# Influence of changing feed flow value on germanium isotope separation in gas centrifuge cascade

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**Abstract.** Modeling of a nonstationary process caused by changing the cascade feed flow value using germanium isotope separation as an example. During a nonstationary process, the concentrations of isotopes with intermediary mass number in light and heavy fraction flows may cross the range limits restricted by the initial and final stationary values and exceed the maximum reachable values for a three-flow cascade. Time for stationary isotope concentrations to settle is different for isotopes and depends on the final values of cascade flow.

## 1. Introduction

During the operation of gas centrifuge (GC) cascade for the multicomponent isotope mixture (MIM) separation there are nonstationary hydraulic processes when the values of pressures and process gas flows change in the cascade stages and in outgoing flows. The nonstationary hydraulic process generates a nonstationary separation process, in the course of which a change takes place in the isotope composition of the process gas in the cascade stages and flows. One of the most frequent origins of nonstationary processes is change of cascade flow rate (feed, light or heavy fraction). Values of pressure, flows of process gas and isotope concentration into cascade are changed during this process. Research of these processes is of interest for determination laws of isotope concentration change.

In the authors' opinion, an experimental approach in this case appears to be quite costly, so it is strongly recommended that nonstationary processes should be investigated by mathematical modeling. A number of publications [1–3] are dedicated to modeling of nonstationary separation processes in cascades for MIM separation. The common drawback of these models [1–3] is that they are of limited use. Modeling is possible only in case of stationary hydraulic parameters of a GC cascade. Besides, the models cannot be used for simultaneous calculation of nonstationary hydraulic and separation processes in cascades for MIM separation. Mathematical model [4-6] is free of above defects and was used to research the specifics of MIM separation during filling of the GC cascade with a process substance [7].

This article contains the research results for nonstationary transfer of isotopic mixture in GC cascade caused by the change in values of cascade feed flow. The research considers the case of

germanium isotopes that are applied in production of semi-conducting materials and research of neutrinoless double β-decay. Germanium isotopes are also used as a starting material for arsenic radioactive isotopes (e.g. $^{70}$Ge is used to obtain radioactive $^{72}$As that finds application in medical diagnostics).

## 2. Mathematical model

The basic equations that are included in the mathematical model of a nonstationary process in MIM separation are described below.

MIM separation occurs in a GC cascade. A process gas contains $n$ components (isotopes) identified by index $j$ ($j=\overline{1;n}$). The cascade consists of $S$ separation stages (figure 1) numbered by index $i$ ($i=\overline{1;S}$). The stages are arranged in a symmetric-countercurrent layout. The input feed flow $F$ enters the cascade, the light fraction flow $P$ and the heavy fraction flow $W$ are withdrawn. The feed flow is fed into the stage $S_F$; the heavy fraction flow is withdrawn from the first stage; the light fraction flow is withdrawn from the stage $S$.

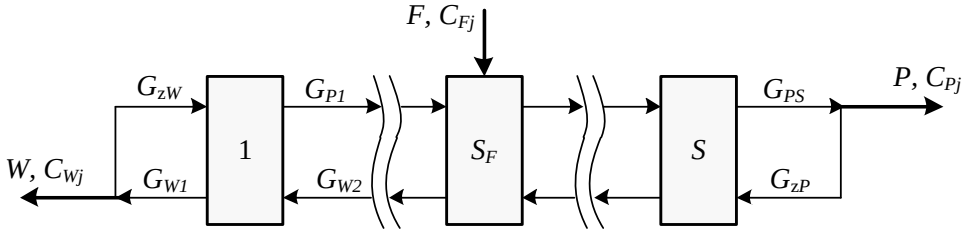


**Figure 1.** Separation cascade.

The separation stage consists of $N_i$ GCs connected in parallel. The feed flow $G_{Fi}$ is fed into the stage. The light fraction flow $G_{Pi}$ and the heavy fraction flow $G_{Wi}$ are withdrawn from the stage. The concentrations of the $j^{\text{th}}$ component in the feed, light and heavy fraction flows of the $i^{\text{th}}$ stage are designated as $C_{Fij}$, $C_{Pij}$, $C_{Wij}$, respectively. Part of the heavy fraction flow of the cascade can be fed into the first stage by return heavy fraction flow which is designated as $G_{zW}$. Part of the light fraction flow of the cascade can be fed into the stage $S$ by return light fraction flow which and is designated as $G_{zP}$.

If no process gas is lost due to corrosion losses, in stationary hydraulic conditions, the stage flows and the concentrations of the components in them can be related by the material and component (isotope) balance equations:

$$G_{Fi} = G_{Pi} + G_{Wi}, \tag{1}$$

$$G_{Fi}C_{Fij} = G_{Pi}C_{Pij} + G_{Wi}C_{Wij}, \tag{2}$$

$$\theta_i = \frac{G_{Pi}}{G_{Fi}}, \tag{3}$$

where $\theta_i$ is the cut of the $i^{\text{th}}$ stage.

The sum of the component concentrations in each flow is equal to 1.

The flow $G_{Fi}$ entering the $i^{\text{th}}$ stage can be calculated by:

$$G_{Fi} = \begin{cases} \delta_i F + G_{zW} + G_{Wi+1}, & i = 1 \\ \delta_i F + G_{Pi-1} + G_{Wi+1}, & i = \overline{2;S-1} \\ \delta_i F + G_{Pi-1} + G_{zP}, & i = S \end{cases}, \tag{4}$$

$$\delta_i = \begin{cases} 1, & i = S_F \\ 0, & i \neq S_F \end{cases}, \tag{5}$$

where $\delta_i$ is the factor showing the input of feed flow $F$ into the $i^{th}$ stage.

The concentration $C_{Fij}$ in the flow $G_{Fi}$ is given by:

$$C_{Fij} = \frac{1}{G_{Fi}} \cdot \begin{cases} \delta_i F C_{Fj} + G_{zW} C_{Wij} + G_{Wi+1} C_{Wi+1\,j}, & i = 1 \\ \delta_i F C_{Fj} + G_{Pi-1} C_{Pi-1\,j} + G_{Wi+1} C_{Wi+1\,j}, & i = \overline{2; S-1} \\ \delta_i F C_{Fj} + G_{Pi-1} C_{Pi-1\,j} + G_{zP} C_{Pij}, & i = S \end{cases}. \tag{6}$$

The rotation frequency of the GC rotors and their operating temperature are assumed constant.

The separation of components in the stage is defined by expressions [8-10]:

$$\chi_{ijl} = \frac{C_{Pij} C_{Wil}}{C_{Pil} C_{Wij}}, \tag{7}$$

$$\chi_{ijl} = \chi_{0i}^{M_j - M_l}, \tag{8}$$

where $\chi_{jl}$ is the separation factor of the $j^{th}$ and $l^{th}$ components in the $i^{th}$ stage; $\chi_{0i}$ is the overall separation factor per unit of mass number difference; $M_j$, $M_l$ are the mass numbers of the $j^{th}$ and $l^{th}$ components, respectively.

The $\chi_{0i}$ value depends on the hydraulic parameters of the stage:

$$\chi_{0i} = f(G_{Fi}, \theta_i). \tag{9}$$

In the stationary hydraulic conditions, the flows of cascade and its component concentrations can be related by the balance equations:

$$F = P + W, \tag{10}$$

$$FC_{Fj} = PC_{Pj} + WC_{Wj}. \tag{11}$$

Weight-average concentration of germanium isotope in stage $\overline{C}_{ij}$ is calculated by:

$$\overline{C}_{ij} = \frac{H_{Fi} C'_{Fij} + H_{Pi} C_{Pij} + H_{Wi} C_{Wij} + H'_i C'_{ij}}{H_{Fi} + H_{Pi} + H_{Wi} + H'_i}, \tag{12}$$

where $H'_i$, $H_{Fi}$, $H_{Pi}$, $H_{Wi}$ are holdup of GC, feed, light and heavy fraction manifolds, kg; $C'_i$, $C'_{Fi}$, $C_{Pi}$, $C_{Wi}$ are concentrations of the $j^{th}$ component of GC, feed, light and heavy fraction lines.

Algorithm for differential equations that describe nonstationary hydraulic and separation processes is provided in [4, 5].

In order to evaluate parameters of a square cascade (SC) that has equal holdup $H$, flow rate $G_F$ and separation factor $\chi_0$ for all stages, the following values are used [1]:

$$\omega = \frac{H}{G_F}, \tag{13}$$

$$\tau = \frac{t \ln \chi_0}{\omega}, \tag{14}$$

where $\omega$ is stage constant, s; $H$ is stage holdup, kg; $\tau$ is dimensionless time.

## 3. Results and discussion

Germanium tetrafluoride $GeF_4$ ($\mu$ = 149 kg/kmol) was used as a process gas for separation of germanium isotopes. $^{72}Ge$ was selected as the desired isotope. Square cascade with the following parameters was considered: GC separation coefficient $\chi_0$ = 1.1; number of stages $S$ = 100; stage for feed flow input $S_F$ = 57; ratio of feed flow to stage flow rate $F/G_F$ = 0.08; ratio of light fraction flow to feed flow $P/F$ = 0.484; stage constant $\omega$ = 0.23 s.

**Table 1.** Isotopic composition of germanium in cascade flows (initial condition).

| Ge isotope mass number (u) | $GeF_4$ molecule mass number (u) | Concentration in flow | | |
|---|---|---|---|---|
| | | heavy fraction (%) | feed (%) | light fraction (%) |
| 70 | 146 | 0.00 | 20.52 | 42.40 |
| 72 | 148 | 0.67 | 27.45 | 56.00 |
| 73 | 149 | 13.58 | 7.76 | 1.56 |
| 74 | 150 | 70.73 | 36.52 | 0.04 |
| 76 | 152 | 15.02 | 7.75 | 0.00 |

Calculation was carried out for a nonstationary process caused by an instantaneous change of feed flow value at time $\tau$ = 0. Ratios of final cascade feed value $F_{кон}$ to initial feed flow value $F$ were taken as equal to 0.8, 0.9, 1.1, and 1.2. Light fraction flow remains unchanged ($P/F$ = const), and heavy fraction flow starts to change through a certain period of time after flow $F$ changes (at time $\tau \approx 1{,}5$) due to cascade holdup and time of response (figure 2) and asympomatically reaches to final stationary value $W/G_{Fend}$.

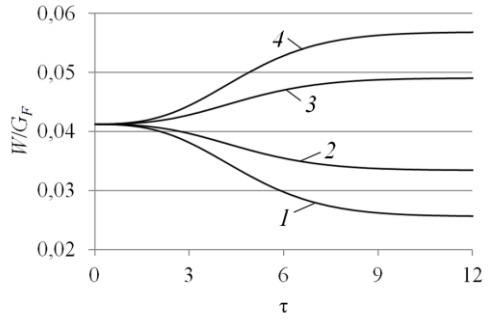


**Figure 2.** Change in time of cascade heavy fraction flow value depending on the final value of cascade feed flow:
*1* – $F_{end}/F$=0.8; *2* – $F_{end}/F$ =0.9;
*3* – $F_{end}/F$ =1.1; *4* – $F_{end}/F$ =1.2.

Final stationary value of the heavy fraction flow in cascade settles at time $\tau \approx 9$ irrespective of final value of $W/G_{Fend.}$

Change of feed flow value affects not only heavy fraction flow in the cascade, but also its isotopic composition (figure 3). The curves of $^{72}Ge$ and $^{74}Ge$ isotope concentration in flow $W$ during a nonstationary process are monotonous irrespective from final values of cascade feed flow $F_{end}$.

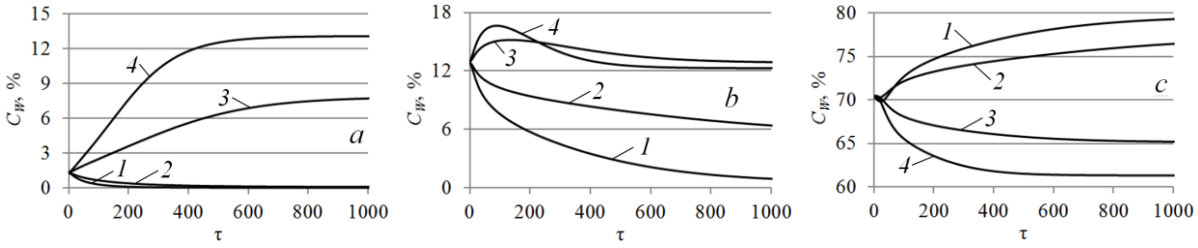


**Figure 3.** Concentration of germanium isotopes in the heavy fraction flow during a nonstationary process: a – $^{72}Ge$; b – $^{73}Ge$; c – $^{74}Ge$; *1* – $F_{кон}/F$ = 0.8; *2* – $F_{кон}/F$ = 0.9; *3* – $F_{кон}/F$ = 1.1; *4* – $F_{кон}/F$ = 1.2.

The increase in feed flow (at $F_{end}/F = 1.1$ and $F_{end}/F = 1.2$) is accompanied with temporary increase in concentration of $^{73}$Ge, which reaches a maximum values of 15.2 and 16.6 % at times $\tau \approx 130$ and $\tau \approx 90$ correspondingly, after that $^{73}$Ge concentration decreases to final stationary values of 12.8 and 12.3%. This pattern of change in $^{73}$Ge concentration in flow $W$ is caused by maximum concentration of this isotope in the stages in the middle part of the cascade (particularly in stage 23) in the initial state. Changing the values of flows $F$ and $W$ (at $F_{end}/F = 1.2$) results in evacuation of the excessive amount of $^{73}$Ge by transferring this isotope from the middle part of the cascade towards stage 1.

Light fraction flow remains constant during a nonstationary process. Nevertheless, during a nonstationary process the concentration of germanium isotopes changes in this flow (figure 4).

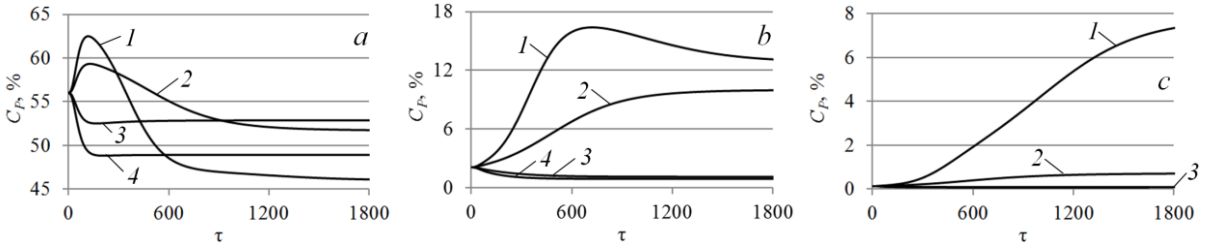


**Figure 4.** Concentration of germanium isotopes in the light fraction flow during a nonstationary process: a – $^{72}$Ge; b – $^{73}$Ge; c – $^{74}$Ge; *1* – $F_{end}/F = 0.8$; *2* – $F_{end}/F = 0.9$; *3* – $F_{end}/F = 1.1$; *4* – $F_{end}/F = 1.2$.

As seen in figure 4a, $^{72}$Ge concentration changes abruptly at $F_{end}/F = 0.8$ to the value of 62.5 %, exceeding the maximum reachable limit of concentration for a three-flow cascade. $^{73}$Ge concentration is unimodal with the maximum value of 16.4 % at time $\tau \approx 720$. It should be noted that the duration of the process of settling stationary concentrations of germanium isotopes depends on the final value of $F_{end}$ (or $F_{end}/F$): maximum duration corresponds to the case of $F_{end}/F = 0.8$ resulting in the increased amount of $^{74}$Ge and $^{76}$Ge isotopes; the amount of other isotopes decreases (figure 5).

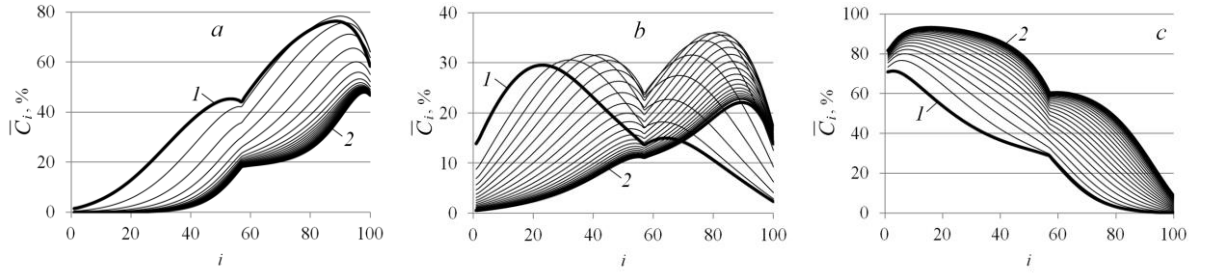


**Figure 5.** The change of the weighted average concentration of germanium isotopes in cascade stages during a nonstationary process: a – $^{72}$Ge; b – $^{73}$Ge; c – $^{74}$Ge; *1* – initial state; *2* – final state.

The change in monotonousness of curves in figure 6 can be explained by mixing inter-stage flows and feed flow of the cascade, which have different isotope composition, at the input of stage 57. During a nonstationary process, $^{73}$Ge isotope transfers from stage 23 to stage 90 and decreases from 29.6 to 22.1 %. Similar situation is observed for $^{72}$Ge isotope. The amount of $^{74}$Ge isotope in the cascade increases approximately 2.2 times (the weighted average concentration of $^{74}$Ge in the cascade holdup increases approximately from 30.3 to 67.8 %).

## 4. Conclusion

There have been carried out a research of a nonstationary process caused by the change of cascade feed flow value using germanium isotope separation as an example. It was determined that during a nonstationary process, the concentrations of isotopes with intermediary mass number in light and

heavy fraction flows may cross the range limits restricted by the initial and final stationary values and exceed the maximum reachable values for a three-flow cascade. Time for stationary isotope concentrations to settle is different for isotopes and depends on the final values of cascade flow.

**References**

[1] Laguntcov N I, Levin E V, Nikolaev B I and Sulaberidze G A 1987 *Atomic energy* **62** 394
[2] Zeng S and Ying C 2001 *Separation science and technology* **36** 3439
[3] Smirnov A Yu, Sulaberidze G A, Borisevich V D, Zeng S and Jiang D 2015 *Chemical engineering and science* **127** 418
[4] Orlov A A, Ushakov A A and Sovach V P 2017 *MATEC Web of Conferences* **92** 01033
[5] Orlov A A, Ushakov A A and Sovach V P 2017 *Journal of engineering physics and thermophysics* **90** 258
[6] Orlov A A, Ushakov A A, Sovach V P and Malyugin R V 2018 *Journal of engineering physics and thermophysics* **91** 565.
[7] Orlov A A, Ushakov A A and Sovach V P 2017 *MATEC Web of Conferences* **110** 01063
[8] De la Garza A, Garrett G A and Murphy J E 1961 *Chemical Engineering Science* **15** 188
[9] Von Halle E 1987 *Proc. Separation Phenomena in Liquids and Gases* p 325
[10] Ying C, Guo Z and Wood H G 1996 *Separation Science and Technology* **31** 2455