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1. INTRODUCTION

The main purpose of a fissile material safeguards system in a fuel reprocessing facility is to prevent or detect any diversion of fissile material from the plant. Of the three basic safeguards measures [1, 2] namely, containment, material balance and other redundant methods, available to a control authority, the second measure plays a key role in a properly developed safeguards system. A complete containment of fissile material may not be economically practicable in a reprocessing plant operating industrially, and an excessive use of the third measure may require undesirable intrusion in the plant or impose a heavy burden of activity on the control authority. Whereas a material balance for a plant can be established relatively economically and without causing an excessive intrusion into the plant or an excessive workload on the control authority.

With the help of a fissile material balance around a reprocessing plant, it is possible to assess the probability of a diversion after it has taken place. This means there is always a finite time lag between the time of diversion and its detection, and a diversion cannot be prevented directly by carrying out a material balance alone. However, with a proper choice of strategic points in a plant and a combination of other safeguards measures, the material balance may decrease the possibility of diversion in an indirect manner and may also reduce the time lag between a diversion and its detection.

2. CHOICE OF STRATEGIC POINTS

To establish a complete fissile material balance in a reprocessing plant over a given period of time, it is essential to determine the amount of fissile material which has flown into the plant, the amount which has come out of the plant and also the amount which has remained inside the plant (denoted as hold-up of the plant) during the same period of time.

2.1. Input and output measurements

The input and output of fissile material can be definitely and completely determined by measuring the fissile material content in all the effluent streams to and from a plant. It is therefore, necessary and also sufficient to locate the strategic points only at the effluent streams to determine the input and output amount of fissile material. In a reprocessing plant these strategic points are:

(a) Fuel element storage area (Pt. a; Fig. 1)

(b) The input accountability vessel containing the dissolver solution and recycle feed stream (Pts. 1b, 1a; Fig. 1)

(c) The product output accountability station (Pts. 3, 4; Fig. 1)

(d) Waste exit points (Pt. 4; Fig. 1)

It is to be noted that of the four points mentioned above only the points b), c), and d) are required to establish the material balance. The first point is required mainly for containment. The fissile material content is not determined at this point but it is only ensured that all the fuel elements received at this point also reach the dissolver tank. The other two points 5 and 6 shown in Fig. 1 are the socalled internal strategic points and are located after the active part of the process and before the end purification step. They are not required for establishing the material balance but may offer some specific advantages in reducing the possibilities of diversion. These advantages are discussed in chapter 4 of this paper. The strategic points for the waste streams are symbolically combined together in point 4 of Fig. 1. In practice some losses also occur from the inactive area.

2.2. Hold-up measurement

The amount of fissile material which enters the plant at the beginning of a reprocessing operation and becomes the plant hold-up, cannot remain and accumulate in the plant for an indefinite period under industrial conditions. It has to appear at some later date in one of the effluent streams. This time lag may not be very high for a campaign type operation in which a certain amount of fuel is reprocessed in a batchwise manner. The plant operator has to guarantee a return of at least 98 - 99% of the fissile material fed into the plant. As the plant is normally started without any appreciable fissile material hold-up, it has to be built up at the beginning of the operation before the plant can start processing purified material under equilibrium conditions. At the end of the campaign, the plant operator has to process the fissile material hold-up also to fulfil his guarantee. This fissile material appears then mainly in the product stream and partly in the waste streams. In a continuously operated plant the hold-up may not appear in any of the effluent streams for much longer periods of time than in a campaign type operation. But it can also be measured in the effluent streams by reprocessing it and emptying the plant from time to time. Besides, the fissile material is continuously renewed in the hold-up of such a plant and a particular fissile atom has to appear in one of the effluent streams after traversing through the various process steps. Therefore, for the establishment of a fissile material balance it is completely adequate to determine the input and output flows as well as the hold-up amounts in a reprocessing plant at the strategic points mentioned under 2.1 only and no additional points are required.

3. RELATIVE IMPORTANCE OF STRATEGIC POINTS

Although fissile material flows at all the effluent strategic points have to be measured, the importance of these points are different.

The range of uncertainties in the integrated amount of fissile material which is obtained at each of these points after a given amount of fuel has been processed, can be regarded as an index for assessing the relative importance of these points. For a given amount of fissile material, the range of uncertainties is mainly a function of the fraction of fissile material passing through a strategic point and the accuracy with which the amount is measured. These accuracies are not randomly distributed among the strategic points but have definite range of values for a particular point because of the characteristics of the material to be measured at this point.

3.1. Overall ranges of accuracies at different strategic points

The strategic points in a reprocessing plant are shown in Fig. 1. Under normal operating conditions two fissile material streams have to be measured at the first strategic point for determining the input. These streams are firstly, the fissile material input to the dissolver from the fuel elements (1b) and secondly, the fissile material contained in the recirculated acid stream (1a). Because of the different types of operations involved in determining the amount (density or volume measurement, sampling, determination of concentration) and the high activity of the solution, the attainable overall accuracies of measurement for the feed solution from the dissolver may be only between 1 - 2 %. The major contribution to the overall inaccuracies may be from the volumetric measurement. The accuracy for the acid recycle stream lies somewhere around 3 %. The plutonium and uranium in the product streams (3, 4) are obtained in highly purified form and the attainable accuracies therefore are fairly high. They may lie in the range of 0.2 - 0.25 %. The waste streams (4) contain very small amounts of fissile material and are highly active. The overall accuracies of fissile material measurements may be in the region of 10-20 %.

3.2. Range of uncertainties for simulated reprocessing campaigns

A number of reprocessing campaigns were simulated with two different plutonium throughputs and different measurement accuracies to assess the range of uncertainties at the different strategic points. The input data for the simulation are summarized in table I. The low Pu-content fuels correspond to an advanced light water type reactor with a burnup of around 20 000 MWd/t whereas, the high Pu-content fuels represent a typical sodium cooled fast breeder type with an averaged core-blanket burnup of around 27 000 MWd/t. The Pu-concentration is that for a mixed core-blanket management. The batch sizes for the two types correspond to three batches per reactor load.

In determining the ranges of uncertainties (expressed in kgs of Pu), four times the standard deviation σ were used. This corresponds to a confidence level of 99.9936 %, whereas all the accuracies in measurements [expressed in %] correspond to a relative standard deviation of one σ.

The ranges of uncertainties for three different accuracies in feed measurements for the two different Pu-throughputs have been presented in tables II and III respectively. The accuracies of measurement at the rest of the strategic points were kept constant for all campaigns and are also shown in table II. Only the uncertainties in Pu-amounts have been presented,as the uncertainties in the U-235 amounts for the low concentrations considered here, are negligible. A summary of all the assumptions and the results is given in table IV.

The most important point to note from both the tables is the fact that the range of uncertainties increases linearly with increasing inaccuracies. The highest amount of uncertainty is contributed by the feed point followed by the recycle acid, product and the waste point respectively. For ranges of accuracies and throughputs considered here the relative importance of the strategic points will also be in the same order.

Although 99 % of the plutonium fed into the plant passes through the product point, the range of uncertainties is less at the product point than at the point for recycle acid, even though only 12.5 % of the total plutonium passes through the latter. This is because of the lower accuracy of measurement at the point for recycle acid. On the other hand, the range of uncertainty in the waste stream is the lowest although the inaccuracy of measurement is the highest there. The main reason is the small amount of fissile material flowing through this point.

Another important point to note is the influence of the number of samples taken. The accuracy improves with increasing number of samples.

For example, extrapolating the feed point accuracy to 0.2 % (table II), the range of uncertainties reduces to 0.25 kg of plutonium as against 0.27 kg for the product point, which also has an accuracy of 0.2 %. Three samples were assumed to be taken daily for the feed stream compared to two samples for the product stream. In table III, 35 samples were assumed to be taken for the product stream and 15 for the feed stream. For the same accuracy, the situation is reversed in this case.

An analysis of the results presented in tables II and III show that maximum amount of effort and care has to be devoted to the measurement of fissile material flow at the feed point. With increasing plutonium throughput the requirement for high accuracy becomes all the more important. The accuracy for the recycle acid stream should also be improved and if this is not possible, the amount of recycled plutonium should be reduced. The present day product point accuracies are quite adequate even for fairly high throughputs of plutonium. No strong incentive exists for the improvement of measurement accuracies in the waste streams.

3.3. Probability of diversion for different accuracies at the feed point

The probability of diversion for different accuracies at the feed point have been given in table V. The campaign for the low Pu-content fuels (table II) has been taken as the basis for these calculations. The results of table V are shown graphically in Fig. 2, in which the constant probability of diversion lines have been drawn as parameter with the amount of plutonium diverted as the ordinate and the measurement accuracies at the feed point as the abscissa. It is to be noted that a given probability of diversion always corresponds to a value which is > the amount of plutonium shown as diverted in table V and Fig. 2 [3]. The probability of diversion, which is a statement made by the safeguarding authority, decreases with the increasing inaccuracy of measurement at the feed point. Since the probability of diversion influences partially the effectiveness of a safeguarding system, the results of table V and Fig. 2 show that the effectiveness of the system would decrease with increasing inaccuracies at the feed point.

4. EFFECT OF INTERNAL STRATEGIC POINTS

It was mentioned under 2.2. that no other strategic points than those mentioned in 2.1. are necessary to establish a complete material balance in a reprocessing plant. Introduction of additional strategic points at (5) and (6) in Fig. 1 may however, give some advantages for the overall safeguarding systems. These points are after the extraction cycle (in which the uranium and plutonium are decontaminated to a fairly high degree and separated from each other) and before the end purification stage. The plutonium in the form of Pu-nitrate is stored in tanks before the end purification step and collection of samples and measurement of the volume of the solution by the controlling authority at this point may not cause an intolerable intrusion for the plant. The advantages of this strategic point are summarized below. The same advantages would be obtained for the uranium stream also (5).

(i) Since the fissile materials from this point onwards are considerably less active, the introduction of a strategic point will devide the plant hold-up into an active and an inactive part. The fissile material hold-up between two consecutive strategic points will be reduced thereby.

(ii) The material balance can be established for the two parts separately by considering the measurement at point (6) as the plutonium output for the active part and as input for the inactive part of the plant.

(iii) Since the plutonium at point (6) can be measured with the same high accuracy as at the product point (3), and the hold-up will be lower than the total plant hold-up, the range of uncertainties in the amount measured will be considerably lower for this part of the plant than for the whole plant. As a result, the probability for the detection of a given amount of diverted material will be higher.

(iv) Normally it would be easier to divert plutonium from the inactive part of the plant as it is present in a readily accessible form. Since the probability of detection for a given amount of diverted material from the inactive part increases by the introduction of a strategic point at this step, the risk of diversion would increase as well. The plant operator would then tend to plan a diversion from the active part of the plant where the risk would be less (because of the lower accuracy of the feed point). The uncontrolled extraction of plutonium from an active part of the plant is associated with a considerably larger amount of effort than is required from the inactive part of the plant. The introduction of the strategic point (6) may therefore exert an indirect influence to prevent diversion.

The main disadvantage of these points may lie in the fact that recycling of fissile materials to other parts of the plant may be required from process considerations. A detailed analysis of the process is necessary to assess properly the virtue of these two strategic points.

5. STRATEGIC POINTS FOR A REPROCESSING PLANT IN A CLOSED FUEL CYCLE

All the considerations carried out sofar have been made for a single reprocessing plant. The only difference, if the plant were to be a part of a closed fuel cycle, would be that the accuracies of measurements at the feed end and the product end, for the same range of uncertainties, need to be slightly less. However, the accuracies for the determination of fissile material content in irradiated fuel elements from a reactor, which forms the previous step for the reprocessing plant in a closed cycle, are expected to be considerably less than the feed point accuracies in the reprocessing plant. Therefore, the introduction of the concept of a closed fuel cycle would not bring any significant advantage for the feed end of a reprocessing plant and it would not be possible to eliminate the strategic point at this end. The accuracies at the product end may however, be reduced by a factor of around 1.4, as the product can be measured with the same accuracy at the entrance of a fabrication plant, which forms the next step of a closed fuel cycle. It is also possible to eliminate the measurement of the fissile material flow at either the product end of a reprocessing plant or the feed end of a fabrication plant in a closed fuel cycle, as one of the measurement is redundant. It would however, be preferable to eliminate it at the fabrication plant as otherwise, the completion of material balance in a reprocessing plant might be delayed considerably, so that the detection of a probable diversion would also be delayed. The control activity at the entrance of a fabrication plant cannot however, be eliminated completely. Even if no fissile material measurements be carried out there, the control authority has to ensure that all the fissile material received at this plant is also processed.

6. USE OF TRACERS TO DETERMINE THE HOLD-UP AND THE RATE OF CHANGE OF HOLD-UP IN A PLANT

The hold-up of fissile material can be determined only with a time lag in case it is measured as a throughput at one of the strategic points for the outgoing streams. It may however, be possible to follow the time behaviour of the hold-up and may even be possible to determine the actual amount of hold-up with the help pf suitable isotopic tracers.

To determine the time behaviour i.e. the rate of change of hold-up with time, a suitable isotope (either of uranium or of plutonium) may be added to the feed solution, with a given periodicity. If the hold-up remains constant during the operation, the same periodicity would be observed and measured in the product stream. In case the hold-up does not remain constant but also shows some periodicity, this will be superimposed on the periodicity introduced at the feed point. The resulting periodicity which will be characteristic of the normal operation, can also be measured at the product point. Any deviation from the feed periodicity (in case of a constant hold-up) or the resulting periodicity (in case of a variable hold-up), for a given throughput would mean that the hold-up has deviated from the normal operation. The nature of the deviation may throw some light on the behaviour of hold-up inside the plant.

With a radioactive tracer with suitable half life and activity, the actual amount of hold-up in the plant may be estimated by adding this tracer to the feed solution and measuring its activity in the product stream.

Data on some uranium and plutonium isotopes which may be of interest in this connection are shown in table VI.

Further work in this field would however be necessary before the suitability of this method can be assessed.

Acknowledgement

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TABLE I. INPUT DATA FOR SIMULATING FUEL REPROCESSING CAMPAIGNS

<table>
<tr><th></th><th>Low Pu content fuel</th><th>High Pu content fuel</th></tr>
<tr><td>Plant capacity [t heavy metal /d]</td><td colspan="2">1</td></tr>
<tr><td>Batch size [t]</td><td>30</td><td>15</td></tr>
<tr><td>Pu-concentration [w %]</td><td>0.44</td><td>7.0</td></tr>
<tr><td>Total amount of Pu-processed [kg]</td><td>132</td><td>1050</td></tr>
<tr><td>Pu lost in the waste [%]</td><td colspan="2">1.0</td></tr>
<tr><td>Amount of Pu in recycle acid [%]</td><td colspan="2">12.5</td></tr>
<tr><td>Hold-up in the plant [t] (total heavy metal)</td><td colspan="2">6</td></tr>
<tr><td>Type of distribution for the measuring accuracies and losses</td><td colspan="2">normal</td></tr>
</table>

TABLE II. RANGE OF UNCERTAINTIES FOR DIFFERENT MEASUREMENT ACCURACIES IN FEED STREAM FOR LOW PU CONTENT FUEL

Total amount of Pu processed: 132 kg;
Accuracies (1σ) at strategic points: 1a : 3%;
3 : 0.2 %; 4 : 10 %;
No of samples at strategic points / per day /:1a: 3;
3 : 2; 4 : 6

	Accuracies at Feed Point (1b) / 1σ; % /		
	0.5	1	2
No. of samples at feed point / per d /	3	3	3
Range of uncertainties / kg Pu; 4σ /			
Strategic points (Fig. 1)			
Feed (1b)	0.63	1.26	2.53
Product (3)	0.27	0.27	0.27
Acid recycle (1a)	0.45	0.45	0.45
Waste (4)	0.10	0.10	0.10
Total uncertainties	0.85	1.37	2.59

TABLE III. RANGE OF UNCERTAINTIES FOR DIFFERENT MEASUREMENT ACCURACIES IN FEED STREAM FOR HIGH PU CONTENT FUEL

Total amount of Pu processed: 1050 kg;
Accuracies (1σ) at strategic points: same as in table II;
No. of samples at strategic points / per day /: 1 a: 15;
3 : 35; 4 : 30

	Accuracies at Feed Point (1b) / 1σ; % /		
	0.5	1.0	2.0
Number of samples at feed point / per day /	15	15	15
Range of uncertainties / kg Pu; 4σ /			
Strategic points (Fig. 1)			
Feed (1b)	3.16	6.33	12.66
Product (3)	0.75	0.75	0.75
Acid recycle (1a)	2.35	2.35	2.35
Waste (4)	0.51	0.51	0.51
Total uncertainties	4.05	6.81	12.84

TABLE IV. RANGE OF UNCERTAINTIES IN PLUTONIUM MEASURED AT STRATEGIC POINTS IN A REPROCESSING PLANT

	Low Pu-Content Fuel			High Pu-Content Fuel		
Amount of Pu Processed [Kg]	132			1050		
Measuring Accuracies [1σ; %]						
Product	0.2 (2)			0.2(35)		
Acid Recycle	3.0 (3)			3.0(15)		
Waste	10.0 (6)			10.0(30)		
Feed	0.5 (3)	1.0 (3)	2.0(3)	0.5(15)	1.0(15)	2.0(15)
Range of Uncertainties [kg Pu]						
Feed	0.63	1.26	2.53	3.16	6.33	12.68
Acid Recycle	0.45	Same	Same	0.35	Same	Same
Product	0.27	Same	Same	0.75	Same	Same
Waste	0.10	Same	Same	0.51	Same	Same
Total	0.85	1.37	2.59	4.05	6.81	12.84

( ) Number of Samples per day

TABLE V. PROBABILITIES OF DIVERSION FOR DIFFERENT MEASURING ACCURACIES AT THE FEED POINT

Pu-processed, accuracies, no. of samples: same as in table II; Amount diverted = 2 kgs of Pu

Accuracy [1σ; %] at the feed point	0.5	1.0	2.0
Range of uncertainty [4σ ; kg]	0.85	1.37	2.59
Probability of diversion	Minimum amounts (or greater) from the diverted Pu, which can be declared as diverted with the corresponding probability of diversion		
[%]	[kg Pu]	[kg Pu]	[kg Pu]
99.9936	1.57	1.31	0.70
95	1.83	1.72	1.41
90	1.86	1.78	1.58
80	1.91	1.86	1.73
70	1.94	1.91	1.83

TABLE VI. DATA ON URANIUM AND PLUTONIUM ISOTOPES WHICH MAY BE CONSIDERED FOR TRACER USE

	Half life	Typical activity	Method of production
I. Uranium			$^{232}Th(p,3n)\rightarrow {}^{230}Pa \xrightarrow{\beta} {}^{230}U$
U-230	21 d	α;5.88 MeV	$^{232}Th(d,4n)\rightarrow {}^{230}Pa \xrightarrow{\beta} {}^{230}U$
			$^{230}Th(d,2n)\rightarrow {}^{230}Pa \xrightarrow{\beta} {}^{230}U$
			Can only be produced in a cyclotron
U-232	72 a	α;5.32 MeV	$^{231}Pa \xrightarrow{n,\gamma} {}^{232}Pa \xrightarrow{\beta} {}^{232}U$
		5.26 MeV	$^{236}Pu \xrightarrow{\alpha(100\ \%)} {}^{232}U$
U-237	6.73 d	γ;0.207 MeV	$^{241}Pu \xrightarrow{\alpha(10^{-3}\%)} {}^{237}U$
II. Plutonium			
Pu-236	2.85 a	α;5.77 MeV	$^{237}Np(n,2n)\rightarrow {}^{236}Np \xrightarrow{\beta} {}^{236}Pu$
			can be produced in a reactor
Pu-246	11 d	γ; 1 MeV	Production method not yet known

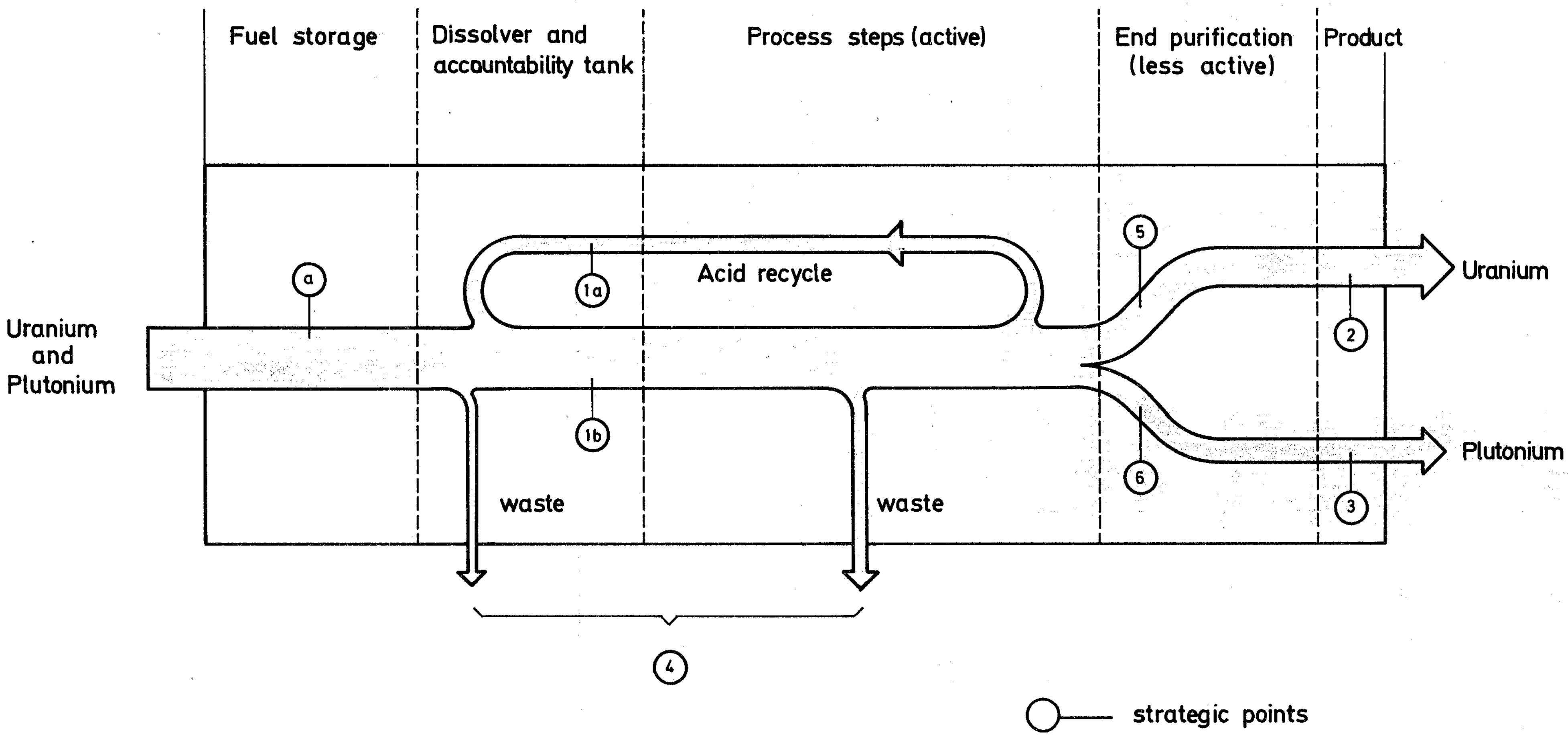

Fig. 1 Fissile material flow and location of strategic points in a reprocessing plant

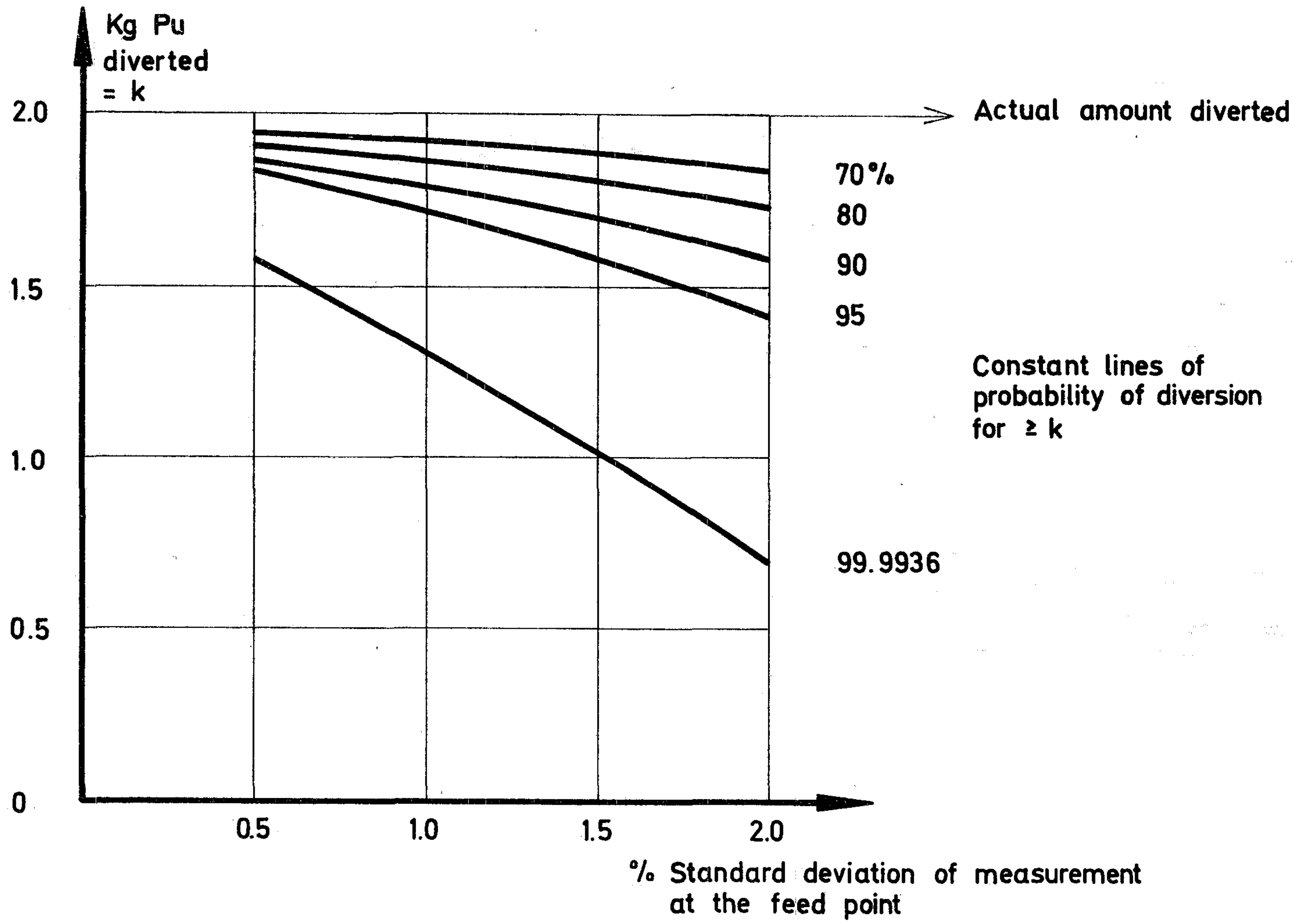

Fig. 2 Amounts of plutonium diverted as a function of feed point measurement accuracy with probability of diversion (%) as parameter.