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### SEPARATING ISOTOPE COMPONENTS OF SMALL ABUNDANCE

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## SEPARATING ISOTOPE COMPONENTS OF SMALL ABUNDANCE

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### ABSTRACT

It is difficult to enrich isotope components of small abundance to a high concentration by means of gas centrifuge cascades. A nonsteady-state approach is studied for the separation of end components and middle components. Unlike ordinary conventional separation cascades, which have two withdrawals at the two ends of the cascades and one feed in between, the cascade used in the nonsteady-state approach has only one withdrawal at the either end and one feed. The nonsteady-state separation approach is analyzed from two aspects: the material recovery and the operation time efficiency, for producing one unit mass of material, and is compared with conventional cascades. The results show that the approach is advantageous over the conventional cascades for obtaining the same amount of a desired component, especially for the separation of end components.

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**Key Words:** Multicomponent isotope separation; Transient process; Gas centrifuge cascade

## INTRODUCTION

In a multicomponent isotope separation, it is very common to encounter cases in which the component of small abundance needs to be enriched to a very high concentration. For example,  $^{124}\text{Xe}$  and  $^{123}\text{Te}$  are needed in the production of the radionuclide  $^{123}\text{I}$  by means of precursor methods for the purpose of radiology. The concentrations of  $^{124}\text{Xe}$  and  $^{123}\text{Te}$  must be very high ( $\geq 99.9\%$ ) in order to reduce the quantities of harmful contaminants such as  $^{124}\text{I}$  and  $^{125}\text{I}$  that are formed in the production process. A way to obtain such highly enriched  $^{124}\text{Xe}$  and  $^{123}\text{Te}$  is by using gas centrifuge cascades.

A cascade can operate in various ways. The most popular one is operating through a conventional way, i.e., with two withdrawals (one product and one waste) at the two ends of the cascade and one feed in between. With a sufficiently long conventional cascade, theoretically it is possible to separate a component in a multicomponent isotope mixture with only one operation run if it is an end component (that is, the lightest or the heaviest component in the mixture), or with two operation runs if it is a middle component. This can be summarized as follows (cf. [1] for details). Let  $N_c$  be the number of components and number the components sequentially from the lightest to the heaviest. Denote the concentrations of the original feed (i.e., the feed for the first separation run) to be  $C_i^F$ ,  $i = 1, 2, \dots, N_c$ . Separating an end component is quite easy, and only one separation run suffices. The product of the cascade contains only the lightest component if the cascade cut  $\Theta$ , defined to be the ratio of the product rate  $P$  to the feed rate  $F$ , is adjusted to  $C_1^F$ , or the waste contains only the heaviest component if the cascade cut is adjusted to  $\sum_{i=1}^{N_c-1} C_i^F$ . For separating a middle component, the  $j$ th component, two separation runs are needed. In the first separation run the cascade cut should be set to  $\sum_{i=1}^{j-1} C_i^F$  to make the component an end component in the waste or  $\sum_{i=j}^{N_c} C_i^F$  in the product. The  $j$ th component is then obtained in the second separation run by using the waste or the product as the feed of the second run.

There are several practical difficulties and inconveniences, however, in realizing the separation of a desired component by a conventional cascade in the above way if the concentration of the component to be enriched is very small, like  $^{124}\text{Xe}$  here, which is 0.093% in the Xe isotope mixture of natural abundance. The first difficulty is that the cascade should be sufficiently long. But, how long a cascade can be thought of being sufficiently long? This of course has something to do with the separation performance of the centrifuges used in the cascade. In



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Ref. [1], the calculations show that a cascade of about 41 stages is already sufficiently long if the unit separation factor  $\gamma_0$  of the centrifuges is 1.4 in the separation of Xe isotopes. Such a cascade is not impractical, but, as long as the same separation effects can be obtained, it is always desirable to use a cascade, which is as short as possible from the point of view of economic considerations in the following two obvious respects: (a) a longer cascade has more centrifuges to operate; (b) a longer cascade has a larger total holdup. These would increase either the operation cost, or the amount of process gas to be separated, or operation time. The second difficulty is that the required cascade cut is hard to control within an acceptable accuracy. In order to guarantee that the product withdrawal contains only  $^{124}\text{Xe}$ , the cascade cut should be no larger than 0.00093 according to the theory, which is hard to be realized. To avoid this difficulty, two ways may be followed: taking larger cascade cuts, or adding a gas additive to the Xe isotope mixture. The first is straightforward but consequently more separation runs (preliminary runs) are needed, with the product of a separation run used as the feed for the next separation run. This is to enhance the concentration step by step. In this way, the quantities of feed for the later separation runs become less and less, and perhaps for the last run, unless a fairly large-scale production is carried out, the quantity may not even be sufficient to establish a steady flow in the cascade. Inconvenience brought in by doing enrichment step-by-step is that the withdrawals (either wastes or products) of the preliminary runs need be stored, because the concentrations of the desired component in these wastes or products may be most likely higher than its natural abundance, if the cascade is not sufficiently long. Another inconvenience is that in the preliminary runs, since the concentrations of the desired component change, to achieve the best possible separation performance and so to reduce the number of preliminary runs the cascade operation status, such as feed location and cascade cut, should be adjusted accordingly probably for every preliminary run. The second way mixes the isotopic gas mixture with another gas additive, called carrier gas.<sup>[2]</sup> The molar weight of the carrier gas is carefully chosen, for example for the separation of  $^{124}\text{Xe}$ , the carrier gas should ideally have a molar weight that is larger than that of  $^{124}\text{Xe}$  but smaller than that of  $^{126}\text{Xe}$ . The product withdrawal comprises the mixture of  $^{124}\text{Xe}$  and the carrier gas, which needs to be separated by means of chemical or/and physical processes. However, such a carrier gas with a proper molar weight as well as chemical and physical properties suitable for gas centrifuges is not always available.

Since separating a middle component needs at least two runs, it is obviously more difficult than separating an end component. In addition to those difficulties encountered in separating an end component, there are a few more other ones that are peculiar in separating middle components. An evident exhibition of these difficulties is that a middle component is still a middle component, even after many separation runs.

The so-called nonsteady-state approaches (see Refs. [2,3]) may be utilized to deal with the above difficulties. But a clear understanding should be gained as to what advantages, the nonsteady-state approaches have, and how much advantageous they are over the traditional methods. This article addresses this topic. The nonsteady-state approaches actually make use of transient properties in cascades. The one used for separating an end component<sup>[2]</sup> is different from that for separating a middle component.<sup>[3]</sup> Here only the former approach is investigated numerically with its application not only to the separation of an end component but also to the separation of a middle component, in two aspects: the material recovery and the operation time efficiency, and is compared with conventional cascades.

### THE PARTIAL DIFFERENTIAL-DIFFERENCE EQUATIONS

The waste end of a rather general cascade for separating isotope mixture is shown schematically in Fig. 1. Here,  $H_n$ ,  $H'_n$ , and  $H''_n$  are, respectively, the holdups in the gas centrifuges, the upstream pipes, and the downstream pipes at the  $n$ th stage,  $m_{n,i}$ ,  $m'_{n,i}$ , and  $m''_{n,i}$  are the rates of material losses for the  $i$ th component in the centrifuges and the pipes, respectively.  $F_n$ ,  $P_n$ , and  $W_n$  are the feed, the head, and the tail flows, respectively, and  $G_n$  is the entering flow, at stage  $n$ . It should not be confused by the general cascade diagram because of its so many feeds and withdrawals. For example, a conventional cascade has only one nonzero feed  $F_{N_F}$  at stage  $N_F$  and two nonzero withdrawals  $W_1$  at stage 1 and  $P_N$  at stage  $N$ ; and all other  $F$ s,  $P$ s, and  $W$ s are zero.  $L'_n$  and  $L''_n$  are the upstream flows of the  $n$ th stage

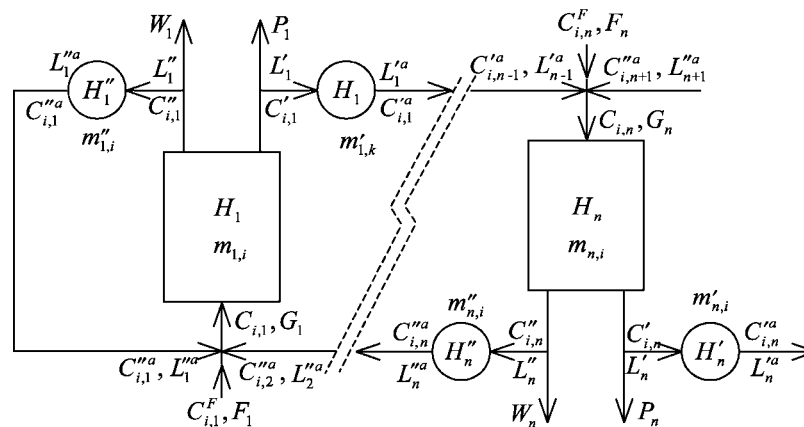


Figure 1. A schematic illustration of part of a cascade.



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before and after passing through the pipe connecting stages  $n$  and  $n + 1$ ;  $L_n''$  and  $L_n''^a$  are the downstream flows before and after passing through the pipe connecting stages  $n$  and  $n - 1$ .  $L_n''^a$  satisfies

$$L_n''^a = L_{n-1}^a - \sum_{l=1}^{n-1} \left[ F_l - P_l - W_l - \sum_i (m_{l,i} + m'_{l,i} + m''_{l,i}) \right]$$

or

$$L_n''^a = L_{n-1}^a + \sum_{l=n}^N [F_l - P_l - W_l - \sum_i (m_{l,i} + m'_{l,i} + m''_{l,i})]$$

$C'_{i,n}$  and  $C''_{i,n}$  are, respectively, the concentrations of the  $i$ th component in the heads and tails flows just leaving the centrifuges, and  $C'^a_{i,n}$  and  $C''^a_{i,n}$  are, respectively, the concentrations in the head and the tail flows just before the mixing points. At the two ends of the cascade, the following relationships hold

$$L_0^a = L_1^a, \quad C'^a_{i,0} = C''^a_{i,1} \quad (1)$$

$$L_{N+1}''^a = L_N^a, \quad C'^a_{i,N+1} = C'^a_{i,N} \quad (2)$$

where  $N$  is the total number of stages. Mass conservation of the  $i$ th component in the centrifuges at the  $n$ th stage gives

$$\begin{aligned} & \frac{\partial}{\partial t} H_n [\theta_n C'_{i,n} + (1 - \theta_n) C''_{i,n}] \\ &= L_{n-1}^a C'^a_{i,n-1} + L_{n+1}''^a C''^a_{i,n+1} + F_n C^F_{i,n} \\ & - (L'_n + P_n) C'_{i,n} - (L''_n + W_n) C''_{i,n} - m_{n,i} \end{aligned} \quad (3)$$

where  $\theta_n$  is the stage cut, defined as

$$\theta_n = \frac{L'_n + P_n}{L'_n + P_n + L''_n + W_n}$$

For the pipes the equations are

$$\frac{\partial H_n' C_{i,n}'^a}{\partial t} = L_n' C_{i,n}' - L_n'^a C_{i,n}'^a - m_{n,i}' \quad (4)$$

$$\frac{\partial H_n'' C_{i,n}''^a}{\partial t} = L_n'' C_{i,n}'' - L_n''^a C_{i,n}''^a - m_{n,i}'' \quad (5)$$

It should be pointed out that these equations about mass conservation are derived from some approximations to avoid the use of concentrations inside the centrifuges and the pipes. Only here concentrations are involved at the places of entering and outgoing flows of centrifuges and at the two ends of pipes. The details of the derivation can be found in Ref. [4]. For gas centrifuges, a well-known relation,<sup>[5]</sup> which associates the concentrations of the  $i$ th component with those of the  $j$ th component in the head and tails flows, is given by

$$\frac{C_{i,n}'/C_{i,n}''}{C_{j,n}'/C_{j,n}''} = \gamma_0^{M_j - M_i} \quad (6)$$

with  $\gamma_0$  being the unit separation factor, and  $M_i$  and  $M_j$ , respectively, the molar weights of the  $i$ th and  $j$ th components. Equations (1)–(6) are the equation system describing the transient behavior of concentrations in a cascade, subject to the following condition

$$\sum_{i=1}^{N_c} C_{i,n}' = \sum_{i=1}^{N_c} C_{i,n}'' = \sum_{i=1}^{N_c} C_{i,n}'^a = \sum_{i=1}^{N_c} C_{i,n}''^a = 1 \quad (7)$$

Discretization of the partial differential-difference Eqs. (3)–(5) is described in Ref. [4]. The resulting algebraic equations together with Eqs. (1), (2), (6), and (7) are solved at each time step by the so-called Q-iteration method. The readers may refer to Refs. [6,7] for the details about the solution of this equation system. The stop criterion for the Q-iterations is

$$\max \left( \left| \sum_n C_{i,n}' - 1 \right|, \left| \sum_n C_{i,n}'' - 1 \right|, \left| \sum_n C_{i,n}'^a - 1 \right| \right) \leq \varepsilon_1 \quad (8)$$

where  $\varepsilon_1$  is set to  $10^{-6}$ . The time marching stops when the following condition is satisfied:

$$\left( \sum_n \sum_i (F_n C_{i,n}^F - P_n C_{i,n}' - W_n C_{i,n}'' - m_{i,n} - m_{i,n}' - m_{i,n}'')^2 \right)^{1/2} \leq \varepsilon_2 \quad (9)$$

where  $\varepsilon_2$  is also set to  $10^{-6}$ .



## SEPARATION OF AN END COMPONENT

## The Idea

Here the separation of  $^{124}\text{Xe}$  in the isotope mixture of natural abundance is used as an example, without loss of generality. The natural concentrations  $C_i^F$  ( $i = 1, 2, \dots, 9$ ) of the nine components of Xe in the mixture are given in Table 1. It has been explained that the cascade cut should be less than  $C_1^F$  in order to have only  $^{124}\text{Xe}$  in the product. Because  $C_1^F$  is too small, controlling the cascade cut is difficult. However, when the cascade cut is extremely small, that is, zero, this problem is resolved. What happens if there is no product? One can expect that the lightest component,  $^{124}\text{Xe}$  here would be accumulated in the product end of the cascade and push all the heavier components out of the cascade through the waste withdrawal. As long as the time of accumulation is long enough, the enriching section of the cascade would be filled with highly enriched  $^{124}\text{Xe}$ . For easy reference, this way of cascade operation is referred to as the single withdrawal method, (SW method for short), as there is only one withdrawal. A cascade of using this method is referred to as an SW cascade, which is actually a conventional cascade without either the waste withdrawal ( $W_1 = 0$ ) or the product withdrawal ( $P_N = 0$ ). Such a cascade is used in Ref. [2] but not investigated thoroughly.

## The Optimal Result

What are the advantages of the SW method over the conventional method? Of course one method can be compared directly with the other, but it is best to know what the optimal result is and compare these methods with reference to the optimum. By this kind of comparison, we not only know which method is relatively better to each other, but also how much better it is. The comparisons are made with the two aspects: the material recovery  $R$  and the operation time efficiency  $E_t$  in producing the same amount of the required product. They are defined as follows. Assume that to obtain 1 unit mass of  $^{124}\text{Xe}$  of a given concentration, a cascade with a feed rate  $F = 1$  unit mass per unit time needs a total amount of material  $M$  unit mass and has to operate for  $T$  unit time. Here for

**Table 1.** Natural Concentrations of the Nine Components of Stable Xe Isotopes

$^{124}\text{Xe}$	$^{126}\text{Xe}$	$^{128}\text{Xe}$	$^{129}\text{Xe}$	$^{130}\text{Xe}$	$^{131}\text{Xe}$	$^{132}\text{Xe}$	$^{134}\text{Xe}$	$^{136}\text{Xe}$
0.00093	0.0009	0.01917	0.2644	0.0408	0.2118	0.2689	0.1044	0.0887



simplicity the values of all quantities are specified in units. Given mass  $M^*$  and time  $T^*$ , we call that mass  $M'$  is  $M$  unit mass if  $M = M'/M^*$ , and time  $T'$  is  $T$  unit time if  $T = T'/T^*$ . All mass and time quantities are nondimensionalized based on the mass  $M^*$  and the time  $T^*$  adopted. To obtain the required concentration, many separation runs may be necessary.  $M$  is actually the amount of feed in the first run, and  $T$  is the sum of the times used in all runs. The material recovery  $R$  is defined to be the ratio of the amount of  $^{124}\text{Xe}$  in the final product to that in  $M$ . For cascades with the same feed rate  $F$ , the best separation is that all  $^{124}\text{Xe}$  in the feed can be extracted into the product. So the recovery rate has its maximum value to be 1 and the cascade needs the least amount  $M$  of the feed material for producing the same amount of product. Consequently, the operation time  $T = M/F = M$  is the least. Now we define the optimal result of  $M_o$  and  $T_o$  to be, respectively, the amount of feed material and operation time of a cascade for producing 1 unit mass of product with the required concentration of the desired component when the feed rate is 1 unit mass per unit time and the material recovery is 1. Then operation time efficiency  $E_t$  is defined to be  $T_o/T$ . Note that the material recovery  $R$  is equal to  $M_o/M$ .

Such an optimal result does exist and can be obtained with a sufficiently long cascade operating in a conventional way. To avoid trivial but complicated computation procedures, the following assumptions are made throughout the analyses in this article: steady flow is already established in any cascade; the transient processes of concentrations are ignored for conventional cascades, because they are very short in comparison with the total operation times; the initial concentration distributions are constant and are taken to be the concentrations in the feed. As discussed before, the shortest time and the maximum material recovery are achieved when all  $^{124}\text{Xe}$  in the feed goes into the product. It is already known that this can be realized by taking the cascade cut  $\Theta$  to be  $C_1^F/C_1^P$  (see Ref. [1]), with  $C_1^P (\geq C_1^F)$ , of course) being the required concentration in the product. To produce 1 unit mass of product, the total amount of feed needed is  $M = C_1^P/C_1^F$ , which makes the material recovery 1. Because the transient time at the beginning of the separation is ignored, the time spent is simply  $T = M/F = M$ . So this is already the optimal result and so no doubt there cannot be any other better results. In the comparisons, we require the concentration  $C_1^P$  of  $^{124}\text{Xe}$  in the final product to be  $C_1^P = 0.999$ . Then  $M_o = T_o \approx 1074$ .

### Calculation of $M$ , $T$ , $R$ , and $E_t$

The results from other methods will be compared with the optimal result. For simplicity of calculation, it is assumed that in a conventional cascade all holdups  $H_n$ ,  $H'_n$ , and  $H''_n$  are zero. If the conventional method produces a final



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product of 1 unit mass of  $^{124}\text{Xe}$  with concentration  $C_1^{P,N_R}$  after  $N_R$  separation runs, it is easy to calculate that the total amount of material needed and also the time spent are

$$M = T = 1 / \prod_{r=1}^{N_R} \Theta^r$$

and the material recovery  $R$  and the operation time efficiency  $E_t$  are

$$R = E_t = C_1^{P,N_R} \prod_{r=1}^{N_R} \Theta^r / C_1^{F,1}$$

Here  $\Theta^r$ ,  $C_1^{P,r}$ , and  $C_1^{F,r}$  represent, respectively, the cascade cut, the product, and the feed concentrations at the  $r$ th run. Here, obviously  $C_1^{F,r} = C_1^{P,r-1}$ . The term “final product” is used to mean the withdrawal, either the product or the waste withdrawal, in which the concentration of the desired component satisfies the requirement in the last separation run, to distinguish it from any withdrawals between the first and the last runs.

The values of  $M$ ,  $T$ ,  $R$ , and  $E_t$  of an SW cascade are calculated in the following way. All holdups  $H_n$ ,  $H'_n$ , and  $H''_n$  except  $H'_N$  are assumed to be zero.  $H'_N$  should not be zero because the enriched  $^{124}\text{Xe}$  accumulates at the product end, and is always taken to be 1 (unit mass) for simplicity. There is no loss of generality for taking  $H'_N = 1$ , because in Ref. [4] it is shown that the transient time is proportional to  $H'_N$  for a fixed feed. Therefore, if a separation run spends  $T$  unit time when  $H'_N = 1$ , then in the case of  $H'_N \neq 1$  the time spent would simply be  $TH'_N$ . Also assuming that all  $H_n$ ,  $H'_n$ , and  $H''_n$  except  $H'_N$  are zero is reasonable, because the desired component should be retained in  $H'_N$  as much as possible, and therefore  $H'_N$  should be designed such that  $H'_N \gg H_n$ ,  $H'_n$ , and  $H''_n$ . Note that it is the material in  $H'_N$  that is used as the feed for the next run. In the  $r$ th run, if the operation time is  $T_r$ , then the total amount of feed material needed is  $T_r F$ . Notice that the amount of withdrawal is  $T_r F - H'_N$ , because  $H'_N$  is collected after the run (remember that  $H'_N = 1$ ). The feed of the  $r$ th run is of course from the  $(r-1)$ th separation, which needs the  $(r-1)$ th separation run to operate for  $T_r F T_{r-1}$  unit times and requires  $T_r F T_{r-1} F$  unit mass of material. So to obtain 1 unit mass of the final product, the total operation time  $T$  and total amount of material  $M$  are, respectively, calculated by

$$T = T_1 \prod_{r=2}^{N_R} (T_r F)$$

and

$$M = \prod_{r=1}^{N_R} (T_r F)$$

Then the material recovery is

$$R = C_1^{P,N_R} / (M C_1^{F,1})$$

In the following, some comparisons are made between the conventional method and the SW method by using the same cascade.

### Numerical Experiments

Assume that the length of a cascade  $N = 21$ , the upstreaming flow rate at each stage  $L_n = 10$ , and the feed rate  $F = 1$  unit mass per unit time. Xenon does not react with the cascade materials, so  $m_{i,n} = m'_{i,n} = m''_{i,n} = 0$ . Because we are considering a cascade that is neither sufficiently long nor very large in scale, the real value of  $F$  is not large, and the cascade cut cannot be too small for the purpose of easy control of the product and waste withdrawals. For different allowable smallest cascade cuts, numerical experiments are carried out to investigate the separation processes that finally yield the required concentration of  $^{124}\text{Xe}$ , which is chosen to be greater than 99.9% here for demonstration purpose. The unit separation factor  $\gamma_0$  is taken to be 1.4 in all numerical experiments. The experiments are shown in Table 2. In each separation run, the cascade cut is given, and feed location is determined by maximizing  $C_1^{P,r}$ . The values of  $M$ ,  $T$ ,  $R$ , and  $E_t$  are also given in Table 2.

A little explanation is in place about the values of the cascade cut, which are chosen rather arbitrarily. It is shown in Ref. [8] that, when several separation runs have to be employed, a better way of achieving the highest concentration may be taking the concentration of the desired component in the final separation run as the optimization object, and the cascade cuts of all separation runs as the optimization variables, instead of optimizing the concentration at each run with a given cascade cut. However, achieving the highest concentration does not necessarily mean obtaining the largest  $R$  and  $E_t$  as well as the smallest  $M$  and  $T$ , and the computation is much more complex, which is especially the case where the number of separation runs increases, as is in the numerical experiments for shorter cascades, which are given later. In practice, it is not always possible to obtain the largest  $R$  and  $E_t$  and meanwhile have the smallest  $M$  and  $T$ . Compromise has to be made among  $M$ ,  $T$ ,  $R$ , and  $E_t$  for best results in some sense, for example, at the lowest cost. Therefore, here we simply specify the cascade cut for each run.

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**Table 2.** Experiments for Separation of  $^{124}\text{Xe}$  with a Conventional Cascade of Length  $N = 21$ 

Experiment	$r$	$N_F$	$\Theta^r$	$C_1^{P,r}$	$T (M)$	$R(E_i)$
1 (No restriction on $\Theta^r$ )	1	11	0.00093	0.9992	1075	0.999
2 ( $\Theta^r \geq 0.01$ )	1	21	0.01	0.0930	1075	0.999
	2	11	0.093	0.9991		
3 ( $\Theta^r \geq 0.1$ )	1	20	0.1	0.0093	1087	0.989
	2	21	0.2	0.0465		
	3	21	0.2	0.2325		
	4	10	0.23	0.9996		

It is clearly seen that, the smaller the allowable cascade cut can be taken, the closer the results are to the optimal one. It should be noted that in an experiment the last run or the last few runs are problematic, because, for example in Experiment 3, the amount of feed in the last run is only  $1/\Theta^r = 1/0.23 \approx 4.35$  unit mass. Depending on what the real dimension of unit mass is, such an amount of feed is probably too little to even fill in the cascade or to establish a steady flow. For example, if one unit mass is taken to be just 1 g, then the feed is only 4.35 g. For gas centrifuge cascades, this problem always exists. But the larger the cascade, more obvious the problem becomes. Regardless of this, a cascade with 21 stages seems to perform quite satisfactorily from the point of view of the material recovery and operation time efficiency. There are no big differences in the results of  $T$  and  $R$  for different cascade cuts. How about a shorter cascade? Table 3 presents the results for a cascade of length  $N = 11$ .

Now it is immediately noticed that the values of  $T$  and  $R$  are very different in different experiments. Compare Experiment 4 with Experiment 5. In the two experiments, which require  $\Theta^r \geq 0.01$  to avoid too small cascade cuts, the first runs both use  $\Theta^1 = 0.01$ . The second run in Experiment 4 takes  $\Theta^2 = 0.035$  and obtains the required concentration of  $^{124}\text{Xe}$ . But in Experiment 5 the second run takes  $\Theta^2 = 0.093$ , and one more run with  $\Theta^3 = 0.95$  is needed to achieve the required concentration. Experiment 4 spends 2857 unit time, with the material recovery being only 37.6%, whereas Experiment 5 consumes 1132 unit time, with the material recovery being up to 94.9%. This suggests that it is more important to plan the separation strategies for a shorter cascade than for a longer cascade.

The results for the SW method are presented in Table 4 for two cascades of length  $N = 21$  and  $N = 11$ , respectively, corresponding to the previous two conventional cascades. Only one separation run is enough to produce the required concentration. Comparing Table 4 with Tables 2 and 3, the SW method is

**Table 3.** Experiments for Separation of  $^{124}\text{Xe}$  with a Conventional Cascade of Length  $N = 11$ 

Experiment	$r$	$N_F$	$\Theta^r$	$C_1^{P,r}$	$T(M)$	$R(E_t)$
1 (No restriction on $\Theta^r$ )	1	1	0.00037	0.9990	2703	0.397
2 ( $\Theta^r \geq 0.001$ )	1	6	0.001	0.9237	1205	0.892
	2	5	0.83	0.9991		
3 ( $\Theta^r \geq 0.001$ )	1	9	0.005	0.1860	3333	0.322
	2	1	0.06	0.9991		
4 ( $\Theta^r \geq 0.01$ )	1	9	0.01	0.0930	2857	0.376
	2	1	0.035	0.9990		
5 ( $\Theta^r \geq 0.01$ )	1	9	0.01	0.0930	1132	0.949
	2	6	0.093	0.9745		
	3	6	0.95	0.9990		
6 ( $\Theta^r \geq 0.1$ )	1	10	0.1	0.0093	3333	0.322
	2	10	0.1	0.0930		
	3	9	0.2	0.4650		
	4	1	0.15	0.9990		
7 ( $\Theta^r \geq 0.1$ )	1	10	0.1	0.0093	2151	0.500
	2	10	0.1	0.0930		
	3	9	0.15	0.6197		
	4	2	0.31	0.9990		

advantageous over the conventional method in at least the following four respects:

1. There is no need for long cascades. Apparently, the differences in  $T$  and  $R$  between the two cascades of 21 and 11 stages are very little and can be ignored in practice. For  $N = 21$  the results are just the optimal ones and for  $N = 11$  the results are very close to the optimal ones. This means that using a cascade longer than 11 stages is not necessary.
2. The operation is simple and convenient. Using the SW method, one does not have to set up several runs, saving the troubles of choosing

**Table 4.** Experiments for Separation of  $^{124}\text{Xe}$  with the SW Method for Two Cascades of Different Length

Experiment	$N_F$	$C_1^P$	$T(M)$	$R(E_t)$
1 ( $N = 21$ )	11	0.9990	1074	1074
2 ( $N = 11$ )	5	0.9990	1083	1083



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appropriate cascade cuts, feed locations, as well as the preparations between any two successive runs.

3. No worries about whether or not the amount of feed is sufficient for the last few runs.
4. Short operation time-consumption and high material recovery. This is obvious as shown in Table 4.

The results of the SW method are close to the optimal one. Therefore, the SW method can be said to be almost optimal.

## SEPARATION OF A MIDDLE COMPONENT

## The Idea

Take the separation of the 5th component,  $^{130}\text{Xe}$ , in the natural Xe isotope mixture as an example. The difficulty of separating  $^{130}\text{Xe}$  lies in not only that it is a middle component, but also that its concentration is much smaller compared with those of its two neighboring components. With a sufficiently long cascade, the separation can be fulfilled in only two runs, as mentioned earlier. But in practice, once again, the problems are the length of cascades as well as the exact control of cascade cut. Here we are not going to investigate the nonsteady-state approach especially used in separating middle components, but instead to explore the feasibility of using the same idea as for separating end components. The idea is explained as follows. Assume that there is only the waste withdrawal in an SW cascade. The lighter components are easier than the heavier components to accumulate in the product end, i.e., in  $H'_N$ . As the cascade operates, more and more lighter components get accumulated in  $H'_N$  and heavier components are gradually expelled from  $H'_N$  one after another. A middle component may first accumulate in  $H'_N$  and then dispel from it. This means that the concentration of the component would reach a maximum at a certain time, which is shown in Fig. 2 for Xe isotopes.

The results plotted are from an SW cascade of length  $N = 21$  and with a feed of natural concentration at  $N_F = 13$ ,  $H'_N = 1$ , and one waste withdrawal  $W_1 = 1$ . For simplicity all  $H_n$ ,  $H'_n$ , and  $H''_n$  except  $H'_N$  are taken to be zero, the same as before. From the figure, one sees that the concentration  $C'_N$  of  $^{130}\text{Xe}$  reaches a maximum of 0.1179 at the time  $t = 1.83$  unit time. At this moment,  $H'_N$  is collected as the product. For better separation performance, the SW method can be exploited in different manners. It is not always the case that  $H'_N = 1$ , it is often the case that in a run  $H'_N = 1$  and  $W_1 = 1$ , and in the next run  $H''_1 = 1$  and  $P_N = 1$ , alternatively collecting  $H'_N$  or  $H''_1$ . Furthermore, when  $^{130}\text{Xe}$  has been enriched to some extent, numerical experiments show that it may be even more

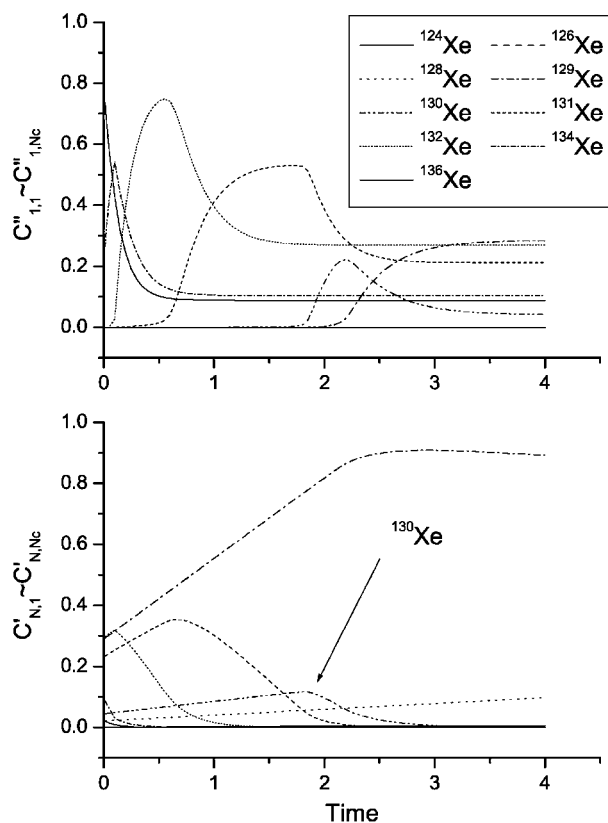


Figure 2. Variation with time of concentrations of Xe isotope components in  $H'_N$ .

efficient to collect the withdrawal  $W_1$  or  $P_N$  as the feed for the following run rather than  $H'_N$  or  $H''_1$ .

### The Optimal Result

With a sufficiently long cascade, the two separation runs proceed as follows. In the first run take the cascade cut to be  $\Theta^1 = \sum_{i=1}^5 C_i^{F,1}$ . Then the first five components go into the product withdrawal, in which the concentration of  $^{130}\text{Xe}$  is enriched to  $C_5^{P,1} = C_5^{F,1} / \sum_{i=1}^5 C_i^{F,1}$ . In the second run, if the required concentration of  $^{130}\text{Xe}$  is  $C_5^{W,2}$ , take the cascade cut to be  $\Theta^2 = 1 - C_5^{P,1} / C_5^{W,2} = 1 - C_5^{F,1} / (C_5^{W,2} \sum_{i=1}^5 C_i^{F,1})$ . It is easy to calculate that the total amount of material



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needed to produce 1 unit mass of  $^{130}\text{Xe}$  is  $M_o = 1/[\Theta^1(1 - \Theta^2)] = C_5^{W,2}/C_5^{F,1}$ , and the time spent is  $T_o = M_o/F = M_o$ . Clearly the material recovery is 1.

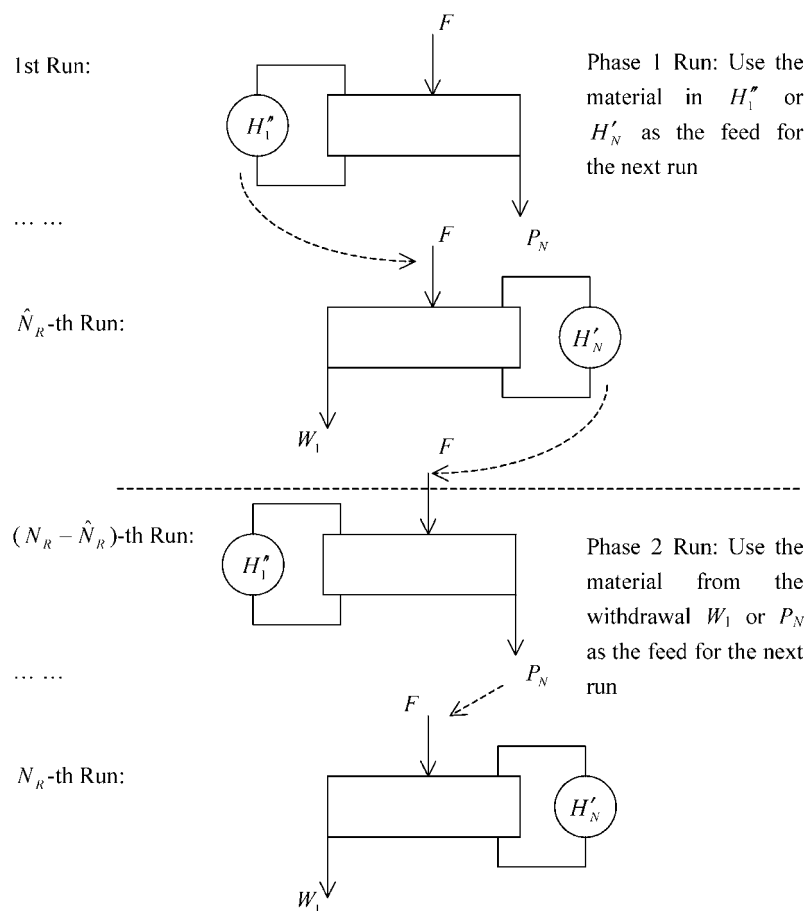
Of course, one does not have to let  $^{130}\text{Xe}$  go with the four lighter components in the first run. It can also be grouped with the four heavier components, in which case all  $^{130}\text{Xe}$  would appear in the waste withdrawal by taking the cascade cut to be  $\Theta^1 = 1 - \sum_{i=5}^9 C_i^{F,1}$ . Then in the second run, using the waste withdrawal of the first run instead of the product withdrawal as the feed for the second run, and taking the cascade cut to be  $\Theta^2 = C_5^{F,1}/(C_5^{P,2} \sum_{i=5}^9 C_i^{F,1})$ , the concentration of  $^{130}\text{Xe}$  in the product withdrawal is  $C_5^{P,2}$ . Therefore,  $M_o = 1/[(1 - \Theta^1)\Theta^2] = C_5^{P,2}/C_5^{F,1}$ . It can be readily found out that no matter how the two runs are arranged, to obtain 1 unit mass of  $^{130}\text{Xe}$  of the same concentration the amount of feed material and the total separation time do not change.

Comparing the result here with that from separating an end component, it is seen that the total mass and the operation time are all correspondingly the same. This means that with a sufficiently long cascade theoretically separating a middle component is just as easy as separating an end component, and the total amount of material and separation time needed only depends on the required concentration and the original concentration of the required component. Therefore, the result here is also optimal in the same sense as before, that is, least material consumption, shortest cascade operation time, and largest material recovery. For  $^{130}\text{Xe}$  and  $C_5^{P,2} = 0.999$ , we have  $M_o = T_o \approx 24.49$ .

Calculation of  $M$ ,  $T$ ,  $R$ , and  $E_t$ 

Suppose that, to obtain 1 unit mass of the final product whose concentration is greater than 99.9% needs a total number  $N_R$  of runs. To improve separation performance, the  $N_R$  runs are split into two phases. In the first  $\hat{N}_R$  runs, which are referred to as Phase 1 Run,  $H'_N$  or  $H''_1$  are collected, and in the following  $N_R - \hat{N}_R$  runs, which are referred to as Phase 2 Run, the withdrawals  $P_N$  or  $W_1$  are collected. See Fig. 3 for such a separation run queue. As before, assume that  $H'_N$  or  $H''_1$  is always 1 unit mass. For simplicity in the later  $N_R - \hat{N}_R$  runs the cascade operation time of each run is fixed and is denoted as  $T_L$ . So in these runs each run needs  $T_L F$  unit mass of material and produces  $T_L F - 1$  unit mass of product (remember  $F = 1$  always); that the amount of material is one unit mass less than the product is due to  $H'_N$  or  $H''_1$ . That is, starting from the  $(\hat{N}_R + 1)$ th run, in every run the amount of withdrawal obtained is always 1 unit mass less than the amount of feed. Therefore, during the last run of  $T_L$  unit time, the final product of  $T_L F - 1$  unit mass is produced, which requires that a total amount of feed needed for the  $(\hat{N}_R + 1)$ th run, i.e., the first run in Phase 2 Run, is  $T_L F + N_R - \hat{N}_R - 1$ . In the first  $\hat{N}_R$  runs of Phase 1 Run, the cascade operation time of a run is determined according to when the concentration of  $^{130}\text{Xe}$  in  $H'_N$  or  $H''_1$  reaches





**Figure 3.** The separation run queue for separating a middle component.

maximum. Let the operation time of the  $r$ th run be  $T_r$ . So the total amount of material needed is  $T_r F$ , just the same as in the case for the separation of an end component. It is clear that to obtain 1 unit mass of product in the  $\hat{N}_R$ th run the total material for the first run is  $\prod_{r=1}^{\hat{N}_R} (T_r F)$  and the total operation time is  $T_1 \prod_{r=2}^{\hat{N}_R} (T_r F)$ . Therefore to produce 1 unit mass of the final product, the total amount of material is

$$M = \frac{(T_L F + N_R - \hat{N}_R - 1)}{T_L F - 1} \prod_{r=1}^{\hat{N}_R} (T_r F)$$

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and the total time consumption is

$$T = \frac{1}{T_L F - 1} \left\{ (T_L F + N_R - \hat{N}_R - 1) T_1 \prod_{r=2}^{\hat{N}_R} (T_r F) \right. \\ \left. + \frac{(N_R - \hat{N}_R)}{F} \left[ T_L F + \frac{1}{2} (N_R - \hat{N}_R - 1) \right] \right\}$$

If the actual concentration of  $^{130}\text{Xe}$  in the final product is  $C_5^{P,N_R}$ , then the material recovery is

$$R = C_5^{P,N_R} / (C_5^{F,1} M)$$

## Numerical Experiments

In the numerical experiments, again two cascade lengths are considered,  $N = 11$  and  $N = 21$ , respectively. When operating in the conventional way, the feed position  $N_F$  and cascade cut  $\Theta$  are determined by maximizing the concentration of  $^{130}\text{Xe}$  in either the product withdrawal or the waste withdrawal. The concentrations of  $^{129}\text{Xe}$ ,  $^{130}\text{Xe}$ , and  $^{131}\text{Xe}$  are presented of successive separation runs in Tables 5 and 6. Alternatively choosing  $C_5^{P,r}$  and  $C_5^{W,r}$  as the quantity for maximization is due to the fact that if  $\sum_{i=1}^5 C_i^{F,r} < \sum_{i=5}^9 C_i^{F,r}$  in the  $r$ th run (remember that  $C_i^{F,r} = C_i^{P,r-1}$  or  $C_i^{F,r} = C_i^{W,r-1}$ , depending on which withdrawal, the product withdrawal, or the waste withdrawal of the  $(r-1)$ th run is collected for the feed of the  $r$ th run),  $C_5^{P,r}$  should be maximized, because the theory for a sufficiently long cascade tells us  $\max(C_5^{P,r}) = C_5^{F,r} / \sum_{i=1}^5 C_i^{F,r} > \max(C_5^{W,r}) = C_5^{F,r} / \sum_{i=5}^9 C_i^{F,r}$ , otherwise  $C_5^{W,r}$  should be maximized. Actually in the later stages of separation runs, only the concentrations of the two neighboring components of  $^{130}\text{Xe}$  remain large enough to affect the concentration of  $^{130}\text{Xe}$ , so that whether  $C_5^{P,r}$  or  $C_5^{W,r}$  should be maximized can be determined by just looking at the concentrations of the two neighboring components. Doing this way leads to a faster enrichment of  $^{130}\text{Xe}$ . For example, in the 6th run as shown in Table 6, if  $C_5^{P,6}$  is chosen to be maximized instead of  $C_5^{W,6}$ , then  $\max(C_5^{P,6})$  is 0.9887, whereas  $\max(C_5^{W,6})$  is 0.9978, because  $C_4^{F,6} (= C_4^{P,5}) = 0.0108 > C_6^{F,6} (= C_6^{P,5}) = 0.0016$ . Note that here no restriction is exerted on the smallest possible cascade cut when maximizing  $C_5^{P,r}$  or  $C_5^{W,r}$ .

The results from the SW method are given in Tables 7 and 8. The choice for the values of  $\hat{N}_R$  here is somewhat arbitrary: Phase 1 Run stops when the concentration of  $^{130}\text{Xe}$  is larger than those of its two neighboring components. To achieve a fast possible enrichment of  $^{130}\text{Xe}$ , the collection of  $H_N$  or  $H_1^I$  in Phase 1 Run and that of  $P_N$  or  $W_1$  in Phase 2 Run depend also on the concentrations of the two neighboring components of  $^{130}\text{Xe}$  in the same way as above. Note that in

**Table 5.** Separation of  $^{130}\text{Xe}$  with a Conventional Cascade of Length  $N = 11$ 

Run Number	Concentration	Cascade				
	Maximized	$N_F$	Cut $\Theta$	$C_4 (^{129}\text{Xe})$	$C_5 (^{130}\text{Xe})$	$C_6 (^{131}\text{Xe})$
1	$C_5^{P,1}$	7	0.3740	0.7049	0.0994	0.1316
2	$C_5^{W,2}$	7	0.7000	0.2756	0.2638	0.4335
3	$C_5^{P,3}$	6	0.5858	0.4675	0.3925	0.1382
4	$C_5^{W,4}$	6	0.4679	0.1289	0.6128	0.2575
5	$C_5^{P,5}$	5	0.7048	0.1817	0.7514	0.0669
6	$C_5^{W,6}$	6	0.2707	0.0400	0.8691	0.0909
7	$C_5^{P,7}$	6	0.6958	0.0498	0.9305	0.0196
8	$C_5^{W,8}$	6	0.1860	0.0100	0.9661	0.0239
9	$C_5^{P,9}$	6	0.8318	0.0119	0.9833	0.0048
10	$C_5^{W,10}$	6	0.1616	0.0023	0.9920	0.0057
11	$C_5^{P,11}$	6	0.8412	0.0028	0.9961	0.0011
12	$C_5^{W,12}$	6	0.1551	0.0005	0.9981	0.0013
13	$C_5^{P,13}$	6	0.8429	0.0006	0.9991	0.0003

$M = T = 235.1, R = E_t = 0.104$

Phase 2 Run, the concentrations given in the tables are averaged values for the period of cascade operation, since they are not constant in the withdrawals. It is worth pointing out that the value of  $T_L$  is an important parameter that affects the material recovery and the operation time efficiency. Being too large or too small is not a wise choice.

**Table 6.** Separation of  $^{130}\text{Xe}$  with a Conventional Cascade of Length  $N = 21$ 

Run Number	Concentration	Cascade				
	Maximized	$N_F$	Cut $\Theta$	$C_4 (^{129}\text{Xe})$	$C_5 (^{130}\text{Xe})$	$C_6 (^{131}\text{Xe})$
1	$C_5^{P,1}$	13	0.3406	0.7763	0.1179	0.4394
2	$C_5^{W,2}$	12	0.8186	0.1437	0.6126	0.2421
3	$C_5^{P,3}$	12	0.7500	0.1916	0.7900	0.0184
4	$C_5^{W,4}$	12	0.2230	0.0102	0.9662	0.0236
5	$C_5^{P,5}$	12	0.9476	0.0108	0.9876	0.0016
6	$C_5^{W,6}$	12	0.0650	0.0004	0.9978	0.0017
7	$C_5^{P,7}$	12	0.9510	0.0004	0.9994	0.0001

$M = T = 32.96, R = E_t = 0.743$



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**Table 7.** Concentrations of  $^{129}\text{Xe}$ ,  $^{130}\text{Xe}$ , and  $^{131}\text{Xe}$  in the Products Collected in Different Separation Runs ( $\hat{N}_R = 3$ ,  $N = 11$ ,  $N_F = 6$ )

Run No.	Collection	$C_4$ ( $^{129}\text{Xe}$ )	$C_5$ ( $^{130}\text{Xe}$ )	$C_6$ ( $^{131}\text{Xe}$ )
1	$H'_N$	0.7435	0.1020	0.0930
2	$H''_1$	0.1326	0.3749	0.4807
3	$H'_N$	0.3032	0.6227	0.0731
$T_L = 2.50$				
4	$W_1$	0.0909	0.8069	0.1020
5	$P_N$	0.1260	0.8603	0.0136
6	$W_1$	0.0193	0.9618	0.0189
7	$W_1$	0.0023	0.9716	0.0261
8	$P_N$	0.0032	0.9936	0.0033
9	$P_N$	0.0044	0.9952	0.0004
10	$W_1$	0.0005	0.9989	0.0005
11	$P_N$	0.0007	0.9992	0.0001
$M = 198.6$ , $T = 130.6$ , $R = 0.123$ , $E_t = 0.106$				
$T_L = 5.00$				
4	$W_1$	0.1795	0.7329	0.0876
5	$W_1$	0.0565	0.8385	0.1049
6	$P_N$	0.0675	0.9102	0.0223
7	$W_1$	0.0123	0.9610	0.0267
8	$P_N$	0.0147	0.9810	0.0043
9	$W_1$	0.0023	0.9926	0.0052
10	$P_N$	0.0027	0.9965	0.0008
11	$W_1$	0.0004	0.9987	0.0009
12	$P_N$	0.0005	0.9994	0.0001
$M = 101.9$ , $T = 122.2$ , $R = 0.240$ , $E_t = 0.200$				
$T_L = 7.50$				
4	$W_1$	0.2206	0.6965	0.0828
5	$W_1$	0.1304	0.7758	0.0937
6	$W_1$	0.0459	0.8480	0.1061
7	$P_N$	0.0518	0.9154	0.0327
8	$W_1$	0.0114	0.9516	0.0370
9	$P_N$	0.0129	0.9793	0.0079
10	$W_1$	0.0024	0.9887	0.0089
11	$P_N$	0.0027	0.9956	0.0017
12	$W_1$	0.0005	0.9976	0.0019
13	$P_N$	0.0006	0.9991	0.0003
$M = 79.59$ , $T = 98.05$ , $R = 0.308$ , $E_t = 0.250$				

**Table 8.** Concentrations of  $^{129}\text{Xe}$ ,  $^{130}\text{Xe}$ , and  $^{131}\text{Xe}$  in the Products Collected in Different Separation Runs ( $\hat{N}_R = 2$ ,  $N = 21$ ,  $N_F = 11$ )

Run No.	Collection	$C_4$ ( $^{129}\text{Xe}$ )	$C_5$ ( $^{130}\text{Xe}$ )	$C_6$ ( $^{131}\text{Xe}$ )
1	$H'_N$	0.7733	0.1164	0.0487
2	$H''_1$	0.0271	0.6626	0.3090
$T_L = 5.00$				
3	$P_N$	0.0325	0.7935	0.1740
4	$P_N$	0.0390	0.9433	0.0177
5	$W_1$	0.0010	0.9779	0.0212
6	$P_N$	0.0012	0.9983	0.0005
7	$W_1$	0.0000	0.9993	0.0007
$M = 39.80$ , $T = 48.56$ , $R = 0.615$ , $E_t = 0.504$				
$T_L = 10.00$				
3	$P_N$	0.0298	0.7281	0.2421
4	$P_N$	0.0328	0.7999	0.1674
5	$P_N$	0.0360	0.8784	0.0855
6	$P_N$	0.0396	0.9549	0.0054
7	$W_1$	0.0013	0.9927	0.0060
8	$P_N$	0.0014	0.9984	0.0002
9	$W_1$	0.0000	0.9998	0.0002
$M = 31.44$ , $T = 41.55$ , $R = 0.779$ , $E_t = 0.589$				
$T_L = 15.00$				
3	$P_N$	0.0289	0.7063	0.2648
4	$P_N$	0.0308	0.7527	0.2165
5	$P_N$	0.0336	0.8210	0.1453
6	$P_N$	0.0361	0.8794	0.0845
7	$P_N$	0.0385	0.9364	0.0251
8	$W_1$	0.0016	0.9716	0.0268
9	$P_N$	0.0017	0.9972	0.0011
10	$W_1$	0.0001	0.9988	0.0012
11	$P_N$	0.0001	0.9999	0.0000
$M = 29.06$ , $T = 41.27$ , $R = 0.843$ , $E_t = 0.593$				

Comparing Table 5 with Table 7, and Table 6 with Table 8, one can conclude that the SW method is, in general, advantageous over the conventional method, especially for shorter cascades. With an appropriate choice of  $T_L$ , the SW method can always be tuned to perform better than the conventional method, but the superiority is not as large as for separating end components. As the length of cascade increases, the advantages of the SW method seem to be gradually lost,



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and the conventional method becomes comparable with it. This is understandable, because a conventional cascade that is long enough is a “sufficiently long cascade” and would yield the optimal result.

## CONCLUSIONS

A method, called the SW method is discussed in separating components of small abundance. Separation of  $^{124}\text{Xe}$  and  $^{130}\text{Xe}$  in the natural Xe isotope mixture is used as an example on two cascades of different lengths  $N = 11$  and 21, respectively. On the basis of producing one unit mass of product of the same concentration, the results are compared in two respects, the material recovery and the operation time efficiency, with the optimal results and those from the same two cascades but operated in a conventional way. For separating an end component of small abundance, here  $^{124}\text{Xe}$ , the SW method is nearly optimal, and its advantages over the conventional method are obvious. For separating a middle component, that is  $^{130}\text{Xe}$  here, the SW method is in general superior to the conventional method but not very satisfactory. The SW method seems to be more suitable for separating end components than for middle components.

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