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SEPARATION OF ISOTOPES BY MASS-DIFFUSION TECHNIQUE

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ABSTRACT

This article reviews and critically analyzes the current state-of-the-art of theory and practical applications of mass-diffusion elements and columns for separation of stable isotopes. The problems of tailored design and optimization of their operation conditions are considered and discussed.

INTRODUCTION

The essence of mass-diffusion separation method consists of the use of distinctions in the diffusion rate of components of the mixture under separation in the flow of an auxiliary gas such as, for example, a previously condensed steam. The development of mass-diffusion method evolved in two directions: 1) realization of separation process in mass-diffusion elements, and 2) separation in mass-diffusion columns. Based on the same general principle, these two versions of mass-diffusion separation method differ essentially in the ways of excitation of the longitudinal flows of vapor and gas in apparatuses and maintenance of the inter-stage

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flows in connections combining the apparatuses in a cascade. Despite higher separation efficiency, the columns appeared to be rather sensitive to the magnitude of product stream (withdrawal), which was an obstacle to their wide application in practice. Therefore, preference was given to mass-diffusion elements in the design of cascades for separation of isotopes.

The major contribution to both theoretical and experimental development of mass-diffusion method was made by a group of Soviet scientists headed by Gverdtziteli. The method developed by this research group was successfully used in the cascade for production of neon isotopes.

According to the modern estimates of separation efficiency, the mass-diffusion method occupies an intermediate place between thermo-diffusion and distillation techniques. Hence, it can be successfully applied for the solution of different separation problems, including production of highly enriched isotopes in laboratory–and pilot–scale.

DEVELOPMENT OF MASS-DIFFUSION SEPARATION METHOD MASS-DIFFUSION SEPARATIVE ELEMENTS AND COLUMNS

Mass-Diffusion Elements

The first work in the field of mass-diffusion separation method dates back to 1922–1923 when Hertz had completely separated the neon-helium mixture by using a jet of water steam as diffusive environment (1). A large number of works devoted to the further research of separation process and development of designs of separative elements were carried out in 1934–1939 (2,3).

In 1934 Hertz, when developing his experimental model set-up, succeeded to combine both a separative element and a circulating pump in one device, and thus realized the cascade process without any additional circulating pumps (4). He used these combined elements (separative pumps) to construct the first mass-diffusion cascade for separation of hydrogen and neon isotopes wherein the 50% concentration of ^{22}Ne was achieved.

Figure 1 shows schematic diagram of the Hertz's separative pump and connection such elements in a cascade. A steam jet of working liquid (in an improved element design mercury was used as a working liquid) is injected from vaporizer 1 to the space between nozzle 2 and pipe 3. The steam flow is divided in this space into two parts of smaller diameter by pipe 3. The central part of the flow enters pipe 3 while the peripheral part is directed to the array between pipe 4 and the surface of condenser 5.

The gas mixture under separation is fed from the bottom of pipe 4. It moves upwards along this pipe and then diffuses in a jet of steam. As the diffusion coef-

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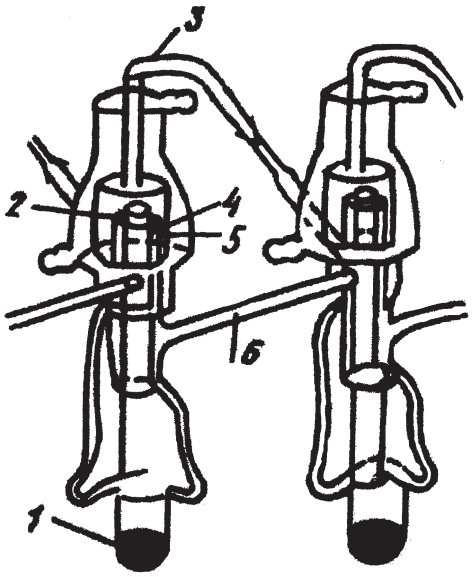

Figure 1. Schematic diagram of Hertz's separative pump and connection of elements in cascade (1,4): 1, vaporizer; 2, nozzle; 3, 4, 6, pipes; 5, surface of condenser.

ficient of a light component of mixture is higher than that of the heavier components, the gas reaching pipe 3 appears to be enriched by this component (light fraction). The gas going out from pipe 6 is enriched by heavy component (heavy fraction). Thus, the transfer of light fraction from the top of connecting pipe 3 to a subsequent element is provided. The transfer of heavy fraction from the bottom of pipe 6 to a previous element is also provided. As the result, the light fraction moves through the system of separative pumps connected in a series (cascades) from the left to the right. The heavy fraction moves in the opposite direction.

The theoretical and experimental investigation of separation process in the Hertz's pumps was carried out by Barvich (5). He determined the value of separation factor, α, for oxygen isotopes (in the $H_2{}^{16}O/H_2{}^{18}{}_{\circ}$ system) by diffusion of water steam in the flow of mercury vapor in a single pump to be of $\alpha = 1.085$. For neon isotopes separation ($^{20}Ne/^{22}Ne$) α value appeared to equal 1.2, and for carbon isotopes separation (in methane CH_4) $\alpha = 1.107$.

In 1937–1939 many isotopes were produced (in display quantities) by using a cascade of Hertz's pumps (3). For example, Kopferman yielded ^{36}Ar isotope with 50% concentration. Krüger succeeded to obtain a 20% enrichment of ^{15}N isotope, and Hampton obtained isotope of ^{13}C with 50% concentration by using methane (3).

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Sherr used the Hertz's pumps with somewhat modified geometry of the area between nozzle 2 and pipe 3 (see Fig. 1), however it did not essentially result in an increase of the separation effect. In his work Sherr used cascades comprising either 14 or 29 stages to separate carbon (in the form of CH_3Br), oxygen (as gaseous oxygen and water), neon and argon isotopes and obtained the following α values for above systems: $\alpha = 1.005$; $\alpha = 1.045$; $\alpha = 1.073$; $\alpha = 1.198$, and $\alpha = 1.154$, respectively.

Despite relatively high values of separation factors, the Hertz's pumps are characterized by poor efficiency (low productivity) due to both low working pressure (several torr) and small effective area of the vapor jet, through which the gas diffusion occurs.

A radical change of the design of mass-diffusion element was achieved in 1939 by Mayer (6), who introduced a porous diaphragm into the element to increase the productivity. Schematic of the Mayer's separative element is presented in Fig. 2. The main part of this apparatus is a cylinder, which is divided into two coaxial cylindrical chambers by means of a porous diaphragm. The gas mixture to be separated enters along the vertical central pipe to the top part of the internal chamber, and then moves along the diaphragm downwards. The steam-flow is fed into the bottom part of the external chamber and moves upwards. Hence, part of the vapor passes through the diaphragm and reaches the bottom condenser along with the flow of heavy fraction. The gas enriched with a light component diffuses against the steam flow, passes through the top condenser, and then moves to the subsequent element together with the remaining vapor. This provides a countercurrent movement of gas and vapor flows in the element along both sides of the diaphragm.

The pressure difference on diaphragm sides and the ratio of values of light and heavy fraction in the Mayer's apparatus were regulated by the use of three gates, shown in Fig. 2. When gate 2 is open the pressure at an external side of the diaphragm decreases in comparison with the pressure inside of the bulk limited by the diaphragm. Therefore, the diffusing part of the gas mixture enriched with light component (the largest part) goes away from the apparatus.

The most remarkable change of light component concentration is achieved when the pressure in both areas is maintained approximately identical. By using this apparatus for separation of Ne-He mixture, the separation factor was determined to equal $\alpha = 5.7$. For $^{12}CH_4 - {}^{13}CH_4$ respective α value appeared to be 1.015. The working pressure in these experiments equaled 1 atm., and the temperature was maintained between 408 and 423 K.

The efficiency of the Mayer's apparatus is proportional to the area of the diaphragm creating an additional diffusion space. For example, the Mayer's separative apparatus with diaphragm area of 1 m^2 is equivalent (in terms of productivity) to approximately several tens of thousands of Hertz's pumps. At the same time, the Mayer's element is characterized by lower α values in comparison with

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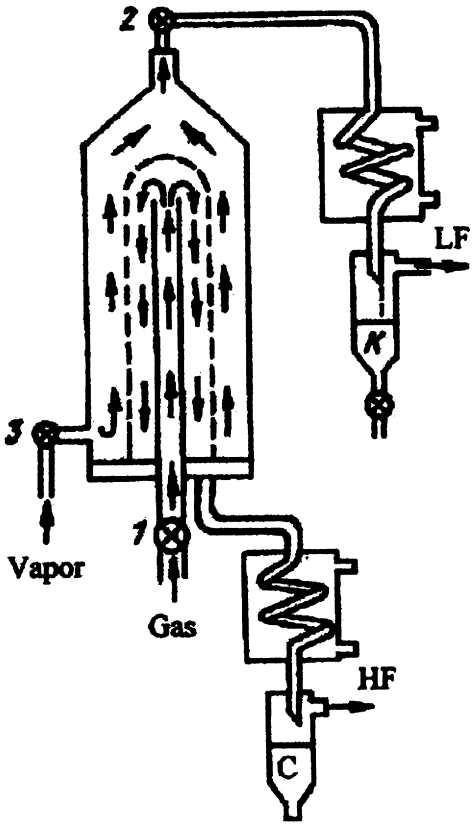

Figure 2. Schematic diagram of Mayer's (6) separating element: 1, 2, 3, gates; LF, light fraction; HF, heavy fraction.

Hertz's pumps. Thus, for carbon isotopes (in CH_4) $\alpha = 1.107$ for the latter while for the former it equals 1.015.

A simpler design of the glass elements, in which the main condenser (internal) was placed outside the diaphragm, was proposed by Gverdziteli et al. (6,7) in 1956. A schematic diagram of two separative elements of this type jointed in a series is shown in Fig. 3. In these elements the diaphragm used was made of steel pipe with diameter of 15 mm and working part of 40 mm height with wall thickness of 0.3 mm. The surface of diaphragm had 500 apertures of 0.4 mm in diameter. The condensing surface was offset by 3 mm from the diaphragm. The mer-

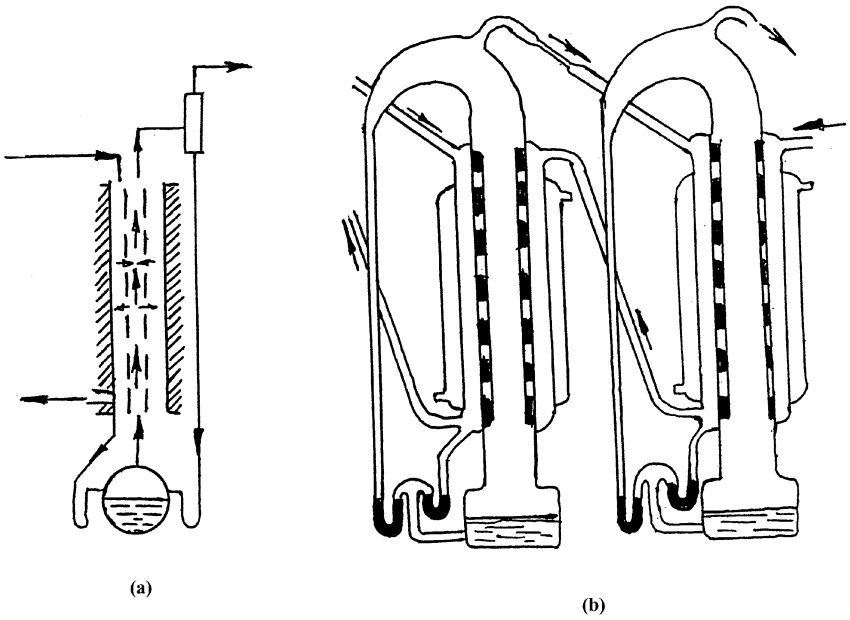

Figure 3. Schematic diagram of Gverdziteli's (7,8) separating element (a) and connection of two elements in series (b).

cury vapor entered the internal cavity of the diaphragm. A part of the vapor passed through the diaphragm aperture created a uniform radial flow, against which the diffusion of components of mixture to be separated occurred.

The other part of the vapor condensed outside the separative element in wide connecting pipes representing the external condenser, which was cooled by the air. The inter-stage flow was provided by the difference of pressure on the diaphragm sides. The value of light fraction flow was determined by the pressure difference in a capillary introduced in an appropriate communication pipe. The glass elements operated at pressure of 10 torr. The cascade comprising 80 elements of this type was used to produce highly enriched isotopes in quantities of 10 g. The main results of this work are collected in Table 1.

One of the merits of the described cascade was in its small gas holding. Approximately 100 n. cm^3 of gas (measured under normal conditions) was required to fill the whole of cascade volume at pressure of 10 torr. Therefore, the time of establishment of the stationary regime for isotopes listed in Table 1 did not exceed 20 h. However, for yielding substantial quantities of enriched isotopes, especially

Table 1. Enrichment of Isotope Mixtures in Cascades Comprising 80 Glass Elements

Enriched Isotope (component)	Separation Factor	Initial Concentration, %	Product Stream, cm^3/day	Enriched Isotope Concentration, %
^{22}Ne	1.20	9.8	50	99.99
^{20}Ne	1.22	91.2	100	99.99
^{36}Ar	1.15	0.3	40	45
$^{13}CH_4$–$^{12}CH_4$	1.095	2.7	30	92
$^{13}CH_4$–$^{12}CH_4$	1.095	2.7	200	42
^{86}Kr	1.033	55	100	55
^{86}Kr	1.033	92	60	92

when their concentration in the initial mixture is small, the productivity of this cascade was insufficient.

The metal mass-diffusion elements of considerably greater productivity were developed in 1965 on the basis of design of glass separative pump. The cascade for production of neon isotopes (9) was constructed of these elements. The schematic of this separative element is presented in Fig. 4. It consists of vaporizer 1 provided with internal electrical heater (heat booster) 2 filled with a thin layer of mercury, separative bulk 3 with diaphragm 4 and ring external condenser 5, and additional top condenser 6, the surface of which is located inside the element. Such design of the top condenser was chosen to reduce the gas holding of the element and to combine all units in the form of a uniform apparatus to make it more compact.

The diaphragm of 58 mm in diameter was demountable and was condensed from both ends by mercury shutters formed in the process of element operation. The height of working part of the diaphragm was 150 mm. The backlash between the diaphragm and the surface of condenser was similar to that in the glass element and equaled 3 mm. The diaphragm was made of metal foil and had 50 apertures per 1 cm^2 with diameter of 0.15 mm. A light fraction was removed from the top condenser through a capillary. Awning 7 and funnel 8 were placed in the internal cavity of the element to hinder trickling out of the mercury on the diaphragm surface.

The cascade, which consisted of 60 elements including 10 elements of the stripping section, was used to produce isotope ^{20}Ne by means of re-enrichment of the waste product formed in the concentration of ^{22}Ne with initial content of $\sim$5% of ^{20}Ne. The steady-state condition in the cascade was established sufficiently fast that it permitted the start of product withdrawal 12 h after starting the cascade operation. Such a short period of achievement of a steady state provided the high

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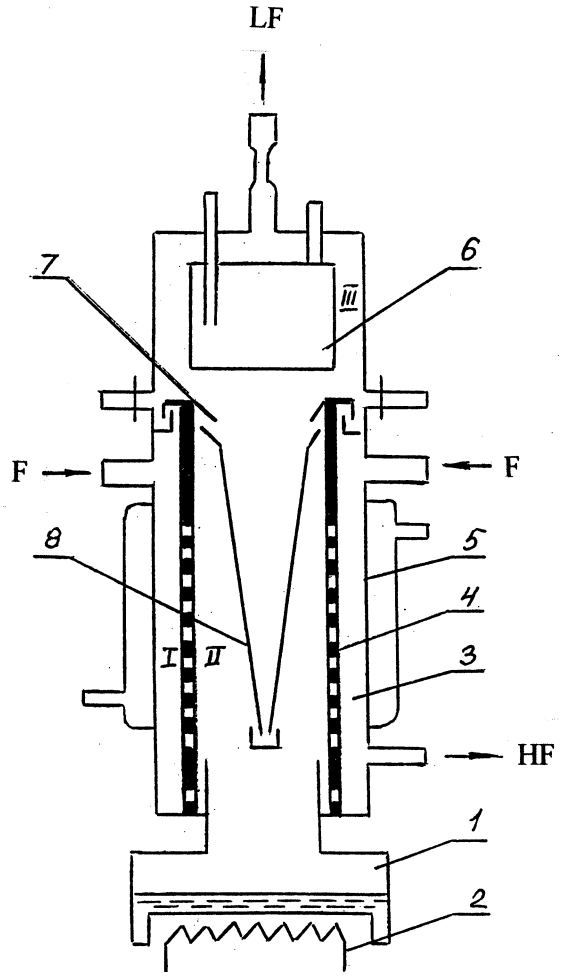

Figure 4. Schematic diagram of metallic mass-diffusion element (9): 1, vaporizer; 2, heat booster; 3, separating bulk; 4, diaphragm; 5, ring external condenser; 6, additional top condenser; 7, awning; 8, funnel; LF, light fraction; F, feed flow; HF, heavy fraction.

reliability of the cascade operation. At the product flow rate of $1.74 \cdot 10^3$ n · cm^3/day the concentration of isotope ^{22}Ne achieved 92%. At the cascade productivity of $2 \cdot 10^4$ n · cm^3/day the concentration of ^{20}Ne was 99.7%. The connection of subsequently parallel additional sections of 20 elements to the head part of this cascade (to ^{22}Ne reception section) allowed for increasing the product flow rate to $3.65 \cdot 10^3$ n · cm^3/day with ^{22}Ne concentration of more than 90%.

Mass-Diffusion Columns

The main studies on separation processes in mass-diffusion columns date back to 1950–1960 (8,10–14). A mass-diffusion column differs from a mass-diffusion element by the design as it contains the vapor chimney (see Fig. 5). A porous or punched tube is used as vapor chimney 1 to provide a uniform distribution of the vapor along the height of the apparatus. The internal wall of the column serves as the condensing surface. The convection flux fulfills the role of "phases" (by analogy with thermo-diffusion column); it ascends in the vapor chimney and descends along the condensing surface. Diaphragm 2 is introduced to make vertical gas-flows adjustable from the outside and to prevent them from hashing in the column.

Benedict and Boas (10) used free convection in the gravitation field to create the circulation (by analogy with Klusius and Dickel's thermo-diffusion column). The convection arose due to the difference in density of the gas and the va-

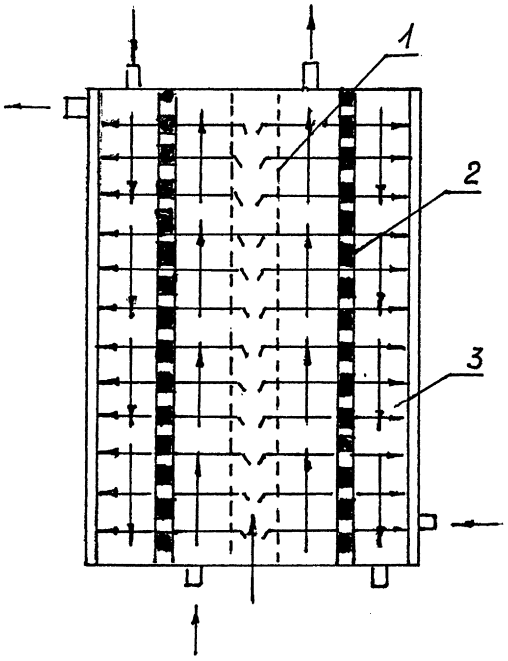

Figure 5. Schematic diagram of mass-diffusion column: 1, porous or punched tube; 2, diaphragm; 3, condensing surface.

por at the column and in the vapor chimney. The separation factor* determined with the use of water and carbon tetrachloride vapor as diffusion environment for separation of the $^{12}CH_4-^{13}CH_4$ mixture in a column supplied with a diaphragm of 100 cm in height was 1.058 and 1.024, respectively. Relatively small values of separation factor (see above) can be associated with unsuccessful choice of working liquids and non-optimal values of longitudinal circulating flows.

In the column design reported by Chichelli, Wheatherford and Bowman (11) the excitation of circulation is provided through the gas withdrawal (uptake) by the liquid film flowing down by the condenser wall (see Fig. 6). The liquid was fed upwards by a special pump and then it was splashed as a uniform layer on the surface of the condenser. The maximal separation factor for the $^{12}CH_4-^{13}CH_4$ mixture when using heptane as working liquid was 1.021.

In a similar column of 90 cm in height and with backlash of 7 mm Heymann and Kistemaker (12) obtained separation factors of 1.5 and 1.22 for the mixture of $^{20}Ne-^{22}Ne$ and of $^{36}Ar-^{40}Ar$, respectively, by using water and methanol as working liquids.

Los and Wet (13) used external centrifugal compressors to create the vertical convection flux in a column of 140 cm in effective height. The column was supplied with a stainless steel diaphragm (of 0.01 mm thickness) having 2500 apertures per cm^2 with an average diameter of apertures of 8 μm. Separation of neon isotope in this column by using xylene vapor at 175 torr was characterized by separation factor of 5.2.

More effective columns were designed by Barvich and Kucherov (14). The peculiarity of their columns consisted of the use of vapor chimney with apertures supplied with periodically located bars and in the way of creating circulating flows. The scheme of this column is presented in Fig. 7. The working vapor enters the column (filled with the mixture to be separated) from vaporizer 1 through vapor chimney 3. The main part of the vapor moves in the radial direction through the pores of diaphragm 4 to internal condenser 5. The liquid flows down the walls of the condenser and returns back to vaporizer 1 through the drainpipe. The working gas enters from the external condenser to the backlash between condensing surface and diaphragm where it moves downwards. In the bottom part of the column gas diffuses back to the internal cavity. The experiments were carried out on the column supplied with a diaphragm of 1 mm in length and diameter of 4 cm with effective thickness of 0.6 cm. The backlashes on both sides of the diaphragm were of 0.35 cm. The maximal separation factors achieved on the column of this type in fractionation of $^{22}Ne/^{20}Ne$, $^{36}Ar/^{40}Ar$ (with xylene vapor), and

*Here and above defined as the ratio of relative concentration of the target component at the ends of a column (or element).

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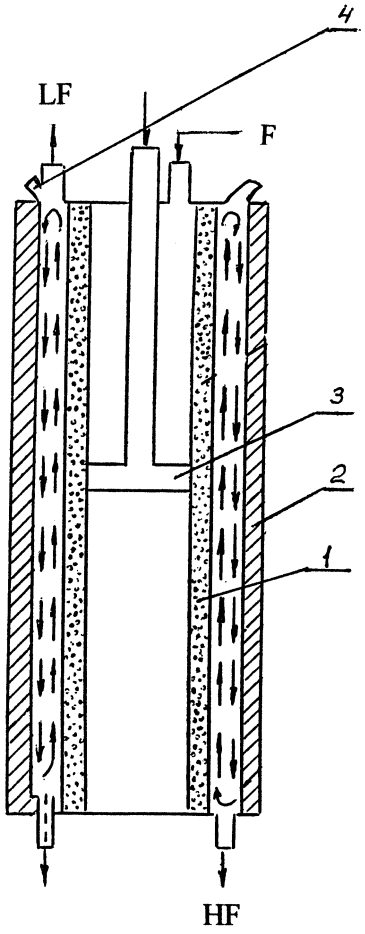

Figure 6. Schematic diagram of Chichelli et al. (11) mass-diffusion column: 1, porous tube; 2, condenser; 3, ring tube for gas supply; 4, tubes for liquid supply; LF, light fraction; HF, heavy fraction.

$^{13}CH_4/^{12}CH_4$ (with nitrobenzene vapor) were 9.5 (for Ne and Ar isotopes) and 2.6 (for $^{13}C/^{12}C$). The cascade, comprising 10 columns, permitted production of 400 cm^3/day of 99% ^{22}Ne. From the comparison of presented results it follows that the column developed by Barvich and Kucherov has obvious advantages over columns of other types.

However, the use of organic compounds as working liquids in all above studies inevitably reduced the efficiency of the separation process. The preference

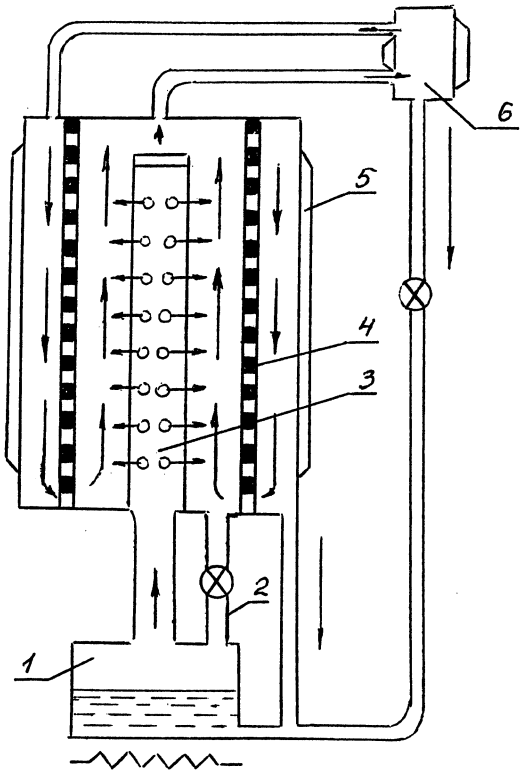

Figure 7. Scheme of Barvich and Kucherov column: 1, vaporizer; 2, line for additional vapor stream; 3, vapor chimney; 4, diaphragm; 5, 6, condensers.

given to organic liquids was based on the ease of selecting an appropriate working liquid with favorable physical and chemical parameters from a wide spectrum of organic substances. At the same time, organic fluids have a number of drawbacks, mainly thermal instability under long-term operation of separative apparatuses. Moreover, high solubility of gases in organic fluids results in considerable reduction of separation effect.

The use of metallic mercury as working liquid in works by Nikolaev et al. (15–17) has allowed overcoming these drawbacks. Mercury has sufficiently high molecular weight and high thermal stability. Mercury vapor can be easily condensed on a water-cooled surface. The solubility of gases in mercury is small. However, high chemical activity of mercury towards constructive materials and high boiling temperature required the search for new technical solutions in the col-

umn design. Figure 8 shows a schematic diagram of mercury mass-diffusion column. The column was constructed with special stainless steel, quartz and molybdenum glass. The diaphragm represented a stainless steel grid having 10400 apertures per cm^2 with cell size of $4 \cdot 10^{-3}$ mm (wire diameters of 64 and 32 μm). The initial grid was processed on rollers and was curtailed in one or several layers. The optimal diagram parameters such as grid thickness and number of grid layers were pre-selected.

The diameters of vapor chimney, diaphragm and internal condenser were 28, 38 and 45 mm, respectively. The length of the working part of the diaphragm was 1 mm. The vapor entered the column through the aperture of vapor chimney. An additional electrical heater was placed inside the vapor chimney to provide a uniform heating of the vapor along the chimney height and to prevent condensation of mercury on the chimney and the diaphragm.

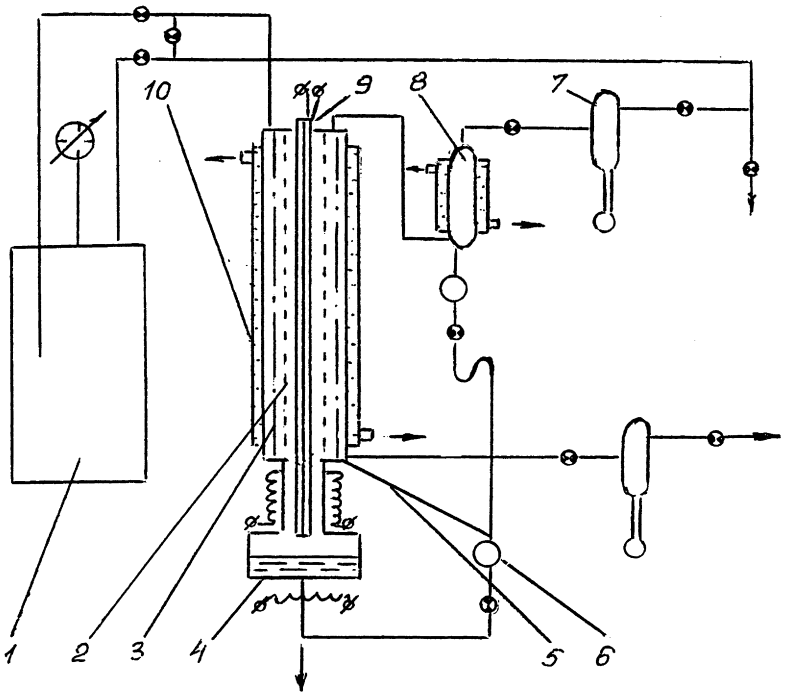

Figure 8. Schematic diagram of mercury mass-diffusion column (15): 1, bulk; 2, vapor chimney; 3, diaphragm; 4, vaporizer; 5, mercury sink; 6, gauge for measurement of mercury expenditure; 7, gauge for gas flow measurement; 8, external condenser; 9, additional heater; 10, internal condenser.

The results of the experiments carried out on this column have shown that the use of vapor chimney with a non-uniform distribution of vapor sources and appropriate selection of diffusive resistance of diaphragm allowed for creating longitudinal gas flows in the column without external sources of excitation of convection flux. The regulation of circulating flows was performed through the change of both resistance of external contour and the difference of pressure on the porous diaphragm.

The proposed way of excitation of circulation made it possible 1) to considerably simplify the design of installation, and 2) to achieve the value of circulating flows close to the optimum one. The column was connected with a 40-litre bulk containing an isotope mixture to be separated that provided the regularity of the pressure during the operation cycle. The overall vapor expenditure was defined by the time of filling of calibrated volume. The operating pressure in the column was in the range from 10 to 60 torr.

For the diaphragm having diffusive resistance of 0.6 cm the maximal separation factors for $^{20}Ne-^{22}Ne$ and $^{36}Ar-^{40}Ar$ mixtures were found to equal to 27.6 and 10, respectively. In a three-step cascade installation composed of separative columns of this type 97% concentration of ^{22}Ne at the product flow rate of $4.4 \cdot 10^{-3}$ cm^3/s was obtained (16,17).

The theory of separation process in mass-diffusion columns has been developed in detail and is well-documented in the above-cited articles. For this reason the theory of mass-diffusion columns will not be considered in this article.

Despite numerous studies of separation process which were carried out in mass-diffusion columns, they have found only limited application in a semi-pilot or laboratory scale. Besides high-sensitivity to the value of product flow rate (see above), parallel connection of several columns aiming to increase the productivity of the separation stage imposes rigid requirements to the uniformity of diaphragm and leads to the necessity of highly precise maintenance of technological conditions.

At the same time, mass-diffusion elements with a short diaphragm developed for the isotope production have sufficiently high technological characteristics. Stable working regime in a cascade along with simplicity and reliability of operation have resulted in the preference given to the practical use of cascades of separative elements. However, despite intensive experimental studies of separation process in mass-diffusion elements it was not possible for a long time to increase considerably the efficiency of the method. The main reason for this was dealing with complexity of establishment of the optimum distribution of the overall vapor-flow entering the element between the main and additional condenser. As the result, the main attempts were primarily directed to the increase of separation factor values of a single apparatus that was achieved, as a rule, to the detriment of the value of product flow (enriched with the target component) from the element outlet. This "gap" was eliminated by the authors in their works (per-

formed in cooperation with other investigators) on theoretical and experimental study of mass-diffusion elements (18–24,26). Some of the results obtained are presented in the following sections of the article.

THEORETICAL AND EXPERIMENTAL STUDY OF SEPARATION PROCESS IN MASS-DIFFUSION ELEMENT

To describe the process of separation of a binary isotope mixture, one needs to consider the diffusion in a ternary gas mixture, two components of which are similar in property. In this case, by neglecting the inessential effects of thermo- and baro-diffusion, one can derive sufficiently simple relations (based on the kinetic theory of gases) for the density flow of gas to be separated in the vapor:

$$\tau = -n \cdot D_{10} \cdot \nabla\gamma + u \cdot \gamma \tag{1}$$

and for the density flow of light isotope molecules in a ternary gas mixture:

$$\tau_1 = -n \cdot D \cdot \gamma \cdot (\nabla c - \varepsilon_0 \cdot c \cdot (1 - c) \cdot \nabla \ln \gamma) + u \cdot c \tag{2}$$

where

$$D = \frac{D_{10} \cdot D_{12}}{D_{10} \cdot \gamma + D_{12} \cdot (1 - \gamma)}$$

$$\varepsilon_0 = \frac{D_{10} - D_{20}}{D_{10}} \approx \frac{m_1 - m_2}{2 \cdot m_1} \cdot \frac{\mu_v}{m_1 + \mu_v}$$

The expression for elementary enrichment coefficient is reasonable to use for the vapor of auxiliary substance, which molecular mass exceeds 4–5 times the molecular mass of gas to be separated. The value of gas diffusion coefficient in the vapor must be as large as possible. Therefore, the mono-atomic vapors are more preferable than the multi-atomic ones with identical molecular masses. Furthermore, from this it follows that mass-diffusion is applicable for separation of isotopes of light and medium masses. Equations (1) and (2) together with continuity equations for τ and τ_1 and hydrodynamic equations form the total system, which is sufficient for the description of the mass-transfer process.

Modifications of construction of apparatuses described by Mayer (6) have led the authors to the apparatus design schematically presented in Fig. 9. The mass-diffusion separative element consists of four main units: vaporizer 1, diaphragm 2 and two condensing surfaces 3 and 4. The vapor flow Q_0 enters the bottom part of cavity II. The part of this flow $(1 - \theta_v)Q_0$ passes through the aperture of diaphragm to cavity I and condenses on the surface of the main condenser (3). The remaining part of the vapor $\theta_v \cdot Q_0$ is removed from the top end of cavity II and condenses in an additional condenser (4). Let us name the θ_v

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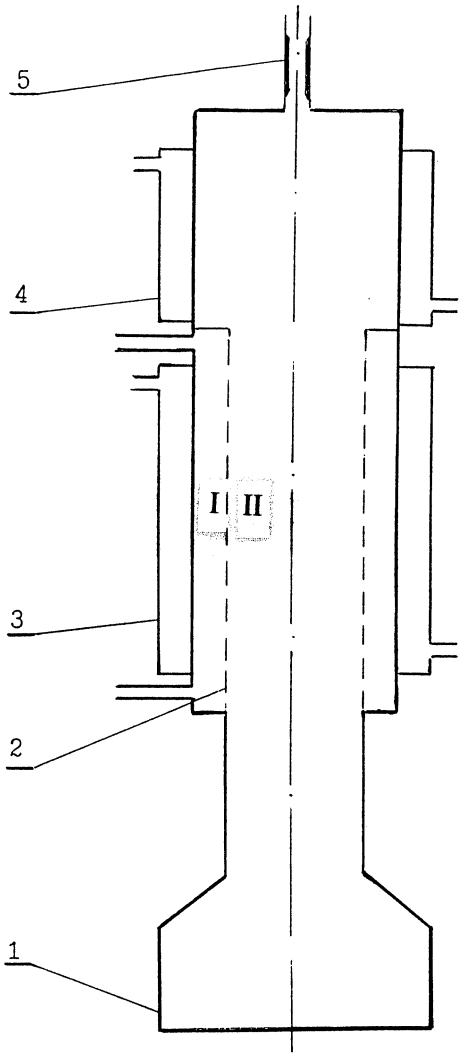

Figure 9. General schematics of mass-diffusion element: I and II, cavities; 1, vaporizer; 2, diaphragm; 3, 4, condensing surfaces; 5, capillary; →, gas stream; ⇒, vapor stream.

value as "steam distribution factor". The flow L of the mixture to be separated with light component concentration of c_o is usually directed to the top of cavity I between the diaphragm and the main condenser. The part of gas $L' = \theta \cdot L$ diffuses towards the vapor flow through the porous diaphragm, thus being enriched with light component until it achieves the concentration c^+ followed by its re-

moval together with vapor from the top end of cavity II. The remaining part of gas $L'' = (1-\theta)L$, depleted with light component to concentration c^- is removed from the bottom end of cavity II.

The solution of the problem of convective diffusion of a ternary mixture in a separative element is hindered both by complexity of joining diffusion and hydrodynamics equations, and by complexity of imposing the boundary conditions. However, in a number of simple cases it is possible to obtain the analytical expressions, connecting working and geometrical parameters of the element with its separative characteristics ε, L and δU.

Thus, if the value of gas-vapor mixture flow through the diaphragm, u, is unknown and the connection of the Peclet diffusion criterion $\ln q = ul_e/nD_{10}$ with total vapor flow Q_0 and steam distribution factor is disregarded, one can derive from Eq. (1) the following useful expressions for the flow density of gas through the diaphragm τ_x:

$$\tau_x = \frac{n \cdot D_{10}}{l_e} \cdot \frac{\gamma_a - q \cdot \gamma_b}{q - 1} \cdot \ln q \tag{3}$$

where

$$\gamma_a = \gamma_0 \cdot \frac{l_e}{a} \cdot \frac{1 - \exp(-u \cdot a/n \cdot D_{10})}{\ln q} \tag{4}$$

$$\gamma_b = \gamma_a \cdot \left(1 - \left(1 - \frac{u \cdot \prod}{Q_0}\right)^{1/(q-1)}\right) \tag{5}$$

$$\ln q = u \cdot l_e/n \cdot D_{10} \tag{6}$$

and the flow of light fraction equals

$$L' = \prod \cdot H \cdot \frac{n \cdot D_{10}}{l_e} \cdot \gamma_a \cdot \theta_v \cdot \frac{1 - \theta_v^{1/(q-1)}}{1 - \theta_v} \cdot \ln q \tag{7}$$

By neglecting the influence of availability of gas in cavity II and its direct movement along the diaphragm (that is true for the diaphragm of small length and small γ_a values) one can derive from Eq. (2) the following simple expression for enrichment factor of the element:

$$\varepsilon = \varepsilon_0 \cdot \frac{q \cdot \ln q}{q - 1} \cdot \frac{1}{\theta} \cdot \ln \frac{1}{1 - \theta} \tag{8}$$

The accounting for γ_a value leads to the following expression for ε, which can be derived only by the using numerical integration:

$$\varepsilon = \varepsilon \cdot \prod \cdot \frac{1}{\theta} \cdot \int_o^H \frac{\alpha \cdot \tau_x}{\delta \cdot G}\, dy \tag{9}$$

where

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$$\alpha = \int_0^l \frac{d \ln \gamma}{dx} \cdot \exp\left(- \int_0^l \frac{\tau_x}{n \cdot D \cdot \gamma} \cdot dx \right) \cdot dx \tag{10}$$

$$\delta = \exp\left(- \int_0^l \frac{\tau}{n \cdot D \cdot \gamma} \cdot dx \right) \tag{11}$$

$$G = (1 - \theta) \cdot L + Q_0 \cdot \gamma_a \cdot \left(1 - \frac{u \cdot \prod}{Q_0} \cdot y \right) \cdot \left(1 - \left(1 - \frac{u \cdot \prod}{Q_0} \cdot y \right)^{1/(q-1)} \right) \tag{12}$$

The analysis of expression for τ_x shows that at sufficiently high concentrations of gas in internal cavity, γ_a, (that is observed at small charges of vapor and small θ_v value) this value can change the sign in top of the element. It corresponds to the case, when return convective transfer of the gas begins to prevail over the molecular diffusion. The internal circulation is known to emerge in the element (see Fig. 10). The internal circulation reduces the light fraction flow leaving the element and thus reduces its productivity.

Therefore it is necessary to maintain the value of γ_a at a sufficiently high level that is connected with additional expenditure of vapor condensed in the top condenser. It should be emphasized that the role of an additional condenser as a necessary constructive unit of mass-diffusion apparatuses is defined by this particular condition.

Expressions (3)–(6) are obtained for the case when both $\ln q$ and θ_v value changes independently from each other. In the real apparatuses they are always strictly interconnected with each other due to the nature of distribution of the total vapor flow between the main and additional condensers. This interconnection, however, can be understood only through accounting for the particular features of separative element design. For the most widespread elements with additional condensers being the continuation of cavity II and vertical arrangement of condensing surface on the basis of the solution to the problem heat and mass transfer in various zones of apparatus the algebraic transcendent equations system (20,22) was obtained. This system makes it possible to calculate simultaneously u, Q_0, L', P, and T, and consequently $\ln q$, θ_v and ε as a function of the entering vapor flow:

$$u \cdot \prod \cdot H = Q_t - L' \tag{13}$$

$$L' = \gamma_a \cdot Q_0 \cdot \left(1 - \frac{Q_t - L'}{Q_0} \right) \cdot \left(1 - \left(1 - \frac{Q_t - L'}{Q_0} \right)^{1/(q-1)} \right) \tag{14}$$

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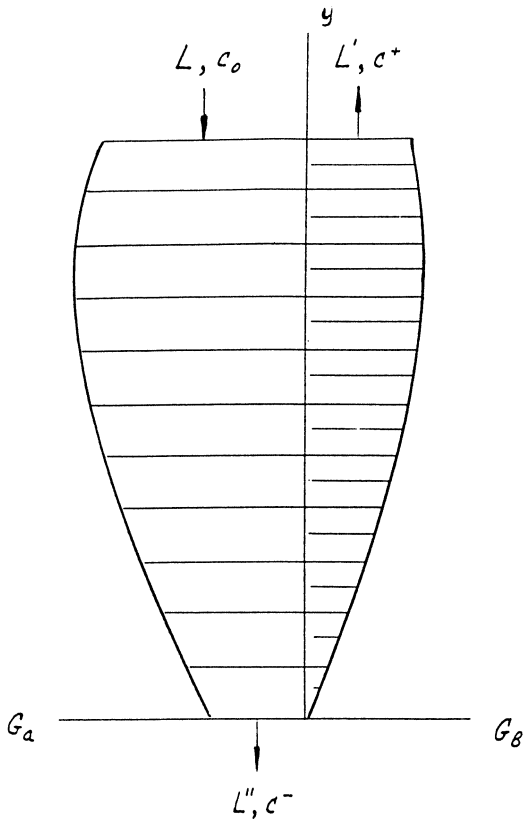

Figure 10. Internal circulation in separating element: G_a and G_b are distributions of gas flow rates in cavities I and II (see Fig. 9), respectively; L is flow of mixture to be separated; L' is light fraction flow; L'' is heavy fraction flow; C_o, C^+, C^- are concentrations of light component in feed flow, light fraction flow, and heavy fraction flow, respectively.

$$u = \sqrt{\frac{16 \cdot \eta \cdot l_c \cdot T_c \cdot L'}{\pi \cdot r_c^4 \cdot \mu_v \cdot (1 - (1 - \mu_g/\mu_v)) \cdot T_p}} \left| \left(\frac{F_1}{\alpha_1 \cdot F_0} - 1 \right) \right| \tag{15}$$

$$P_p = \frac{P_0 \cdot V_0}{V_{com} + \pi \cdot x_c^2 \cdot (H_b - \delta \cdot (Q_0 - Q_t)/(nD_{10} \cdot \prod \cdot \ln(\gamma_a/\gamma_b)))} \tag{16}$$

$$\ln P_p = 7.20 - 2760/T \tag{17}$$

$$\gamma_d = \frac{\gamma_a - \gamma_b}{\ln q} - \frac{\gamma_a - q\gamma_b}{q - 1}, \quad a_1 = 0.63 + 0.37 \cdot (F_0/F_1)^3 \tag{18}$$

$$n = P_p/(RT_p) \tag{19}$$

The system of Eqs. (4)–(6), (9)–(17) complemented with the boundary conditions on the walls of main and additional condensers, permits the determination of the main separative characteristics of element ε, L and δU. The following relation was used to calculate the gas concentration at condensing surfaces:

$$\gamma_0 = 1 - P_c/P_p \tag{20}$$

where P_c is the pressure of saturated vapor at temperature of the wall. To establish the connection between vapor flows in the main and additional condensers the continuity condition for the gas concentration in gas-vapor mixture on their borders was used.

Finally, for the complete description of the separation effect in the element it is also necessary to take into account the direction of movement of gas flows on both sides of the diaphragm which can be either co-current or counter-current.

The comparison of theoretical and experimental data was carried out for elements operating with mercury as working liquid in separation of neon isotopes. The elements had a cylindrical diaphragm of 5.8 cm in diameter, a height of 15 cm, and a diffusive resistance of 1.3 cm (50 apertures per 1 cm^2 with diameter of 0.014 cm). The height of the top condenser was 15 cm, and the capillary had the following parameters: $d_c = 0.28$ cm and $l_c = 5$ cm.

Figure 11 shows experimental and calculated dependencies of Q_0, L' and ε on the total vapor flow Q_0. The Q_0 and L' values were determined by measuring the expenditure of mercury condensate from the main condenser and the pressure difference in capillary, respectively. The value of ε was calculated by the results of mass-spectrometric isotope analysis of enriched and depleted fraction flows. The comparison made shows a satisfactory correlation between calculated and experimentally determined values of the above characteristics. This confirms the applicability of the developed model of separation process in the mass-diffusion element for practical calculations.

The separative power of element, δU, is one of the basic characteristics of separative apparatuses. This parameter was calculated by using the values of light fraction flow and the enrichment coefficient as follows:

$$\delta U = L'\varepsilon^2/4 \tag{21}$$

The variation of separative power with total vapor flow shown in Fig. 12 indicates that its increase is mainly due to the rise of light fraction flow since the enrichment coefficient in the range of large Q_0 values changes slowly. The nature of δU dependence on the total vapor flow (see Fig. 12) is stipulated by the behavior of L' and ε parameters as functions of Q_0.

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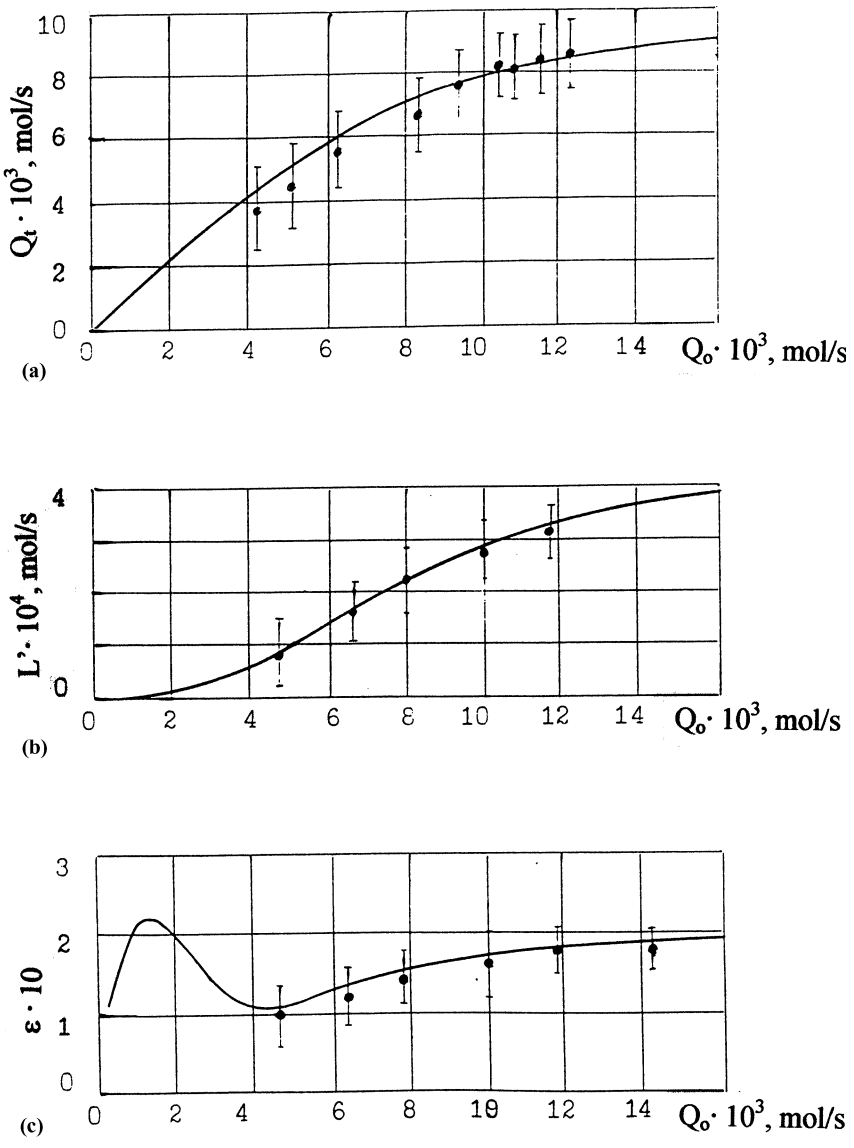

Figure 11. Experimental (points) and computed (lines) $Q_t = \theta_v Q_0$ (a), L' (b), and ε(c) versus Q_0 dependencies (see text).

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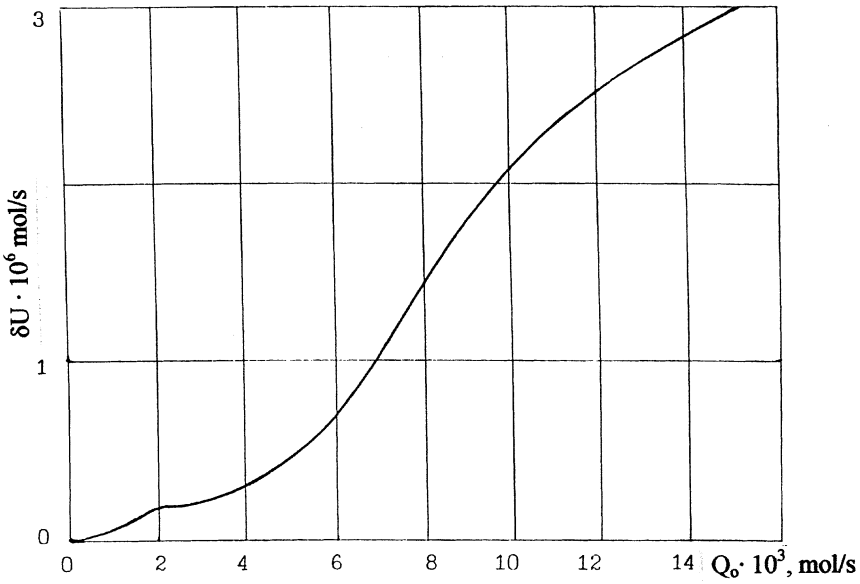

Figure 12. Separative power of element versus Q_0 dependencies for $\theta = 0.5$.

CONNECTION OF MASS-DIFFUSION ELEMENTS IN CASCADES

For the production of the enriched product from the initial raw material with required concentration of target isotope a definite number of elements connected in a cascade is needed. The mass-diffusion elements can be combined in a cascade in three various ways. The mode of their connection considerably affects the character of gas and vapor flows inside elements and their separative efficiency (see Fig. 13). For example, when the feed flow enters into the top of cavity 1 the elements work in the counter-current mode of operation (see Fig. 13a). This type of connection is the most widespread. However, at a high diaphragm length (height) it seems to be non-expedient since it may result in remarkable isotope mixing effects at the points of flow entrance. Indeed, the element with a long diaphragm can be represented as a part of the cascade consisting of several counter-current stages. Hence, removal of light fraction flow through these stages will evidently cause the isotope mixing. For this reason it seems more appropriate to introduce such elements according to so-called column connection scheme (see Fig. 13a). In this case both direct and return interstage flows form a closed contour, connecting the outlet of the previous element with the inlet of subsequent elements. In Fig. 14 the flow profiles inside the elements are shown for different variants of their connection.

As follows from Fig. 14b, by using the column connection scheme the junction of interstage flows is to be made at some distance from the bottom end of the previous element as in this area the ascending and descending parts of circulating flow are small. As at the column type of connection the value of internal flow in separative element defines the profile of the cascade, the narrowing of this profile at the points of connection of the elements is a considerable drawback of such a scheme.

Nevertheless, due to the self-multiplication effect of elements with a long diaphragm the column connection scheme can be used in the cascade sections of the final concentrating. By using the co-current connection scheme of elements (see Fig. 14c) the multiplication of the primary separation effect on the diaphragm is absent while the losses caused by the gradual depletion of gas with light com-

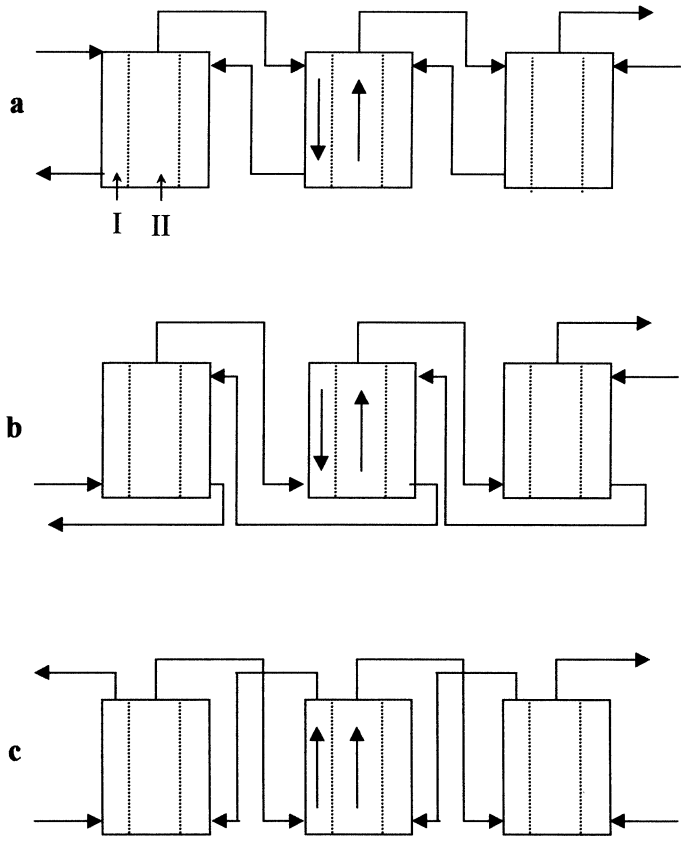

Figure 13. Schematic diagrams of connection of mass-diffusion elements in cascade: a, counter-current mode; b, column mode; c, concurrent mode; I and II, cavities.

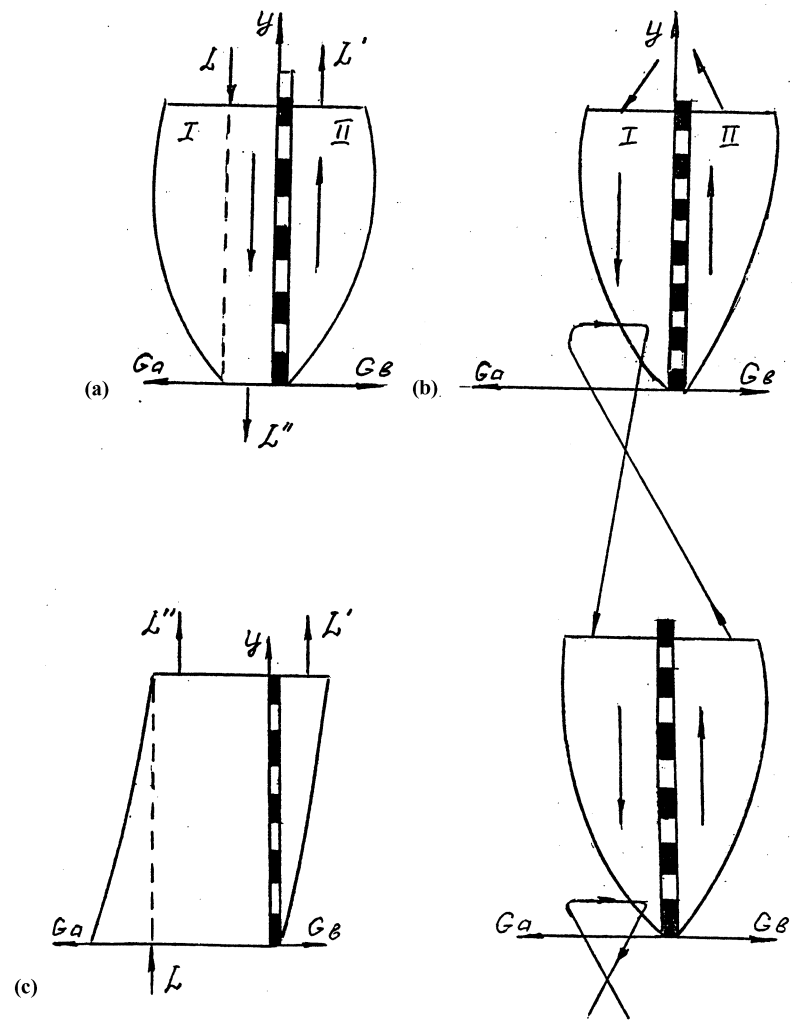

Figure 14. Profiles of gas flow inside separating elements at various modes of their connection: a, counter-current connection; b, column mode; c, concurrent mode. For designations see Fig. 10.

ponent during its flowing along the diaphragm in cavity 1 still remain. Therefore, the co-current connection of elements is not usually applied.

The interstage flows in mass-diffusion cascades are created through the use of pressure difference on the diaphragms of separative elements. The regulation of these flows is performed by means of variation of hydrodynamic resistance of capillaries on the lines of light fraction. At all types of connections of elements the flow of light fraction from the cavity of an external condenser of the previous el-

ement (area of higher pressure) through the capillary moves to cavity 1 of the subsequent element (to the area with lower pressure).

OPTIMIZATION OF MASS-DIFFUSION ELEMENT PARAMETERS

The number of elements required for achievement of the desired enrichment of the target isotope in the product is proportional to $1/\delta U$ (25) and defines the installation costs of a separative set-up. The construction of a cascade with minimum volume (minimum number of elements) requires selecting the mode of operation and geometrical parameters of the element providing the maximum value of separative power. However, its increase is connected with production of large vapor flows that results in an increase of energy expenditure. Therefore, it is necessary to choose the working regime of the element on the basis of criterion accounting for the ratio between energy expenditure and operating costs providing the required quantity of isotope production (24). The specific cost function, S_{sp}, was used as such a criterion. Their ratio can be written as follows:

$$S_{sp} = \frac{1}{\eta f \cdot \delta U} \cdot \left(\Phi(c_P) + \frac{c_P - c_F}{c_F - c_W} \cdot \Phi(c_W) - \frac{c_P - c_W}{c_F - c_W} \cdot \Phi(c_F) \right) \times \left(\frac{a_k \cdot (1 + k_1)}{t_c} + (a_e + a_c) \cdot Q_0 \right) + \frac{c_P - c_W}{c_F - c_W} \cdot a_F + \frac{a_{zp}}{P} \tag{22}$$

where $\Phi(c) = (2c - 1)\ln(c/(1 - c))$, a_k and k_1 are the factors accounting for amortization and reparation of instruments, a_e and a_c are the value factors of energy and coolant, a_F is the cost of feed flow unit, and a_{zp} is the personnel wages per unit time.

The optimum working and geometrical parameters of mass-diffusion elements calculated by using objective function (22) are collected in Table 2. The value of separative power corresponding to parameter values given in Table 2 equals $4 \cdot 10^{-6}$ mol/s that exceeds by more than 75% the magnitude of separative power of the element described by Gverdtziteli et al. (8).

The influence of the main parameters of the mass-diffusion element on specific cost was also studied in the course of solution of optimization problem. The dependencies of specific cost value and its power and hardware components on the diameter of capillary, diffusion resistance and height of the diaphragm and total vapor flow shown in Figs. 15 and 16 illustrate the results obtained. The value

Table 2. Optimum Parameters of Separative Element

$Q_o \times 10^2$, mol/s	l_e, cm	d_c, cm	H, cm	P_p, torr
0.9	1.55	0.44	25.4	26

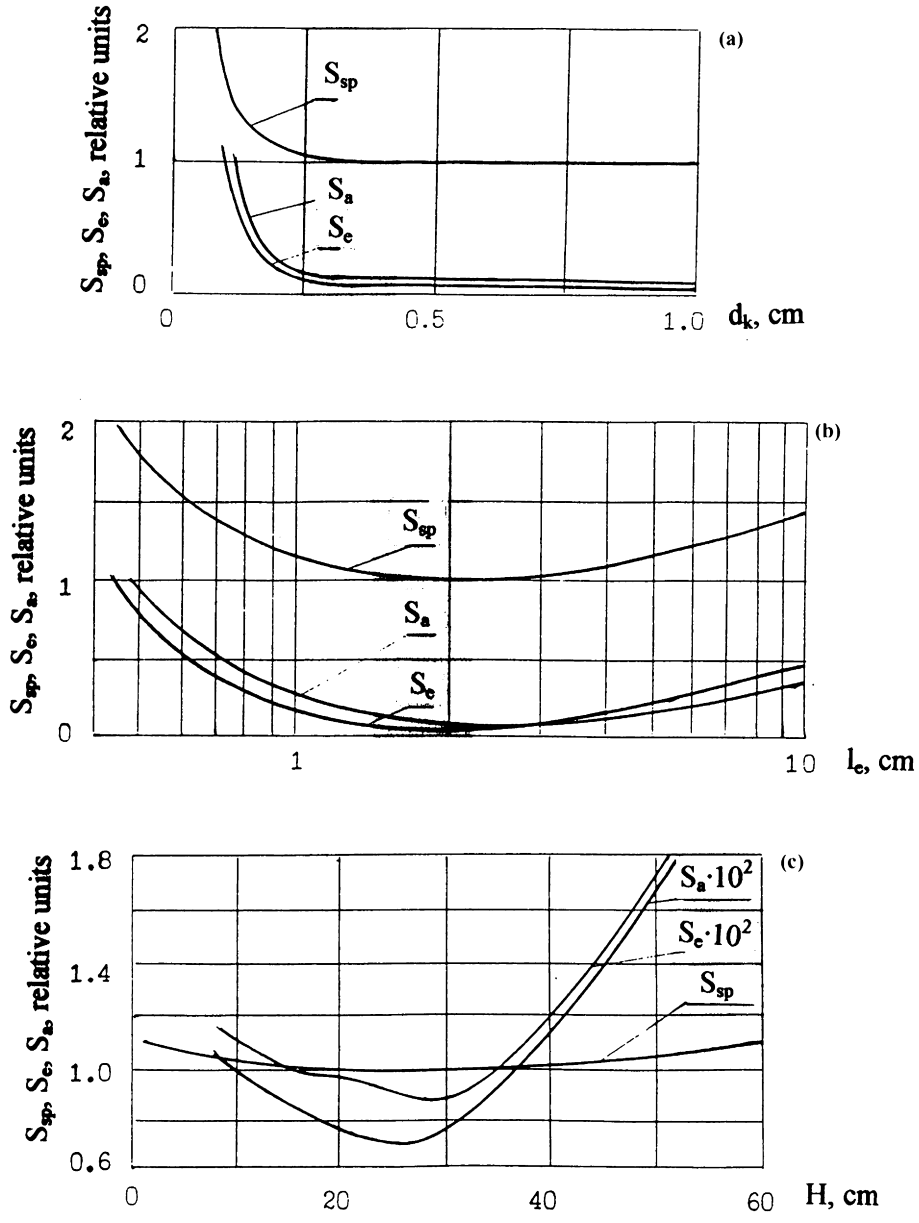

Figure 15. Dependence of specific cost S_{sp} on capillary diameter d_c (a): l_e (b) is diffusive resistance of diaphragm; H (c) is height of diaphragm.

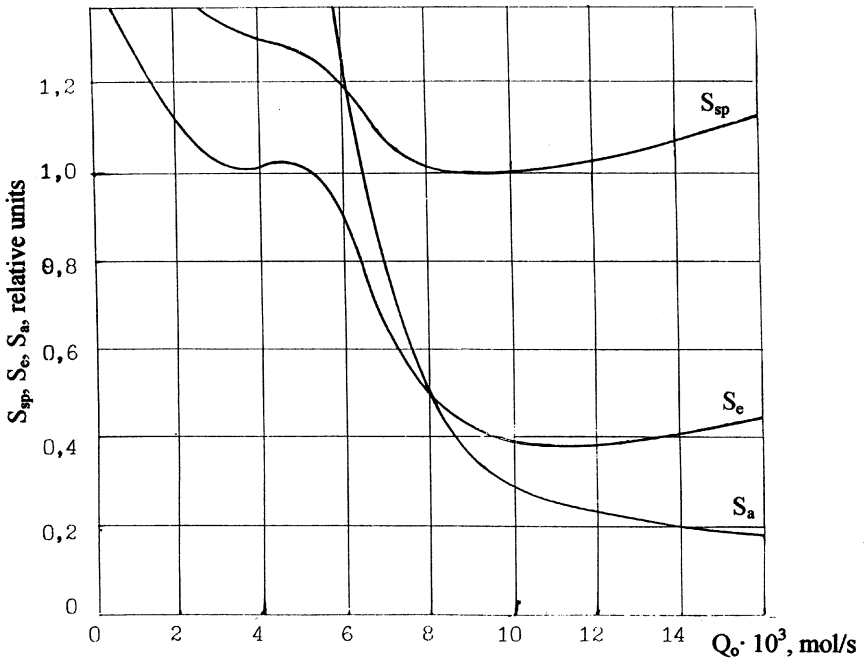

Figure 16. Dependence of specific cost S_{sp} and specific cost components, S_e and S_a, on total vapor flow Q_0.

of specific cost and their components were normalized by the minimum specific cost value. The calculation of dependencies for each parameter was carried out at the optimum values of other parameters.

As follows from Fig. 15, the increase of capillary diameter to 0.4–0.5 cm leads to substantial reduction of specific cost, which remains constant at a large value of capillary diameter. Such behavior of S_{sp} function can be explained as follows: the growth of d_c in the range of small values of capillary diameter primarily leads to the reduction of pressure difference on the diaphragm and, as a consequence, to the increase of the steam distribution factor θ_v. It causes, in turn, the growth of light fraction flow and, hence, results in the growth of separative power. The further increase of capillary diameter hardly causes any change of separative power, because with large d_c values the light fraction flow is defined only by diffusion process through the diaphragm and slightly depends on the steam distribution factor θ_v. The occurrence of a minimum at the dependence of specific cost on the diffusive resistance of the diaphragm and its lengths (see Fig. 15, b and c) is explained by the competitive influence of both enrichment coefficient and light fraction flow on separative power with the change of l_c and H.

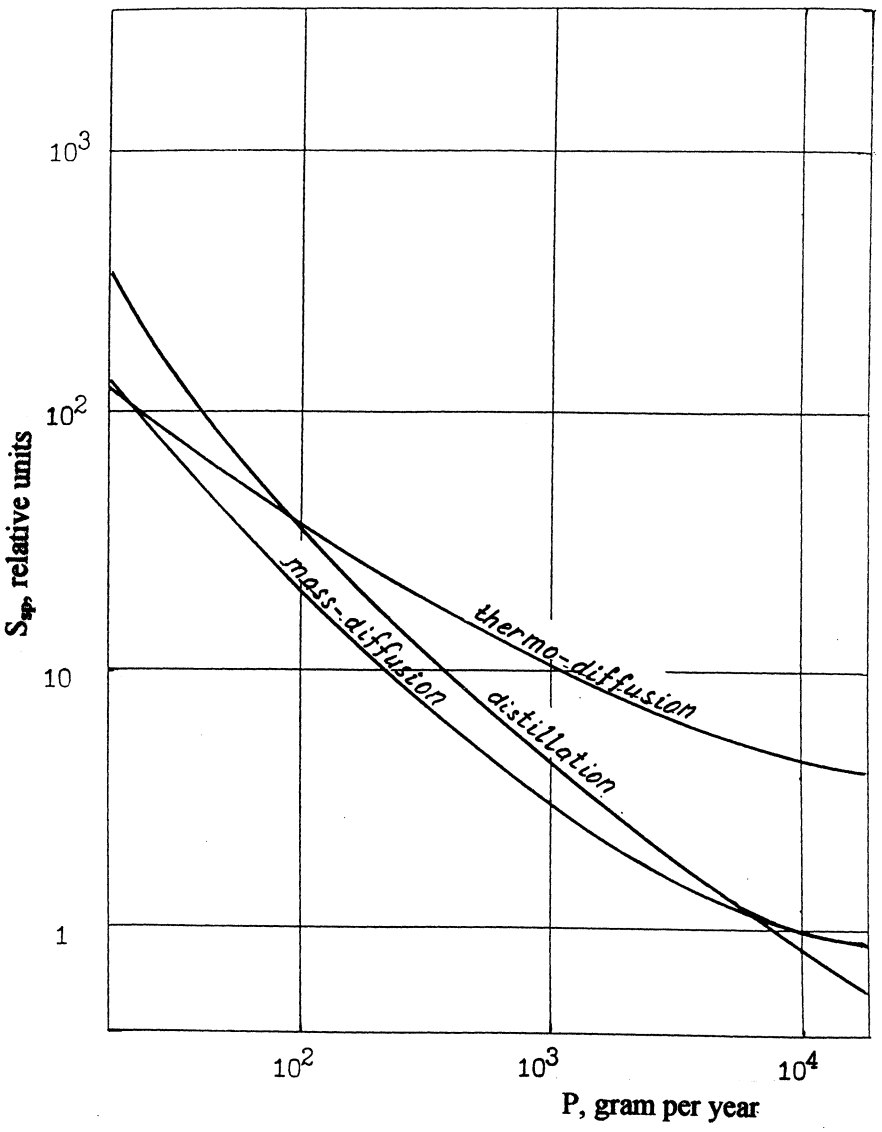

Figure 17. Comparison of different separating methods (thermo-diffusion, distillation, and mass-diffusion) for production of 95% ^{22}Ne.

COMPARISON OF MASS-DIFFUSION WITH OTHER SEPARATION METHODS

The proposed specific cost function can be applied not only to search for optimum operating conditions and optimum value of geometrical parameters of mass-diffusion element, but also to evaluate efficiency of mass-diffusion process in comparison with other separation methods. Such a comparison was carried out through the evaluation of specific cost functions for production of identical quantities of isotope product with enrichment of 95% ^{22}Ne by mass-diffusion, distillation and thermo-diffusion techniques. The comparison was carried out with accounting for the value factors (see Eq. 22), which are considered to be typical for the thermo-diffusion and distillation methods.

The results of calculations are given in Fig. 17 in the form of the dependence of specific cost on the values of product flow corresponding to enrichment of neon-22 isotope to not lower than 95%. As follows from the analysis of dependencies shown in Fig. 17, the enrichment of ^{22}Ne isotope by mass-diffusion method occupies an intermediate position between thermo-diffusion and distillation techniques in terms of their comparative efficiency in the range of product flows from 50 g/year to 8 kg/year. Note that the results under discussion are just estimates, however this approach can be applied to select an appropriate separation method for a particular scale of isotope production.

The analysis of dependence of specific cost on parameters of mass-diffusion element also permits distinguishing the main routs for decreasing the operating and the hardware costs in the production of a given isotope product. For example, it seems reasonable to use in certain instances the spent water steam or other energy sources of industrial installations to decrease the total energy consumption. The water steam can be used either directly as a diffusion media or for the heating up of the working liquid. In the first case it is necessary, however, to take into account the probable decrease of the enrichment coefficient.

The other way of providing the decrease of the specific cost value is the perfection of mass-diffusion stages. The fact is that the productivity of the considered mass-diffusion element is still limited as the increase of perimeter of the diaphragm results in deviation of the gas flow from uniformity in cavity 1 which decreases the element's separative power.

The increase of separative power of the element can be achieved by introducing in its design a device to supply a part of the separated mixture into the internal cavity of the diaphragm. This allows the elimination of the reverse gas transfer through the diaphragm and the creation of a four-line scheme for the mixture flux in the element (26). This improvement of the mass-diffusion element results in an increase of its separative power by more than 20%. The use of elements of the above design allows decreasing both the hardware and the operating costs connected with the isotope production.

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NOMENCLATURE

c	concentration of target isotope, mol fraction
D_{10}, D_{20}	diffusion coefficient of isotopes to be separated in vapor, cm^2/s
D_{12}	mutual diffusion coefficient of the components of mixture to be separated, cm^2/s
D	effective diffusion coefficient, cm^2/s
d_c	capillary diameter, cm
F	feed flow rate in cascade, mol/s
F_0	area of aperture diaphragm, cm^2
F_1	part of the diaphragm surface per aperture, cm^2
G	flow of mixture to be separated in arbitrary section of element, mol/s
H	height of the diaphragm and main condenser, cm
H_b	height of additional condenser, cm
L	flow of mixture to be separated entering element, mol/s
$L' = \theta L$	light fraction flow, mol/s
l_c	length of capillary, cm
$\ln q = ul/nD_{10}$	the Peclet's diffusion number
l_e	diffusive resistance of the diaphragm, cm
m_1	molecular mass of the light component, g/mol
m_2	molecular mass of the heavy component, g/mol
n	density of gas-vapor mixture, mol/cm^3
P	product flow rate, mol/s
P_0	initial pressure in element, torr
P_p	working pressure in element, torr
Q_0	vapor flow entering element, mol/s
Q_t	vapor flow passing through diaphragm, mol/s
R	universal gas constant
S, S_e, S_a	specific cost, operational and hardware components costs respectively, relative units
T_p	working temperature in element, K
T_{ct}	temperature of condensing surfaces, K
t_c	service life of separative apparatus, years
u	density of gas-vapor mixture flow through the diaphragm, $mol/(cm^2 \cdot s)$
V_0, V_{com}	volume of element and communications, cm^3
W	waste flow rate, mol/s
x, y	cross and longitudinal co-ordinates
α, δ	internal transfer characteristics of mixture in element (see Eqs. 10 and 11)

δU	separative power of element, mol/s
γ_0	gas concentration on condensing surfaces, mol fraction
γ_a, γ_b	gas concentration in gas-vapor mixture in external backlash and internal cavity of the element, respectively, mol fraction
γ	current value of gas concentration in gas-vapor mixture, mol fraction
$\varepsilon_0 = (D_{10} - D_{20})/D_{10}$	elementary enrichment coefficientε
ε	enrichment coefficient of element
μ_g, μ_v	molecular mass of gas and vapor, respectively, g/mol
η	viscosity of mixture to be separated, g/(cm·s)
η_f	form factor of cascade
θ	cut number
θ_v	vapor distribution coefficient
Π	perimeter of diaphragm, cm
τ	density of gas flow , mol/(cm^2·s)
τ_1	density of molecules flow of allocated isotope, mol/(cm^2·s)

The indexes x and y denote the cross and longitudinal components of values, respectively. The other designations are explained in the text.

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