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Minor isotope safeguards techniques (MIST): Analysis and visualization of gas centrifuge enrichment plant process data using the MSTAR model

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ABSTRACT

This paper presents a development related to the use of minor isotope safeguards techniques (MIST) and the MSTAR cascade model as it relates to the application of international nuclear safeguards at gas centrifuge enrichment plants (GCEPs). The product of this paper is a derivation of the universal and dimensionless MSTAR cascade model. The new model can be used to calculate the minor uranium isotope concentrations in GCEP product and tails streams or to analyze, visualize, and interpret GCEP process data as part of MIST. Applications of the new model include the detection of undeclared feed and withdrawal streams at GCEPs when used in conjunction with UF_6 sampling and/or other isotopic measurement techniques.

1. Introduction

This paper presents a development related to the use of minor isotope safeguards techniques (MIST) and the MSTAR cascade model as it relates to the application of international nuclear safeguards at gas centrifuge enrichment plants (GCEPs). MIST is an umbrella term for the variety of ways that the minor uranium isotopes (i.e., ^{232}U, ^{234}U, and ^{236}U) can be utilized in safeguards, typically with an emphasis on GCEP safeguards.

The product of this paper is a universal and dimensionless form of the existing MSTAR model. The new model can be used to calculate the minor uranium isotope concentrations in GCEP product and tails streams or to analyze, visualize, and interpret GCEP process data as part of MIST.

Any changes in GCEP operation (e.g., process flow rates; feed, product, or tails concentrations; addition of side feed and withdrawal streams) will change the minor uranium isotope concentrations of the product and tails streams. MIST, using the new model, could be used to monitor and interpret these characteristic changes to detect undeclared activities at GCEPs. The need for such tools and techniques is outlined in the International Atomic Energy Agency (IAEA) Department of Safeguards Long-Term R&D Plan [1] and the IAEA safeguards objectives for GCEPs [2].

This paper begins by briefly introducing the reader to the background of MIST. The core of the paper is a derivation of the universal and dimensionless MSTAR cascade model. The paper concludes with a demonstration of how MIST, using the new MSTAR model in conjunction with the destructive analysis of UF_6 process samples, could be implemented to monitor GCEPs.

2. Background

Natural uranium is nominally 0.7200 at% ^{235}U, 0.005420 at% ^{234}U, and 99.2745 at% ^{238}U [3]. While the ^{235}U concentration in natural uranium varies from 0.7198 to 0.7207 at%, a variation of ~0.13%, the ^{234}U concentration varies from 0.0050 to 0.0059 at%, a variation of ~17% [3]. ^{234}U concentrations above this range have been reported for in-situ leach mined samples [4]. ^{232}U and ^{236}U are not found in natural uranium but are present in uranium recycled from reactor tails. In reactor tails, ^{232}U appears in trace quantities while ^{236}U concentrations are on the order of 0.1 to 1 at% [5,6].

^{232}U, ^{234}U, and ^{236}U can be thought of as tracers present in the nuclear fuel cycle. Because ^{234}U is the only naturally occurring tracer and is always present in the feed material for enrichment plants, it will be the focus of the remainder of this paper.

In centrifuge enrichment cascades, ^{234}U enriches 'faster' than ^{235}U and consequently the ^{235}U-to-^{234}U ratio of the feed material is altered by the process. This results in the ^{235}U-to-^{234}U ratio of the product/tails being lower/higher than that of the feed, respectively.

A superficial introduction to the cascade terminology used in this paper is provided here; a more formal background on multicomponent and two-component cascade theory can be found in [7–10] and [11–14], respectively. The cascades considered in this paper are symmetric cascades (i.e., one-up, one-down cascades) where the tails stream from stage $n+1$ and the heads stream from stage $n-1$ merge at a mixing point to form the feed stream to stage n (see Fig. 1 in [10]). The relative abundance (or abundance ratio) is the mole ratio of two components,

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which for UF_6 is equivalent to the isotope ratio (e.g., $R = {}^{235}UF_6/{}^{238}UF_6 = {}^{235}U/{}^{238}U$). Matched-abundance cascades are designed to match the relative abundance of two components at the mixing points. Matched-concentration cascades are designed to match the mole fraction of one component (e.g., ${}^{235}UF_6/UF_6 = {}^{235}U/U = {}^{235}U$ enrichment) at the mixing points. Ideal cascades are mathematical cascades with non-integer numbers of separators and do not incur mixing losses at the mixing points. Squared-off cascades are real cascades with integer numbers of separators and do incur mixing losses at the mixing points. Well-designed, squared-off cascades closely approximate ideal cascades. The separative efficiency of a cascade is the ratio of the separative capacity (in separative work units (SWU) per cascade) of a squared-off cascade to an ideal cascade and is a measure of the separative work lost due to mixing. The overall stage separation factor is the stage heads-to-tails relative abundance ratio (i.e., $\alpha = R_P/R_W$) and is the same for all stages. The stage enrichment factor and stage depletion factor are the stage heads-to-feed and feed-to-tails relative abundance ratios (i.e., $\beta = R_P/R_F$ and $\gamma = R_F/R_W$), respectively, which for an ideal cascade are equivalent and equal to the root of the overall stage separation factor (i.e., $\beta = \gamma = \alpha^{1/2}$).

The MSTAR model is a multicomponent cascade model which can be used to calculate the flow rates and uranium isotope concentrations for ideal, symmetric, matched-abundance, steady-state, gaseous centrifuge (and gaseous diffusion) uranium enrichment cascades. The MSTAR model was developed by de la Garza at the Oak Ridge Gaseous Diffusion Plant (GDP) in the 1960s [7–9]. The work was continued by Von Halle through the 1970–80s [10]. The MSTAR '96 and MSTAR '12 modeling software packages were produced for the IAEA by Starr at the Oak Ridge GDP in 1996 [15] and by Weber at Oak Ridge National Laboratory (ORNL) in 2012 [16,17], respectively.

The MIST series of studies theoretically and experimentally explored and validated the results of the MSTAR model using U.S. GDP data. The MIST studies were performed in the 1970–80s by Blumkin and Von Halle at the Oak Ridge GDP [18,19], and showed how variations in the ${}^{235}U$-to-${}^{234}U$ ratio (and ${}^{235}U$-to-${}^{236}U$ ratio) in the product and tails streams of enrichment plants are related to changes in cascade operation. Particular attention was given to how the introduction of side feed and withdrawal streams perturb the minor isotope ratios and how the safeguards community could exploit this property to detect undeclared side feed and withdrawal streams. It was analytically shown that the MSTAR model is applicable to GCEPs even though the experimental validation was based on GDP data [18,19]. A key qualification of MIST (and MSTAR) for use in safeguards is that they can be used without knowledge of sensitive parameters, thus, MIST are expected to be acceptable to plant operators and technology holders.

During IAEA safeguards inspections of GCEPs, inspectors collect UF_6 samples from process streams and UF_6 cylinders. UF_6 samples are shipped back to the IAEA's analytical laboratory at Seibersdorf for destructive analysis.

Nondestructive assay (NDA) techniques for measuring/utilizing the ${}^{235}U$-to-${}^{234}U$ ratios of the GCEP process streams appear in the literature. Gamma-neutron techniques utilizing the 186 keV gamma from ${}^{235}U$ and neutrons from the ${}^{234}U(\alpha,n){}^{19}F$ reaction have been demonstrated for liquid UF_6 streams at the Portsmouth GDP [20]. Gamma-only techniques which use the gammas from ${}^{234}U$ and ${}^{235}U$ are theoretically possible for some online monitoring cases but are not known to have been experimentally investigated [21]. Neutron-only techniques which use the spontaneous fission of ${}^{238}U$, the ${}^{234}U(\alpha,n){}^{19}F$ reaction, and a known/assumed value for the ${}^{235}U$-to-${}^{234}U$ ratio to indirectly determine U mass and ${}^{235}U$ enrichment are currently being investigated [22].

The following sections proceed to derive, demonstrate, and discuss the new MSTAR model.

3. Method

In this section, the universal and dimensionless MSTAR model is derived. This derivation is an extension of the MSTAR model derivation by Von Halle [10] and uses consistent notation. This derivation is presented in three parts. The objective of the derivation is to re-organize the existing MSTAR model into a universal and dimensionless form which is independent of separator- (i.e., centrifuge) and cascade-specific parameters (e.g., separation factor, number of stages, number of separators).

The starting point for the first part of the derivation is Eqs. 37, 39, 48, and 51 from Von Halle [10], shown here as Eqs. (1) through (4)

$$x_{i,P} = \frac{\frac{E_i x_{i,F}}{E_i+S_i}}{\sum_{i=1}^{J} \frac{E_i x_{i,F}}{E_i+S_i}} \tag{1}$$

$$x_{i,W} = \frac{\frac{S_i x_{i,F}}{E_i+S_i}}{\sum_{i=1}^{J} \frac{S_i x_{i,F}}{E_i+S_i}} \tag{2}$$

where x_i is the mole fraction of component i of the J distinguishable components (i.e., ${}^{234}UF_6$, ${}^{235}UF_6$, ${}^{238}UF_6, \ldots$), the subscripts F, P, and W indicate the feed, product, and tails streams, respectively, and E_i and S_i are defined as

$$E_i = \frac{\left(\alpha_i^*\right)^{-1}}{1-\left(\alpha_i^*\right)^{-N}} \tag{3}$$

$$S_i = \frac{\left(\alpha_i^*\right)^{-1}}{\left(\alpha_i^*\right)^{M+1}-1} \tag{4}$$

where α_i^* is the effective stage separation factor of component i which is defined later in Eq. (8), N is the number of enriching stages and M is the number of stripping stages. The feed stage is the first enriching stage which is also the $M+1$ stripping stage; there are $N+M$ stages in the cascade considered here (which is different from some other conventions used in the literature where there are $N+M+1$ stages in the cascade with the feed stage indexed as stage 0). See Figs. 1 and 2 in Von Halle [10] for further information.

Diverging from Von Halle's derivation, $x_{5,P}$ is divided by $x_{4,P}$ and $x_{5,W}$ by $x_{4,W}$ and reorganized, resulting in Eqs. (5)–(7)

$$\frac{\frac{x_{5,P}}{x_{4,P}}}{\frac{x_{5,F}}{x_{4,F}}} = \frac{1+\frac{S_4}{E_4}}{1+\frac{S_5}{E_5}} = \frac{1+\frac{1-\left(\alpha_4^*\right)^{-N}}{\left(\alpha_4^*\right)^{M+1}-1}}{1+\frac{1-\left(\alpha_5^*\right)^{-N}}{\left(\alpha_5^*\right)^{M+1}-1}} \tag{5}$$

$$\frac{\frac{x_{5,F}}{x_{4,F}}}{\frac{x_{5,W}}{x_{4,W}}} = \frac{1+\frac{E_5}{S_5}}{1+\frac{E_4}{S_4}} = \frac{1+\frac{\left(\alpha_5^*\right)^{M+1}-1}{1-\left(\alpha_5^*\right)^{-N}}}{1+\frac{\left(\alpha_4^*\right)^{M+1}-1}{1-\left(\alpha_4^*\right)^{-N}}} \tag{6}$$

$$\frac{\frac{x_{5,P}}{x_{4,P}}}{\frac{x_{5,W}}{x_{4,W}}} = \frac{\frac{E_5}{S_5}}{\frac{E_4}{S_4}} = \frac{\frac{\left(\alpha_5^*\right)^{M+1}-1}{1-\left(\alpha_5^*\right)^{-N}}}{\frac{\left(\alpha_4^*\right)^{M+1}-1}{1-\left(\alpha_4^*\right)^{-N}}} \tag{7}$$

to arrive at the dimensionless ${}^{235}UF_6$-to-${}^{234}UF_6$ mole ratios (left side) expressed in terms of α_4^*, α_5^*, N, and M (right side). This ends the first part of the derivation.

Clearly, α_4^*, α_5^*, N, and M are separator- and cascade-specific parameters which must be excluded to arrive at a separator- and cascade-independent model. The second part of the derivation is an intermediary step which works to replace the α_4^* and α_5^* variables in terms of the overall stage separation factor per unit mass difference, α_0.