Generalized Modeling of Enrichment Cascades That Include Minor Isotopes*

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Abstract

The monitoring of enrichment operations may require innovative analysis to allow for imperfect or missing data. The presence of minor isotopes may help or hurt—they can complicate a calculation or provide additional data to corroborate a calculation. However, they must be considered in a rigorous analysis, especially in cases involving reuse. This study considers matched-abundance-ratio cascades that involve at least three isotopes and allows generalized input that does not require all feed assays or the enrichment factor to be specified. Calculations are based on the equations developed for the MSTAR code but are generalized to allow input of various combinations of assays, flows, and other cascade properties. Traditional cascade models have required specification of the enrichment factor, all feed assays, and the product and waste assays of the primary enriched component. The calculation would then produce the numbers of stages in the enriching and stripping sections and the remaining assays in waste and product streams. In cases where the enrichment factor or feed assays were not known, analysis was difficult or impossible. However, if other quantities are known (e.g., additional assays in waste or product streams), a reliable calculation is still possible with the new code, but such nonstandard input may introduce additional numerical difficulties into the calculation. Thus, the minimum input requirements for a stable solution are discussed, and a sample problem with a non-unique solution is described. Both heuristic and mathematically required guidelines are given to assist the application of cascade modeling to situations involving such non-standard input. As a result, this work provides both a calculational tool and specific guidance for evaluation of enrichment cascades in which traditional input data are either flawed or unknown. It is useful for cases involving minor isotopes, especially if the minor isotope assays are desired (or required) to be important contributors to the overall analysis.

Introduction

There are a number of facilities around the world that are engaged in the process of enriching natural uranium for peaceful and non-peaceful purposes. Declared facilities are monitored under international safeguard agreements by the International Atomic Energy Agency (IAEA) via onsite inspections, nondestructive survey tools, material control and accountability procedures, as well as environmental sampling. These procedures are designed to ensure the material is being used for its intended and declared purpose and not for proliferation activities. Computational toolsets provide additional monitoring capability, since measurements of samples in or near a facility can be used along with model predictions to verify declared activities at the site.

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In general, monitored facilities publicly declare sufficient information to perform reliable verification calculations. Such information includes the feed assays for all components and the product and waste assays for the primary enriched component (^{235}U). Often additional information is also available, such as the enrichment factor or the number of stages in the cascade. However, there are occasions when such information is not available, in error, or suspect. In these cases it is desirable to use other information such as multiple assays in the product or feed stream in lieu of feed assays. It may also be possible to use minor isotope assays in place of the ^{235}U assays to verify (or contradict) declared operations.

This paper seeks to investigate alternatives to the traditional inputs, in order to understand the mathematical capabilities of simple cascade models. Recall that the traditional input is comprised of the enrichment factor, the feed assays for all components, and product and waste assays for ^{235}U. This work will evaluate many various alternative assay inputs, including those in which little is known about the feed stream or the primary enriched component. In such cases, a reliable description of the cascade will depend on minor isotope inventories, possibly in the product or waste streams instead of the feed. It will also assess possible alternatives if the enrichment factor is unknown. These possibilities will be illustrated by two examples that describe enrichment of natural uranium and enrichment of reprocessed fuel.

Cascade model

The model to be used is the matched-abundance-ratio cascade, which is a natural analog to the ideal cascade for problems that include more than two isotopes. We appropriate the algebraic form developed by Von Halle,[1] which has been encoded using the traditional input assumptions to produce the Original MSTAR code.[2] This work appropriates the same set of algebraic equations but configures them in such a way as to allow more flexibility in the input. Note that this model does not involve detailed machine performance but rather describes the system of identical machines that make up operational stages. Each stage may consist of multiple machines operating in parallel, with different stages connected in series. The code calculates the number of stages and stagewise flow rates and isotopic assays for a given enrichment operation.

We begin by defining the isotopic assays (mole fractions) and overall flow rates in feed, product, and waste streams:

x_i, y_i, z_i = assay in feed, product, and waste streams of isotope i
F, P, W = total flow rates in feed, product, and waste streams

Using molar and flow balances on different stages in both enriching and stripping sections of a cascade, von Halle derives algebraic equations that describe the relationship between these variables.[1] The reader is referred to his work, which is not repeated here. The resulting equations [see Eqs. (46) and (49) in Ref. 1 or in the Appendix to Ref. 2] are

$$\frac{P}{F} y_i = \frac{E_i x_i}{E_i + S_i} \quad \text{and} \quad \frac{W}{F} z_i = \frac{S_i x_i}{E_i + S_i}, \qquad i = 1, 2, \ldots, I, \tag{1a,b}$$

where I is the total number of isotopes considered. Also, E_i and S_i are special quantities involving the separation factors α_i and the numbers of stages N and M in the enriching and stripping sections, respectively:

$$E_i = \frac{\alpha_i^* - 1}{1 - (\alpha_i^*)^{-N}} \quad \text{and} \quad S_i = \frac{\alpha_i^* - 1}{(\alpha_i^*)^{M+1} - 1}\ , \quad i = 1, 2, \ldots, I, \tag{2a,b}$$

where $\alpha^* = \alpha\ (\alpha_j)^{-1/2}$ and the index j represents the component whose abundance ratio is matched (usually ^{235}U). In addition, the overall flow and molar balances for the entire cascade are easily seen to be

$$P + W = F, \quad P y_i + W z_i = F x_i\ , \quad i = 1, 2, \ldots, I. \tag{3a,b}$$

Likewise, the isotopic balances in each stream are straightforward:

$$\textstyle\sum_i x_i = 1\ , \qquad \sum_i y_i = 1\ , \qquad \sum_i z_i = 1\ . \tag{4a,b,c}$$

It is important to note that Eq. (3a) is not linearly independent of the others, since it can be obtained by summing Eq. (3b) over all isotopes (i.e., over the index i), and using Eqs. (4). We also note that adding Eq. (1a) to Eq. (1b) and simplifying yields Eq. (3b), so that only two of these three equations are independent. Finally, we recognize that dividing Eq. (3a) or Eq. (3b) by the feed flow yields the flow ratios P/F and W/F, which also appear in Eqs. (1). Hence, the problem only has two independent variables involving flow, namely, these two ratios (the flow rate, if given, is used only to carry units for the desired output). While the derivation of Eqs. (1) and (2) was done using discrete stages, the resulting mathematical formulation does not require the numbers of stages (N and M) to be integers. The ideal cascade itself is an approximation, and allowing non-integer numbers of stages does not compromise the value of this approximate solution. Finally, the separation factors for minor isotopes are determined using the well-known exponential relation

$$\alpha_i = \alpha^{\left[(M_k - M_i) / (M_k - M_j) \right]}\ , \tag{5}$$

where M_i = atomic weight of isotope i and α is the overall stage separation factor; j = index of the isotope whose abundance is matched in the cascade (usually ^{235}U); and k = index of the "key" component (usually ^{238}U), the isotope against which all others are compared.

Traditional input and output requirements are listed in Table 1. This formulation of the problem [i.e., Eqs. (1) – (4), together with the required inputs] allows for a generally stable algorithm that attains a reliable solution in a wide variety of problems.

Revised Calculational Approach

From a mathematical perspective, nothing in Eqs. (1) – (5) suggests that all feed assays are essential, or that any of them are. It might well be possible to substitute other assays (in product and waste) and still achieve a reliable solution. In addition, Eqs. (2a,b) suggest interrelationships

between α, N, and M so that numbers of stages might be given in lieu of the separation factor. An alternate viewpoint may be that the input described in Table 1 defines a "forward" problem that follows the flow of material through the system; it permits a unique and stable solution. Reliance on other inputs instead of those in Table 1 may constitute an "inverse" problem, whose solution may be elusive, unstable, or even non-unique. As we shall see in the second example below, there may be a small number of problems for which this viewpoint has merit.

Table 1. Traditional Input and Output Variables

Input Variables	**Description**
F or P	Feed or product stream flow rate (NOTE: feed rate must be negative)
x_i	Inlet feed assays (mole fractions) for all isotopes
y_j, z_j	Product and waste stream assays for ^{235}U
α	Stage separation factor (must be >1, usually $1 < \alpha < 2$)
Output Variables	
W and F or P	Waste stream flow rate, and either feed or product flow rate (whichever was not specified as input)
N, M	Number of stages in enriching and stripping sections
y_i, z_i	Product and waste assays for additional components (if more than two isotopes)
F_k, P_k, W_k	Stagewise flow rates at stage k
y_{ki}, x_{ki}, z_{ki}	Stagewise assays: mole fraction of isotope i in stage k

From Eqs. (1) – (5), there are $2I+3$ independent equations. The unknown variables are α, N, M, P, W, and the assays for feed, product, and waste streams, totaling $3I+5$ in all. It is thus apparent that, at a minimum, it is necessary to specify $(3I+5) - (2I+3) = I+2$ of these variables as input. It can also be shown, and has been verified in practice, that one of the inputs must be α, N, M, or $N+M$. The interchangeability of the other inputs is examined in the next section.

Since the set of variables specified as input is flexible, the resulting equations are (almost assuredly) nonlinear; hence, a simple algebraic solution algorithm is not practical. Instead, it is straightforward to solve them as a system of nonlinear equations using iterative methods. In this approach, the deviations of Eqs. (1) – (5) from equality form error residuals; these are squared and summed to obtain the familiar Sum-of-Squared-Error (SSE), which is minimized. The detailed numerical procedure is beyond the scope of this document, but relies on several algorithms from the optimization literature.

Sample Problems

In this section, problems are selected in order to understand the limits of generalized input. Hence, they reflect difficulties or unusual combinations of parameters. While some of the input combinations may not reflect normal applications, it is useful to understand the extent of the code's capabilities, and the circumstances under which it will fail.

Problem 1. This problem is a fairly simple problem in enrichment of natural uranium to 20% ^{235}U. A base case was run using the Traditional Input parameters, shown in bold in Table 2. Recall that for three isotopes (I = 3), it is necessary to supply I + 2 = 5 independent inputs. The base case does this by supplying α, all feed assays, and the product and waste assays for the matched component ^{235}U. Other parameters were then determined by running the code.

Table 2. Cascade Parameters for Problem 1

Overall Cascade				Assays		
Variable	Value		Isotope	Feed	Product	Waste
α	**1.5**		^{234}U	**0.000055**	0.0020161	1.3214E-5
N	17.537		^{235}U	**0.00711**	**0.2**	**0.003**
M	3.2769		^{238}U	0.992835	0.7979839	0.9969868
P	0.020863					
W	0.979137					

Assumes feed flow F=1.

Testing of different input parameter combinations is done by substituting other parameters for those in bold and calculating the remaining cascade parameters. Virtually every possible combination of parameters is considered in order to assess the essential requirements, and to gauge the merits of different input combinations. In most cases, the algorithm produced excellent results (at least 5 digits of accuracy, which was the limit of the output verification). In a small number of cases, the calculation produced parameters of lower accuracy, but still fairly good representations of the original cascade. A small number of cases either produced high error or failed to converge. The failed cases constitute guidance for limitations on the flexibility of input information and help to establish minimal requirements for input. The following are summaries of various input combinations and the results they achieved.

1. Input: 4 assays and 1 from the Overall Cascade group (Columns 1 and 2) of Table 2. If the fifth input was α, N, M, or N+M, then all results were excellent (good to 5 significant figures), regardless of which assays were specified. If P or W was specified instead of α, N, M, or N+M, all results were very bad—no convergence or very large error.

2. Input: 3 assays and 2 from the Overall Cascade group. Most cases yield excellent results; however, there are more failures than in Case 1 above. These can be generalized as follows:

a. All assays are for the same isotope—failure always occurs. There is no unique distribution of the remaining two isotopes.

b. ^{234}U assay given in both Feed and Product—always fails if P or W is input, regardless of third assay. The reason is not exactly clear, since the same is not true for ^{235}U and ^{238}U.

c. The following combinations fail for understandable reasons:

1) α, N given—fails if no assay is in waste stream

2) α, *M* given—fails if no assay is in the product stream
3) α, *N*+*M* given—fails if no assay is in the feed stream

d. No ^{234}U assay given—usually fails. Some cases do work; it is unclear why most do not.

3. Input: 2 assays, α, *N*, and *M*. Very good or excellent results, except for situations analogous to a. and d. from case 2 above.

4. Input: 5 assays and nothing else. Results always fail.

Exhaustive simulations have not been run for more than three isotopes, although some cases are considered in the next section. However, as a precaution and aid to users, all of these situations form the basis for error or warning messages in the code.

Problem 2. This problem involves four isotopes and is analogous to enrichment of reprocessed uranium to highly enriched uranium (HEU). In addition, the enrichment is driven to the point where the key component (^{238}U) is very small, which presents difficulties for the solution algorithm. All possible input options have not been evaluated, as this would require more than 12,000 combinations. However, a wide variety of assay inputs have been examined. As done for Problem 1, the base case is consistent with the Traditional Input, except that the specified waste assay is ^{238}U instead of ^{235}U. Input quantities are shown in bold in Table 3; the other values in the table were computed using these inputs. The waste assay of ^{238}U is specified instead of that for ^{235}U simply to illustrate the variety of possible inputs.

Table 3. Cascade Parameters for Problem 2

Overall Cascade				Assays		
Variable	Value		Isotope	Feed	Product	Waste
α	**1.81**		^{234}U	**0.00025**	0.013857	2.2215E-6
N	31.537		^{235}U	**0.015**	**0.79**	0.000888
M	8.5893		^{236}U	**0.0055**	0.19168	0.0021098
P	0.017883		^{238}U	0.97925	0.0044589	**0.997**
W	0.98212					

The feed stream assays are similar to what might be expected from reprocessed light water reactor (LWR) fuel in that inventories of minor isotopes ^{234}U and ^{236}U are more significant. In fact, they prevent further enrichment of ^{235}U past the 79% indicated in the table, since they make up nearly all of the remaining uranium present in the product stream. The key component ^{238}U is perilously close to zero, which creates some numerical difficulties since it appears in the denominator of assay ratios used in von Halle's formulation.

This example presents another difficulty not obvious from Table 3 but illustrated well in Figure 1. Shown are the assays for product streams from each stage of the enriching section. All three of the lighter components are enriched relative to the key component ^{238}U (not shown), and so all three

initially increase. However, as the stages increase, the amount of ^{238}U decreases sufficiently that eventually ^{234}U and ^{235}U begin to enrich relative to ^{236}U. At this point, the ^{236}U product assays begin to decrease (the maximum is reached at stage 21, with an assay of 0.29). This behavior is both reasonable and understandable, but it presents a difficulty in establishing cascade information from minor isotope assays. For example, if the only cascade product information is the ^{236}U assay, the solution is not unique for a large range of values. The value from Table 3 of ^{236}U = 0.19168 occurs at the exit of the highest stage (stage 31.537); it also occurs at the exit of Stage 13. With no other information, it is impossible to know which stage is truly the highest one in the cascade.

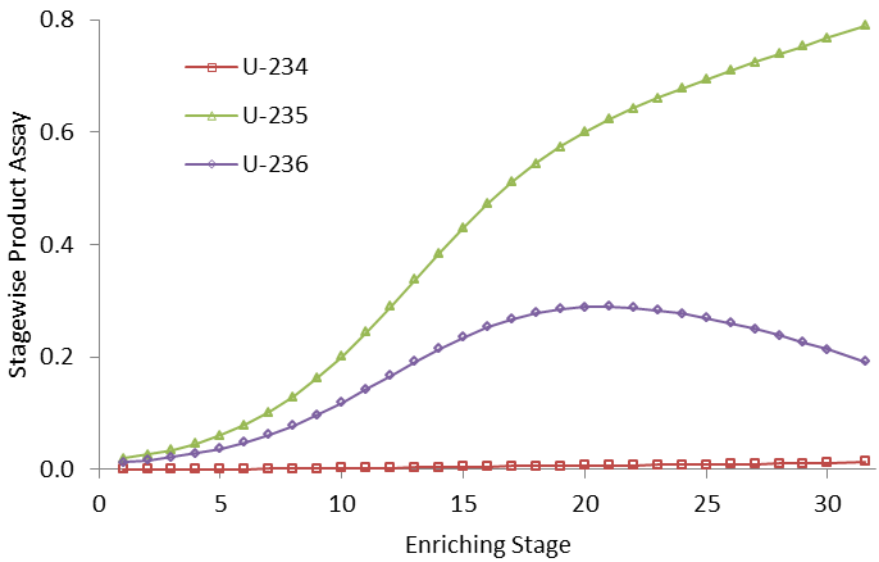

Figure 1. Problem 2: Stagewise Product Assays in Enriching Section

To evaluate the flexible input option for Problem 2, every combination of five assays was considered that included at least one assay for each isotope. Also specified was the enrichment factor α =1.81. This approach resulted in 180 trials (including the base case), of which nearly all matched the base case to four or five significant figures. In a small number of cases, one assay would match to only two or three significant figures. In eight cases, the product assay for ^{238}U exhibited between 10 and 60% error (actual value between 0.0035 and 0.0055), yet all other parameters had less than 1% error. This level of error for ^{238}U is fairly inconsequential in actual applications, since reduction to this extremely low level is not likely to occur. Also, the primary concerns are not precision of the low ^{238}U inventory but rather the elevated amounts of other components, which were estimated reliably. Finally, a few cases were randomly selected to use N, M, or N+M in place of α, and gave very similar results. From the evaluation of Problem 1, it seems apparent that the four quantities α, N, M, and N+M are routinely interchangeable in almost all input schemes, and this observation was apparent in the evaluation of Problem 2 as well.

In these trials, there were 16 results that did not converge at all to the values in Table 3 but were likely affected by the non-uniqueness issues noted in Fig. 1. The input assays for these 16 results are

given in Table 4, where the headings 4, 5, 6, and 8 refer to isotopes as noted in the table. In about half of the cases, a ^{236}U assay is the lone assay given for a particular stream, usually in the product stream. In the other cases, it is accompanied by another assay that is not a principal assay in that stream, usually ^{234}U. In most cases, the number of stages is between 19–23, although in one case (row 10) the confusion caused a perturbation of the number of stages in the stripping section instead. In every one of these cases, the Sum-of-Squared-Error (SSE) of the minimization procedure was very low, indicating a legitimate solution for the given data. Thus, under circumstances illustrated in Fig. 1, the possibility of multiple solutions under generalized input cannot be ignored.

Table 4. Input Assays for Cases Not Converged to Original Problem

Feed[a]					Product					Waste				
4	**5**	**6**	**8**		**4**	**5**	**6**	**8**		**4**	**5**	**6**	**8**	*N*[b]
		x			x					x	x	x		23
x							x			x	x	x		23
	x						x			x	x	x		23
		x					x			x	x	x		19
x	x						x			x			x	23
x	x						x				x		x	23
x	x						x					x	x	23
x			x		x	x						x		21
x			x				x				x		x	22
	x	x					x	x		x				M=13.7
		x			x	x				x			x	14
x					**x**		**x**				**x**		**x**	**19.513**
		x			x		x				x		x	20
	x				x		x				x		x	23
	x				x		x					x	x	23
			x		x		x				x		x	22

[a] Heading numbers refer to uranium isotopes; that is, "**4**" = ^{234}U, "**5**" = ^{235}U, etc.
[b] N = number of enriching stages in final converged solution (exact solution: N = 31.537)

Because of this non-uniqueness concern, one of these cases was examined in greater detail by supplying additional information. The case selected is that described by row 12 in Table 4 (highlighted in blue boldface), with ^{234}U given in feed and product, ^{236}U given in product, and ^{235}U and ^{238}U given in waste. In addition to specifying α and five assays, the number of enriching stages N was imposed as an additional input parameter. (The resulting system is overspecified, but since the algorithm is configured as a nonlinear minimization problem, a solution is still straightforward.) A number of different values for N were used, and the results were judged by examining the Sum-of-Squared-Error (SSE) for the numerical optimization. These results are shown in Fig. 2 (note the logarithmic scale), where it is seen that a very low SSE occurs in two places: N=19.513 and

N=31.537. The former is the value obtained by the code during its normal iteration process without any specification of *N*. The latter is, of course, the correct solution that matches the original problem.

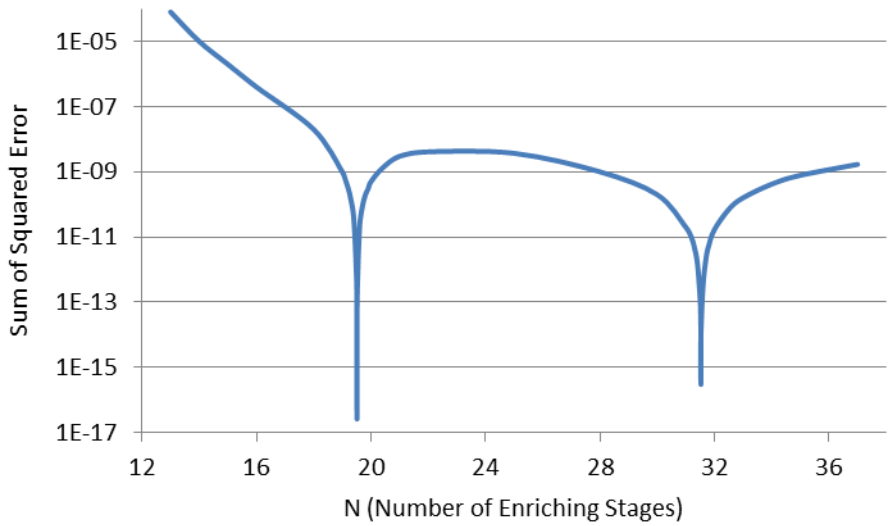

Figure 2. Problem 2 Error Analysis for Double Solution Case

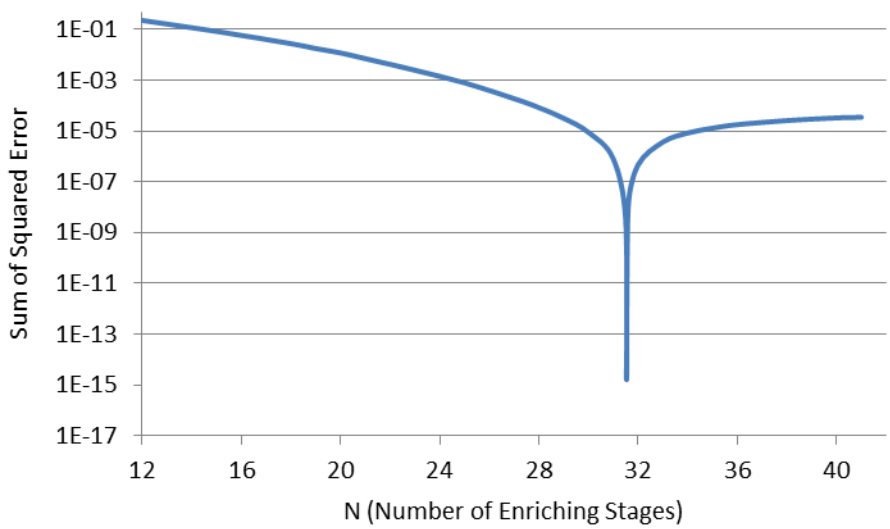

Figure 3. Problem 2 Error Analysis for Single Solution Case

Since a double solution exists in this case, an additional check was made for a case in which the original calculation produced excellent results. This case was very similar to the base case, differing only in that the waste assay for ^{235}U was given instead of that for ^{238}U. Imposing varying values of *N* as additional input and recording the SSE for various values of *N* gave the results shown in Fig. 3. From the figure, it appears that a single solution exists, coinciding exactly with the value of *N*=31.537 determined from the original solution. No other cases were examined, but it is assumed that the solutions near the exact solution are all single solution points.

Summary and Conclusions

The algebraic formulation of von Halle for a mixed-abundance-ratio cascade formed the basis for a new algorithm which allows more general and flexible input. Extensive trials with the more general input options generally produced excellent results, although there were a few notable exceptions. The new method is illustrated with two sample problems, one for enriching natural uranium to 20%, the other for enriching reprocessed uranium to a very high (79%) level. Numerous trials with both problems were conducted in order to push the limits of the algorithm and identify any difficulties in obtaining a solution.

From Problem 1, certain input combinations were insufficient to obtain a solution, and these are noted in the section describing this problem. Analysis of Problem 2 illustrated that unusual circumstances in the problem itself may create a non-unique solution for certain input combinations. This situation reflects difficulties that can arise generally with "inverse problems," as opposed to forward problem formulations. The sample problems illustrate the situations in which generalized input is likely to succeed. The example with three isotopes (Problem 1) covered nearly every input combination. However, the example with four isotopes (Problem 2) covered only a subset of different assay combinations even though 180 cases were investigated. In general, the method was successful for most inputs, even those that were unusual and notably difficult to solve.

References

1. E. Von Halle, "Multicomponent Isotope Separation in Matched Abundance Ratio Cascades Composed of Stages with Large Separation Factors," pp. 325-56 in K. G. Roesner, Ed., *Proceedings of the 1st Workshop on Separation Phenomena in Liquids and Gases*, Institut für Mechanik, Darmstadt, FRG (1987).

2. D. F. Starr, *MSTAR-IAEA Enrichment Code User Guide*, K/NSP-386, National Security Programs Office, Oak Ridge K-25 Site (June 1996).

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