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SEPARATION OF MULTICOMPONENT ISOTOPE MIXTURES BY GAS CENTRIFUGE

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ABSTRACT

This article represents an analytical review of the state-of-the-art on separation of multicomponent isotope mixtures in a gas centrifuge. The main physical principles, physical-mathematical models, research techniques and peculiarities of separation of multicomponent isotope mixtures in a gas centrifuge are considered and discussed.

INTRODUCTION

It is known that sixty-two of eighty-four naturally existing stable chemical elements are polyisotopic. They comprise 264 stable isotopes (1). Nature provided every isotope with its unique properties such as, mass, spin, neutron interaction cross-section and Mossbauer lines. Each of these isotope features has found an application in different fields of science and technology (2–4). First investigations on separation of multicomponent mixtures of stable isotopes by gas centrifuge

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techniques were started in The Kurchatov Institute of Atomic Energy, Moscow, under supervision of academician Kikoin in parallel with the development of centrifugal technology for uranium isotope enrichment. The first centrifuges were designed for operation with uranium hexafluoride and, as a rule, could not be efficiently used for separation of other isotopes existing in different chemical compounds. For this reason in the former Soviet Union and now in Russia special centrifuges with their own specific characteristics have been designed for separation of stable isotopes by using either existing or specially synthesized gaseous chemical compounds.

The basic condition for process gases to be applicable in the newly designed centrifuges for separation of stable isotopes is their vapor pressure, which cannot be less than 5–10 mm Hg at normal temperature. At the same time, the substance should be non-corrosive to the structural materials of the centrifuge and thermally stable. Moreover, this substance should preferably possess the maximum possible content of the separated element in the molecule. The list of such substances includes fluorides and oxyfluorides of metals and non-metals, metal-organic and complex compounds, phosphorus hydrides and carbonyls, boron hydrides, freons and some others. The synthesis of these volatile compounds is in certain instances difficult, because their properties are difficult to foresee. Hence, the search for new compounds is basically carried out by empirical means.

The accumulated experiments on separation of isotopes of both light (B, N, C, O, Kr, Cl) and heavier elements has shown that gas centrifuge technology is extremely promising. Probably one of the most important features brought in by this method is a large-scale isotope production that opens a new era of their use in industry, science and medicine. The centrifuge cascade used for separation of non-uranium isotopes usually has a level of productivity ten times higher than that of the electromagnetic installation with a comparative feed flow (1). It allowed producing isotopes of germanium, chromium, zinc and tungsten in quantities of 10 or more kilograms. Only during recent years The Research Scientific Center “Kurchatov Institute” produced more than 40 kgs of Cr-50 with enrichment higher than 38% and more than 15 kg of Ge-75 enriched to 86–90% for unique experiments on studying the double beta-decay and investigation of solar neutrino (5,6). At present, for experiments on improvement of radiation safety of nuclear stations one needs thousands of kilograms of zinc depleted with Zn-64 isotope capable for strong activation.

Stable isotopes are widely used as starting materials in the production of radioactive isotopes, which occupy a separate niche in medicine especially for radiopharmaceutical purposes (7). Thus, highly concentrated Mo-98 is used for producing Tc-99 (1). This process does not produce any radioactive wastes and, consequently, is ecologically safe. Radioactive Xe-127 used for diagnostics in pulmonology has been obtained from highly enriched Xe-126. Owing to a short half-life and moderate radiation energy, Xe-127 ensures a low dosage loading on a patient. For production of final targets containing radioisotope Ni-63, an efficient separation technique based on gas centrifuge technology has been developed (8). Radioisotope I-123, widely used in medicine, was offered to be produced from highly enriched by gas centrifuges Te-123 (up to 96.5%) after irradiation with protons in a cyclotron (9).

The gas centrifuge technology ensures the production of isotopes with accessible limit concentrations. For example, Xe-124 with concentration of 99.9% (natural abundance (NA) of 0.096%), 99.999% Xe-136 (NA 8.87%), Sn-112 of 99.5% (NA 0.96%), and Sn-124 of 99.9% (NA 5.94%) have been produced by gas centrifuges. More and more rigid requirements to the isotope purity as well as a sharp rise in demands for isotopes put gas centrifuge technology in a leading position.

An industrial scale complex separation of iron, carbon and oxygen isotopes from iron pentacarbonyl, $Fe(CO)_5$, realized at The Electrochemical Plant in the city of Krasnoyarsk can be considered to be the best achievement in the development of a centrifuge technology for enrichment of stable isotopes. Because all elements composing the $Fe(CO)_5$ molecule are polyisotopic (Fe contains 4, C-2, and O-3 isotopes), iron pentacarbonyl of natural isotope abundance represents a mixture of 284 molecular species with different degrees of isotope replacement. These species are distributed throughout 20 components with molecular masses ranging from 194 to 213. Almost all of them contain several isotopes of all of the above chemical elements. Such isotopic overlapping substantially limits the possibility of direct enrichment of iron isotopes with intermediate masses and heavy carbon and oxygen isotopes by using a centrifugal cascade. This limitation has been overcome due to the introduction of an additional stage into the process consisting of isotope exchange between $Fe(CO)_5$ molecules in so-called photo-reactors. The combination of isotope separation in gas centrifuges with isotope exchange reaction has allowed yielding in one process several highly enriched isotopes: Fe-57 (concentration >99%), C-13 and O-18 (with concentrations up to 80%) (10).

Separation of stable isotopes has transformed into an independent area of development of gas centrifuge technology, whose progress depends on the solution of the following problems: 1) gas centrifuge design; 2) chemical synthesis of process gases; 3) conversion of enriched isotope to desired (demanded) chemical compound; 4) theoretical description of separation of multicomponent isotope mixture, and 5) further development of theory and practice for complex separation of isotopes. Significant distinctions in chemical and physical properties of process gases lead to the necessity to create a wide set of different gas centrifuge constructions optimized for various ranges of molecular masses through accounting for a circulating flow, which substantially multiplies the radial separation effect by centrifugal force field. The development of these constructions strongly depends on the theoretical study and numerical calculations of gas dynamics and separation processes in a rotor of gas centrifuge. For this reason, the present article considers and discusses these particular problems.

PHYSICO-MATHEMATICAL MODEL FOR MASS TRANSFER IN GAS CENTRIFUGE

Description of Centrifuge and Main Separation Principles

A classic schematic of a gas centrifuge is presented in Fig. 1. A gas centrifuge separates in radial direction the lighter molecules from the heavier ones by using the difference in centrifugal force exerted on two or more species. The fundamental advantage of centrifuge arises from the fact that the primary (radial) isotope separation effect occurs at thermodynamic equilibrium. At a constant temperature a thermodynamic equilibrium distribution of concentration for each component of the isotope mixture along the rotor's radius is described by the following equation:

$$n_i\,(r) = n_i\,(0)\,\exp(m_i \omega_0^2 r^2/2\mathrm{RT})$$

(for notations in this and other equations throughout the text see List of Symbols at the end). As follows from this equation, the process gas will be enriched with the light components of mixture near the axis of rotation and depleted with the same components near the side wall of the rotor. The axial counter-current flow pattern to be established by the rotating gas leads to the multiplication of the radial separating effect to the axial direction in accordance with the well-known principle of the counter-current separation column. The circulation flow is excited by the slack action of the stationary scoop used for collection of the fraction enriched with heavier components of the isotope mixture.

The description of the flow and diffusion of a multicomponent isotope mixture in the rotor of a gas centrifuge (moving at a high rotation speed) can be based on the axisymmetric approximation. The flow pattern in the gas centrifuge rotor can be conditionally divided into three subregions in radial direction with various flow features (11): 1) the viscous flow near the side wall; 2) the transient flow area, and 3) a strongly rare gas (called the Knudsen gas) near the axis of rotation. A small part of the transient subregion (as a rule, a small percent of the rotor radius) permits to consider only two subregions without substantial decrease of accuracy. The subregion A is a zone of viscous flow and the subregion B is a zone where the gas is in a state of free-molecular flow (see Fig. 2). The interface between subregions is modeled by an infinitely thin cylindrical surface on which the mass, the momentum and the energy fluxes through this boundary, as well as the tangential components of the viscous stress tensor are assumed to vanish. The radius of the interface of two subregions r_{Kn} is determined from estimates of the lo-

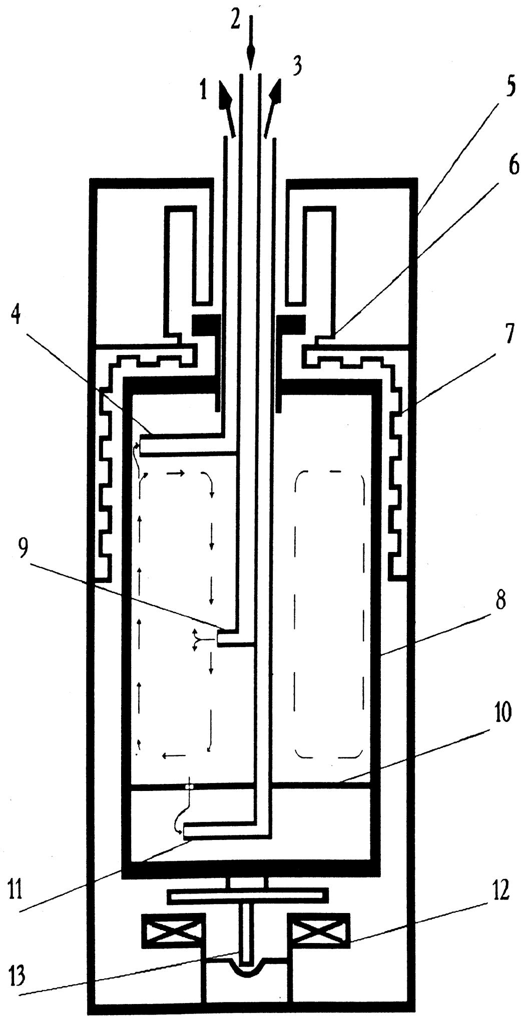


Figure 1. Schematic diagram of gas centrifuge. 1 – heavy fraction; 2 – feed; 3 – light fraction; 4 – top scoop; 5 – casing; 6 – magnet bearing; 7 – molecular pump; 8 – rotor; 9 – feed inlet; 10 – diaphragm; 11 – bottom scoop; 12 – stator; 13 – needle.

cal mean-free path for the process gas molecules corresponding to a transient from regime of the free-molecular flow to a continuous medium (11–13):

$$\alpha x_{Kn} \exp[\alpha(1 - x_{Kn}^2)] = A$$

where $x_{Kn} = r_{Kn} / r_0$; $\alpha = m\omega_0^2 r_0^2 / 2\mathrm{RT}$ is the gas compression ratio, and A is a constant.

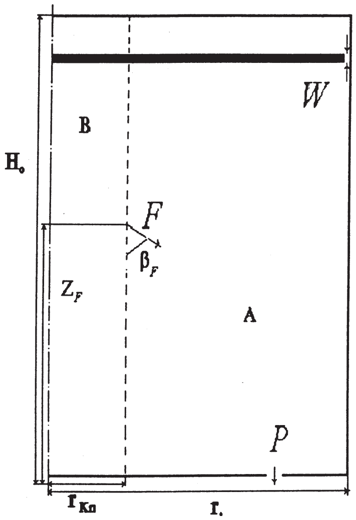


Figure 2. Schematic diagram of zones and external flows in gas centrifuge (see text).

Unlike the uranium isotope separation, where the linear speed of rotor rotation is around 600 m/s and the corresponding subregion B occupies ~3/4 of the radius, in a gas centrifuge for stable isotopes it can be considerably less or even absent. On the border of subregions, the feedflow to a gas centrifuge, F is simulated as in a real apparatus where it moves through a stationary gas-inlet pipe located along the axis of rotor rotation (see Fig. 1). In a typical case the feedflow in addition to a flow rate and its position on the z axis Z_F is characterized by a tilt angle β_F and a relative angular velocity ω_F (so-called "swelling"). Parameter Θ characterizes the division of a feedflow into two streams: of the light, P, and the heavy, W, fractions.

It is also considered that the flow and the diffusion in a subregion B do not affect the separation of the isotope mixture, which is completely defined by a circulation flow in a subregion A excited by a stationary upper scoop extending in a rapidly rotating gas. Within the chosen axisymmetric approximation it is impossible to simulate shock waves arising as a result of interaction between a rotating gas and the edge of the molecule extractor. Therefore, the stationary was modeled by a disk, which is transparent for all axial flows of a process gas. The degree of process gas slack by the scoop was modeled by either a disk system of sinks for the azimuthal component of momentum and energy or by a toroidal system of mass sinks at the boundary of the disk (14,15).

The boundary conditions corresponded to the standard ones, which were regularly used in the studies of internal flows of gas mixtures: 1) 'no-slip' condition on the rigid surfaces; 2) prescribed temperature on the boundaries of the computational region; 3) all parameters of external flows entering into the centrifuge rotor are given explicitly; 4) density, radial and axial components of the velocity vector are also given explicitly, and 5) azimuthal stagnation and temperature gradient in the outgoing extraction flow of the light fraction P are disregarded. The diffusion problem was based on the assumption of an absence of any diffusion flow at all boundaries of the computational region. The temperature boundary conditions on the baffle and outer wall were set from the known experimental data. The bottom chamber of a centrifuge with the lower scoop (to extract the light fraction) was, as a rule, not studied, since perturbations in this chamber do not reach the separating chamber and thus do not influence the separation process. The upper scoop (for extraction of the heavier fraction) can be situated directly in the separating chamber or in the special product chamber separated by its own baffle provided with two co-axial systems of holes to introduce the gas in and to withdraw it from the separating chamber.

System of Equations for Movement of Multicomponent Gas Mixture

The equations describing the movement of multicomponent gas mixture in a gas centrifuge can be either derived from the general principles of the field theory (16) or based on the kinetic theory of gases (17). The latter approach seems to be more preferable due to the knowledge of the main properties of distribution function by velocities and coordinates for molecules. This permits to derive not only the system of equations for the gas mixture but also the expressions to calculate so-called kinetic factors of transfer (coefficients of viscosity, diffusion and thermal conductivity). As the gas mixture under consideration is far from equilibrium, various properties of the distribution function can be used and, hence, different forms for a system of transfer equations can be obtained. For example, a remarkable deviation of a gas mixture can be a consequence of a large difference in molecule masses and appearance of strong gradients in a gas flow. This may lead to refusal of one partial and to the use of group distribution functions (18) requiring the description of a gas mixture either as a multicomponent or a multi-velocity and a multi-temperature gas.

Numerical theoretical studies of flow and diffusion in a gas centrifuge (15,19–21) have demonstrated that in a centrifugal field of force (existing in practice of isotope mixture separation) the conditions for occurrence of such kinds of non-equilibrium are usually absent. Even at a large difference in masses of component molecules the gas can be considered to be close to equilibrium (22). The exception, which must be made for the regions of shock-wave flows next to the edges of stationary scoops (23), will not be considered in the present review.

The equations of dynamics for an n-component viscous compressible gas mixture in a classical form are written as follows (17):

$$\frac{\partial \rho}{\partial \mathrm{t}} + \frac{\partial}{\partial \mathbf{r}} \cdot \rho \mathbf{V} = 0 \tag{1}$$

$$\frac{\partial \mathbf{V}}{\partial \mathrm{t}} + \left(\mathbf{V} \cdot \frac{\partial}{\partial \mathbf{r}}\right)\mathbf{V} = -\frac{1}{\rho}\left(\frac{\partial}{\partial \mathbf{r}} \cdot \dot{\Pi}\right) + \sum_{i=1}^{n} c_i \frac{m_i}{\overline{m}} \mathbf{F}_i \tag{2}$$

$$\rho \frac{\partial E}{\partial \mathrm{t}} + \rho \mathbf{V} \cdot \frac{\partial E}{\partial \mathbf{r}} = -\frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{q} - \left(\dot{\Pi} \cdot \frac{\partial}{\partial \mathbf{r}}\right) \cdot \mathbf{V} + \rho \sum_{i=1}^{n} c_i \frac{m_i}{\overline{m}} \mathbf{F}_i \mathbf{v}_i \tag{3}$$

$$\frac{\partial}{\partial \mathrm{t}}\left(\rho c_i \frac{m_i}{\overline{m}}\right) + \frac{\partial}{\partial \mathbf{r}} \cdot \left(\rho \mathbf{V} \frac{m_i}{\overline{m}} c_i + \mathbf{J}_i\right) = 0 \tag{4}$$

where $i = 1, \ldots, n$.

The first three equations describe the movement of the mixture as a whole. The fourth represents the continuity equation for i-th component. All together they allow calculation of the mole fractions of components c_i. To make this system complete it is necessary to add an equation of a gas mixture state:

$$\mathrm{P} = \rho \frac{\mathrm{RT}}{\overline{m}} \tag{5}$$

The expression for the pressure tensor in Eq. 3 is written as follows:

$$\dot{\Pi} = \mathrm{P}\dot{\mathrm{I}} - 2\eta \dot{\mathrm{S}} \tag{6}$$

where $\dot{\mathrm{I}}$ is the unit tensor, and $\dot{\mathrm{S}}$ is the tensor of deformations.

The following formula was proposed in (24) to calculate the viscosity of a multicomponent gas mixture:

$$\eta = \sum_{i=1}^{n} \frac{\eta_i c_i}{\sum_{k=1}^{n} G_{ik} c_k} \tag{7}$$

where

$$G_{ik} = \frac{[1 + (\eta_i/\eta_k)^{1/2} (m_k/m_i)^{1/4}]^2}{2^{3/2} (1 + m_i/m_k)^{1/2}} \tag{8}$$

and

$$\eta_i = 266.93 \cdot 10^{-7} \frac{\sqrt{m_i T}}{}, \quad \mathrm{g/cm \cdot s} \tag{9}$$

is the viscosity of a “pure” i-th component (17).

The diffusion flow density vector is defined by the following expression (25):

$$\mathbf{J}_i = \rho \frac{m_i}{\overline{m}} c_i \mathbf{v}_i = -\rho \frac{m_i}{\overline{m}} c_i \left(\sum_{j=1}^{n} \mathrm{D}_{\mathrm{ij}} \mathbf{d}_j + \mathrm{D}_{\mathrm{i}}^{\mathrm{T}} \frac{\partial \ln \mathrm{T}}{\partial \mathbf{r}}\right) \tag{10}$$

in which

$$\mathbf{d}_j = \frac{\partial\, c_j}{\partial \mathbf{r}} + c_j\left(1 - \frac{m_j}{\overline{m}}\right)\frac{\partial \ln \mathrm{P}}{\partial \mathbf{r}} - \frac{m_j}{\overline{m}}\frac{c_j}{\mathrm{P}}\left(\rho\mathbf{F}_j - \sum_{k=1}^{n}\rho\mathbf{F}_k\right) \tag{11}$$

and the generalized diffusion coefficients D_{ij} are determined from the solution of the following equations:

$$\begin{cases} \displaystyle\sum_{k=1}^{n}\frac{c_i c_k}{D_{\mathrm{ik}}}(\mathrm{D_{ij}} - \mathrm{D_{kj}}) = \delta_{ij} - \frac{m_i}{\overline{m}}c_i, \\ \displaystyle\sum_{i=1}^{n} m_i c_i \mathrm{D_{ij}} = 0 \end{cases} \tag{12}$$

Here δ_{ij} is the Kronecker delta, and the binary diffusion coefficients D_{ik} are calculated by the following formula (17):

$$D_{\mathrm{ik}} = 2.68\cdot 10^{-3}\,\frac{\mathrm{T}^{3/2}\sqrt{(m_i + m_k)/(2m_i m_k)}}{\mathrm{P}\sigma_{ik}^{2}\Omega_{ik}^{(1,1)*}}, \quad \mathrm{cm}^2/\mathrm{s} \tag{13}$$

where $\sigma_{ik} \approx 1/2(\sigma_i + \sigma_k)$ in Å, P in atmospheres.

The heat flux density vector in Eq. (3) is expressed as follows (17):

$$\mathbf{q} = -\nu\frac{\partial\,\mathrm{T}}{\partial \mathbf{r}} + \sum_{i=1}^{n}\mathbf{J}_i h_i + \mathrm{P}\sum_{\mathrm{i=1}}^{\mathrm{n}} k_{Ti}\mathbf{v}_i \tag{14}$$

where

$$h_i = \int_{T_0}^{T} c_{Pi}\,\mathrm{dT} + h_i^0 \tag{15}$$

is the enthalpy for i-th component per unit of mass, and k_{Ti} is the thermodiffusion ratio, defined by the following set of equations:

$$\begin{cases} \displaystyle\sum_{j=1}^{n}\mathrm{D_{ij}}k_{\mathrm{Ti}} = \mathrm{D_i^T} \\ \displaystyle\sum_{i=1}^{n} k_{\mathrm{Ti}} = 0 \end{cases} \tag{16}$$

The heat conductivity of the gas mixture can be calculated through the use of the following equation (26):

$$\nu = \sum_{i=1}^{n}\frac{\nu_i\,(0.115 + 0.354c_{Pi}/\mathrm{R})}{(1 + 1.065\,\Sigma_{k\neq i}^{n}\,\mathrm{G}'_{ik}c_k/c_i)} \tag{17}$$

where

$$G'_{\mathrm{ik}} = \frac{[1 + (\nu_i/\nu_k)^{1/2}\,(m_i/m_k)^{1/4}]^2}{2^{3/2}\,(1 + m_i/m_k)^{1/2}} \tag{18}$$

and

$$\nu_i = \frac{25(\pi\, m_i \mathrm{RT})^{1/2}}{32\pi\sigma_i\Omega_i^{(2,2)*}\,N_A}\cdot\frac{3\mathrm{R}}{2m_i} \tag{19}$$

is the heat conductivity of a pure *i*-th component of the mixture.

As seen from Eqs. (1–19), all parameters of the multicomponent mixture flow are complexly interconnected with each other and depend on the space distribution of component concentrations. The definition of flow parameters and distribution of concentrations is a sophisticated problem even with the use of modern numerical methods. However, referring to the isotope mixtures, the statement of the problem and its solution can be simplified through the use of a special approach known as "isotope approximation".

Isotope Approximation

The values of σ_i, $\Omega_{ij}^{(1,1)*}$, $\Omega_{\mathrm{i}}^{(2,2)*}$, c_{Pi}, h_i^0 for components of isotope mixtures are practically identical. Thus, in this case the differences between components in terms of the mass, impulse and energy transfer are determined by distinctions of their molecular masses. For the majority of isotope mixtures the correlation between molecular masses of components satisfies the following inequality:

$$|m_i - m_j| \ll \overline{m} \tag{20}$$

By using inequality (Eq. 20) one can show the following dependence of viscosity and heat conductivity on molecular masses:

$$\eta \approx \overline{\sqrt{m}}, \qquad \nu \approx \overline{\sqrt{m}} \tag{21}$$

Here an average size is defined as $\overline{\sqrt{m}} = \Sigma_{i=1}^{n} \sqrt{m_i} c_i$. Because in the modern ultracentrifuges the absolute changes of component concentrations in isotope mixtures are not as great, one can see from the ratio (Eq. 21) that the dependence of viscosity and heat conductivity on mole fractions and, hence, on coordinates, can be neglected.

Taking into account the following identities

$$\sum_{i=1}^{n} \mathbf{J}_i = \mathbf{0}, \quad \sum_{i=1}^{n} k_{Ti} = 0, \quad \mathbf{F}_i = \omega_0^2\,\mathbf{r}, \quad \mathbf{v}_i = \mathbf{U}_i - \mathbf{V}$$

it is possible to show the validity of the following equalities for the terms standing in the set of Eqs. (1–4)

$$\begin{aligned}
&\sum_{i=1}^{n} c_i m_i \mathbf{F}_i = \omega_0^2 \mathbf{r},\\
&\sum_{i=1}^{n} c_i m_i \mathbf{F}_i \mathbf{v}_i = \mathbf{0},\\
&\sum_{i=1}^{n} \mathbf{J}_i h_i \approx \mathbf{0},\\
&\sum_{i=1}^{n} k_{Ti} \mathbf{v}_i = \sum_{i=1}^{n} k_{Ti}\, \mathbf{U}_i \approx \mathbf{e} \sum_{i=1}^{n} \frac{k_{Ti}}{\sqrt{m_i}} \cong 0
\end{aligned} \qquad (22)$$

Thus, taking into account Eq. (22) one may consider that Eqs. (1–3), (5) (required to determine a gas-dynamic flow) can be solved quite independently from the system of diffusion Eq. (4). As the result, the complete solution of a problem for the mass transfer in multicomponent isotope mixtures in a gas centrifuge may be divided in two stages. First, the gas-dynamic problem is solved for an average-mass velocity, density, and temperature of the mixture as a whole. Then, the values found for these parameters are employed to solve the diffusion equations, written for each component of the mixture. This means that both the space distributions of mole fractions and the separative characteristics of a centrifuge will be determined. A similar approach is known as the "isotope approximation", which is widely used in the theoretical studies of uranium isotope separations. The statement and the solution methods for the gas dynamics and diffusion problems in the case of uranium isotopes are presently well-developed (19–21,27–32). For the multicomponent uranium and other non-uranium isotope mixtures the solution of a gas dynamics problem does not have any additional peculiarities. The statement and the solution methods of the multicomponent diffusion problem are at present not so well known. It calls for a special discussion, which is given in the following sections.

Statement of Diffusion Problem

The main problem in the solution of the diffusion Eqs. (4) consists in the dependence of the generalized diffusion coefficients (Eq. 12) on concentrations and also in the dependence of the diffusion flow densities (Eq. 10) on the thermodiffusion transfer. Because of this, and due to the dependence of the molar mass of the isotope mixture, $\overline{m}$, on the component mole fractions, the expression for a vector of diffusion flow densities (Eq. 10) appears to be nonlinear. In this case the solution of Eq. (4) is accompanied by significant mathematical difficulties.

For isotope mixtures separated in an ultracentrifuge the thermodiffusion effects in Eq. (10) can be neglected in comparison with the barodiffusion ones. To simplify system Eq. (4) it is possible to take advantage of some properties of the matrix for generalized diffusion coefficients. It is shown (33) that for isotope mixtures under condition Eq. (20) the system (Eq. 12) has the following approximate solution:

$$\mathrm{D}_{ij} = \frac{f_{ij}}{c_i} - \frac{3}{2}\,\frac{m_i}{\overline{m}(3m_i + \overline{m})}\sum_{k=1}^{n} f_{kj}\,(3m_k + \overline{m}) \tag{23}$$

when $(m_j - m_i) > 0$,

$$\mathrm{D}_{ij} = \frac{f_{ij}}{c_i} - \frac{5}{6}\,\frac{m_i}{\overline{m}(5m_i - \overline{m})}\sum_{k=1}^{n} f_{kj}\,(5m_k - \overline{m}) \tag{24}$$

when $(m_j - m_i) < 0$, and

$$f_{ij} = \left(\delta_{ij} - \frac{m_i c_i}{\overline{m}}\right)\overline{D}_i \tag{25}$$

where

$$\overline{D}_i = \left(\sum_{k=1}^{n}\frac{c_k}{D_{ik}}\right)^{-1} \tag{26}$$

It is more convenient to present parameter D_{ij} as follows:

$$\mathrm{D}_{ij} = A_{ij} + B_i \tag{27}$$

Equation (27) is a matrix of the generalized diffusion coefficients as a sum of two matrixes. In the general case, the elements of the first matrix differ from zero and have various values. In the second, all elements in each line are equal. The use of A_{ij} instead of D_{ij} in the expression for a vector of diffusion flow density (Eq. 10) appears not to change this vector (33). In other words, it is possible to make equivalent transformations in the matrix of the generalized diffusion coefficients consisting in addition to each line of the matrix of an arbitrary constant. The significance of this theorem (about equivalent transformations) for solution of a system of diffusion equations can be evidently demonstrated by taking more rigid restrictions (instead of Eq. 20) for the difference of molecular masses of the isotope mixture components:

$$\sqrt{|m_i - m_j|} \ll \sqrt{m} \tag{28}$$

In this case the solution for Eq. (12) can be reduced to the following simpler version:

$$\mathrm{D}_{ij} \cong \left(\delta_{ij} - \frac{m_i c_i}{\overline{m}}\right)\frac{\overline{D}_i}{c_i} \tag{29}$$

The additions in Eq. (27) will be defined as follows:

$$A_{ij} \cong \delta_{ij}\frac{\overline{D}_i}{c_i},\ \mathrm{B}_i = \frac{m_i}{\overline{m}}\overline{D}_i \tag{30}$$

Thus, due to the theorem on equivalent transformation one can use an equivalent diagonal matrix $[A_{ij}] = [(\overline{D}_i/c_i)\ \delta_{ij}]$ in approximation (Eq. 28) instead of a complete matrix for coefficients $[\mathrm{D}_{ij}]$ in Eq. (4). At the same time, in a more exact solution (Eqs. 23–25) the equivalent matrix $[\mathrm{A}_{ij}]$ is not strictly diagonal. Its non-diagonal elements differ from zero and are not equal to each other. It was shown for isotope mixtures (33) that predomination of diagonal elements over the non-diagonal ones consists of several orders of magnitude.

The opportunity to use the equivalent diffusion coefficients A_{ij} permits making a number of important conclusions on the convective-diffusion processes in isotope mixtures. Due to a strong diagonal predomination observed in matrix $[\mathrm{A}_{ij}]$, it is possible in many instances to use its approximate form (Eq. 30). Thus, in Eq. (10) the sum $\Sigma_{j=1}^{n}\ \mathrm{D}_{ij}\mathbf{d}_j$ transforms into only one term $\mathrm{A}_{ij}\,\mathbf{d}_i$ and the mutual influence of diffusion flows for all mixture components is displayed through an average molecular mass and coefficients (Eq. 26) $\overline{D}_i$. When the isotope mixtures are separated, the deviations of concentrations and consequently, the values of $\overline{m}$ and $\overline{D}_i$ are fairly small, which confirms a quasi-independence of the mixture component transfer.

The use of equivalent A_{ij} values instead of the generalized diffusion coefficients leads to the appearance of an imaginary contradiction. If coefficients D_{ij} are strictly symmetric (25), i.e. $\mathrm{D}_{ij} = \mathrm{D}_{ji}$ the symmetry for coefficients A_{ij} is not saved. It is not pursuant to the Onsager reciprocity relations. The contradiction can be removed by the use of the theorem proven by De Groot and Mazur (34). This theorem shows that if a linear correlation exists between forces and flows the phenomenological factors such as diffusion coefficients, in particular, are not unique. Hence, one can select any of those that satisfy the Onsager relations.

SOLUTION METHODS OF DIFFUSION PROBLEM

Substantial difficulties arise in solution of nonlinear diffusion Eq. (4) by using exact mathematical methods. Therefore, the solution must be found either by using analytical methods through the linearization of respective equations or by numerical methods using a more complete system of equations.

Analytical Methods

The main analytical method used for solution of a diffusion problem is based on the principle of an average mole fraction along the radius of a gas centrifuge rotor. The method was developed by Cohen for calculation of uranium isotope separation (35) and thereafter improved by Soubbaramyer for the case of variable circulating flow along the height of a centrifuge (36). The main point of the method consists in construction of the solution for an average value

$$\bar{c}(z) = \frac{1}{\pi r_0^2} \int_0^{\mathrm{r}_0} c(\mathrm{r}, z) 2\,\pi\,\mathrm{rdr} \tag{31}$$

instead of two-dimensional mole fraction distribution in cylindrical coordinates (r, z). Neglecting the radial convective transfer, the diffusion equation can be reduced to a one-dimensional ordinary differential one comprising the variable $\bar{c}(z)$. Using the solution of a gas-dynamic problem for the stream function Ψ we have found that this equation can be easily solved.

The method of an average mole fraction along the radius for multicomponent mixtures was applied for the first time in 1992 (37). The use of a mole fraction of each component in Eq. (31) allows for reducing the system (Eq. 4) to a system of ordinary differential equations, which are interconnected with each other through the nonlinear terms containing the molar mass of a gas mixture $\overline{m}$. To solve this nonlinear problem, it was proposed to use an iterative procedure with the specification at each iterative step the value of $\overline{m}$ using the mole fraction values found at the previous iterative step. Thus, the found system of the ordinary differential equations is solved. The iterations are repeated as long as the mole fractions do not cease to vary. The dependence of diffusion coefficients on concentrations in the solution was not taken into account.

Let us view now the main principles of this method for the case when the dependence of diffusion coefficients on mole fractions is taken into account (38). The system (Eq. 4) for a stationary case in cylindrical coordinates is written as follows:

$$\frac{1}{\mathrm{r}} \frac{\partial}{\partial \mathrm{r}} \left[\frac{m_i}{\overline{m}} c_i \mathrm{r} \rho \mathrm{V}_{\mathrm{r}} + \mathrm{r} \mathrm{J}_{\mathrm{ir}} \right] + \frac{\partial}{\partial \mathrm{z}} \left[\frac{m_i}{\overline{m}} c_i \rho \mathrm{V}_{\mathrm{z}} + \mathrm{J}_{\mathrm{iz}} \right] = 0 \tag{32}$$

Let us first not to limit our consideration by a special kind of a matrix of the equivalent diffusion coefficients $[\mathrm{A}_{ij}]$. As to solve Eq. (32) it is sufficient to find a solution for only $(n-1)$ equations, the mole fraction of the n-th component will be determined by the following expression:

$$c_n = 1 - \sum_{i=1}^{n} c_i \tag{33}$$

Using Eq. (27), let us introduce the following new quantities:

$$\overline{\mathrm{D}}_{ij}^{0*} = \rho c_i \left(\mathrm{D}_{ij} - \mathrm{D}_{in} \right) \tag{34}$$

The use of the second equation of Eq. (12) leads to the following form of Eq. (32):

$$\begin{aligned}&\frac{1}{\mathrm{r}}\frac{\partial}{\partial \mathrm{r}}\left\{\frac{m_i}{\overline{m}}-\left[\mathrm{rD}^0_{in}\frac{\partial c_i}{\partial \mathrm{r}}-\frac{m_i\omega_0^2\mathrm{r}_0^2}{\mathrm{RT}}c_i\left(\overline{\mathrm{D}}^{0*}_{ii}+\mathrm{D}^0_{in}+\sum_{\mathrm{j}\neq \mathrm{i}}^{\mathrm{n}-1}\overline{\mathrm{D}}^{0*}_{ii}\frac{c_jm_j}{c_im_i}\right)\right]\right\}\\&+\frac{\partial}{\partial \mathrm{z}}\left\{\frac{m_i}{\overline{m}}\left[c_i\rho \mathrm{V}_{\mathrm{z}}-\overline{\mathrm{D}}^{0*}_{ii}\frac{\partial c_i}{\partial \mathrm{z}}-\sum_{\mathrm{j}\neq \mathrm{i}}^{\mathrm{n}-1}\overline{\mathrm{D}}^{0*}_{ij}\frac{\partial c_j}{\partial \mathrm{z}}\right]\right\}=0\end{aligned} \tag{35}$$

where $\mathrm{D}^0_{in} = \rho \mathrm{D}_{in}$. The main properties of the isotope mixture flow in a gas centrifuge, namely the minority of $\rho \mathrm{V}_{\mathrm{r}}$ in comparison with J_{ir} and the minority of axial barodiffusional flow in comparison with the convective and mass diffusion flows are taken into account in Eq. (35). In addition, the following conditions are also included in Eq. (35): 1) the radial distribution of pressure is rather close to the equilibrium, i.e. $\mathrm{P}_i(\mathrm{r}) = \mathrm{P}_i(0)\exp[m_i\omega_0^2\mathrm{r}^2/(2\mathrm{RT})]$ and 2) the gradient of total pressure corresponds to gradients of partial pressures as follows: $\partial/\partial \mathrm{r} \ln \mathrm{P} = \partial/\partial \mathrm{r} \ln \mathrm{P}_i - \partial/\partial \mathrm{r} \ln c_i$. The boundary conditions for a system (Eq. 35) are defined as follows:

$$\begin{gathered}\left.\frac{\partial c_i}{\partial_r}\right|_{r=0}=0,\\ \left.\mathrm{D}^o_{in}\frac{\partial c_i}{\partial \mathrm{r}}-\frac{m_i\omega_0^2\mathrm{r}_0}{\mathrm{RT}}c_i\left[\overline{\mathrm{D}}^{0*}_{ii}+\mathrm{D}^o_{in}+\sum_{j\neq i}^{n-1}\overline{\mathrm{D}}^{0*}_{ij}\frac{c_jm_j}{c_im_i}\right]\right|_{\mathrm{r}=\mathrm{r}_0}=0,\\ \left.\int_0^{r_0}\frac{m_i}{\overline{m}}\left(\rho \mathrm{V}_zc_i-\overline{\mathrm{D}}^{0*}_{ii}\frac{\partial c_i}{\partial z}-\sum_{j\neq i}^{n-1}\overline{\mathrm{D}}^{0*}_{ij}\,\partial\frac{c_j}{\partial z}\right)2\pi\,\mathrm{rdr}+Wc_i^W\right|_{z=H_0}=0,\\ \left.\int_0^{r_0}\frac{m_i}{\overline{m}}\left(\rho \mathrm{V}_zc_i-\overline{\mathrm{D}}^{0*}_{ii}\frac{\partial c_i}{\partial z}-\sum_{j\neq i}^{n-1}\overline{\mathrm{D}}^{0*}_{ij}\frac{\partial c_j}{\partial z}\right)2\pi\,\mathrm{rdr}-Pc_i^P\right|_{\mathrm{z}=0}=0\end{gathered} \tag{36}$$

where W is the flow of a heavy fraction from a gas centrifuge, and P is the light fraction flow, c_i^W and c_i^P are the mole fractions in W and P flows. To make the solution procedure for Eqs. (35) and (36) reasonably simple it is necessary to consider the features of terms included in these equations. First, as the multiplication by $\rho \mathrm{D}_{ij}$ does not depend on the density by virtue of Eq. (12), the coefficients $\overline{\mathrm{D}}^{0*}_{ij}$ and D^0_{in} must also not depend on the density and will smoothly vary along the radius. Secondly, in the isotope separation it is possible to consider the relative changes of mixture molar mass to be small. Therefore, as the first approximation of solution for Eqs. (35) and (36) one can consider the linear boundary problem. All coefficients giving their contribution in nonlinearity can be presented as constant values calculated for definite mole fractions. A more exact solution of this nonlinear problem can be found through the use of the iterative procedure described above with correction at each iterative step of all coefficients depending on mole fractions.

Using now the mean procedure in Eq. (31) for the mole fraction of each component and introducing besides the new independent variables $\xi = 1 - (\mathrm{r}/\mathrm{r}_{\mathrm{o}})^2$ and $z = z/r_o$ to transform Eq. (35) to the system of ordinary differential equations relative to averaged mole fractions, one obtains:

$$\Lambda\left(\left\{\frac{\overline{m}}{m_i}\right\}\widetilde{\widetilde{c_i}} - \overline{c}_i\right) = 2\varepsilon_{0i}Q_1\,(\overline{z})\overline{c}_i + (K_{1i} + K_{2i}Q_2\,(\overline{z}))\,\frac{\mathrm{d}\overline{c}_i}{\mathrm{d}z} - \pi\,\mathrm{r}_0\,\{E_i\} \quad (37)$$

where

$$\Lambda = \begin{cases} -P, 0 < \overline{z} < \overline{z}^F \\ W, \overline{z}^F < \overline{z} < \overline{z}_0 \end{cases}; \quad \widetilde{\widetilde{c_i}} = \begin{cases} c_i^P, 0 < \overline{z} < \overline{z}^F \\ c_i^W, \overline{z}^F < \overline{z} < \overline{z}_0 \end{cases}$$

and $\overline{z}^F$ is the feed-point for a feed-flow F entering a centrifuge with mole fractions c_i^F. The coefficients in Eq. (37) are expressed as follows:

$$\begin{aligned} &Q_1(\overline{z}) = \int_0^1 \psi\,\mathrm{d}\,\xi, \quad Q_2(\overline{z}) = \int_0^1 \psi^2\,\frac{\mathrm{d}\,\xi}{1-\xi}, \\ &\psi = -\frac{r_0^2}{2}\int_1^\xi \rho\,\mathrm{V}z\mathrm{d}\xi, \quad \varepsilon_{0i} = -\pi\alpha_i\{A_i\}, \quad \alpha_i = \frac{m_i\omega_0^2\mathrm{r}^2{}_0}{2\,\mathrm{RT}}, \\ &\{A_i\} = 1 + \frac{\{\overline{\mathrm{D}}_{ii}^{0*}\}}{\{\mathrm{D}_{in}^o\}} + \sum_{j\neq i}^{n-1}\frac{\{\overline{\mathrm{D}}_{ij}^{0*}\}}{\{\mathrm{D}_{in}^o\}}\cdot\left\{\frac{c_j}{c_i}\right\}\cdot\frac{m_j}{m_i}, \\ &\{E_i\} = \sum_{j\neq i}^{n-1}\{\overline{\mathrm{D}}_{ij}^{0*}\}\left\{\frac{\mathrm{d}\overline{c}_j}{\mathrm{d}\overline{z}}\right\} \\ &K_{1i} = \pi\,\mathrm{r}_0\,\{\overline{\mathrm{D}}_{ii}^{0*}\} \quad K_{2i} = -\pi/\mathrm{r}_0\,\{\mathrm{D}_{in}^o\} \end{aligned} \quad (38)$$

The terms in brackets { } depend on the mole fractions and are to be specified in each new iteration. The general soluti1on of the problem is given as follows:

$$\overline{c}_i(\overline{z}) = \exp(-\varphi_i(\overline{z}_1))\left[\overline{c}_i(\overline{z}_1) + \int_{\overline{z}_1}^{\overline{z}}\frac{\Lambda\left\{\frac{\overline{m}}{m_i}\right\}\tilde{\tilde{c}} + \pi\,\mathrm{r}_0\{E_i\}}{}\exp(\varphi_i(\overline{z}'))\mathrm{d}\,\overline{z}'\right] \quad (39)$$

where

$$\varphi_i(\overline{z}) = \int_{\overline{z}_1}^{\overline{z}}\frac{\Lambda + 2\varepsilon_{0i}Q_1(\overline{z}')}{K_{1i} + K_{2i}Q_2(\overline{z}')}\,\mathrm{d}\,\overline{z}' \quad (40)$$

Coefficients $\overline{\mathrm{D}}_{ij}^{0*}$ can be calculated by Eqs. (23–25) or by means of the diagonal matrix (Eq. 30) that is to be much faster. Application of the diagonal matrix in Eq. (30) obtains the stationary solution with the relative mole fraction fluctuations of the order of 10^{-7} in 5–7 iterations. The high accuracy of the results is due to the use of the exact values of generalized diffusion coefficients obtained from the solution of Eq. (12). For example, the accuracy of calculations of separative efficiency for uranium isotopes in UF_6, sulfur isotopes in SF_6, and carbon and oxygen isotopes in CO_2 was 10^{-3}%, 10^{-1}% and about 1%, respectively.

Numerical Methods

The numerical calculations of Eq. (4) are based, as a rule, on the use of two types of finite-difference techniques.

Finite-Difference Technique for Steady Convective and Diffusion Flows

The use of this technique supposes that derivatives by time standing in Eq. (4) are equal to zero, i.e., the convective and diffusion flows of the mixture components are steady. The various modifications of this technique are based on application of actually one finite-difference procedure, which has been developed for the binary mixture of uranium isotopes (19). According to this procedure the flow region is covered by a non-uniform grid with the beforehand known set of gas dynamic parameters on it. Eq. (4) is substituted by finite-difference analogs forming therefore, a system of nonlinear algebraic equations with block three-diagonal matrix of coefficients. The solution of this system is determined by using a modified iterative Newton procedure. A special procedure based on the Jacoby matrix perturbation can be used to make the steady convergence of the solution (15). The practice of such calculations has shown a relatively good convergence of this technique for both binary (13) and multicomponent mixtures (14).

However, this approach has one essential drawback, which is connected with the fact that the solution of the system of nonlinear algebraic equations can be obtained at least though a single conversion of the Jacoby matrix for any initial approximation. It requires a big computer memory and a long time of routine computing work, which is proportional to $(n - 1)^2 N_z \cdot N_r^2$ where N_r and N_z are the number of grid steps in the radial and axial directions, respectively. If the initial approximation appears to be rather far from the final solution (e.g., this situation frequently takes place in practice), the conversion of the Jacoby matrix should be repeated. All this imposes the restrictions on the number of the grid steps (and, hence, on the accuracy of solution) and the number of mixture components, n. For example, for $n = 5$ the volume of computation work on solution of the diffusion problem becomes close to that for the gas-dynamic problem and even exceeds it by following the square law.

Finite-Difference Technique for Unsteady Flows

The use of this technique suggests that in Eq. (4) the gas dynamic parameters are steady while the diffusion flows at the initial moment of a time are completely unsteady. The convective flows of components are also unsteady because of the unsteady values of mole fractions. More economical numerical methods (21,39) (in comparison with those described above) were used to solve the problem of this kind. These methods include the factored finite-difference schemes or so-called splitting techniques. A comprehensive description of a wide spectrum of these methods can be found in Refs. 40 and 41. In brief, the gist of the splitting method applied to the solution of Eq. (4) consists in the following: the conservation Eq. (4) is written as follows:

$$\frac{\partial \mathbf{c}}{\partial \mathrm{t}} + \frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{F}(\mathbf{c}) = \frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{G}(\mathbf{c}) \tag{41}$$

where **F** and **G** are the vectors of convective and diffusion flows, respectively, for each mixture component, and **c** is the vector of mixture mole fraction. The following scheme is used for numerical integration by time (21):

$$\begin{aligned} \frac{\mathbf{c}^{k+1} - \mathbf{c}^{k}}{\tau} &= \frac{\theta}{1+\zeta} \frac{\partial \mathbf{c}^{k+1}}{\partial \mathrm{t}} + \frac{1-\theta}{1+\zeta} \frac{\partial \mathbf{c}^{k}}{\partial \mathrm{t}} + \frac{\zeta}{1+\zeta} \frac{\mathbf{c}^{k} - \mathbf{c}^{k-1}}{\tau} \\ &+ \mathrm{O}\left[\left(\theta - \frac{1}{2} - \zeta\right)\tau^{2} + \tau^{3}\right] \end{aligned} \tag{42}$$

where τ is the step of integration by time, θ and ζ are the parameters, and k is the number of iterations (number of steps by time). After decomposition of convective and diffusion flows in the Taylor series and using the deviations of mole fractions $\Delta \mathbf{c}^{k} = \mathbf{c}^{k+1} - \mathbf{c}^{k}$ from Eqs. (41) and (42), one obtains the following matrix equation for $\Delta \mathbf{c}^{k}$:

$$\left\{ \mathbf{I} + \frac{\theta\tau}{1+\zeta} \sum_{l=1}^{2} \Omega_{l}^{1} \right\} \Delta \mathbf{c}^{k} = \frac{\tau}{1+\zeta} \sum_{l=1}^{2} \Omega_{l}^{2} + \frac{\zeta}{1+\zeta} \Delta \mathbf{c}^{k-1} \tag{43}$$

where **I** is a unit matrix, $l = 1, 2$ denotes the numbers of space coordinates (r and z), and Ω_{l}^{1}, Ω_{l}^{2} are the matrix differential operators for each space variable. Applying the approximation procedure with the second order of accuracy by time

$$\left\{ \mathbf{I} + \frac{\theta\tau}{1+\zeta} \sum_{l=1}^{2} \Omega_{l}^{1} \right\} = \prod_{l=1}^{2} \left\{ \mathbf{I} + \frac{\theta\tau}{1+\zeta} \Omega_{l}^{1} \right\} \tag{44}$$

to Eq. (43) allows one to construct the two-step search scheme for $\Delta \mathbf{c}^{k}$ with solution at each step of the following one-dimensional matrix equation:

$$\begin{aligned} &\left\{ \mathbf{I} + \frac{\theta\tau}{1+\zeta} \Omega_{1}^{1} \right\} \Delta \mathbf{c}^{*} = \frac{\tau}{1+\zeta} \sum_{l=1}^{2} \Omega_{l}^{2} + \frac{\zeta}{1+\zeta} \Delta \mathbf{c}^{k-1}, \\ &\left\{ \mathbf{I} + \frac{\theta\tau}{1+\zeta} \Omega_{2}^{1} \right\} \Delta \mathbf{c}^{\mathrm{k}} = \Delta \mathbf{c}^{*}, \\ &\mathbf{c}^{k+1} = \mathbf{c}^{k} + \Delta \mathbf{c}^{k} \end{aligned} \tag{45}$$

The scheme in Eq. (45) splits the solution consistently for each space variable. In the course of solution of Eq. (4) the operators Ω_{l}^{1}, Ω_{l}^{2} represent $(n-1)(n-1)$ rank matrices. Acting on a massive of variables in the three-point finite-difference approximation for the first and the second space derivatives, they lead to a system of algebraic equations with block tree-diagonal matrix. To solve this system at each space point it is necessary to converse the matrixes with dimension $(n - 1)$ $(n - 1)$. This requires the volume of calculations of the order of $(n - 1)^2$. The common volume of computational work at each iteration can be estimated to equal $(n - 1)^2\ N_z{\cdot}N_r$. This permits the square law dependence and restrictions on the quantity of grid steps in the radial direction inherent to the first finite-difference technique to be overcome.

The square law dependence for the volume of calculations on the number of mixture components is overcome with through the simultaneous application of two approaches: the use of the diagonal form of matrix of diffusion coefficients (Eq. 30) and the use of molar mass and diffusion coefficients calculated by the values of mole fraction found at the previous iterative step. In this case the matrices Ω_l^1, Ω_l^2 are diagonal and the solution of the problem breaks up in solution (Eq. 45) by the two-step scheme for $(n - 1)$ independent scalar equations for $\Delta\mathbf{c}^k$. In this case the conversions of matrixes are not required; the quantity of calculations at each iteration is proportional to $(n - 1)N_z{\cdot}N_r$, which makes the calculations for isotope mixtures with arbitrary number of components more effective.

To make use of this numerical technique, the compulsory requirement for the operators Ω_l^1, Ω_l^2 on the maximum stability for the numerical scheme either at each intermediate or external iterative step by time is imposed. The stability is provided by a kind of finite-difference approximation of the differential operators and special factors θ and ζ along with the addition to Eq. (41) of special stabilizing terms tending to zero when approaching the stationary solution (40).

COMPUTER EXPERIMENT IN STUDIES ON SEPARATION OF MULTICOMPONENT MIXTURE

The numerical techniques are the most universal and the most attractive when either determining the separative characteristics of a gas centrifuge with sufficient for practice accuracy or optimizing its operation parameters. The analytical methods do not allow for accomplishing these tasks since they do not account for nonlinear phenomena and have a limited opportunity to take into account the real design features of a gas centrifuge.

A regular study of separative power of real gas centrifuges by numerical techniques in many cases includes a computer experiment. The computing experiment is more preferable compared with the real one because it is far cheaper and more convenient. In addition, it permits studying the characteristics of centrifuges under design and making calculations for hypothetical isotope mixtures. Both the statement and realization of a computing experiment require the solution of a number of problems, which will be considered below.

Parameters for Adjustment of Numerical Model

A physico-mathematical model for a gas centrifuge involves two parameters that cannot be set without a preliminary adjustment: 1) the gas content (mass of gas) in the centrifuge rotor, and 2) the parameter simulating the interaction of a rotating gas with the stationary scoop. The gas content defines the value of density ρ_W near the side-wall and the Eckman number $E = \eta/(\rho_W \omega_o r_o^2)$, which is the criterion of similarity when describing the relationship between viscous and inertial forces and a structure of the secondary flows.

Strictly speaking, the value of a gas content is perfectly defined by the feed-flow and a centrifuge design, particularly the extractor of a light fraction and the baffle shielding it from the working chamber. However, these elements of a real construction are not axisymmetric and can be defined just approximately within the axisymmetric physico-mathematical model. In this context, the problem of selection of gas flow parameters that maximally correspond to the real gas content value is particularly important.

In practice, a gas content value is determined by several methods. For example, it can be evaluated by measuring the gas weight in a centrifuge rotor or by the pressure value on a certain rotor radius. Due to a very high speed of rotor rotation, these kinds of experiments are very difficult and their accuracy is low. According to mathematical and numerical models, the value of gas content is set by the Eckman number and from some boundary conditions. In some computer experiments, the extractor of a light fraction can be disregarded. In this case, the boundary conditions simulating the product flow *P* are set for a baffle. This model is physically justified because a scoop for a light fraction brings much less disturbance in the rotating gas than the extractor for heavier fractions. The disturbances from the lower chamber practically do not penetrate into the working volume. To replace a ring system of holes on the lower baffle into a ring gap and to make an obvious determination for ρV_z in the mathematical model, the value of light fraction flow is established as follows:

$$P = \int_{r_1}^{r_2} 2\pi \, r \, \rho \, V_z dr \tag{46}$$

The gas content is determined by the density value in Eq. (46). The modeling of extractors is a very important problem as the stationary scoops are the main reason for the excitation of axial secondary flows inside the centrifuge rotor that defines the multiplication of a radial separation effect in the axial direction. In the computer experiment, the scoop parameters are to be included in the model. Let us consider in detail the basic properties of the scoop model described, for example, by Kai and Borisevich et al. (14,15).

Because of a fast gas spin in the centrifuge, the disturbances produced by a stationary gas inlet are very quickly distributed in the azimuthal direction and can be considered as axisymmetric ones. The sink-sources terms defining the density of mass sink, the impulse sink, and sinks and sources of energy should be added to Eqs. (1–3) in the cylindrical axisymmetric area of rotating gas and scoop contact. The mass sink exactly corresponding to the value of the evacuating flow is located in the toroidal volume shaped in rotation of the open scoop edge. The azimuthal component of the volume density for impulse sinks is determined by the resistance force of a stationary scoop to the upward flow of the rotating gas. Due to a supersonic speed of rotation, the viscous effects can be neglected and only wave-drag is to be taken into account. Thus, the sink for azimuthal impulse component I_φ can be determined from the following relation:

$$\int \mathrm{I}_\varphi \, 2\pi \, \mathrm{rdrdz} = \frac{\chi}{2} \int \rho \, \mathrm{V}_\varphi^2 \, \mathrm{drdz} \tag{47}$$

where the integration is carried out by the cylindrical area of the scoop location. The expression χ determines the value of the wave-drag factor, which the calculation of is a difficult problem. An energy sink in the toroidal volume is caused by a mass sink, and an energy source in the cylindrical volume is produced by a resistance force. By neglecting the thermal conductivity of scoop material the energy sources are such that $V_\varphi \, I_\varphi$.

The factor value for a wave-drag χ and the gas content Γ will determine the flow in a gas centrifuge and are the adjustment parameters, which should be defined from the correlation condition for calculated and experimentally determined separating characteristics. For a more detailed description of the adjustment technique for a concrete type of a gas centrifuge it is necessary to define the separative characteristics that will be used for comparison of theoretical and experimental data.

Separative Characteristics of Gas Centrifuge for Separation of Multicomponent Isotope Mixtures

The most important and commonly used criteria of the efficiency of a gas centrifuge for separation of binary uranium isotope mixture are the value function, V, and the separative power, δU, (35), which are defined as follows:

$$V = (2c - 1)\ln\left(\frac{c}{1 - c}\right) \tag{48}$$

$$\delta U = PV(c^P) + WV(c^W) - FV(c^F) \tag{49}$$

They allow for numerical estimation of the separative work necessary to change the mixture mole fraction.

There is no uniform universal concept on the criterion describing the separation process in the case of multicomponent isotope mixtures. A standard approach is based on the use of separation factors, which are written as follows:

$$\alpha_{ij} = \frac{c_i^P}{c_j^P} / \frac{c_i^F}{c_j^F}; \quad \beta_{ij} = \frac{c_i^F}{c_j^F} / \frac{c_i^W}{c_j^W}; \quad \gamma_{ij} = \alpha_{ij}\beta_{ij} \tag{50}$$

The knowledge of their values allows for calculating the cascades for multicomponent isotope separation but it is not sufficient even to estimate separative power. This kind of estimation is presently carried out by using two performance characteristics defined by Eqs. (48) and (49) or their equivalents, which are considered below.

In theoretical study of separation of the third component of uranium isotope mixture, Kai (14) suggested using the following analogs of the above values to calculate the value function and the separative power for any couple of components with numbers i and j:

$$V(c_i, c_j) = (c_i - c_j)\ln\left(\frac{c_i}{c_j}\right) \tag{51}$$

$$\delta U_{ij} = PV(c_i^P, c_j^P) + WV(c_i^W, c_j^W) - FV(c_i^F, c_j^F) \tag{52}$$

In this case the value function of Eq. (51) characterizes the degree of mixture order in relation to a chosen pair of components. The δU_{ij} value is proportional to the work necessary to maintain changes of mole fractions of these components in P and W flows compared with the feed-flow by a separative process. It is supposed that the mixture is not completely in order and its "cost" (value) is minimal in the case when component mole fractions equal each other, i.e., $c_i = c_j$. The separative power equals zero when no simultaneous changes of difference and relative concentration values of components are observed in flows P and W compared with a feed-flow. This situation may theoretically exist for multicomponent mixtures of more than three components for some component couples. The zero separative work can be found if the pair of components with the numbers i and j is enriched in a flow P and simultaneously depleted in a flow W or vice versa. On the other hand, even in this case a gas centrifuge produces the separative work to fractionate these two and other mixture components. To expel the situations characterized by zero separative work, the following formula has been proposed to calculate a complete separative power as a linear combination of respective partial δU_{ij} values:

$$\delta U_T = \sum_{\substack{i,j=1 \\ i<j}}^{n} \frac{2C_{ij}}{n(n-1)} \frac{\delta U_{ij}}{c_i^F + c_j^F} \tag{53}$$

Here C_{ij} is an arbitrary constant independent from the mole fraction values. A sum $(c_i^F + c_j^F)$ in the denominator of Eq. (53) is used to maximally reduce the dependence of δU_T on mole fractions of the initial mixture. If Eq. (52) is written through the separation factors in Eq. (50) this dependence becomes more obvious:

$$\delta U_{ij} = F\left[\frac{\alpha_{ij}(\beta_{ij}-1)\ln\alpha_{ij}-(\alpha_{ij}-1)\ln\beta_{ij}}{\gamma_{ij}-1}c_i^F\right.$$
$$\left.+\frac{\beta_{ij}(\alpha_{ij}-1)\ln\beta_{ij}-(\beta_{ij}-1)\ln\alpha_{ij}}{\gamma_{ij}-1}c_j^F\right] \tag{54}$$

The partial separative powers in Eq. (53) are strictly physically justified. However, probably this generalized measure of a separative work with weight factors of $C_{ij}/(c_i^F + c_j^F)$ requires an additional physical justification. The most difficult contradictions are caused by the formal use of the term $(c_i^F + c_j^F)$.

Another approach to calculate the separative power of a gas centrifuge for multicomponent isotope separation is used by Filippov and Sosnin and Aisen et al. (42,43). The separative power is set as an approximate value by an analogy to separation of a binary mixture assuming minor changes of component mole fractions within one centrifuge (35):

$$\delta U_T = \frac{\Theta(1-\Theta)}{2}F\frac{\varepsilon_0^2}{2} \tag{55}$$

The ε_0^2 value can be given either as a mean value of the effective enrichment factors for all mixture components taken in the second power:

$$\varepsilon_0^2 = \frac{1}{n^2}\left[\sum_{i=1}^{n}\frac{c_i^P - c_i^W}{c_i^F\sum_{j=1}^{n}(m_j-m_i)c_j^F}\right]^2 \tag{56}$$

or as an average value of all effective enrichment factors in the second power:

$$\varepsilon_0^2 = \frac{1}{n^2}\sum_{i=1}^{n}\frac{(c_i^P - c_i^W)^2}{(c_i^F)^2\left[\sum_{j=1}^{n}(m_j-m_i)c_j^F\right]^2} \tag{57}$$

The quantities in Eqs. (53) and (55) formally differ from each other but both represent the same characteristics—the generalized separative power of a gas centrifuge that accounts for the work spent to simultaneously separate all components of the isotope mixture. In this sense both characteristics can be used as the optimization criteria in computer experiments.

Computer Experiment Techniques

The adjustment of the numerical technique by the working parameters of a gas centrifuge can be carried out as follows. The numerical calculations of the

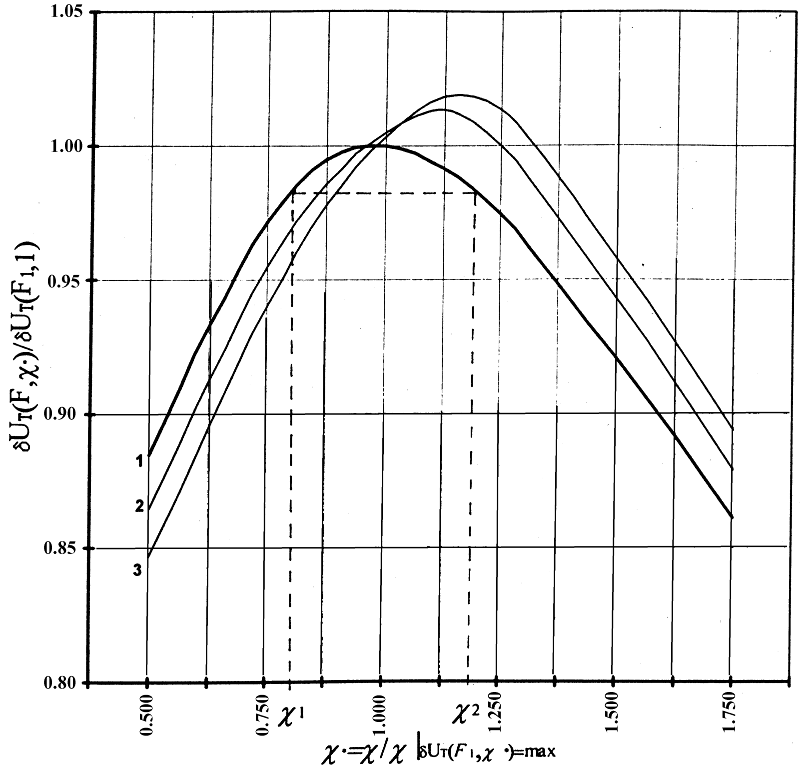


Figure 3. Relative separative power versus relative wave-drag coefficient for different feed-flows: $F_1 > F_2 > F_3$.

variation of separative power δU_T with the wave-drag coefficient are shown in Fig. 3 (see curve 1). This dependence allows for determination of two values of the wave-drag coefficient, χ_1 and χ_2, that satisfy experimental values of separative power calculated through concentrations c_i^P, c_i^W, and c_i^F. The χ_1 value corresponds to a regime with insufficient circulation flow to achieve the maximum value of a separative power (subcirculation) whereas χ_2 relates to the regime of the redundant circulation (hypercirculation) characterized by a lower value of separative power. Hence, to determine the true value of wave-drag coefficient one needs to have additional experimental data. These data must correspond to the dependence δU_T on some working parameter whose value does not influence the wave-drag coefficient. Besides this parameter must be measurable with a high accuracy to make it of use in the numerical model. The feed-flow F appears to be a suitable parameter to study the experimental dependence $\delta U_T(F)$. An increase of

F value leads to a growth of the maximum value $\delta U_{\mathrm{T}}^{\max}(F)$ and to the shift of this dependence to higher wave-drag coefficient values (see Fig. 3). Hence, the decreasing $\delta U_T(F)$ dependence will correspond to the subcirculation regime whereas with the hypercirculation one will be characterized by a growing $\delta U_T(F)$ dependence with a maximum corresponding to transition F values at which this regime transfers to another one. To achieve a good concurrence between experimental and theoretical dependencies one is required to carry out the correction of experimental results by the gas content value Γ. The typical dependence of separative power on Γ is shown in Fig. 4. The growth of circulation intensity is stipulated by an increase of either wave-drag coefficient or gas content. Therefore, at higher Γ values, the optimal intensity of circulating flow can be achieved at a smaller value of wave-drag factor. As a result, by varying Γ and χ parameters one can achieve a good correlation between experimental and theoretically determined

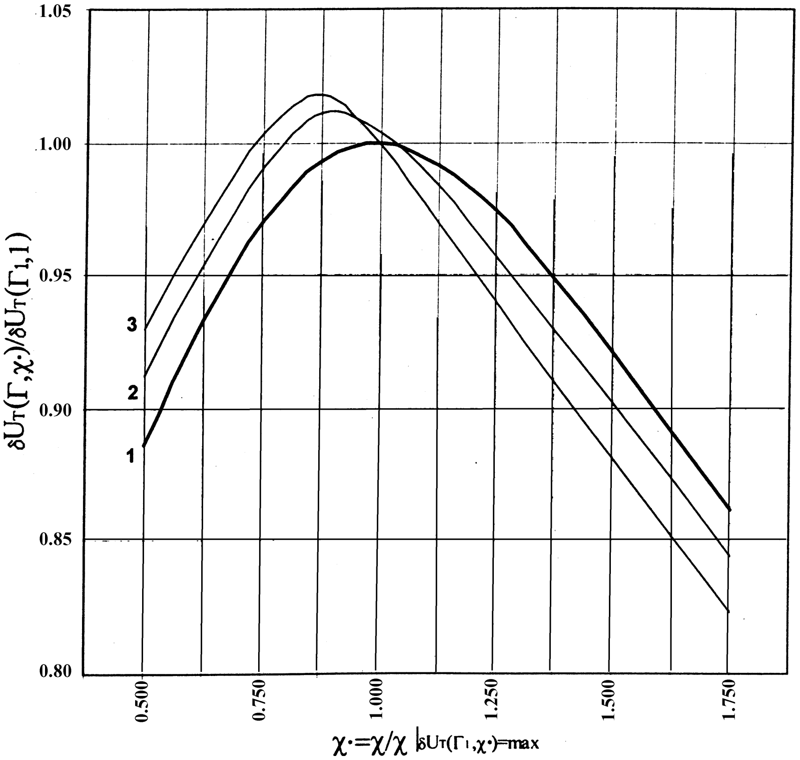


Figure 4. Relative separative power versus relative wave-drag coefficient for different gas contents: $\Gamma_1 > \Gamma_2 > \Gamma_3$.

δU_T values. The basic Γ and χ values selected in the above fashion can be used in a wide spectrum of computer experiments by studying the influence of controlled parameters on separative characteristics of a gas centrifuge.

Due to the complexity and high cost of measurements, the available experimental data can be found only for a limited number of process gases. At the same time, computer experiments can be carried out for an unlimited number of gases. However, in this case the dependencies reflecting the influence of process gas parameters on Γ and χ values are also required. The χ values for an arbitrary process gas can be calculated by using the Ackeret formula for a thin profile streamlined by a supersonic flow (44):

$$\chi = a/\sqrt{M^2 - 1} \tag{58}$$

where $M = \sqrt{\dfrac{2\alpha}{\gamma}}$ is the Mach number, $\alpha = \overline{m}(\omega_0 r_0)^2/(2\ \mathrm{RT})$ is a constant, and $\gamma = c_p/c_v$ is the ratio of the specific heats.

For any isotope mixture, χ_1 coefficient can be calculated from the known χ value by using the following expression:

$$\chi_1 = \chi\sqrt{M^2 - 1}/\sqrt{M_1^2 - 1} \tag{59}$$

Equation (59) accounts for the possibility of varying both the angular velocity, ω_o, and the rotor radius, r_o. The recalculation of Γ values for known temperature and pressure near the side wall of a rotor for a new process gas can be carried out by using the following equation:

$$\Gamma_1 = \Gamma \frac{\overline{m}_1}{\overline{m}} \frac{1 - \exp\left(-\dfrac{\overline{m}_1\,\omega_0^2\,\mathrm{r}_0^2}{2\ \mathrm{RT}}\right)}{} \tag{60}$$

Special attention must be paid to the fact that Γ value is used as an adjustment parameter for experimental data only with the purpose of maximally precise determination χ coefficient. When χ value is determined the gas content should be considered as one of the parameters for optimization of a gas centrifuge.

Figure 5 illustrates the results of adjustment of the numerical model for separation of sulfur isotopes in SF_6 by the experimental data adapted from Aisen et al. (43). The parameters obtained in this computer experiment have been used to simulate separation of tellurium isotopes in TeF_6 in the same centrifuge. The comparison of the results shows that in separation of sulfur isotopes, the scoop parameters correspond to the subcirculation regime, whereas in tellurium isotopes separation, the hypercirculation regime is realized due to an increase of gas content value associated with a higher molecular mass of TeF_6.

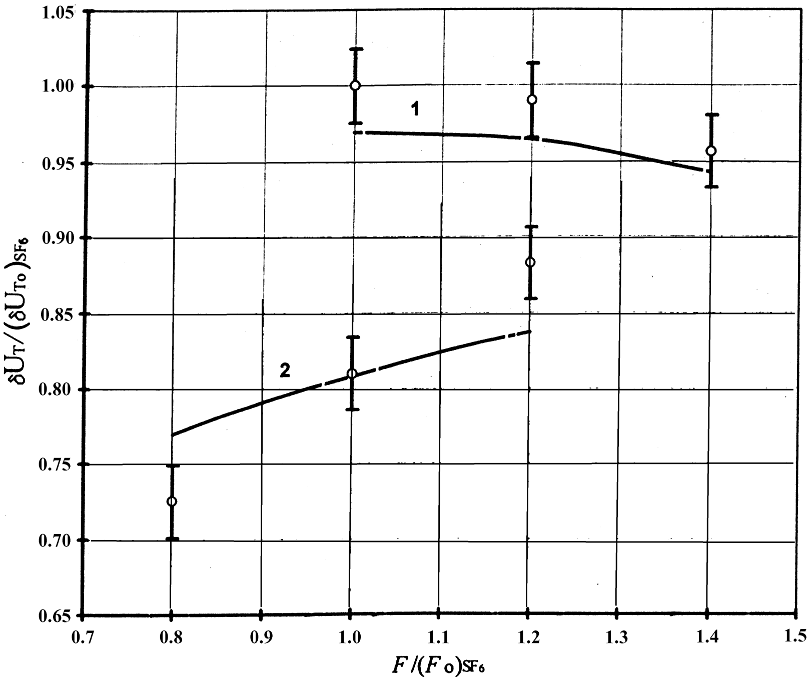


Figure 5. Theoretical (curves) and experimental (points) dependencies of relative separative power on relative feed-flow for SF_6 (1) and TeF_6 (2) isotopic mixtures.

RESULTS OF STUDIES ON SEPARATION OF MULTICOMPONENT ISOTOPE MIXTURES

Three groups of results are of the most practical interest in separation of multicomponent isotope mixtures:

1. dependencies describing the influence of centrifuge working (mainly, regulating) parameters on its separative characteristics;
2. results obtained by the analysis of the influence of separating mixture composition (mole fractions of mixture component and molecular masses of process gases) on separative power, and
3. dependencies characterizing the efficiency of gas centrifuge of pre-selected type for separation of a wide range of isotope mixtures.

Influence of Gas Centrifuge Parameters

The most important parameters of a centrifuge design are those connected with the way of introduction of a feed-flow (the feed-flow F, the cut Θ, axial feed-point Z_F , the relative angular velocity $\overline{\omega}_F$, and the slope angle to the axis of rotation β_F) and also parameters determining the circulating flow (rotating gas slack by a scoop, the temperature gradients on rotor surfaces and the gas content Γ).

The influence of the above parameters has been considered in the previous section describing the methodology of computer experiment. Thus, it has been shown that an increase of rotating gas slack by a scoop leads to a growth of the separative power up to the maximum followed by its decrease at an excessive value of circulating flow (see Figs. 3 and 4). The rotating gas slack is proportional both to the wave-drag factor and the geometrical sizes of a scoop (product of its diameter and length). When the slack is insufficient, an increase of feed-flow results in reduction of separative power and under the same conditions, an increase of gas content leads to its increase. At excessive gas slack, the action of both of the above effects becomes opposite.

The study of different ways of gas introduction in the centrifuge through a stationary central column assembly is important because this assembly can be

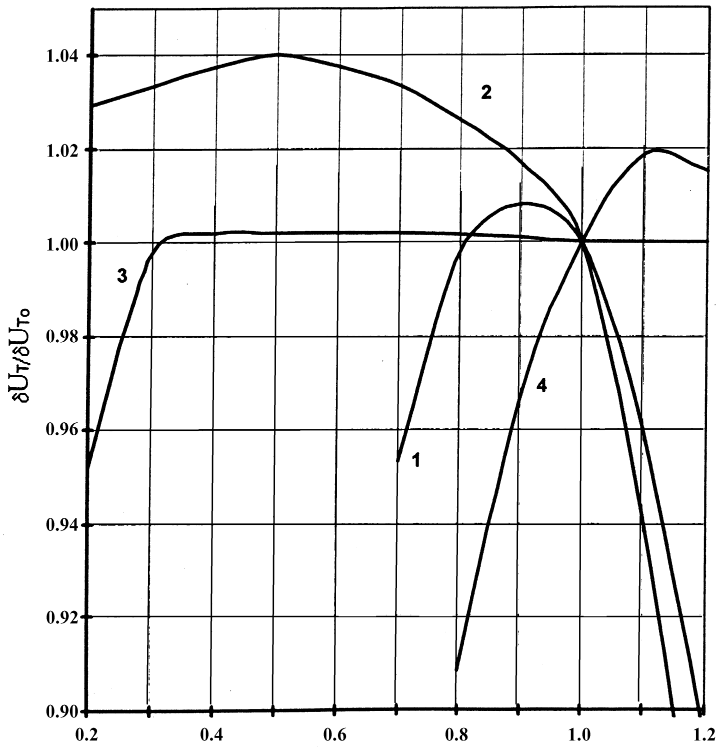


Figure 6. Relative separative power versus gas centrifuge parameters: 1 − θ/θ_0; 2 − β_F/β_{F0}; 3 −, $\overline{\omega}_F^0/\overline{\omega}_F$; 4 −, $\overline{Z}_F/\overline{Z}_F^0$.

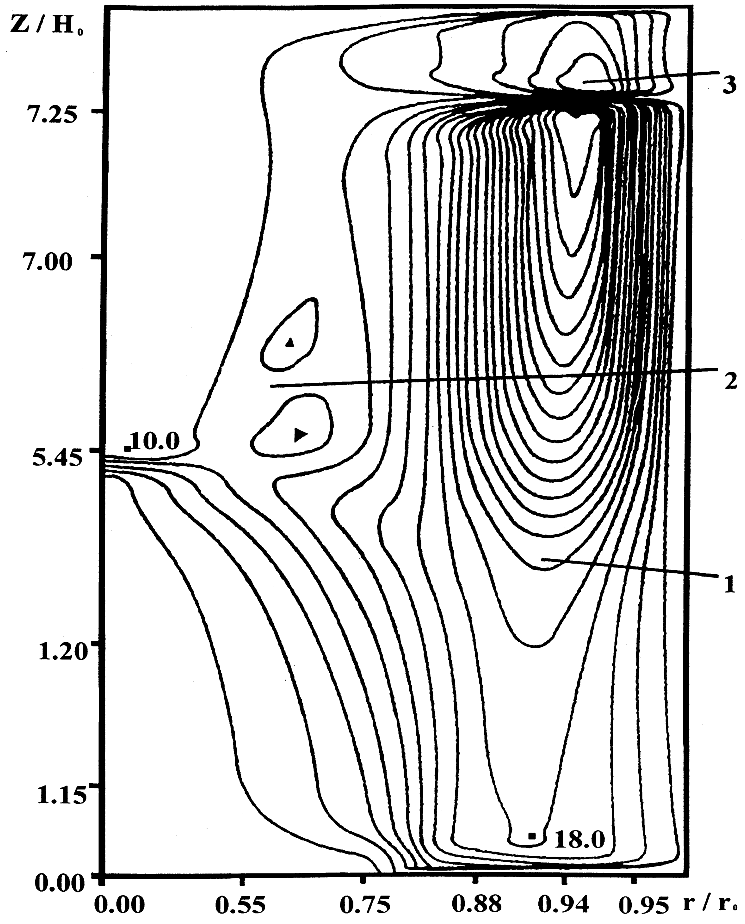


Figure 7. Typical distribution of circulation flow in gas centrifuge.

easily modified if necessary. The results of computer experiments on separation of sulfur isotopes from SF_6 of the natural isotope content (45) are presented in Fig. 6 for the relative values: Θ/Θ^o, β_F/β_F^o, $\bar{\omega}_F^o/\bar{\omega}_F$, $\bar{Z}_F$, $\bar{Z}_F^o$. The following centrifuge parameters were chosen as the basic set: the relative coordinate of the feed-flow inlet, $\bar{Z}_F^0 = 0.61$; the relative angular velocity of the gas in the feed-flow, $\bar{\omega}_F^0 = 0.1$; the input angle of the feed-flow in the rotor, $\beta_F^o = 90°$, and the cut, $\Theta^o = 0.45$. The analysis of circulation flow structure in the centrifuge rotor (the typical structure is shown in Fig. 7) can help to explain the computed dependencies. The structure of circulation flow consists of the main flow 1 excited by the stopping action of the gas-inlet and encompassing the main part of the separation chamber volume. The flow is regular and represents the main "mechanism" responsible for multiplication of the radial separation effect in the isotope mixture. The interaction of the feed-flow with flow 1 leads to the formation of an additional system of weak vortices 2. This system negatively affects the efficiency of the centrifuge. Circulation 3, which is separated from the main flow in the region above the gas scoop, also decreases the efficiency. The center of flow 1 is located near the edge of the scoop. This flow is so strong that it does not permit the formation of a special circulation flow excited by the temperature gradient on the side-wall of the rotor in the Stewartson layer, which is observed in the centrifuge for uranium isotope separation.

According to curves 2 and 4 in Fig. 6, the location of the feed-point and its slope angle to rotation axis appreciably influence the separation characteristics of the centrifuge. In the example considered, the optimal angle equals approximately 45°. The radial feed-flow employed in the basic centrifuge version, is less effective as it leads to excitation of the system of secondary flows 2. In addition, a part of the feed-flow moving opposite to the main flow 1 direction immediately reaches extractor of the heavy fraction. This effect can be eliminated by decreasing the angle from 90° to 45°. The optimal feed-flow in this case is found to correspond to $\overline{Z}_F \sim 0.7$ that is far higher than the theoretical value reported by Olander and Borisevich et al. (46,47). This discrepancy is caused by a sharp asymmetry of distribution of the circulation flow along the rotor length. According to the results of von Halle (48), the maximum of the circulation flow is to be formed in the feed region. Then, its profile approaches that of an ideal cascade, which corresponds to the highest separation capability. In our case, the maximum of the circulation flow is located near the scoop. This necessitates displacing the feed-point in the same direction. Therefore, the optimal cut Θ (see curve 1 in Fig. 6) also differs appreciably from 0.5.

Let us now consider curve 3 in Fig. 6 describing the effect of the azimuthal velocity of the gas in the feed-flow on the separative efficiency. As seen, the angle velocity of the feed-flow does not affect the separation in a wide range of its values. This is associated with conditions corresponding to the introduction of the gas into the rotor. An incompletely swirled gas is rapidly swirling and the momentum is redistributed between the azimuthal and radial components. The momentum loss results in the capture of feed-flow (accompanied by small perturbations) in the main circulation flow 1. In the cases when the value of feed-flow is close to ω_o remarkable perturbations of flow 1 resulting in the loss of separative efficiency, can be expected. The last conclusion is evidently confirmed by the dependence obtained.

Influence of Mixture Composition on Separative Characteristics of Gas Centrifuge

The influence of a mixture composition on the separative power and the separation factors was studied for the first time by computer experiment on separation of the third component of uranium isotope mixture (14). It was shown that variation of the component mole fractions within up to 0.5 could result in the change of the total separative power by up to 2.5%. The variations of partial separative powers and separation factors were shown to follow the linear law. Moreover, the partial separative powers remain constant when the mole fraction of the third component varies. Note once again that the above results have been obtained for the case of the three-component uranium isotope mixture, of which molar mass is rather high. A more general case of separation of an arbitrary multicomponent isotope mixture is considered by Levin and Ying (49). The solution of the problem was carried out by using an analytical method in linear approximation.

To understand the influence of a mixture composition on the separative power of a gas centrifuge, a diffusion problem for the mole fractions in Eq. (31) averaged along the rotor radius at small P and W values (compared with intensity of the circulation flow) was solved. The solution obtained has the following form:

$$\gamma_{ij} \approx (1 + \varepsilon_{\mathrm{i}})(1 - \varepsilon_{\mathrm{j}}) \tag{61}$$

where $\varepsilon_i = \varepsilon_{0_i}(\overline{m} - m_i)$ are the effective enrichment factors, and

$$\varepsilon_{0i} = \frac{\varepsilon_{or} H_0}{r_0^2(m^2 + 1)/(m\pi\rho\overline{D}_i)/L_{oi}} \approx \varepsilon_{\mathrm{o}}, \varepsilon_{\mathrm{or}} = \frac{\pi\omega_0^2}{RT}\int_0^{\mathrm{r}} \frac{\psi(r)}{L(r)}\, rdr,$$

$$L_{oi} = r_0\rho\overline{D}_i\left[2\int_0^{r_0}\left(\frac{\psi}{L}\right)^2 \frac{dr}{r}\right]^{-1/2} \approx L_0, L = \int_0^r |\,\rho V_z\,|\, 2\pi r dr, \tag{62}$$

$$\psi = \int_0^r 2\pi r\, \rho V_z dr, \quad m = L/L_0$$

Expressions standing in Eq. (62) account for variations of mole fraction in a gas centrifuge that are not big enough to lead to appreciable changes of coefficients $\overline{D}_{\mathrm{i}}$ in Eq. (26). Let us assume that arbitrary changes of mole fractions in a feed-flow c_i^F equal $\delta(c_i^F)$ and result in the changes in molar mass of the mixture of $\delta(\overline{m})$ and, respectively, to the changes $\delta(\overline{D}_{\mathrm{i}})$ of coefficients $\overline{D}_{\mathrm{i}}$. Using the expression for the diffusion coefficients $D_{\mathrm{ij}} \cong m_{ij}^{-1/2}$, $m_{ij} = m_i m_j/(m_i + m_j)$, variations of the total separation factors, which is defined by the following equation:

$$\delta(\gamma_{ij}) = \frac{\partial\gamma_{ij}}{\partial\overline{m}}\delta(\overline{m}) + \frac{\partial\gamma_{ij}}{\partial\overline{D}_{\mathrm{i}}}\delta(\overline{D}_{\mathrm{i}}) + \frac{\partial\gamma_{ij}}{\partial\overline{D}_{\mathrm{j}}}\delta(\overline{D}_{\mathrm{j}})$$

can be written as follows:

$$\delta(\gamma_{ij}) \approx -\varepsilon_0(\varepsilon_i + \varepsilon_j)\delta(\overline{\mathrm{m}}) + \left[\varepsilon_i \frac{\delta(\overline{m_{ik}^{1/2}})}{} - \varepsilon_j \frac{\delta(\overline{m_{jk}^{1/2}})}{}\right] \tag{63}$$

where $\delta(f) = f[c_k + \delta(c_k)] - f(c_k)$, $\mathrm{k} = 1,..n$; $\delta(\overline{m_{ik}^{1/2}}) = \sum_k m_{ik}^{1/2}\delta(c_i)$.

The first term in Eq. (63) reflects the influence of $\delta(\overline{m})$ on $\delta(\gamma_{ij})$ value. The value of this term almost does not depend on the molar mass of gas mixture. The second term in Eq. (63) shows the influence of changes of effective diffusion coefficients $\overline{D}_i$. Their contribution increases with the decrease of the molecular mass.

As follows from Eq. (54), the complete separation factors γ_{ij} have the strongest influence on the partial separative power values. From this, the variation of the separative power can be represented as follows:

$$\delta(\delta U_{ij}) \approx -\delta U_{ij} \frac{\delta(\gamma_{ij})}{\gamma_{ij} - 1} \tag{64}$$

Using approximate expressions $\gamma_{ij} - 1 \approx \varepsilon_i - \varepsilon_j$; $\overline{m_{ik}^{1/2}} \approx \overline{m^{1/2}}$ and Eq. (63) one obtains the following relationship between relative deviations of the separative power and changes of mole fractions:

$$\frac{\delta(\delta U_T)}{\delta U_T} \approx \left[\varepsilon_0 \delta(\overline{m}) - 2\varepsilon_0 \delta(\overline{m}) \sum_{j>i}\sum \frac{\overline{m} - m_i}{} + \frac{\delta(\overline{m_{ik}^{1/2}})}{} \right] \tag{65}$$

The maximum relative changes of separative power are described by the following equation:

$$\left. \frac{\delta(\delta U_T)}{\delta U_T} \right|_{\max} = -\left[\varepsilon_0 \Delta(\overline{m}) + \frac{\Delta(\overline{m^{1/2}})}{} \right] \tag{66}$$

where $\Delta(\overline{m}) = m_n - m_1, \quad \Delta(\overline{m}^{1/2}) = m_n^{1/2} - m_1^{1/2}$.

As follows Eq. (66), for modern gas centrifuges possessing $\varepsilon_o \geq 0.05$ the expected influence of mole fraction values on the separative power can reach 10% and more, whereas that of changes of mole fraction (through the change of diffusion coefficients for isotope mixture) can be around 5%. The results of calculations using Eq. (66) are collected in Table 1.

The qualitative dependencies of Eqs. (63) and (65) were examined by the results of calculations carried out by using the analytical model described under *Solution Methods of Diffusion Problems.* The compositions of tested isotope mixtures are represented in Table 2.

The dependencies δU_T and γ_{ij} for the process gases UF_6, SF_6, and CO_2 are presented in Fig. 8 in the form of surfaces in the space of mole fractions in the

Table 1. Maximum Value of $\frac{\Delta(\overline{m^{1/2}})}{\overline{m}^{1/2}}$ for Different Isotope Mixtures

	UF_6	SF_6	TeF_6	Xe	Kr
$\frac{\Delta\,(m^{1/2})}{\overline{m}^{1/2}}$	0.004	0.014	0.02	0.032	0.05

Table 2. Compositions of Feed Flow (Mole Fractions) for Studied Isotope Mixtures

i	1	2	3
	$^{235}UF_6$	$^{236}UF_6$	$^{238}UF_6$
C_i^{0F}	10^{-2}	10^{-2}	0.98
	$^{32}SF_6$	$^{33}SF_6$	$^{34}SF_6$
C_i^{0F}	0.98	10^{-2}	10^{-2}
	$^{12}C^{16}O_2$	$^{13}C^{16}O_2$	$^{12}C^{16}O^{18}O$
C_i^{0F}	0.98	10^{-2}	10^{-2}

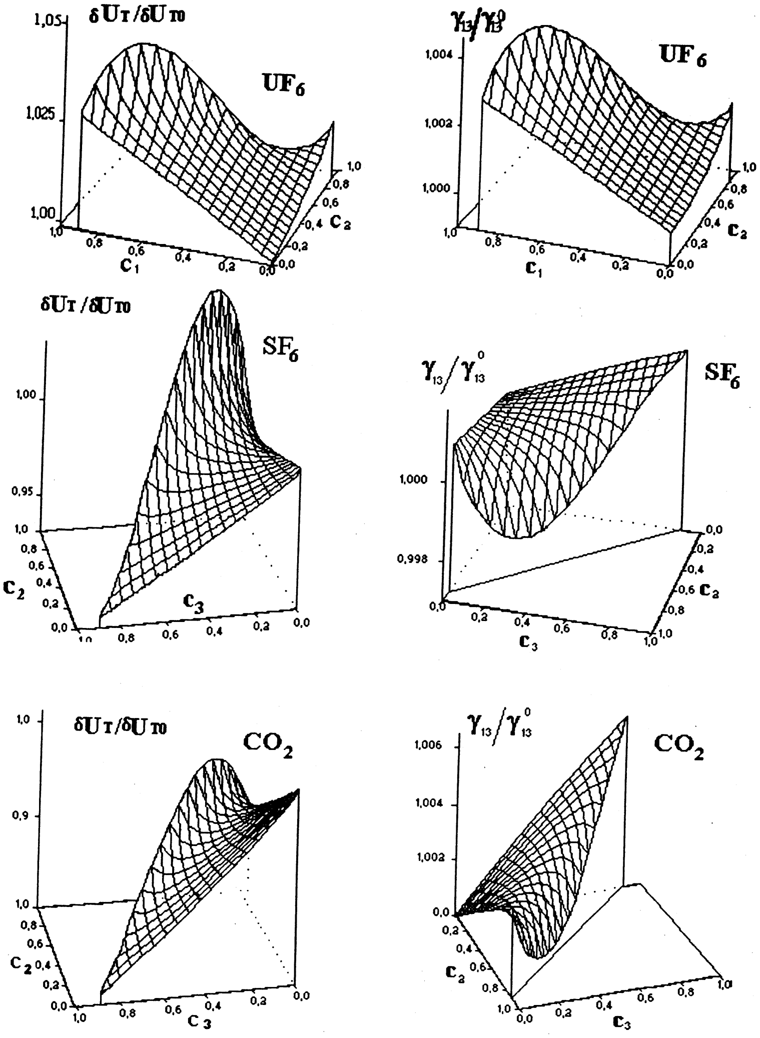
feed-flow c_i^F. The results shown in Fig. 8 clearly indicate the case when each process gas consists of only three isotopes. All surfaces are normalized by mole fraction values in the feed-flow of natural composition. The dependencies presented evidently illustrate the dependence of separation characteristics on concentrations. As seen, small changes of the separation factors cause substantial changes of the separative power. Note that the decrease of the separative performance is connected with an increase of the mixture molar mass due to an increase of mole fractions of heavier components.

The dependence of separation factors and separative power on composition of the mixture is of particular importance for the design of cascades of gas centrifuges. Small deviations of γ_{ij} can cause big changes in δU_T (see Fig. 8). Hence, this must be taken into account when calculating the cascade installations.

As follows from Eqs. (61) and (63) the separation factors γ_{ij} in the first approximation are the square-law functions on mole fractions in the feed-flow. This can be used to derive approximate formulae for calculation of the separation factors on the basis of experimental data obtained. The practical calculations show that to achieve a satisfactory accuracy when using a limited set of experimental data, the approximate formulae should be given as a polynomial of the third power written, for example, in the following form:

$$\gamma_{ij} \approx \gamma_{ij}^0(c_i^{F0})\left\{1 + \sum_{\substack{k=1\\k\neq n_1}}^{n}(c_k^F - c_k^{F0})\left[d_k^{ij} + \sum_{\substack{l=k\\l\neq n_1}}^{n}(c_{\mathrm{l}}^F - c_{\mathrm{l}}^{F0}) \times\left[b_{kl}^{ij} + \sum_{\substack{m=l\\m\neq n_1}}^{n}(c_m^F - c_m^{F0})\, a_{klm}^{ij}\right]\right]\right\} \tag{67}$$

where c_i^{F0} are the mole fractions of a feeding mixture with a basic composition of, for example, natural isotope content. To find the factors $a_{\mathrm{klm}}^{\mathrm{ij}}$, $b_{\mathrm{kl}}^{\mathrm{ij}}$ and $d_{\mathrm{k}}^{\mathrm{ij}}$ it is necessary to use some special numerical algorithms to minimize the deviation from



(a) (b)

Figure 8. Dependence of separative power (a) and separation factors (b) on mole fractions for different isotope mixtures.

the surface formed by a set of experimental points. For a big quantity of experimental data corresponding different c_i^F one needs to accomplish this procedure. At the same time, this problem can be far easier solved through the use of computer experiment technique. After determination of coefficients, Eq. (67) can be used to calculate cascade installations for separation of multicomponent isotope mixtures.

CONCLUSIONS

The gas-centrifuge technology has not yet become a universal tool for separation of stable isotopes. Isotopes of not every chemical element can be separated by using gas centrifuges because the volatile chemical compound for many elements of the Periodic Table have not been found so far. Nevertheless, at present the possibility of using the gas centrifuge to separate isotopes of 29 elements is feasible. The processes for isotope separation of 20 chemical elements have been developed in Russia by now, and 70 isotopes can be successfully separated by using this technology. Nevertheless, there are still a lot of scientific and technological problems to be solved that determines the prospects for the future activity in this field.

LIST OF SYMBOLS

c_i	mole fraction of *i*-th component
c_i^F	mole fraction of *i*-th component in a feed flow
c_i^P	mole fraction of *i*-th component in output of light fraction
c_i^W	mole fraction of *i*-th component in output of heavy fraction
c_P	specific heat at constant pressure, J/(kg·K)
c_V	specific heat at constant volume, J/(kg·K)
c_{Pi}	specific heat at constant pressure of *i*-th component, J/(kg·K)
D_{ij}	generalized multicomponent diffusion coefficient, $m^2 \cdot s$
D_{ij}	binary diffusion coefficient, m^2/s
$\overline{\mathrm{D}}_i$	effective diffusion coefficient, m^2/s
D_i^T	thermal diffusion coefficient, m^2/s
E	internal energy, J/m^3
E	Eckman number
$\mathbf{e}$	unit vector
F	feedflow, kg/s
$\mathbf{F}_\mathrm{i}$	spatial force density vector, N/kg
H_0	height of a rotor, m
h_i	enthalpy of *i*-th component, J/kg
h_i^0	standard enthalpy, J/kg

$\dot{\mathrm{I}}$	unit tensor
I_{φ}	sink density of azimuthal momentum, N/m^3
$\mathbf{J}_{\mathrm{i}}$	diffusion flow density vector, kg/(m^2·s)
J_{ir}	radial diffusion flow density, kg/(m^2·s)
k_{Ti}	thermal diffusion ratio
M	Mach number
m_i	molar mass of *i*-th component, kg/kmol
$\overline{m}$	molar mass of a gas mixture, kg/kmol
n	number of components in a gas mixture
n_i	number density of molecules, m^{-3}
N_A	Avogadro number, kmol^{-3}
P	light fraction flow, kg/s
P	gas mixture pressure, N/m^2
P_i	partial pressure of *i*-th component, N/m^2
$\mathbf{q}$	heat flow density vector, W/(m^3·kg)
R	gas constant, J/(kmole·K)
r	radial co-ordinate, m
r_0	radius of a rotor, m
$\mathbf{r}$	radius vector, m
r_{Kn}	Knudsen zone radius, m
$\dot{\mathrm{S}}$	deformation tensor, s^{-1}
t	time, s
T	absolute temperature, K
$\mathbf{V}$	mass average velocity vector, m/s
V	value function
V_r	radial mass average velocity, m/s
V_z	axial mass average velocity, m/s
V_{φ}	azimuthal mass average velocity, m/s
$\mathbf{v}_{\mathrm{i}}$	diffusion velocity vector of *i*-th component, m/s
$\mathbf{U}_i$	velocity vector of *i*-th component, m/s
W	heavy fraction flow, kg/s
z	axial co-ordinate, m
z_F	axial feed co-ordinate in a gas centrifuge, m
$\bar{z} = z/H_0$	dimensionless axial co-ordinate
α	gas compression ratio in a gas centrifuge
α_i	compression ratio for *i*-th component
α_{ij}	isotope mixture separation factor in light fraction flow
β_F	slope angle of feedflow admission, rad
β_{ij}	isotope mixture separation factor in heavy fraction flow
γ_{ij}	total separation factor
Γ	gas content in a gas centrifuge, kg
γ	ratio of the specific heats

δU	separative power, kg/s
δ_{ij}	Kronecker delta
δU_T	total separative power, kg/s
δU_{ij}	partial separative power, kg/s
ε_0	effective enrichment factor
η	viscosity, $N \cdot s/m^2$
η_i	viscosity of pure *i*-th component, $N \cdot s/m^2$
θ	cut
λ	mean free path, m
ν	heat conduction, $W/(m \cdot K)$
ν_i	heat conduction of pure *i*-th component, $W/(m \cdot K)$
$\dot{\Pi}$	pressure tensor, N/m^2
ρ	gas mixture density, kg/m^3
ρ_W	gas mixture density at side wall, kg/m^3
σ_i	collision diameter for *i*-th species of molecules, m
χ	wave-drag coefficient
Ψ	stream function, kg/s
ω_0	angular velocity of a gas centrifuge, rad/s
$\overline{\omega}_F = \omega_F/\omega_0$	relative angular velocity of a gas centrifuge
ω_F	angular velocity in a feedflow, rad/s
$\Omega_{ij}^{(1,1)*}$	reduced collision integral for mass transfer
$\Omega_{i}^{(2,2)*}$	reduced collision integral for momentum transfer
φ	azimuthal co-ordinate, rad

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