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A METHOD FOR ESTIMATING THE INVENTORY OF AN
ISOTOPE SEPARATION CASCADE BY THE USE OF
MINOR ISOTOPE TRANSIENT CONCENTRATION DATA

S. Blumkin

E. Von Halle

**UNION
CARBIDE**

OAK RIDGE GASEOUS DIFFUSION PLANT
OAK RIDGE, TENNESSEE

*prepared for the U.S. DEPARTMENT OF ENERGY under
U.S. GOVERNMENT Contract W-7405 eng 26*

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OAK RIDGE GASEOUS DIFFUSION PLANT
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ABSTRACT

An indirect method for estimating the inventory of a uranium enrichment cascade which presumably can be performed by the International Atomic Energy Agency within the Non-Proliferation Treaty limitations on its safeguards activities has been devised and tested at the Oak Ridge Gaseous Diffusion Plant (ORGDP). This method involves the feeding of a cascade with uranium that is significantly richer in one component than the normal cascade feed for a short period of time, and the measurement of the subsequent transient concentrations of this component in the cascade withdrawal streams. The inventory estimate is then obtained from a comparison of the observed data with parallel data calculated for an appropriate but non-identical cascade model. The validity of the method is demonstrated numerically by parallel calculations made for two nearly ideal hypothetical cascade models.

A test of this method conducted in the ORGDP cascade, utilizing U-236 as the spike component, yielded an estimate of the cascade inventory greater by 3.0% than that determined by the usual measurement method. The single numerical proof of the method showed a bias in the inventory estimate of 0.5%. The difference between this and the indicated error in the plant test is attributed to measurement error. This result is regarded as a very good one since it represents, in effect, just a single result obtained in a first test of the method. The determination of confidence limits for the method would require results from several additional tests.

The proposed method has been devised for use at enrichment plants that are subject to safeguarding by the IAEA and for which the proprietors will permit the inventories to be known. The method does not require knowledge or determination of any technology information that may be considered to be sensitive. For it to be applied, a plant proprietor must be willing to spike his cascade with a non-normal feed and do or permit the extensive sampling of its feed and withdrawal streams that is entailed.

The method has been tested at the ORGDP though it is not subject to IAEA safeguards and the disclosure of its inventory is restricted. For this reason any information that might divulge the ORGDP cascade inventory is excluded from this report.

The numerical demonstration of the validity of the method rests on a single comparison of two nearly ideal hypothetical cascades. It is also shown that if the two cascades being compared differ greatly in separative efficiency, such as a square cascade versus a nearly ideal one, a basic discrepancy in the inventory estimate not attributable to any measurement error would result. Additional numerical comparisons of cascades of varied designs should be made to provide further confirmation of the validity of the technique and to determine its limitations.

FOREWORD

This document is a report on one phase of a study relating to the use of minor uranium isotope concentration data in cascade external streams as an enrichment plant safeguards technique (MIST) conducted for the United States Arms Control and Disarmament Agency (ACDA) under Agreements No. ACDA/NWT/RA-181 and No. AC6AA705.

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A METHOD FOR ESTIMATING THE INVENTORY OF AN
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INTRODUCTION

In its responsibility, under the Nuclear Non-Proliferation Treaty (NPT), to develop and apply methods for the safeguarding of nuclear materials at uranium enrichment plants, the International Atomic Energy Agency (IAEA) is limited by the treaty provision which denies it access to the separation equipment and to any data which would disclose what are considered to be sensitive features of the process technology. This restriction makes it impossible for the Agency to verify the physical inventory of a separation cascade by the usual method of measurement. The inability to verify the physical inventory of an enrichment cascade leaves a gap in the IAEA's nuclear material accountancy data, which it is seeking to close. Thus, as part of its activities in support of the IAEA programs for nuclear safeguards, the U.S. Arms Control and Disarmament Agency (ACDA) has sponsored a study at the Oak Ridge Gaseous Diffusion Plant (ORGDP) to develop a non-intrusive alternate method.

An indirect method for estimating the inventory of an isotope separation cascade, which presumably can be performed within the NPT regulations on safeguards activities, has been devised and tested at the ORGDP. This report describes the proposed method, its application at the ORGDP and the results obtained.

The testing of the proposed method was the major but not the only objective of a third plant test conducted in 1975 at the ORGDP in connection with the measurement of the minor uranium isotope concentrations in cascade feed and withdrawal streams for use as a safeguards technique (MIST). Only those aspects and data from the test relevant to the inventory measurement are discussed in this report.

SUMMARY AND CONCLUSIONS

An indirect method for estimating the inventory of a uranium enrichment cascade which presumably can be performed by the IAEA within the NPT limitations on its safeguards activities has been devised and tested at the ORGDP. This method involves the feeding of a cascade with uranium that is significantly richer in one component than the normal cascade feed for a short period of time, and the measurement of the subsequent transient concentrations of this component in the cascade withdrawal streams. The inventory is then obtained from a comparison of the observed data with parallel data calculated for an appropriate but non-identical cascade model. The validity of the method is demonstrated numerically by parallel calculations made for two nearly ideal hypothetical cascade models.

A test of this method conducted in the ORGDP cascade, utilizing ^{236}U as the spike component, yielded an estimate of the cascade inventory greater by 3.0% than that determined by the usual measurement method. The single numerical proof of the method showed a bias in the inventory estimate of 0.5%. The difference between this and the indicated error in the plant test is attributed to measurement error. The result is regarded as a very good one since it represents, in effect, just a single result obtained in a first test of the method. The determination of confidence limits for the method would require results from several additional tests.

The proposed method has been devised for use at enrichment plants that are subject to safeguarding by the IAEA and for which the proprietors will permit the inventories to be known. The method does not require knowledge or determination of any technology information that may be considered to be sensitive. For it to be applied, a plant proprietor must be willing to spike his cascade with a non-normal feed and do or permit the extensive sampling of its feed and withdrawal streams that is entailed.

The method has been tested at the ORGDP though it is not subject to IAEA safeguards and the disclosure of its inventory is restricted. For this reason any information that might divulge the ORGDP cascade inventory is excluded from this report.

The numerical demonstration of the validity of the method rests on a single comparison of two nearly ideal hypothetical cascades. It is also shown that if the two cascades being compared differ greatly in separative efficiency, such as a square cascade versus a nearly ideal one, a basic discrepancy in the inventory estimate not attributable to any measurement error would result. Additional numerical comparisons of cascades of varied designs should be made to provide further confirmation of the validity of the technique and to determine its limitations.

THE METHOD AND ITS BASIS

The concept for this method was suggested by the fact that the ratio of the equilibrium times of two ideal isotope separation cascades is equal to the ratio of their inventories provided the two cascades have the same product and tails concentrations and the same separative capacities. The thesis is that if each of two such cascades is fed for a short period with measured quantities of a feed which is significantly richer in one component than the normal cascade feed, the times required to remove the same fraction of the transiently enhanced component from the two cascades should be in the same ratio as the inventories of the two cascades.

This premise has been used to devise an indirect method for measuring the uranium inventory of a ^{235}U enrichment cascade for which data of the kind that are usually collected for inventory determinations are unavailable to those who need to verify the inventory as a safeguards measure. The proposed method is non-intrusive and does not require knowledge or determination of any technology information that may be considered

sensitive such as operating pressures and temperatures or numbers, sizes and arrangement of separating units. For the method to be applied, a plant proprietor obviously must be willing to permit the process inventory of his cascade to be known to others and also be willing to spike his cascade with a non-normal feed and do or permit the extensive sampling of the feed and withdrawal streams that is entailed.

Briefly, the method involves the comparison of the time taken for an imposed transient change in the isotopic concentrations in the withdrawal streams of the subject cascade with that calculated for a cascade model that is identical to the subject cascade only in its flowsheet. It is assumed that the subject cascade is approximately ideal and that the rates and isotopic concentrations of all feed and withdrawal streams can be measured. Work that has been done to demonstrate the validity of the method is presented subsequent to the description of the procedure which follows.

Description of the Method

- A. As a first step, a sufficient quantity of uranium significantly richer in ^{234}U or ^{236}U relative to its ^{235}U content than in the normal feed is obtained. This uranium will be referred to as the spike and the process of feeding it to the cascade will be called spiking.
- B. The spike uranium is fed to the cascade at a known and preferably constant rate for a short period of time. The spiking time should be long enough for the spike component to build up to concentrations in the withdrawal streams so that its concentration can be measured with sufficient precision. The rates of all of the feed and withdrawal streams need to be measured during this period and for a time thereafter long enough to permit an appreciable fraction of the spike to be withdrawn from the cascade.
- C. Samples of all the cascade withdrawal streams should be taken at roughly regular intervals during the test period. The magnitude of the sampling interval depends on the length of the process equilibrium time: for gaseous diffusion once daily should be adequate; for centrifugation about six to twelve times daily would probably be desirable.
- D. The concentration of the spiking component in the samples plus that in any feed other than natural uranium should be measured with an accuracy equal to or better than $\pm 1\%$. The ^{235}U concentration in the spike uranium needs to be known for operational reasons. It is desirable also to determine the ^{235}U concentrations in the product and tails samples for assurance of otherwise stable cascade operation during the test.
- E. Computations are then made using the sample concentration data and the stream rate data to obtain the fraction of the spike component that has been withdrawn at each sampling time.

- F. A cascade model is designed to represent the cascade for which an inventory estimate is desired. The measured or declared flowsheet for the subject cascade and an arbitrarily assumed separation factor, within the range of what may normally be expected for the given separation process, is used to design the cascade model. Though the actual cascade and its model have the capacity to do the same separation job, the two will almost certainly differ as to the number of cascade sections, numbers of stages and the magnitudes of the interstage flows.
- G. A stage hold-up time is assumed for the cascade model. Though it is not critical, it is preferable that the value chosen be one that is within the range of what is normally expected for the given separation process. The assumed hold-up time specifies the inventory of the cascade model.
- H. Multicomponent transient productivity calculations are then made for the cascade model using the same feed and withdrawal stream rates and the same spiking time as was reported for the subject cascade. The fraction of the spike that has been withdrawn is computed for the cascade model for various times during the test period.
- I. A plot is made of the spike fraction removed versus time for the cascade model. Similar values already computed from the data for the real cascade are also plotted. Then for a given fraction of spike removed, one can read from the two curves the times required to do so for the cascade model and the real cascade. The estimate of the inventory of the subject cascade is then equal to the inventory of the cascade model multiplied by the ratio of the time read off the curve for the subject cascade to that of the cascade model.
- J. As is shown subsequently, the premise for the method is valid if the spiking periods for the two cascades are in the ratio of their inventories. Consequently, the procedures in paragraphs H and I need to be repeated with a new spiking time for the cascade model based on the ratio of its inventory to that of the inventory estimated for the real cascade until no significant change in the spiking time for the cascade model calculation is required. In practice, just one iteration, after the initial estimate is obtained, should suffice.

Validity of the Method

One can easily demonstrate analytically that the premise on which the method is based holds for single separation stages. But whether it holds also for cascades comprised of many stages of various sizes and arrangements was open to question and therefore required substantiation. Initial efforts to develop a rigorous analytical proof of the premise for cascades showed that, at the very least, it is a difficult and laborious task which may prove to be infeasible to complete successfully. It was decided instead to employ a numerical approach to demonstrate the validity of the method. Use was made of a computer code which is designed to calculate the transient concentration gradient and inventory of each component of a multicomponent mixture in a separation cascade.

The initial numerical test of the method was based on two hypothetical gaseous diffusion multi-section cascade models designed for different stage separation factors but identical flowsheets, that is, with the same flow rates and ^{235}U concentrations in the feed and withdrawal streams. The value for $(\alpha-1)$ for one of the cascades was set at $3/4$ of the other. (α is defined in the usual manner as the ratio of the ^{235}U to the ^{238}U concentrations in the enriched stream leaving a stage divided by the corresponding ratio in the depleted stream from the same stage.) Thus, though the two cascade models were designed to do the same separation job, they differ appreciably in size. Each model is a minimum total flow cascade for a design with seven enricher and seven stripper sections so that each is nearly ideal: the separative efficiency of one is 98.2% and the other 98.3%. The number of stages and the stage upflow rates in each section of the cascade with the lesser $(\alpha-1)$ are $4/3$ of that in the other cascade. The assumption was made that the stage hold-up times in all of the stages of both cascades are identical. Consequently, the uranium inventory of the cascade with the smaller separation factor would be $(4/3)^2$ or 1.7778 times the other one. Due to round-off to an integral number of stages in each section of the cascade, the actual ratio of the inventories came out to be 1.7756.

The isotopic composition of the uranium to be used as a spike in the calculations was assumed to be 0.89% ^{235}U and 0.393% ^{236}U . The feed rate for the spike was chosen arbitrarily to be one-tenth that of the normal cascade feed rate.

The following calculations were then made for each cascade model using the computer code for transient concentrations in a multicomponent system except where noted otherwise.

1. The concentration gradients for ^{235}U and ^{238}U for each cascade at steady-state when fed with natural U only were obtained. The same feed rates and the same product rates were used for both cascades. The two steady-state gradients did not turn out to be exactly identical, probably because of round-offs in cascade design, the ^{235}U product concentrations differing by 8 parts per thousand and the tails concentration by 20 parts per thousand. However, the ratio of the ^{235}U inventories in the two cascades was equal to the ratio of the U inventories, that is, 1.7756. For these initial calculations, none of the spike component, ^{236}U , was present.
2. Starting from the steady-state concentration gradients in the two cascade models, the changes in the cascade concentrations resulting from the feeding of the spike U were computed. The analytical treatment that had been made for single stages had shown that the premise for the methods holds if the spiking periods are in the ratio of the stage inventories. On the presumption that this may also be true for multi-stage cascades, two sets of calculations were made, one with identical spiking times and the other with the spiking times for the two cascade models in the ratio of their inventories.

In an actual cascade operation, the product and feed rates would be adjusted continually during the spiking period to maintain the ^{235}U product and tails concentrations at their target values. For these calculations, instead, the natural U feed rate and the product withdrawal rate were set during the spiking period at the values which would result in the targeted ^{235}U concentrations if the spike were to be fed until the cascade concentrations reach their new steady-state values. These rates, which are the same for both cascades, were determined from calculations with a multi-component steady-state productivity computer code. Thus, the ^{235}U concentrations in the cascade withdrawal streams deviated somewhat from their targeted values during the spiking period and for a time thereafter.

3. At the end of the spiking period for each cascade, the product and feed rates were restored to the values pertaining before the spiking phase and the transient calculations were continued until most of the ^{236}U was removed from the cascade.

The postulate to be demonstrated is that the times required to remove equal fractions of the spike component from two nearly ideal cascades performing the same separation job are in the ratio of their inventories. In connection with the transient productivity code, the fraction removed is expressed more conveniently in terms of the spike component inventory in the cascade rather than in terms of the quantity removed in the withdrawal streams. Thus,

$$\phi \equiv \frac{F_S X_{FS} t' - I_S(t)}{F_S X_{FS} t'} = 1 - \frac{I_S(t)}{F_S X_{FS} t'}$$

where ϕ = the fraction of the spike component (^{236}U) withdrawn,

F_S = the feed rate of the spike U,

X_{FS} = the concentration of the spike component in the spike U,

t_S = the duration of the spiking period,

I_S = the inventory of the spike component in the cascade.
The computer code computes this value at the end of each specified time interval by numerical integration over the cascade.

t and t' = time: $t' \leq t_S$; when $t < t_S$, $t' = t$;
when $t \geq t_S$, $t' = t_S$. At start of spiking
period $t' = t = 0$.

Then for the premise to be valid, the times t_A and t_B for all values where $\phi_A = \phi_B$ for two cascades A and B, have to be in the ratio of the cascade inventories, that is

$$\frac{t_A}{t_B} = \frac{I_A}{I_B}$$

where I = the uranium inventory of the cascade.

Computations were then made from the transient code data output to obtain the times at which equal values of ϕ for the two cascade models occurred. This yielded the time ratios for equal spike fractions withdrawn, which should be equal to the cascade inventory ratios. The time ratios obtained are given in Table 1 for the case in which the spiking times are identical and for the one in which the spiking times for the two cascades are in the ratio of their inventories.

TABLE 1
TIME RATIOS FOR VARIOUS FRACTIONS OF SPIKE REMOVAL
FOR THE TWO NEARLY IDEAL CASCADE MODELS

$$\frac{\alpha_B - 1}{\alpha_A - 1} = \frac{3}{4}$$

$$I_B/I_A = 1.7756$$

ϕ	t_B/t_A	
	Identical Spiking Periods	Spiking Periods in the Ratio of the Cascade Inventories
.10	1.4623	1.7740
.20	1.4775	1.7720
.30	1.5149	1.7705
.40	1.5507	1.7690
.50	1.5829	1.7674
.60	1.6118	1.7658
.70	1.6383	1.7646
.80	1.6636	1.7630
.90	1.6905	1.7615
Avge Ratio	1.5769	1.7675
Avge Error	-11.2%	-0.46%

It is apparent that the ratio of the times is not a constant but varies somewhat with the ϕ value, much more so in the case of identical spiking times than when the spiking times are in the ratio of the cascade inventories. For this reason, an average value for the time ratios was computed for each case and is also tabulated. The deviation, in per cent, of the average time ratio from the cascade inventory ratio is also tabulated under the heading of average error.

Thus, for the two cascade models, which are designed for the same separation job, the average times required to withdraw identical fractions of a cascade spike are in a ratio which differs from the ratio of their inventories by 0.46% if the spiking periods are in the ratio of the cascade inventories. When the spiking periods are identical the two ratios differ by 11%. In applying this method to estimate the unknown inventory of a cascade, one will therefore initially have to assume identical spiking times, and then repeat the calculations one or more times with appropriately corrected spiking time ratios.

Cascade Non-ideality and the Validity of the Method

To check the intuitive expectation that the premise for the method may not be valid if the two cascades, real and model, differ significantly in separative efficiency, the extreme situation, where one cascade is square and the other is nearly ideal, was examined. The two cascades for this comparison were low enrichment ones (4% ^{235}U) designed for the same flow-sheet and the same $^{235}\text{U}/^{238}\text{U}$ separation factor. The square cascade was an optimum square cascade with the stage upflows 1.36 times the required minimum stage upflow at the feed point. Its separative efficiency was 76.4% versus 98.2% for the nearly ideal cascade. The total flow of the square cascade was greater by a factor of 1.286 than that of the nearly ideal one and since identical stage hold-up times were assumed for both, the inventory of the square cascade was larger than that of the ideal one by the same factor. The spiking times used were in the ratio of the cascade inventories. The results of the calculations are given in Table 2.

Thus if, in the unlikely case, the real cascade is a square one and the assumed cascade model is made nearly ideal this method would result in an overestimation of the cascade inventory of the order of 14% giving no consideration for other sources of error that may enter into the determination.

Additional numerical comparisons of pairs of cascades of varied designs need to be made to provide further confirmation of the validity of the method and to determine its limitations.

TABLE 2
TIME RATIOS FOR VARIOUS FRACTIONS OF SPIKE REMOVAL
FOR A SQUARE CASCADE AND A NEARLY IDEAL ONE

$$I_{SQ}/I_A = 1.2860$$

$$\text{Spiking Times Ratio} = 1.2860$$

ϕ	t_{SQ}/t_A
.10	1.3004
.20	1.3300
.30	1.3674
.40	1.4112
.50	1.4601
.60	1.5103
.70	1.5609
.80	1.6117
.90	1.6664
Avg Ratio	1.4687
Avg Error	+14.2%

THE PLANT EXPERIMENT

The proposed method for estimating the inventory of a uranium enrichment cascade was tested at the Oak Ridge Gaseous Diffusion Plant (ORGDP). The time chosen for the test was one during which cascade operational changes were expected to be at a minimum.

Cascade Streams and Concentrations

In normal operation at the time, the external streams of the ORGDP cascade consisted of two products, two feeds and tails. One of the feeds was partially enriched product from another installation and the other was natural uranium. The ^{235}U concentrations targeted for the two products and the tails, and present in the two feeds are given in the following tabulation:

TABLE 3
 ^{235}U CONCENTRATIONS IN THE CASCADE STREAMS

Products:	Top	3.1
	Side	2.5 & 2.8
Feeds:	Partially Enriched U	1.9
	Natural U	0.71
Tails		0.30

About half-way through the test period, production requirements necessitated a change in the target side product concentration. Two values are listed for the side product for this reason.

The Spike Feed

Since it is absent in natural feed and is present in significant concentrations in uranium recovered from spent nuclear fuel, ^{236}U is a logical choice for the spiking component. Thus, uranium recovered from nuclear reactor tails, that was on hand in the form of UF_6 in sufficient quantity to conduct the test, was chosen as the spike. Determinations on samples of this uranium from the cylinders in which it was delivered showed that the isotopic composition differed to some degree from cylinder to cylinder. From the point of view of test simplicity, it was desirable to mix the contents of the cylinders into a single batch of uniform isotopic composition. This was impractical, however, and consequently the concentration of the ^{235}U and the ^{236}U in the spike uranium varied during the test. The isotopic concentrations of the uranium in the cylinders used to spike the cascade are tabulated below.

TABLE 4
 ISOTOPIC COMPOSITION OF THE SPIKE URANIUM

Cyl #	Concentration, %		Ratio of ^{235}U Conc. to that of ^{236}U
	^{235}U	^{236}U	
1	1.7960	.3304	5.44
2	1.8065	.2294	7.88
3	1.7086	.2942	5.81
4	1.2492	.2204	5.67
5	1.2447	.2192	5.95
6	1.1887	.2151	5.53
7	1.1848	.2150	5.51
8	1.1840	.2106	5.62

The Partially Enriched Cascade Feed

Some ^{236}U in very small but variable concentrations was present in the partially enriched uranium fed to the cascade before and during the test. The contents of the cylinders containing this uranium that were fed during the test ranged in isotopic composition as follows:

TABLE 5
RANGE OF ISOTOPIC COMPOSITION
OF THE PARTIALLY ENRICHED FEED

<u>Isotope</u>	<u>Concentration Range, %</u>
^{235}U	1.921 - 1.927
^{236}U	.000310 - .000905

Thus, the ^{236}U in this feed varied, from the highest to the lowest concentration, by a factor of three but was for all cylinders no more than .5% of its concentration in any of the spike uranium. Thus the cascade had a very small inventory of ^{236}U at the time of test initiation. The ^{235}U concentrations in this feed were essentially constant for all practical purposes.

The Spike U Feed Rate

The precision of the three-stage mass spectrometer which was to be employed for the high precision ^{236}U measurements was estimated to be about $\pm 1\%$ for samples with no less than 100 ppm ^{236}U and $\pm 10\%$ for samples with about 1 ppm of the isotope. With none of the spike being fed to the cascade, the ^{236}U concentrations in the cascade top product and tails streams were running about 6 and 0.9 ppm, respectively. To obtain reasonably reliable ^{236}U determinations the spike uranium should be fed at a large enough rate and for a sufficient length of time to bring the ^{236}U concentration in the top product up to at least several hundred ppm. Calculations were made to estimate the rate at which the spike would need to be fed to achieve the desired ^{236}U concentration enhancement during the test.

Test Execution

The spike uranium was fed to the cascade from the cylinders containing it, in the sequence of their listing in Table 3. The contents of each cylinder were fed to a stage in the cascade where the in-cascade ^{235}U concentration matches as closely as practical to that in the uranium being fed.

The input rates of the two regular feeds, that is, the partially enriched and natural uranium, were adjusted for each cylinder of spike uranium to maintain the two product rates and concentrations and also the tails concentration as constant as possible. Such operations are, of course, outside the domain of a safeguards inspection team. However, once a cascade

operator has consented to the use of the special feed, he would almost certainly want to adjust the cascade feed rates in the manner described. The flow rates of all of the cascade feed and withdrawal streams were measured and recorded.

Beginning several days before the feeding of the spike was initiated and continuing until the test was completed, samples were taken at approximately the same time each day of the two product streams and the tails stream. Each of the cylinders containing the spike uranium and the partially enriched uranium fed to the cascade during the test was also sampled. No samples were taken of the natural uranium feed, it being considered to be of constant and known isotopic composition.

All samples were analyzed for their ^{235}U and ^{236}U concentrations. The ^{235}U determinations were made with a single-stage dual-collector mass spectrometer using certified standard samples for comparison. The ^{236}U determinations were made by pulse counting ion detection on a three-stage mass spectrometer, yielding the results in the form of ratios of the abundance of ^{235}U to that of ^{236}U .

An operational problem resulted in the spike uranium being fed to the cascade at a sharply reduced and variable rate. The spiking time was lengthened in accommodation. Thus, the ^{236}U concentrations in the cascade streams did not increase quite to the originally planned levels.

About mid-way during the spiking period the plant operator increased the target side product concentration from 2.5 to 2.8% ^{235}U . The operator, of course, adjusted the product rates and the input rates of the feeds other than that of the spike uranium to the extent required to effect this without disturbing the ^{235}U top product and tails concentrations.

Calculations Using the Measured ^{236}U Data

The observed ^{236}U concentration data were used to obtain a curve for the quantity of ^{236}U withdrawn from the cascade in the product and tails streams expressed as a fraction of the ^{236}U fed to the cascade versus the elapsed time. The cumulative quantity of ^{236}U fed to the cascade at successive points in time was computed from the average daily feed rate recorded for the spike and the partially enriched uranium and the ^{236}U concentrations in them. Similarly, the cumulative amount of ^{236}U withdrawn from the cascade at various times was computed from the average daily product and tails rates and the average of the ^{236}U concentrations in the given stream at the beginning and end of the given day.

The ^{236}U concentrations in each of the streams were plotted versus time. The concentrations in the first two top product samples after feeding of the spike was started were grossly anomalous. The two anomalous values were replaced for the computations with values taken from a curve drawn to smooth out the data. The plot of the fraction of the ^{236}U spike removed from the ORGDP cascade versus time is shown as curve A in the figure (see page 24).

Design of a Hypothetical Cascade Model

For the next step, a nearly ideal, squared-off cascade was designed to simulate the ORGDP cascade, such as a safeguards inspection team with no access to the real cascade might do it. The design was based on the average measured ^{235}U concentrations in the two products, in the partially enriched U feed and in the tails, and the average measured withdrawal rates of the two products. All of the values were averaged over the time period from test start to the change in the target side product concentration. The feed rates for the partially enriched and natural U were determined on the basis of the target ^{235}U tails concentration by material balance calculations with the ratio of the two feeds specified as that of their average measured values during the same time period. The spike feed did not enter into the cascade design.

The theoretical separation factor for $^{235}\text{U}/^{238}\text{U}$ for the gaseous diffusion process ($\alpha = 1.00429$) was assumed for the model. The squared-off design was made up arbitrarily of six square enriching and four square stripping sections, resulting in a cascade with a separative efficiency of 97.8%. The fictional cascade model, designed on the basis described, matches the actual cascade essentially in the operating flowsheet and separative capacity, but differs significantly in the number of square sections, number of stages, and the magnitudes of the stage upflows.

An arbitrary but reasonable stage hold-up time was then selected for the cascade model. Since the stage upflows were already set, this assumption had the effect of specifying the inventory of the cascade model.

Feed Rate Adjustment for the Model

Preliminary calculations showed that the measured feed rates were in general not consistent on a daily basis with the reported ^{235}U tails concentrations and the rates and ^{235}U concentrations of the products. For this reason it was decided not to use the measured data as is for the transient computations for the cascade model. Steady-state productivity calculations were then made to obtain a balanced operating cascade flowsheet for each batch of spike feed and for the periods before and after the spike was fed. For these calculations the withdrawal rates and ^{235}U concentrations of the two products and the ^{235}U concentration in the tails streams were set at their average measured values for the periods before and after the target side product concentration was changed. Then given the average measured input rate and the ^{235}U concentration for the spike U in a particular cylinder, the rates of the other two feeds, that is, the partially enriched U and the natural U, were computed. Eleven flowsheets were determined in this fashion: one each for seven of the spike U cylinders fed; two for the remaining cylinder which was on stream at the time the side product target concentration was changed; and one each for the period before the feeding of the spike U was begun and after it was discontinued.

These steady-state productivity calculations also provided the cascade feed stage locations for the two feeds and that of the side product withdrawal stream for each of its two target concentrations, to be used in the calculations with the computer code for transient concentrations.

Calculations of Transient Concentrations for the Model

Transient, multicomponent calculations were performed next with the safeguards cascade model to parallel the test procedure that had been carried out on the actual cascade. The first step in this set of computations was to establish the steady-state concentration gradient in the cascade at the time feeding of the spike U was begun. To do this, the transient productivity code was run, using the flowsheet that the safeguards model was based on, for sufficient cascade operating time so that the isotopic concentrations no longer changed significantly with time.

With this steady-state concentration gradient now representing that at time zero, the transient computer code was utilized to obtain the variation of the isotopic concentrations in the cascade withdrawal streams and of the component inventories with time, as each of the cylinders containing the spike U was fed in the same order and for the same period of time as had been recorded for its feeding to the real cascade.

From the data on the feed rates, feeding times and ^{236}U concentrations in the batches of the spike U and in the partially enriched U, the cumulative quantity of ^{236}U fed to the cascade at various times since time zero was computed. From the ^{236}U cascade inventory data obtained in the calculations, the cumulative quantity of ^{236}U removed from the cascade at these times was also computed. These two sets of numbers then yielded the quantity of ^{236}U removed from the cascade versus time expressed as a fraction of the ^{236}U fed. These results were then plotted on the same graph with the corresponding data derived from the actual cascade measurements.

Estimate of the Cascade Inventory

The times required to remove the same fraction of ^{236}U from the actual cascade and the cascade model can be read off from the two curves. If t_r and t_m are the times read off for the real cascade and the cascade model respectively, and the corresponding cascade inventories are denoted by I_r and I_m , then the cascade inventory can be estimated using the following equation:

$$I_r = \frac{t_r}{t_m} I_m$$

I_m has been specified by the cascade design and the assumption of the stage residence time.

It was evident from the curves that the time ratio at equal values of ϕ is not a constant. To obtain an average ratio, five pairs of values for t_r and t_m were read off from the two curves, each pair for a different value of the ordinate. This yielded five ratios of t_r to t_m , from which an average value of t_r/t_m (or I_r/I_m) was obtained.

As had been shown in the numerical demonstration of the validity of the method, the spiking times should be in the ratio of the cascade inventories. The initial calculations, of course, had to be made with the spiking time for the cascade model set equal to that used for the real (ORGDP) cascade. After an average value for t_r/t_m was obtained from the first set of calculations, the transient calculations for the cascade model were then repeated with the on stream time for each cylinder of spike uranium feed corrected by this ratio. A new curve of the fraction of spike component withdrawn, ϕ , versus time was obtained. This is curve B in the figure (see page 24). The corresponding data obtained from the test measurement on the ORGDP cascade is shown as curve A in the figure. Eleven pairs of values of t_r and t_m were then obtained from curves A and B, from which a second average value for t_r/t_m was computed. The set of time ratios and their average value are presented in Table 6.

TABLE 6
TIME RATIOS FOR VARIOUS FRACTIONS OF SPIKE REMOVAL
FOR THE ORGDP CASCADE
AND THE FICTIONAL CASCADE MODEL

ϕ	t_r/t_m
.30	1.083
.35	1.084
.40	1.094
.45	1.124
.50	1.131
.55	1.109
.60	1.111
.65	1.116
.70	1.114
.75	1.112
.80	1.103
Average Ratio	1.107

The spiking time ratio which was used for the second calculation was considered to be close enough to the second value obtained for t_r/t_m that it was concluded that a second iteration would probably yield an insignificantly different result and was therefore unnecessary. This value of t_r/t_m was then used in the above equation to obtain an estimate of the ORGDP cascade uranium inventory (I_r).

Accuracy of the Result

The inventory of the ORGDP cascade had been measured by the normal method at the beginning and end of the test. The value reported for the inventory at the end of the test was less than that at the beginning by 0.56%. Taking the average of these two values as the true inventory of the cascade, denoting it by I^* , and comparing it with I_r obtained from the test, it turns out that I_r is greater than I^* by 3.0%.

It should be noted that the method measures the process gas inventory only in the on-line equipment, that is, in the operating stages, and associated compressors, coolers, valves, and interconnecting piping. The inventory in off-line equipment such as traps, by-pass lines, inoperative stages, etc., is not included in the measurement.

The single numerical demonstration of the validity of the method showed a bias in the inventory estimate of the order of 0.5% and the plant test measurement came out to be 3% greater than what is considered to be the correct value. This difference, in all probability, reflects measurement error. In consideration of the fact that this was a first test of the method, the result is considered to be a very good one. The inventory estimate obtained from the many concentration and flow measurements made for the plant test represents just a single result. This being the case, there is insufficient data to determine confidence limits for the proposed method. To do so would require several additional tests.

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