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# Entropy of the Isotopic Enrichment Process for $^{235}\text{UF}_6/^{238}\text{UF}_6$

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**Abstract** — This work presents the effects that produce the change in entropy during separation processes; it takes into account the dilution of  $\text{UF}_6$  in a carrier gas ( $\text{H}_2$ ,  $\text{He}$ ,  $\text{N}_2$ ,  $\text{Ar}$ ,  $\text{Xe}$ ,  $\text{SF}_6$ , etc.). Comparisons were made between two technologies: one a mature process currently used, i.e., centrifugation (process A), and the other in development, i.e., processes based on a laser [Condensation Repression Isotope Separation by Laser (CRISLA), Molecular Laser Isotope Separation (MLIS), etc.] (process B). The calculations were made using the principles of mix thermodynamics. The results indicate that entropy expenditure is two orders of magnitude higher than that necessary to separate isotopes when the amount (of isotopes) is the same in both process A and process B.

**Keywords** — Entropy, isotopic enrichment, uranium hexafluoride.

**Note** — Some figures may be in color only in the electronic version.

## I. INTRODUCTION

From a thermodynamic point of view, the most appropriate method for diagnosing industrial facilities is an entropic diagnosis<sup>1</sup> because it makes direct use of the first and second principles of thermodynamics. However, from a practical point of view, entropy is a difficult function to assimilate at a quantitative level. On the other hand, entropic data are largely scarce for substances of industrial interest, which normally are mixtures of various pure substances, gaseous, liquid, or solid.

The following ideal processes will be analyzed from the second principle of thermodynamics. The first process is isotopic separation or enrichment by a laser, specifically the Third Generation Laser Uranium Enrichment Technology, which involves a set of steps clearly described elsewhere.<sup>2–7</sup> The most well-known processes are the Separation of Isotopes by Laser-Assisted Retardation of Condensation (SILARC) in its various versions and the Condensation Repression Isotope

Separation by Laser (CRISLA)—both involve the formation of clusters.

Studies conducted in the 1970s, 1980s, and 1990s indicated that it was necessary to dilute with a carrier gas by 2% and 15% to enrich  $^{32}\text{SF}_6/^{34}\text{SF}_6$  or  $^{235}\text{UF}_6/^{238}\text{UF}_6$ , respectively. This requirement is necessary for the formation of a cluster-like dimer with Van der Waals interactions forces. The typical experimental setup is shown in Fig. 1, initially published by Snyder.<sup>2</sup> These techniques require an expenditure of energy to prepare the mixture, separate the isotopes, and then separate the enriched isotopes from the carrier gas mixture. The latter step, according to our knowledge,<sup>3</sup> has not been taken into account when assessing the efficiency of the enrichment process.

The second process (to be compared or contrasted with the laser method) is based on the use of centrifugal rotor forces, and it does not require the use of gaseous mixtures (carrier gas/ $\text{UF}_6$  or carrier gas/ $\text{SF}_6$ ). It uses the pure compound alone.<sup>8–11</sup> The gas centrifuge method, shown in Fig. 2, includes a vertical rotor spinning at high speed. The  $\text{UF}_6$  gas is fed inside the rotor, and it is

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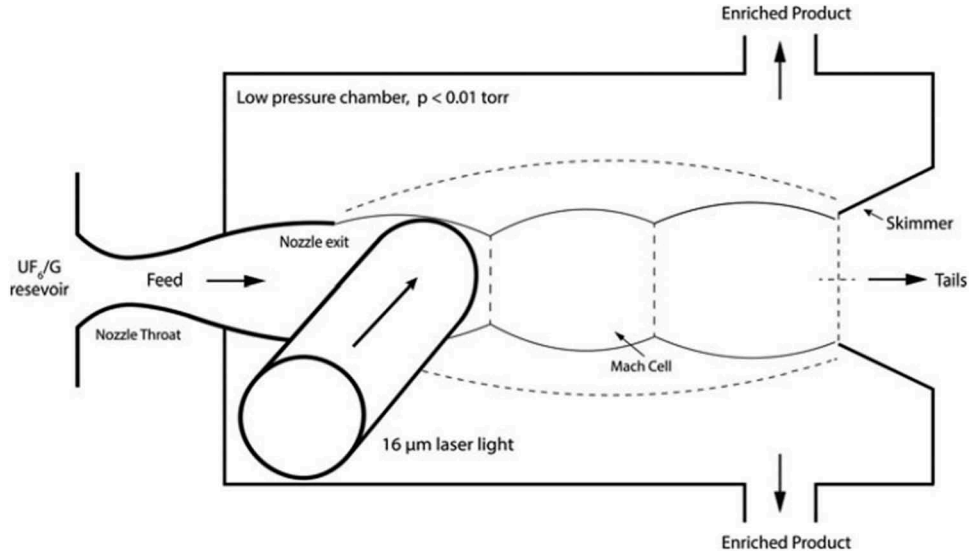


Fig. 1. The process of laser excitation, dimer formation, dimer dissociation, and migration to produce enriched product. Taken from Ref. 2.

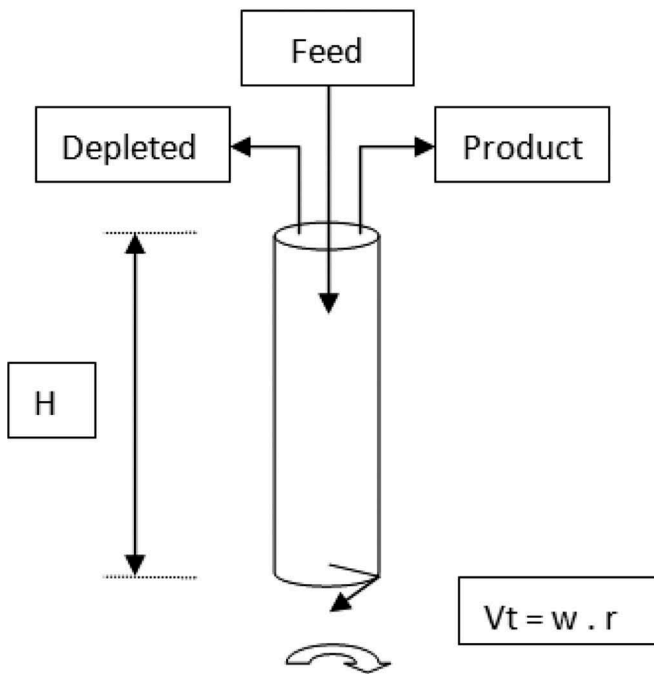


Fig. 2. Schematic view of the gas centrifuge process.

separated into two streams: one enriched in the light isotope and the other depleted in the light isotope.<sup>8</sup>

Two important variables that affect separation are the angular velocity  $w$  (rad/s) and the length of the rotor  $H$  (m). Many centrifuges are arranged in arrays, called cascades, to obtain a separation similar to distillation.<sup>8-11</sup>

The separative work unit (SWU) is a useful function that expresses the change in achieved enrichment, a measure of how much an individual separation unit or global plant

can actually separate. The basic concept was developed by Dirac during the Manhattan Project,<sup>9-11</sup> and this function is useful in assessing economic, mass, and energy issues:

$$\Delta U(\text{SWU}) = P \times V(C_p) + W \times V(C_w) - F \times V(C_f), \quad (1)$$

where

$P$  = amount of product (in tonne <sup>238</sup>U/yr)

$C_p$  = fraction product (in  $p1/Pt$ )

$W$  = waste amount (in tonne <sup>238</sup>U/yr)

$C_w$  = fraction waste (in  $p1/Pt$ )

$F$  = feed amount (in tonne <sup>238</sup>U/yr)

$C_f$  = fraction feed (in  $p1/Pt$ )

$p1$  = partial pressure of <sup>235</sup>UF<sub>6</sub> (N/m<sup>2</sup>)

$p2$  = partial pressure of <sup>238</sup>UF<sub>6</sub> (N/m<sup>2</sup>)

$Pt = p1 + p2$  (N/m<sup>2</sup>)

and  $V(C)$  is a value function that takes the form

$$V(C) = (2 \times C - 1) \ln(C/(C - 1)), \quad (2)$$

where  $C$  is a fraction in  $p1/Pt$  [1 SWU = 1 kg SW (kg <sup>238</sup>U) (Refs. 8 through 11)].

In this work, the entropy function will be used as a complement to the classical SWU evaluation, highlighting special subprocesses to study, in this case the gases. Change in entropy (second law of thermodynamics) is an

important rate or index used when comparing various processes using gas flows.

This work presents the effect that produces a change in entropy during separation processes when it accounts for the dilution of  $UF_6$  in the carrier gas (e.g.,  $SF_6$ ), in our case through some ideal processes and gases. The scope of this work is limited to systems that, under any circumstances, require diluting any of the gas's isotopes to be enriched with a carrier gas.

## II. ENTROPY CALCULATIONS FOR PURE $UF_6$

Entropy calculations are needed to establish the initial and final conditions in the enrichment process. In this sense, we realize the calculations for two processes: process A and process B. Process A is the isotope physic separation. Process B is process A + process separation of a mix of two different gases ( $UF_6$  + carrier), in general form without studying in depth each of the elements necessary in both systems. The separation unit will be considered a black box with the ability to separate isotopes to the desired degree and can be optimized to a specific concentration goal. When an enrichment process is based on the use of centrifugal force, the diagram in Fig. 3 will be used.

With the scheme in Fig. 3, it is possible to simplify the isotope separation process, as shown in Fig. 4. According to

the last scheme, the entropy change can be calculated for a small commercial plant with 200 000 SWU/yr (an economically profitable scale plant has a capacity of around 3 000 000 SWU/yr).

To perform the calculations, we define the following:  $X_{235}$  is the mole fraction of the  $^{235}UF_6$  feed, and  $X_{238}$  is the mole fraction of the  $^{238}UF_6$  feed. Assuming that the black box produces isotopic separation efficiently,  $Y_{p235}$  is the mole fraction of  $^{235}UF_6$  in the product,  $Y_{p238}$  is the mole fraction of  $^{238}UF_6$  in the product,  $Y_{t235}$  is the mole fraction of  $^{235}UF_6$  in the tails, and  $Y_{t238}$  is the mole fraction of  $^{238}UF_6$  in the tails.

The cutting factor  $\theta$  is defined as<sup>8-11</sup>

$$\theta = \frac{X_{235} - Y_{t235}}{Y_{p235} - Y_{t235}} \quad (3)$$

Assuming that the separation system shown in Fig. 4 has the following values for feed, product, and tails:

$$\begin{aligned} \text{Feed:} & \quad X_{235} = 0.711\% \quad ; \quad X_{238} = 99.289\% \\ \text{Product:} & \quad Y_{p235} = 3.5\% \quad ; \quad Y_{p238} = 96.5\% \\ \text{Tails:} & \quad Y_{t235} = 0.3\% \quad ; \quad Y_{p238} = 99.7\% \end{aligned}$$

$\theta$  can be solved:  $\theta = 0.128$ .

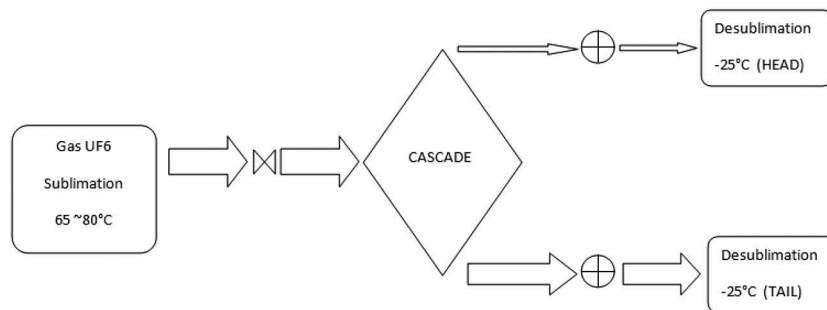


Fig. 3. Typical scheme for a centrifuge isotopic enrichment gas process (process A).

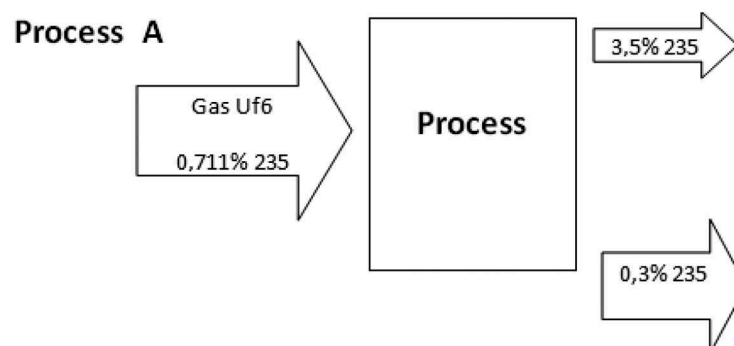


Fig. 4. A simplified scheme of separation process A for centrifugal force.

Because the plant can produce 200 000 SWU/yr, the following feed conditions are necessary:

$$\text{Input flow feed/yr: } F_{gasUF_6} = 530.69 \cdot \text{tonnes},$$

and solving Eqs. (1) and (2) with the inputs from the calculation in Eq. (3),

$$\text{Exit flow product/yr: } P_{gasUF_6} = 68.16 \cdot \text{tonnes}$$

and

$$\text{Exit flow tails/yr: } T_{gasUF_6} = 462.53 \cdot \text{tonnes}.$$

To calculate entropy, the well-known function for a mixture of ideal gases is used:

$$\Delta S = -nR \sum_{i=1}^n x_i \ln x_i, \quad (4)$$

where

$n$  = total gas mol number

$R$  = ideal gas constant

$x_i$  = molar fraction of the gas  $i$  in the gas phase solution.

### III. INPUT ENTROPY

For a feed mixture of  $UF_6$ , the corresponding molecular weight is

$$M_{in} = \left[ X_{235} \left( 349 \frac{\text{g}}{\text{mol}} \right) + X_{238} (352) \frac{\text{g}}{\text{mol}} \right];$$

$$M_{in} = 0.352 \frac{\text{kg}}{\text{mol}},$$

and using the data from Eq. (4),

$$S_{in} = -R \cdot \left( \frac{F_{gasUF_6}}{M_{in}} \right) \cdot (X_{235} \cdot \ln(X_{235}) + X_{238} \cdot \ln(X_{238})). \quad (5)$$

The input entropy becomes

$$S_{in} = 5.296 \times 10^5 \frac{\text{J}}{\text{K}}.$$

### III.A. Output Entropy

To determine output entropy, two final states must be considered: the tail  $S_T$  and the enriched product  $S_P$ .

#### III.A.1. Tail Entropy of $UF_6$

For a given feed mixture of  $UF_6$ , the corresponding molecular weight for the tail is

$$M_T = \left[ Y_{t235} \left( 349 \frac{\text{g}}{\text{mol}} \right) + Y_{t238} \left( 352 \frac{\text{g}}{\text{mol}} \right) \right];$$

$$M_T = 0.352 \frac{\text{kg}}{\text{mol}}.$$

Using the data from Eq. (4),

$$S_T = -R \left( \frac{F_{gasUF_6} \cdot (1 - \theta)}{M_T} \right) \cdot (Y_{t238} \cdot \ln(Y_{t238}) + Y_{t235} \cdot \ln(Y_{t235})). \quad (6)$$

This yields the tail entropy:

$$S_T = 2.31 \times 10^5 \frac{\text{J}}{\text{K}}.$$

#### III.A.2. Product Entropy

In the same way, for a given feed mixture of  $UF_6$ , the corresponding molecular weight for the product is the molecular weight of the product mixture of  $UF_6$ :

$$M_P = \left[ Y_{p235} \left( 349 \frac{\text{g}}{\text{mol}} \right) + Y_{p238} \left( 352 \frac{\text{g}}{\text{mol}} \right) \right];$$

$$M_P = 0.352 \frac{\text{kg}}{\text{mol}}.$$

Using the data from Eq. (4),

$$S_P = -R \cdot \left( \frac{F_{gasUF_6} \cdot \theta}{M_P} \right) \cdot (Y_{p238} \cdot \ln(Y_{p238}) + Y_{p235} \cdot \ln(Y_{p235})). \quad (7)$$

This yields the product entropy:

$$S_P = 2.443 \times 10^5 \frac{\text{J}}{\text{K}}.$$

Because the total entropy can be considered an additive function state, the total output entropy  $S_{out}$  is the direct sum of  $S_{T6}$  and  $S_{P6}$ :

$$S_{out} = (S_P + S_T)$$

and

$$S_{out} = 4.674 \times 10^5 \frac{\text{J}}{\text{K}}.$$

Finally, the entropy change for process A, using pure  $\text{UF}_6$  (g), can be determined using Eq. (8):

$$\Delta S_A = S_{out} - S_{in}$$

and

$$\Delta S_A = -6.229 \times 10^4 \frac{\text{J}}{\text{K}}. \quad (8)$$

Using the exit flow product per year and input flow feed per year, the entropy per mole of  $\text{UF}_6$  product is  $-0.322 \text{ J/K}\cdot\text{mol}$ , and the entropy per mole of  $\text{UF}_6$  feed is  $-0.041 \text{ J/K}\cdot\text{mol}$ , respectively. The negative change in entropy results because the entire process requires an amount of work to enrich  $\text{UF}_6$  to 3.5% in  $^{235}\text{UF}_6$ . But, this calculation allows only an estimation of the minimum expenditure of energy required to enrich  $\text{UF}_6$  because it is necessary to add the energy required in the sublimation and desublimation processes that are shown in Fig. 3, as well as all the consuming systems that are present in an enrichment plant.

#### IV. ENTROPY CALCULATIONS FOR A MIXTURE OF M (INERT GAS) AND $\text{UF}_6$

In entropy calculations for a laser-based procedure, the initial and final conditions of the enrichment process must be established. Calculations similar to those used for the

system shown in Fig. 2 will be carried out for other hypothetical processes,<sup>2,3,7</sup> without studying in depth each of the elements that are needed (e.g., laser issues or photo reactor efficiency processes). Again, the separation unit will be considered a black box with the ability to separate isotopes to the desired degree and that can be optimized for a specific concentration. Where the enrichment process is based on laser use, the diagram shown in Fig. 5 will be utilized as a simplification of the laser-based isotope separation process.

These techniques would require energy for preparing the mixture, for enriching the  $^{235}\text{U}/^{238}\text{U}$  isotopes in  $\text{UF}_6$  gas, and for separating the  $\text{UF}_6$  gas from the carrier.

Therefore, the hypothetical process would need two kinds of cascades/separators: the main one for isotope enrichment and a secondary one for separating the  $\text{UF}_6$  gas from the carrier gas (e.g.,  $\text{SF}_6$ ).

#### IV.A. INPUT ENTROPY OF M (Inert Gas) and $\text{UF}_6$

To get an idea of the dimension of entropy expense, in our example, a mixture of 95%  $\text{SF}_6$  and 5%  $\text{UF}_6$  is used in the laser separation process. Using these values, the entropy of the mixture can be calculated for process B [process A + process separation of a mix of two different gases ( $\text{UF}_6$  + carrier)], shown in Fig. 6. For a plant that produces 200 000 SWU/yr, the required materials can be determined:

1. ( $\text{SF}_6$  +  $\text{UF}_6$ ) input total
2.  $\text{UF}_6$  input gas 5%
3.  $F_{\text{gas}\text{UF}_6} = 531 \cdot \text{tonnes}$
4.  $\text{SF}_6$  input inert gas (carrier) 95%:

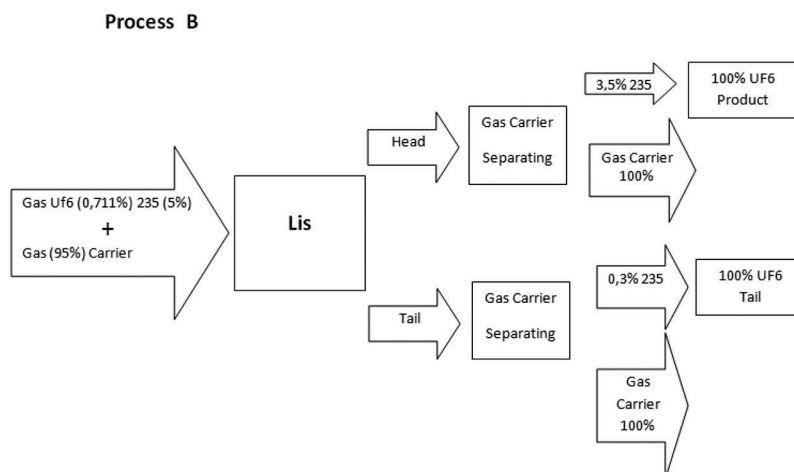


Fig. 5. Scheme of the core separation process using the laser method (process B).

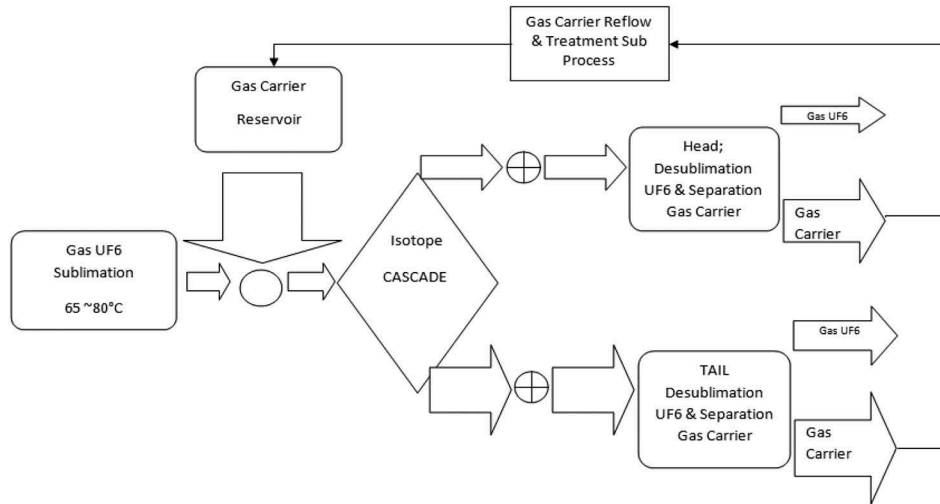


Fig. 6. Best typical scheme for a laser isotopic enrichment gas process.

$$F_{gasSF_6} = \frac{F_{gasUF_6}}{5\%}; \quad F_{gasSF_6} = 10614 \cdot \text{tonnes} . \quad S_{TM_{SF_6}} = 0.00 \frac{J}{K} .$$

Using these values in Eq. (4), the input entropy [process separation of a mix of two different gases (UF<sub>6</sub> + carrier)] can be determined:

$$\Delta S_{Mix} = -R \cdot \left[ \left( \frac{F_{gasUF_6}}{352 \frac{g}{mol}} \right) \cdot (5\% \cdot \ln(5\%)) + \left( \frac{F_{gasSF_6}}{146 \frac{g}{mol}} \right) \cdot (95\% \cdot \ln(95\%)) \right] . \quad (9)$$

In this case, the mole fraction of UF<sub>6</sub> corresponds to that of a mixture with natural abundance.

The entropy of the mixture is therefore

$$\Delta S_{Mix} = -2.986 \times 10^7 \frac{J}{K} .$$

#### IV.B. OUPUT ENTROPY OF M (Inert Gas) and UF<sub>6</sub>

As with pure UF<sub>6</sub>, two final states are considered for output entropy: the tail mixture  $S_{TM_{SF_6}}$  and the enriched product mixture  $S_{PM6}$ .

##### IV.B.1. Tail Mixture Entropy M/UF<sub>6</sub>

Assuming it is possible to completely separate M (SF<sub>6</sub>) from the enriched  $^{235}UF_6/^{238}UF_6$ , the tail mixture entropy corresponds to that for pure SF<sub>6</sub> reflows:

##### IV.B.2. Product Mixture Entropy M/UF<sub>6</sub>

As with the tail mixture, assuming it is possible to separate completely M (SF<sub>6</sub>) from the enriched  $^{235}UF_6/^{238}UF_6$ , the product mixture entropy corresponds to that for pure enriched  $^{235}UF_6/^{238}UF_6$ :

$$S_{PM} = 0.00 \frac{J}{K} .$$

Finally, the entropy change for process B, a mixture of M and UF<sub>6</sub> (g), can be found using Eq. (4):

$$\Delta S_{Mix} = (S_{TM_{SF_6}} + S_{PM}) - \Delta S_{Mix_{SF_6}} . \quad (10)$$

The negative change in entropy results from the fact that the entire process requires work to carry out the separation of pure carrier gas from the enriched product,  $^{235}UF_6/^{238}UF_6$ :

$$\Delta S_B = \Delta S_A + \Delta S_{Mix} = -2.992 \times 10^7 \frac{J}{K} .$$

This calculation allows only an estimate of the minimum expenditure of energy required by process B. The entropy per mole of UF<sub>6</sub> product is -154.5 J/K mol, and the entropy per mole of UF<sub>6</sub> feed is -19.8 J/K mol.

## V. ANALYSIS OF THE RESULTS

The results obtained can be used to determine the relative variation in entropy between process A (enrichment of  $^{235}\text{UF}_6/^{238}\text{UF}_6$  from pure  $\text{UF}_6$ ) and process B (enrichment of  $^{235}\text{UF}_6/^{238}\text{UF}_6$  from a mixture 5%  $\text{UF}_6$  and 95%  $\text{SF}_6$ ).

Using the values found in Eqs. (8) and (10), process B has greater entropy than process A in the following proportions:

$$\text{Separation of } \text{UF}_6 + \text{carrier: } S_{\text{Mix}} = -2.986 \times 10^7 \frac{\text{J}}{\text{K}}$$

$$\text{Process A: } \Delta S_A = -6.229 \times 10^4 \frac{\text{J}}{\text{K}}$$

$$\text{Process B: } \Delta S_B = \Delta S_A + \Delta S_{\text{Mix}} = -2.992 \times 10^7 \frac{\text{J}}{\text{K}}.$$

The percent entropy change  $\Delta S(B/A)$  of process A relative to process B is given by

$$\Delta S(B/A)\% = \left[ \frac{\Delta S_B}{\Delta S_A} \right] \times 100; \quad \Delta S(B/A)\% = 48033\%.$$

Such an inequality expresses the greater expense of entropy for gas separating  $n$  moles  $\text{UF}_6$  gas from the  $N$  moles carrier ( $\text{SF}_6$ ) (where  $N \gg n$ ), rather than the isotope separation  $^{235}\text{UF}_6/^{238}\text{UF}_6$ . In the case of Ar (or any monatomic gas) used as a carrier, the percentage of entropy change obtained is  $\Delta S(B/A)\% = 167152\%$  [see Eq. (9)]. The difference in entropy values spans at least three or four orders of magnitude, assuming ideal processes and gases. *Extrapolating this to an actual strongly irreversible process will provide even greater differences.* For a better comparison Table I shows the calculations of the entropy for both systems.

The variation of mixture entropy of isotopes is the same for both processes A and B as was show before. The largest expense in mixture entropy in process B is due to the large amount of carrier gas to be used (95% M and 5%  $\text{UF}_6$ ).

In the actual situation—real machines and real gases—the carrier gas separation would involve some

irreversible processes, for example, the work done by the vacuum pump, friction, etc. Too much high-energy consumption cannot be recovered in steady-state production. The  $\text{UF}_6$  gas must be separated from the carrier gas. An accepted method is cooling in a cryogenic heat exchanger; however, much more energy would be needed, and the additional entropy generated is not negligible. For these reasons the expenditure of total entropy must be greater than only the expenditure of entropy to separate the gas carrier M from the mixture. This would involve processing in a vacuum or a rarified gas requiring high-volume chambers, a huge heat exchanger coefficient, large exchanger surfaces, and large pipes for reasonable production rates. These would be taken into account in an overall evaluation regardless of the challenges faced by issues with photonic laser techniques. These types of calculations should be done for the design a separation plant and are out of the scope of this work.

## VI. CONCLUSIONS

The Condensation Repression Isotope Separation by Laser (CRISLA) process and Molecular Laser Isotope Separation (MLIS) process need two separation processes: isotopic separation and separation or treatment of  $\text{UF}_6$  mixed with a carrier (M). The challenge to overcome is separating two gases where a large amount of carrier gas plays a very important energy role in the whole process and is reflected in the entropy variation of gases. According to the calculations made and taking into account the scheme shown in Fig. 6, the entropy of the mixture plays a fundamental role in the process of separation of isotopes for process B. In this case, the use of a carrier gas as pure as possible is required to reduce the increase of the entropy of the mixture given by Eqs. (4) and (9). If gases such as HF or other gaseous impurities are generated during the separation process, then Eq. (4) will show an increase in the entropy of the mixture. To reduce this effect in a real system, it will be necessary to carry out a set of operations such as diffusion, adsorption, or distillation cycle to eliminate traces of  $\text{UF}_6$ , HF, or other impurities in the carrier gas. Without this precaution, the entire separation process will lose enrichment power (SWUs) or availability, increasing the input entropy by mixing traces of impurities in feedback carrier gas at the entrance.<sup>8,9</sup> This is another important challenge that is difficult to resolve during plant design and engineering scale-up of processes. Returning to the ideal system, the thermodynamics of the mix calculations carried out in this work clearly show the expense of entropy to separate

TABLE I

Entropy Calculations for Process A and Process B

	Process A	Process B
Entropy variation	$-6.229 \times 10^4 \text{ J/K}$	$-2.992 \times 10^7 \text{ J/K}$
Entropy/(mol $\text{UF}_6$ product)	$-0.322 \text{ J/(K)}$	$-154.5 \text{ J/(K)}$
Entropy/(mol $\text{UF}_6$ feed)	$-0.041 \text{ J/(K)}$	$-19.8 \text{ J/(K)}$



100% gas carrier from a mixture. This expenditure is two orders of magnitude higher [see Eqs. (8), (9), and (10)] than what is necessary to separate isotopes when the amount (of isotopes) is the same in both process A and process B. The important change in entropy (as the second law of thermodynamics index between two techniques) highlights the laser isotope separation challenge in working with large mixed gas flows, and additional energy is required to separate the main gas from the carrier.

The energy required to achieve a separation of quality can be assessed by the change of entropy. This calculation allows us to infer a lower limit of expenditure of energy assuming that the mixtures are ideal and there are no other energy expenses. This would take into account a hypothetical scale-up plant in engineering design. Controlling energy and evaluating energy are good practices in determining dissipated energies, improving efficiencies, or comparing various actual processes or when in the development phase. When they have been minimized or optimized, complex evaluations must be performed, so other techniques or tools are required, such as this proposed entropy analysis.

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