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USE OF MINOR URANIUM ISOTOPE MEASUREMENTS AS AN AID IN SAFEGUARDING A URANIUM ENRICHMENT CASCADE

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INTRODUCTION

Material accountability is the basic and most direct means for safeguarding nuclear facilities against clandestine diversions of nuclear materials. Surveillance and containment, which are indispensable supporting measures for material accountability, do not provide those charged with safeguarding an installation with the assurance beyond the shadow of a doubt that all of the input and output uranium will in fact be measured. For this reason, some method to complement material accountability is desirable.

Those who are concerned with developing non-intrusive techniques for safeguarding uranium enrichment plants under the Nuclear Non-Proliferation Treaty have perceived the possibility that data on the ^{234}U concentrations, and that of ^{236}U when present, in addition to that of the ^{235}U in the cascade withdrawal and feed streams may provide a means to either corroborate the material accountability results or indicate that the integrity of the declared cascade operation is suspect. A basic theoretical study has been conducted to determine whether complete isotopic measurements on enrichment cascade streams may be useful for safeguards purposes. Numerous steady-state multicomponent concentration gradient and productivity calculations have been made to develop the fundamental data for this study. The results of the calculations have been reported in a series of five reports.[1] In this paper, a brief review and appraisal is made of the potential value of utilizing the measurement of ^{234}U and ^{236}U concentrations as well as that of ^{235}U in enrichment cascade feed and withdrawal streams as a safeguard technique.

SUMMARY AND CONCLUSIONS

The results of the calculations made to determine the behavior of the minor uranium isotopes in separation cascades and the results of the three plant tests made to substantiate the validity of the calculations are reviewed briefly. Based on the fact that the ^{234}U and ^{236}U concentrations relative to that of ^{235}U in cascade withdrawal streams depend on the

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cascade flowsheet, it is concluded that the use of minor isotope concentration measurements (MIST) is a potentially valuable adjunct to material accounting for safeguarding a ^{235}U enrichment cascade. Another characteristic of MIST which qualifies it for safeguards applications under the NPT is the fact that its application would be entirely non-intrusive on process technology and proprietary information.

REVIEW OF THE BEHAVIOR OF THE MINOR URANIUM ISOTOPES IN ^{235}U ENRICHMENT CASCADES

Minor Isotope Concentration Dependence

The study has shown that the steady-state concentrations of the uranium minor isotopes relative to that of the ^{235}U in the withdrawal streams of a separation cascade are determined by the following parameters:

A. The ^{235}U top product enrichment. The ^{234}U concentration relative to that of ^{235}U in both the product and tails streams increases with increasing ^{235}U concentration in the product stream. This is illustrated in Figure 1 which is a plot of the ^{235}U to ^{234}U concentration ratios in the product and tails streams of a squared-off cascade, designed to yield product at 4% ^{235}U and tails at 0.25% ^{235}U from natural uranium, when the cascade is operated to withdraw product with ^{235}U concentrations greater than the design product concentration.

   The ^{236}U concentration relative to that of ^{235}U decreases in the product and tails streams with increasing ^{235}U product concentration. Figure 2 is shown to illustrate this. The ratios of the ^{235}U to ^{236}U concentrations in the product and tails streams represent the results of calculations for various ^{235}U product concentrations for the same cascade when it is fed only with a reactor tails uranium (RTU1)[1] which has a ^{235}U concentration of 2.5%.

B. The ^{235}U depletion in the tails stream. The ^{234}U concentration relative to that of ^{235}U in both the product and tails streams increases with increasing ^{235}U tails concentration. This is illustrated in Figure 3 in which the ^{235}U to ^{234}U concentration ratios in the withdrawal streams of the same fixed cascade of the two preceding figures are plotted versus the ^{235}U tails concentration. For this set of data, the product concentration was maintained at its design value of 4% ^{235}U and the tails concentration was varied by changing the feed rate. The ^{236}U concentration relative to that of ^{235}U increases in the product stream but decreases in the tails stream with increasing ^{235}U tails concentration. Figure 4 demonstrates this behavior. The feed to the cascade is comprised of reactor tails U (RTU1) and natural U in constant proportions while the total rate of feed is varied to obtain the various ^{235}U tails concentrations.

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[1]RTU1 is one of two species of reactor tails uranium used in this study to calculate the effects of non-normal uranium feeds.

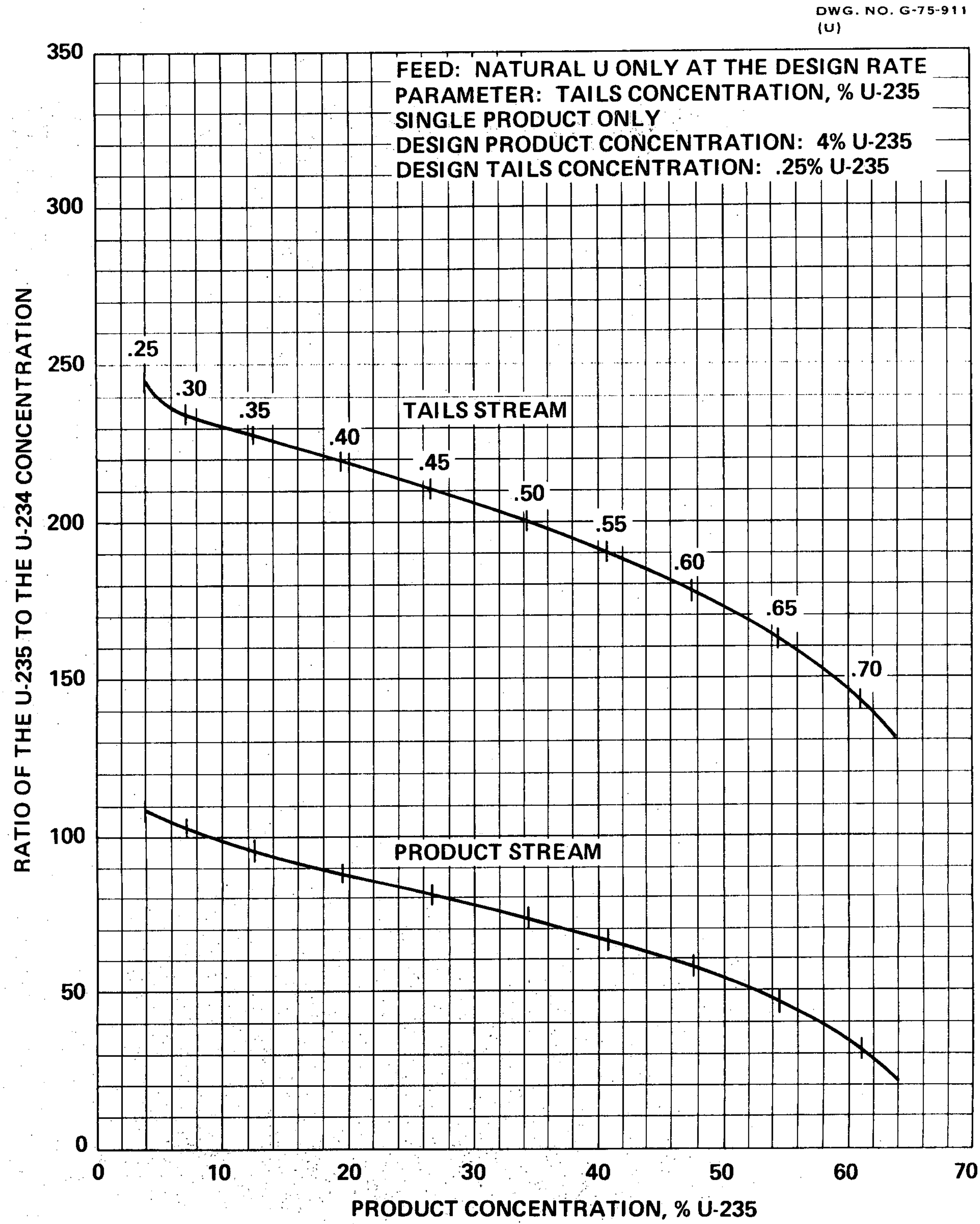

Figure 1

VARIATION OF THE U-235 TO THE U-234
CONCENTRATION RATIO IN THE PRODUCT AND WASTE STREAMS
WITH THE PRODUCT CONCENTRATION OF A FIXED CASCADE

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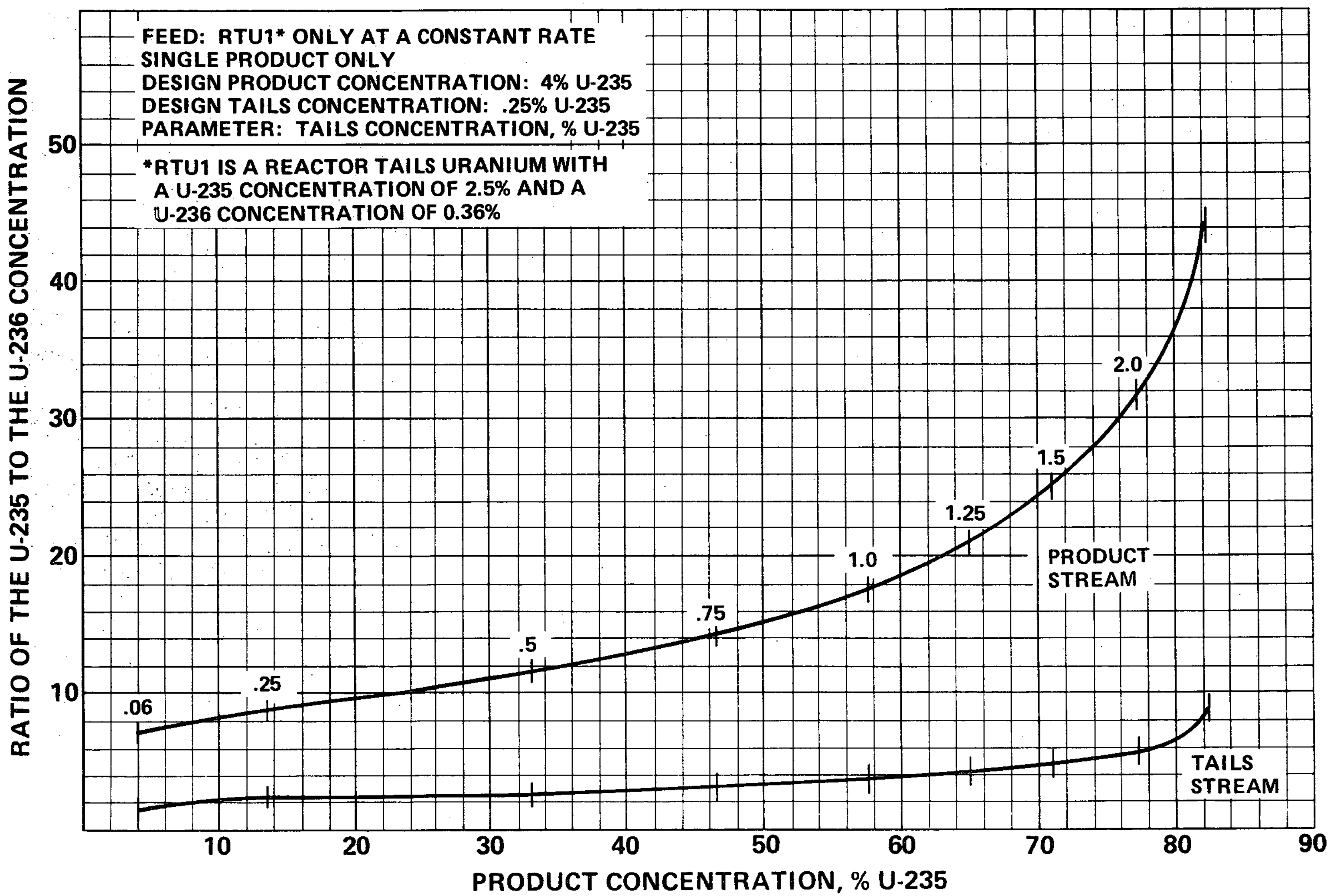

Figure 2

VARIATION OF THE U-235 TO THE U-236 CONCENTRATION RATIO IN THE
PRODUCT AND WASTE STREAMS WITH THE PRODUCT CONCENTRATION OF A FIXED CASCADE

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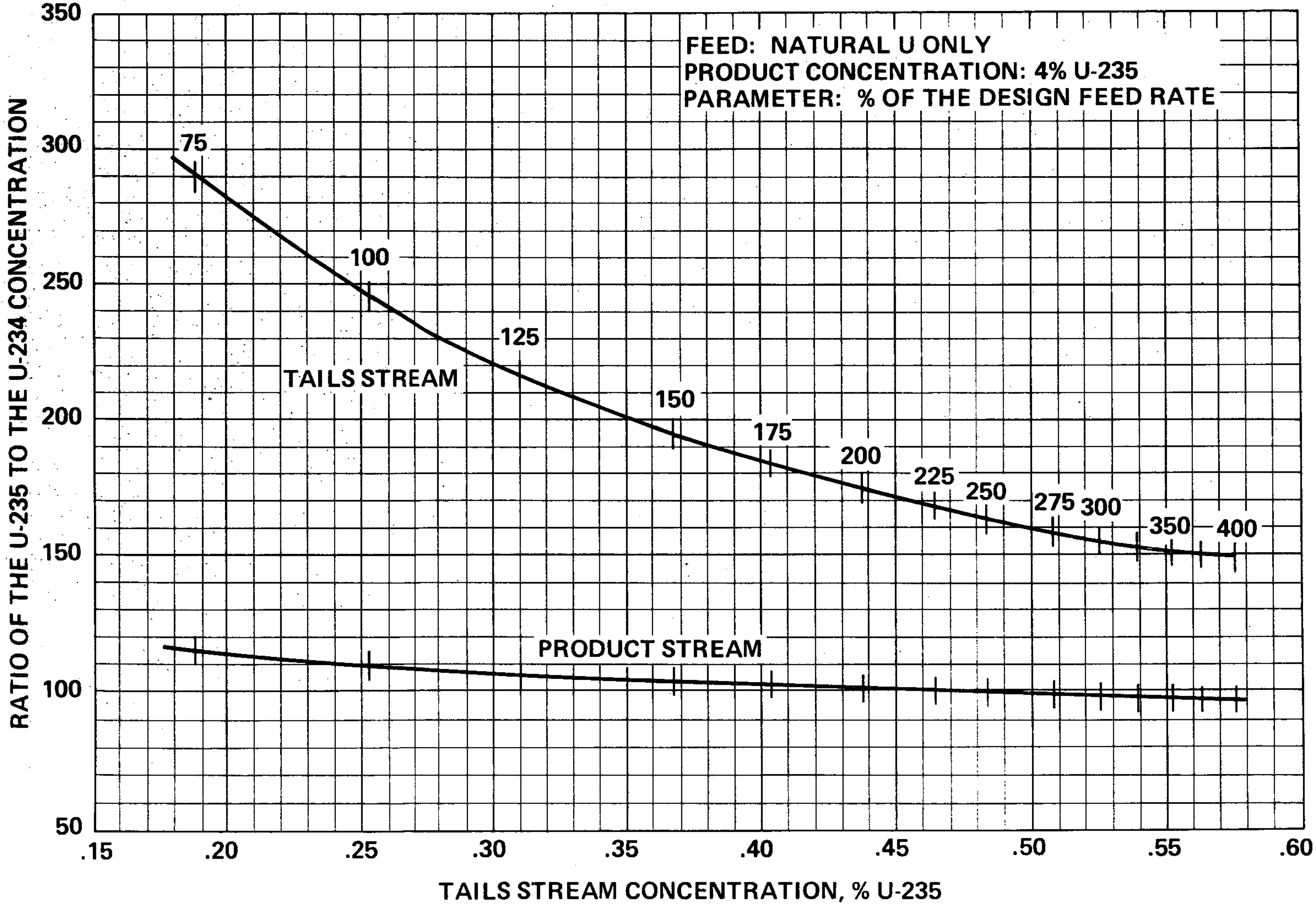

Figure 3

THE VARIATION OF THE U-235 TO THE U-234 CONCENTRATION RATIOS IN THE PRODUCT AND TAILS STREAMS WITH THE U-235 TAILS CONCENTRATION OF A FIXED CASCADE

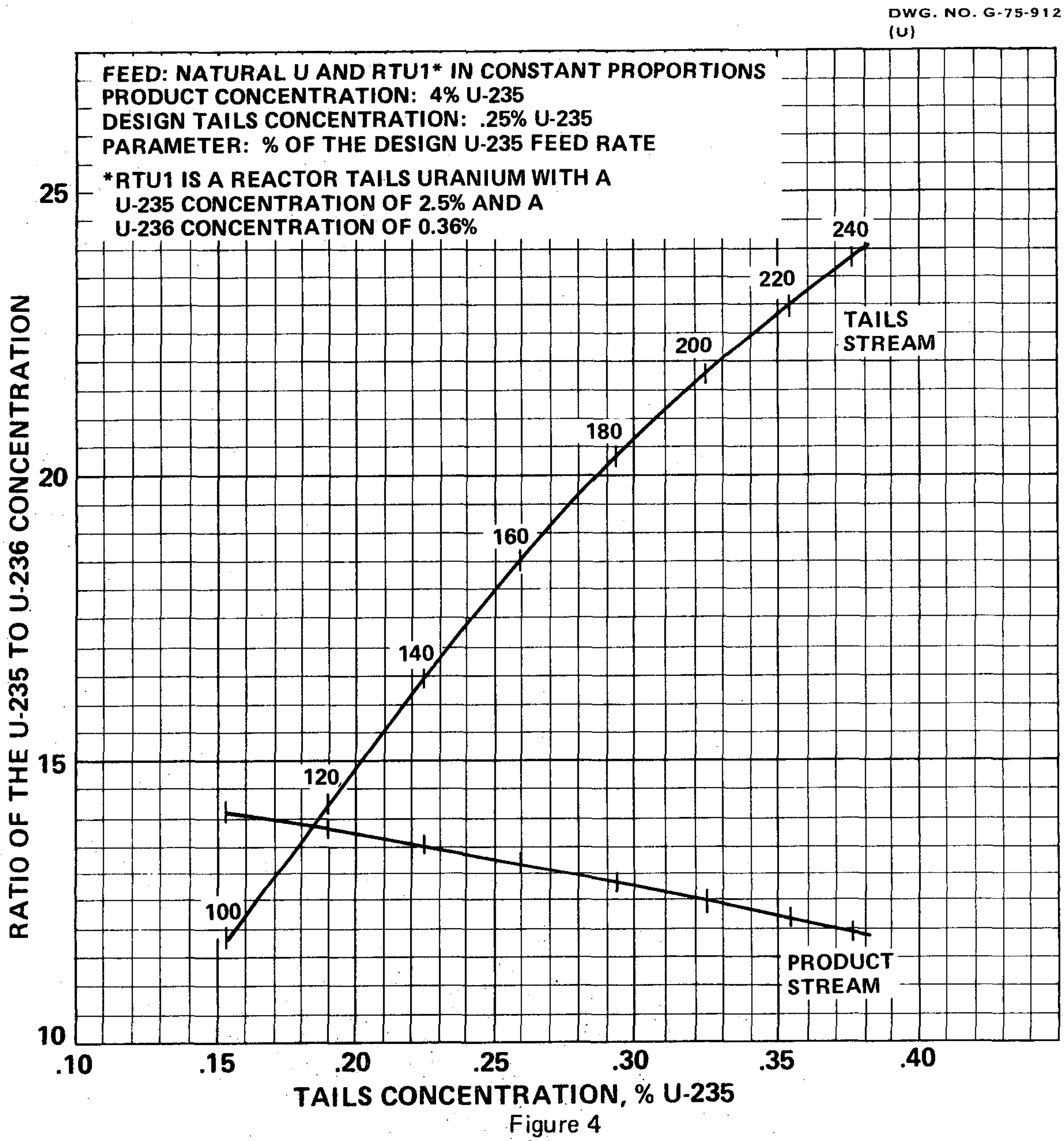

Figure 4

THE VARIATION OF THE U-235 TO THE U-236 CONCENTRATION RATIOS IN THE PRODUCT AND TAILS STREAMS WITH THE U-235 TAILS CONCENTRATION OF A FIXED CASCADE

C. The use of uranium other than natural as a feed to the cascade. Obviously ^{236}U will appear in cascade withdrawal streams only if uranium containing the isotope, such as uranium recovered from spent reactor fuel, is or has been fed to the cascade. When non-normal uranium is fed, the ^{234}U and ^{236}U concentrations relative to that of ^{235}U in cascade withdrawal streams are affected by both the ratio of the concentration of the ^{235}U to that of the minor isotope and the ^{235}U concentration in the non-normal uranium, and by the fraction of the total feed it comprises.

The ^{234}U concentration relative to that of ^{235}U may be depressed or enhanced by the use of the non-normal uranium depending upon its isotopic composition, that is, upon the ^{235}U to ^{234}U concentration ratio and the ^{235}U concentration. Figure 5 demonstrates the effect of feeding one or the other of two species of reactor tails uranium in various proportions with natural uranium on the ^{235}U to ^{234}U concentration ratios in the withdrawal streams of a fixed cascade. The reactor tails uranium, designated as RTU1, has a considerably higher ^{235}U concentration and ^{235}U to ^{234}U concentration ratio than does that designated as RTU2. Figure 6 is the counterpart to Figure 5 illustrating the behavior of the ^{235}U to ^{236}U concentration ratios in the product and tails streams. The ^{235}U to ^{236}U concentration ratio in RTU1 is also considerably greater than it is in RTU2.

D. The existence of an additional product stream. When the withdrawal of a second product stream is begun, the minor isotope concentrations in the first product stream will be affected even if the ^{235}U concentration in it remains unchanged. The minor isotope concentrations relative to that of ^{235}U in the tails stream are also affected. The magnitude of this effect depends upon the degree of enrichment of the additional product and the fraction this product represents of the total cascade production. Figure 7 is presented to illustrate how the ^{235}U to ^{234}U concentration ratios in the first product stream and the tails stream are affected by the existence of an additional product stream.

E. The ratio of feed streams and/or the ratio of product streams in the case of more than one feed and/or more than one product stream. When the cascade mode of operation normally consists of more than three external streams, that is, more than one feed stream and/or more than one product stream, there is an infinite set of combinations of external stream rates for which the ^{235}U concentration in the cascade withdrawal streams can be maintained constant. However, the minor isotope concentrations in the withdrawal streams will be affected by the changes in the stream rates. Figures 5 and 6, which have already been presented for item C, above, illustrate this. While the ^{235}U concentrations in the cascade product and tails stream are constant at their design values, and the yield of product is increasing as the proportion of reactor tails in the feed is increasing, the minor isotope concentrations are changing.

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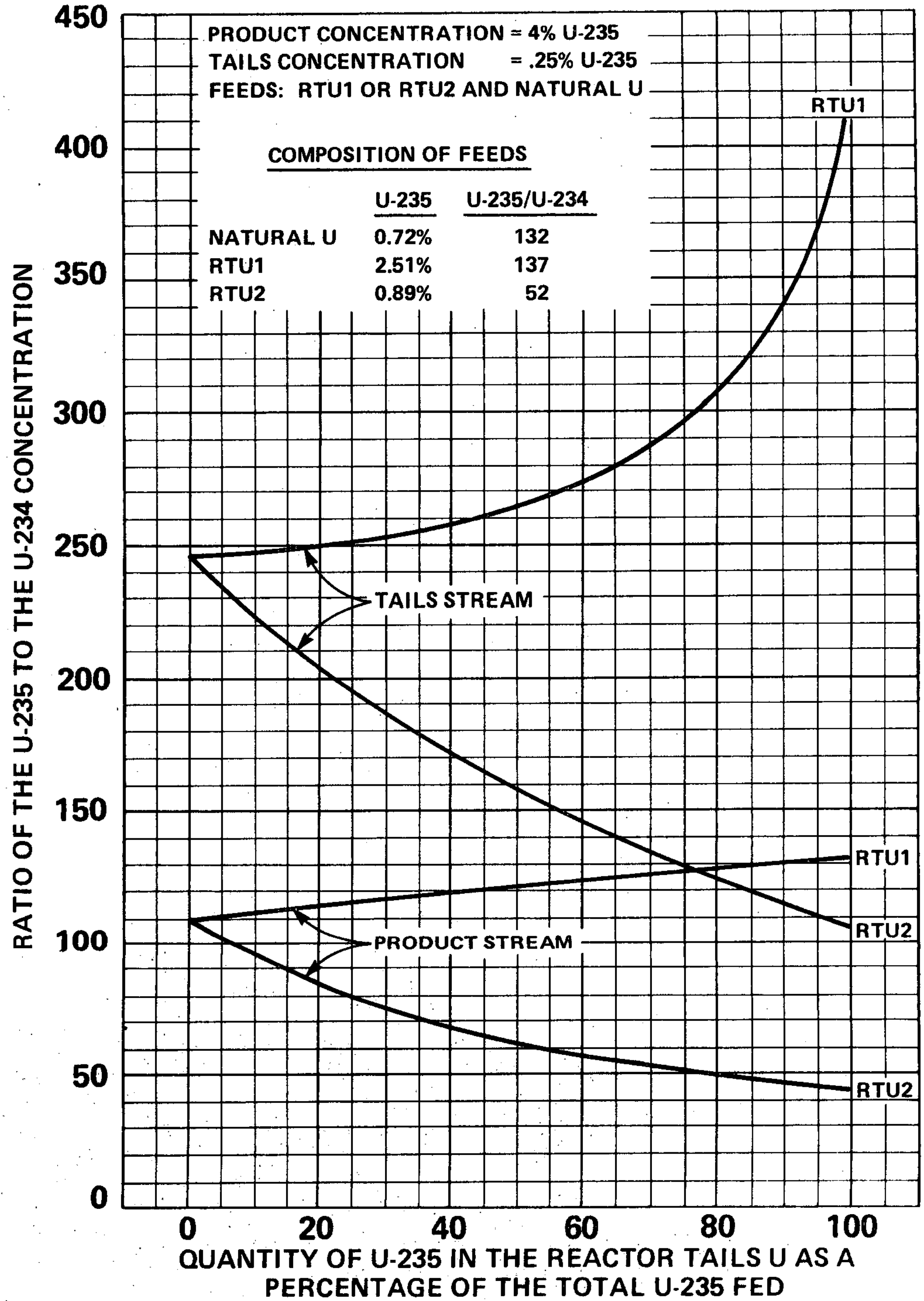

Figure 5

COMPARISON OF THE EFFECTS OF TWO SPECIES OF REACTOR TAILS URANIUM ON THE U-234 CONCENTRATIONS IN THE WITHDRAWAL STREAMS OF A FIXED CASCADE

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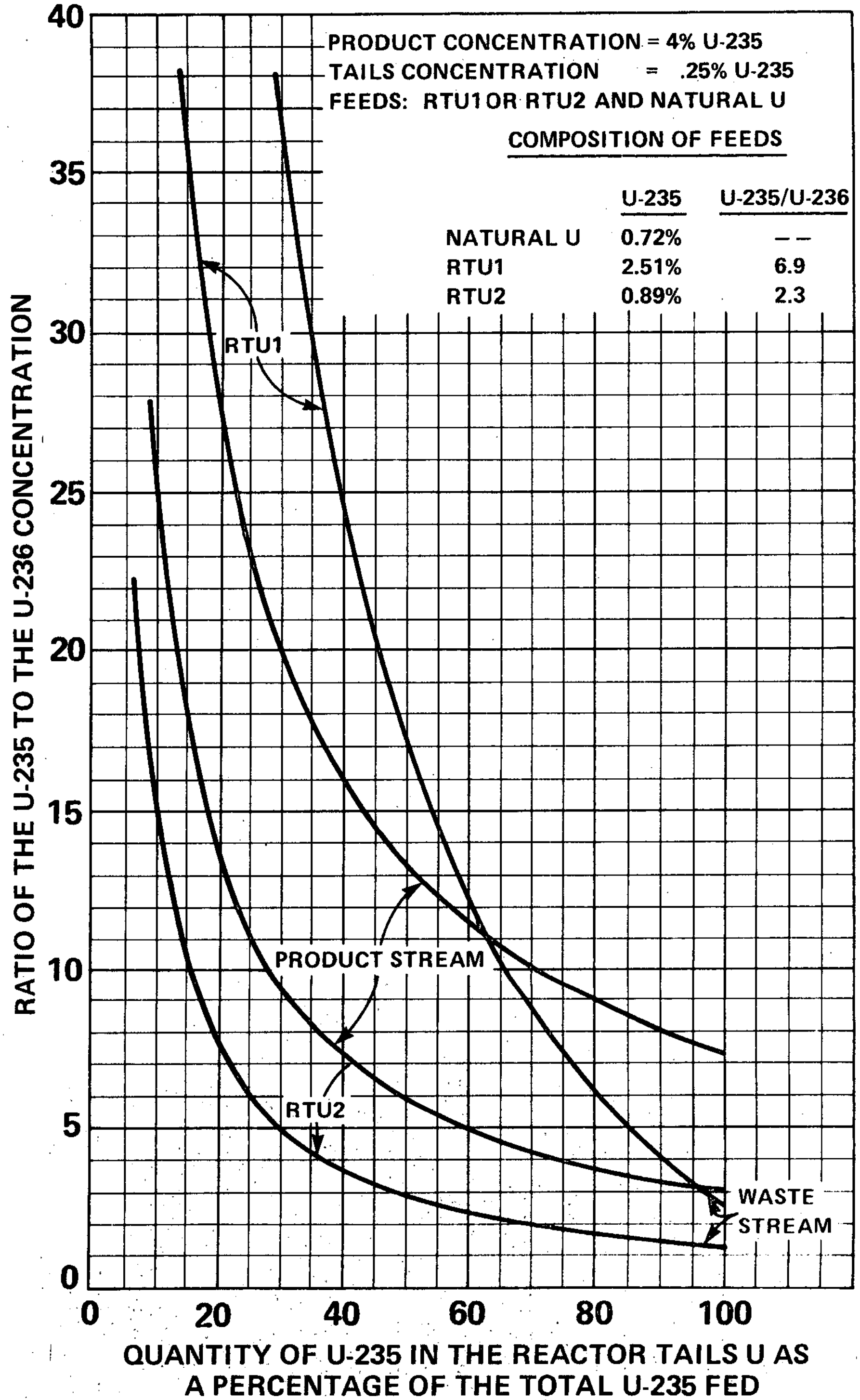

Figure 6

COMPARISON OF THE EFFECTS OF TWO SPECIES OF
REACTOR TAILS URANIUM ON THE U-236 CONCENTRATIONS IN THE
WITHDRAWAL STREAMS OF A FIXED CASCADE

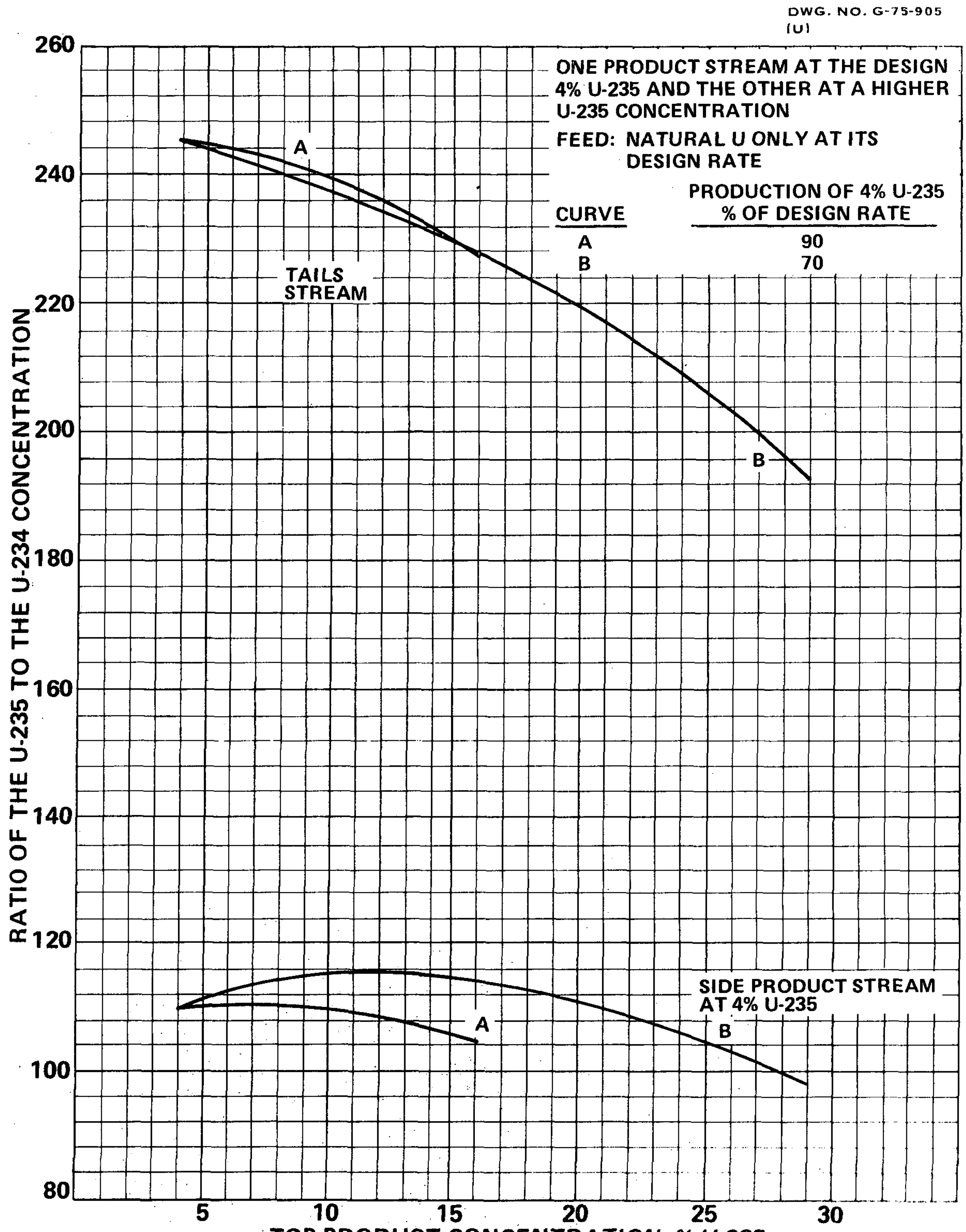

Figure 7

THE U-235 TO U-234 CONCENTRATION RATIOS
IN THE DESIGN PRODUCT STREAM AND THE TAILS STREAM
FOR TWO PRODUCT OPERATION

F. The cascade design only when it deviates considerably from ideality. In general, the steady-state ^{235}U concentrations relative to that of ^{235}U in the withdrawal streams of a squared-off cascade differ measurably from those of the corresponding ideal cascade only when the separative efficiency of the squared-off cascade is less than 90%. The ^{236}U concentrations are somewhat more sensitive, becoming measurably different when the separative efficiency of the cascade is less than 96%.

G. Process gas losses. Calculations show that the minor isotope concentrations relative to that of ^{235}U in cascade withdrawal streams are affected by process gas losses that are uniformly distributed along the cascade. This effect becomes barely measurable only when the loss rate is greater than about 1.5% of the cascade feed rate.

Minor Isotope Concentration Independence

The study has demonstrated that the steady-state concentrations of the uranium minor isotopes are independent of:

A. The stage separation factor. It has been shown that the minor isotope concentrations relative to that of ^{235}U in the withdrawal streams of a gaseous diffusion cascade and a gas centrifuge cascade are independent of the magnitude of the $^{235}U/^{238}U$ separation factor. Furthermore, the minor uranium isotope concentrations in the withdrawal streams of a gas centrifuge cascade are essentially identical with those of a gaseous diffusion cascade when the ^{235}U concentrations in the corresponding withdrawal streams are the same and the isotopic composition of any non-normal feeds and their feed rates in proportion to that of the natural feed are the same for both cascades. Consequently, the utilization of the uranium minor isotope concentrations in cascade feed and withdrawal streams as a safeguard technique will not require any disclosure of sensitive technology or proprietary data for the cascade being safeguarded.

B. The cascade size. The minor uranium isotope concentrations relative to that of ^{235}U in cascade streams are independent of the cascade size. Two well-designed cascades using feeds of the same isotopic composition and in the same proportion to natural feed and yielding the same ^{235}U concentrations in their withdrawal streams will have the same ^{234}U and ^{236}U concentrations in corresponding product and tails streams regardless of how much the separative capacity of one cascade is greater than that of the other.

CASCADE TEST DATA

The basic background information on the behavior of the minor uranium isotopes in separation cascades which has been presented was developed from solutions of sets of simultaneous multicomponent concentration gradient and material balance equations. It is therefore important to establish the validity of the reported calculations by comparisons of measured isotopic gradients with predicted values. Three plant tests have been conducted at the Oak Ridge Gaseous Diffusion Plant for this purpose. The sample data from the first test which was obtained by standard routine analyses on a single stage mass spectrometer proved to be too badly scattered to make a satisfactory comparison. However, very good agreement was obtained in the second test[2] and third tests [3] between the measured and predicted values for the ^{235}U to ^{234}U concentration ratios when the determinations were made by high precision scans on a three-stage mass spectrometer. The results for ^{234}U in the second test are presented in Figure 8. The observed ^{235}U to ^{236}U concentration ratios in the second test on the other hand showed a large deviation from the predicted values. This was due to the ^{236}U concentrations not being at steady-state when the samples were drawn from the cascade. Unfortunately, the ^{236}U concentrations in the several cylinders containing the partially enriched uranium fed to the cascade during the three days just prior to the time of sampling had varied by a factor of three. At the time of the cascade sampling at the start of the third test, the primary objective of which was to make an indirect measurement[4] of the cascade inventory by means of transiently enhanced ^{236}U concentrations, the ^{236}U concentrations along the cascade were everywhere < 10 ppm. Due to the imprecision of the analyses at such low concentrations, the ^{236}U data did not closely match the calculated cascade curve, but was scattered on both sides of it, thus grossly verifying it.

APPRAISAL AND APPLICABILITY OF MIST

The subject of utilizing measurements of the uranium minor isotopes as a safeguard technique has come to be known by its acronym: MIST. The data which have been obtained in this study definitely indicate that MIST has the potential of being a useful adjunct to, but not a substitute for, material accountability for safeguarding a ^{235}U enrichment facility. Because the ^{234}U and ^{236}U concentrations relative to that of ^{235}U in the cascade withdrawal streams reflect the cascade flowsheet[2] in a specific way, MIST can be quite valuable in confirming or contradicting the declared flowsheet and can serve as a means of determining the actual one when it fails to confirm the declared one. In order to utilize MIST, the flow rate of at least one stream (product, feed or tails) must be known. The most useful application of MIST would be for a multi-feed and/or multi-product case wherein there exists unlimited combinations of stream feed and withdrawal rates that will yield ^{235}U concentrations in the withdrawal streams that are unchanged from the declared values but minor isotope concentrations that vary with the stream rates.

Particular Qualifying Features of MIST

The calculations and correlations which have been made and reported demonstrate that the use of ^{234}U and ^{236}U concentration measurements in enrichment cascade feed and withdrawal streams has three features which qualify it as a safeguards technique. These are:

(1) The procedure does not require any disclosure of process technology or sensitive proprietary information.

(2) It requires data only on cascade external streams and therefore there would be no need for access to process equipment by safeguards inspectors.

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[2] In its usage in this study, the word *flowsheet* refers to the flow rate and the ^{235}U concentration of each of the cascade feed and withdrawal streams.

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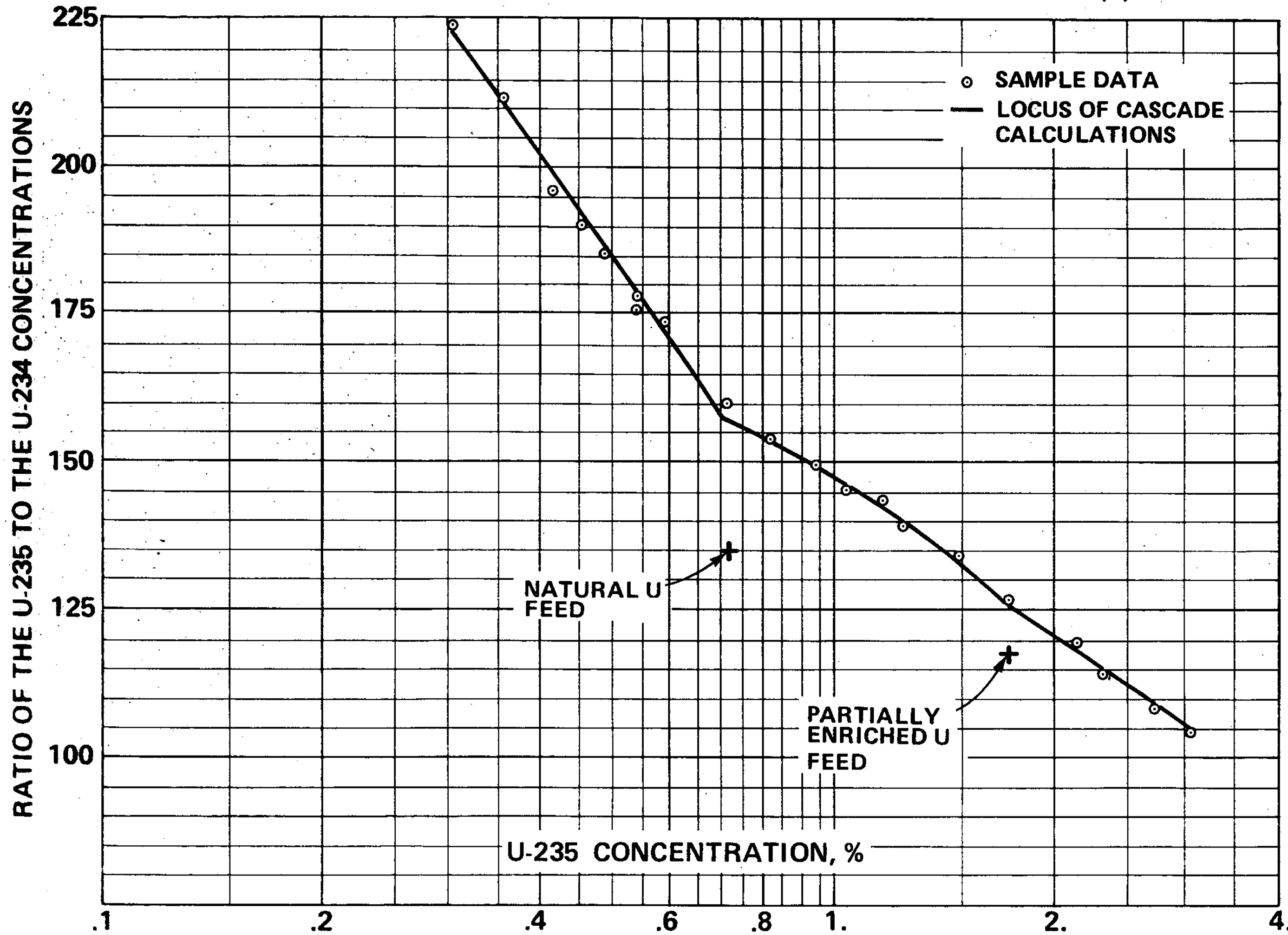

Figure 8

COMPARISON OF MEASURED AND CALCULATED
U-234 AND U-235 CASCADE CONCENTRATIONS

(3) The technique is applicable to both of the enrichment processes currently in use: gaseous diffusion and centrifugation.

Capability Required for the Application of MIST

An organization vested with the responsibility of applying MIST to a ^{235}U enrichment facility would have to possess personnel, equipment and know-how to perform the following tasks:

(1) Make sufficiently accurate analyses on cascade feed and withdrawal samples for the ^{234}U, ^{235}U, and ^{236}U concentrations therein.

(2) Design an optimum or nearly optimum squared-off cascade to do the separation job corresponding to the design flowsheet declared for the cascade under safeguards supervision.

(3) Carry out four-component concentration gradient and productivity calculations for various flowsheets for any specified cascade.

Application Costs

No attention has been given in this study to the cost of utilizing MIST. Matters such as the number of samples which will have to be analyzed, the cost of high precision isotopic analyses, the number of inspectors, requirements for back-up personnel and the cost of computing will determine the application cost. The number of inspectors and the numbers of samples required should not be in excess of what is necessary to carry out material accounting procedures. The incremental cost of MIST will largely be due to analytical costs and the requirements of back-up personnel to set up the appropriate calculations and interpret the results.

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[3] Results on cascade concentration gradient data for the third plant test have not been published.

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