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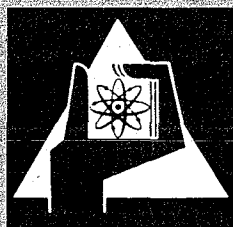
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Projekt Spaltstoffflußkontrolle

**User's Manual for the Application of
the Dynamic Process Inventory Determination
Using Isotopic Step Signals in Reprocessing Plants**

E. Drosselmeyer, R. Kraemer, A. Rota



GESELLSCHAFT FÜR KERNFORSCHUNG M. B. H.

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User's Manual for the Application of
the Dynamic Process Inventory Determination
Using Isotopic Step Signals in Reprocessing Plants

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This paper was prepared in the framework of the joint research program of the association EURATOM/GFK/CEN/CNEN/RCN (contract no. OOI-69-I TDR D) of safeguards and was presented at the IAEA Working Group Meeting on the Use of Isotopic Composition Data in Safeguards, Vienna, 10. - 14. April 1972.

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Abstract

The manual describes the technical feasibility and procedures of a dynamic heavy material process inventory technique which correlates isotopic composition data of subsequent input and product batches in reprocessing plants. This technique was developed for the application in safeguards with the objective to close the nuclear material balance in adequate time intervals during a running campaign.

The manual is divided in 7 chapters covering preprocess information requirements on the material balance area and the spent fuel to be processed, optimisation procedures for the generation of an adequate isotopic stepsignal and the procedures to determine with the help of the actual measured input- and product signal the physical inventory of heavy material. This inventory is compared with the corresponding book inventory by suitable statistical procedures (Monte Carlo technique) in order to obtain the Material Unaccounted For (MUF) and its range of uncertainty. The annexes give a short description of four computer codes for suitable data handling of this inventory technique.

Zusammenfassung

Das Handbuch beschreibt die Maßnahmen und technischen Grenzen einer Methode zur dynamischen Bestimmung des Prozeßinventars von Uran oder Plutonium in Wiederaufarbeitungsanlagen. Diese Methode wurde für die Spaltstoffflußkontrolle entwickelt mit dem Ziel, die Materialbilanz während einer laufenden Aufarbeitungskampagne in geeigneten Zeitintervallen zu schließen.

Das Handbuch ist in 7 Kapitel aufgeteilt, die zunächst die Beurteilung der Materialbilanzzone und des abgebrannten Kernbrennstoffes im Hinblick auf die Größe und Beschaffenheit des notwendigen Isotopensprungsignals behandeln. Danach werden die Maßnahmen zur statistischen Auswertung (Monte-Carlo-Technik) der gemessenen Isotopensignale im Eingangs- und Produktstrom beschrieben, mit dem Ziel, den realen Bestand an Uran bzw. Plutonium zu bestimmen. Über den Vergleich mit dem entsprechenden Buchbestand erhält man quantitativ das nicht nachgewiesene Material (MUF) und dessen Unsicherheitsbereich. Im Anhang werden 4 Rechenprogramme zur statistischen Auswertung von Teilproblemen dieser Inventurtechnik beschrieben.

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1. General

1.1 Introduction

This manual was prepared after considerable theoretical and experimental work /1,2,3/ on the validity of a new inventory determination technique which employs the correlation of isotopic data of subsequent input and output batches. This new technique described here had a number of different names in the past some of which are:

- a) Dynamic Inventory Determination in order to express the fact that this technique does not need static conditions during inventory taking, but may be performed during a running campaign,
- b) In-Process Inventory Determination in order to emphasize the point that this technique is applicable only in the process area and not in the storage area of a reprocessing plant because it is based on nuclear material flow measurements,
- c) Self Tracing Technique, because the step signal used for the evaluation of the inventory is the inherent isotopic composition of the heavy material and not an artificial tracer.
- d) Finally this technique was simply called Physical Inventory Technique in order to demonstrate the fact that this technique represents an independent inventory measurement which can be compared with the book inventory determination in order to make a statement on MUF. Therefore this independent inventory determination meets the specifications defined in the model agreement /4/ paragraph 113:

"Physical inventory" means the sum of all measured or derived estimates of batch quantities of nuclear material on hand at a given time within a material balance area, obtained in accordance with specified procedures. -

It is our intention to specify the procedures and technical limitations of this inventory technique which we will call from here on Dynamic Process Inventory Determination (DPID).

1.2 Basic Considerations

For ready reference, a short description of the basic principles of this method is given below.

This technique makes use of the fact that the heavy material inventory between input- and product-accountability-tanks, which is replaced by the incoming material, can be measured quantitatively in subsequent product batches, provided that the isotopic composition of the inventory differs sufficiently from that of the new input material. The problem is to generate a step function in the isotopic composition of the input flow by loading the dissolver in such a way that a sufficient number of fuel elements of equal initial enrichment and irradiation history will be followed by fuel elements with isotopic abundances different from the first superbatches¹⁾. It is also possible to use the different isotopic composition of irradiated fuel elements from two different reactor batches which are processed in close sequence.

The evaluation of the physical inventory is a simple sum-up of product batches weighted by a factor which indicates the fraction of the inventory material in each individual product batch according to equ. (1.1).

$$(1.1) \quad H(t_1) = \sum_{i(t>t_1)} M_i^0 \frac{x_i - c_2}{c_1 - c_2} \quad \text{[kg U, Pu]}$$

t_1 = time of introducing the step input signal

M_i^0 = product batch size [kg U, Pu]

c_1, c_2 = weight fraction of the tracer isotope in the two consecutive superbatches¹⁾ which form the step signal [%]

x_i = weight fraction of the tracer isotope in product batch i [%]

¹⁾ "superbatch" is defined by a certain number of input batches with similar weight fraction of the tracer isotope.

The weight factor of subsequent product batches illustrates the operator's individual material management during the passage of the signal through the plant. Simulation studies and also experimental results indicate that this factor converges to zero in a similar way as an exponential function does.

The dispersion of the input step signal can be minimized if the operator runs the process according to a small number of specified procedures, mainly by special operation of the headend and product catch tanks but only during the residence time of the signal in the plant.

This technique may also be applied when the use of three superbatches and, consequently, two tracer isotopes is necessary. In this case the proper formalism is more complicated (1), but its derivation does not need the introduction of further basic concepts.

2. Technical Feasibility

2.1 MBA-System

A definition of "Material Balance Area" ("MBA") is given in the basic report of the IAEA INFCIRC/153 /4/ in article 110:

"MBA means an area in or outside of a facility such that:

- (a) the quantity of nuclear material in each transfer into or out of each MBA can be determined; and
- (b) the physical inventory of nuclear material in each MBA can be determined when necessary, in accordance with specified procedures,

in order that the material balance for Agency safeguards purposes can be established".

In connection with the application of the DPID the MBA has been defined for two different plants. The first one was the American NFS plant, the second one was the Belgian EUROCHEMIC where the method was very successful in two integral experiments.

Fig. 2-1 shows the example of the NFS-MBA, Fig. 2-2 shows the EUROCHEMIC-example.

In both cases the MBA system covered the process part only and excluded the product storage area because emphasis has to be laid on the condition that the system response can be measured adequately. In addition it is advisable to have only one significant input stream into the defined MBA. When the process needs auxiliary inputs of heavy material (e.g. the U^{IV} reducing agent in separation units) the DPID must be suitably corrected.

2.2 Design Limitations

In order to allow a fruitful application of the method a few limitations should be kept in mind. They apply to the design and the operation of the plant in question and to the material which is to be reprocessed. Of course there are interconnections between these parameters, e.g. it is

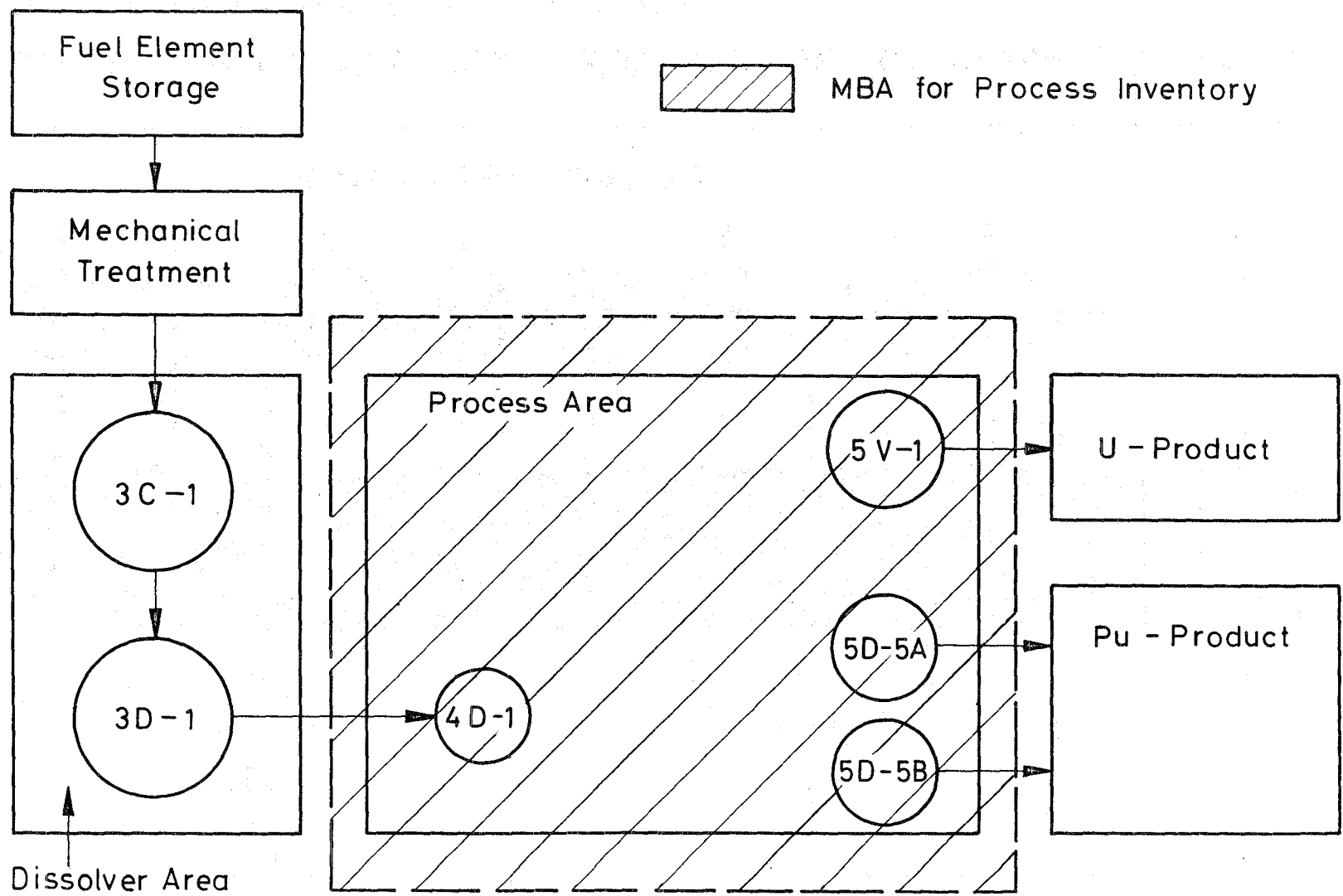


Fig.2-1: Definition of Material Balance Area for Process Inventory in NFS, USA

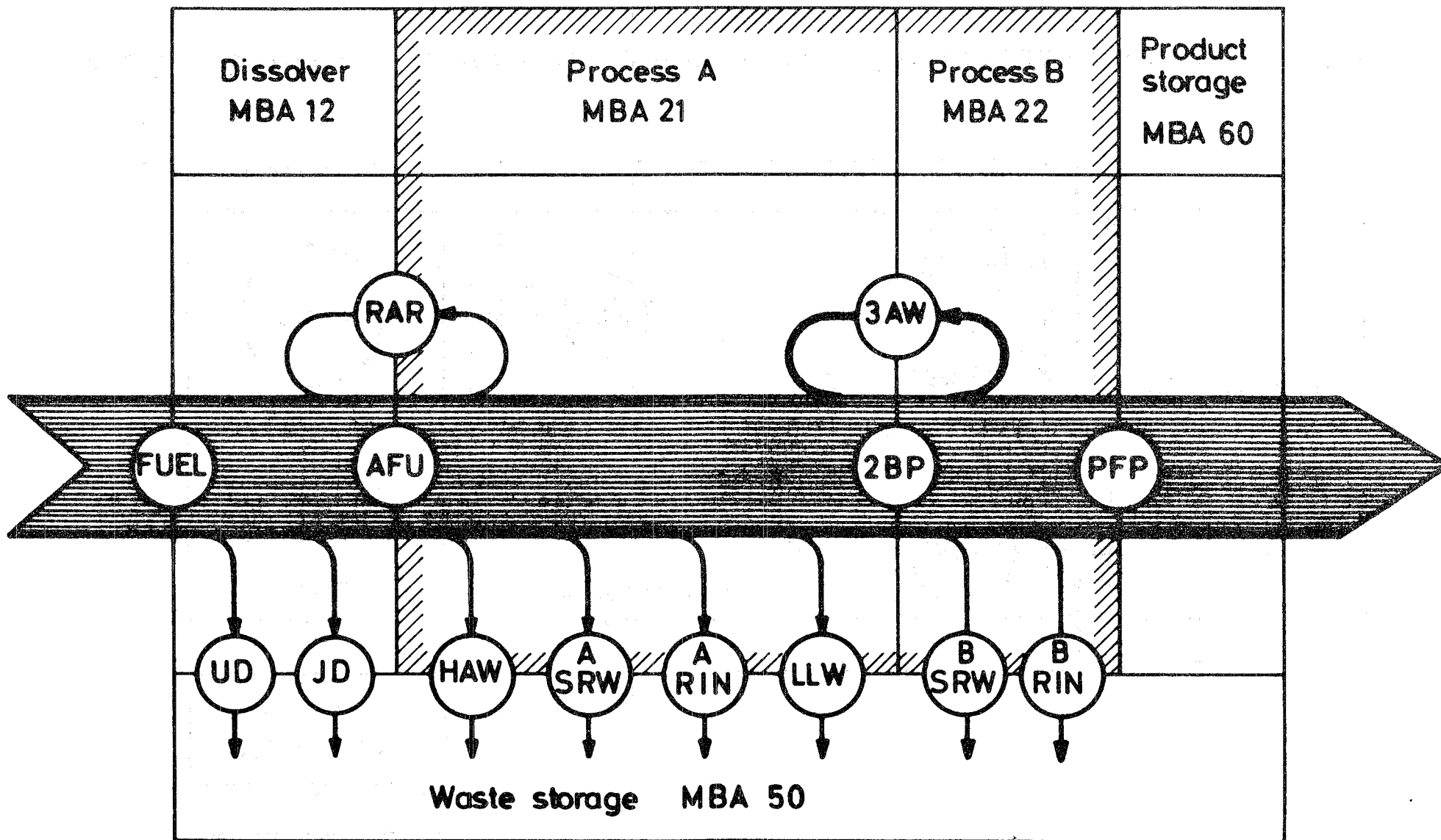


Fig. 2-2 Scheme of Pu-flow and MBA-system for Process Inventory in EUROCHEMIC, MOL, BELGIUM

evident that when large amounts of heavy material are mixed in big tanks the superbatches must also be large.

The limiting quantities for the superbatch sizes become smaller when the mixing of material inside the plant is reduced. As far as the design is concerned, the following parameters must be taken into account:

- a) The main influence on the dispersion of the step signals comes from the mixing inside the tanks with a great hold-up of heavy material, as buffer tanks, evaporators or product catch tanks.
- b) A second contribution is due to the heels in the tanks which can be inherent in the plant design or can be operation parameters. The condition of low heels - in order to have low mixing - should be mainly respected during the passage of the step through the respective units.
- c) Mixing inside the continuously working units of the plant (as mixer-settlers) cannot be avoided without a washout. But since this mixing is small, it can be left out. With the models which were constructed in order to describe the flow of fissile material /5/ relatively good correspondence of the theoretical and experimental results could be obtained, in spite of the fact that these units were neglected.

2.3 Operational Limitations

Generally speaking, the optimization of operations in a given plant with objective to allow the application of the DPID method must be solved case by case if there is only a limited quantity of material available. Simulation studies, which use a proper model of the plant under consideration, or historical experience of operation can be of great help. The only general rule is that the mixing should be maximized inside the superbatches and minimized between two consecutive superbatches. It should be attempted to homogenize the material inside the superbatches as much as possible.

The simulation studies of the Pu-cycle of the EUROCHEMIC plant - and their results which were applied during the Mol III experiment - give some hints on relevant operation parameters with respect to the influence of mixing. The studies were made on:

- a) Residual heels in the head-end tanks.
- b) Mixing procedures in the feed tanks between the accountability units and the first extraction cycle. This is possible in a plant with parallel intermediate storage tanks.
- c) Recycles to the second purification cycle.

ad a) Increasing the mixing in the head-end tanks by enlarging the heels can help to achieve a smoothing of the tracer concentration within one superbatch⁺⁾ . Obviously the prescription of minimum heels at the step passage must be kept.

Heels of 1 % and 10 % were studied.

ad b) Since the study of the mixing strategy in the feed adjustment tanks of the first extraction cycle was connected with the variation of other process parameters like the quantity of recycled material or the number of batches taken into account for the calculation of the concentration mean values for the superbatches, the results are not completely clear. There seems to be a tendency by which the variance of the measured inventory is lowered by the mixing strategy /5/.

ad c) Reprocessing campaigns with (5 %) and without continuous Pu-recycling have been studied. The results show no significant differences. The only important remark concerns the magnitude of the superbatches: a Pu recycle obviously increases the mixing so that larger superbatches are needed in order to estimate the process inventory with the same accuracy.

Two further important points are:

- d) It has been shown in /6/ that the DPID method can also be applied successfully in case of additional inputs, as e.g. U^{IV}-streams.

⁺⁾ This strategy for the input accountability tank has been called external mixing in /5/.

It is possible to adjust the model for additional inputs either by modification of the input or of the output signal.

- e) A further operation requirement is to avoid batchwise recycling of heavy material as far as possible.

2.4 Material Requirements

It has been mentioned before that one of the most important conditions for the application of the DPID method is the availability of enough material in each of the superbatches. To give an estimate of the required superbatch sizes one needs either practical experience with the plant or a rough model of the different mixing mechanisms. According to Fig. 2-3 it can be deduced /5/ that for the case of two superbatches the first one should not be smaller than $Q_2 - Q_0$ and the second one not smaller than $Q_2 - Q_1$.

Here Q_0 is the quantity of material which is produced up to the introduction of the step, Q_1 the quantity between the introduction of the step and the first influence of the concentration c_2 in the product, and Q_2 the quantity of material which is produced up to the point where only material of superbatches 2 comes out.

It is quite clear that the difference $Q_2 - Q_1$ gives the amount of material which is in a mixed state of superbatches 1 and 2. For the EUROCHEMIC plant this amount is about 3 times as large as the biggest heavy material hold-up of any unit of the plant.

In order to get some more detailed information, simulation studies on the EUROCHEMIC plant - under normal operation conditions, beginning with an empty plant, and for a certain type of material - have been done in order to estimate the amounts of material in superbatches 1 and 2 which should be present for a predetermined recovery of the inventory. The results are condensed in Table I:

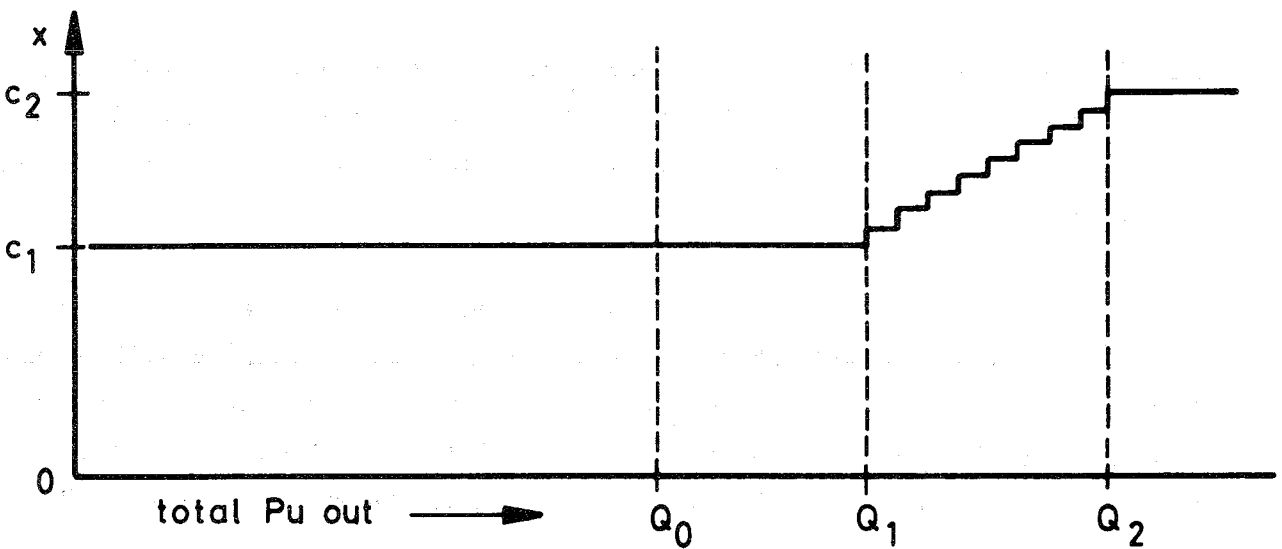
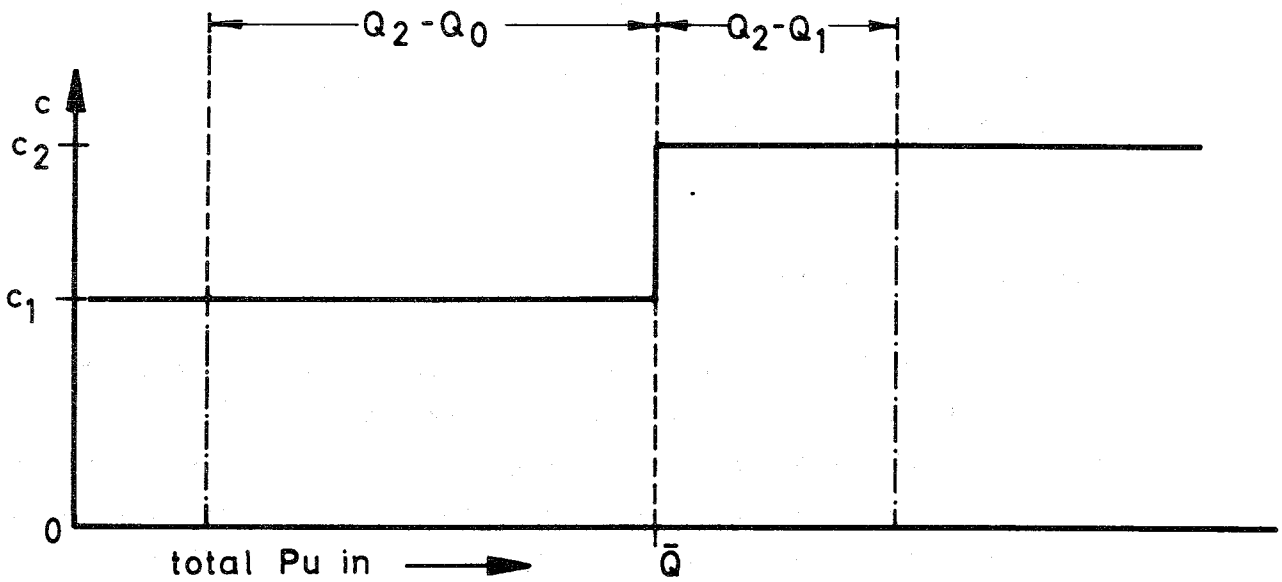


Fig. 2-3 : Determination of superbatches size for the application of the DPID method.

Table I: Minimum size of the two superbatches

No. of output batches considered	Percentage of the inventory measured	Superbatch size I ^o (Q ₂ -Q ₀)	(kg of Pu) II ^o (Q ₂ -Q ₁)
4	70.6	11.32	0
5	86.9	14.15	2.83
6	96.9	16.98	5.66
7	99.44	19.82	8.50
8	99.90	22.65	11.34
9	99.98	25.47	14.15
10	100.	28.30	16.98

So, if 97 % of the inventory is to be measured, superbatches 1 and 2 should contain 17 kg and 6 kg of Pu. For reasons given in section 4.2 it is not meaningful in actual situation to increase the superbatches size so that the recovery would be 100 %.

Analogous results for the U-cycle of the EUROCHEMIC are given in /3/. They are dependent on the strategy of recycling or not recycling. With recycling - and that means with a greater mixing - the superbatches have to be much larger for the determination of a prefixed percentage of the inventory.

All these numbers are results which come out for a specific plant under specific operation conditions. They are given in order to indicate the order of magnitude of the superbatches in a plant of this size and to show the trends which can be expected with certain parameter variations.

3. Generation of Adequate Input Signal

3.1 Information Requirements on Fuel Elements

The maximum benefit from the use of the illustrated technique is achieved when the physical inventory is measured with the lowest uncertainty.

A certain number of factors, which influence the statistical error of the inventory, have been already indicated /7/. They concern essentially the dissolution of the fuel elements available for reprocessing and the choice of suitable procedures for the plant operations. This second group of requirements was treated in 2.3.

The choices, which must be made a priori concern:

- a) the isotope to be used as tracer,
- b) the grouping of fuel elements in individual dissolution batches, and
- c) the dissolution sequence of the batches.

It can be shown /6/ that the minimum variance of the inventory is achieved when the ratio

$$(3.1) \quad r = \frac{(\sigma_1^2 + \sigma_2^2)^{1/2}}{|c_1 - c_2|} \text{ is minimum.}$$

Here

$$(3.2) \quad c_i = \frac{\sum_j c_{ij} M_{ij}^i}{\sum_j M_{ij}^i} \quad i = 1, 2$$

where the sum is performed on j batches of the i-th superbatches, c_{ij} is the weight fraction of the tracer isotope in the j-th batch of the i-th superbatches

M_{ij}^i is the total mass of the heavy element inside one input batch

Here the variances are

$$(3.3) \sigma_i^2 = \frac{1}{1 - \frac{\sum_j M_{ij}^2}{(\sum_j M_{ij})^2}} \frac{\sum_j M_{ij} (c_{ij} - c_i)^2}{\sum_j M_{ij}}$$

The expression

$\sigma_1^2 + \sigma_2^2$ is an indicator of the batch to batch variation in the weight fraction of the tracer isotope, and

$|c_1 - c_2|$ is the step size.

As long as every isotope of fissile material (of Pu or of U) may be used as tracer, a suitable analysis of the material to be reprocessed must be performed. For this reason the following information must be available for proper treatment:

- The number of fuel elements to be reprocessed, divided according to the type of reactor and of initial enrichments, if they differ.
- The number of fuel elements of each type, that are dissolved together as a "single" batch.
- Final quantities of Pu and U, contained in each fuel element (or assembly) and their isotopic composition, as calculated by the reactor operator.

The above mentioned data are usually communicated as shipper data by the reactor operators to the reprocessing plant management and, for power reactors, it seems that there is no reason for their classification.

On the basis of these data the batches will be constructed and ordered.

In those cases where the results of input analysis show that another tracer is more suitable the tracer can be different from the one chosen on a priori basis.

3.2 The Determination of "Super Batches"

When our method of inventory determination is used, the first constraint of "sufficient quantity" of material ⁺⁾ must be satisfied. In practice two cases are possible:

- a) The step in the weight fraction of the tracer isotope will take place between the reprocessing of two different types of reactor fuels.
- b) The step will be created by suitable selection and ordering of the fuel elements of one reactor.

The two cases are conceptually not different, but, in practice, it is better to follow two different approaches. In both of them it will be possible to profit from the use of a computer code illustrated in Annex 1.

It is necessary to stress that the optimization procedure, that will be described in the following pages, allows the optimal sequence determination for the measurement of the Pu or U inventory. Nevertheless it is possible that the best input sequence for one of the elements (Pu, e.g.) is apt to generate a "step" useful for the other element inventory. This second "step", however, is not necessarily optimal.

Let us assume that a Pu inventory measurement is required. In the first case a), we have the following problem: There are N fuel elements (of equal type), that must be used to create a superbatches, containing at least a prescribed quantity of Pu. In other words, the minimum number of batches n, which are necessary to create a superbatches of the prescribed quantity Q, can be calculated as follows:

$$(3.4) \quad n = j \left(\frac{Q}{mq} \right)$$

In this formula the following quantities are given:

- n = number of batches for the construction of a superbatches
- Q = quantity of Pu inside this superbatches
- m = number of fuel elements in one batch
- q = mean quantity of Pu in one element (the computer code has been written for equal amounts of Pu in all the elements, see below)

⁺⁾ see section 2.4.

j is a function that defines the minimum integer number with Q/mq as the argument.

The "sufficient quantity" constraint can be expressed as

$$(3.5) \quad N \geq n \cdot m$$

The computer code given in Annex 1 has been written with the constraint that the amounts of Pu (and U) in all the elements are equal. Nevertheless, in many actual cases, the mass differences of different elements are not so large to render relevant changes in the results. It must be recalled that the batch construction is made on the basis of reactor data which can be affected by not negligible errors: for this reason, in most cases, a more sophisticated analysis on such data is useless.

The n batches must be constructed in such a way that the batch to batch variation of the tracer isotope is minimal. This operation must be performed on all of the Pu (U) isotopes in order to select the tracer isotope. This tedious exercise is automatically performed by the GROUP code (Appendix 1). For each Pu (and U) isotope the computer code gives:

- the list (if any, when $N > n$) of the $N-n$ elements not considered,
- the optimum grouping of the elements in n batches of each containing m fuel elements,
- the expected relative weight fractions of each isotope in each batch, and
- their expected mean values and variances for the superbatches.

(The latter may be interpreted as a measure of the batch to batch variation.)

When more than n batches can be constructed with the N elements it is suggested to run the code for superbatches composed also of $n+1$, $n+2$, ... batches. In fact it is possible that no significant change in c (mean value of the tracer weight fraction) and no increase in its variance occur. In this case the use of a larger superbatches is advised, because this guarantees that the "sufficient quantity" constraint is better satisfied.

The calculation described heretofore must be performed for both superbatches independently.

When the results for all the isotopes are available the proper choice of the optimal Pu isotope to be used as tracer can be made. Obviously that tracer will be chosen for which the ratio (3.1) is minimized.

In the second case, b), the problem consists in the construction of two superbatches, with minimum quantities Q_1 and Q_2 , by putting the N available elements in groups of m . In this case the two superbatches will consist respectively of

$$n_1 = j \left(\frac{Q_1}{mq} \right)$$

and

$$n_2 = j \left(\frac{Q_2}{mq} \right)$$

batches. The "sufficient quantity" constraint can be expressed as $N \geq (n_1 + n_2)m$.

The following results for each Pu and U are obtained when the proper values of the input parameters of the GROUP program are used,

- the list (if any, when $N > (n_1 + n_2)m$) of the elements not taken into consideration for the determination of the superbatches,
- the optimum grouping of the elements in the two superbatches of n_1 and n_2 batches respectively, containing m fuel elements each,
- the expected relative weight fractions of all the isotopes (for both U and Pu) in each batch, and
- their expected mean values (for each superbatch) and variances.

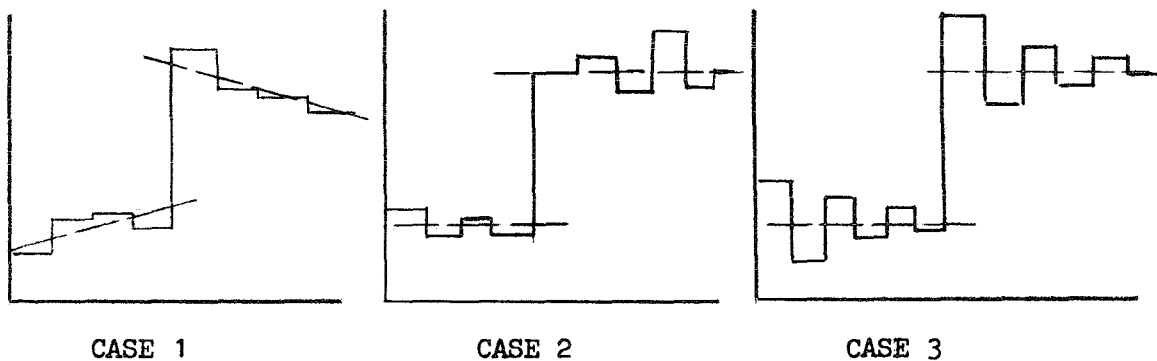
When more than $n_1 + n_2$ batches may be constructed with the N available elements, it is suggested to run the computer code increasing the parameters n_1 and/or n_2 and to decide, from the examination of the results, if an increase in the quantities Q_1 and/or Q_2 is possible. However, it must be recalled that this increase of the "minimum quantities" is possible and advisable only when no significant increase of the ratio (3.1) occurs.

3.3 The Determination of the Input Sequence

The problems concerning the tracer isotope selection and the composition of the single dissolution batches have been solved. The last decision, that must be taken before the dissolution, is the determination of the input sequence for the batches of the two superbatches.

Two rules should be respected:

1. The regression lines, which approximate the histogram of the tracer weight fraction versus total material quantity, must be as flat as possible for both superbatches. It has been demonstrated that any divergence of the regression line from the horizontal decreases the precision of the final result. (Avoid the case 1, illustrated in the figure.)



2. When the rule number 1 is respected (case 2 and case 3 of the figure) it is preferable to shift the maximum tracer oscillations towards the first batches of the superbatches (case 3 of the figure must be preferred to case 2).

3.4 Practical Problems

Sometimes it happens that the required grouping of elements for the construction of the different batches and/ or the prescribed input sequence can not be fulfilled because of handling difficulties inherent to the storage location of the reprocessing plant. In such cases the inventory taking should be planned sufficiently early, to solve the problems indicated at 3.2 and 3.3 before the fuel element shipment. In this case the shipment and the storage may be, in time, planned in such a way that the handling difficulties at the reprocessing storage pond are overcome.

4. Procedures to Determine Process Inventory

4.1 Analysis of the Measured Input Signal

Because of the lack of adequate shipper data the realized input step signal may show considerable differences from the provided input signal as specified in section 3. All further evaluations will be based on the measured input signal. Thus we proceed as follows:

- A - Specify the nuclear material (U or Pu) which is to be balanced by book and physical inventory comparison.
- B - Make a chronological list of relevant input batch data:
 - batch identification,
 - date and hour of transfer (as specified in 5.1),
 - total mass of element expressed in grams,
 - available isotopic measurements in weight percent.
- C - Specify the single input batches forming the two superbatches as given in section 3.
- D - Select that particular tracer isotope which shows the smallest ratio r as given in equations (3.1-3.3)⁺.
- E - Plot the weight fraction of the tracer isotope versus the mass of the element of subsequent input batches for the two superbatches as in Fig. 5-1:

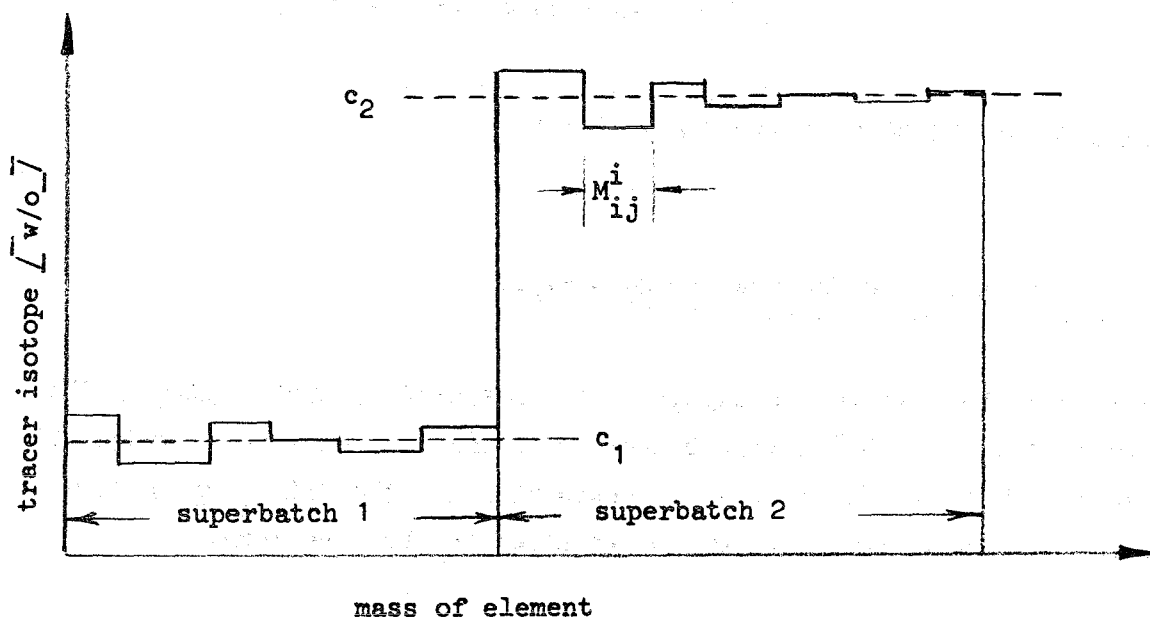


Fig. 5-1: Isotopic input signal

⁺) r_{\min} gives already a first estimate of the relative standard deviation of DPID

- F - Select one additional auxiliary isotope which shows the second smallest ratio r .
- G - Plot the weight fractions of the tracer isotope vers. the auxiliary isotope of all input batches forming the two superbatches and establish the maximum possible mixing area according to Fig. 5-2.

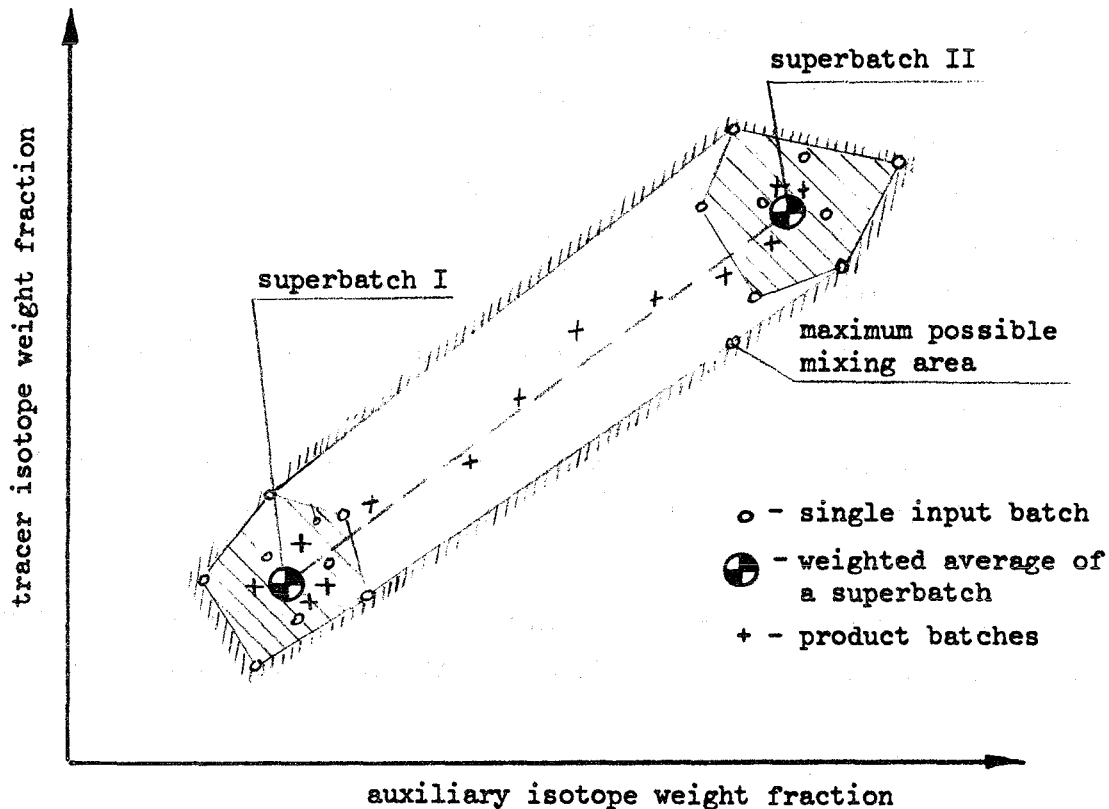


Fig. 5-2: Mixing area of two superbatches

4.2 Analysis of the Measured Product Signal

The main problem in this analysis of the measured product signal is the determination of the "cut-off batch" which will not show any significant fraction of inventory material. Consequently all the following product batches will also not give any contribution to the inventory.

We therefore propose to proceed as follows:

- A - Make a chronological list of all relevant product batch data starting from the first product batch after initial physical inventory measurement at time t_0 :
- batch identification,
 - day and hour when the transfer out of the defined MBA is finished,
 - total mass of element in grams,
 - available isotopic measurements in weight percent.
- B - Find the first product batch after the step time t_1 as given in 5.1.
- C - Plot the weight fractions of the tracer isotope and the auxiliary isotope of the subsequent product batches, which are released from the MBA after t_1 (see B).⁺) Note that every product point must be in the indicated maximum possible mixing area if the isotopic measurements are unbiased and no other material than the fuel of both superbatches is mixed together.
- D - Proceed plotting as specified in C until the isotope vector merges with the weighted average point within the dashed mixing area of superbatches I and II. If the next few product batches come roughly to the same point, one can be sure that all inventory material passed the product accountability tank. The "cut-off batch" is consequently defined to be the first product batch which reaches the weighted average of superbatches I and II.

4.3 Separation of Mixing Types in the Relevant Product Batches

The relevant product batches may contain either

- a) pure inventory material of superbatches I or,
- b) mixtures of material from both superbatches I and II.
- c) In the case that the superbatches I and II were too small to replace completely the inventory material (superbatch I) three component mixtures may occur including material of a third superbatches.

⁺) This information should be summarized in Fig. 5-2.

The separation of the two (or three) groups is an important procedure with respect to the inventory evaluation (Sec. 4.4). With the help of Fig. 5-2 the grouping can be arranged very quickly. Product batches belonging to group a) are roughly within the dashed mixing area of superbatches I. As soon as the product isotopic vector leaves the mixing area of superbatches I following roughly the dashed line between the weighted averages of superbatches I and II the product batch will belong to group b).

Complications arise if a third superbatches is involved as the solution of three component mixing equations requires that the coefficient determinant is sufficiently different from zero [1]. This condition can be demonstrated graphically in Fig. 5-3.

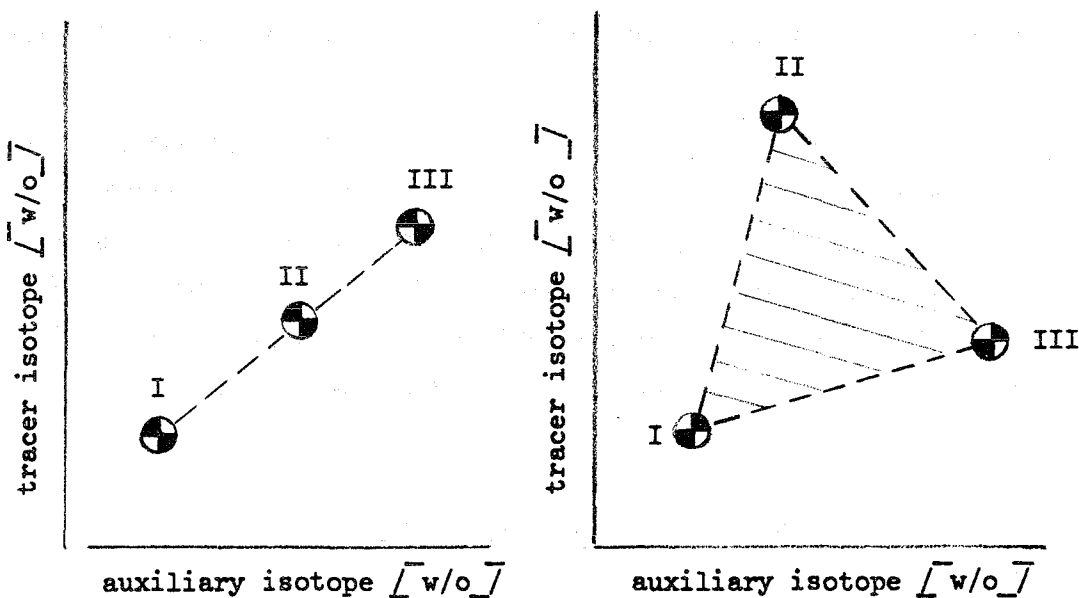


Fig. 5-3: Three component mixing

The figure on the left demonstrates the conditions of an unsolvable equation system where the three components (I, II, III) are linear dependent. The figure on the right shows an example of a solvable equation system where the coefficient determinant which is proportional to the shadowed mixing area is sufficiently different from zero.

It is recommended to avoid the three component mixing system by enlarging the size of superbatches II. Only for reasons of completeness the relevant procedures for the evaluation of the inventory including a three component mixture is also outlined in the following section. Product batches which contain components of three superbatches can be identified by plotting the corresponding isotopic data in Fig. 5-3. In case of a solvable equation system (figure on the right) the product point is located clearly within the maximum possible mixing area.

4.4 Evaluation of the Physical Inventory

From the analysis of the measured input signal (Sec. 4.1) it is possible to perform the final choice of the tracer isotope (or isotopes, when three superbatches must be considered).

The analysis of the output signal allow the separation of the output batches in two (or three) groups, according to their content (Sec. 4.2 and 4.3).

The results of the previous analysis allow a simplification in the determination of the inventory. In fact the inventory value, H , may be expressed as

$$(4.1) \quad H = H_1 + H_2 (+H_3)$$

where H_i is the contribution to the total inventory of the i -th group.

In the following each single contribution will be evaluated independently. In 4.4.4 the evaluation of the whole inventory and its statistical error will be outlined.

4.4.1 H₁ Evaluation

The contribution of the first group of batches to the inventory is simply given by

$$(4.2) \quad H_1 = \sum_{i=1}^{n_1} M_i^0$$

where n_1 is the number of batches of the first group and M_i^0 are the masses of heavy material (of the first superbatches only) present in the i -th output batch.

The contribution H_1 is independent of the c values (weight fractions of tracer isotope), so that the statistical error of H_1 may be easily calculated from the errors of the single mass measurements. These errors are usually small ($\approx <1\%$) and normally distributed. Thus, it follows that

$$(4.3) \quad \sigma_{H_1}^2 = \sum_{i=1}^{n_1} \sigma_{M_i^0}^2$$

4.4.2 H₂ Evaluation

The definition of H_2 is given (see 1.1) by

$$(4.4) \quad H_2 = \sum_{i=1}^{n_2} M_i^0 \frac{x_i - c_2}{c_1 - c_2}$$

n_2 being the number of batches of the second group. All the other symbols have been already defined.

The use of the weighted average concentrations c_1 and c_2 may have a biased estimate of H_2 ⁺⁾ . Due to the fact that the theory is still not sufficiently developed to indicate how to calculate these values in order to obtain a better estimate of H_2 , the following approach may be used.

⁺⁾ L. Farese - private communication.

Let us assume that the independent variables M_1^0 , x_1 , c_1^i and c_2^i are realizations of random variables each of which has a known prescribed pdf (probability density function). For the values M_1^0 and x_1 the pdf's are assumed to be normal with mean values equal to the measured ones and standard deviations equal to the standard errors of the related measurements.

The variables c_1^i and c_2^i are given by

$$(4.5) \quad c_j^i = \frac{\sum_k M_{kj}^i R_{kj} c_{kj}}{\sum_k M_{kj}^i R_{kj}} ; \quad j = 1, 2$$

where the sums are performed on all the input batches of the j -th superbatches and where

M_{kj}^i is the input mass of the heavy material contained in the k -th batch of the j -th superbatches,

c_{kj} is the weight fraction of the tracer isotope in the same batch and

R_{kj} are random numbers extracted from a (0,1) interval with uniform distribution.

Consequently, c_1^i and c_2^i are also random variables, and their mean values and standard deviation may be calculated. Note that the previous formula assures that $Ec_j^i = c_j$; and that

$$(4.6) \quad \min(c_{kj}) \leq c_j^i \leq \max(c_{kj})$$

The repeated use of the formula $\sqrt{4.4}$ for a high number of realizations of the independent variables M_1^0 , x_1 , c_j^i allows the computation of a high number of contributions of type H_2 , related to the real input signal. It is then possible to calculate the frequency curve, the distribution, the mean value and the variance of the realizations of H_2 . The frequency curve may be interpreted as the probability density function of H_2 , so that it is possible to determine its expectation value and the error limits, according to a prescribed confidence interval.

These operations may be easily effectuated by a computer, employing the computer code CC2, described in Annex 2.

4.4.3 H₃ Evaluation

The DPID requires sometimes the use of a "three component" system (three subsequent input superbatches, with two selected isotope tracers). In this case a further contribution to the inventory is given by

$$(4.7) \quad H_3 = \sum_{i=1}^{n_3} M_i^o \frac{T_i}{D}$$

where the sum is performed over the n₃ batches of the third group and where

$$(4.8) \quad T_i = \begin{vmatrix} 1 & 1 & 1 \\ x_i & c_2 & c_3 \\ y_i & d_2 & d_3 \end{vmatrix}$$

and

$$(4.9) \quad D = \begin{vmatrix} 1 & 1 & 1 \\ c_1 & c_2 & c_3 \\ d_1 & d_2 & d_3 \end{vmatrix}$$

c_j and d_j (j = 1,2,3) are the concentrations of two tracers (e.g. Pu-241 and Pu-242) in the j-th input superbatches. x_i and y_i are the concentrations of the same isotopes in the i-th output batch.

The inadequacy of this theory in respect to the treatment of real cases results from the same reasons pointed out in the case of the "two component" systems. It follows that the evaluation of the experimental data may be obtained following a procedure similar to the one outlined in the previous paragraph.

In this case, however, it is necessary to pay some attention to the fact that the selection of random values for the pairs c_jⁱ, d_jⁱ must respect the physical correlation existing between these parameters. The use of the following system

$$(4.10) \quad c_j^i = \frac{\sum_k M_{kj}^i R_{kj} c_{kj}}{\sum_k M_{kj}^i R_{kj}}$$

$$j = 1,2,3$$

$$(4.11) \quad d_j^i = \frac{\sum_k M_{kj}^i R_{kj} d_{kj}}{\sum_k M_{kj}^i R_{kj}}$$

logically justified by the equation [4.5] (the symbols have the same meaning) indicates that single values of c'_j and d'_j are obtained as randomly weighted means of the concentrations of the input batches c_i and d_i . It follows that, in every realization of the pair of random variables c'_j, d'_j , the same contribution of the individual input batches to the means must be considered. The use of the same weighting factors (M_{kj}^i, R_{kj}) in both equations of the system assures that the physical correlation existing between c and d inside each single superbatches is respected.

Similarly to the previous case of H_2 , the contribution H_3 is evaluated many times as function of independent variables randomly selected from the mentioned distributions. The analysis of the results of this application of the Monte Carlo technique gives the expectation value of H_3 , its pdf, and the error limits, according to a prescribed confidence interval.

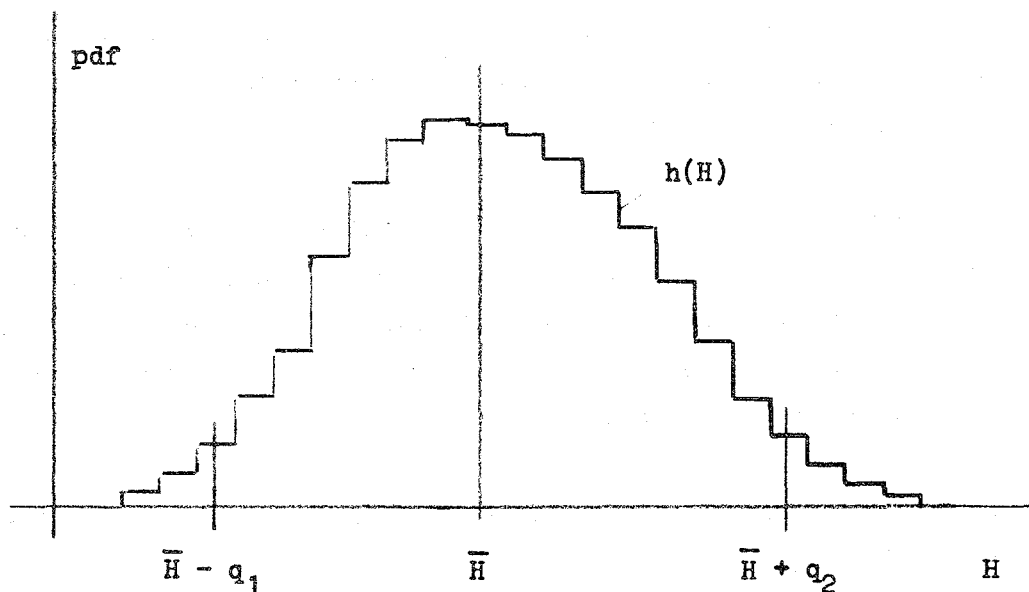
These calculations may easily be effectuated by a computer, using the computer code CC3, described in Annex 3.

4.4.4 Total Inventory Determination and Error Limit Estimation

In the previous sections it has been indicated how to calculate the different contributions to the total process inventory and how to interpret each of them. The pdf's that have been associated to the measurement of each H_1 ($1 = 1, 2, 3$) represent the confidence, that can be attributed to the measured figure. When a variable is normally distributed, its pdf is simply specified by the value of the standard deviation parameter. This is the reason that 1σ or 2σ values are often used when confidence intervals are given. The approximation of the pdf of H_2 and H_3 with a normal function is, generally, not valid. Therefore a more sophisticated error propagation analysis is given below.

Finally the pdf must be calculated for the whole inventory H . This will give an immediate picture of the measurement significance. This kind of pdf is obtained by subsequent convolutions of the pdf's of the addends H_1, H_2 and H_3 . The definition of these convolutions is given in Appendix 4. For the moment, however, it is sufficient to remember that in this case the pdf's are given in form of histograms and that the convolutions reduce to simple sums. The pertinent formulae are also given in Appendix 4.

However, it is important to stress the physical significance of the formalism connected with the pdf of the quantity H. In the figure an example of such a type of function: h(H) is plotted. The area under the curve is, by definition, equal to 1.



As expectation value the mean value \bar{H} is chosen. The error limits connected with a prescribed confidence p are the values $\bar{H} - q_1$ and $\bar{H} + q_2$, such that:

$$(4.9) \quad \int_{\bar{H}-q_1}^{\bar{H}} h(H) dH = p/2 = \int_{\bar{H}}^{\bar{H}+q_2} h(H) dh$$

5. Book Inventory Determination Procedures

5.1 Definition of the Time Interval

The physical inventory determined by DPID (section 4) represents the inventory situation within the defined MBA at the time (t_1) when the isotopic step signal is introduced.

As this time (t_1) is very important for closing the material balance, more specifications must be given:

- Case I: The input accountability tank (IAT) is located outside the defined MBA-system. The relevant time point is defined at the beginning of the transfer of the first input batch of the second superbatches from the IAT to the next process unit.
- Case II: The IAT is located inside the defined MBA which is sometimes so the case that the adequate shipper-receiver differences can be established. The relevant time point is defined at the beginning of the transfer of the first input batch of the second superbatches from the dissolver tank to the IAT. In the case that there are relatively great heels in the IAT one has to relate the isotopic signal to the incoming material and not to outgoing material as it is done in case I.

Correspondingly the book inventory (B), which is to be compared with the physical inventory (H) (determined according to the procedures specified in section 4), has to be related to the time point defined above.

5.2 Evaluation of Book Inventory

The evaluation of the book inventory is a well known procedure which needs not to be specified in detail here. The relevant equation is given below:

$$(5.1) \quad B(t_1) = BI(t_0) + \sum_i I - \sum_p P - \sum_w W \quad \text{[kg heavy element]}$$

where $BI(t_0)$ = beginning inventory at time t_0
 I, P, W = input, product and waste batches
 i, p, w = number of input, product and waste batches
 between t_0 and step time t_1 (as defined in section 5.1)

The input accountability stops with the last batch of the first superbatches. However, only those product and waste batches are accounted for, which are completely transferred out of the defined MBA between t_0 and t_1 .

5.3 Determination of the Variance of Book Inventory

The Relative Standard Deviation (RSD) associated with each flow can be calculated with

$$(5.2) \quad \delta_F^2 = \left(\delta_c^2 + \frac{\delta_r^2}{n} \right) \frac{\text{volume}}{\text{weight}} + \left(\delta_c^2 + \frac{\delta_r^2}{n \cdot m} \right) \text{analysis} \quad (\text{RSD})^2$$

where

δ_c	= calibration error	}	RSD
δ_r	= precision of the measurement		
n	= number of batches		
m	= number of analyses per batch		

Equ. (5.2) is valid if all the batches have equal volumes (weights) and concentrations. This assumption appears to be justified. Calibration error and precision have to be estimated from the experience of quality control procedures.

The resulting variance of the book inventory is the sum of all the variances:

$$(5.3) \quad \sigma_B^2 = \sum_F \delta_F^2 \cdot (\Sigma F)^2 ; F = I, P, W, BI$$

6. Procedures to Determine the MUF

6.1 Book-Physical Inventory Difference

From the point of view of the safeguards control the main interest of the DPID is the comparison between the measured value (H) and the accountability value (B), usually referred to as book inventory.

The difference between these two quantities is the well known MUF (Material Unaccounted For), defined as

$$(6.1) \quad \text{MUF} = B - H$$

The calculation of such a quantity and of its statistical errors is of capital importance for the control activities. In fact the article 30 of INFCIRC/153 /4/ explicitly requires a statement on "the amount of material unaccounted for over a specific period, giving the limits of accuracy of the amounts stated".

6.2 Confidence Intervals

As illustrated in 4.4.4, the determination of the limits of accuracy for the MUF is deduced from its pdf. Such a function is obtained, as in the case of the whole inventory, by a suitable convolution between the pdf.s associated to H and B. The first has been calculated in 4.4.4; the second is simply a normal distribution, the mean value and standard deviation of which have been deduced in sect.5. Indicating by $h(H)$ and $b(B)$ such pdf.s, the pdf for the MUF, $m(\text{MUF})$ is given by

$$(6.2) \quad m(\text{MUF}) = \int b(\text{MUF}+x) \cdot h(x) dx$$

or by a suitable approximation of this integral with a sum.

This operation is described in more detail in Appendix 4, where the performances of a computer code, which may be used for calculations, are illustrated.

6.3 The "Statement" on the MUF

In theory, if a facility has a perfect accountancy system for all the inputs and outputs, the MUF should be only the consequence of cumulated measurement errors. Its value, if not zero, should be "consistent" with zero. The analysis of the MUF measurements must then state a conclusion about such "consistency".

The determination of the required statement is straight forward when the error distribution (the pdf) around the MUF (the mean value of such a distribution) is given.

Let p be the value fixed a priori of the probability level requested for the statement ⁺⁾ . With the employment of the same technique reported in 4.4.4 it is possible to define the two error limits m_1 and m_2 for the measured value of \overline{MUF} .

Two cases must be considered

$$(a) \quad \overline{MUF} - m_1 < 0 < \overline{MUF} + m_2$$

$$(b) \quad \overline{MUF} - m_1 > 0$$

In the case (a) it is possible to state that the measured MUF is "consistent" with 0 (with a confidence p); in the case (b) the measured MUF is "not consistent" with 0 (with a confidence p).

A third case is theoretically possible: $\overline{MUF} + m_2 < 0$. This last case, in spite of the fact that it gives a piece of information on the "safe side" from the safeguards point of view, should impose deeper investigation on the process accountancy techniques (measurement included) or on hidden inventories inside the process area.

⁺⁾ The "working group meeting on quantitative data and results of systems analysis and integral tests" (Vienna 4-8.10.71) suggested values of p in the range $.90 \div .99$; usually 95 % is adapted.

In the case (a) mentioned before it is possible to calculate the probability that the MUF is equal or lower than 0. Such value is simply

$$(6.3) \quad p(\text{MUF} \leq 0) = \int_{-\infty}^0 m(\text{MUF}) d(\text{MUF})$$

This is a further information, that quantifies the agreement between B and H at the moment of closing the balance.

Another interesting statement, from the point of view of the safeguards control, concerns the comparison of the measured MUF with a threshold amount (e.g. the LOA referred to in ref. / 8 /). Such a statement is obtained in the identical way in which the previous has been obtained. It is sufficient only to substitute the 0 by the actual value of the threshold.

Acknowledgement

The authors are grateful for the help of Nebojsa Nakićenović who has reviewed the manuscript.

7. Literature

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A/CONF.49/P/773 July 1971

Annexes

In the following pages a short description of the computer codes, suitable for data handling of the DPID, is given.

The listings of such codes, written in FORTRAN language, may be found, together with the solution of sample problems, in the external report EUR 4833.e : "Four Computer Codes for the Dynamic Process Inventory Determination - Application in Safeguards" by A. Rota (1972).

Annex 1

The Computer Code GROUP

The computer code GROUP has been designed to provide the information necessary for the composition of the single input batches, when a DPID is planned.

The problem has been outlined in paragraphs 3.1 and 3.2.

The input data which define the problem are:

- the total number, N , of the available fuel elements (of the same reactor and approximatively of the same enrichment);
- the number of elements, m , of which an input batch is composed;
- the total number of batches desired in superbatches 1 and superbatches 2 (N_1 and N_2 respectively);
- the name of each fuel element available for reprocessing together with the calculated values of its content in Pu and U isotopes.

When both N_1 and N_2 are different from zero, the code defines the optimum grouping for the construction of two superbatches. When either N_1 or N_2 is zero, only one superbatches is constructed. If $N_1 = 0$ the superbatches is constructed in such a way that the higher tracer concentration is selected. When $N_2 = 0$ the minimum tracer concentration is selected.

Note that when both N_1 and N_2 are different from zero, the first superbatches will be constructed with the minimum tracer concentration and the second with the maximum one. When a going down "step" is desired, the two superbatches selected by the code will be used in inverse order.

The selection of the grouping of the element for each batch of the required superbatches is made for each isotope, both of U and Pu, present in the burned fuel elements. For each of these isotopes, and according the specifications required (N_1 , N_2 , N), the GROUP code gives:

- the list of the elements not utilized (when $(N_1 + N_2) \cdot m < N$),
- the list of the groups of fuel elements, which constitute a batch,
- for every constructed batch the expected abundance of each Pu and U isotope,
- for every superbatches the mean values and the standard deviations of such abundances.

The block diagram of the code is reported in fig. A1-1.

Fig. A1-2 shows an example of batch grouping for a case of 12 elements which are to be divided in groups of 3.

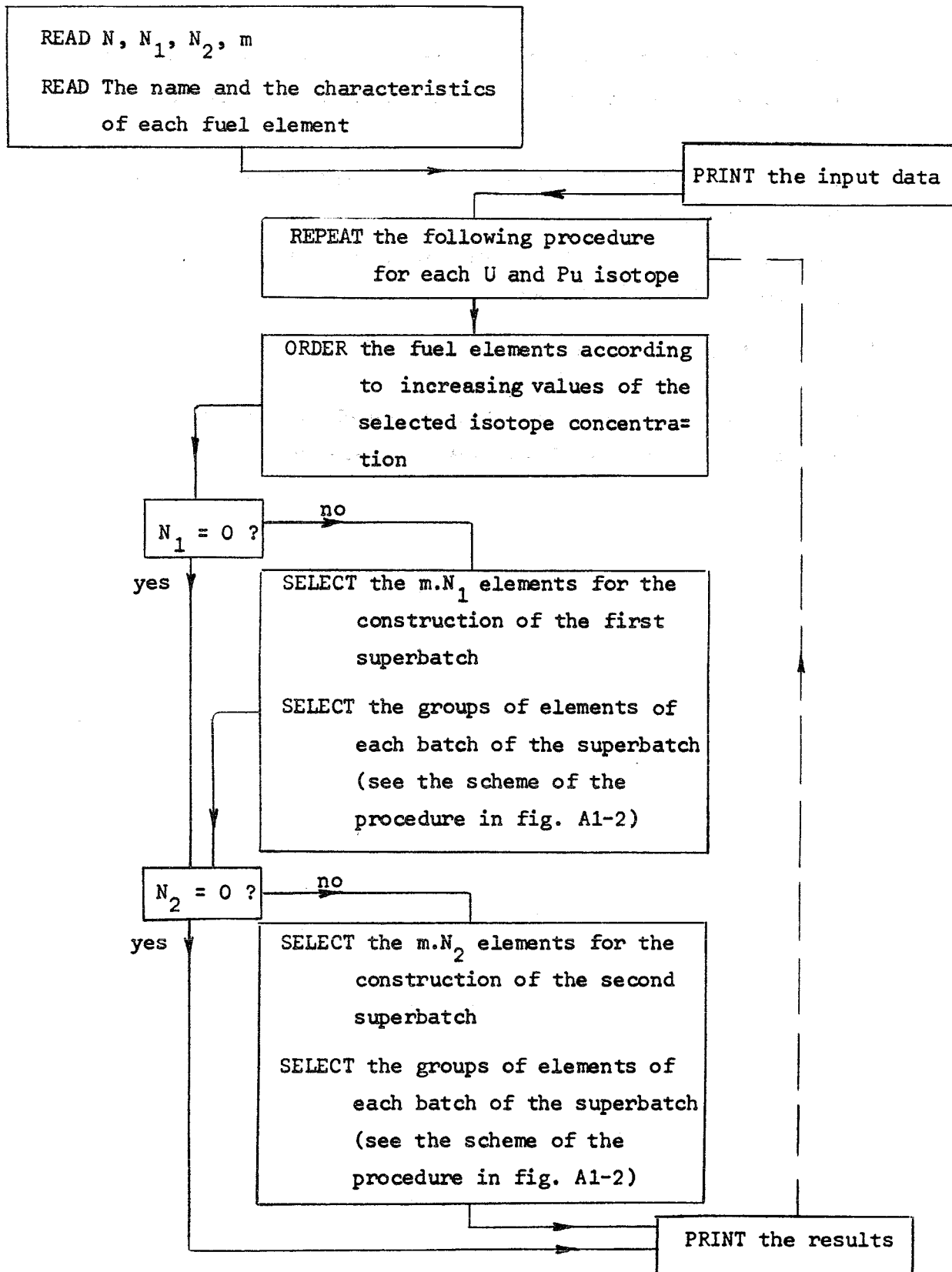


Fig. A1-1: Block diagram of the GROUP computer program

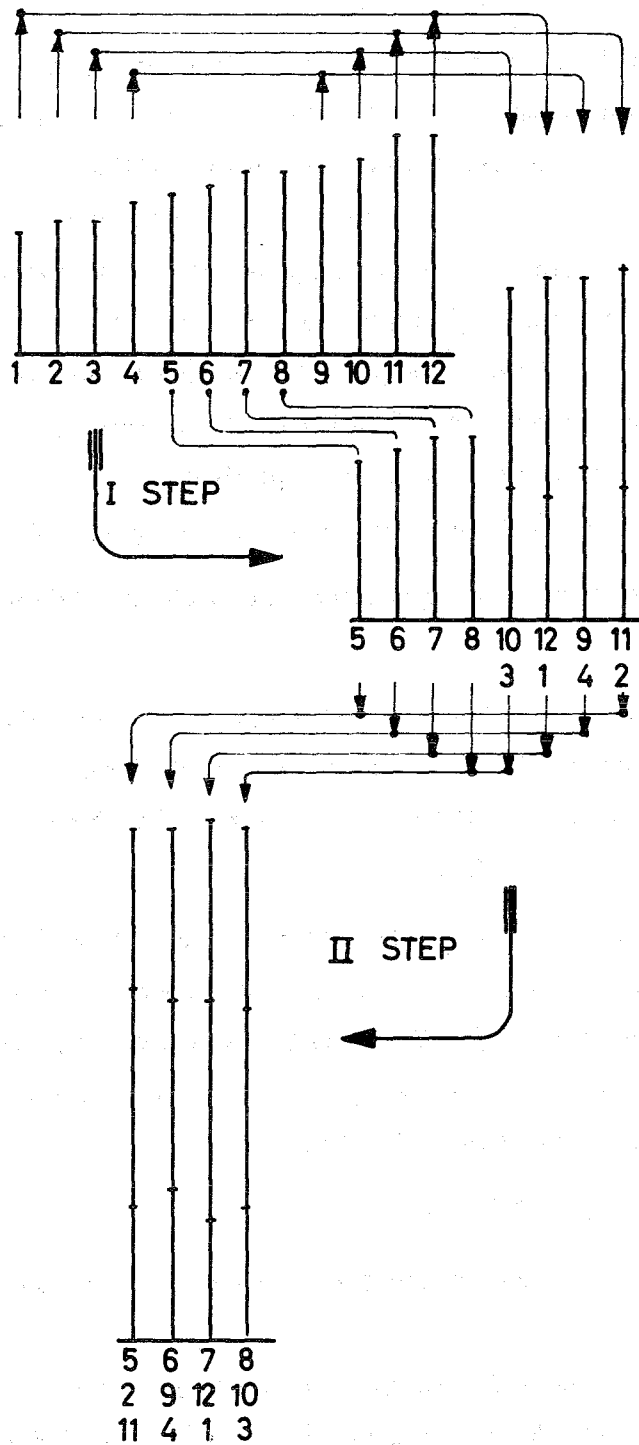


Fig. A1-2: Scheme of the procedure for batch grouping: Case of 12 elements to be divided in groups of 3.

Annex 2

The Computer Code CC2 for H_2 Evaluation

The calculation of the "two component" contribution H_2 to the physical inventory, according to the formula (4.4) does not in itself justify the use of a computer and the construction of a computer code. However, this becomes unavoidable when further information is required. The computer code CC2 has been in fact designed to calculate the measurement uncertainty. By a proper use of a Monte Carlo technique, the pdf and the distribution function of the parameter H_2 are calculated. H_2 is considered as the realization of a random variable depending, on the set of random variables M_i^0 , x_i , c_1 , c_2 , as given in (4.4). For each of these variables the code requires the definition of its distribution.

M_i^0 and x_i ($i = 1$, number of output batches) are assumed to be random variables with normal distributions individuated by a proper value of the standard deviations ($\sigma_{M_i^0}$, σ_{x_i})

c_1 and c_2 are random variables, the distributions of which are implicitly defined by the equations (4.5). This formalism is used by CC2. It follows that the input batch characteristics (M_{kj}^i , c_{kj}) must be introduced as input data for the program.

Further input data for the CC2 are:

- the number N of "histories" requested for the Monte Carlo procedure;
- the range that should be covered by the error analysis;
- the step magnitude for the frequency distribution, and then for the p. d. f.;
- the list of the particular values of the probability for which an error boundary definition is particularly interesting;

- a list of parameters of minor interest (initialization values for the random number generation routines, print-out definitions, optional perforation of the results, etc.).

Fig. A2-1 reproduces a logical flow-sheet of the program.

The output of the program contains the following information

- a summary of the input batch data
- a summary of the "two component mixture" output batch data
- the median value $\bar{H}_2 = f(\bar{M}_i^0, \bar{x}_i, \bar{c}_1, \bar{c}_2)$
- the contribution to \bar{H}_2 of each output batch
- the frequency histogram
- the table of the pdf of H_2
- a rough plot of such pdf
- the table of the distribution of H_2
- a rough plot of such distribution
- the main characteristics of the statistical analysis:
 \bar{H}_2, σ_{H_2} , relative standard deviation
- the table of the values of H_2 corresponding to the prescribed values of the probability.

The last table can look e.g. like this:

	P(X)	X - \bar{H}_2	X
1	2.5	-528.93	2 320.94
2	5.0	-433.08	2 416.79
3	10.0	-332.67	2 517.20
4	20.0	-209.17	2 640.70
5	80.0	179.53	3 029.40
6	90.0	261.62	3 111.49
7	95.0	321.26	3 171.13
8	97.5	365.57	3 215.44

The values of the column P(X) are input data. In this sample case they have been chosen in order to select confidence intervals of

95 % , 90 % , 80 % , 60 % ; in fact the error limits for a 95 % confidence are located at $P(X) = 2.5$ (%) and 97.5 (%); this gives

$$H_2 = \bar{H}_2 \begin{matrix} +365.57 \\ -528.93 \end{matrix}$$

The same procedure is used for the other confidence intervals.

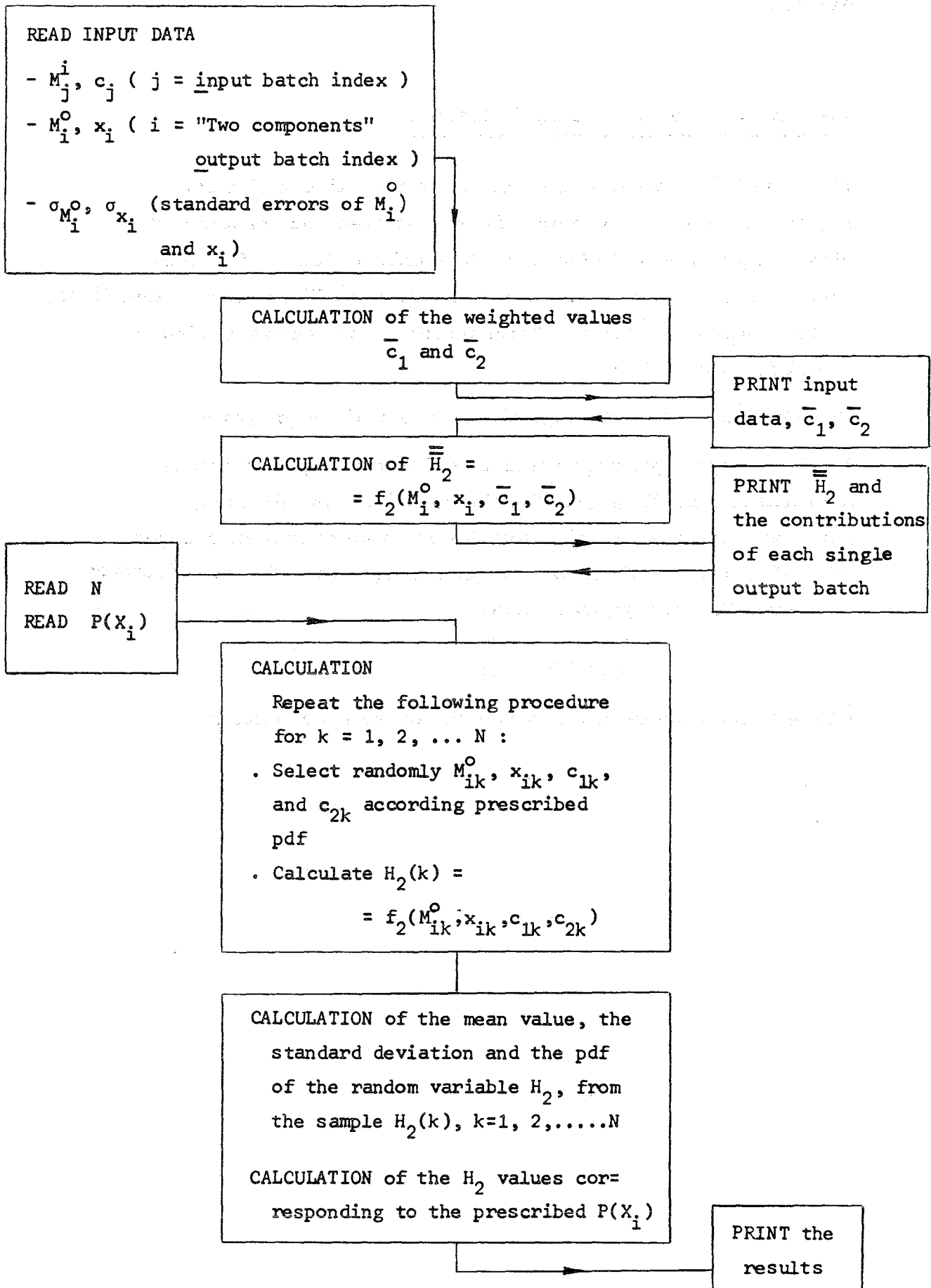


Fig. A2-1: Block diagram of the CC2 computer program

Annex 3

The Computer Code CC3 for H₃ Evaluation

The code CC3 evaluates the "three component" contribution, H₃, to the physical inventory. It is conceived and constructed identically to the CC2 (Annex 2). The only obvious differences concern the formula for the H₃ calculation (4.7) and the system (4.10, 4.11) which defines the distributions of the random variables c_j and d_j , $j = 1, 2, 3$.

Fig.A3-1 illustrates the logical flow sheet of the program.

The list of the input requirements and of the output results is identical to that illustrated for the code CC2. In this case, obviously, the input batch characteristics must include 3 superbatches ($j = 1, 2, 3$) and two weight percent ages of tracer isotopes. In particular, the program needs:

$$M_{j,k}^i, c_{jk}, d_{j,k}$$

where k is the index of the k -th batch of the superbatches j .

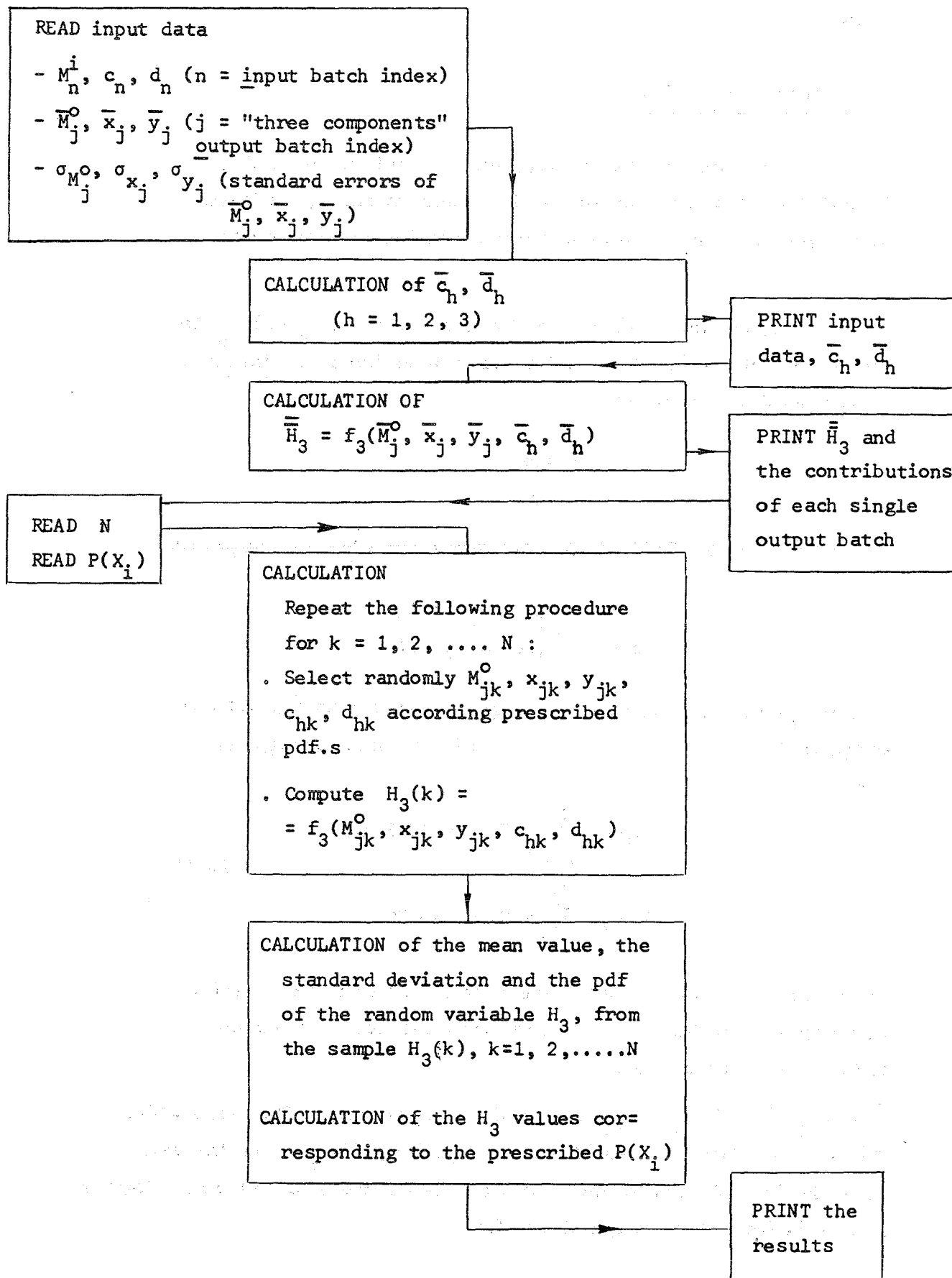


Fig. A3-1: Block diagram of the CC3 computer program

Annex 4

The Computer Code MUF

The present code has been designed to calculate the p. d. f. (probability density function) of a random variable, defined as an algebraic sum of others random variables with known pdf.s.

Let p and q be independent random variables and f₁ and f₂ the corresponding pdf. s. The analytical expressions for the pdf. s of the random variables

$$s = p + q$$
$$r = p - q$$

are given, respectively, by the following convolution integrals:

$$g(s) = \int f_1(t) \cdot f_2(s-t) dt$$
$$h(r) = \int f_1(r+t) \cdot f_2(t) dt$$

In this particular case we will express the f.s in the form of histograms. The previous integrals then become :

$$G(S) = \sum_{K1}^{K2} (F1(K) \cdot F2(S-K))$$
$$H(R) = \sum_{J1}^{J2} (F1(R-J) \cdot F2(J))$$

(A4.1)

(The capital letters used for the function names and variables correspond to the small letters, which indicate continuous function and variables).

Let N₁₁, N₁₂ (N₁₁ < N₁₂) define the integer interval outside which F1 is identically zero and let N₂₁, N₂₂ (N₂₁ < N₂₂) have the same meaning for F2. From these limits it is possible to deduce analogous intervals for the functions G and H:

$$N_{11} + N_{21} < S < N_{12} + N_{22}$$

$$N_{11} - N_{22} < R < N_{12} - N_{21}$$

Ignoring those terms which certainly do not give any contribution to the sums (A4.1), the sum limits become:

$$K1 = \text{Max} (N_{11}, S - N_{22})$$

$$K2 = \text{min} (N_{12}, S - N_{21})$$

$$J1 = \text{Max} (N_{21}, N_{11} - R)$$

$$J2 = \text{min} (N_{22}, N_{12} - R)$$

These relationships are used in the MUF computer code to evaluate the sequence of convolutions necessary for a statement on the "material unaccounted for". This program, however may be used, independently of the safeguards problems, for the evaluation of any sequence of convolution integrals of the mentioned type.

Great care must be taken to correctly define the histogram intervals. They must be equal for every considered pdf.

The code uses as input the subsequent pdf. s (defined through their histograms or through their mean value and standard deviation, when they are of normal type). The process of the data is sequential: a first pdf $f_1(p)$ is defined (it is read or it is the result of a previous convolution); second pdf, $f_2(q)$ is read together with an indication; the possible indications and their significance are the following:

- + when the pdf of the random variable $s = p + q$ is required
- 1 - 2 when the pdf of the random variable $s = p - q$ is required
- 2 - 1 when the pdf of the random variable $s = q - p$ is required.

For every convolution, the code delivers the table of the resulting pdf, together with its relevant parameters (mean value, standard dev., etc.);

on request, the following further specifications may be obtained:

- a rough plot of the resulting pdf
- a rough plot of the resulting distribution function
- the table of the s values, corresponding to the prescribed values of the probability (as described in Annex 2 for the code CC2).