MSTAR2019: URANIUM ENRICHMENT CASCADES WITH SIDESTREAMS

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ABSTRACT

The MSTAR2019 code, an ideal cascade model of uranium enrichment that includes sidestreams, has recently been delivered to the environmental sampling unit at the International Atomic Energy Agency (IAEA). The code builds on previous versions of the MSTAR code, but now includes options for a side feed and/or side product streams. Based on equations derived by Ed Von Halle, the current version includes several additional features such as flexible input options and integer numbers of stages. Applications that are relevant to safeguards analysis of enrichment plants are demonstrated, including enrichment of natural uranium to reactor fuel or to high enrichments, enrichment of recycled uranium, and various simulations of undeclared operations. Finally, this work describes limitations of ideal cascade models generally and discusses unusual situations and how they could be simulated using this code.

INTRODUCTION

Facilities around the world are engaged in the process of enriching natural uranium. The facilities in countries that have signed international safeguards agreements with the IAEA are monitored via an assortment of technical measures, including on-site inspections, nondestructive survey tools, material balance evaluations, design information verification, as well as environmental sampling. These procedures are designed to assure the material is being used for its intended and declared purpose and not for proliferation activities. The use of computational tool sets allows for the confirmation of declared activities in many of these areas, particularly in environmental sampling where measurements of small samples in or near a facility can be combined with model predictions to verify declared activities at the site. It is particularly useful for IAEA staff to be able to run numerical simulations of undeclared behavior as well as normal operations. Hence, the ability to simulate an enrichment plant employing a side feed or side product stream is a necessary tool.

Shown in Figure 1 is a simplified schematic of connections between stages in an enrichment facility. Cascade models do not involve detailed machine performance but assume basic attributes of machine operations that are obtained from testing data or from simulations of machine performance. They assume that operational stages are made up of a number of identical machines operating in parallel, and that different stages are connected in series. The more machines there are in a stage, the greater throughput can be achieved. Each stage increases the product enrichment, so more stages result in higher overall enrichment. The concept of an *ideal cascade* presents both a simple and an optimized description of cascade performance for separation of ^{235}U from ^{238}U. The ideal cascade assumes that no mixing losses occur when a depleted stream from a higher stage is combined with an enriched stream from a lower stage (see Figure 1); hence, it constitutes the most efficient possible operation and is generally a very good approximation to commercial facilities. The ideal cascade model also simplifies calculations because the assumption of equality for these two streams reduces the number of variables that must be calculated. When more than two isotopes are considered, the assumptions of the ideal cascade result in a matched-abundance-ratio cascade

(MARC), in which mixing losses can be eliminated for only one isotope. This isotope is termed the "matched" isotope and is almost always ^{235}U. The additional isotopes create more complexity in modeling, but MARC still represents a simplification of the enrichment processes. Both the ideal cascade and MARC also assume that the separation performance at each stage is the same (i.e., the separation factor is constant for all stages of the cascade).

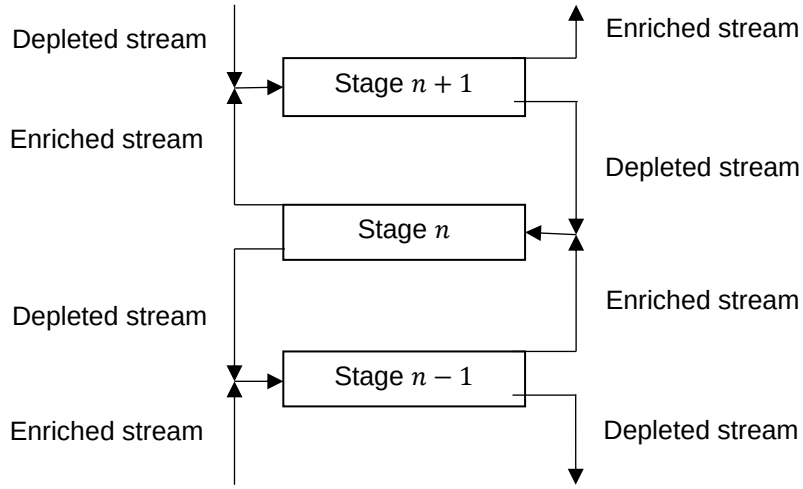

Figure 1. Stage arrangement schematic

Early efforts in cascade modeling were led by Ed Von Halle, who derived algebraic equations for cascades of machines with large separation factors [1]. He did additional development to include side feed and side product streams into his MARC model [2]. This work was innovative and elegant but apparently was never published or presented in an open forum. However, it formed the basis for the original MSTAR code [3], written in Visual Basic and distributed to the IAEA in 1996 as a deliverable of the U.S. Support Program. In 2012, an update to the MSTAR code was commissioned by the IAEA [4], which relaxed some of the strict input requirements and provided code that would operate on modern operating systems including Windows 10, Linux, and Mac. The input flexibility was described in an open publication [5], and limitations to the basic cascade model were noted therein. Unlike the original code, the MSTAR'12 code did not include sidestreams, which are a valuable resource much desired by IAEA.

This work describes an update to the MSTAR'12 code that preserves the generality of input and also includes sidestreams. The basic mathematical formulation relies on the previously unpublished derivations of Von Halle [2] and is described briefly in the next section; a full detailed description can be found elsewhere [6]. The remainder of this paper illustrates some of the capabilities and results of MSTAR2019 code calculations.

IDEAL CASCADE MODEL

The MSTAR2019 code calculates the number of stages, stagewise flow rates, and isotopic assays for a given enrichment operation. It is based on standard mass balances, as is common in chemical process modeling (i.e., isotopic mass and flow balances are taken over different control volumes in the system). The code is configured to separation of uranium isotopes ^{232}U, ^{234}U, ^{235}U, ^{236}U, and ^{238}U, although the numerical algorithm could handle any number of isotopes. The partitioning of

isotopes in a single separating machine is determined by a *separation factor*α, assumed constant throughout the plant.

The separation factors for different isotopes are related by the following formula:

$$\ln(\alpha_i) = \left[\frac{M_k - M_i}{M_k - M_m}\right] \ln(\alpha), \qquad i = 1, 2, \ldots, I, \tag{1}$$

where
$M_i =$ atomic weight of isotope i,
$m =$ index of the isotope whose abundance is matched in the cascade (usually ^{235}U),
$k =$ index of the *key* component (usually ^{238}U), the isotope against which all others are compared.

It is easy to see from Eq. (1) that the quoted separation factor α is identical to the separation factor for the matched component α_m. This usage was justified in an early report by Blumkin and Von Halle [7], which was part of a series known as the MIST reports (Minor Isotope Data as a Safeguards Technique). The appropriateness of Eq. (1) was also established by earlier references using only two components [8,9] and Harink-Snijders [10], who does so for three components.

Table 1 lists variables associated with external streams and the stages where they enter or exit the cascade. Shown in Figure 2 are the principal regions and a schematic illustrating these variables in an enrichment cascade with sidestreams. The waste (or tails) stream is always at stage 1. As depicted in Figure 2, all flow rates are shown as leaving the cascade; hence, feed rates (F and E) need to be negative numbers for calculational purposes. Thus, the arrows showing exiting streams should actually be reversed to indicate streams entering the cascade. However, as entering streams, they would flow directly into the feed of the next higher stage. Hence, under conventional parlance, the main feed stage is $N_F + 1$, and the side feed stage is $N_E + 1$. The distinction between side and main feeds is somewhat academic—both are feed streams, and we usually consider the main feed to be the largest. The ordering above suggests that the side feed is located below the main feed, but it could just as well occur above it.

Table 1. Principal variables

External stream	Assays	Stage numbers	Flow rates
Top product	X_{iP}	N_P	P
Side product	X_{iG}	N_G	G
Main feed	X_{iF}	N_F	F
Side feed	X_{iE}	N_E	E
Waste (or tails)	X_{iW}	1	W

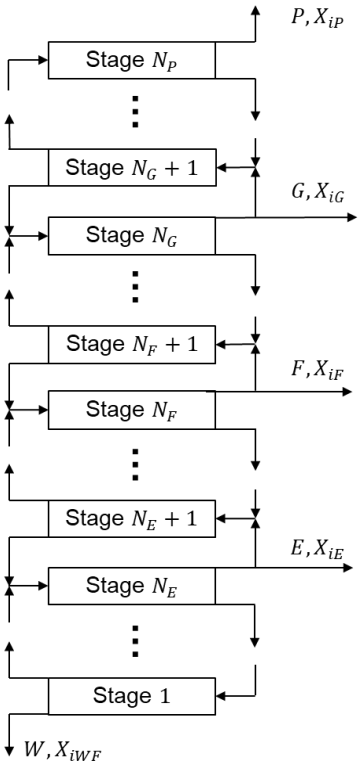

Figure 2. Cascade external streams and variables

The flow rates represent mass or molar flows of UF_6 and can be given in any useful unit such as kilograms per year (kg/y), milligrams per second (mg/s), or moles per hour (mol/h). The interchangeability of mass and molar flow units may seem incongruous on the surface. The internal calculations are based on gas dynamics and therefore use molar balances. Enrichment capability is usually related to mass of natural uranium feed, an easily measured quantity; hence, mass flow terms are often preferred for practicality. The molecular weights of UF_6 for the two principal isotopes are 349 and 352 (for ^{235}U and ^{238}U, respectively). The relative difference in these values is small enough that mass and mole fractions are nearly identical and can be interchanged with little impact on the accuracy (or utility) of calculations.

Real enrichment cascades have integer numbers of stages and integer numbers of machines in each stage. A significant consequence of the ideal cascade assumption is that these variables cease to be integers and must be treated as real numbers. If integer numbers of stages are desired, a modification is discussed as one of the example problems. However, for most examples, noninteger results occur, and must be treated as approximations, which is of course, exactly what they are.

SAMPLE PROBLEMS

In this section, several sample problems illustrate the capabilities and flexibility of the code. These include standard problems typical of declared facilities, as well as undeclared operations and

unusual calculations. Some of the bounds and limitations on possible calculations are also presented.

Standard Production of Low-Enriched Uranium

This example consists of natural uranium enriched to 3.5% with a waste stream of 0.35% in ^{235}U, a typical scenario for declared enrichment operations. The solid black line in Figure 3 shows the stagewise feed flow rates, normalized to unit flow of main feed into the cascade. If we add a side feed of 0.1 (one-tenth of the main feed rate), the upper dashed curve results. As seen in the figure, all stages exhibit higher stagewise flow rates. If a side product is considered instead of side feed, the lower dotted curve results. The side stream locations are characterized by non-smooth behavior of the rate curves (at 2.6 for side feed, and 9.5 for side product).

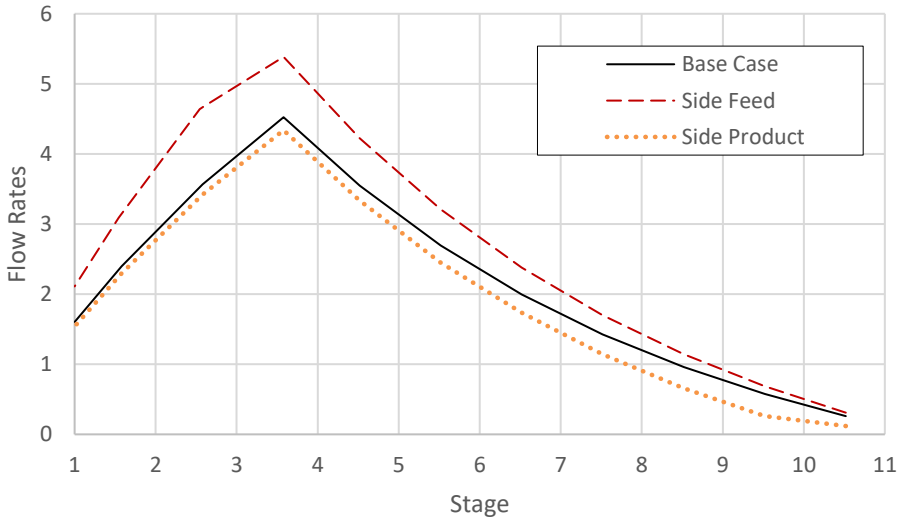

Figure 3. Flow rate comparison

The ^{235}U assays for these three cases are nearly identical, as the feed, product, and tails have been specified as input. However, the ^{234}U values do differ somewhat—the cascade product is about 4% higher for the side product case and 2% lower for the side feed case.

Integer Stages. Figure 3 clearly illustrates that some of the stage numbers are non-integer—the feed stage is 3.6 and the product stage is 10.5. This is an artifact of the ideal cascade assumptions but does not reflect actual enrichment facilities, which would always have an integer number of stages. If we desire an integer number of stages, then we must relax some other constraint; otherwise, no solution exists. In many applications it is practical to relax the specification of product and tails assays, which can be done for the current example as an illustration.

When specifying integer stages with no sidestreams, the MSTAR2019 model differs from the base case as shown in Table 2. The stage numbers are integers as desired. However, the ^{235}U assays do vary slightly from their specified values. Minor variations in other parameters also occur.

Table 2. Comparison of base case with integer stages

	Base case	Integer stages
Number of stages	10.52	11
Feed stage	3.58	4
Product ^{235}U assay	3.5	3.54
Tails ^{235}U assay	0.35	0.322

Nonstandard Input. We have mentioned that MSTAR2019 includes input flexibility, and here we present several examples of this feature. The standard input assays are ^{235}U in tails, product, and side products, together with all assays in main feed and side feed. It is possible to use different combinations of assays if necessary, although the combinations are subject to the following constraints:

1) Every isotope must have at least one assay specified in some stream.
2) Every external stream must have an assay or stage number specified.

For example, we can substitute the minor isotope ^{234}U for ^{235}U in tails and product assays, and the code will provide results nearly identical to those with ^{235}U specified. Various other combinations are also possible and will almost always provide good results.

The MSTAR model uses assays in the external streams to calculate the stage numbers where these streams should occur. However, it is also possible to perform an inverse calculation—specify the stage numbers, and the code will calculate the assays for streams at those stages. Thus, if we specify integer stages and the stage numbers from Table 2 for product and feed stages, then we get exactly the assays shown in Table 2.

Production of Highly Enriched Uranium from Recycled Uranium

Enrichment of recycled uranium will include noticeable amounts of ^{236}U, and this can complicate the evaluation of scenarios. In this section, we investigate enrichment far beyond declarations to produce a product of highly enriched uranium. This is done through two scenarios: by using a single cascade configured to produce highly enriched uranium and by using successively higher enrichments as feed. The starting material is listed in the feed column of Table 3.

Table 3. Recycled uranium to high enrichment

	Tails	Feed	Product
^{234}U	0.00524	***0.02***	2.57
^{235}U	***0.35***	***0.78***	***75.0***
^{236}U	0.372	***0.49***	20.93
^{238}U	99.2732	***98.71***	1.50
Flow rate	0.99424	***1***	0.00576
Stage number	1	4	46

Note: Quantities in bold italics were supplied as input.

Single Cascade. Rather than the default value of 3.5% enrichment, we specify a top product of 75%. The resulting cascade is depicted in Table 3 and contains more than 43 stages in the enriching section and almost 3 in the stripping section. More than 99% of the flow is out the tails, as the product flow rate is quite small. Note, the ^{238}U assay in the product stream has been reduced to only 1.5% and cannot be reduced much further without incurring an error. This effect precludes enrichment to a product assay much higher than 75%.

Batch Recycle. We again consider input of recycled uranium as feed (feed assays from Table 3) and enrich to 3.5%; the full cascade results are given in the first row of Table 4. However, we now take this standard (declared) product of 3.5% enrichment and use it as feed into another cascade identical to the first. Results are shown in the second row and indicate that a product of nearly 14% enrichment is produced. In the same way, this 14% product is again used as feed to an identical cascade, the result is a product of almost 40% enrichment (third row of Table 4). An additional enrichment of this feed produces a product of 67% enrichment. If the product at 67% enrichment is used as feed in an attempt to produce an even higher enrichment, an error occurs. This is the same difficulty that was mentioned previously, where enrichment in a single cascade beyond 75% ^{235}U was problematic.

Table 4. Batch recycle to obtain highly enriched uranium from reprocessed uranium

Feed description	Product assays (%)		
	^{234}U	^{235}U	^{236}U
1. Initial recycled U	0.11058	3.5	1.56023
2. Product from step 1	0.544	13.9739	4.42038
3. Product from step 2	1.90771	39.7702	8.92732
4. Product from step 3	3.98201	67.3713	10.7315
5. Product from step 4	Error incurred—key isotope too small		

This problem only occurs with reprocessed feed where ^{236}U has a sizable inventory, and the ^{238}U inventory gets very low. The model describes all enrichment relative to a key isotope, which is almost always ^{238}U. Hence, a major computed quantity is the abundance ratio—the inventory of any isotope divided by the inventory of the key isotope. Because the key isotope is in the denominator of a fraction, when it becomes very small, a singularity develops. See [6] for a more thorough discussion of this issue.

SUMMARY AND CONCLUSIONS

The MSTAR2019 model has been described briefly and illustrated on examples for enriching natural uranium and reprocessed uranium. A short overview of enrichment cascades and the assumptions of the ideal cascade model has been included. References for the mathematical derivations were supplied and have followed the original equations of Von Halle [5], which for the basis of the original MSTAR code. The current version includes sidestreams and the flexible input options, and extensive quality analysis has been performed. The code itself has been supplied to the environmental sampling team at the IAEA to assist monitoring of declared facilities.

ACKNOWLEDGEMENT

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