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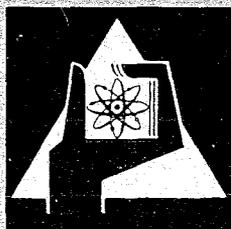
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Methods for Independent Determination of Process  
Inventory in Nuclear Facilities

- Fabrication Plant -

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METHODS FOR INDEPENDENT DETERMINATION OF  
PROCESS INVENTORY IN NUCLEAR FACILITIES  
- FABRICATION PLANT -

by

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<sup>x)</sup> delegated from EURATOM



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INTRODUCTION

A modern safeguards system for fissionable material has been described in [1, 2, 3, 4]. Its purpose is to prevent or to detect a diversion of nuclear fuel from the domain of peaceful use of nuclear energy. In this system, the measurement of the flow of fissionable material at strategic points plays a key role (second safeguard measure). This measurement allows the calculation of the nominal value of the process inventory of a nuclear plant, based on the assumption of mass conservation. The third safeguard measure, the inventory taking, gives the real value of the process inventory of the plant at a certain time. This value is compared with the result of the second safeguard measure and from this comparison result the statements of a control authority about a diversion of nuclear material.

Methods for the determination of process inventory should be

- nonintrusive
- accurate
- feasible without any considerable time lag.

In this paper different methods of determining process inventory have been investigated. Physical inventory taking and process inventory determination during wash-out-campaigns are not discussed. The first method violates the requirement of non-intrusivity and the second is not feasible without time lag,

because the time between two wash-outs of a nuclear plant (fabrication plant and reprocessing plant) can be in the order of a year.

In order to discuss these different methods to apply the third safeguard measure, we investigate the flow of fissile material through a fuel element fabrication plant. Particularly we refer to a plant for the fabrication of plutonium bearing fuel.

In the first chapter we define the basic notions needed for a description of the fissile material flow through a fabrication plant. In the second chapter different possibilities for mathematical model system representations of a fabrication plant are discussed. In the third chapter different possibilities for process inventory determination and the use of tracer methods for this purpose are described. In the fourth chapter we give an application of the ideas outlined in the preceding chapters to an existing fabrication plant for plutonium bearing fuel elements.

## 1. BASIC CONCEPTS

Even if plutonium, which is produced in reactors from U-238, is completely free of fission products, it has to be handled in glove-boxes to avoid health hazards by its  $\alpha$ -activity and toxicity. Its processing is therefore much more difficult than the processing of the weakly active uranium. The maximum Pu-content of the glove-boxes is limited by criticality reasons to few kg.

In a fabrication plant the fuel has to be converted into reactor fuel elements by changing its chemical and geometrical form and by surrounding it by a can. In the case of Pu-bearing fuel, this chemical, mechanical or heat treatment is performed in glove-box lines. We refer to such a glove-box line as a fabrication unit<sup>x)</sup>. For oxide-type fuel elements the pressing, sintering and grinding are examples for fabrication units.

A glove-box can be represented schematically by Fig. 1. It contains a machine which performs one of the different steps of production. The "machine" may be a mechanical press, a sintering furnace or simply a set of instruments for the testing of the pellets. The production rate of the "machine" determines the output of the glove-box.

It is always possible to store a certain amount of fuel in the interior of a glove-box. This was symbolized in Fig. 1 by the introduction of a "store". Of course a fabrication unit can be schematically represented by the same Fig. 1. The store of a fabrication unit is then the sum of the stores of the glove boxes which are contained in the unit, and its "machine" is the chain of the single machines of the boxes. In general the output of a fabrication unit is determined by the production rate of the last machine of the unit.

In order to have a mathematical description of the flow of fuel through a fabrication unit, we introduce three functions of time  $t$ :

- the inventory function,  $h(t)$ , which gives the mass of fuel [in kg] contained at the time  $t$  in the fabrication unit.
- the output function,  $k(t)$ , which gives the rate of mass flow [in kg/h e.g.] leaving the fabrication unit at the time  $t$ .

<sup>x)</sup> Of course, the simplest fabrication unit is a glove-box, and the most complicated one is the whole plant

- the residence time function,  $T(t)$ , which indicates, how long the fuel, entering the fabrication unit at the time  $t$ , stays in this unit.

A fabrication plant can be represented as a chain of  $N$  fabrication units, each of which being characterized by a triple of functions  $h_i(t)$ ,  $k_i(t)$  and  $T_i(t)$ ,  $i = 1, 2, \dots, N$ . The inventory,  $h^{\text{tot}}(t)$ , of the whole plant is

$$(1.1) \quad h^{\text{tot}} = \sum_{i=1}^N h_i(t).$$

## 2. MODEL SYSTEM REPRESENTATION OF A FABRICATION PLANT

In fabrication plants considerable amounts of fuel (U and Pu) accumulate. Moreover, the fuel is directly accessible, which is a very important fact for the safeguard problem. As the time variation of the accumulated amount of fuel in the plant is described by the inventory function  $h^{\text{tot}}(t)$ , introduced in the preceding chapter, it will be highly interesting to study this function and its relation to the output and residence time functions, starting from a detailed knowledge of the plant, the fabrication process etc. This will give us a good insight to what are the important facts which determine the amount of fuel accumulating in different fabrication units.

There are two different mathematical approaches to construct a model system:

- Deterministic description. Here the basic functions  $h$ ,  $k$  and  $T$  are deterministic functions of time  $t$ .
- Stochastic description. The basic functions are random functions of time.

In both cases there exists a continuous and a discontinuous description of the material flow. In a continuous model, the basic functions are continuous functions of time with the properties of differentiability

and integrability needed for our purposes. In a discontinuous model, the basic functions are defined for discrete values  $t_n$ ,  $n = 1, 2, \dots, M$  of the independent time variable  $t$ . In the interval  $\overline{t_n, t_{n+1}}$  the variations of the basic functions have no influence upon the behaviour of the system.

Nowadays fabrication plants for Pu-fuel elements work batchwise. Thus a discontinuous description seems to be the right way. But such a description is often mathematically difficult to handle, so that a continuous approximation may be justified. Furthermore, if we believe in the predictions, the market of nuclear energy is considerably expanding in the next future, so that it may be justified to assume, that the fabrication plants will introduce continuous instead of batch processes with expanding capacity. Then the continuous one is the correct description.

### 2.1.1 A deterministic and continuous model

The inventory functions  $h_i(t)$ , the output functions  $k_i(t)$  and the residence time functions  $T_i(t)$  are continuously differentiable and integrable functions of the time  $t$ . (We can even admit, that these conditions are not fulfilled in a finite number of points  $t_n$ ). The fabrication plant consists of  $N$  fabrication units. The aim is, to calculate the  $3N$  unknown functions  $h_i(t)$ ,  $k_i(t)$  and  $T_i(t)$ ,  $i = 1, 2, \dots, N$  from a known input  $X(t)$  into the system. Fig. 2 shows an example for such a plant. There is a feed-back in the system. The fraction  $\kappa$  of the outcoming fuel of the second fabrication unit is fed back (after a reprocessing step in the third unit) to the entrance of the second unit. In general such feed-backs arise, because a certain fraction of the produced pellets (or fuel elements) do not correspond to the specifications prescribed by the operator of the reactor for which the fuel elements are destined. Unfortunately, the fraction  $\kappa$  of the fuel, which is fed back, is not always the same.  $\kappa$  can be considered as a random variable. This aspect of the model system representation is dealt with in detail in the next part of this chapter. Here we will assume, that  $\kappa$  is constant - corresponding to the mean value of the stochastic variable. This assumption does not affect the inventory function  $h^{\text{tot}}$  of the whole plant as we will see later on.

It only results in a small shift (because of the smallness of  $\kappa$ ) of the inventory of the second, the third and the fourth fabrication unit.

The first step of our model system representation is the calculation of the 2 N functions  $h_i$  and  $k_i$ . In a second step we will show, that there exists a functional relationship between the residence time function  $T_i$  and the inventory and output functions  $h_i$  and  $k_i$  of each fabrication unit.

For each fabrication unit, there exist two characteristic values of its inventory function  $h_i$ :

- a minimum value of inventory,  $h_i^{\min}$ , which is the amount of fuel, necessary for the production step performed by the machine. This can be a very small quantity. For the press, this is the amount of fuel to form one pellet, for a sintering furnace this may be several hundred of pellets etc.
- a maximum value of inventory,  $h_i^{\max}$ , which is the maximal amount of fuel that can be contained in the fabrication unit. This amount is limited to some kg in the case of Pu-fuel by criticality reasons for a glove-box.

In general, a fabrication unit can work with different values of the output function. There is only an upper limit,  $k^{\max}$ , for this function which is attained when the machines work at maximum power. We assume, that the operator of the plant has an economical interest to use at maximum the capacity of his machines. Thus, we can describe the machines by means of machine characteristics. Fig. 3 shows examples for such characteristics. The first one describes a machine, which does not work if the inventory  $h$  of the corresponding fabrication unit is smaller than the minimum value  $h^{\min}$ . When  $h > h^{\min}$  then it works at maximum power,  $k^{\max}$ . By criticality reasons  $h < h^{\max}$  must hold. The mechanical press, the sintering furnace and the grinder are machines of this type. The second characteristic of Fig. 3 describes a machine with variable capacity. In the inventory range  $0 < h < h^{\min}$  of the corresponding fabrication unit, the output  $k$  of the machine is proportional to the inventory  $h$ :

$$(2.1) \quad h = \tau k \text{ with } \tau = \text{const.}$$

In the range  $h^{\min} < h < h^{\max}$  the machine works at maximum power  $k^{\max}$ . The mixer, a set of machines of the same type with the first characteristic started in parallel are examples for machines of this second type. In the case of a set of machines, when  $h^{\min} < h < h^{\max}$  all machines of the set work at maximum power.

Knowing the characteristics of the machines, we are now enabled to calculate the inventory and the output functions. In the case of  $N$  fabrication units, forming the fabrication plant, we have  $2N$  unknown quantities  $h_i$  and  $k_i$ ,  $i = 1, 2, \dots, N$ . The mass conservation equation will hold for each fabrication unit, which gives us  $N$  equations. The machine characteristics of the  $N$  machines contained in the units complete the set of  $2N$  equations needed for the calculation of the  $2N$  unknown quantities  $h_i$  and  $k_i$ ,  $i = 1, 2, \dots, N$ .

To illustrate our ideas we will now consider some examples.

1<sup>st</sup> EXAMPLE. Fig. 4 shows a plant (or a part of it) consisting of 2 fabrication units with a feed-back. The incoming flow,  $X(t)$ , crosses the first and the second unit. The fraction  $\kappa$  of the second unit's output  $k_2$  goes back to the entrance of the first unit. The rest,  $(1-\kappa)k_2$ , goes out of the system. We can think of the combination of a glove-box with a mixer and a glove-box containing the press. The fraction  $\kappa$  of the pellets, formed by the press does not correspond to the specifications. We assume  $\kappa$  to be constant in agreement with our remark at the beginning of this section. Let the two machines have the first characteristic, shown in Fig. 3.

Thus we have the four equations

$$(2.2) \quad \begin{aligned} \frac{dh_1}{dt} &= X + \kappa k_2 - k_1 \\ \frac{dh_2}{dt} &= k_1 - k_2 \end{aligned}$$

$$k_1 = \begin{cases} 0 & h_1 < h_1^{\min} \\ k_1^{\max} & \text{for } h_1^{\min} \leq h_1 \leq h_1^{\max} \end{cases}$$

$$k_2 = \begin{cases} 0 & h_2 < h_2^{\min} \\ k_2^{\max} & \text{for } h_2^{\min} \leq h_2 \leq h_2^{\max}. \end{cases}$$

The first couple of equations are mass conservation equations, the second couple of equations describes the characteristics of the machines.

$h_1^{\min}$ ,  $h_1^{\max}$ ,  $h_2^{\min}$ ,  $h_2^{\max}$ ,  $k_1^{\max}$ ,  $k_2^{\max}$ ,  $X(t)$  and  $\kappa$  are known quantities.

Now let

$$(2.3) \quad \begin{cases} X = 0 & \text{for } t < 0 \\ X = X_0 \text{ (constant)} & \text{for } t \geq 0. \end{cases}$$

Then at the time  $t_1 = h_1^{\min}/X_0$  the machine of the first fabrication unit begins to work with the output  $k_1^{\max}$ . If  $X_0 > k_1^{\max}$ , it will continue

to run and at the time  $t_2 = t_1 + \frac{h_2^{\min}}{k_1^{\max}}$  the machine of the second

fabrication unit begins to work. The fraction  $\kappa$  of its output  $k_2^{\max}$  then goes back to the first unit. We have the following system of differential equations:

$$\begin{aligned}
 \frac{dh_1}{dt} &= X_0 && \text{for } 0 \leq t < t_1 \\
 \frac{dh_2}{dt} &= 0 && \text{for } 0 \leq t < t_1 \\
 \frac{dh_1}{dt} &= X_0 - k_1^{\max} && \text{for } t_1 \leq t < t_2 \\
 \frac{dh_2}{dt} &= k_1^{\max} && \text{for } t_1 \leq t < t_2 \\
 \frac{dh_1}{dt} &= X_0 + \kappa k_2^{\max} - k_1^{\max} && \text{for } t \geq t_2 \\
 (2.4) \quad \frac{dh_2}{dt} &= k_1^{\max} - k_2^{\max} && \text{for } t \geq t_2
 \end{aligned}$$

For the initial conditions  $h_1(t=0) = h_2(t=0) = 0$  we find the solutions

$$\begin{aligned}
 h_1 &= X_0 t && \text{for } 0 \leq t < t_1 \\
 h_2 &= 0 && \text{for } 0 \leq t < t_1 \\
 h_1 &= \{X_0 - k_1^{\max}\} \{t - t_1\} + h_1^{\min} && \text{for } t_1 \leq t < t_2 \\
 h_2 &= k_1^{\max} \{t - t_1\} && \text{for } t_1 \leq t < t_2 \\
 h_1 &= \{X_0 + \kappa k_2^{\max} - k_1^{\max}\} \{t - t_2\} + \\
 &\quad + \{X_0 - k_1^{\max}\} \{t_2 - t_1\} + h_1^{\min} && \text{for } t \geq t_2 \\
 (2.5) \quad h_2 &= \{k_1^{\max} - k_2^{\max}\} \{t - t_2\} + h_2^{\min} && \text{for } t \geq t_2.
 \end{aligned}$$

Fig. 5 shows the time dependence of the inventory functions  $h_1$  and  $h_2$  for the following set of constants:

$$\kappa = \frac{1}{8}; X_0 = 10 \frac{\text{kg Pu}}{\text{d}}; k_1^{\text{max}} = 9 \frac{\text{kg Pu}}{\text{d}}; k_2^{\text{max}} = 8 \frac{\text{kg Pu}}{\text{d}};$$

$$h_1^{\text{min}} = 5 \text{ kg Pu}; h_2^{\text{min}} = 4,5 \text{ kg Pu}; h_1^{\text{max}} = 15 \text{ kg Pu and}$$

$$h_2^{\text{max}} = 10 \text{ kg Pu.}$$

As we can see at once, this system cannot work for an unlimited time. At the time  $t = 5,75$  days the inventory of the first fabrication unit reaches the critical value  $h_1^{\text{max}} = 15 \text{ kg Pu}$  and now the operator of the plant has to diminish the input  $X_0$  into the system, e.g.

There exists a steady state operation of this system, as we can see at once from the equations (2.4). For  $t > t_2$ , we have  $h_1 = \text{const.}$  and  $h_2 = \text{const.}$  if

$$k_1^{\text{max}} = k_2^{\text{max}}$$

$$(2.6) \quad X_0 = (1-\kappa)k_2^{\text{max}}$$

because then  $dh_1/dt = dh_2/dt = 0$ .

These equations (2.6) indicate the fact, that in a steady state operation of a plant, all machines work with the same capacity and that the input ( $X$ ) into the plant must be equal to its output ( $\sqrt{1-\kappa}k_2$ ).

Thus in steady state operation the input into the system cannot be changed arbitrarily but is a fixed quantity.

This steady state can be reached at any inventory  $h^{\text{tot}} = h_1 + h_2$  of the plant within the following range:

$$(2.7) \quad h_1^{\text{min}} + h_2^{\text{min}} \leq h^{\text{tot}} \leq h_1^{\text{max}} + h_2^{\text{max}}.$$

To show this, let us consider once more our numerical example, this time in respecting the condition (2.6) of steady state operation.

Thus

$$\kappa = \frac{1}{8}; X_0 = \begin{cases} 8 \text{ kg Pu/d} & \text{for } 0 \leq t < t_2 \\ 7 \text{ kg Pu/d} & \text{for } t_2 \leq t \end{cases}$$

$$k_1^{\max} = k_2^{\max} = 8 \frac{\text{kg Pu}}{d} \quad \text{and } h_1^{\min} \quad \text{and } h_1^{\max} \quad (i = 1,2) \text{ as before.}$$

Fig. 6 shows how the operator can drive his plant at different inventory levels. In the first case he starts the machines of the two fabrication units at once when the minimum inventory  $h_1^{\min}$  is reached. Thus he works in a steady state with the minimum inventory 9,5 kg Pu. (The steady state condition (2.6) is fulfilled). In the second case he starts the machines when the inventory of the fabrication units has reached the maximum value  $h_1^{\max}$  resp.  $h_2^{\max}$ . Thus he works in a steady state with the maximum inventory 25 kg Pu.

For economical reasons it can be expected, that industrial fabrication plants will work with a minimum inventory, but as we have shown, this is by no means a necessity from the technical point of view.

2<sup>nd</sup> EXAMPLE. We consider the same system as shown in Fig.4. But now the two fabrication units contain two machines with the second characteristic shown in Fig. 3.

Thus we have the four equations

$$\frac{dh_1}{dt} = X + \kappa k_2 - k_1$$

$$(2.8) \quad \frac{dh_2}{dt} = k_1 - k_2$$

$$k_1 = \begin{cases} h_1/\tau_1 & \text{for } h_1 < h_1^{\min} \\ k_1^{\max} & \text{for } h_1^{\min} \leq h_1 \leq h_1^{\max} \end{cases}$$

$$k_2 = \begin{cases} h_2/\tau_2 & \text{for } h_2 < h_2^{\min} \\ k_2^{\max} & \text{for } h_2^{\min} \leq h_2 \leq h_2^{\max} \end{cases}$$

to determine the four unknown quantities  $h_1$ ,  $k_1$ ,  $h_2$  and  $k_2$ . The first couple are the mass conservation equations (the same as in the 1<sup>st</sup> example), the second couple of equations describe the machine's characteristics (see Fig. 3). We have to consider four different cases:

$$\begin{aligned} \text{1<sup>st</sup> case:} \quad & h_1 < h_1^{\min} \\ & h_2 < h_2^{\min}. \end{aligned}$$

Then the system (2.8) reduces to

$$\begin{aligned} (2.9) \quad & \frac{dh_1}{dt} = X + \kappa k_2 - k_1 \\ & \frac{dh_2}{dt} = k_1 - k_2 \\ & h_1 = \tau_1 k_1 \\ & h_2 = \tau_2 k_2. \end{aligned}$$

Hence with  $\tau_1$  and  $\tau_2$  constant (we assume  $X = X_0$  const.):

$$\begin{aligned} (2.10) \quad & \frac{dh_1}{dt} = X_0 + \frac{\kappa}{\tau_2} h_2 - \frac{1}{\tau_1} h_1 \\ & \frac{dh_2}{dt} = \frac{1}{\tau_1} h_1 - \frac{1}{\tau_2} h_2. \end{aligned}$$

This is an inhomogeneous system of two first order differential equations for the inventory functions  $h_1$  and  $h_2$ . The homogeneous system is

$$\begin{aligned} (2.11) \quad & \frac{dh_1}{dt} = -\frac{1}{\tau_1} h_1 + \frac{\kappa}{\tau_2} h_2 \\ & \frac{dh_2}{dt} = \frac{1}{\tau_1} h_1 - \frac{1}{\tau_2} h_2 \end{aligned}$$

and its characteristic equation is

$$(2.12) \quad (1 + \tau_1 \lambda) (1 + \tau_2 \lambda) = \kappa.$$

As  $0 < \kappa < 1$  and  $\tau_1, \tau_2 > 0$  hold and  $\kappa, \tau_1, \tau_2$  real, the roots  $\lambda_i$  of the characteristic equation (2.12) are always negative and real:

$$\lambda_1, \lambda_2 < 0 \text{ and real.}$$

Thus the solutions of the homogeneous system (2.11) tend to zero when  $t \rightarrow \infty$ .

There exists a steady state solution of the system (2.10):

$$(2.13) \quad h_{1\text{stead.}} = \frac{\tau_1 X_o}{1 - \kappa}$$

$$h_{2\text{stead.}} = \frac{\tau_2 X_o}{1 - \kappa}$$

which corresponds to the steady state in the first example (2.6), as we can see by using the last two equations of (2.9).

The general solution of the inhomogeneous system (2.10) is

$$(2.14) \quad h_1 = A e^{\lambda_1 t} + B e^{\lambda_2 t} + \frac{\tau_1 X_o}{1 - \kappa}$$

$$h_2 = \frac{\tau_2}{\kappa \tau_1} \left\{ A (1 + \lambda_1 \tau_1) e^{\lambda_1 t} + B (1 + \lambda_2 \tau_1) e^{\lambda_2 t} \right\} + \frac{\tau_2 X_o}{1 - \kappa}$$

Here  $\lambda_1$  and  $\lambda_2$  are the roots of the characteristic equation (2.12) and

$$A = h_1^o - \frac{\tau_1 X_o}{1 - \kappa} - B$$

$$B = \frac{1}{\tau_1 (\lambda_1 - \lambda_2)} \left\{ (h_1^o - \frac{\tau_1 X_o}{1 - \kappa}) (1 + \lambda_1 \tau_1) + \frac{\kappa \tau_1 X_o}{1 - \kappa} - \frac{\kappa \tau_1 h_2^o}{\tau_2} \right\}$$

with  $h_1^o$  and  $h_2^o$  as the initial values  $h_1(t = 0)$  resp.  $h_2(t = 0)$ . The output functions  $k_1$  and  $k_2$  can be calculated by dividing (2.14) by  $\tau_1$  resp.  $\tau_2$ .

If  $h_1^o = h_2^o = 0$ , then we have

$$\begin{aligned}
 h_1(t=0) &= 0 & ; & & h_2(t=0) &= 0; \\
 \frac{dh_1}{dt}(t=0) &= X_0 > 0; & & & \frac{dh_2}{dt}(t=0) &= 0; \\
 \frac{d^2h_2}{dt^2}(t=0) &= \frac{X_0}{\tau_1} > 0
 \end{aligned}
 \tag{2.15}$$

as can be seen from (2.10). In this case the inventory function of the first fabrication unit begins at zero for  $t = 0$ , but with a positive slope ( $X_0 > 0$ ). The inventory function of the second fabrication unit begins at zero too, for  $t = 0$ . Its slope is zero, but as  $\frac{d^2h_2}{dt^2}(t=0) > 0$ ,

it tends to positive values for  $t > 0$ .

Both functions  $h_1$  and  $h_2$  tend to the stationary solution (steady state 2.13) for  $t \rightarrow \infty$  if

$$\frac{\tau_1 X_0}{1 - \kappa} < h_1^{\min}$$

$$\frac{\tau_2 X_0}{1 - \kappa} < h_2^{\min}.$$

Fig. 7 shows the time development of the inventory functions  $h_1$  and  $h_2$  for the initial conditions  $h_1(t=0) = h_2(t=0) = 0$  and with the conditions (2.16) fulfilled.

2<sup>nd</sup> case.

$$\begin{cases} h_1 < h_2^{\min} \\ h_2^{\min} \leq h_2 \leq h_2^{\max} \end{cases}$$

Then the system (2.8) reduces to

$$\begin{aligned} \frac{dh_1}{dt} &= X_0 + \kappa k_2 - k_1 \\ \frac{dh_2}{dt} &= k_1 - k_2 \\ (2.17) \quad h_1 &= \tau_1 k_1 \\ k_2 &= k_2^{\max} (= h_2^{\min} / \tau_2). \end{aligned}$$

Hence

$$\begin{aligned} \frac{dh_1}{dt} &= X_0 + \kappa k_2^{\max} - \frac{1}{\tau_1} h_1 \\ (2.18) \quad \frac{dh_2}{dt} &= \frac{1}{\tau_1} h_1 - k_2^{\max}. \end{aligned}$$

This is a very simple type of a system of two coupled differential equations. Its general solution is

$$\begin{aligned} (2.19) \quad h_1 &= \left\{ h_1^0 - \tau_1 (X_0 + \kappa k_2^{\max}) \right\} e^{-t/\tau_1} + \tau_1 (X_0 + \kappa k_2^{\max}) \\ h_2 &= - \left\{ h_1^0 - \tau_1 (X_0 + \kappa k_2^{\max}) \right\} e^{-t/\tau_1} + \left\{ X_0 - (1-\kappa) k_2^{\max} \right\} t + \\ &\quad + h_2^0 + h_1^0 - \tau_1 (X_0 + \kappa k_2^{\max}), \end{aligned}$$

with  $h_1^0$  and  $h_2^0$  as initial values of  $h_1$  and  $h_2$ .

The system (2.18) has a steady state solution if

$$\begin{aligned} h_1 (t \rightarrow \infty) &= \tau_1 k_2^{\max} \text{ and} \\ h_1 (t \rightarrow \infty) &= \tau_1 \left\{ X_0 + \kappa k_2^{\max} \right\} \end{aligned}$$

hold. This means that

$$X_0 = (1 - \kappa) k_2^{\max}$$

must be fulfilled, in agreement with our preceding considerations. As can be seen, in this case the linear part of  $h_2$  in (2.19) is vanishing.

$$\text{3rd case.} \quad \begin{cases} h_1^{\min} \leq h_1 \leq h_1^{\max} \\ h_2 < h_2^{\min}. \end{cases}$$

Now we have the following system

$$\frac{dh_1}{dt} = X_0 + \kappa k_2 - k_1$$

$$\frac{dh_2}{dt} = k_1 - k_2$$

(2.20)

$$k_1 = k_1^{\max} (= h_1^{\min} / \tau_1)$$

$$h_2 = \tau_2 k_2$$

This leads to

$$\frac{dh_1}{dt} = \frac{\kappa}{\tau_2} h_2 + X_0 - k_1^{\max}$$

(2.21)

$$\frac{dh_2}{dt} = k_1^{\max} - \frac{1}{\tau_2} h_2.$$

This is a system analogous to (2.18) with the roles of  $h_1$  and  $h_2$  interchanged in a certain sense. The general solution of (2.21) with initial values  $h_1^0$  and  $h_2^0$  is

$$h_1 = -\kappa \left\{ h_2^0 - \tau_2 k_1^{\max} \right\} e^{-t/\tau_2} + \left\{ X_0 - (1-\kappa)k_1^{\max} \right\} t \\ + h_1^0 + \kappa \left\{ h_2^0 - \tau_2 k_1^{\max} \right\}$$

(2.22)

$$h_2 = \left\{ h_2^0 - \tau_2 k_1^{\max} \right\} e^{-t/\tau_2} + \tau_2 k_1^{\max}.$$

This time  $h_1$  contains a linear part, which disappears if  $X_0 = (1 - \kappa)k_1^{\max}$ , which means steady state operation of the system.

$$\text{4}^{\text{th}} \text{ case.} \quad \begin{cases} h_1^{\min} \leq h_1 \leq h_1^{\max} \\ h_2^{\min} \leq h_2 \leq h_2^{\max}. \end{cases}$$

In this case, both inventory functions have exceeded the minimum value. We have the following system of equations.

$$(2.23) \quad \begin{aligned} \frac{dh_1}{dt} &= X_0 + \kappa k_2 - k_1 \\ \frac{dh_2}{dt} &= k_1 - k_2 \\ k_1 &= k_1^{\max} \\ k_2 &= k_2^{\max} \end{aligned}$$

which is identical to the system (2.4) for  $t \geq t_2$ .

The general solution is (as indicated in (2.5))

$$(2.24) \quad \begin{aligned} h_1 &= \left\{ X_0 + \kappa k_2^{\max} - k_1^{\max} \right\} t + h_1^0 \\ h_2 &= \left\{ k_1^{\max} - k_2^{\max} \right\} t + h_2^0, \end{aligned}$$

with  $h_1^0$  and  $h_2^0$  as initial values of  $h_1$  and  $h_2$ .

The time development of the inventory functions  $h_1$  and  $h_2$  can now be found in the following way:

The system of two fabrication units starts at the zero value of the inventory functions. For a certain time it works in the proportional part of the characteristics of both machines with increasing inventory (corresponding to the considerations of case 1). In general after a certain time, one of the two fabrication units reaches the minimum value  $h^{\min}$  and then continues to operate at maximum capacity  $k^{\max}$  corresponding to the case 2 or 3. If both inventories have exceeded the minimum value of their inventory, the system will behave corresponding to the fourth case.

The system will reach the steady state after an initial period, if the quantities  $X_0, \kappa$ ,  $k_1^{\max}$  and  $k_2^{\max}$  fulfil the steady state condition (2.6), i.e.

$$(2.25) \quad \begin{aligned} k_1^{\max} &= k_2^{\max} \\ X_0 &= (1 - \kappa)k_2^{\max}. \end{aligned}$$

If these conditions are not fulfilled, the system can reach the steady state, if

$$\begin{aligned} k_1^{\max} &> \frac{X_0}{1 - \kappa} \\ k_2^{\max} &> \frac{X_0}{1 - \kappa}. \end{aligned}$$

Then the steady state is reached in the linear part of the machine's characteristic. The system works with the stationary values

$$(2.26) \quad \begin{aligned} k_1^{\text{stat}} &= \frac{X_0}{1 - \kappa} < k_1^{\max} \\ k_2^{\text{stat}} &= \frac{X_0}{1 - \kappa} < k_2^{\max} \\ h_1^{\text{stat}} &= \tau_1 k_1^{\text{stat}} \\ h_2^{\text{stat}} &= \tau_2 k_2^{\text{stat}} \end{aligned}$$

in accordance with (2.13) and (2.9).

3<sup>rd</sup> EXAMPLE. Fig. 8 shows a fabrication plant, consisting of five fabrication units. We can think of the first one containing the fuel store, the reprocessing part for pellets, the ceramic glove-box line up to the mechanical press, the second one containing the sintering furnace, the third one the grinder, the washing, drying and final control facilities, the fourth one the whole canning technology and the final product store (for the fabricated fuel elements) and the fifth one containing the waste store.

We will assume that the first, the second and the third fabrication unit contain machines with the first characteristic (see Fig. 3). As the fourth and the fifth fabrication unit do not have an output (during the period of time which we consider), they do not contain a "machine".

We have the following system of equations to describe the mass flow through the plant:

$$(2.27) \quad \left. \begin{aligned} \frac{dh_1}{dt} &= \kappa_2 k_2 - k_1 \\ \frac{dh_2}{dt} &= k_1 - k_2 \\ \frac{dh_3}{dt} &= (1-\kappa_2)k_2 - k_3 \\ \frac{dh_4}{dt} &= (1-\kappa_3)k_3 \\ \frac{dh_5}{dt} &= \kappa_3 k_3 \end{aligned} \right\} \text{mass conservation equations}$$

$$\left. \begin{aligned} k_1 &= \begin{cases} 0 & \text{for } t \leq t_1 \\ k_1^{\max} & \text{for } t > t_1 \end{cases} \\ k_2 &= \begin{cases} 0 & \text{for } t \leq t_2 \\ k_2^{\max} & \text{for } t > t_2 \end{cases} \\ k_3 &= \begin{cases} 0 & \text{for } t \leq t_3 \\ k_3^{\max} & \text{for } t > t_3 \end{cases} \end{aligned} \right\} \text{machine characteristics}$$

where  $t_1$  is the time when the unit 1 starts to work. Then  $t_2 = t_1 + h_2^{\min}/k_1^{\max}$  and  $t_3 = t_2 + h_3^{\min}/k_2^{\max}(1-\kappa_2)$ .

The general solution of the system (2.27) of differential equations can be found at once (because the  $k_i$  are constant):

$$\begin{aligned}
 h_1 &= \left\{ \kappa_2 k_2 - k_1 \right\} t + h_1^0 \\
 h_2 &= \left\{ k_1 - k_2 \right\} t + h_2^0 \\
 (2.28) \quad h_3 &= \left\{ (1-\kappa_2)k_2 - k_3 \right\} t + h_3^0 \\
 h_4 &= (1-\kappa_3)k_3 t + h_4^0 \\
 h_5 &= + \kappa_3 k_3 t + h_5^0,
 \end{aligned}$$

with  $h_i^0$  as initial values of the inventory functions  $h_i$ .  $h_4$  and  $h_5$  are linear increasing time-functions, as  $(1-\kappa_3)k_3 > 0$  and  $\kappa_3 k_3 > 0$ . This corresponds to the physical identification of the fourth and the fifth fabrication unit. In chapter four this example will be used as a rough description of the ALKEM plant. The solutions (2.28) will then be discussed.

There is only one point of interest here. As we can see immediately from (2.28) by adding all equations, we have for the total inventory of the

$$\text{plant } h^{\text{tot}} = \sum_{i=1}^5 h_i(t):$$

$$(2.29) \quad h^{\text{tot}} = \sum_{i=1}^5 h_i^0 = \text{const.} \quad x)$$

This means, that the nature of  $\kappa_2$  and  $\kappa_3$  - whether these quantities are stochastic variables or constants - does not affect the value of the total inventory  $h^{\text{tot}}$ . Thus our assumption, that  $\kappa_i$  is constant - corresponding to the mean value of the stochastic variable  $\kappa_i$  - can be justified. The only difference between a stochastic approach and the deterministic one is a small shift of the inventory of the fabrication units. The shift is small because of the smallness of the mean value of  $\kappa_i$  ( $\approx 0,05$ ).

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<sup>x)</sup> It can be seen directly from the mass conservation equations (2.27) that  $dh^{\text{tot}}/dt = 0$ .

### 2.1.2 The residence time function in a continuous model

As defined in the first chapter, the residence time  $T(t)$  indicates, how long the fuel, entering the fabrication unit at the time  $t$ , stays in this unit. We assume that the fuel crosses a fabrication unit in the form of pellets (or fuel elements). Then two pellets, entering the fabrication unit at the same time generally will not have the same residence time, as the pellets are in general mixed in the interior of the store contained in the fabrication unit.

We have to consider two possibilities:

- the pellets do not change their succession in the interior of the fabrication unit.
- there is a mixing of the pellets, changing their succession in the fabrication unit.

The first case will be considered in this section. The second one will be dealt with in the next chapter.

If the succession of the pellets is not changed in a fabrication unit with the output function  $k(t)$ , the pellet entering this unit at the time  $t_0$  has to wait, until all the inventory  $h(t_0)$  of the unit has left the unit. In mathematical terms

$$(2.30) \quad \int_{t_0}^{t_0 + T(t_0)} k(t) dt = h(t_0).$$

Here  $T(t_0)$  gives the residence time of our pellet. As we have shown in the preceding part of this chapter,  $k(t)$  and  $h(t)$  can be calculated. Thus (2.30) enables us, to calculate the residence time function  $T(t_0)$  for a fabrication unit, the inventory- and output functions of which are known.

The functional relationship (2.30) is in general not easy to handle, because the unknown quantity appears in the upper limit of an integral. But when the fabrication unit regarded here works with a constant output - in the case of a machine with the first characteristic and  $h(t_0) > h^{\min}$ , or in steady state operation ( $k$  and  $h$  constant) - the functional

relationship (2.30) reduces to the very simple equation

$$(2.31) \quad T(t_0) = \frac{h(t_0)}{k} .$$

This is the well known definition of residence time in fluid dynamics. The residence time of a pellet in two fabrication units can then be found as the sum of the residence times of each fabrication unit.

### 2.2.1 A stochastic model

This approach takes account of the fact that at the outputs of the press, the sintering furnace and the grinding the pellets must fulfil certain specifications before the next treatment, or before being definitively accepted. If a pellet has not fulfilled one of these conditions, it is returned into the mixer. We are interested in the investigation of stationary processes, i.e. processes with probability laws independent of time. It should be noted that this is the most simple case of stable processes, in which the different functions of time are bounded, but we shall not deal with this more complicated problem here. The great interest of stationary processes, is that they permit the use of statistical tests easier to handle, and this is the justification of such a study.

In the following pages we describe two discrete models. Then we shall discuss the extension of the preceding continuous models to the stochastic case.

### 2.2.2 Discrete case

General description. It should be remarked at the beginning that time does not play any particular role in the description of this model. We have to replace this continuous variable by a discrete one,  $n = 1, 2, \dots$ , the particular value  $n$  characterizing the  $n^{\text{th}}$  "operation" which we have to describe now. For this purpose, we must first establish the transfer function of each glove-box. Then we shall write the transfer function of the plant, define the concept of an operation and deduce the behaviour of the plant.

- a) The mixer. We consider a mixer which runs always at full capacity. This means, that during a certain time  $v_1$  we fill the mixer with a fixed quantity  $h$  of material coming from the two inputs  $x$  and  $y$  (Fig. 9). During the time  $v_2$  the mixer works and during a third interval of time  $v_3$  we fill the press with this quantity  $h$  of material.

- b) The press. The press box transforms the quantity  $h$  into the form of pellets. Each pellet is subjected to a test which decides if the pellet fulfils, or not, some conditions to be accepted for the sintering. A suitable assumption is that a pellet is accepted with the probability  $p_1$  or rejected with the probability  $q_1 = 1 - p_1$ .  $p_1$  is constant on a long period of time and can be estimated with sufficient precision, for a given press, by the proportion of "good" pellets in a large number of pellets manufactured by this press. If the mass  $h$  is approximately required for  $N$  pellets, we shall observe, when this total amount  $h$  will be treated in the press box,  $n_1$  "bad" pellets in the output  $u'$  (Fig. 10) and  $N - n_1$  "good" pellets in the output  $u$ , with the following binomial probability:

$$\Pr \left\{ u = N - n_1 \right\} = \Pr \left\{ u' = n_1 \right\} = \binom{N}{n_1} q_1^{n_1} p_1^{N - n_1}$$

$v_3$  is the time required for treating the amount  $h$  of material.

- c) The sintering furnace. The sintering box receives from the press (input  $u$ ) the random number  $u_1 = N - n_1$  of pellets. We can suppose that under the same conditions as in the case of the press there exists a probability  $p_2$  that a pellet, after treatment in this box, will fulfil the conditions to be accepted in the grinding box. We have to make another assumption, which is quite acceptable:  $p_2$  does not vary with the number of pellets present in the sintering furnace during this treatment. Consequently, after having treated  $u_1$  pellets, we observe in the output  $v$  a total number  $v_1$  of "good" pellets, and in the output  $v'$  a number  $v'_1$  of "bad" pellets (Fig. 11a) with the probability:

$$\Pr \left\{ v = u_1 - v'_1 \right\} = \Pr \left\{ v' = v'_1 \right\} = \binom{u_1}{v'_1} q_2^{v'_1} p_2^{u_1 - v'_1}; \quad q_2 = 1 - p_2.$$

Such a glove-box can run in two different manners:

- i) Running at variable capacity. The sintering furnace works each time with a number  $u_1$  (not necessarily fixed) of pellets.

In this case we see that the probability law of the output  $v$  depends on the actual value of the input  $u_1$ . This way of running

includes the case where the pellets are treated one after the other, or the case where the sintering process begins only when  $u_1$  pellets coming from the press are arrived in the sintering furnace.

- ii) Running at constant capacity. In this case the sintering begins only if a constant number  $N_2$  of pellets are available. A storage facility  $H_2$  is needed in this case (Fig. 11b). The output  $v$  is then the product  $v = a \cdot t$  of two random variables. The first one follows the binomial law:

$$\Pr \left\{ a = N_2 - v_1 \right\} = \binom{N_2}{v_1} q_2^{v_1} p_2^{N_2 - v_1}, \quad 0 \leq a \leq N_2.$$

The second one can take the values 0 or 1 with the probability

$$\Pr \left\{ t = 1 \right\} = \text{Prob} \left\{ \bar{h}_2 > N_2 \right\}$$

$$\Pr \left\{ t = 0 \right\} = \text{Prob} \left\{ \bar{h}_2 < N_2 \right\}$$

where  $h_2$  is the number of pellets in the storage facility  $H_2$  just before the sintering process. The same argumentation is valid for  $v' = a \cdot t$ , with

$$\Pr \left\{ a' = v_1 \right\} = \binom{N_2}{v_1} q_2^{v_1} p_2^{N_2 - v_1}$$

And we denote by  $v_4$  the time required for treating the variable quantity  $u_1$  of the constant one  $N_2$ .

- d) The grinding. The running of the grinding box is similar to the preceding one. We shall denote the corresponding variables by  $\omega$ ,  $\omega'$ ,  $H'_3$ ,  $N_3$ ,  $h_3$ ,  $p_3$ ,  $q_3$  (Fig. 12).

- i) When the grinder works at variable capacity, the probability law is

$$\Pr \left\{ \omega = v_1 - \omega_1 \right\} = \Pr \left\{ \omega' = \omega_1 \right\} = \binom{v_1}{\omega_1} q_3^{\omega_1} p_3^{v_1 - \omega_1}$$

ii) When the grinder works at constant capacity we have

$$\omega = bt' ; \omega' = b' t' \quad \text{with}$$

$$\Pr \{ b = N_3^{-\omega_1} \} = \Pr \{ b' = \omega_1 \} = \binom{N_3}{\omega_1} q_3^{N_3 - \omega_1} p_3^{\omega_1}$$

$$\Pr \{ t' = 1 \} = \Pr \{ \bar{h}_3 \geq N_3 \}$$

$$\Pr \{ t' = 0 \} = \Pr \{ \bar{h}_3 < N_3 \};$$

$v_5$  is the time needed to treat the quantities  $v_1$ , or  $N_3$ .

e) The storage. The outputs  $u'$ ,  $v'$  and  $\omega'$  are collected in a storage facility (Fig. 13). The state of this storage will be defined by the number  $Z$  of pellets present in this box before the arrival of the quantities  $u'$ ,  $v'$  and  $\omega'$ .  $\bar{Z}$  will denote the state of the storage when these quantities have been added. So we have:

$$\bar{Z} = Z + u' + v' + \omega'.$$

It is clear that  $Z$  and  $\bar{Z}$  can denote the equivalent mass of material. From this storage facility the quantity  $y$  is filled into the mixer according to the following rule:

$$y = \xi h \quad \text{if } \bar{Z} \geq \xi$$

$$y = \bar{Z} \quad \text{if } \bar{Z} < \xi h ; 0 < \xi \leq 1, \xi \text{ constant.}$$

Thus we have described in a), b), c), d), and e) a sequence of elementary operations. At the first step, we fill the mixer with material coming from  $x$  and  $y$ ; the quantity  $y$  is given by the preceding law and  $x = h - y$ . At the last step we collect in the output of the plant a random number of the pellets meeting all the required specifications.

The other pellets are collected in a storage facility for a next cycle of elementary operations. By convention, we shall define this cycle as an "operation". The running of the plant from the beginning to the end of the  $n^{\text{th}}$  operation is then described by the sequence of the

first, the second, until the  $n^{\text{th}}$  operation. We characterize each quantity by an index  $n$  indicating that it corresponds to the  $n^{\text{th}}$  operation. The investigation of the functioning of a plant is thus reduced to the study of the sequence of the random variables  $u_n; u'_n; v_n; v'_n; \omega_n; \omega'_n; Z_n; \bar{Z}_n; x_n; y_n; h_{2n}; h_{3n}$ . We shall remark that the time necessary for one operation, i.e.  $v_1 + v_2 + v_3 + v_4 + v_5$ , can vary arbitrarily. This is the reason for which the time does not play any particular part here. If by

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chance  $\sum_{i=1}^5 v_i$  follows a particular law (a statistic or deterministic one) then we could interpret the running of the plant as a function of time. But this interpretation is not at all necessary.

In the following pages, we consider two different runnings for a fabrication plant. The first one, called running with variable capacities, corresponds to a sintering process and a grinding process with variable capacities as described above. The second one, the so called running with constant capacities, corresponds to the running of these glove-boxes with constant capacities.

### 1<sup>st</sup> EXAMPLE: A fabrication plant running with variable capacities

In this case the plant is represented by Fig. 13. We have three boxes, the mixer, the storage facility, and a fabrication unit consisting of the press, the sintering furnace and the grinding box. Conventionally, all the variables will be expressed in terms of the corresponding number of pellets. The probability that a pellet has fulfilled all the required conditions for being definitively accepted is obviously  $p = p_1 p_2 p_3$ . Let  $q = 1-p$  be the probability that at least one condition is not fulfilled. The outputs  $\omega$  and  $c = u' + v' + \omega'$  of the fabrication unit are the following random variables:

$$\begin{cases} \omega = N-k \\ c = k \end{cases} \quad \text{with}$$

$$\Pr \{ \omega = N-k \} = \Pr \{ c = k \} = \binom{N}{k} q^k p^{N-k}$$

In Fig. 14 we give a diagram of the different notations for the variables and the sequence of their measurement.

Equations. The mass conservation equations are

$$(2.32) \quad x_n + y_n = N \quad \text{for the mixer box}$$

$$(2.33) \quad \begin{cases} \omega_n & = N - k_n \\ c_n & = k_n \end{cases} \quad \text{for the fabrication unit}$$

$$(2.34) \quad \begin{cases} \bar{z}_{n-1} - y_n & = z_n \\ z_n + c_n & = \bar{z}_n \end{cases} \quad \text{for the storage facility}$$

The solution of the system (2.33) is obvious: the  $\omega_n$  are independent and identically distributed random variables. This is also true for the  $c_n$  and we have:

$$\Pr \{ \omega_n = N - k_n \} = \Pr \{ c_n = k_n \} = \binom{N}{k_n} q^{k_n} p^{N - k_n}.$$

On the other hand,

$$\begin{cases} y_n = \xi N & \text{if } \bar{z}_{n-1} - \xi N \geq 0 \\ y_n = \bar{z}_{n-1} & \text{otherwise.} \end{cases}$$

Consequently, we have:

$$\begin{cases} z_n = \bar{z}_{n-1} - \xi N & \text{if } \bar{z}_{n-1} - \xi N \geq 0 \\ z_n = 0 & \text{otherwise.} \end{cases}$$

Or equivalently:

$$z_n = \text{Max} \left\{ 0; \bar{z}_{n-1} - \xi N \right\}$$

$$z_n = \text{Max} \left\{ 0; z_{n-1} + k_{n-1} - \xi N \right\}.$$

Taking account of the fact that  $Z_1 = 0$  and putting:

$$F_n(z) = \Pr \{ Z_n < z \},$$

$$g(k' = m - \xi N) = \binom{N}{m} q^m p^{N-m}$$

it follows from a straight forward calculation that

$$(2.35) \quad F_n(k) = \sum_{k' \leq k} F_{n-1}(k-k') g(k').$$

And, following D.V. LINDLEY [7], we know that a non degenerate limit distribution

$$F(k) = \lim_{n \rightarrow \infty} F_n(k)$$

exists, if and only if  $E(k') = E(k) - \xi N = q^{N-\xi N} < 0$ ;  $\bar{E}(y)$  is the mean value of  $y$ ; or equivalently  $p_1 p_2 p_3 > 1 - \xi$ . In this case, the inventory function of the plant is approximatively, at a long run, a stochastic variable with a well defined distribution function  $F(k)$ , solution of the equation

$$F(k) = \sum_{m < k + \xi N} F(k - m + \xi N) \binom{N}{m} q^m p^{N-m}.$$

Otherwise,  $\lim_{n \rightarrow \infty} F_n(z) = 0$  for all  $z > 0$ . This means, that it is sure to observe an inventory greater than all possible values  $z$  given in advance, after a certain time which can be calculated. Now:

$$\Pr \{ \bar{Z}_n = z \} = \sum_{a+c=z} \Pr \{ Z_n = a \} \Pr \{ c_n = c \}$$

$$\Pr \{ y_n = \xi N \} = \Pr \{ x_n = N - \xi N \} = \Pr \{ \bar{Z}_{n-1} > \xi N \}$$

$$\Pr \{ y_n = z \} = \Pr \{ x_n = N - z \} = \Pr \{ \bar{Z}_{n-1} = z \} \text{ for } z < \xi N.$$

The behaviour of the plant for  $k$  constant is defined by:

$$\left\{ \begin{array}{l} z_n = (n-1) (k-\xi N) \\ z_n = (n-1) (k-\xi N) + k \\ y_n = \xi N \\ x_n = N - \xi N \end{array} \right. \quad \text{if } k-\xi N > 0.$$

$$\left\{ \begin{array}{l} z_n = 0 \\ z_n = y_n = \xi N \\ x_n = N-\xi N \text{ for all } n > 1, \text{ if } k-\xi N < 0. \end{array} \right.$$

2<sup>nd</sup> EXAMPLE: A fabrication plant running with constant capacities

In this case, we have to take account of the two transfer functions described in c) ii) and d) ii). We shall discuss here only the interesting case where  $h \leq N_2 \leq N_3$ . The physical meaning of this condition is clear: the output of each glove-box is less than or equal to the capacity of the storage facility of the successive box. Intuitively, we can think that this condition is sufficient for leading to an equilibrium state in this storage facilities. We shall show that this is true and, furthermore, that in this state the amounts of the storage facilities  $N_2$  and  $N_3$  are stochastic variables uniformly distributed between 0 and  $N_2-1$ , for the first one, 0 and  $N_3-1$  for the second one.

If  $h_{2,n}$  is the number of pellets in  $N_2$  at the end of the  $n^{\text{th}}$  operation, we have

$$\left\{ \begin{array}{l} h_{2,n+1} = h_{2,n} + u_n \quad \text{if } h_{2,n} + u_n < N_2 \\ \quad \quad \quad = h_{2,n} + u_n - N_2 \quad \text{otherwise.} \end{array} \right.$$

For illustrating these considerations, we give in Fig. 15 a the first six steps of such a process, with  $N_2 = 5$  and  $u_n$  taking the values 0, 1, 2 and 3 with positive probabilities. The algebraic operation

$$h_{2,n+1} = h_{2,n} + u_n \quad \text{modulo } N_2$$

corresponds to a mapping of the  $N$  axis on a circle of length  $N_2$ , as we can see on Fig. 15b. It follows that the random walk defined by the initial state  $h_{2,0} = 0$  and by the transition probabilities  $P(0, h_2) = g(u)$  is a random walk on a finite cyclic group. If  $M$  is the markov matrix, the state of the storage facility  $H_2$  at the end of the  $n^{\text{th}}$  operation, i.e. the number  $h_{2,n}$  of the pellets present in  $H_2$  at this time, is given by the probability vector:

$$\left\{ \Pr(h_{2,n} = 0), \dots, \Pr(h_{2,n} = N_2 - 1) \right\} = \{1, 0, \dots, 0\} M^n.$$

At the same time,  $\bar{h}_{2,n}$  is given by

$$\Pr\{\bar{h}_{2,n} = z\} = \sum_{h+n=z} \Pr\{h_{2,n-1} = h\} g(u)$$

and the auxiliary stochastic variable  $t_n$  is defined by:

$$\Pr\{t_n = 0\} = \Pr\{\bar{h}_{2,n} < N_2\} = \sum_{z < N_2} \sum_{h+u=z} \Pr\{h_{2,n-1} = h\} \binom{N}{u} q_1^{N-u} p_1^u$$

$$\Pr\{t_n = 1\} = \Pr\{h_{2,n} > N_2\} = \sum_{z > N_2} \sum_{h+u=z} \Pr\{h_{2,n-1} = h\} \binom{N}{u} q_1^u p_1^{N-u}.$$

The output of the sintering furnace is then described by:

$$\begin{cases} \Pr\{v_n = N_2 - v\} = \binom{N_2}{v} q_2^v p_2^{N_2 - v} \cdot \text{Prob}\{t_n = 1\} & \text{for } v \neq N_2 \\ \Pr\{v_n = 0\} = q_2^{N_2} \text{Prob}\{t_n = 1\} + \text{Prob}\{t_n = 0\} \end{cases}$$

$$\begin{cases} \Pr\{v'_n = v\} = \binom{N_2}{v} q_2^v p_2^{N_2 - v} \cdot \text{Prob}\{t_n = 1\} & \text{for } v \neq 0 \\ \Pr\{v'_n = 0\} = p_2^{N_2} \text{Prob}\{t_n = 1\} + \text{Prob}\{t_n = 0\}. \end{cases}$$

Now, it is well known (see for instance FELLER [8]) that the limit matrix

$$Q = \lim_{n \rightarrow \infty} M^n \text{ exists and is such that each element } Q_{ij} \text{ is equal to } 1/N_2.$$

This fact is a direct consequence of the group structure of the points over a circle with the usual addition as group operation, this implying that  $M$  is bistochastic, i.e., the sum of the elements of a line or a column is equal to 1. In Fig. 16 we give the corresponding matrix of the markov chain described above, and its limit matrix.

Let  $\bar{h}_2, h_2, t, t', v$  and  $v'$  be the limits of the variables  $\bar{h}_{2,n}, h_{2,n}, t_n, t'_n, v_n$  and  $v'_n$  for  $n \rightarrow \infty$ , at a long run, the behaviour of the sintering process is approximatively described by:

$$\Pr \{h_2 = h\} = \frac{1}{N_2} \text{ for } h = 0, 1, \dots, N_2-1.$$

$$\Pr \{\bar{h}_2 = Z\} = \frac{1}{N_2} \sum \left\{ \binom{N}{u} q_2^{N-u} p_2^u / h + u = Z, h = 0, 1, \dots, N_2-1 \right\}$$

$$\Pr \{t = 0\} = \frac{1}{N_2} \sum_{Z < N_2} \sum \left\{ \binom{N}{u} q_2^{N-u} p_2^u / h + u = Z, h = 0, 1, \dots, N_2-1 \right\}$$

$$\Pr \{t = 1\} = \frac{1}{N_2} \sum_{Z \geq N_2} \sum \left\{ \binom{N}{u} q_2^{N-u} p_2^u / h + u = Z, h = 0, 1, \dots, N_2-1 \right\}$$

$$\left\{ \begin{array}{l} \Pr \{v = N_2 - k\} = \binom{N_2}{k} q_2^k p_2^{N_2 - k} \cdot \Pr \{t = 1\} \text{ for } k \neq N_2 \\ \Pr \{v = 0\} = q_2^{N_2} \cdot \Pr \{t = 1\} + \Pr \{t = 0\} \end{array} \right.$$

$$\left\{ \begin{array}{l} \Pr \{v = k\} = \binom{N_2}{k} q_2^k p_2^{N_2 - k} \cdot \Pr \{t = 1\} \text{ for } k \neq 0 \\ \Pr \{v' = 0\} = p_2^{N_2} \cdot \Pr \{t = 1\} + \Pr \{t = 0\}. \end{array} \right.$$

The description of the running of the grinding box follows the same lines.

We have:

$$\begin{cases} h_{3,n+1} = h_{3,n} + v_n & \text{if } h_{3,n} + v_n < N_3 \\ = h_{3,n} + v_n - N_3 & \text{otherwise.} \end{cases}$$

But now, the random variables,  $v_n$  are no longer identically distributed for  $n = 0, 1, \dots$ . This means, that in the markov chain on the circle of length  $N_3$ , each step  $n$  is performed following the particular matrix of transitions  $M_{3,n}$  of which the element  $(i, j)$  is defined by

$$\begin{aligned} m_{3,n}(i, j) &= \Pr \{ v_n = j - i \} && \text{if } j \geq i \\ &= \Pr \{ v_n = N_3 + j - i \} && \text{if } j < i. \end{aligned}$$

Thus, at the  $n^{\text{th}}$  step the vector of probabilities of the states is:

$$\left\{ \Pr \{ h_{3,n} = 0 \}, \dots, \Pr \{ h_{3,n} = N_3 - 1 \} \right\} = \left\{ 1, 0, 0, \dots, 0 \right\}_{k=1}^n \prod_{k=1}^n M_{3,k}.$$

The other variables are defined by:

$$\Pr \{ \bar{h}_{3,n} = Z \} = \sum_{h+v=Z} \Pr \{ h_{3,n-1} = h \} \Pr \{ v_n = v \}$$

$$\Pr \{ t'_n = 1 \} = \Pr \{ \bar{h}_{3,n} \geq N_3 \} = \sum_{Z > N_3} \sum_{h+v=Z} \Pr \{ h_{3,n-1} = h \} \Pr \{ v_n = v \}$$

$$\Pr \{ t'_n = 0 \} = \Pr \{ \bar{h}_{3,n} \leq N_3 \} = \sum_{Z < N_3} \sum_{h+v=Z} \Pr \{ h_{3,n-1} = h \} \Pr \{ v_n = v \}.$$

The probability law of the outputs is:

$$\begin{cases} \Pr \{ \omega_n = N_3 - \omega \} = \binom{N_3}{\omega} q_3^\omega p_3^{N_3 - \omega} \Pr \{ t'_n = 1 \} & \text{for } \omega \neq N_3 \\ \Pr \{ \omega_n = 0 \} = q_3^{N_3} \cdot \text{Prob} \{ t' = 1 \} + \Pr \{ t'_n = 0 \} \end{cases}$$

$$\begin{cases} \Pr \left\{ \omega'_n = \omega \right\} = \binom{N_3}{\omega} q_3^\omega p_3^{N_3 - \omega} \cdot \Pr \left\{ t'_n = 1 \right\} \text{ for } \omega \neq 0 \\ \Pr \left\{ \omega'_n = 0 \right\} = p_3 \sum_{t'_n=1}^{N_3} \Pr \left\{ t'_n = 1 \right\} + \Pr \left\{ t'_n = 0 \right\}. \end{cases}$$

Now, when  $n \rightarrow \infty$  we have seen that  $v = \lim_{u \rightarrow \infty} v_n$  exists. It follows

that

$$\lim_{n \rightarrow \infty} M_{3,n} = M_3$$

exists.

Furthermore, it has been shown by J. WOLFOVITZ [9], that the infinite product of stochastic matrices with one ergodic class is a matrix with identical lines. The bistochastic property being invariant under multiplication, this result leads to the conclusion that the completely simple semi group of the compact semi-group of finite bistochastic matrices is reduced to the idempotent matrix of which all the elements are equal.

Then

$$\lim_{n \rightarrow \infty} \prod_{k=1}^n M_{3,k} = Q_3 \text{ with } \|Q_3\|_{ij} = \frac{1}{N_3}$$

$$\lim_{n \rightarrow \infty} \Pr \left\{ h_{3,n} = h \right\} = \frac{1}{N_3} = \Pr \left\{ h_3 = h \right\}, \quad h_3 = \lim_{n \rightarrow \infty} h_{3,n}.$$

The other limits are easily derived:

$$\begin{cases} \Pr \left\{ \bar{h}_3 = Z \right\} = \frac{1}{N_3} \sum \left\{ \Pr(v) / h + v = Z; h = 0, 1, \dots, N_3 - 1 \right\} \\ \Pr \left\{ t' = 1 \right\} = \frac{1}{N_3} \sum_{Z \geq N_3} \sum \left\{ \Pr(v) / h + v = Z; h = 0, 1, \dots, N - 1 \right\} \\ \Pr \left\{ t' = 0 \right\} = \frac{1}{N_3} \sum_{Z < N_3} \sum \left\{ \Pr(v) / h + v = Z; h = 0, 1, \dots, N - 1 \right\} \end{cases}$$

And:

$$\left\{ \begin{array}{l} \Pr \{ \omega = N_3 - k \} = \binom{N_3}{k} q_3^k p_3^{N_3 - k} \cdot \Pr \{ t' = 1 \} \text{ for } k \neq N_3 \\ \Pr \{ \omega = 0 \} = q_3^{N_3} \cdot \Pr \{ Z' = 1 \} + \Pr \{ t' = 0 \} \end{array} \right.$$

$$\left\{ \begin{array}{l} \Pr \{ \omega = k \} = \binom{N_3}{k} q_3^k p_3^{N_3 - k} \cdot \Pr \{ t' = 1 \} \text{ for } k \neq 0 \\ \Pr \{ \omega = 0 \} = p_3^{N_3} \cdot \Pr \{ t' = 1 \} + \Pr \{ t' = 0 \}. \end{array} \right.$$

Let us consider now the variables defining the stochastic inventory function in the storage box, and firstly the input  $c'_n = u'_n + v'_n + \omega'_n$ .

The random variables  $u'_n$  are obviously independent and identically distributed;  $v'_n$  depends on  $t_n$ , or equivalently on  $\bar{h}_{2,n}$ , or equivalently on  $h_{2,n-1}$  and  $u_n$ , or on  $h_{2,n-1}$  and  $u'_n$ ;  $u'_n$  depends also on the result of an independent random experiment obeying to the binomial law  $(N_2, p_2)$ ; similarly  $\omega'_n$  depends on  $h_{3,n-1}$ ,  $v'_n$  and on the result of the independent random experiment  $(N_3, p_3)$ . Consequently,  $c'_n$  is the sum of three variables  $u'_n$ ,  $v'_n$ ,  $\omega'_n$ , each of them depending on the previous one. So the law of  $c'_n$  depends on  $h_{2,n-1}$ ,  $h_{3,n-1}$  and  $u'_n$ . The  $c_n$  are thus always independent, but not more identically distributed. The probability law of  $c_n$ , for instance, is

$$\Pr \{ c'_n = c' \} = \sum \left\{ \Pr \{ u' = u'_1 \} \cdot \Pr \{ v'_n = v'_1 / u' = u'_1 \} \Pr \{ \omega'_n = \omega'_1 / v'_n = v' \} \right. \\ \left. / u'_1 + v'_1 + \omega'_1 = c' \right\}.$$

This expression can be deduced from the preceding equations, but we do not give it here, because of the length of the formula.

We have to note that the limit  $\lim_{n \rightarrow \infty} \Pr \{ c'_n = c' \}$  exists, as a consequence of the fact that all the summations are finite and each term in these sums has a limit. This expression seems to be difficult to handle in analytical form. But the passage to the limit shows that there exists a stochastic variable  $c' = \lim_{n \rightarrow \infty} c'_n$ , of which the probability law can be calculated. Denoting

by  $k_n(c')$  the frequency function of  $c'_n$  and  $k(c')$  the one of  $k(c')$  we have again

$$F_u(z) = \sum_{c' \leq z} F_{n-1}(z-c') k_n(c')$$

$$k(c') = \lim_{n \rightarrow \infty} k_n(c').$$

The stationary conditions in this case are not so easy to formulate as the previous one. In a forthcoming paper, we shall discuss this case.

### 2.2.3 Some remarks on continuous stochastic models

Adopting the notations of the section 2.1.1 we want to show how to solve an equation of the type

$$(2.36) \quad \tau \frac{dk}{dt} + (1-\kappa) k - X = 0$$

when  $\kappa$  is not constant but a random variable. The equation (2.36) can be deduced for the system given in Fig. 4 with the hypothesis

$$\begin{cases} h_1 = \tau_1 k_1 \\ h_2 = \text{constant} \\ X = \text{constant.} \end{cases}$$

This stochastic equation can be considered as giving an approximate description of material flow through a fabrication unit. Here we are interested uniquely in the solution of (2.36) under various assumptions on  $\kappa$ :

Let  $\kappa$  be constant in each interval of time  $\{n\gamma; (n+1)\gamma\}$ ,  $n = 0, 1, \dots$ .

If  $\kappa_n$  is the value of  $\kappa$  in this interval, the random variables  $\{\kappa_n\}$  are independent and identically distributed; the common probability law is given by

$$\begin{cases} \kappa_n = \alpha & \text{with probability } p \\ \kappa_n = \beta & \text{with probability } q = 1 - p \\ 0 \leq \alpha, \beta \leq 1. \end{cases}$$

Let us assume that  $\alpha \neq 1$  and  $\beta \neq 1$ . It is clear that if  $k(n) = k_n$  is the value of  $k(t)$  at time  $t=n$ , there are two possibilities for the solution of (2.36) in the interval  $\underline{[n\tau; (n+1)\tau]}$ :

$$(2.37) \quad \left\{ \begin{array}{l} k(t) = e^{-\frac{1-\alpha}{\tau}(t-t_n)} \cdot k_n + \frac{X}{1-\alpha} \left( 1 - e^{-\frac{1-\alpha}{\tau}(t-t_n)} \right) \\ k(t) = e^{-\frac{1-\beta}{\tau}(t-t_n)} \cdot k_n + \frac{X}{1-\beta} \left( 1 - e^{-\frac{1-\beta}{\tau}(t-t_n)} \right) \end{array} \right.$$

The first solution is chosen with probability  $p$ , the second one with probability  $q$ . The equations (2.37) and the initial condition  $k_0=0$  define the random solutions of (2.36). They consist of a set of curves compounded of pieces of exponentials (2.37), see Fig. 17. This set of curves is bounded, when  $\alpha < \beta$ ,

$$\text{below by: } k(t) = \frac{X}{1-\alpha} \left( 1 - e^{-\frac{1-\alpha}{\tau} t} \right),$$

$$\text{above by: } k(t) = \frac{X}{1-\beta} \left( 1 - e^{-\frac{1-\beta}{\tau} t} \right).$$

We can note that a realization of the stochastic process defined by (2.36), i.e. one of the solutions of this equation, reaches one and only one time a value  $k(t)$  in the interval  $\underline{[0, \frac{X}{1-\alpha}]}$ . This set of values is the so called transient set. But, in the interval  $\underline{[\frac{X}{1-\alpha}; \frac{X}{1-\beta}]}$  the same value of  $k(t)$  can possibly be reached several times. So, we are led to the problem of the determination of the probability law of the possible values  $k(t)$ . We introduce the following abbreviations:

$$\left\{ \begin{array}{l} a = e^{-\frac{1-\alpha}{\tau} v} ; b = \frac{X}{1-\alpha} \left( 1 - e^{-\frac{1-\alpha}{\tau} v} \right) \\ c = e^{-\frac{1-\beta}{\tau} v} ; d = \frac{X}{1-\beta} \left( 1 - e^{-\frac{1-\beta}{\tau} v} \right) \end{array} \right.$$

Let  $A$  be a stochastic variable which takes the value  $a$  with probability  $p$  and  $c$  with probability  $q$ , and  $B$  a stochastic variable which takes the value  $b$  with probability  $p$  and  $d$  with probability  $q$ . If, furthermore, we note  $A_n, B_n$  the random variables corresponding to  $t_n$ , (2.37) can be written:

$$(2.38) \quad k_{n+1} = A_n k_n + B_n.$$

And we have

$$\begin{aligned} k_{n+1} &= A_n (A_{n-1} k_{n-1} + B_{n-1}) + B_n \\ &= A_n A_{n-1} k_{n-1} + A_n B_{n-1} + B_n. \end{aligned}$$

But according to the hypothesis of independence, we can take the mean value of this equation:

$$\bar{k}_{n+1} = \bar{A}_n \bar{A}_{n-1} \bar{k}_{n-1} + \bar{A}_n \bar{B}_{n-1} + \bar{B}_n.$$

Putting

$$\begin{cases} \bar{A}_n = pa + qc = M \\ \bar{B}_n = pb + qd = N \end{cases} \quad \text{for all } n,$$

we have:

$$\bar{k}_{n+1} = M^2 \bar{k}_{n-1} + MN + N.$$

Generalizing this result we obtain finally:

$$\begin{aligned} \bar{k}_{n+1} &= M^n \bar{k}_0 + N \frac{M^n - 1}{M - 1}, \\ (2.39) \quad \bar{k}_{n+1} &= N \frac{M^n - 1}{M - 1} \quad \text{because } k_0 = 0. \end{aligned}$$

A similar calculation can be developed for all the moments  $\bar{k}_{n+1}^u$ . We note that

$$(2.40) \quad \lim_{n \rightarrow \infty} \bar{k}_{n+1} = \frac{M}{1-M}$$

exists.

If  $k_{n+1}$  converges towards a random variable  $k$  when  $n$  goes to infinity, this limit must be the mean value of  $k$ . To prove that  $k$  exists, it needs to note that, considering in (2.39)  $k_{n+1}$  defined by the application of the random linear transformation  $Q = (A, B)$  on  $k_n$ , it is justified to apply the Banach fixed point theorem [10]. Consequently, we know that  $k = \lim_{n \rightarrow \infty} k_n$  exists almost surely, and that if  $F(z)$  is the distribution function of  $k$ , writing that the two variables  $\zeta$  and  $\xi$  related by the equality  $\zeta = A_n \xi + B_n$ , have the same  $F(z)$ , we obtain:

$$\frac{p}{a} dF\left(\frac{z-b}{a}\right) + \frac{q}{c} dF\left(\frac{z-d}{c}\right) = dF(z).$$

From this equation we can for instance deduce the following algebraic equation for the first moment  $k$ :

$$\bar{k} = \int_{-\infty}^{+\infty} z dF = \frac{p}{a} \int_{-\infty}^{+\infty} z dF\left(\frac{z-b}{a}\right) + \frac{q}{c} \int_{-\infty}^{+\infty} z dF\left(\frac{z-d}{c}\right), \text{ or}$$

$$\bar{k} = \frac{pb+qd}{1-pa-qc}$$

which is identically equal to (2.40). The same calculation can be performed for each moment.

We also note that the mean solution of (2.36) differs from the solution of this same equation written with the mean value  $\kappa = pa + q\beta$ . The reason of that is easy to find. It suffices to remark, that when we take the mean value of each term in (2.36), we have

$$\overline{\kappa \cdot k(t)} = \bar{\kappa} \cdot \bar{k}(t) \quad \text{only for } t = t_n.$$

We can also verify that the difference  $\Delta$  of the two solutions:

$$\Delta = \frac{\frac{pX}{1-\beta} \left(1 - e^{-\frac{1-\alpha}{\tau} v}\right)}{1 - pe^{-\frac{1-\alpha}{\tau} v}} + \frac{\frac{qX}{1-\alpha} \left(1 - e^{-\frac{1-\beta}{\tau} v}\right)}{-qe^{-\frac{1-\beta}{\tau} v}} - \frac{X}{1-pa - q\beta}$$

converges towards zero with  $\frac{v}{\tau}$ .

On the other hand, the preceding argumentation is also valid if  $\kappa$  takes its values in a countable or continuous set between 0 and 1, 1 being excluded. If  $\kappa$  takes the value one with a positive probability, the upper bound of the set of solutions (2.37) is the straight line

$$k(t) = \frac{X}{\tau} t.$$

In this case we can again calculate  $\bar{k}_n^u$ , but the random fixed point theorem does not hold, and then the preceding asymptotic behaviour is not justified.

Finally we want to underline another way of investigation. We can for instance assume that  $\kappa$  changes its value whenever a fixed quantity of material is available in the box 2 (Fig. 4). If  $m$  is this quantity of material, the sequence  $\{t_n\}$  is defined recursively by

$$m = \int_{t_n}^{t_{n+1}} k(t) dt, \quad t_0 = 0.$$

We shall note only that if the value 1 is excluded,  $k(t)$  is always bounded below and above as we have seen and so  $v_n = t_{n+1} - t_n$ . But if the value 1 is permissible, we have with a positive probability the solution

$$k(t) = \frac{X}{\tau} (t - t_n) + k_n,$$

for arbitrary large values of  $k_n$ . Then the equation

$$m = \int_{t_n}^{t_{n+1}} k(t) dt = \frac{X}{\tau} \frac{(t_{n+1} - t_n)^2}{2} + k_n (t_{n+1} - t_n) = \frac{X}{\tau} \frac{v_n^2}{2} + k_n v_n$$

has always (because of the nature of the physical process) a unique positive solution with the behaviour  $v_n \sim \frac{m}{k_n}$  for large  $k_n$ .

Thus the interval of time between two discontinuities of  $\kappa$  can be as small as we want with, indeed a probability also small, but always positive.

### 3. PROCESS INVENTORY DETERMINATION AND APPLICATION OF TRACER METHODS

The considerations of the preceding chapter are based upon a detailed knowledge of the fabrication process in the fabrication units.

Statements of a control authority, however, concerning the diversion of fissile material from the domain of peaceful use of nuclear energy must be based upon measurable quantities. In the sense of the safeguard system described in [1, 2, 3, 4] the controller should be able to measure these quantities, needed for the detection of a diversion, at strategic points without penetrating into the fabrication units.

#### 3.1 Measuring the output functions at strategic points

The output functions can directly be measured at strategic points. Look at the fabrication plant shown in Fig. 4. Assume that there are 4 strategic points in the plant: one at the entrance (A), one at the exit (B) and two others (C and D) between the fabrication units. These four strategic points allow the measurement of  $X(t)$ ,  $k_1(t)$ ,  $k_2(t)$  and  $\kappa(t)$ . The feedback parameter,  $\kappa$ , being a measurable quantity, its nature (stochastic variable or constant) is no longer of importance for the considerations of this chapter.

The total inventory  $h^{\text{tot}} = h_1 + h_2$  of the plant can be calculated, if  $X(t)$  and  $(1-\kappa)k_2$  are known, by integrating the mass conservation equation

$$(3.1) \quad \frac{dh^{\text{tot}}}{dt} = X - (1-\kappa)k_2.$$

Thus

$$h^{\text{tot}}(t) = \int_0^t [X - (1-\kappa)k_2] dt + h^{\text{tot}}(t=0).$$

For this, only the measurements of two strategic points (A and B) are needed. From the flow measurements at the strategic points, C and D, we get a more detailed information about the inventory function: we are enabled to calculate the inventory  $h_1(t)$  and  $h_2(t)$  of the two fabrication units from the measured quantities  $X$ ,  $\kappa$ ,  $k_1$  and  $k_2$ . The inevitable inaccuracy of the flow rate measurement leads to an inaccuracy of the calculated inventory functions. The propagation of this error can be deduced from the mass conservation equations.

### 3.2 Measuring the inventory function at strategic points

The inventory function,  $h(t)$ , of a fabrication unit or a plant can be measured for discrete time points,  $t_i$ , at strategic points in two different ways:

- the input into the unit (the plant) is stopped at  $t_i$ . The unit is emptied and the outcoming flow is measured, which gives  $h(t_i)$ . This operation interrupts the fabrication process and thus causes undesirable costs.
- the input into the unit (the plant) is traced in a suitable way, beginning at  $t_i$ . The outcoming untraced amount of material after  $t_i$  is measured, which gives  $h(t_i)$ . In this way, the fabrication process has not to be interrupted.

In mathematical terms, this means, that we introduce into the system (fabrication unit or plant) a signal in form of a step function and analyse the system response (the outcoming flow). This is represented in Fig. 18. The input is traced beginning at  $t_0$ . The traced material appears in the output after a certain time, in general with a very small concentration in the beginning, as indicated in the part b of Fig. 18. The indicated area then gives the value  $h(t_0)$  of the inventory function at  $t_0$ .

Both methods of measuring the inventory function at strategic points, indicated in this section, are affected with an inevitable inaccuracy. Concerning the tracer method of inventory measuring, great physical and experimental work has still to be done to determine the nature of tracers to be utilized (radioactive isotopes, fuel with a significantly shifted isotopic vector, etc.), their concentration in the input needed for their detection in the exit stream, and the accuracy of this method.

### 3.3 Measuring the residence time function at strategic points

The residence time function,  $T(t)$ , can be measured for discrete time points,  $t_i$ , at strategic points by using a tracer method similar to the preceding section. This is a wellknown procedure in industry and technics [5].

Fig. 19 shows the principle of this method: at the time  $t_0$ , a delta function signal is fed into the system. The traced material appears in the output after a certain time, beginning in general with a very small concentration, as indicated in the part b of Fig. 19. Different atoms of the tracer material have different residence times. They come out of the system at different times between  $t_1$  and  $t_2$ . In the section 2.1.2 we found a simple relationship between the inventory function, the output function and the residence time function (2.31), for the case of a fabrication unit in steady state operation and without mixing the fuel in the interior of the fabrication unit.

The broadening of the tracer signal, as shown in Fig. 19, is due to a mixing of the particles in the interior of the fabrication unit.

We are now considering the simplified case of a fabrication unit in steady state operation (see section 2.1.1), as shown in Fig. 20. In order to find an equation analogous to (2.31), we follow the ideas described in [5]. Let  $\bar{t}$  be the nominal mean residence time, defined as

$$(3.2) \quad \bar{t} \equiv \frac{h}{k} .$$

Here  $h$  is the inventory- and  $k$  the output-function of the fabrication unit under consideration. (Both are constant in steady state operation.)

Then the time,  $\theta$ , can be expressed in units of the nominal mean residence time  $\bar{t}$  :

$$(3.3) \quad \theta = \frac{t}{\bar{t}} .$$

Let the internal age distribution function,  $I(\theta)$ , define the fraction of particles (pellets, fuel elements or atoms in the case of a fabrication plant) in the fabrication unit having at any instant ages between  $\theta$  and  $\theta + d\theta$ , to be  $I(\theta)d\theta$ . The age of a particle is the time it has spent in the fabrication unit. The exit age distribution function,  $E(\theta)$ , defines the fraction of particles with ages between  $\theta$  and  $\theta + d\theta$  at the moment of leaving the fabrication unit, as  $E(\theta)d\theta$ . Thus

$$(3.4) \quad \int_0^{\infty} I(\theta) d\theta = 1 \quad \text{and} \quad \int_0^{\infty} E(\theta) d\theta = 1.$$

Let  $F(\theta)$  be the fraction of particles in the exit stream which entered the fabrication unit after  $\theta = 0$ . Then at any time  $\theta > 0$

$$\frac{d}{dt} \int_0^{\theta} I(\theta') d\theta' = k - F(\theta)k.$$

Hence with (3.2) and (3.3)

$$(3.5) \quad I(\theta) + F(\theta) = 1.$$

We also have

$$(3.6) \quad F(\theta) = \int_0^{\theta} E(\theta') d\theta'.$$

From (3.5) and (3.6)

$$(3.7) \quad E(\theta) = - \frac{dI(\theta)}{d\theta}.$$

Now let us calculate the mean residence time,  $\bar{\theta}$ , in reduced time units:

$$\begin{aligned} \bar{\theta} &= \int_0^{\infty} \theta E(\theta) d\theta \\ &= - \int_0^{\infty} \theta \frac{dI}{d\theta} d\theta = - \int_0^{\infty} d(\theta I) + \int_0^{\infty} I d\theta \\ &= 1 - \int_0^{\infty} \theta I d\theta \end{aligned}$$

The term in paranthesis disappears, if  $\lim_{\theta \rightarrow \infty} \overline{\theta I} = 0$ ,

which is always fulfilled in physical systems, as  $I(\theta) = 0$  for  $\theta > \theta_N$ , with a finite  $\theta_N$ .

Thus

$$(3.8) \quad \bar{\theta} = 1,$$

which proves, that  $\bar{t}$  as defined by (3.2) is the real mean residence time of particles in the fabrication unit.

Thus we have the following relationship

$$(3.9) \quad \bar{t} = \frac{h}{k}$$

between the mean residence time,  $\bar{t}$ , the inventory function,  $h$ , and the output function  $k$ .

Now, the exit age distribution,  $E(\theta)$ , can directly be measured by means of a tracer signal in the form of a  $\delta$ -function. The tracer concentration - vs. - time curve measured at the exit of the fabrication unit gives the exit age distribution  $E(\theta)$ , as indicated in Fig. 20. The mean residence time,  $\bar{t}$ , can then be obtained as time coordinate of the gravity center of this tracer-concentration-vs.-time curve.

Thus we come to the following conclusion: The residence time function  $T(t)$ , can be measured for discrete time points  $t_i$  at strategic points by using a  $\delta$ -tracer signal. In general, we obtain an exit age distribution function,  $E(\theta, t_i)$ , as indicated in Fig. 19. If the fabrication unit in consideration works in steady state, then the exit age distribution  $E(\theta)$ , is independent of the time point  $t_i$ , as indicated in Fig. 20.

If there is no mixing of the particles in the interior of the unit, then  $E(\theta)$  will be a delta-response. If there is a mixing  $E(\theta)$  will have a form similar to Fig. 20.

The time coordinate of the gravity center of the exit age distribution function,  $E(\theta)$ , gives us the mean residence time,  $\bar{t}$ . Then (in steady state operation) the very simple equation (3.9) holds between the mean residence time, the inventory and the output function.

### 3.4 Different methods to determine the inventory function at strategic points

In the preceding sections we have seen that there are different approaches to determine the inventory function,  $h$ , of a fabrication unit, without penetration into the unit and only measuring at strategic points:

1. Calculation of the inventory function,  $h(t)$ , out of the directly measurable output functions (Section 3.1).
2. Measuring of the inventory function,  $h(t)$ , for discrete time points  $t_i$ , by emptying the fabrication unit and measuring the outcoming flow (Section 3.2).
3. Measuring the inventory function,  $h(t)$ , for discrete time points  $t_i$ , by tracing the input into the unit beginning at  $t_i$ , and measuring the outcoming amount of untraced material after  $t_i$  (Section 3.2).
4. Calculation of the inventory function,  $h(t)$ , for discrete time points,  $t_i$ , by analysing the system response to a tracer signal in form of a delta function, as indicated in Sec. 3.3. If the system operates in steady state, then the simple equation (3.9) can be used to calculate the inventory out of the measured quantities  $\bar{t}$  and  $k$  (mean residence time and output function).

Each of these methods leads to an inaccuracy in the knowledge of the inventory function, as the measurements can always be performed only with a finite accuracy.

Only the first method allows the calculation of the inventory function for all time values. The other methods allow the determination of the inventory function for discrete time points  $t_i$ . But the distance between the time points  $t_i$  can be made very small, by using periodic tracer signals e.g., so that this methods give a good picture of the time development of the inventory function.

### 3.5 Statements of a control authority about the process inventory of a plant

There is a fundamental difference between the values of the process inventory, determined by the four methods indicated in the preceding section. With the first method, a nominal value of the inventory is determined, because this

method is based on the assumption of mass conservation in the fabrication unit. Thus if there has been a diversion of fuel, this does not appear in the result of this method. On the contrary, the other three methods give the real value of the inventory, and thus a diversion of fuel in the fabrication unit does affect the result of this method.

Statements of a control authority about a diversion of fissile material from the domain of peaceful use of nuclear energy, arise from a comparison of the values of the inventory function,  $h(t)$ , which he obtains by the second, third or fourth method indicated in the preceding section, with the nominal value of the process inventory, determined by the first method.

Imagine that an inspector measures the material flow at the strategic points and calculates the inventory function by means of the first method (Sec. 3.1). From time to time he performs a tracer experiment (the third or the fourth method of the preceding section). The resulting values of the inventory function can be compared with the result of the first method and thus statements about a diversion of material can be made, as it has been described in [1].

In this way, the inventory of a plant can be surveyed during the period between two natural wash-outs, which are performed from time to time to clean the fabrication lines.

### 3.6 A simple example of detection of a diversion without physical entering of the plant

Now we want to apply the general considerations developed in 2.2.1, to a simple example of detection of a diversion in a fabrication plant on the basis of the input  $X$  and the output  $\omega$ . The basic idea is to reveal the differences between the behaviours of  $X$  when there is diversion and when there is no diversion. To this end, we consider a plant running at variable capacities. For simplifying the calculations we assume that  $N = 2$ ,  $\xi = \frac{1}{2}$ . This means that from the storage facility we can take only one pellet for each operation. The stationary condition is

$$qN - \xi N < 0, \text{ or}$$

$$(3.10) \quad p = p_1 p_2 p_3 > 1 - \xi = \frac{1}{2}.$$

Writing  $F_k$ ,  $g_m$  for  $F(k)$  and  $g(m)$ , the equation (2.35) becomes

$$\left\{ \begin{array}{l} F_0 = g_1 F_0 + g_0 F_1 \\ F_1 = g_2 F_0 + g_1 F_1 + g_0 F_2 \\ \dots \\ F_n = g_2 F_{n-1} + g_1 F_n + g_0 F_{n+1}. \end{array} \right.$$

Let  $f_n$  be the frequency function, then from  $F_n = \sum_{k=0}^n f_k$ , we obtain after normalization:

$$(3.11) \quad f_n = \frac{p-q}{p^2} \left( \frac{q}{p} \right)^n = \Pr \{ Z = n \}.$$

Now assume that a mass  $\Delta \cdot N$  is diverted from the storage facility each time when this is possible. We suppose here that  $\Delta \cdot N$  is less than one pellet for sake of simplicity; but this is not at all necessary. Then, the running of the plant corresponds to the preceding one but now with

$$\xi' = \xi + \Delta = \frac{1}{2} + \Delta.$$

Consequently, the stationarity condition (3.10) holds for the same  $p$  value. The storage facility instead of containing  $0, 1, 2, \dots, n, \dots$  pellets, with the respective probabilities  $f_0, f_1, f_2, \dots, f_n, \dots$  can contain now  $0, 1-\Delta, 2-\Delta, \dots, n-\Delta, \dots$  corresponding masses of pellets but with the same probability  $f_0, f_1, f_2, \dots, f_n$ . This last fact is essentially due to the preceding assumption:  $\Delta \cdot N$  is less than one pellet. But

$$Z_N = \sum_{i=1}^N X_i - \sum_{i=1}^N \omega_i.$$

For large  $N$ , the process is approximatively stationary, and the mean value  $\bar{Z}$  of the inventory in the storage facility is given by

$$(3.12) \quad \bar{Z} = \lim_{N \rightarrow \infty} \frac{1}{N} \sum_{i=1}^N (X_i - \omega_i)$$

If there is no diversion  $\bar{Z}$  must be

$$(3.13) \quad \bar{Z} = \sum_{n=1}^{\infty} n f_n = \frac{q^2}{p-q} .$$

If there is diversion  $\bar{Z}$  becomes

$$(3.14) \quad \bar{Z}' = \sum_{n=1}^{\infty} (n-\Delta) f_n = \bar{Z} - \Delta (1-f_0) = \frac{q^2}{p-q} - \Delta \left(1 - \frac{p-q}{p^2}\right) .$$

On the basis of the sequence of random variables  $\left\{ \omega_i \right\}$ ,  $i = 1, 2, \dots$  we can estimate  $q = 1-p$ , because of the equality  $\lim_{k \rightarrow \infty} \frac{1}{k} \sum_{i=1}^k \omega_i = pN$ .

The estimation of  $p$  by means of (3.13) must be statistically equivalent. Otherwise the calculation of  $\bar{Z}'$  gives an estimation of  $\Delta$  by (3.14).

In a forthcoming paper we shall describe how such a detection of diversion can be rapidly performed by a tracer signal.

#### 4. APPLICATION TO ALKEM - A FABRICATION PLANT FOR PLUTONIUM BEARING FUEL ELEMENTS

In order to apply the ideas of the preceding chapters, we consider an existing fabrication plant for plutonium bearing fuel elements, the ALKEM plant. In spite of its relatively small annual throughput (200 kg Pu/a) ALKEM works under completely industrial conditions.

##### 4.1 Fabrication of fuel elements of the oxide type

In the following sections we refer to a special production campaign, the fabrication of fuel elements for the subcritical assembly PLATR, USA. These were fuel elements of the oxide type with a mixture of uranium and plutonium. The pellet weight was

19 g before grinding  
18,2 g after grinding,

and the plutonium concentration in the pellets was 2,03 %. The total amount of plutonium for this campaign was 5035,05 g<sup>x)</sup>.

<sup>x)</sup> Actually, neither the pellet weight nor the plutonium concentration or the total amount of plutonium are known exactly because of measuring errors. For our calculations we take the above values (mean values given by ALKEM).

During the production phase of this campaign, the uranium/plutonium powder was subjected to the following production steps:

- The fuel powder was homogenized in the mixer.
- Green compacts were pressed from the powder.
- The green compacts were sintered.
- The sintered pellets were ground, washed and dried.
- After a statistical quality control and pre-stacking of the pellets columns, these columns were loaded into the can tubes. The can tubes finally were welded.

The glove-box line of ALKEM, in which this production campaign was carried out, is divided into 5 fabrication units for our calculations.

The first fabrication unit contains the fuel store, the reprocessing laboratory for pellets, the ceramic glove-box line up to the box with the press.

The second fabrication unit contains the glove-boxes with the sintering furnaces.

The third fabrication unit contains the grinder, the washing, drying and quality control facilities and the prestacking of the pellet columns.

The fourth fabrication unit contains the whole part of the plant, concerned with can tube fabrication and final product storage.

The fifth fabrication unit contains the waste store.

This corresponds exactly to the third example which we considered in section 2.1.1. Fig. 8 shows the flow of fuel through these five fabrication units. The feed-back parameter,  $\kappa_2$ , of the system shown in Fig. 8, corresponds to the fraction of sintered pellets, which do not fulfil the specifications of the PLATR order. These pellets are returned to the first fabrication unit, where they undergo a reprocessing treatment and then are fed back into the production process. The parameter  $\kappa_3$  in Fig. 8 gives the fraction of fuel leaving the third fabrication unit, which goes to the waste store. This waste stream contains the pellets which are not accepted by the final quality control and the waste from grinding.

This is a rather simplified scheme of the fuel flow through ALKEM. In reality there are streams between the different fabrication units and the analytical laboratories of the plant and many samples were taken to determine the isotopic composition, the chemical composition of the fuel after the different production steps. But all these streams between the main system, as indicated in Fig. 8, and other parts of ALKEM are an order of magnitude smaller than the main stream shown in Fig. 8. Thus we neglect them in order to simplify the mathematical representation.

#### 4.2 The machine characteristics of the 1<sup>st</sup>, 2<sup>nd</sup> and 3<sup>rd</sup> fabrication unit

The flow of fuel through the system of fabrication units shown in Fig. 8 is determined by the output of the first, the second and the third fabrication unit. Thus we have to study the machine characteristics of these fabrication units.

The first fabrication unit contains the mixer and the press. The mixer of ALKEM works batchwise and the press in a quasi continuous manner (80-120 green compacts/hour). The output of the first fabrication unit is determined by the capacity of the press. The minimum amount of fuel needed by the press is the mass of one pellet. At the beginning of the fabrication campaign, all the fuel is contained in the first fabrication unit. But inspite of this, the press cannot begin to work at once after the beginning of the production campaign. It has to wait for one batch operation of the mixer (about 4 h). Thus we have Fig. 21 describing the machine characteristic of the first fabrication unit.

The second fabrication unit contains two sintering furnaces. Each one can be loaded for a batch-process with 600 pellets, which are sintered during 16 - 18 h. Assuming that the furnaces are loaded not more than once a day, we have a maximum production rate of 1200 pellets per day. Thus we have the machine characteristic shown in Fig. 22 for the second fabrication unit.

The third fabrication unit's output is determined by the speed of pre-stacking of the pellet columns. Since the capacity of the grinder is smaller than the speed of pre-stacking, for a steady state operation with maximum continuous output, we have to take the grinder's throughput as a limit for  $k_3^{\max}$ , which is about 600 pellets/day. (The speed of pre-stacking of the pellets columns is about 20 - 30 columns per day, each column containing 82 pellets for the PLATR order). Thus we have the

machine characteristic shown in Fig. 23 for the third fabrication unit.

#### 4.3 The PLATR production campaign

It should be pointed out that the machine characteristics describe how the machines of the different fabrication units can function. As there is an economic interest for the operator in running his plant at minimum process inventory and maximum throughput, the knowledge of the machine characteristics enables us to calculate the mass flow through the fabrication units and the process inventory.

In order to compare the real values of throughput and process inventory during the PLATR production campaign with the values expected from a knowledge of the machine characteristics, we consider the output functions of the fabrication units during this campaign, as given by the ALKEM operator:

Fig. 24, 25 and 26 show the integrated flow of pellets from the first to the second fabrication unit (Fig. 24), from the second to the third (Fig. 25) and from the third to the fourth fabrication unit (Fig. 26) during the PLATR campaign.

Assuming that the material flow is as indicated in Fig. 8, we can calculate the mean values of the system constants  $\kappa_2$  and  $\kappa_3$  for the PLATR campaign:

$$\kappa_2 = 0,0844; \kappa_3 = 0,0769.$$

$\kappa_3$  contains the loss of weight of the pellets due to the grinding. From the figures 24, 25 and 26 can be seen, that the output functions of the fabrication units were not constant during the PLATR campaign. A linear interpolation indicates - as shown in these figures - that there were two different phases during this campaign. The first one with relative small values of the output functions and the second phase with higher values of the output functions.

The slopes of the linear interpolated curves in Fig. 24, 25 and 26 allow the calculation of the mean values of the output functions for the two phases of the production campaign. We assume, that  $t = 0$  is the beginning

of the production. Then the first fabrication unit's output starts at  $t = 4$  h. The output of the second and third fabrication unit starts at  $t = 28 \frac{1}{2}$  h (4 h mixing,  $7 \frac{1}{2}$  h pressing and 17 h sintering). The residence time of the pellets in the third fabrication unit has been neglected. Thus we obtain the following mean values of the output functions  $k_i$ ,  $i = 1, 2, 3$  of the different fabrication units:

$$k_1(t) = \begin{cases} 104,77 \text{ g Pu/d} & 0,167 \text{ d} \leq t < 16 \text{ d} \\ 181,06 \text{ g Pu/d} & 16 \text{ d} \leq t \leq 34 \text{ d} \end{cases}$$

(4.1)

$$k_2(t) = \begin{cases} 82,89 \text{ g Pu/d} & 1,188 \text{ d} \leq t < 19 \text{ d} \\ 202,45 \text{ g Pu/d} & 19 \text{ d} \leq t \leq 36 \text{ d} \end{cases}$$

$$k_3(t) = \begin{cases} 28,69 \text{ g Pu/d} & 1,188 \text{ d} \leq t < 21 \text{ d} \\ 196,74 \text{ g Pu/d} & 21 \text{ d} \leq t \leq 41 \text{ d} \end{cases}$$

This shows, that during the PLATR campaign the mean values of the output functions varied over a wide range. The variations of the real values (the variation of the slope of the non-interpolated curves in Figs. 24, 25 and 26) were even greater.

Thus we can conclude that the flow of fuel through the fabrication units was determined not by the machine characteristics, but by other influences.

Now let us calculate the process inventory of the different fabrication units during the PLATR campaign. As the system equations were solved already in the second chapter, we only have to insert (4.1) to (2.28) to find the corresponding inventory functions  $h_1$ ,  $h_2$ ,  $h_3$ ,  $h_4$  and  $h_5$  as functions of  $t$ :

$$0 \leq t < 0,167 \text{ d}$$

$$\begin{aligned} h_1 &= 5035,05 \quad \underline{[g \text{ Pu}]} \\ h_2 &= 0 \\ h_3 &= 0 \\ h_4 &= 0 \\ h_5 &= 0 \end{aligned}$$

$$0,167 \leq t < 1,188 \text{ d}$$

$$\begin{aligned} h_1 &= 5035,05 - 104,77 (t-0,167) \\ h_2 &= \quad \quad + 104,77 (t-0,167) \\ h_3 &= 0 \\ h_4 &= 0 \\ h_5 &= 0 \end{aligned}$$

$$1,188 \leq t < 16 \text{ d}$$

$$\begin{aligned} h_1 &= 4928,08 - 97,78 (t-1,188) \\ h_2 &= 106,97 + 21,88 (t-1,188) \\ h_3 &= \quad \quad + 47,21 (t-1,188) \\ h_4 &= \quad \quad + 26,48 (t-1,188) \\ h_5 &= \quad \quad \quad 2,21 (t-1,188) \end{aligned}$$

$$16 \leq t < 19 \text{ d}$$

$$\begin{aligned} h_1 &= 3479,76 - 174,07 (t-16) \\ h_2 &= 431,07 + 98,17 (t-16) \\ h_3 &= 699,27 + 47,21 (t-16) \\ h_4 &= 392,22 + 26,48 (t-16) \\ h_5 &= 32,73 + 2,21 (t-16) \end{aligned}$$

$$19 \leq t < 21 \text{ d}$$

$$\begin{aligned} h_1 &= 2957,55 - 163,98 (t-19) \\ h_2 &= 725,58 - 21,38 (t-19) \\ h_3 &= 840,90 + 156,67 (t-19) \\ h_4 &= 471,66 + 26,48 (t-19) \\ h_5 &= 39,36 + 2,21 (t-19) \end{aligned}$$

$$21 \leq t < 34 \text{ d}$$

$$\begin{aligned} h_1 &= 2629,59 - 163,98 (t-21) \\ h_2 &= 682,82 - 21,38 (t-21) \\ h_3 &= 1154,24 - 11,38 (t-21) \\ h_4 &= 524,62 + 181,61 (t-21) \\ h_5 &= 43,78 + 15,13 (t-21) \end{aligned}$$

$$34 \leq t < 36 \text{ d}$$

$$\begin{aligned} h_1 &= 497,85 + 17,08 (t-34) \\ h_2 &= 404,88 - 202,44 (t-34) \\ h_3 &= 1006,30 - 11,38 (t-34) \\ h_4 &= 2885,55 + 181,61 (t-34) \\ h_5 &= 240,47 + 15,13 (t-34) \end{aligned}$$

$$36 \leq t < 41 \text{ d}$$

$$\begin{aligned} h_1 &= 532,01 \\ h_2 &= 0 \\ h_3 &= 983,54 - 196,73 (t-36) \\ h_4 &= 3248,77 + 181,61 (t-36) \\ h_5 &= 270,73 + 15,13 (t-36) \end{aligned}$$

$$41 \leq t$$

$$\begin{aligned} h_1 &= 532,01 \\ h_2 &= 0 \\ h_3 &= 0 \\ h_4 &= 4156,82 \\ h_5 &= 346,38. \end{aligned}$$

We have represented these functions in Figs. 27, 28, 29 and 30. They are compared with the real process inventory during the production campaign, as calculated directly from the ALKEM data.

At the end of the campaign, 532 g of plutonium remained in the first fabrication unit. About 250 g of this remaining fuel were used in a later period of the production campaign to produce 550 pellets (this is indicated in Figs. 24, 25 and 26). Also during the PLATR campaign, there was an authorized diversion of plutonium (see  $\overline{[6]}$ ), as this campaign was safeguarded in order to apply the ideas developed in  $\overline{[1]}$  to  $\overline{[4]}$ . The quantity of plutonium remaining in the first fabrication unit was due to the insufficient accuracy of our model representation of the fuel stream in ALKEM.

Let us compare the output of the fabrication units, as calculated from the machine characteristics to the real mean values of the output functions:

Output function $\overline{[g \text{ Pu/d}]}$	$k_1$	$k_2$	$k_3$
Calculated from the machine characteristics <sup>x)</sup>	264	330	158
Real value during the first phase of PLATR	104	82	28
Real value during the second phase of PLATR	181	202	196

<sup>x)</sup> Assuming an 8-hour day and a 5-day week

This comparison shows that during this campaign the flow of fissile material through the fabrication plant was not determined only by the machine characteristics.

#### 4.4 Analysis of the isotopic composition of plutonium during a fabrication campaign

During the PLATR production campaign fuel samples were taken, in order to check the isotopic composition of the plutonium after different production steps. The campaign consisted of 12 batch operations and from each batch three samples were taken - one after mixing, one after pressing and one after sintering. These samples were analysed by means of a mass spectrometer. The following table shows the result of this analysis:

No. of the batch	Sample after mixing	Sample after pressing	Sample after sintering
Isotopic composition of the plutonium: Pu 40/Pu 39 (Pu 41/Pu 39)			
1	0,237 (0,0659)	0,235 (0,0637)	0,238 (0,0636)
2	not analysed	0,217 (0,0566)	0,239 (0,0640)
3	0,228 (0,0598)	0,241 (0,0653)	0,239 (0,0642)
4	0,235 (0,0645)	0,233 (0,0632)	0,240 (0,0643)
5	0,233 (0,0641)	0,239 (0,0657)	0,239 (0,0643)
6	0,234 (0,0647)	0,239 (0,0636)	0,239 (0,0645)
7	not analysed	0,238 (0,0638)	0,239 (0,0658)
8	0,233 (0,0632)	0,235 (0,0639)	0,239 (0,0646)
9	0,239 (0,0645)	0,236 (0,0634)	0,237 (0,0644)
10	0,236 (0,0631)	0,237 (0,0632)	0,239 (0,0645)
11	0,238 (0,0656)	0,237 (0,0634)	0,238 (0,0635)
12	0,234 (0,0625)	0,238 (0,0638)	0,239 (0,0644)

Each one of these values is the mean value of several measurements (6 to 16). The relative mean quadratic deviation was in the range 0,1 % to 1 %. We have applied a test for homogeneity of variance, devised by Bartlett (Proc. of Roy.soc., A, 160, 1937).

This test was applied to the rows and columns of the above table. The result was, that there was no significant difference in the isotopic composition, neither between different batches nor between three samples of the same batch.

This is not surprising, because the source material was mixed and homogenized before starting the fabrication process. But it shows that there was no artificial shift of the plutonium vector (by adding another type of Pu e.g.) during the fabrication process.

## 5. CONCLUSIONS

We have seen that there are different methods of determining the process inventory of a fuel element fabrication plant. The use of tracer methods has been discussed. In order to study the efficiency of these different methods of the detection of a diversion, a model system representation of a fabrication plant will be very helpful.

The problem of a diversion of fissile material can be studied in two different ways:

- A restricted mathematical model for a fabrication plant is considered, which is sufficiently precise to solve the diversion problem and then step by step the number of restrictions is diminished to approach the model to real plants.
- A real complex representation of a plant is studied and then it is tried to find out what type of restrictions and modifications must be applied to increase the efficiency of the safeguards system and facilitate the detection of a diversion.

In both cases a simulation of the model on a computer could be very helpful.

With regard to tracer methods, the physical problems - what to use as tracers, what concentration of tracers needed for an accurate detection etc. - should be studied in detail.

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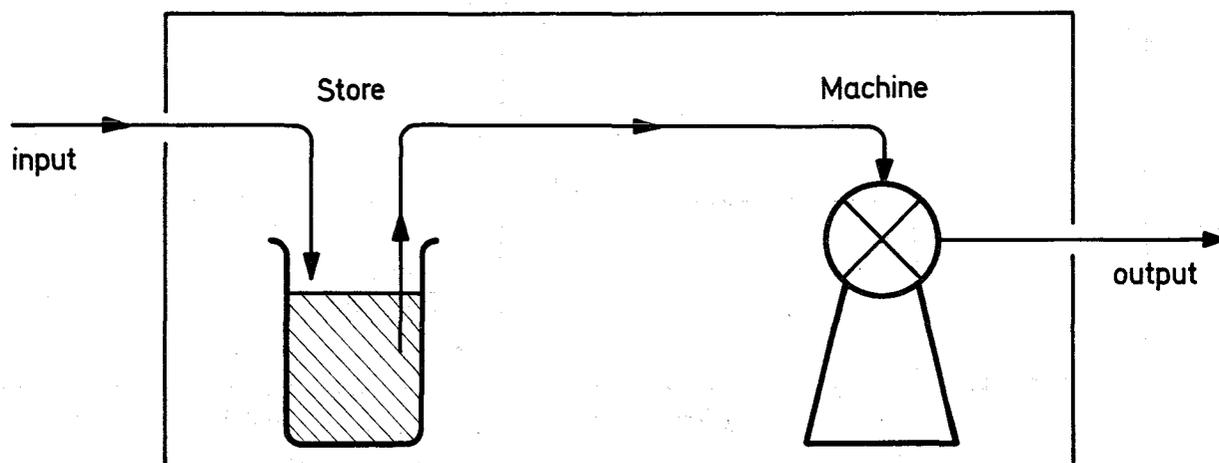


Fig. 1 Schematic representation of a glove box or a fabrication unit

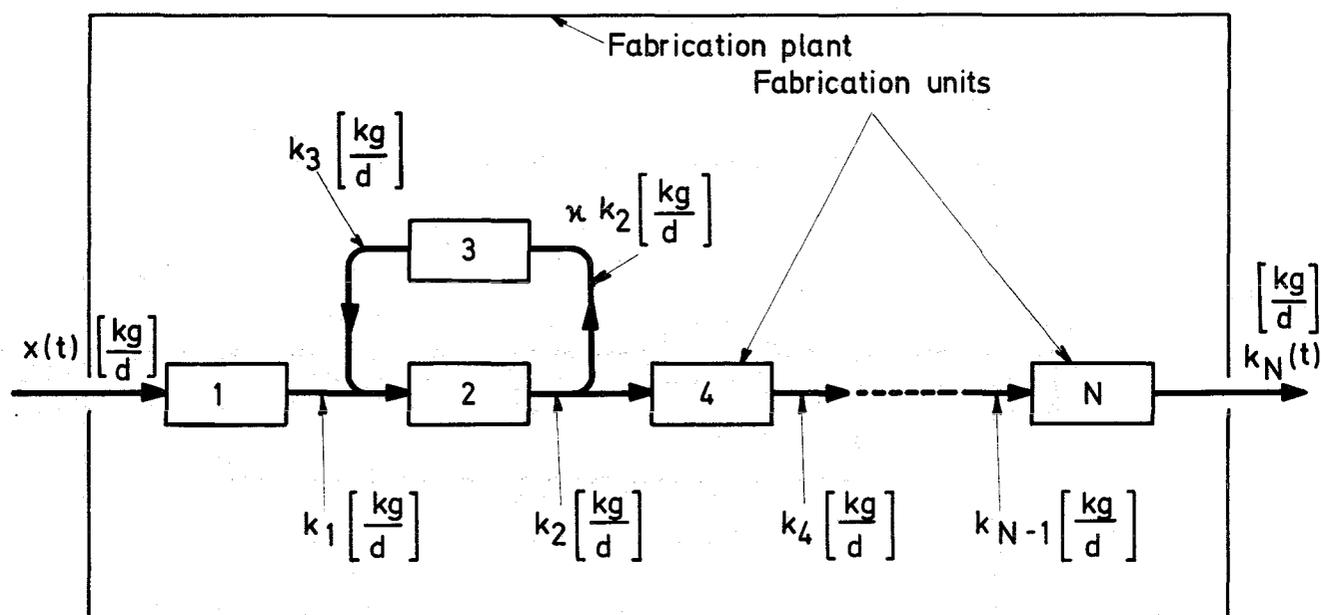
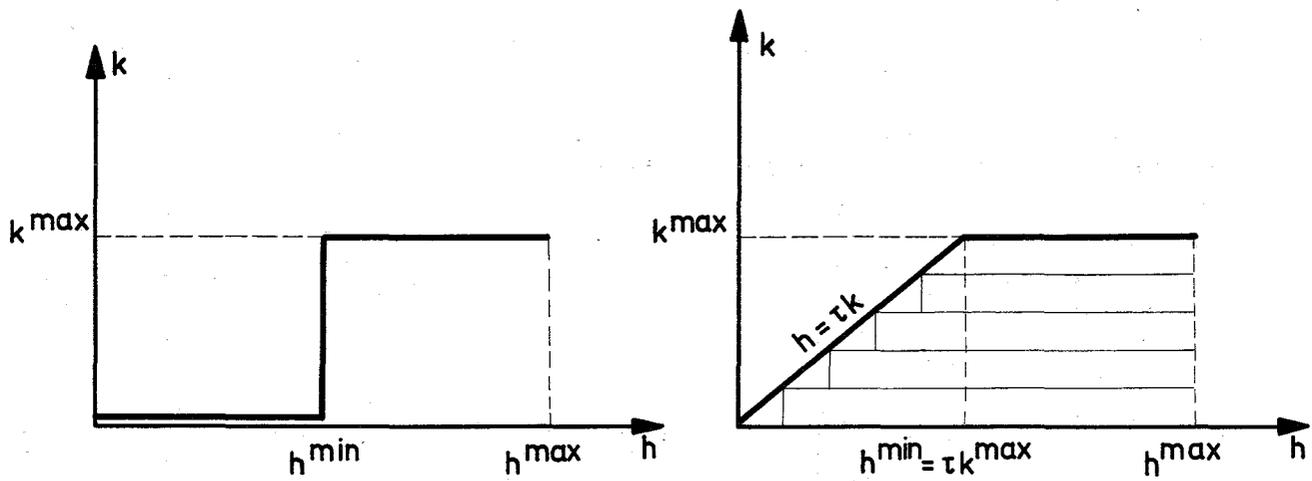


Fig. 2 A fabrication plant, consisting of  $N$  fabrication units



a) constant capacity

b) variable capacity

Fig. 3 Machine characteristics :  
 a) constant capacity machine  
 b) variable capacity machine

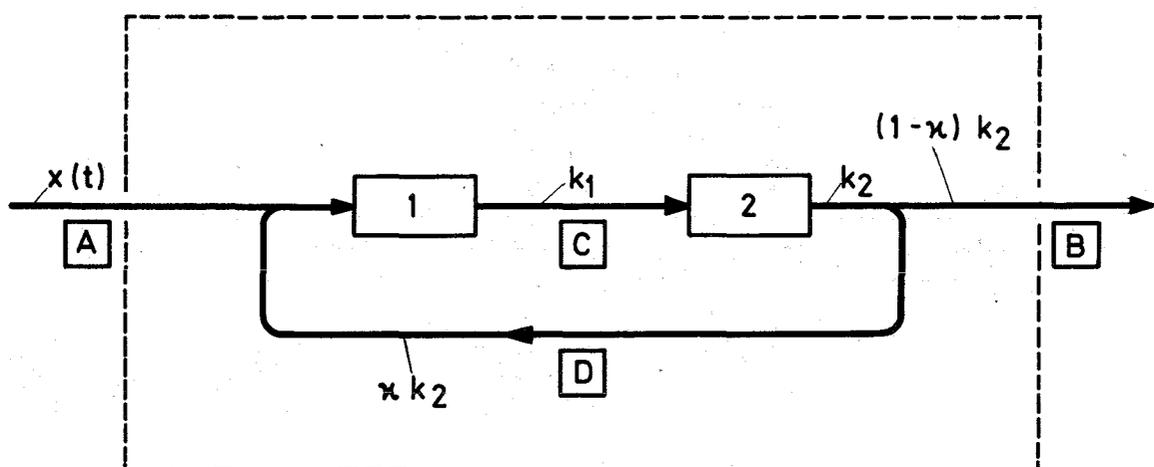


Fig. 4 Two fabrication units with feed back

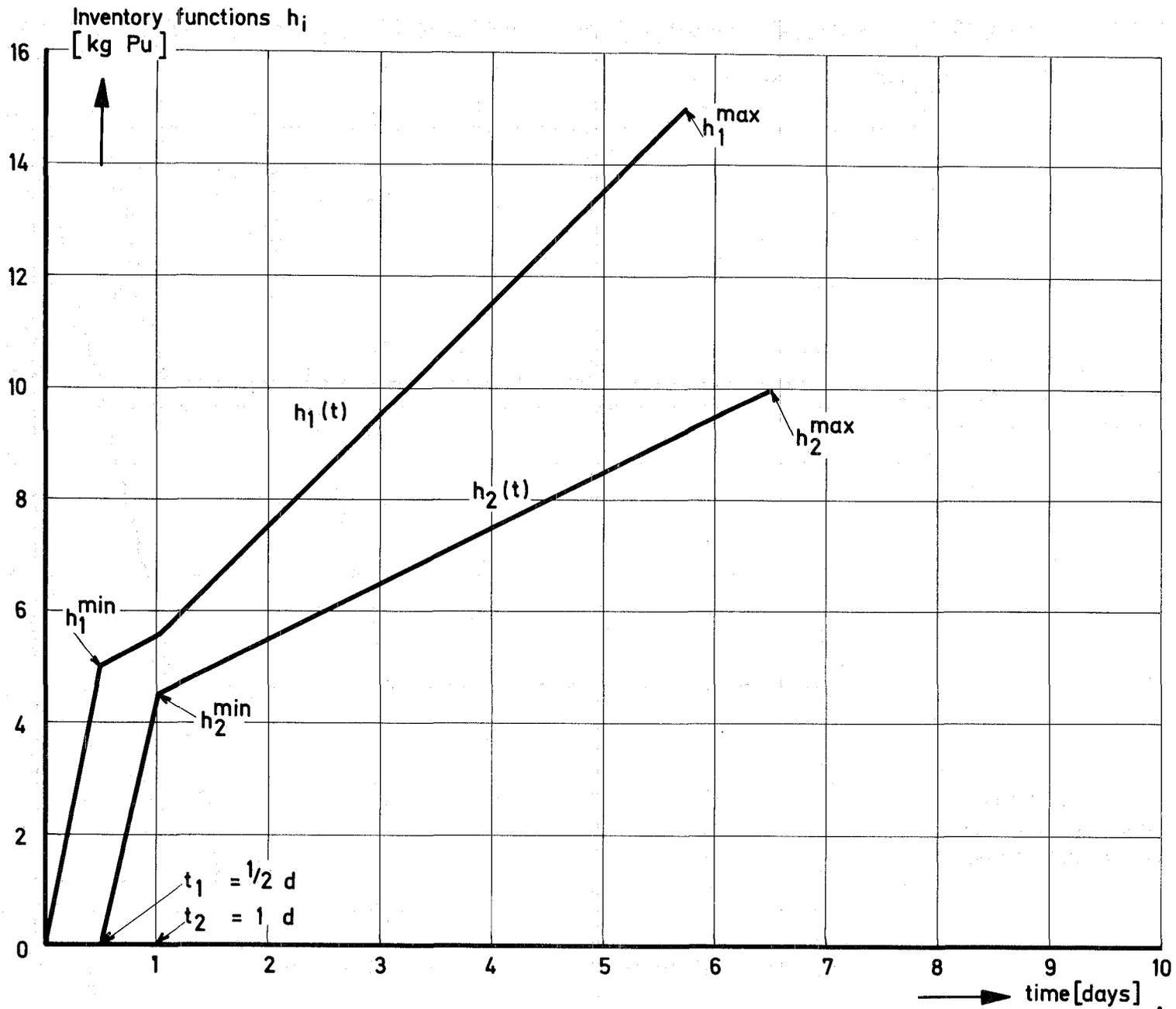


Fig. 5 Time dependence of the inventory function  $h_1$  and  $h_2$  of the 1<sup>st</sup> example

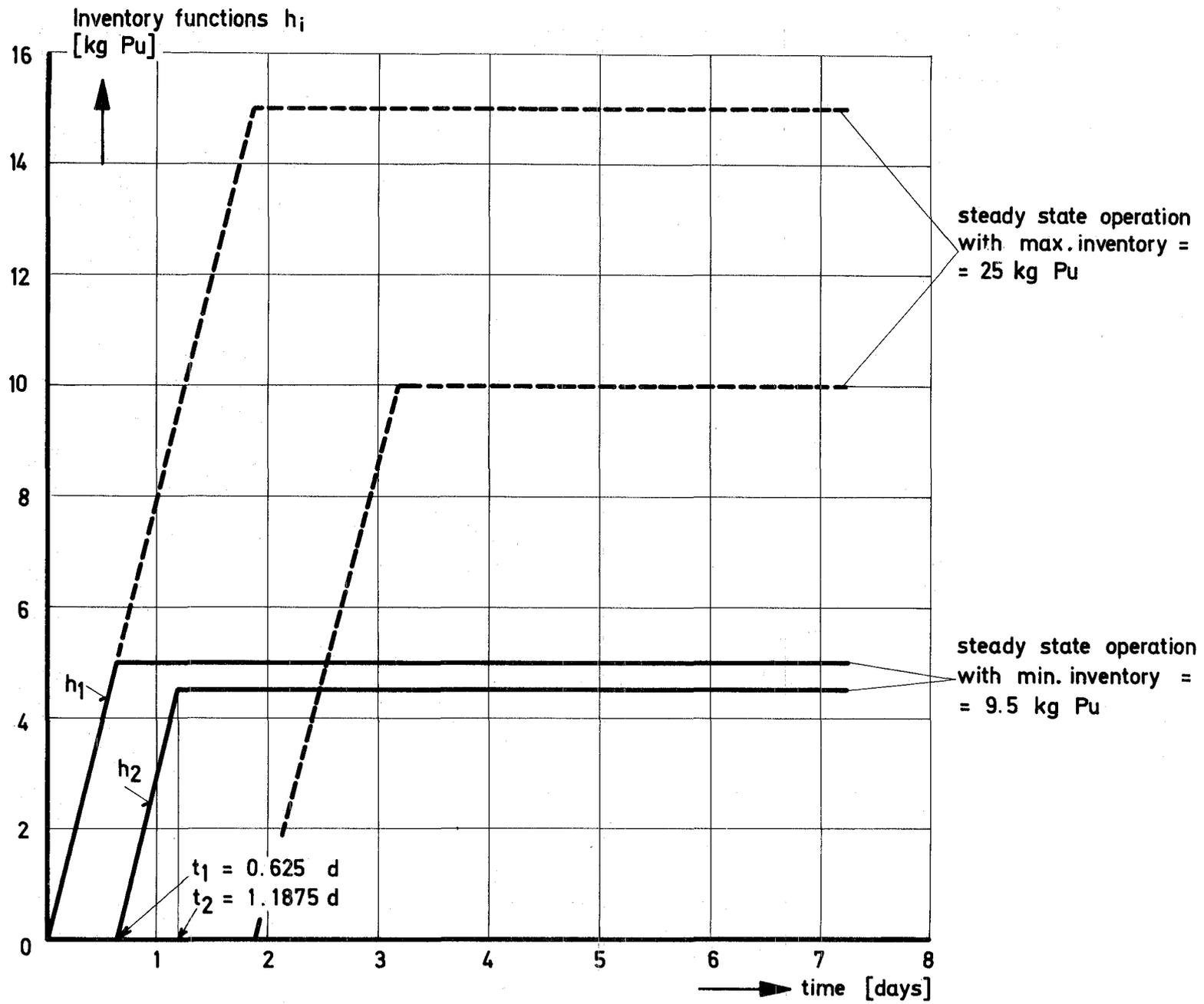


Fig. 6 Steady state operation at different process inventory levels

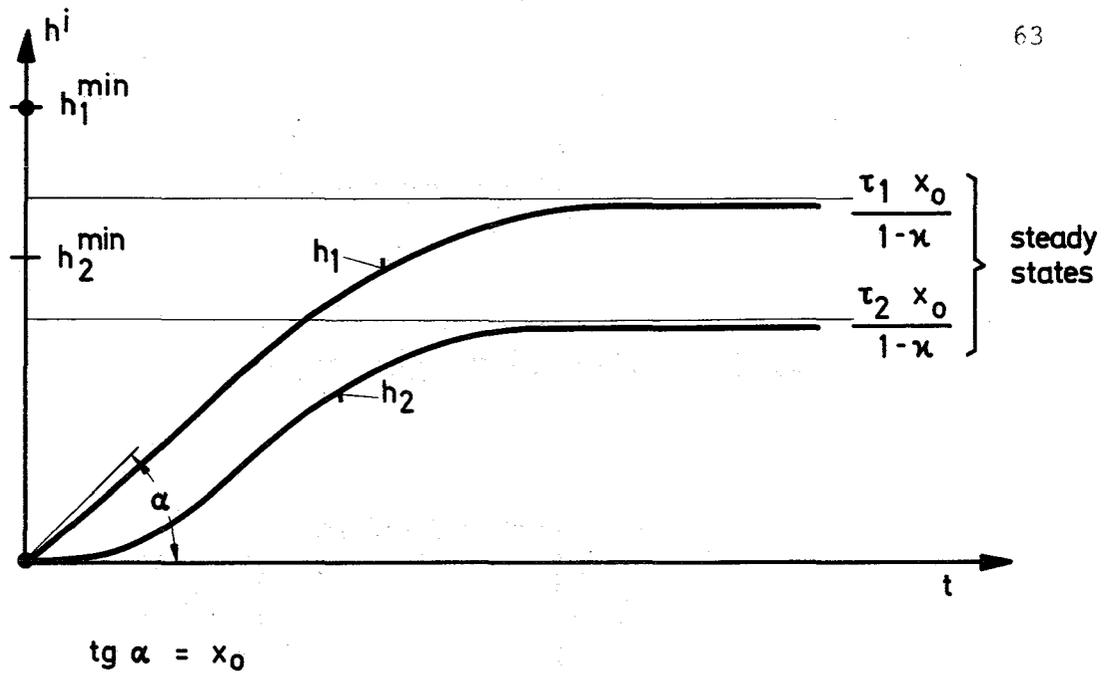


Fig. 7 Time development of the inventory functions  $h_1$  and  $h_2$

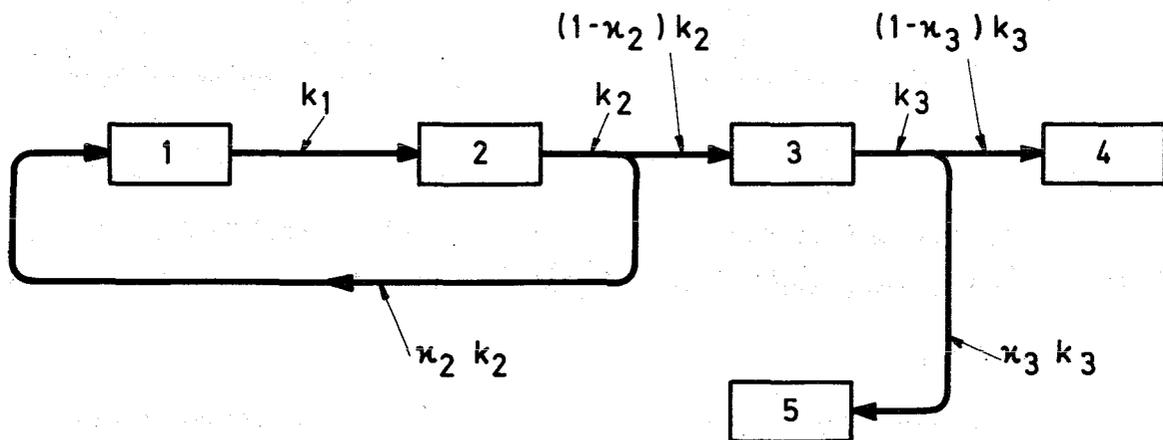


Fig. 8 A fabrication plant consisting of 5 fabrication units

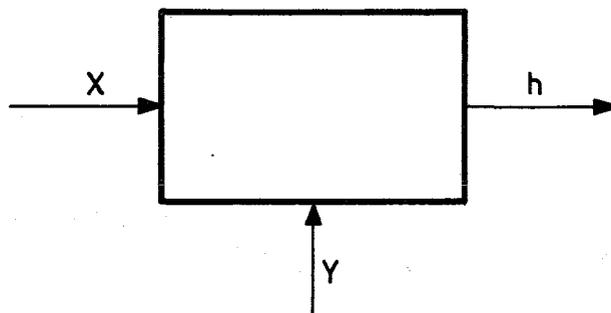


Fig. 9 Glove box with mixer

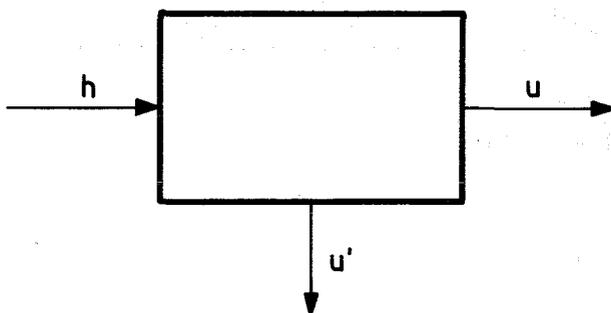
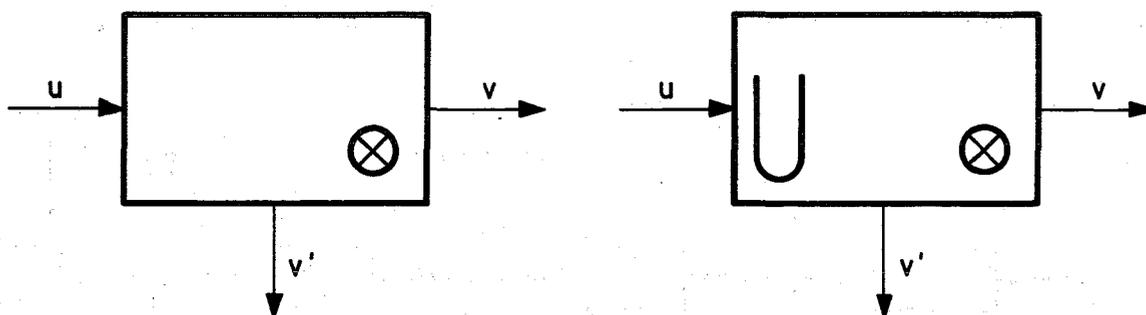


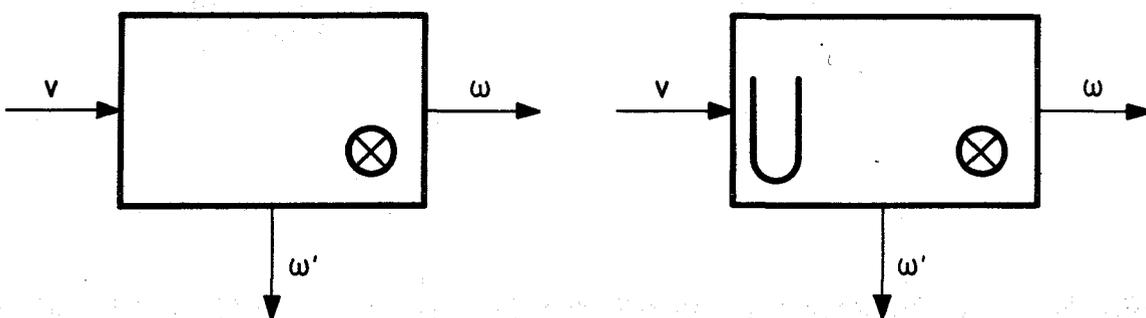
Fig. 10 Glove box with press



a) variable capacity

b) constant capacity

Fig. 11 Glove box with the sintering furnace



a) variable capacity

b) constant capacity

Fig. 12 Glove box with the grinder

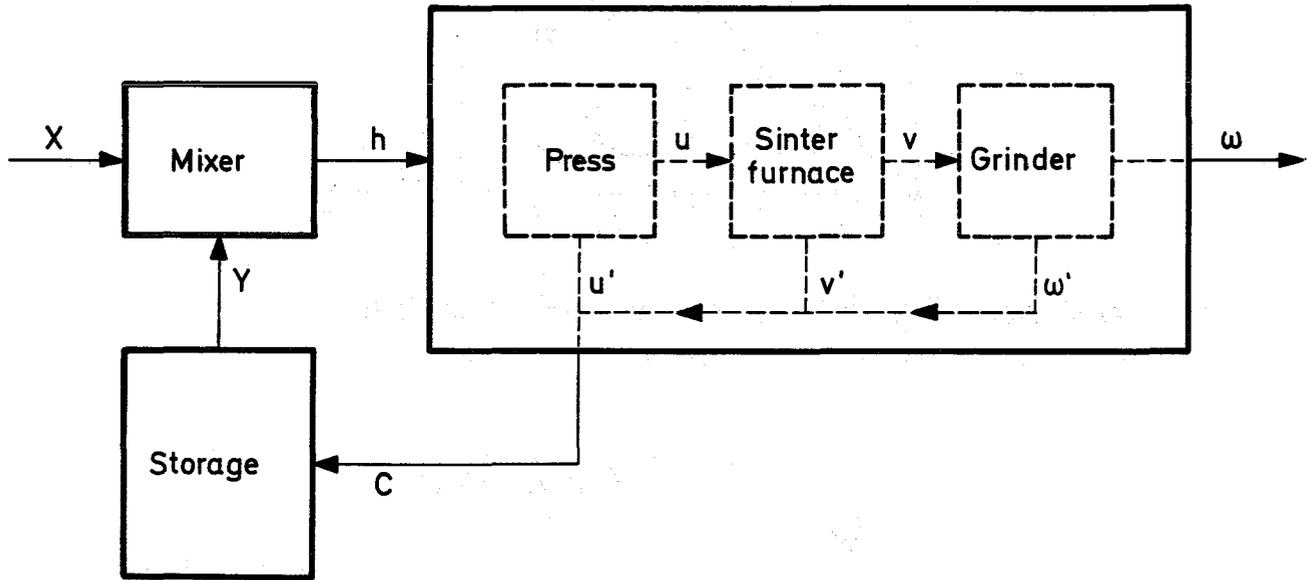


Fig. 13 Complete system with storage facility

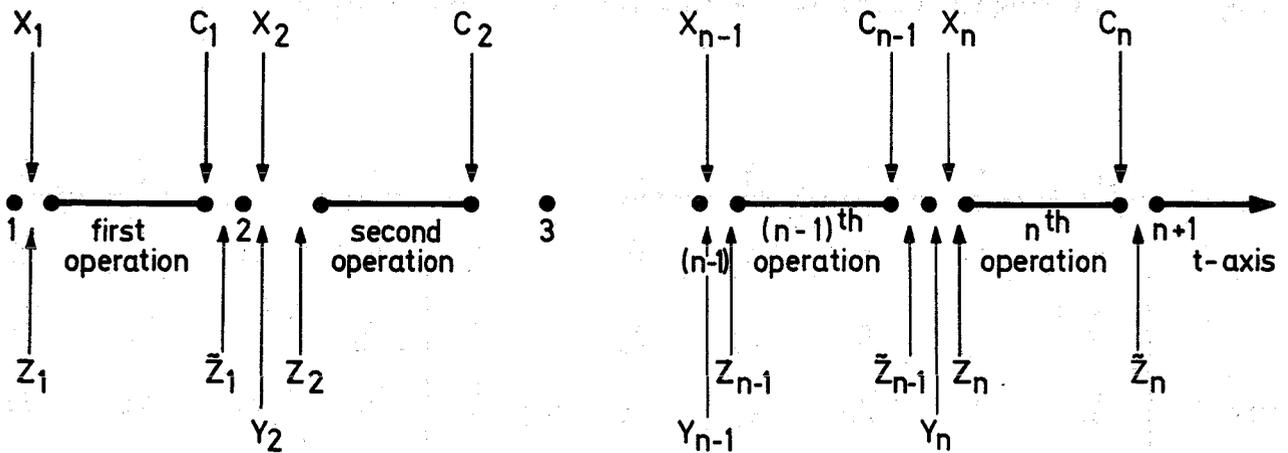


Fig. 14 Notations for different operations



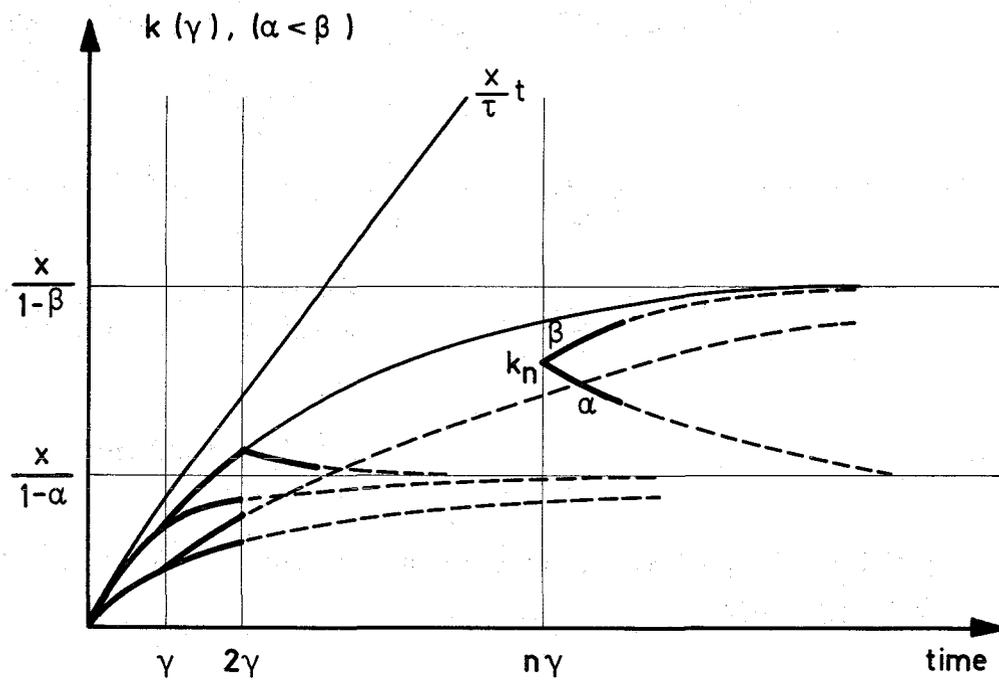
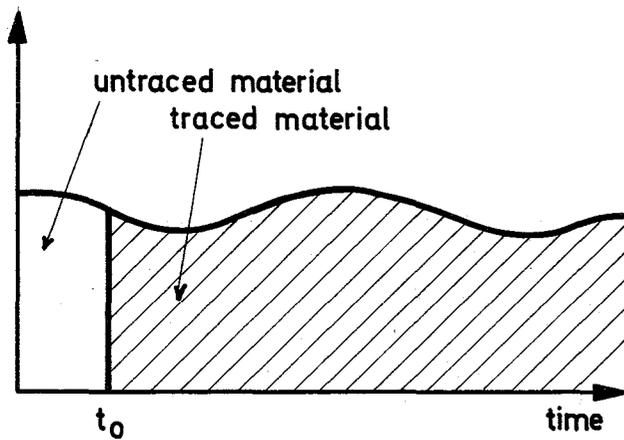
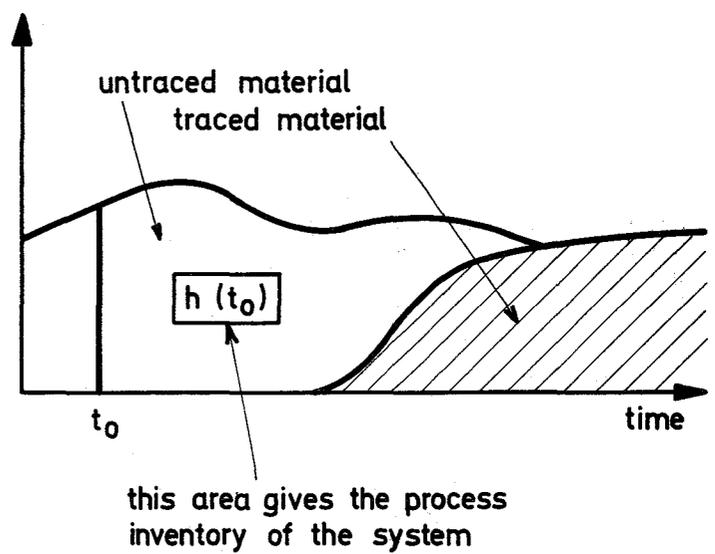


Fig. 17 Set of curves ( 2.37 ) as random solutions of ( 2.36 )

input of the system



output of the system

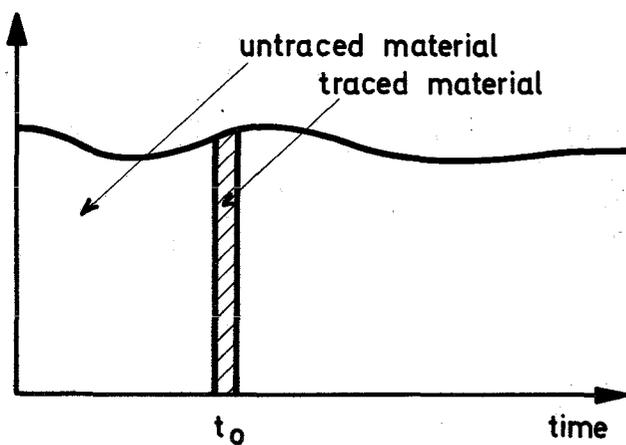


a) Signal

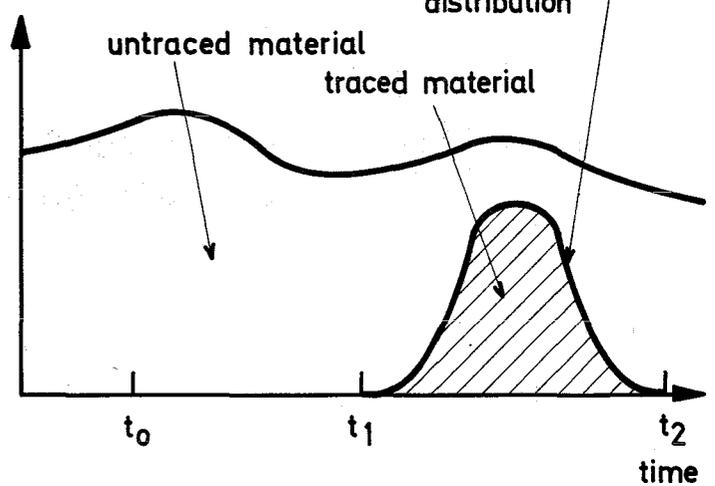
b) System response

Fig. 18 System response to a tracer signal in form of a step function

input of the system



output of the system



a) Signal

b) System response

Fig. 19 System response to a tracer signal in form of a delta function

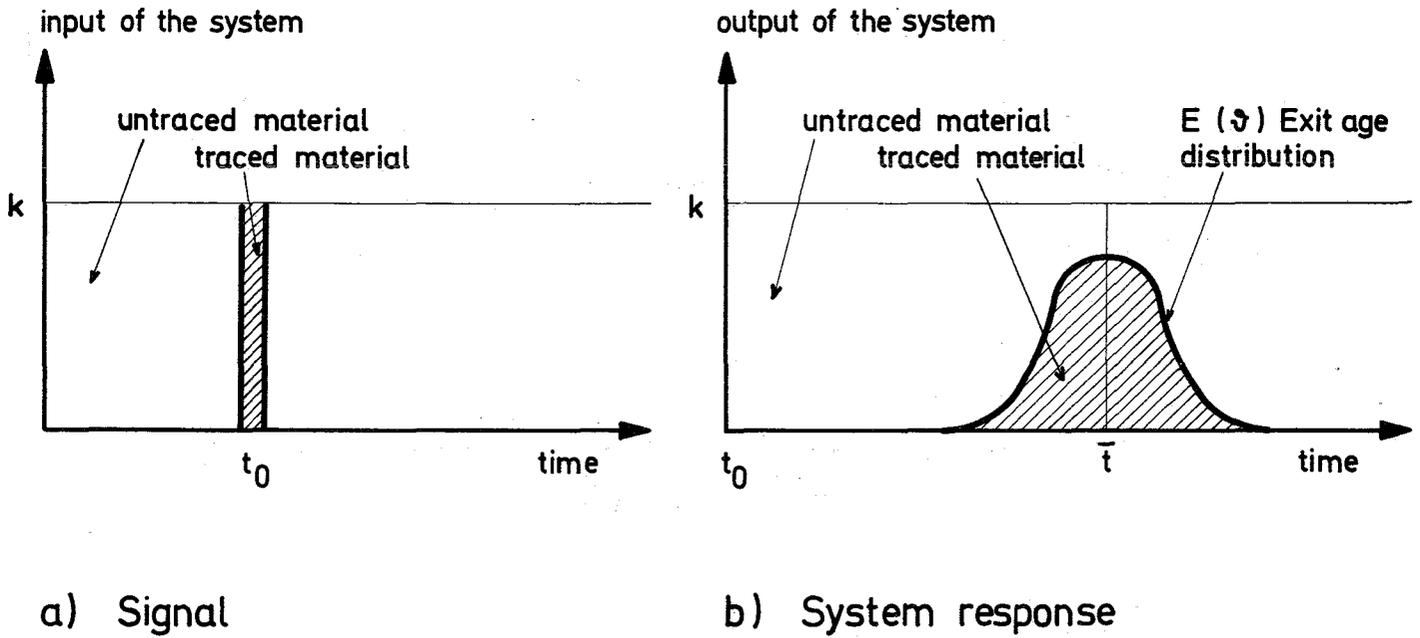


Fig. 20 System response to a tracer signal in steady state operation

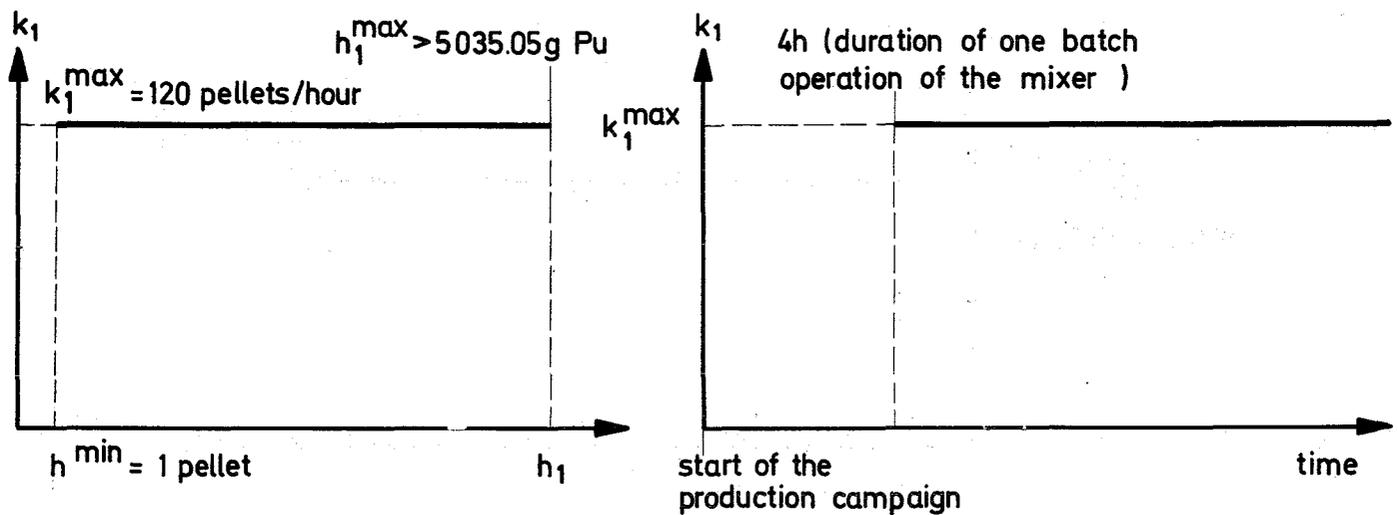


Fig. 21 Machine characteristic of the first fabrication unit



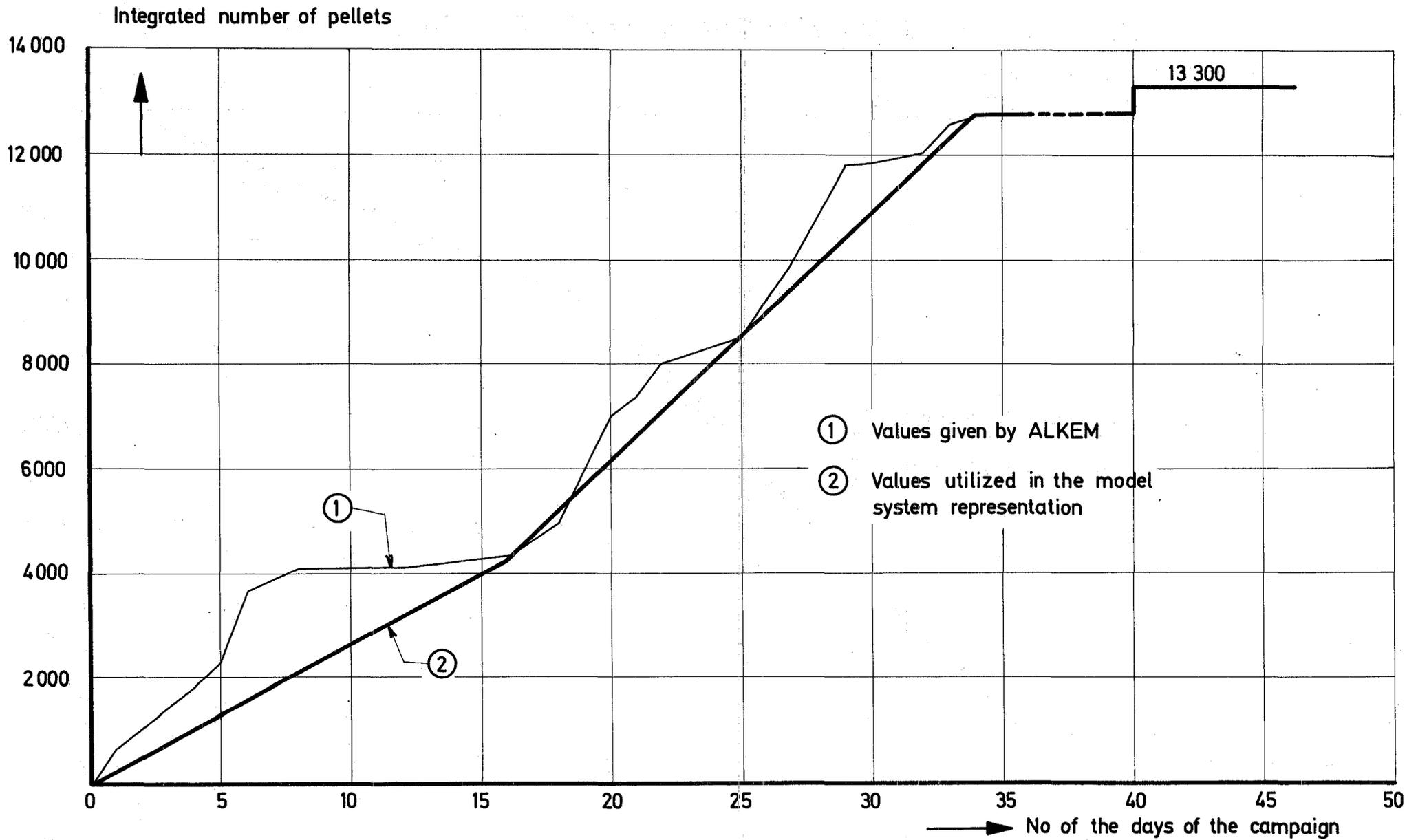


Fig. 24 Integrated flow of pellets from fabrication unit 1 to 2

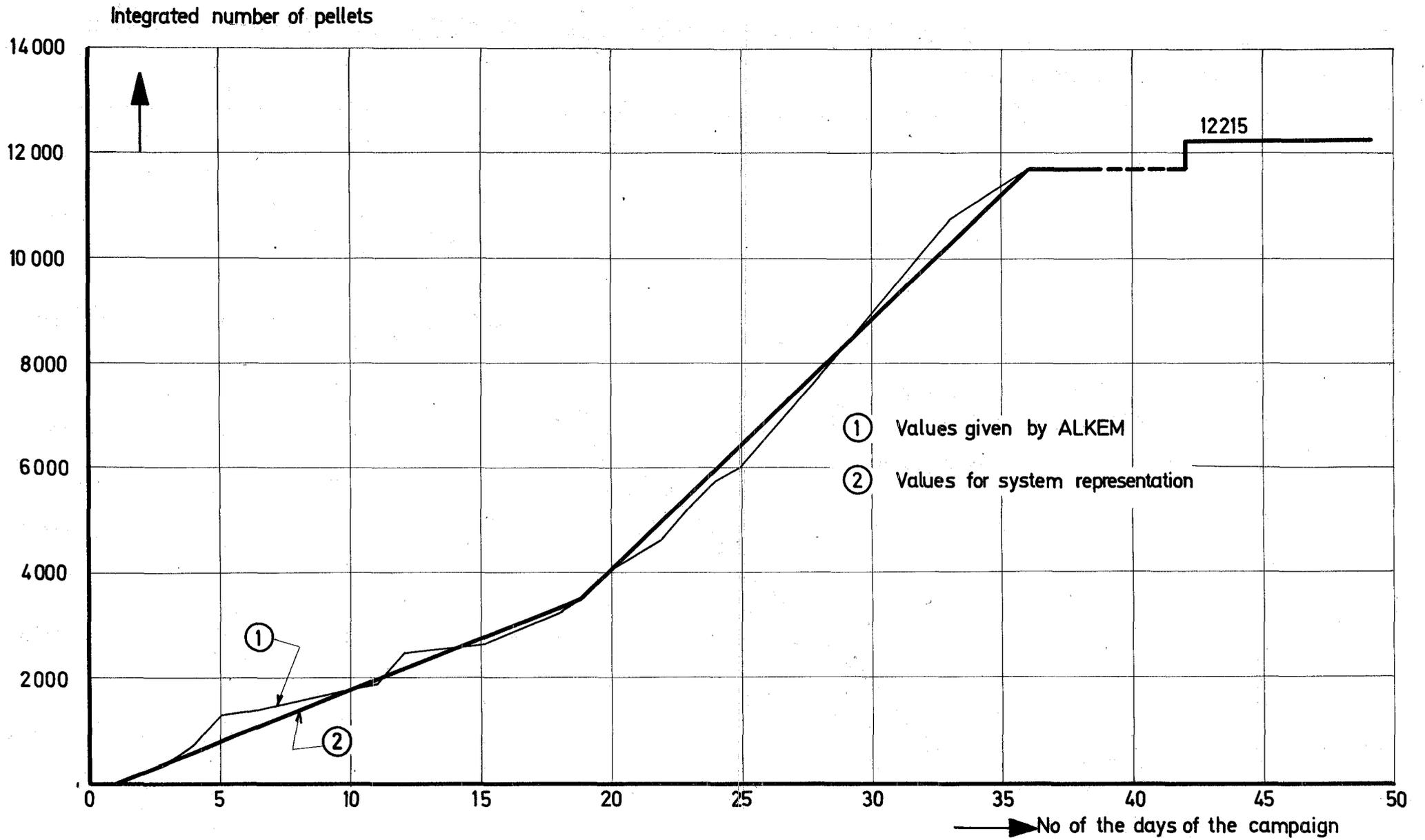


Fig. 25 Integrated flow of pellets from the fabrication unit 2 to 3

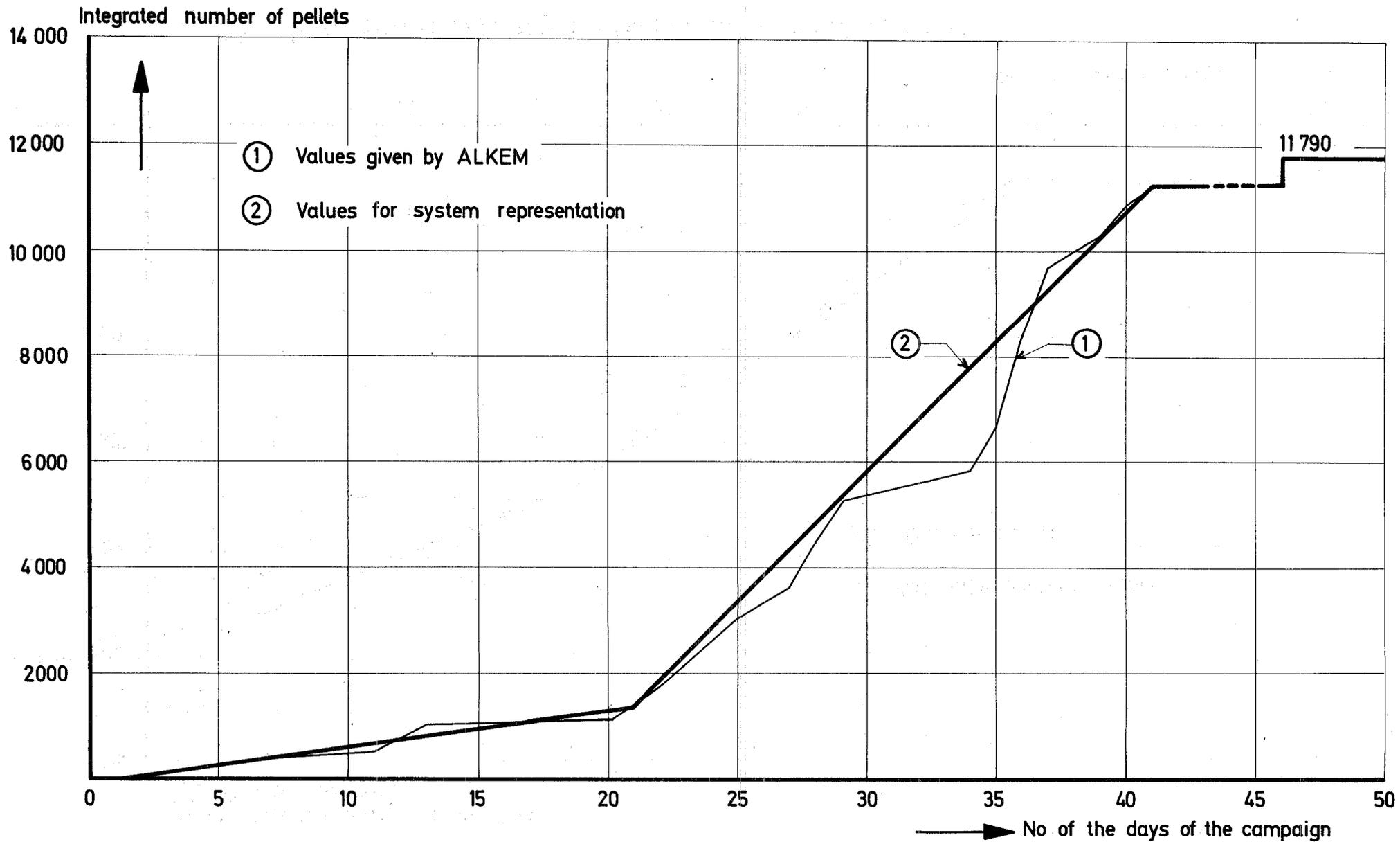


Fig. 26 Integrated flow of pellets from the fabrication unit 3 to 4

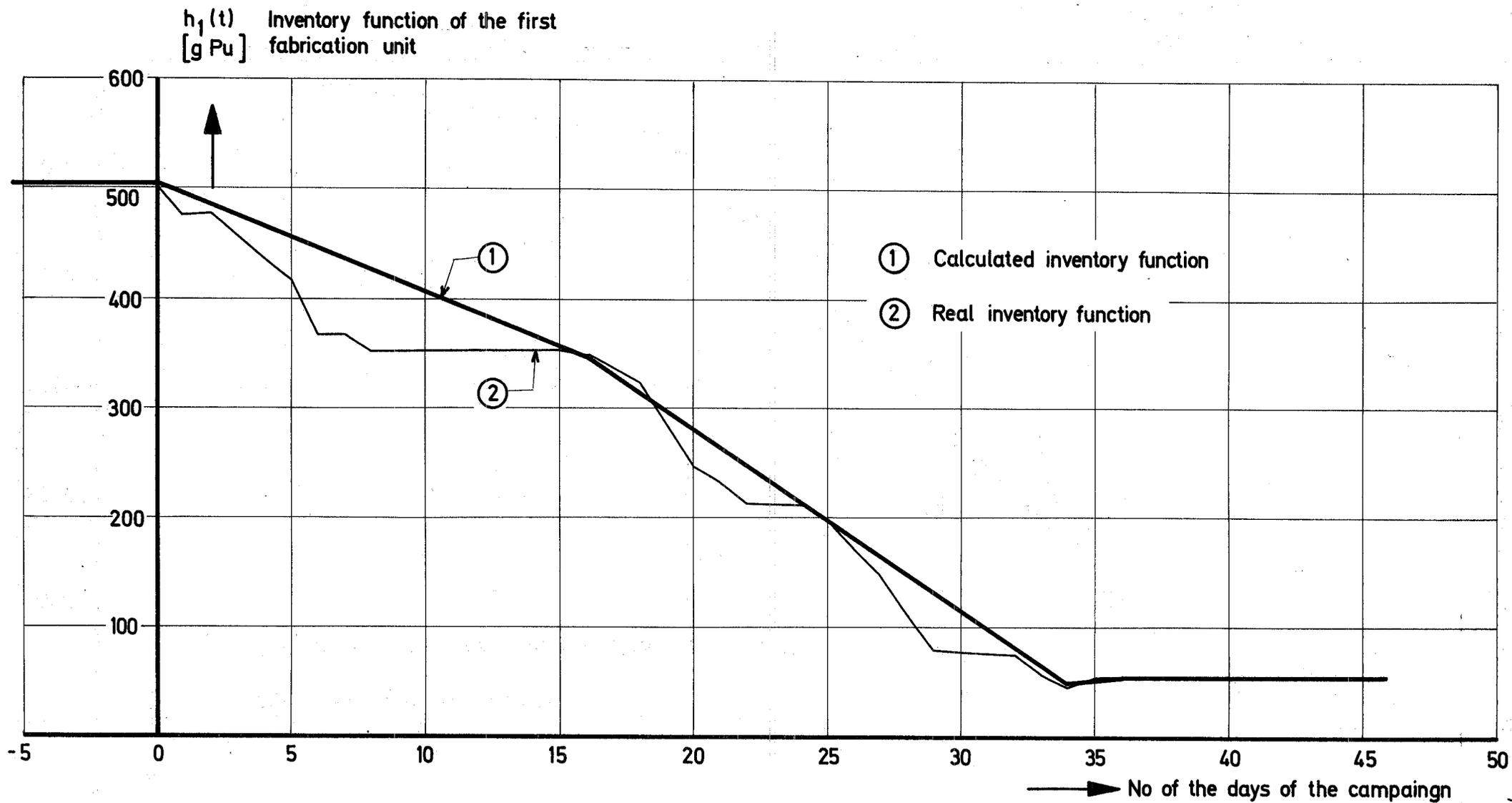


Fig. 27 Process inventory of the first fabrication unit

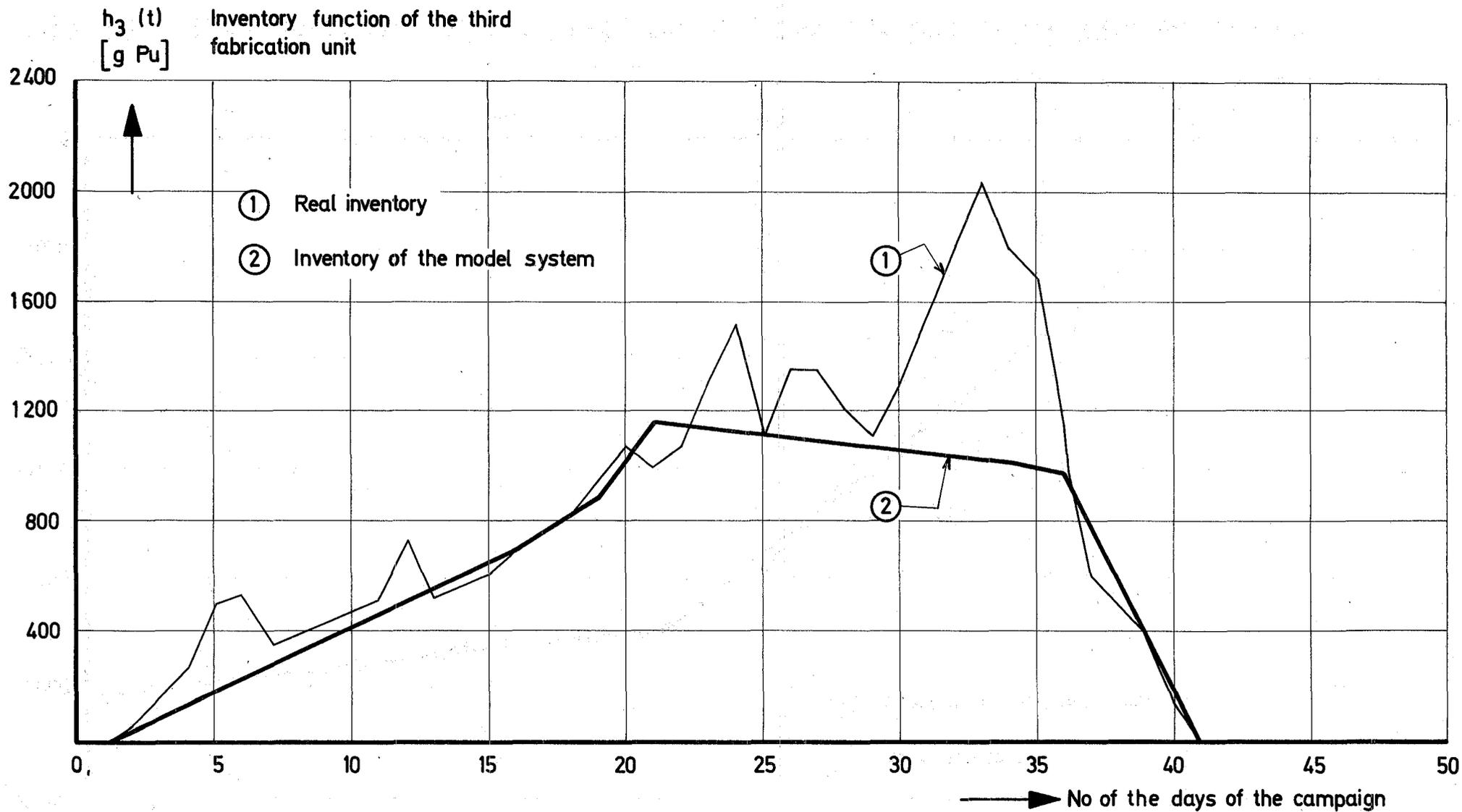


Fig.28 Process inventory of the third fabrication unit

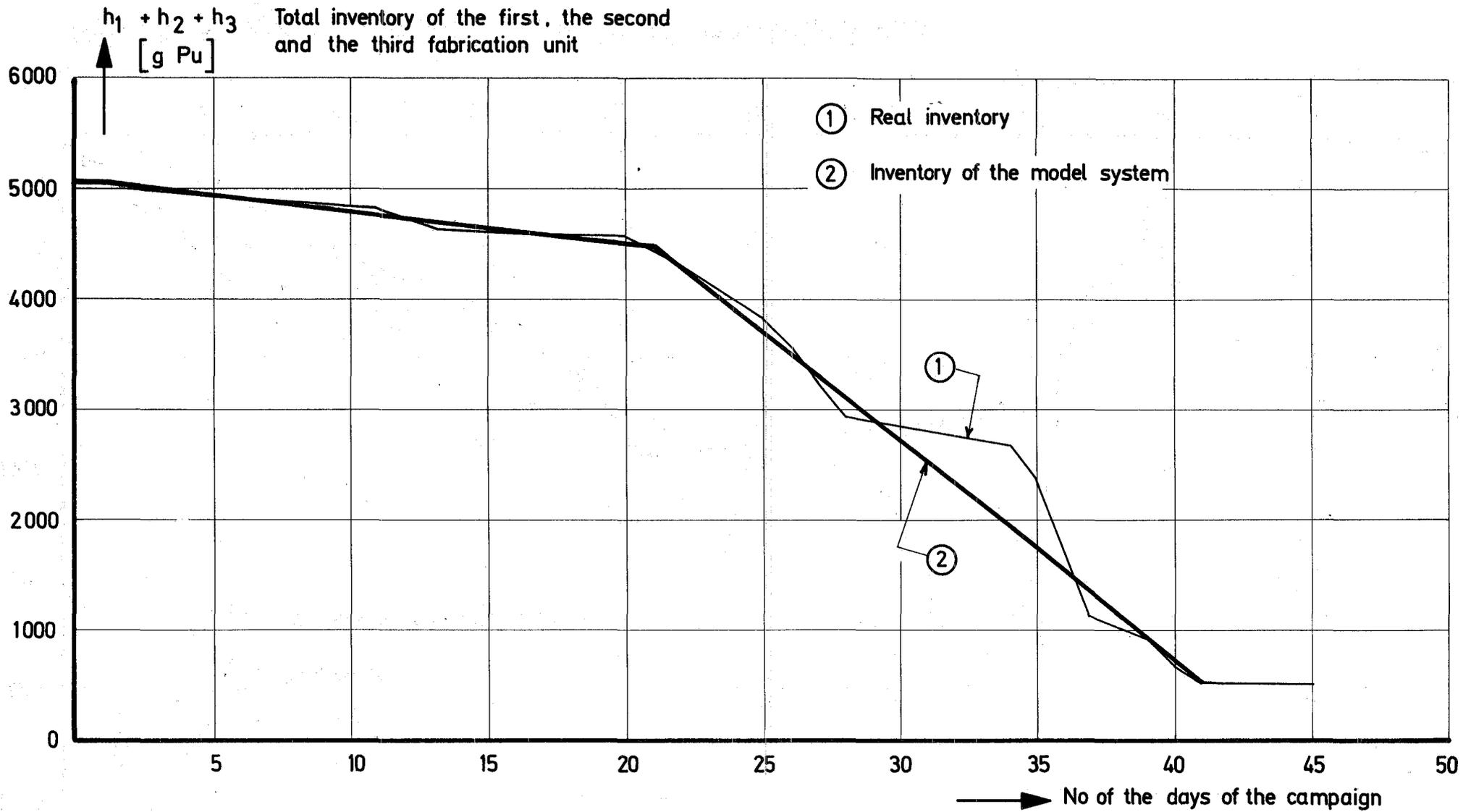


FIG. 29 Total inventory of the first, the second and the third fabrication unit

① Inventory function  $h = h_1 + h_2 + h_3$

② Inventory function  $\tilde{h} = h_4 + h_5$

Ⓐ Calculated inventory

Ⓑ Real inventory

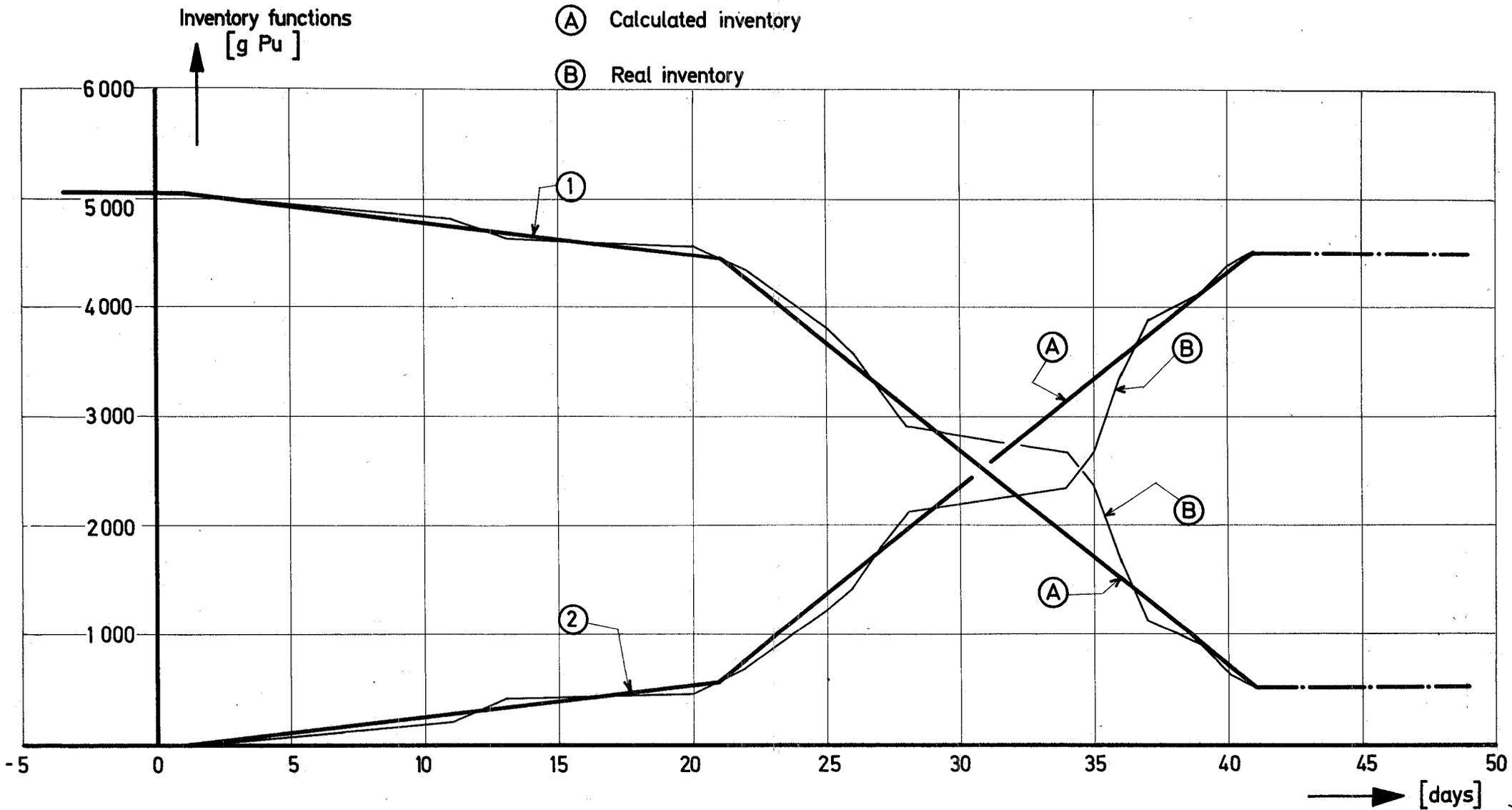


FIG. 30 Behavior of the total inventory as a function of time