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Purging Metal Impurities from Nuclear Fuel using Centrifuge Cascades with Additional Feed Component

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Abstract: Nuclear fuel can be purged, that is metal impurities can be extracted from the uranium hexafluoride, by feeding an additional component, a kind of Freon, C_6F_{12}, which is chemically stable with respect to uranium hexafluoride, into the centrifuge purge cascade. The metal impurities are then continuously withdrawn from the purge cascade as a result of the additional feed. Centrifuges can not be easily used for the purge process without the Freon because the amount of metal impurities in the nuclear fuel is usually very small, thus the flow of impurities withdrawn from the product end of the purge cascade needs to be periodically closed down to minimize the loss of uranium hexafluoride. This results in the accumulation of light impurities such as air and HF in stages near the product end of the purge cascade. These light impurities inside the centrifuges may reduce the working effectiveness of the centrifuges or even put them out of service. The additional Freon eliminates this problem, because the light gas can be extracted from the cascade as soon as it reaches the product end of the cascade. Numerical calculations using a typical metal impurity CrO_2F_2 illustrate the effect of the additional Freon feed in the purge process.

Keywords: Nuclear fuel, metal impurities, purge cascade

INTRODUCTION

There are three different kinds of impurities in nuclear fuel: metal impurities such as CrO_2F_2 and WF_6 which enter in previous steps of the nuclear fuel

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cycle, hydrocarbons as the gases from lubricants in the enrichment plant equipment, and light impurities such as air and HF which come into the cascade or containers from outside plus products from chemical reactions with these impurities. The last two impurities are removed from the uranium hexafluoride by making use of their different saturated vapor pressures with the fractionation method. However, the fractionation method is not effective for eliminating metal impurities from uranium hexafluoride. Gas diffusion plants commonly use a purge cascade to purge impurities from the intermediate products, including these three kinds of impurities. The light impurities have little detrimental effect on the running of the gas diffusers. However, the concentrations of the light impurities in the centrifuge cylinders must be kept low for safe running of the centrifuges, because the centrifuges are very sensitive to light gases. The molar mass of the process gases separated by the centrifuges must be greater than a specified mass. Otherwise, the pressure at the center of the centrifuges would be high enough to force some gas through the molecular seal at the top of the cylinder into the space between the cylinder and the outer casing, which leads to unstable operation or even failure.

Two methods can be used to remove metal impurities in the feed when their concentrations exceed the quality specification limits. The first is to treat the feed material outside of the separation process. The second way is to purge the enriched product from the quality feed material unqualified in a purge cascade. Gas diffusers can be used for a purge cascade, but this makes the cascade quite complex. In addition, unmodified centrifuges can not be used in a purge cascade due to the leakage problems, but modifications give rise to higher costs. This paper presents a different, more cost effective method to use centrifuges without modifications.

In gas diffuser purge cascades, the intermediate product flows from the main cascade into the purge cascade at a point near the waste end of the purge cascade with the final product continuously withdrawn from the waste end. The impurities are not continuously withdrawn from the product end to reduce the loss of uranium hexafluoride. In a normal cascade, the end with the enriched lighter components is called the product end while the end with the enriched heavier components is called the waste end. In a purge cascade, the product end has the enriched impurities, while the uranium hexafluoride, which is the desired product, is enriched at the waste end. A gas diffuser purge cascade can be operated in different ways; one of them is as follows. The working process for the gas diffuser purge cascade has several steps. First, the valve for withdrawing impurities is closed so that impurities accumulate near the product end of the cascade as the purge cascade works with one feed flow and one waste flow. When the impurities reach a specified concentration near the product end, which means low concentration of uranium hexafluoride in the last few stages near this end, the valve is opened and some impurities are withdrawn from the cascade. The valve is then closed again to start another cycle. So the purge cascade is actually working in an unsteady mode. The similar unsteady process can not be realized in a centrifuge cascade because the centrifuge cascade can not work if its product end is blocked.

If there were an additional feed of a component with a molar mass between that of uranium hexafluoride and the metal impurities, there should always be a sufficient amount of gases occupying the stages near the product end of the purge cascade so the operate would not have to wait for the impurities to accumulate. This gives a possibility of continuous flow withdrawal from the product end of the purge cascade. The additional component acts as an isolator between the uranium hexafluoride and the metal impurities. Sosnin et al. (1) called this component a carrier gas. If the product end of the cascade is kept open, in principle, centrifuges can be used to construct a purge cascade.

THE CARRIER GAS AND ITS PHYSICOCHEMICAL PROPERTIES

The compound to be used as a carrier gas must satisfy the following conditions:

1. The molar mass of the carrier gas must be less than that of $^{235}UF_6$ 349 and greater than that of the heaviest impurity. In the present work, CrO_2F_2 (122) was used as a typical metal impurity in the nuclear fuel for the calculations.
2. The carrier gas must not condense on the interior surfaces of the equipment and its adsorption must be low.
3. The compound must be chemically stable with respect to UF_6.
4. The compound must have a proper vapor pressure. Sosnin et al. (1) used $C_8H_3F_{13}$ as a carrier gas for purging the highly radioactive isotope ^{232}U from recycled nuclear fuel. Utrobin et al. (2) analyzed a series of appropriate compounds for this process. In this work C_6F_{12} with a molar mass of 300.1 was chosen as the carrier gas. Other compounds could be used as the carrier gas for purging metal impurities from uranium hexafluoride, because the difference between the molar masses of the impurities and uranium hexafluoride is much larger than in the case of ^{232}U removal. In fact, a mixture consisting of several compounds is also feasible which would reduce costs.

THE BASIC SEPARATION THEORY AND HYPOTHESIS

For isotope separation, Ying et al. (3) found that the relative separation factor of two components in a centrifuge satisfies the following relation:

$$\gamma_{i,j} = \gamma_0^{(M_j - M_i)} \tag{1}$$

where γ_0 is the separation factor per unit molar mass difference. For a mixture having R components with M_i and M_j as the molar mass of the i-th and j-th components, then the relative separation factors are

$$\gamma_{i,j} = \frac{C_i^+}{C_j^+} \bigg/ \frac{C_i^-}{C_j^-} \tag{2}$$

where C, C^+ and C^- are the concentration in the feed, the up flow and the down flow of each stage. A separation stage has one inflow, i.e. the feed, and two outflows. The up flow is the outflow in which the lighter components are enriched while the down flow denotes the outflow in which the heavier components are enriched.

Burris and Tunstall (4) defined the relation for a diffuser stage as:

$$\gamma_{i,j} = 1 + \psi_{i,j} = \frac{C_i^+/C_j^+}{C_i^-/C_j^-} \tag{3}$$

The relative enrichment factor

$$\psi_{i,j} = \psi_0(M_j - M_i) \tag{4}$$

where ψ_0 is the enrichment factor per unit molar mass difference. For the diffusion separation of uranium, the separation factor is close to unity. Hence, Eqs. (3) and (4) can be rewritten as:

$$\gamma_{i,j} = 1 + \psi_{i,j} = 1 + \psi_0(M_j - M_i) \approx \gamma_0^{M_j - M_i} \tag{5}$$

$$\gamma_0 = 1 + \psi_0 \tag{6}$$

Therefore the expressions for the diffuser separation and the centrifuge separation can be written in the same form.

Equations (1–4) can be used for isotope separation. However, accurate relative separation factors between components of two different gases can not be easily measured. The distribution of the mixture components in a cascade were calculated here by assuming that the relative separation factors of the two components satisfies Eq. (1) or Eq. (5) for a centrifuge as well as for a diffuser, regardless of whether the two components are the same gas. The separation factor for $^{235}UF_6$ and $^{238}UF_6$ is known, so other factors can be evaluated using Eq. (1). If the molar mass difference is relatively large, the relative separation factor evaluated from Eq. (1) will be too large, so the calculations will break down. In such a case, the calculations assumed reasonable values for the relative separation factors, while keeping the values of the separation factors for the uranium isotopes unchanged. This assumption does not influence the calculated results.

The finite difference equation for calculating the distributions in a cascade is

$$\begin{aligned} H_n \sum_j a_{i,j} \frac{C_{n,j}^{+m} - C_{n,j}^{+m-1}}{\Delta t} &= [\theta_{n-1} G_{n-1} C_{n-1,i}^{+m} - (1-\theta_n) G_n b_{n,i} C_{n,i}^{+m}] \\ &\quad - [\theta_n G_n C_{n,i}^{+m} - (1-\theta_{n+1}) G_{n+1} b_{n+1,i} C_{n+1,i}^{+m}] \end{aligned} \tag{7}$$

Eq. (7) is an implicit difference scheme where

$$a_{i,j} = \frac{\partial C_{n,j}}{\partial C_{n,j}^{+}} = \begin{cases} \theta_n + (1-\theta_n) \dfrac{\sum\limits_l \gamma_{i,l} C_{n,l}^{+} - C_{n,i}^{+}}{\left(\sum\limits_l \gamma_{i,l} C_{n,l}^{+}\right)^2}, & (i = j) \\ -(1-\theta_n) \dfrac{\gamma_{i,l} C_{n,i}^{+}}{\left(\sum\limits_l \gamma_{i,l} C_{n,l}^{+}\right)^2}, & (i \neq j) \end{cases}$$

$$b_i = \frac{1}{\sum\limits_l \gamma_{i,l} C_l^{+}}$$

Δt—time step
n—stage number in a cascade
H—holdup in a stage
G—flow rate in a stage
θ—flow cut in a stage
m—calculation of step

PURGE PROCESS IN A DIFFUSER PURGE CASCADE

If the purge cascade is long enough and the impurity concentrations in the feed are constant, theoretically, the up flow in the last stage of the purge cascade can be entirely impurities by appropriately adjusting the impurity withdrawal rate from the product end. The flow rate should satisfy:

$$P' = C_F{}' F \tag{8}$$

where P' is the impurity flow rate, F is the feed flow rate, and $C_F{}'$ is the total of the impurity concentrations in the feed.

In fact, the amount of impurities in the feed is very low, so the withdrawal is very small and difficult to accurately control. In addition, the amount of impurities in the feed and in the cascade varies because of variation in the amount of air and HF coming into the cascade from outside and from the feed and because

Table 1. Feed composition

Composition	Molar mass	Percentage
HF	20	0.10 (volume)
Air(N_2)	28	0.10 (volume)
CrO_2F_2	122	0.01 (volume)
$^{235}UF_6$	349	5.00 (isotopic)
$^{238}UF_6$	352	95.00 (isotopic)

of variation in the amount of metal impurities coming into the cascade from the feed. Thus, the purge cascade usually works in an unsteady mode with intermittent withdrawal of the impurities. Initially, the impurities accumulate in the cascade. They are withdrawn only when their concentration reaches an allowable limit, since uranium is also withdrawn together with the impurities and the objective is to minimize the loss of uranium.

The purge process in a diffuser purge cascade will be illustrated with an example to illustrate the main disadvantage of using centrifuges for nuclear fuel purging. The numerical calculation assumes that the composition for the product from a main cascade in the plant (the intermediate product) is listed in Table 1.

The feed point is assumed to be near the waste end of the purge cascade as in Fig. 1. The purge cascade must be frequently closed to allow accumulation of the metal impurities, thus the stages near the product end are full of air and HF, so that the gas withdrawn periodically from the product end is mostly a mixture of air and HF. The metal impurities accumulate in the middle section of the purge cascade with the purged fuel (the final product) withdrawn from the waste end. Figures 2 and 3 show the distributions of the components in a purge cascade at two different moments to illustrate the accumulation of the metal impurities in the purge cascade. In Fig. 2 the impurities have only just begun to accumulate while in Fig. 3 the metal impurity concentrations have reached a significant level. The stage with the maximum metal impurity concentrations changes from the product end towards the direction of the waste end as the accumulation process proceeds. At the same time, the uranium concentration at the product end decreases while the amount of impurities at the waste end increases.

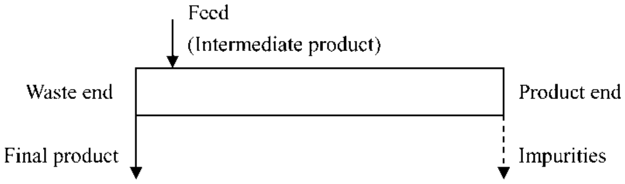


Figure 1. Schematic of a diffuser purge cascade.

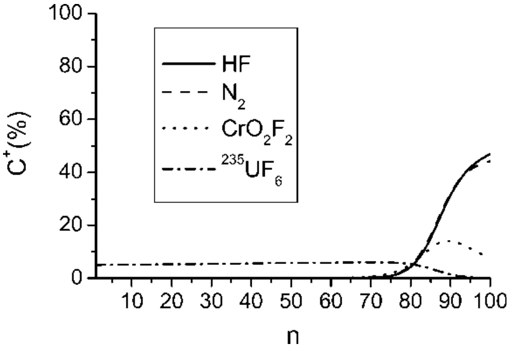


Figure 2. Composition distribution as the metal impurities begin to accumulate.

The opening and closing of the valve at the product end to remove the impurities should ensure that the metal impurity concentrations in the final product provide the desired quality and that the amount of uranium withdrawn from the product end together with the impurities is minimized. Calculations show that the metal impurity concentrations in the final product are reduced several hundred fold relative to the feed concentrations with the amount of $^{235}UF_6$ in the impurities withdrawal flow less than several ppm. For an average impurities flow rate of about 0.2% of the feed flow rate, the $^{235}UF_6$ loss should be very small when the diffuser purge cascade alternates between accumulation and withdrawal of impurities.

CENTRIFUGE PURGE CASCADE WORKING PRINCIPLE

As discussed in the previous section, a diffuser purge cascade operates in an unsteady mode. Ordinary centrifuge cascades are not used in this way, but

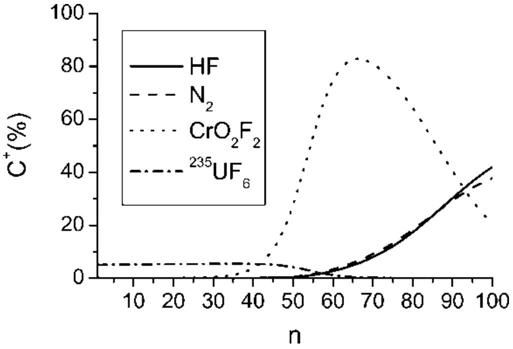


Figure 3. Composition distribution after significant accumulation of metal impurities.

the carrier gas allows the use of centrifuges for purging. Fig. 4 shows a schematic illustration of a centrifuge purge cascade.

The present analysis considers a centrifuge cascade consisting of 5 stages. The intermediate product to be purged is fed into the 1st stage with the carrier gas fed into the 5th stage. The flow rate ratios are assumed to be:

$$F_{C_6F_{12}}/F_{UF_6} = 0.05$$
$$P/(F_{C_6F_{12}} + F_{UF_6}) = 0.04$$

Figure 5 presents the calculated results. Due to the relatively larger separation factor for the centrifuges, the concentrations of the metal impurities and the light impurities in the waste flow of the centrifuge purge cascade are very low and can be neglected even though the cascade only contains a few stages. The air and HF concentrations in the last stage feed are about 2%. Increases of the proportion of C_6F_{12} in the feed and the product flow rate reduce the air and HF concentrations in the last stage, so the centrifuges in the last stage of the purge cascade are safer. The C_6F_{12} feed proportion depends on the air and HF concentrations in the intermediate product and the limits on the air and HF concentrations in the centrifuges. The $^{235}UF_6$ concentration in the impurities withdrawal flow is about 5 ppm, so the loss of ^{235}U is about 0.001%. The C_6F_{12} concentration in the waste flow, i.e. in the final product, is about 1%. The loss of ^{235}U was reduced by higher C_6F_{12} feed proportion, longer cascades and smaller product flow rate (i.e. impurities flow rate). The Freon feed point should be in the last stage for the optimal results. The impurities flow rate, P, depends on the air and HF concentrations in the feed which can be reduced by carefully treating the intermediate product by fractionation in advance.

Note that the component concentrations plotted in Fig. 5 are the values in the up flows in these stages. The difference between the component concentrations in the two outflows of each stage is significant due to the large centrifuge separation factors. The C_6F_{12} concentration is plotted in Fig. 5(a) with the other component concentrations plotted in Fig. 5(b).

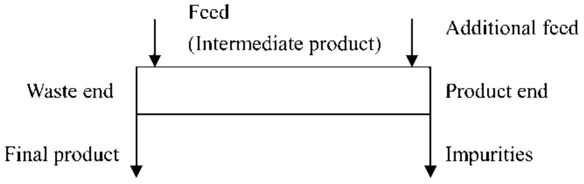
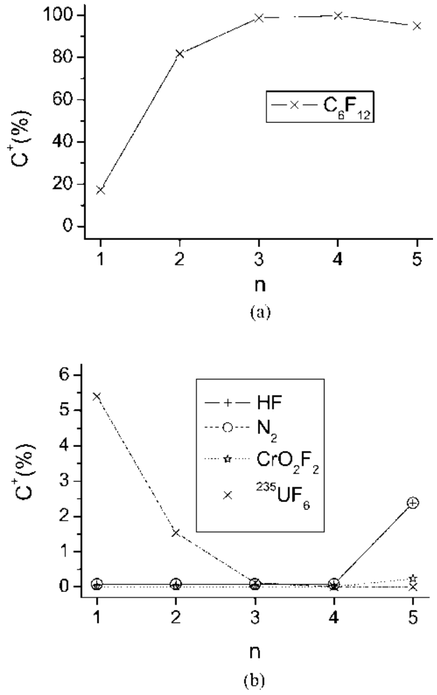


Figure 4. Schematic of a centrifuge purge cascade.



(a)

(b)

Figure 5. Composition distribution in a centrifuge purge cascade with additional feed of C_6F_{12}.

CONCLUSION

The additional feed of a carrier gas is able to establish continuous product flow in a purge cascade, which enables the use of centrifuges in a purge cascade. The role of the carrier gas is to push the uranium hexafluoride away from the product end and to maintain the impurities flow to a controllable quantity. The carrier gas will be more effective if its molar mass is close to the molar mass of the heaviest impurity. Thus there should be other Freons beside C_6F_{12} for this purpose. The Freon feed rate depends on the concentrations of the metal impurities, air and HF in the intermediate product, the desired final product quality and the centrifuge design limitations. The Freon, that is withdrawn from the purge cascade can then be easily extracted by fractionation from the uranium hexafluoride or from the impurities and reused.

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