

**Senior Sophister Physics Project  
Report for Trinity College Dublin**

**“Safeguard Analysis  
of Material Flows in  
a Gas Centrifuge  
Plant”**

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## **Abstract**

A system for the detection of material diversion in a uranium Gas Centrifuge Enrichment Plant (GCEP) based on real-time mass evaluation of the inputs and outputs has been theoretically examined. The response of this Continuous Mass Monitoring System (CMMS) to two specific material diversion scenarios has been simulated and in each case, the diversion could be detected. As a result of these basic simulations, the CMMS-based method of diversion detection could prove to be a promising technique for use in safeguards verification of GCEP's.

## **Acknowledgements**

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## Introduction

Under the terms of the Non-Proliferation Treaty, the International Atomic Energy Agency (IAEA) carries out inspections of nuclear industry facilities in all countries that have signed the treaty. In particular, facilities where weapons-grade uranium or plutonium can be produced require a lot of attention to ensure that there are no undeclared activities with regard to nuclear materials.

At the moment, there are 3 main possible diversion scenarios that can occur at a GCEP which normally produces declared amounts of Low Enriched Uranium (LEU,  $^{235}\text{U}$  content  $< 20\%$ ) which concern the IAEA <sup>[1a]</sup>, namely:

- 1.) The production of large quantities of undeclared Highly Enriched Uranium (HEU,  $^{235}\text{U}$  content  $\geq 20\%$ )
- 2.) The diversion of a significant amount of declared LEU
- 3.) The production of undeclared LEU from undeclared feed

In order to prevent these from occurring, the IAEA conducts inspections both inside and outside the halls containing the enrichment centrifuges. These inspections involve Limited Frequency Unannounced Access (LFUA) visits to the interior of the cascade hall<sup>[1b]</sup> to ensure that the internal arrangement of centrifuges and piping networks is unchanged. They also involve activities outside the cascade hall, such as examination of records and reports, non-destructive analysis of samples, along with containment and surveillance techniques.

In addition, equipment is employed by inspectors which is capable of detecting HEU in containers using gamma spectrometry.  $^{235}\text{U}$  nuclei emit gamma rays with a characteristic energy of 186keV and these photons can be registered by a detector attached to pipes or cylinders containing  $\text{UF}_6$ <sup>[9a]</sup>. These detectors are used as part of Continuous Enrichment Monitors (CEMO's), which are usually placed on product header pipes and send a regular signal to an IAEA inspector. Based on the peak intensity obtained, the CEMO gives a go/no-go signal, referenced to 20% enrichment<sup>[9b]</sup>.

Although these methods of detection of enriched uranium diversion have been in standard use for a number of years, they are far from perfect. For instance, the LFUA

principle is intrusive and sometimes operators of GCEP's are reluctant to provide access to the cascade hall interior. There is also an additional problem with the CEMO system because the low pressure of the UF<sub>6</sub> gas in the cascade means that the sample geometry cannot be considered infinitely thick. Corrections for the gas pressure have to be made with the use of radioactive <sup>109</sup>Cd measurements but even these have considerable uncertainty and only lead to an approximate assay.

A new approach to diversion detection has been put forward by J Delbeke, J Howell et al at the European Commission Joint Research Centre (JRC) in Ispra, Italy which is based on continuous mass measurement of the feed, product and tails <sup>[2a]</sup>. By measuring the mass of these cylinders against time, the flow rates of each can be calculated. In normal steady-state operation, the feed rate should equal exactly the sum of the product and tails flow rates. Changes in these flow rates should indicate a new set of operational characteristics and will show up in the cumulative mass balance (explained further in the next section). This technique could become an invaluable way to monitor a GCEP with minimal intrusion. The JRC team examined 3 specific diversion scenarios, which are as follows:

- 1.) The protracted diversion of LEU from a cascade by increasing the feed flow and skimming off the excess product flow into an unmonitored station <sup>[2b] & [6a]</sup>.
- 2.) Increasing the product assay by means of increasing the separative capacity of the centrifuges (and hence the cascade) while maintaining a constant feed rate <sup>[2b]</sup>.
- 3.) Increasing the product assay above 20% (i.e. HEU) by maintaining the separative capacity but decreasing the feed rate <sup>[4a]</sup>.

In all 3 cases, their proposed Real Time Mass Evaluation System (RTMES) had a specific response which indicated the undeclared activity.

This method of continuously measuring the feed, product and feed weights could be used to detect further diversion scenarios, albeit not necessarily via the cumulative mass balance. Instead of determining the latter, it is proposed that for 2 more important scenarios outlined below, the flow data is used to detect diversions using other calculations:

- 1.) Reconfiguration of the entire plant to produce HEU efficiently, i.e. an ideal cascade.
- 2.) Initially using the cascade to produce LEU as per normal operation but then using batch recycling of the product to produce higher enrichments.

The first of these could be noticed by finding the equilibrium time of the cascade or by measuring the product to feed ratio while the second can be detected by the analysis of the successive emptying-out and filling-up of the plant. All of this will be explained in much more detail in the Experimental Methods section of the report. However before that, the next section will outline the basic properties of both individual centrifuges and entire cascades in a GCEP.

## Theory and Background

### 2.1 Basic Definitions

In any isotope separation plant, like most chemical engineering processes, there are always 3 main streams: the feed, the product and the tails (waste)<sup>[3a]</sup>. In this report, the following labeling is used for the properties of these 3 quantities:

F = feed cylinder mass	f = feed flow rate	N <sub>F</sub> = feed concentration of <sup>235</sup> U
P = product cylinder mass	p = product flow rate	N <sub>p</sub> = prod. concentration of <sup>235</sup> U
W = tails cylinder mass	w = tails flow rate	N <sub>w</sub> = tails concentration of <sup>235</sup> U

Graphs of a typical cylinder mass history can be seen below in Figures 2.1 and 2.2. In the case of the feed cylinder, the mass decreases from its maximum value to the tare value of the empty cylinder, whereas for the product and tails cylinders, the mass increases from the tare value upwards until the cylinders are full.

Fig 2.1

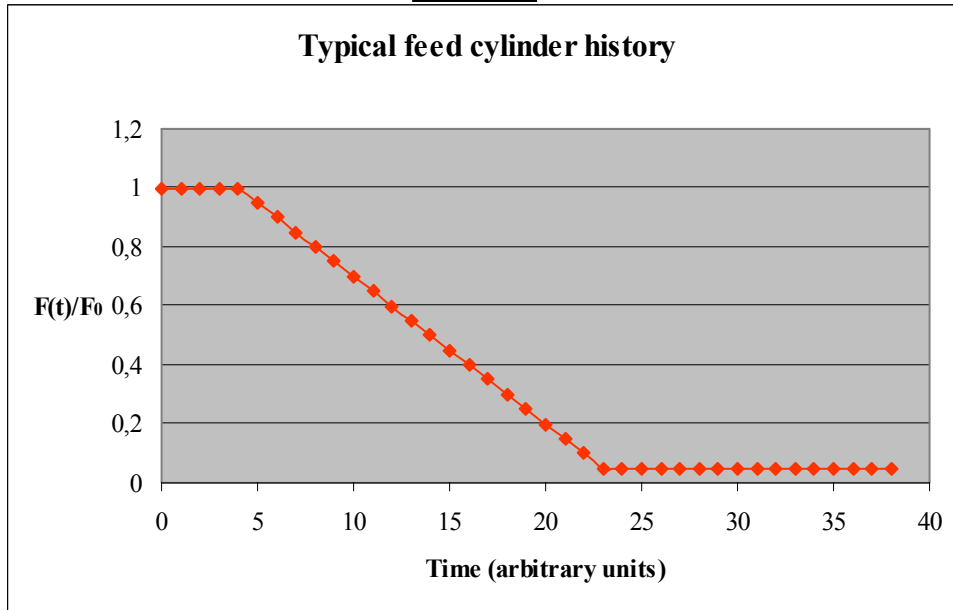
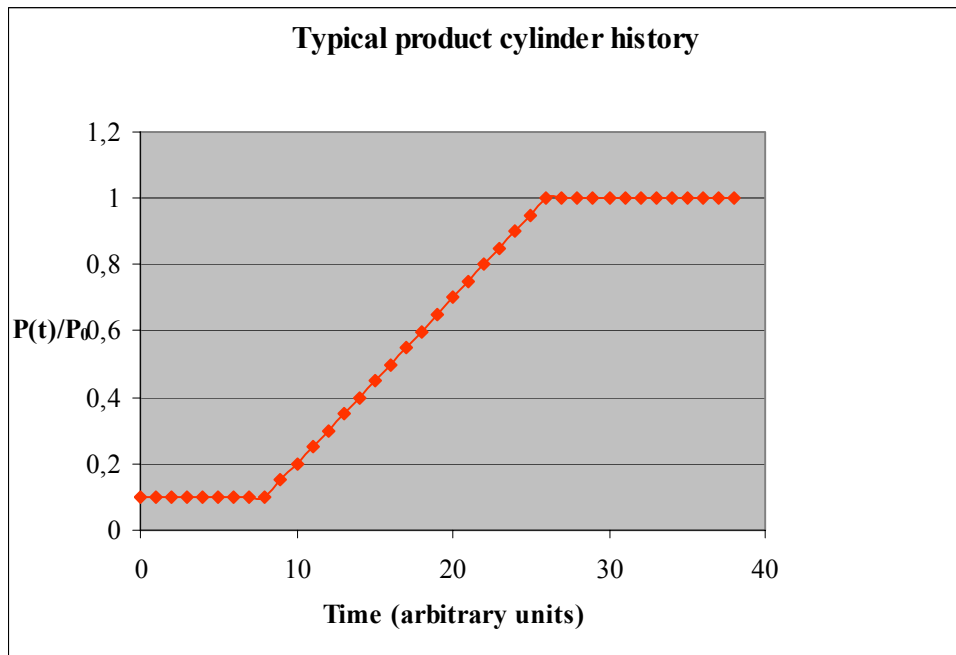


Fig 2.2



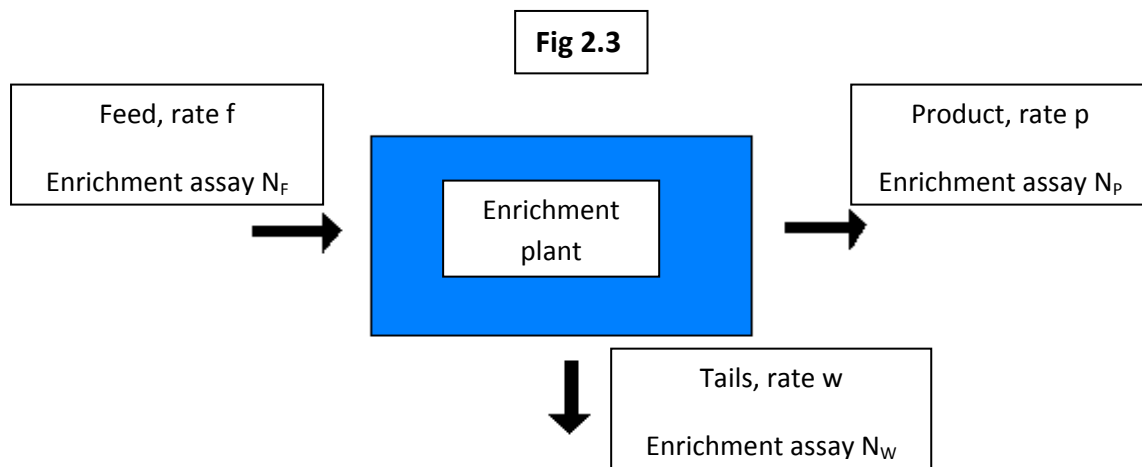
The graph for the waste (tails) cylinder against time would be very similar to that of the product cylinder, although the actual quantity of material in the tails flow will normally be greater. Using these plots, the feed rates can be calculated from the slopes:

$$f = -dF/dt$$

$$p = dP/dt$$

$$w = dW/dt$$

At the most basic level, any enrichment process can be treated in the manner shown in Figure 2.3.



In Figure 2.3, when operating in steady state conditions, the following equations must hold because of mass conservation<sup>[7a]</sup>:

$$f = p + w \quad (1) \quad \text{and} \quad fN_F = pN_P + wN_W \quad (2)$$

Using these relations, it can be shown that the following are also true, regardless of the exact enrichment process:

$$f/p = (N_P - N_W)/(N_F - N_W) \quad (3)$$

$$w/p = (N_P - N_F)/(N_F - N_W) \quad (4)$$

The inverse of equation (3) is known as the cut,  $\theta = p/f$ . Another formula that will prove to be useful later on is the "relative isotopic abundance" which is defined as follows<sup>[3a]</sup>:

$$R = N/(1-N) \quad (5)$$

In this equation,  $N$  is the enrichment assay, referred to earlier in this section.

On top of this, an important concept that has to be dealt with is that of "separative work". Ultimately, enrichment is based on altering the entropy of a system in that energy is spent on reducing the entropy of the feed. However, calculating this change in entropy is difficult and has no simple connection to the energy consumption of an element<sup>[3b]</sup>. As a result of this, the idea of separative work was developed by P.A.M. Dirac and other scientists who



helped to develop uranium enrichment during and after World War II. Separative work is a property of an element or plant that relates the rates of material flow to the energy requirements of the production unit. It is given by the following<sup>[3c]</sup>:

$$\Delta U' = p.V'(N_p) + w.V'(N_w) - f.V'(N_f) \quad (6)$$

Here,  $V'(N)$  is known as the "elementary value function" and is described by<sup>[8a]</sup>:

$$V'(N) = (2N-1) \ln(N/(1-N)) \quad (7)$$

With these simple equations in mind, it is now possible to discuss the actual operations within an enrichment facility. One final point that must be made is that the mass and flow rates are in terms of uranium metal, not uranium fluoride gas. If  $UF_6$  is the working medium then the actual mass of gas transported will be larger<sup>[3a]</sup>. The relationship between the two quantities is quite straightforward because uranium constitutes approximately 0.676 of the mass of a  $UF_6$  molecule.

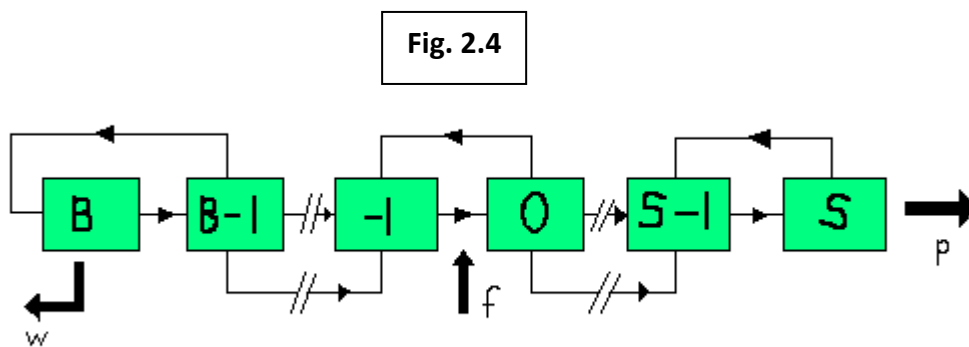
## 2.2 Cascade design

In most facilities designed to produce enriched uranium, for instance ones using gaseous diffusion, centrifugal and aerodynamic separation mechanisms, the enrichment is conducted in many individual, relatively small elements rather than one large one. In an average GCEP that usually produces LEU on a commercial scale, there are usually thousands of elements (centrifuges) within the production hall.

In designing a GCEP, it is desirable to create a plant that minimizes energy losses. In order to do this, elements are connected together in certain arrangements known as cascades. An ideal cascade is one in which there is no mixing of material with different  $N$ <sup>[3d]</sup>. Although this is a theoretical notion, it is reasonable to treat a GCEP cascade as ideal because there are so many elements that it can be constructed in an arrangement very close to the ideal situation<sup>[3e]</sup>.

Cascades are arrays of elements connected in series and in parallel. In a GCEP production hall, there may be just one cascade or there could be several small cascades that are brought into operation one-by-one as they are built. A cascade consists of a number of stages in series, where a stage refers to many elements grouped together in parallel. Figure

2.4 shows a schematic of a symmetric cascade. It points out where the feed, product and tails are introduced / withdrawn and it also shows the detailed inter-stage connections<sup>[3f]</sup>.



The part from the 0<sup>th</sup> stage to the S<sup>th</sup> stage is known as the enricher whereas the part from the -1<sup>st</sup> stage to the B<sup>th</sup> stage is called the stripper. One issue with this picture is that it does not highlight the fact that some stages handle much more material flow than others. Since an element has a limited material flow (usually about  $0.02\text{g}\cdot\text{s}^{-1}$  of  $\text{UF}_6$ <sup>[3g]</sup>), this means that more flow in a stage implies more elements in parallel. The equations governing a GCEP cascade will be presented in the "Experimental Method" section.

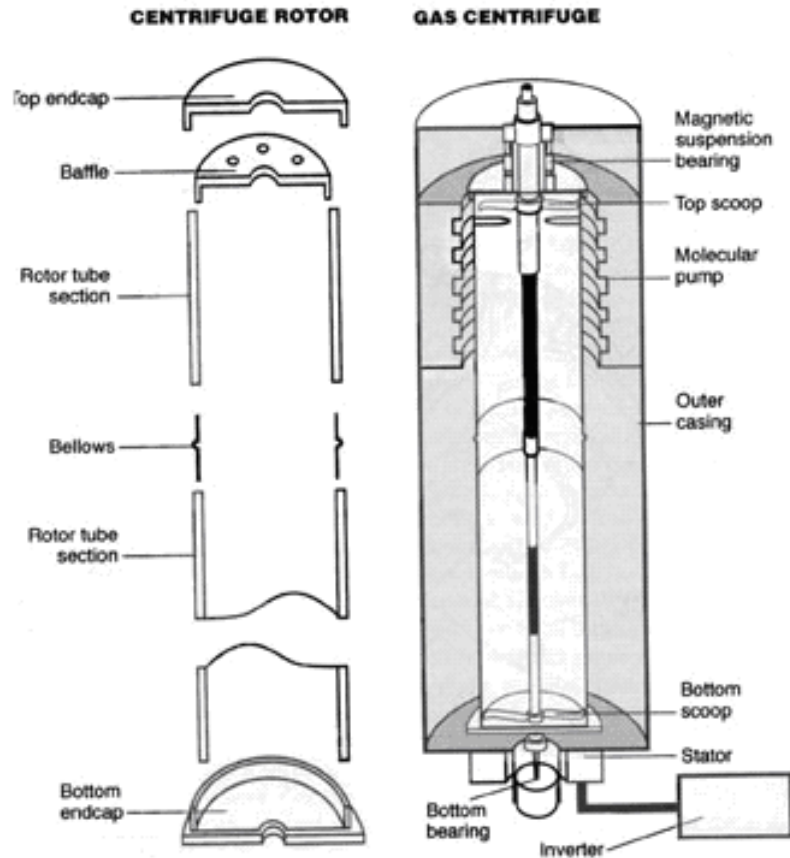
### 2.3 Centrifuge Structure

It is useful at this point to include a brief description of gas centrifuges. This is done to demonstrate its inherent advantages and limitations when employed in undeclared activities.

The idea that centrifugal fields could be used to separate isotopes was first advanced by Lindemann and Aston in 1919. They calculated that the Earth's gravitational field naturally separates out isotopes of neon to a measurable extent at different heights in the atmosphere.

This degree of isotope separation is greatly increased in a gas centrifuge because of the much larger centrifugal fields present. A centrifuge is simply a hollow, vertical tube made of very high tensile materials set in motion about its central axis at high angular velocity (see Figure 2.5).

Fig. 2.5



The angular velocity is so large that a special assembly consisting of a magnetic bearing at the top and a thin needle-point bearing at the bottom, together with a high vacuum chamber are required to prevent the centrifuges from disintegrating during operation.

In the absence of the 2 scoops, i.e. in a concurrent centrifuge, the ideal radial separation factor is given by the expression<sup>[8b]</sup>:

$$\alpha_0(r) = \exp [ -(M_1 - M_2)(\omega r)^2 / (2RT) ] \quad (8)$$

where M is molecular mass, T is temperature

$\omega$  is angular velocity, R is the universal gas constant

r is distance from the central axis

This expression does not give the exact separation factor as most of the gas stays in a thin layer near the inner wall surface. The actual separation factor in a centrifuge can be vastly improved by introducing a well-ordered countercurrent flow and this is performed in almost

all modern centrifuges with the use of specially-designed scoops, such as those in the one in Figure 2.5. By accurately modelling and predicting gas flow within the element, separation factors of 1.5 to 1.7 are commonly achievable today.

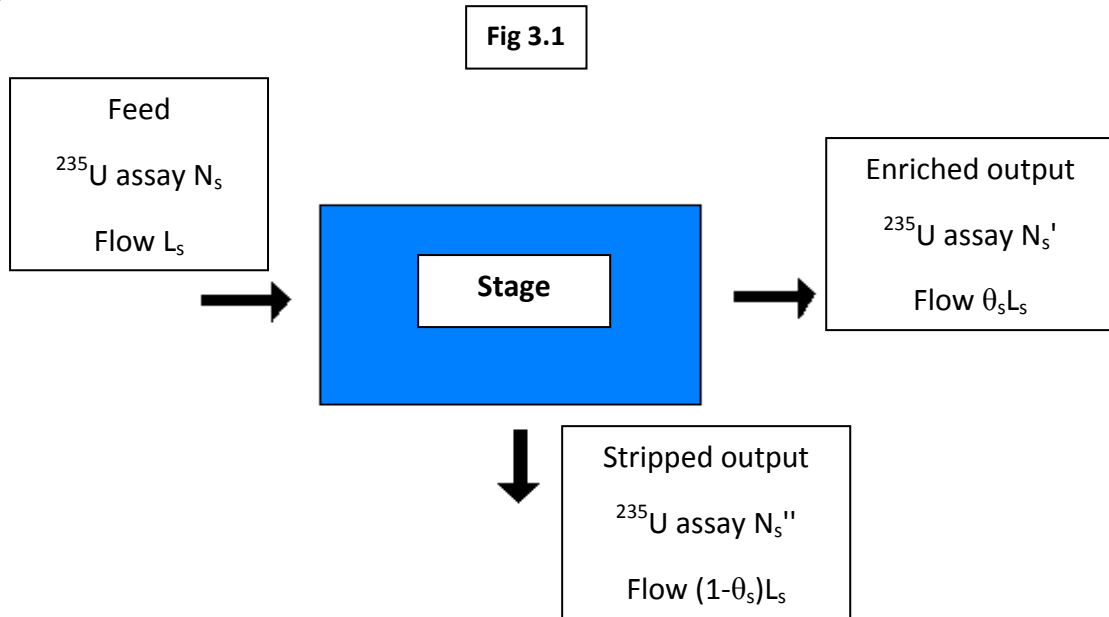
With regard to production of HEU or excess production of LEU, the gas centrifuge has several advantages from the plant operator's viewpoint. It has a high separation factor, a short hold-up time and quite low power consumption compared to other methods<sup>[3e]</sup>, especially the main competing method of gaseous diffusion. These properties mean that an operator, in a "break-out" scenario whereby undeclared operation occurs, could alter the plant quickly and easily. Another distinct advantage for the operator is that cascades can be built inside small, easily-concealed spaces so clandestine facilities could be constructed to contain them<sup>[5a]</sup>.

Nevertheless, there are a few disadvantages too. In order to reach the optimum angular velocities, the centrifuges have to be accelerated through so-called critical frequencies, at which the machines are susceptible to destructive internal vibrations<sup>[3h]</sup>. This means that it is not an easy task to stop and restart them. Yet another problem with centrifuges is the low throughput per element, which means that huge numbers of centrifuges are needed to get a decent overall production rate. As a result of the latter, while it may be straightforward to construct a small cascade in a short time, it can take many years to build an entire plant.

## Experimental Method

### 3.1 Modelling a cascade

In order to predict the response of the CMMS, it is necessary to look at the processes that go on inside a cascade. This in turn requires use of equations describing the material flow.



The above diagram, Figure 3.1, is quite similar to Figure 2.3. It shows the inputs and outputs of a single stage rather than an entire plant (the subscript  $s$  refers to stage number). Earlier on, in Section 2.3, the term "separation factor" was mentioned and for one stage, it is defined as follows<sup>[3a]</sup>:

$$\alpha = R_s' / R_s'' \quad (9)$$

Another closely-related quantity, the "enrichment factor" is defined as<sup>[8c]</sup>:

$$\beta = R_s' / R_s \quad (10)$$

In fact, for an ideal cascade, it can be demonstrated that  $\beta = \sqrt{\alpha}$ . As a result of (10), the relative isotopic abundance of the final product is<sup>[3i]</sup>:

$$R_p = \beta^{S+1} R_f \quad (11)$$

Also, that of the final tails flow is given by:

$$R_W = \beta^B R_F \quad (12)$$

Conservation of matter dictates that:

$$N_s = \theta_s N_s' + (1-\theta_s) N_s'' \quad (13)$$

From (7) and (8), an expression for the stage cut is obtained:

$$\theta_s = (1+\beta R_s)/((\alpha+1)(1+R_s)) \quad (14)$$

By conservation of mass, it can be proven that the stage material flow,  $L_s$ , in the enriching and stripping sections are given by<sup>[8d]</sup> & <sup>[8e]</sup>:

Enricher / Rectifier:  $L_s = p((\beta+1)/(\beta-1))(N_p-N_s)/(N_s(1-N_s))$

$$= pN_p((\beta+1)/(\beta-1))[1 - \beta^{s-S-1} + (1/R_F)(\beta^{-s} - \beta^{-S-1})] \quad (15)$$

Stripper:  $L_s = w((\beta+1)/(\beta-1))(N_s-N_w)/(N_s(1-N_s))$

$$= wN_w((\beta+1)/(\beta-1))[(\beta^{B+S+1} - 1) + (1/R_F)(\beta^{B+1} - \beta^{-S})] \quad (16)$$

Also, the total flow in both sections is calculated to be:

Enricher:  $L_{enr} = \sum_{s=0}^S (L_s)$

$$= p((\beta+1)/(\beta-1))[(2N_p-1)\ln(R_p/R_F)/(\ln \beta) + \{(\beta-(\beta+1)N_F)/(\beta-1)\}\{(N_p-N_F)/(N_F(1-N_F))\}] \quad (17)$$

Stripper:  $L_{str} = \sum_{s=-1}^B (L_s)$

$$= w((\beta+1)/(\beta-1))[(1-2N_w)\ln(R_F/R_w)/(\ln \beta) - \{(\beta-(\beta+1)N_F)/(\beta-1)\}\{(N_F-N_w)/(N_F(1-N_F))\}] \quad (18)$$

All of these formulae govern the flow in an ideal cascade for a set of feed, product and tails enrichment levels. The flow is used to calculate the separative capacity of the cascade which determines the number of centrifuges required. In a given plant, the total material flow will remain approximately constant even if the cascade is reconfigured.

In Section 2.1, separative capacity was calculated with the aid of the concept of a value function. This only gives a more or less precise number for the separative capacity when the separation factor is very close to unity. It turns out that although more

complicated equations are needed for the value function of cascades with a large enrichment factor per stage<sup>[8f]</sup>, when the total flow from both the enricher and the stripper are added up, the extra terms in the value function cancel out and a relatively simple formula holds for the total flow<sup>[8e]</sup>:

$$L_{\text{tot}} = (L_{\text{enr}} + L_{\text{str}}) = (\beta+1)/((\beta-1)\ln\beta) \Delta U' \quad (19)$$

With these equations in mind, it is possible to imitate the behaviour of a GCEP cascade. As the stage enrichment factor  $\beta$  is constant and the total flow in a plant is limited by the fixed number of centrifuges, then  $\Delta U'$  will remain the same regardless of cascade reconfiguration, provided it is approximately ideal. In the case where the cascade is reconnected to create an ideal cascade for the production of HEU from NatU, it is assumed that an enrichment of 90% is desired (this is weapons-grade material). In Table 4.1, the data for 2 types of cascade are given, along with graphs of the material flow in each stage. The first depicts a cascade performing NatU→5%LEU while the second performs the operation NatU→90%HEU. The tails assay is set at 0.2% and their separative capacity is approximately 1000tSWU.yr<sup>-1</sup>.

**Table 4.1**

<b>f (in t.yr<sup>-1</sup>)</b>	<b>p (in t.yr<sup>-1</sup>)</b>	<b>w(in t.yr<sup>-1</sup>)</b>
1100.00	119.17	980.83
<b>f (in kg.s<sup>-1</sup>)</b>	<b>p (in kg.s<sup>-1</sup>)</b>	<b>w(in kg.s<sup>-1</sup>)</b>
0.035	0.004	0.031
<b>N<sub>F</sub></b>	<b>N<sub>P</sub></b>	<b>N<sub>W</sub></b>
0.0072	0.0500	0.002
<b>ΔU' (tSWU.yr<sup>-1</sup>)</b>		<b>θ<sub>tot</sub></b>
1043.9		0.108
<b>α</b>	<b>β</b>	<b># Elements</b>
1.6	1.26	89013
<b>S</b>	<b>B</b>	<b>Stages</b>
7.4	5.4	13.9

Fig 3.2

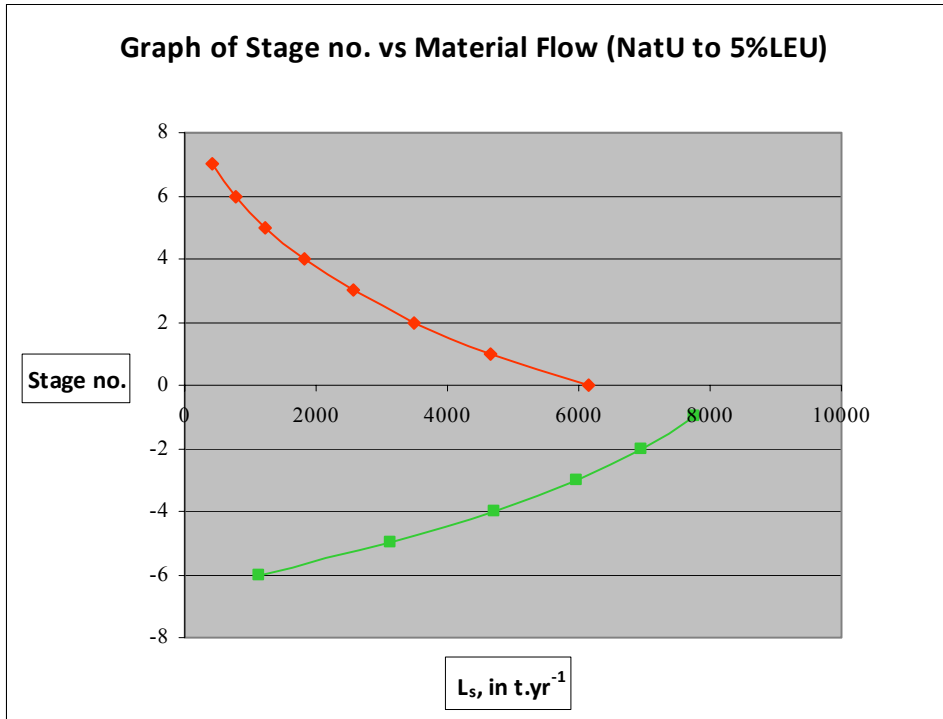
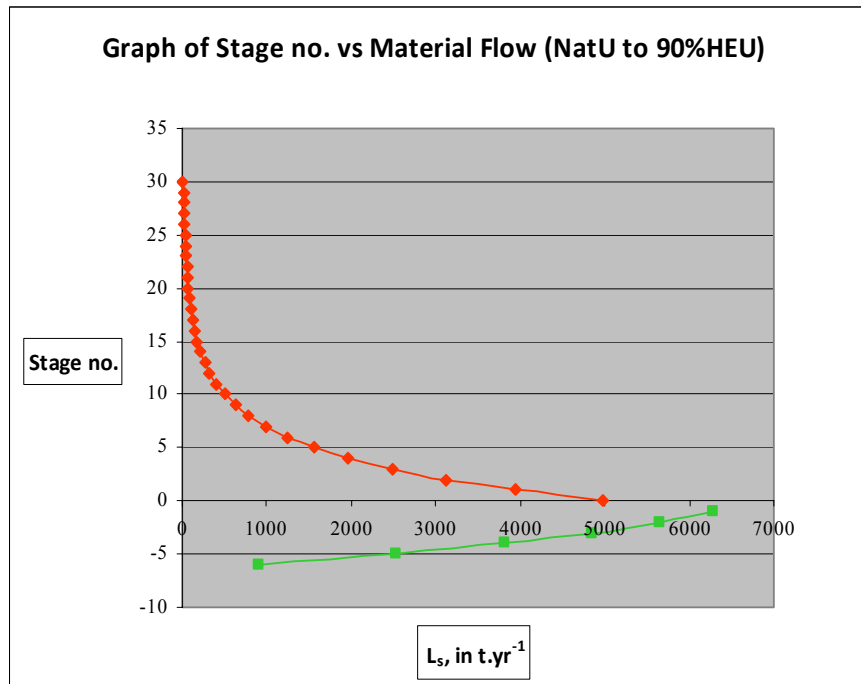


Table 4.2

<b>f (in t.yr<sup>-1</sup>)</b>	<b>p (in t.yr<sup>-1</sup>)</b>	<b>w(in t.yr<sup>-1</sup>)</b>
800	4.6	795.4
<b>f (in kg.s<sup>-1</sup>)</b>	<b>p (in kg.s<sup>-1</sup>)</b>	<b>w(in kg.s<sup>-1</sup>)</b>
0.025	0.0001	0.025
<b>N<sub>f</sub></b>	<b>N<sub>p</sub></b>	<b>N<sub>w</sub></b>
0.0072	0.9000	0.0020
<b>ΔU' in t SWU yr<sup>-1</sup></b>		<b>θ<sub>tot</sub></b>
1045		0.006
<b>α</b>	<b>β</b>	<b>No. Elements</b>
1.6	1.26	89131
<b>S</b>	<b>B</b>	<b>Stages</b>
29.3	5.5	35.8



Fig 3.3



As for the batch recycling system, it is postulated that the plant produces declared 4.2% LEU at first, then this enriched product is placed in the feed stations and the plant is run again. This process is performed twice more to yield a final product of 90.6%. There are 2 main strategies that could be employed by the operator: they could run the plant for a fixed amount of time (e.g. 6 months) for each enrichment step or they could run it for successively shorter periods of time to maintain the appearance of a constant feed rate. Both of these situations are investigated.

### 3.2 Validity of GCEP models

In this short section, the choice of parameters given above is explained. First of all, the assumption that the operator would choose to create an ideal cascade in each case is based on the fact that it is by far the most efficient way to yield a product based on the assays of the feed, product and tails. It is possible to produce HEU using the unchanged NatU→LEU cascade by reducing the feed rate and indeed, this case has already been examined by J Delbeke et al<sup>[4a]</sup>. However, this strategy is not only wasteful of huge amounts of energy but it would also be much more time-consuming.

Regarding the batch recycling system, it is assumed that the cascade would need to be emptied and cleaned out of material after each cycle, as this prevents dilution of LEU

feed with NatU feed. It is assumed that at maximum capacity levels for the plant, the feed is kept at  $1200 \text{ t.yr}^{-1}$  as this leads to a  $\Delta U'$  of just over  $1000 \text{ tSWU.yr}^{-1}$  for a site producing LEU from NatU, a typical value for the separative capacity of a commercial facility.

### 3.3 Model of CMMS

As proposed by J Delbeke et al, the RTMES is in principle able to detect 3 diversion scenarios (outlined in the Introduction). The reason the RTMES is able to do this is because it is capable of registering changes in the inventory of a plant. In non-steady state conditions, the following equation holds:

$$f(t) - p(t) - w(t) = \Delta I(t) \quad (20)$$

Here,  $\Delta I(t)$  refers to the change in inventory of a plant and the other symbols are as before. If the feed is increased at a certain rate for example, this will increase the inventory of the plant for a short time and will manifest itself in the cumulative mass balance:

$$\Delta x = \int_{t=0}^T \Delta I(t) \quad (21)$$

In the above formula, the cumulative mass balance is calculated for a period of time from  $t=0$  to  $t=T$ . Unfortunately, this is limited to situations where the operator either siphons off product into an unmonitored product station or where the enrichment is suddenly switched from one to another. The aim of this study is to analyze the mass data in situations for which the cumulative mass balance is inappropriate. The only difference between the CMMS outlined in this report and the RTMES is the analysis of the data.

For the cascade reconfiguration, 2 possible options are explored. The first option is based on measuring the equilibrium time of the cascade when it is started up. At the beginning of the operation of a GCEP, the cascade is filled with feed at a constant rate. The system is then run in total reflux, which means that no feed, product or tails are withdrawn. Then, once the concentrations at the top and bottom of the cascade reach the desired level at  $t_2$  and  $t_1$  respectively, product and tails are withdrawn slowly. The feed, product and tails flow rates are then gradually increased to the full steady state rates. The exact equations that govern the start-up procedure are complex and beyond the scope of this study. All the

same, Figures 3.2 to 3.4 offer a reasonably fair portrayal of the feed, tails and product flow rates at the beginning of plant operation<sup>[7b]</sup>.

Fig 3.4

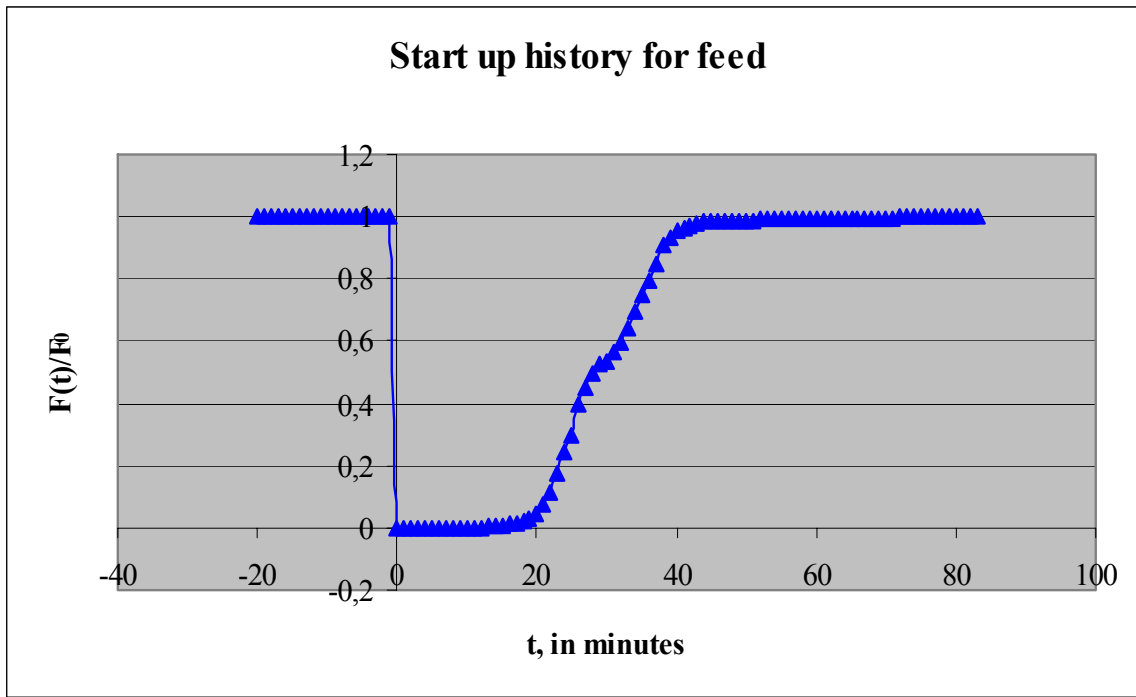


Fig 3.5

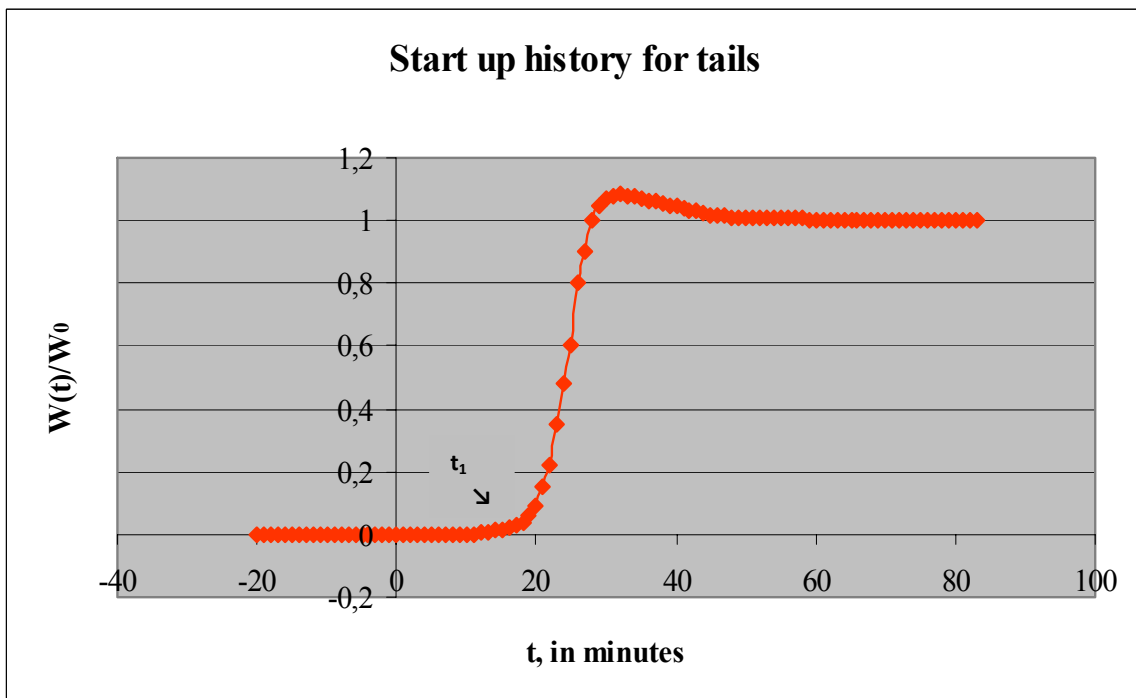
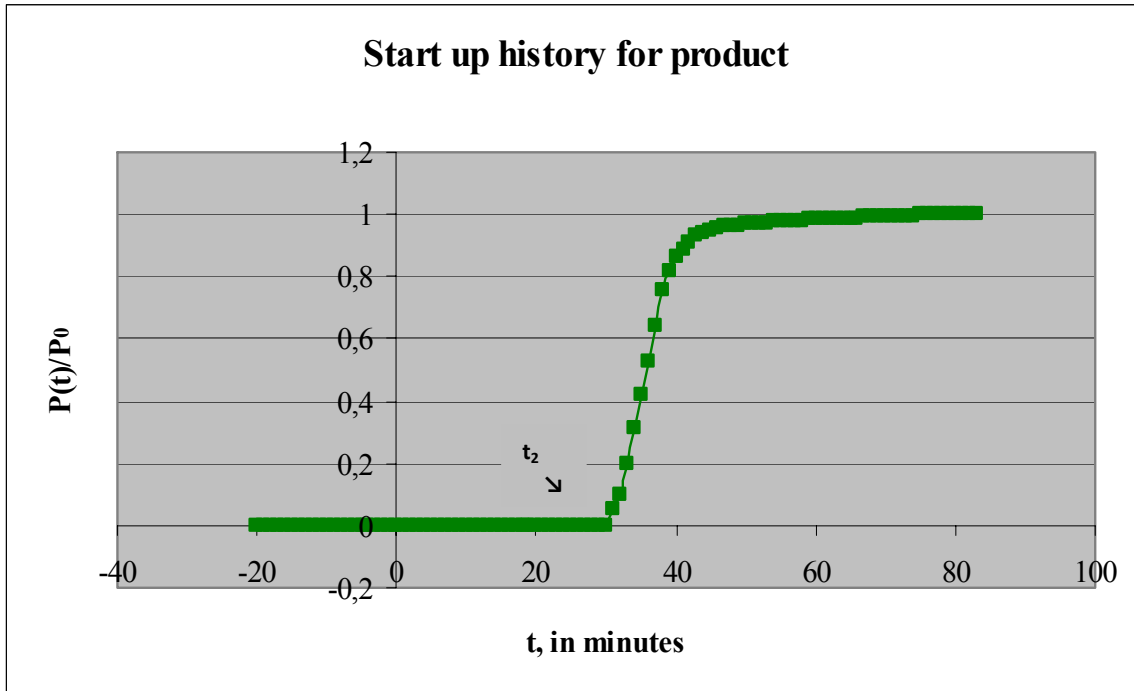


Fig 3.6



It can be proven that the equilibrium time,  $t_p$ , can be calculated quite accurately using this formula<sup>[7c] & [8g]</sup>:

$$t_p = [(\beta+1)t_h/((\beta-1)\ln\beta)] E(N_p, N_F) \quad (22)$$

$$\text{where } E(N_p, N_F) = [-2 + (N_p - 2N_p N_F + N_F) \ln(R_p/R_F)/(N_p - N_F)]$$

If it is assumed that  $t_h$ , the hold-up time of a centrifuge and  $\beta$ , the enrichment factor can be estimated within a low margin of error, measuring the equilibrium time of a cascade could be a good indicator of whether or not a cascade is producing HEU from NatU.

The definition of  $t_p$  is the time from the initial start-up up to the point at which the product flow rate reaches half its asymptotic value<sup>[8h]</sup>. The equilibrium time could be estimated by measuring  $t_2$ , the time elapsed between the cessation of initial feed flow (i.e.  $t=0$ ) and the time when product begins to be withdrawn at the top of the cascade, which is almost equal to  $t_p$ . If more accuracy is desired,  $t_p$  could be calculated exactly by noting  $P_0$  and determining the time when  $P(t) = 1/2 P_0$ .

The second option for detection of reconfiguration would be to simply measure the overall cut  $\theta$  of the cascade. As equation (3) demonstrates, this is dependent on all 3 assays of the inputs and outputs.

$$\theta = p/f = (N_F - N_W)/(N_P - N_W) \quad (23)$$

Even if the operator tried to adapt the tails assay to try and conceal the reconfiguration, it would be almost impossible to fully hide the effect on the cut without incurring huge losses of energy. The measurement of the cut would show a marked difference between a plant processing NatU→LEU from one processing NatU→HEU. One situation to be careful of would be processing LEU→HEU, as  $\theta$  for this is very similar to that for NatU→LEU. However, the CMMS could be utilized to detect this via criticality considerations. Different types of container have to be used for the various levels of enriched material in order to prevent the contents from going critical and starting a chain reaction within. Table 3.1 gives a list of containers used by commercial facilities for containment of UF<sub>6</sub> [9c].

**Table 3.3**

<b>Cylinder Model</b>	<b>Tare mass(kg)</b>	<b>Max. enrichment</b>	<b>Max. Fill Limit(kg of UF<sub>6</sub>)</b>
1S	0.79	100%	0.45
S	1.91	100%	2.22
5A & 5B	24.95	100%	24.95
8A	54.43	12.5%	115.67
12B	84	5%	208.7
30B	635	5% †	2277
48G	1179	1%	12701
48X	2041	4.5% †	9539
48Y	2359	4.5% †	12501

Note: † These enrichment levels are only achievable with the use of a moderator. Without this, the maximum safe level of enrichment is 1%.

If LEU was being used as feed, as it is usually stored in 30B cylinders, the total mass of a feed station loaded with this type of container would be much lower than one loaded

with the 48Y cylinder that is usually used to store NatU fluoride gas. Similarly, if HEU was being produced, it would have to be stored in very small containers such as the 5A, 2S or 1S cylinders and this would be noticeable in the total mass of the product station. Surveillance of the cell stations could also ensure that only 48Y cylinders are being used for the feed and tails and 30B ones used for the product.

For the batch recycling mechanism, as mentioned earlier, there are 2 strategies that could be used to partially obscure diversion are examined. In both of them, the CMMS history of all the feed, product and tails flows should indicate whether or not recycling is occurring.

## Results

### 4.1 CMMS detection of cascade reconfiguration

#### 1.) [ Measurement of equilibrium time] Table of $t_p$ for ideal cascades

**Table 4.1**

	$\alpha$	$\beta$	$t_h$	$\delta\beta$	$\delta t_h$		
	1,6	1,26	13	0,05	1		
$N_F$	$N_P$	$R_F$	$R_P$	$\ln(R_P/R_F)$	$E(N_P, N_F)$	$t_p$ (in mins)	$\delta t_p$ (in mins)
0,0072	0,03	0,007	0,031	1,45	0,34	3	1
0,0072	0,05	0,007	0,053	1,98	0,62	5	2
0,0072	0,25	0,007	0,333	3,83	2,00	16	6
0,0072	0,5	0,007	1,000	4,93	3,00	24	10
0,0072	0,9	0,007	9,000	7,12	5,14	40	17
0,03	0,25	0,031	0,333	2,38	0,86	7	3
0,03	0,5	0,031	1,000	3,48	1,70	13	6
0,03	0,9	0,031	9,000	5,67	3,71	29	12
0,05	0,25	0,053	0,333	1,85	0,54	4	2
0,05	0,5	0,053	1,000	2,94	1,27	10	4
0,05	0,9	0,053	9,000	5,14	3,20	25	10
0,25	0,5	0,333	1,000	1,10	0,20	2	1
0,25	0,9	0,333	9,000	3,30	1,55	12	5
0,5	0,9	1,000	9,000	2,20	0,75	6	2

Note that the values for time given are decimal units, not minutes and seconds. This table of equilibrium times demonstrates that even with the large margin of error (the calculation of this is discussed in Section 4.3), it is quite noticeable when a plant is producing HEU from NatU.

2.) [Measurement of the cut]

The values of cuts for different feed, product and tails assays are listed in Table 4.2.

**Table 4.2**

$N_F$	$N_P$	$N_W$	$\theta$
0.0072	0.03	0.0001	0.24
0.0072	0.03	0.0005	0.23
0.0072	0.03	0.001	0.21
0.0072	0.03	0.002	0.19
0.0072	0.03	0.003	0.16
0.0072	0.03	0.004	0.12
0.0072	0.03	0.005	0.09
0.0072	0.03	0.006	0.05
0.0072	0.03	0.007	0.01

$N_F$	$N_P$	$N_W$	$\theta$
0.0072	0.2	0.0001	0.036
0.0072	0.2	0.0005	0.034
0.0072	0.2	0.001	0.031
0.0072	0.2	0.002	0.026
0.0072	0.2	0.003	0.021
0.0072	0.2	0.004	0.016
0.0072	0.2	0.005	0.011
0.0072	0.2	0.006	0.006
0.0072	0.2	0.007	0.001

$N_F$	$N_P$	$N_W$	$\theta$
0.0072	0.04	0.0001	0.18
0.0072	0.04	0.0005	0.17
0.0072	0.04	0.001	0.16
0.0072	0.04	0.002	0.14
0.0072	0.04	0.003	0.11
0.0072	0.04	0.004	0.09
0.0072	0.04	0.005	0.06
0.0072	0.04	0.006	0.04
0.0072	0.04	0.007	0.01

$N_F$	$N_P$	$N_W$	$\theta$
0.0072	0.5	0.0001	0.014
0.0072	0.5	0.0005	0.013
0.0072	0.5	0.001	0.012
0.0072	0.5	0.002	0.010
0.0072	0.5	0.003	0.008
0.0072	0.5	0.004	0.006
0.0072	0.5	0.005	0.004
0.0072	0.5	0.006	0.002
0.0072	0.5	0.007	0.0004

$N_F$	$N_P$	$N_W$	$\theta$
0.0072	0.05	0.0001	0.14
0.0072	0.05	0.0005	0.14
0.0072	0.05	0.001	0.13
0.0072	0.05	0.002	0.11
0.0072	0.05	0.003	0.09
0.0072	0.05	0.004	0.07
0.0072	0.05	0.005	0.05
0.0072	0.05	0.006	0.03
0.0072	0.05	0.007	0.005

$N_F$	$N_P$	$N_W$	$\theta$
0.0072	0.9	0.0001	0.008
0.0072	0.9	0.0005	0.007
0.0072	0.9	0.001	0.007
0.0072	0.9	0.002	0.006
0.0072	0.9	0.003	0.005
0.0072	0.9	0.004	0.004
0.0072	0.9	0.005	0.002
0.0072	0.9	0.006	0.001
0.0072	0.9	0.007	0.0002

As the table points out, the cut is a number that can vary a lot with the value of the tails enrichment and based purely on the numbers above, it appears as though the production of HEU could be hidden by tailoring the tails enrichment. For instance, the operation of the plant going from NatU→4%LEU with a tails assay of 0.7% has a similar  $\theta$  as for the case of NatU→90%HEU with a tails assay of 0.01%. However, this does not take into account the energy requirements of a facility and this is a factor that needs to be considered if a realistic plant is to be imitated. In short, it is unlikely that this could occur as the former operation

would waste large amounts of uranium while the latter would require a massive stripping section. To get an idea of how unrealistic a tails assay of 0.01% is, the stripping section would have to have 18 stages instead of the norm of 5 or 6 stages. As a result the stripper would demand a larger amount of energy than the more important enriching section. Generally, the tails enrichment is kept at 0.2% or 0.3% but the operator might try to vary this to 0.1% at minimum or 0.5% maximum.

#### 4.2 CMMS Detection of Batch Recycling

If batch recycling occurs in an unchanged plant designed to product LEU from NatU, the number of stages in the enricher and the stripper (i.e. S and B) will both remain the same. From equations (11) and (12), it is clear that if S and B remain unchanged, then the ratios  $R_p/R_f$  and  $R_w/R_f$  will be constant. This fact can be used to predict the number of cycles that would need to occur for weapons grade material to be produced. The following table gives the various  $^{235}\text{U}$  assays of the feed, product and tails for each of the 4 steps in the batch recycling system which uses a plant originally designed to product 4.2% LEU from 0.72% NatU feed and tails of 0.2%.

**Table 4.3**

	$N_f$	$N_p$	$N_w$	$\theta$
<b>Cycle 1</b>	0.0072	0.042	0.002	0.13
<b>Cycle 2</b>	0.042	0.209	0.012	0.152
<b>Cycle 3</b>	0.209	0.616	0.068	0.257
<b>Cycle 4</b>	0.616	0.906	0.307	0.516

Thus the final product has an assay of 90.6%  $^{235}\text{U}$ . As the product is always less than the feed, the amount of feed material available for each cycle decreases. For instance, if the initial tranche of NatU feedstock is 600 tonnes and this is processed in 6 month cycles, then the table below gives the mass of feed, product and tails in each step. In Table 4.4,  $\Delta F$ ,  $\Delta P$  and  $\Delta W$  refer to the total mass of uranium put in or taken out for the entire 6 months. As it involves subtraction of the masses of cylinders, the tare mass of the cylinders will automatically cancel out.



Table 4.4

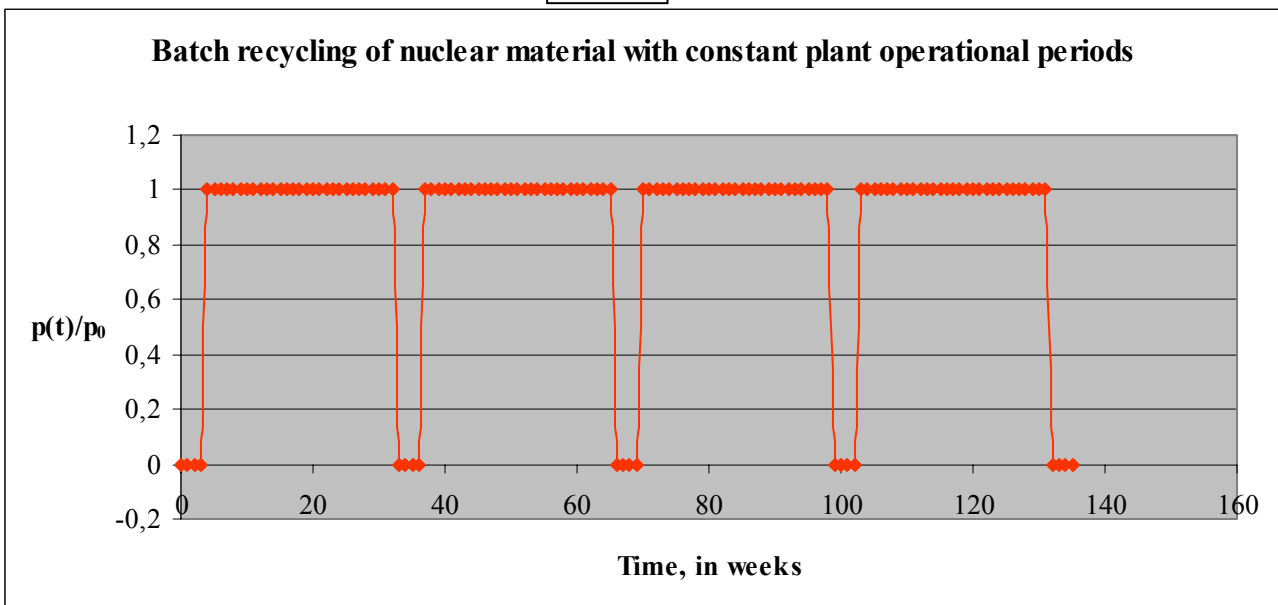
	$\Delta F$ (in kg)	$\Delta P$ (in kg)	$\Delta W$ (in kg)
<b>Cycle 1</b>	600000	78000	522000
<b>Cycle 2</b>	78000	11856	66144
<b>Cycle 3</b>	11856	3047	8809
<b>Cycle 4</b>	3047	1572	1475

So, after 4 cycles of production, a final product of 1572 kg of 90.6% HEU would be produced. It is quite possible that the operator would then process the tails from Cycles 3 and 4 as these are relatively enriched materials, although this is not explored in the report.

(1) [Batch recycling via plant operation for fixed periods of time]

In Figure 4.1, it is assumed that the plant will operate for 6 months at a time, as this would allow appreciable amounts of feed to be processed. The equilibrium time does not appear on the time graph as it is negligible compared to the operation time of the plant (the equilibrium time of the enriching section is much less than a day for any ideal plant).

Fig 4.1



The plant would run for 26 weeks at a time with 4 weeks in between to allow for emptying the cascade of material and possibly stopping the centrifuges to clean them out. The equilibrium time will remain roughly constant. However, this recycling routine could be detected by measuring the total mass of product, feed and tails being taken in or out. In the above diagram,  $p_0$  is the steady state value flow each step, which is different for each one as

$p_0$  for the first 6 month-long operation is larger than that for the second 6 month-long operation.

**Table 4.5**

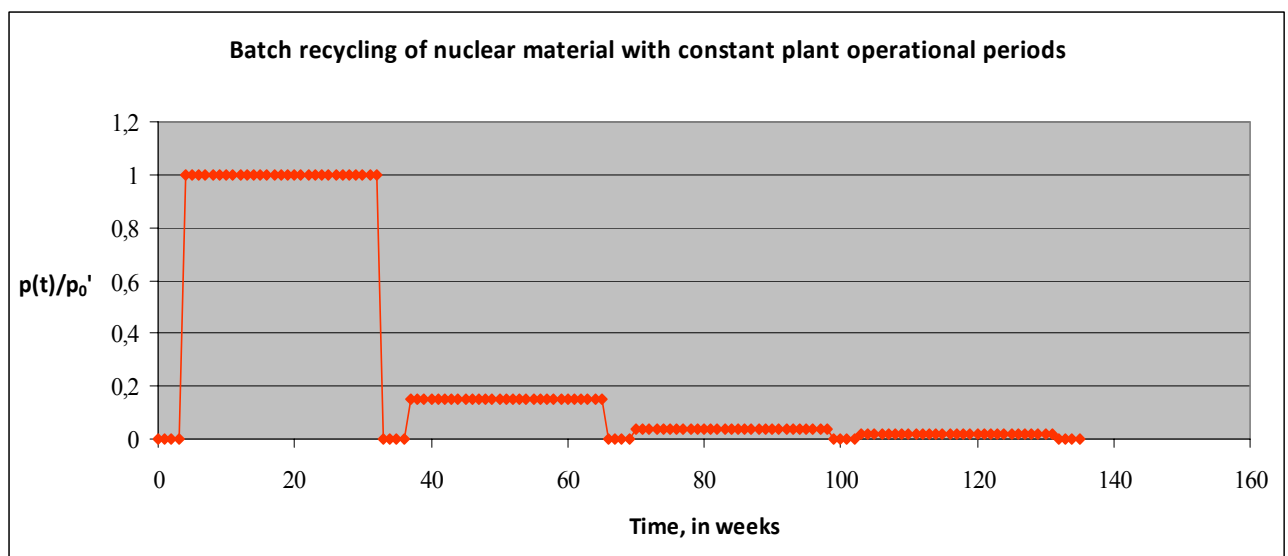
	$f$ (in $\text{kg}\cdot\text{s}^{-1}$ )	$p$ (in $\text{kg}\cdot\text{s}^{-1}$ )	$w$ (in $\text{kg}\cdot\text{s}^{-1}$ )
<b>Cycle 1</b>	38.03	4.94	33.09
<b>Cycle 2</b>	4.94	0.75	4.19
<b>Cycle 3</b>	0.75	0.193	0.557
<b>Cycle 4</b>	0.193	0.10	0.093

If  $p_0'$  refers to the initial steady state flow, then the recycling operation would be visible in a long term history of the product flow, listed in Table 4.5 and plotted as a graph in Figure 4.2.

**Table 4.5**

	$f(t)/f_0'$	$p(t)/p_0'$	$w(t)/w_0'$
<b>Cycle 1</b>	1	1	1
<b>Cycle 2</b>	0.13	0.152	0.127
<b>Cycle 3</b>	0.020	0.039	0.017
<b>Cycle 4</b>	0.005	0.02	0.003

**Fig 4.2**



The only way to make this system work would be to divide the plant into a number of separate cascades. In the first step, all of the cascades would be used to process the initial feedstock while in the second step, some of the cascades would not be required as the level of feedstock is lower than that for the first step. In the fourth step, only a few cascades would be needed to produce the final batch of high-grade HEU.

(2) [Batch recycling via constant flow rates for each step]

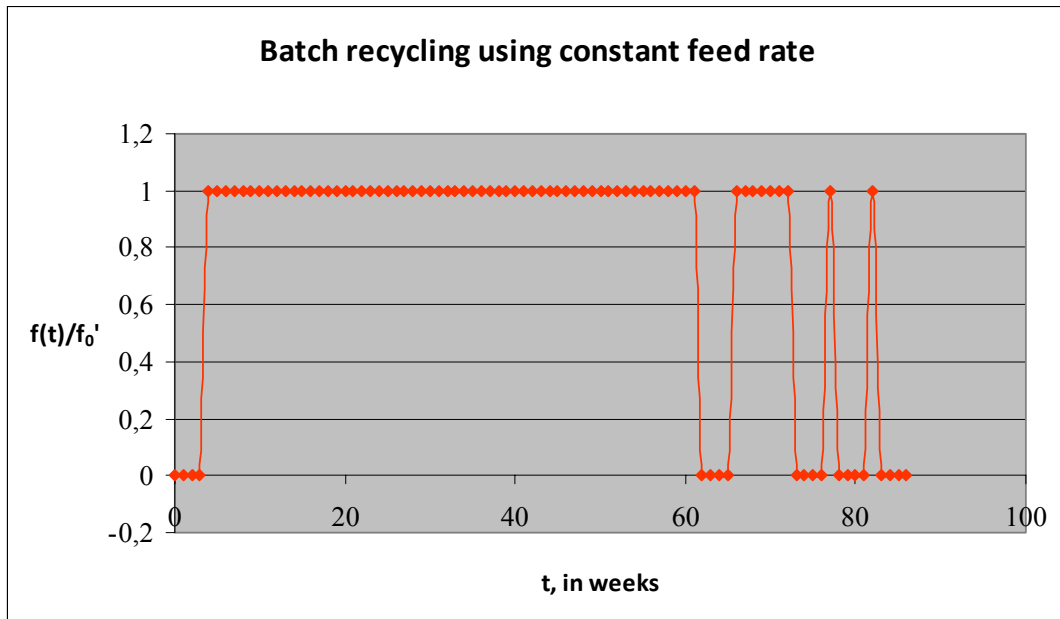
The previous strategy has a number of disadvantages, most notably the fact that the full capacity of the plant is not being used in the second, third and fourth cycles. An alternative strategy to the one above is to use the entire plant to process the initial tranche of feedstock for a certain length of time such that the feed rate remains constant in each step, while the product and tails rates would stay approximately fixed. The implication of this is that the plant would have to be operated for a lengthy period at first and then followed by successively shorter periods of operation (see Table 4.5). It is postulated that the plant would initially produce 4.2%LEU for one year.

**Table 4.5**

	<b>Operational time</b>
<b>Cycle 1</b>	52 weeks
<b>Cycle 2</b>	6.76 weeks
<b>Cycle 3</b>	1.03 weeks
<b>Cycle 4</b>	1.85 days

By measuring this, it would be possible to show that an operator is using the product from one step as the feed for the next step, as Figure 4.3 demonstrates.

Fig 4.3



This system is a slightly more efficient scenario as it uses all of the plant's separative capacity without having to reconfigure it or change the number of centrifuges being used. However, it is very easy to detect using the CMMS as the plant would have a characteristic signature for the feed flow history. One thing to note about Figure 4.3 is that it does not show the initial filling of the plant with feed before main plant operation, although this only takes a short time (of the order of minutes), so it can be neglected. The product and tails histories would show a similar pattern.

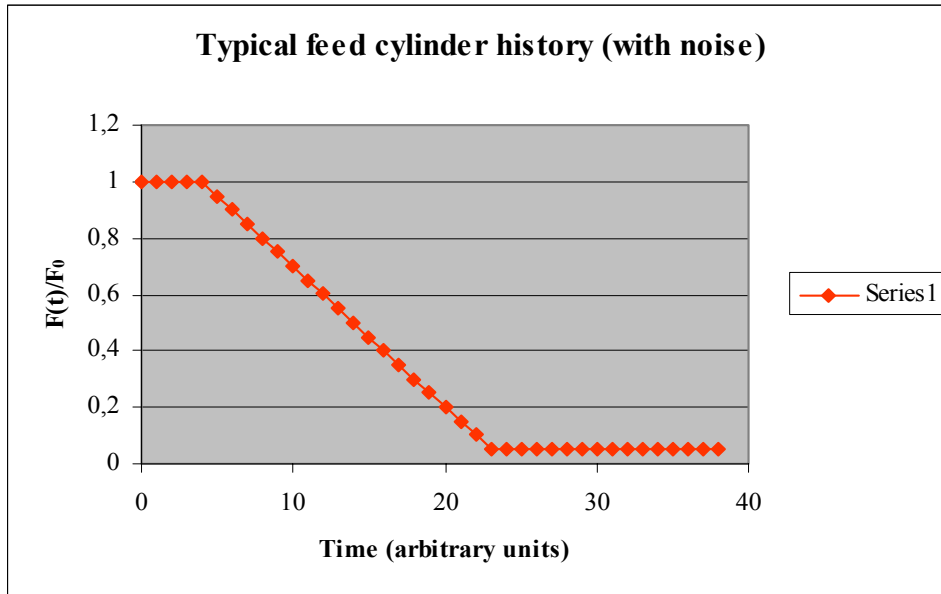
Another means of confirming use of a batch recycling scheme is the gradual increase in the cut for each cycle, which can also be measured by the CMMS.

#### 4.3 Error Analysis

As this is a theoretical study as opposed to an actual experimental one, errors have not yet been dealt with fully so as not to obscure the analysis. Obviously though, in a real GCEP, sources of uncertainty will be present and the purpose of this section is to identify these potential sources of error and to try and predict their effects on the data.

One source of error is the load cell's accuracy, which is mostly due to non-linearity and hysteresis, and is typically within  $\pm 0.1\%$ . To highlight the effect of this noise on the CMMS data, Figure 2.1 is shown with artificial noise of this order is generated by a pseudo-random algorithm on Microsoft Excel within this range.

Fig 4.4



It is visible from this graph that the uncertainty is hardly noticeable and its resulting effect on the CMMS response is negligible. In fact, the main source of error is in the estimation of constants for the process. Logically, it is reasonable to assume that the operator cannot be relied upon to supply detailed information on the centrifuges as it is generally classified. Therefore, constants such as the enrichment factor and the hold-up time per centrifuge would have to be independently calculated. This is not quite as difficult as it may seem at first as it is possible to do this reasonably accurately using models of centrifuges outlined in "Nuclear Chemical Engineering" by Benedict, Pickford and Levi<sup>[7d]</sup>, if the size of the centrifuges in the facility is known and certain assumptions about efficiency are made.

Nevertheless, there will still be a margin of error in this calculation of  $t_p$  which can be estimated using the concept of differentials. For any function  $f = f(\lambda, \mu)$ , the differential is given by  $df = d\lambda(\partial f/\partial \lambda) + d\mu(\partial f/\partial \mu)$ , and the error in  $f$ , i.e.  $\delta f$ , is approximately given by  $df$ . Since  $t_p = t_p(t_h, \beta)$ , the error in the equilibrium time,  $\delta t_p$ , is described by the following equation (which is used for the error calculations in the table of Section 4.1).

$$\begin{aligned} \delta t_p &= \delta t_h \cdot \left| \frac{\partial t_p}{\partial t_h} \right| + \delta \beta \cdot \left| \frac{\partial t_p}{\partial \beta} \right| \\ &= \delta t_h \cdot E(N_p, N_F) \left| \frac{\beta+1}{(\beta-1)\ln\beta} \right| \\ &\quad + t_h E(N_p, N_F) \cdot \delta \beta \left| \frac{1}{(\beta-1)\ln\beta} - \frac{\beta+1}{(\beta-1)^2 \ln\beta} - \frac{\beta+1}{\beta(\beta-1)(\ln\beta)^2} \right| \end{aligned} \quad (24)$$

It is also worth noting that the uncertainty in  $t_p$  in Table 4.1 is quite pessimistic as the hold-up time should easily be known within  $\pm 1s$  along with the enrichment factor being known within  $\pm 0.05$ .

With regard to the equilibrium time, there is one more potential source of error that should be mentioned. The ability to measure the equilibrium time is ultimately based on the premise that the moment when the plant initially reaches its maximum inventory is the

exact time when the machines start enriching the feed. This is not necessarily true because it is likely that the centrifuges will be put in motion before the feed is initially injected, in which case the enrichment process will have partially begun before the plant has been entirely filled. As this is difficult to quantify, it has not been included in the report. However, this problem should be a minor one as the plant is filled quite rapidly and large levels of enrichment should not occur until the system is in total reflux (no inputs or outputs).

As for uncertainty in the cut, this is dependent on the level of tails enrichment, which could be varied by the operator. However, assuming that the tails can only be adapted to a certain extent, the cut for a plant enriching from NatU to 5%LEU or so can be confined to a reasonably narrow range and likewise for one enriching from NatU to 90%HEU. Once more, error due to the actual mass measurements can be neglected.

With regard to the detection of batch recycling, aside from the mass measurement errors, there is no major source of uncertainty as such.

## Discussion

### 5.1 Conclusions

The purpose of this study was to extend the potential use of the ideas behind the RTMES and to simulate its response to new diversion scenarios from those already considered. On the basis of this work and that of the EU Commission JRC team, the CMMS could be installed in GCEP's and this would dispense with the need for destructive analysis of samples.

To summarize, the response of the CMMS to 2 main types of diversion scenarios has been simulated using simple material-flow-based models of GCEP's. For the case of the cascade reconfiguration, there are 2 ways in which the CMMS can detect the new state of affairs. The first is based on measuring the equilibrium time of the cascade. If the cascade is performing the operation NatU→3 to 5%LEU,  $t_p$  should be within the range of  $4 \pm 3$  mins or less. If it is enriching to higher levels,  $t_p$  will be longer and in the case of NatU→90%HEU,  $t_p$  will be of the order of  $40 \pm 17$  mins. Advantages of this method include the calculations' independence from the tails assay, the ability to know that the cascade is reconfigured before any major levels of product have been obtained and the fact that the number of centrifuges in the plant does not need to be known. One major disadvantage of this technique is the need to estimate the hold-up time and separation factor of the centrifuges, as these values have to be determined independent from the operator; this estimation leads to a large percentage error in the equilibrium time. Two other downsides to this method are the fact that it can only be used when the plant is starting up and also, the assumption that

the cascade is ideal. The latter could be an issue if non-ideal, energy-inefficient cascades were used, for which the equilibrium time is always larger than that of an ideal cascade.

An alternative method to ascertain whether or not the cascade has been altered is to continuously measure the cut  $\theta$ , i.e. the ratio of product rate  $p$  to feed rate  $f$ . If the plant is working in the mode NatU $\rightarrow$ 3 to 5%LEU with a reasonable tails assay,  $\theta$  should be inside the range:  $(0.07 \leq \theta \leq 0.20)$ . If the configuration is in the NatU $\rightarrow$ 90%HEU mode,  $\theta$  should be within:  $(0.002 \leq \theta \leq 0.007)$ . Among the benefits of this method is its ability to show up the reconfiguration without needing to have the CMMS installed before the reconfigured cascade has begun operation. Another notable advantage is the fact that no assumptions about ideality are made so this method of detection is still valid regardless of the efficiency of the plant. One disadvantage is the relatively large uncertainty due to the potential variation in the tails assay by the operator. A final point that ought to be made is that it is a complimentary detection technique to the equilibrium time measurement. Thus, if there is a long equilibrium time and a low value for the cut, these 2 pieces of information would provide a strong indication that HEU is being produced directly from NatU.

With regard to the batch recycling scenario, the CMMS should have no difficulty in showing up the diversion via long-term monitoring of the product, tails and feed rates. One possible way to circumvent the CMMS would be to avoid emptying the plant out after each cycle. Although there is nothing to stop the operator from doing this, it is an unlikely situation as the dilution of highly-enriched feed with much less-enriched feed would flagrantly waste the large amounts of time and energy put into the production of the former.

In addition to using the CMMS for calculating the flow rates, the fact that it measures the absolute mass of the cylinders at the entry and exit points would also be useful in ensuring that only certain canisters are used at specific points. For instance, 48Y containers should usually be found at the feed points only, and if the tare mass of a cylinder in a feed station was less than that of a 48Y, this would suggest that NatU is not being used as the feedstock. This could be supplemented by camera surveillance of the feed, product and tails areas.

## 5.2 Future Work

As a result of this study, the principle of real time mass measurement on a continuous basis could be considered for employment in GCEP's in the future. More work may be required to ensure that the system cannot be circumvented. For instance, several feed, tails or product cylinders could be placed on a station mass balance to disguise the use of smaller cylinders. The use of cameras could eliminate this problem but if the use of surveillance techniques was rejected by an operator, it may pose a challenge to determine

whether or not smaller cylinders than usual are being used at the feed, product or tails areas. Another possible way to circumvent the system would be to use only one small cascade within a facility for producing HEU while the rest of the cascade produces reactor-grade LEU. However, this would require siphoning off some of the LEU from the other cascades and should manifest itself in the cumulative mass balance as a result.

At this point in time, the use of real time mass measurement in GCEP safeguards has only been examined in principle. In practice, such a system could encounter a number of complications. For instance, at each of the feed, tails and product entry or exit points, there are usually several load cell stations rather than one, so as to allow the plant to keep running while cylinders are being changed. Thus, a mechanism for deciding which stations are online would have to be figured out. As well as this, there is the issue of large noise levels in the CMMS when cylinders are being changed, a problem that could mask undeclared activity.

In terms of practical implementation, the CMMS would consist of a set of calibrated mass balances located at each load cell station, with knowledge as to whether they are feed, tails or product points. The mass at each station would be measured at regular intervals, for example every 10 seconds, and the information sent to a central tamper-resistant computer on site via a secure communication system. This computer could then store the data and send it to an IAEA office in Europe or elsewhere every day or every week for analysis. Alternatively, the computer could perform the analysis itself using defined algorithms and alert the IAEA in the event of undeclared operations occurring.

Experimental validation is planned at JRC Ispra and this could contribute towards a more comprehensive evaluation of the method.

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| (d.) Chapter 5, pg 104 | (e.) Chapter 6, pg 134 | (f.) Chapter 5, pg 103 |
| (g.) Chapter 6, pg 130 | (h.) Chapter 6, pg 130 | (i.) Chapter 5, pg 105 |

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(a.) pg 3

[6] "Gas Centrifuge Enrichment Plant Safeguards System Modeling"; HA Elayat, WJ O'Connell, BD Boyer; Lawrence Livermore National Laboratory, California USA; 47<sup>th</sup> Annual Meeting for the Institute of Nuclear Materials Management, Nashville USA (2006)

(a.) pg 6 and 7

[7] "Nuclear Chemical Engineering"; M Benedict, TH Pickford, HW Levi; McGraw Hill Series in Nuclear Engineering; 2<sup>nd</sup> Edition (1981)

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| (a.) Chapter 12, pg 655     | (b.) Chapter 12, pg 679 | (c.) Chapter 12, pg 680 |
| (d.) Chapter 14, pg 847-875 |                         |                         |

[8] "The Theory of Isotope Separation as Applied to the Large Scale Production of <sup>235</sup>U"; K Cohen; McGraw Hill National Nuclear Energy Series (1951)

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| (a.) Chapter 1, pg 17 | (b.) Chapter 6, pg 104 | (c.) Chapter 1, pg 5  |
| (d.) Chapter 1, pg 7  | (e.) Chapter 1, pg 23  | (f.) Chapter 1, pg 19 |
| (g.) Chapter 1, pg 26 | (h.) Chapter 1, pg 25  |                       |

[9] "Safeguards Training Course Manual"; United States Program for Technical Assistance to IAEA safeguards; Vienna International Conference Centre, Austria (1987)

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| (a.) Part 4, pg 177 | (b.) Part 4, pg 169 | (c.) Part 3, pg 16-35 |
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