign	
	min /‾wt %_7	max	min /‾wt %_7	max
Pu 238	0.092	0.765	0.03	0.099
Pu 239	72.713	90.190	90.51	92.44
Pu 240	7.734	15.525	6.77	8.49
Pu 241	1.858	9.101	0.51	0.76
Pu 242	0.126	1.896	0.05	0.24

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