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Separation of the Isotopes of Uranium by the Separation Nozzle Process

BY E. W. BECKER, K. BIER, W. BIER, R. SCHÜTTE, AND D. SEIDEL [*]

The separation nozzle process is based on the partial spatial separation of components of different mass in an expanding supersonic jet stream. The process is of especial interest for the separation of uranium isotopes. Details of a systematic experimental determination of the most favorable operating conditions for such a separation are given and the construction and testing of a closed circulation system, the basic unit of a ten membered pilot cascade separator for uranium isotope separation, is described. The optimum values of the specific cost factors obtained experimentally for the separation nozzle process are compared with the corresponding values estimated for the gaseous diffusion process.

Introduction

The light isotope of uranium, ^{235}U, required for the production of nuclear power, is currently enriched on a large scale by means of the gaseous diffusion process [1]. This method is based on the difference between the velocities with which $^{235}UF_6$ and $^{238}UF_6$ molecules migrate through porous membranes. It is also possible to separate the isotopes of uranium with gas centrifuges [2]; however, as far as is known, no such production facilities have as yet been constructed.

In the course of the past few years we have developed a process for the separation of the isotopes of uranium which avoids both the problems of the delicate membranes required by the diffusion process and also

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[1] *E. W. Becker*, Chemie-Ing.-Techn. *29*, 365 (1957).
[2] *W. Groth*, Chemie-Ing.-Techn. *31*, 310 (1959).

those of the high mechanical stresses which occur in the centrifuge process [3–10]. This process is based on the partial spatial separation of the components of a gaseous mixture that takes place in an expanding supersonic jet. The separating effect of the jet may be explained qualitatively by the fact that the streamlines of an expanding jet are curved and, as in a centrifuge, the isotopic mixture is subjected to the action of a centrifugal acceleration, that is, to the pressure gradient which arises as a result of the centrifugal forces.

Figure 1a is a sketch of the arrangement which was used initially in the earlier experiments [3–5]. A convergent nozzle with either a circular or slit-shaped cross section was used in these experiments. A diaphragm, either conical or V-shaped, depending on the nozzle cross section, and having an aperture the same shape as that of the nozzle opening, was positioned directly in front of the nozzle. The diaphragm divided the jet emerging from the nozzle into two fractions, a core fraction and a peripheral fraction, which were then pumped off separately from the separating element. The peripheral gas fraction is enriched in the light component of the mixture when the streamlines are curved away from the axis of the jet. When a reversal in the curvature of the streamlines occurs, as is the case in the recompression region of an over-

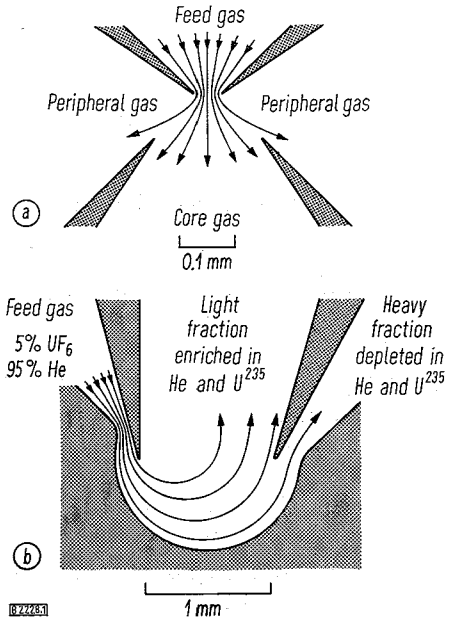

Fig. 1. Cross sections of the separation nozzle arrangements used in the earlier experiments (with a schematic representation of the streamlines):
a) a freely expanding jet from a circular or slit-shaped nozzle.
b) a UF_6 jet with helium as the auxiliary gas deflected by a curved wall.

expanded jet, the sign of the separation effect can also change [6].

Experiments with uranium hexafluoride as the process gas, using the arrangement pictured in Figure 1a, showed that the elementary separation effect for the nozzle process could, in fact, be somewhat larger than that for the gaseous diffusion process [5]. However, due to the higher expansion ratio required and to the appreciably lower absolute pressure which had to be used, the method, in this form, could only be economically competitive with the gaseous diffusion process in certain special cases.

A theoretical investigation of the separating processes taking place in supersonic jets [4, 7] indicated that the separation effect should increase if the angle through which the streamlines turn in the region between the nozzle and the diaphragm could be increased and if the Mach number of the jet could be increased.

An attempt to improve the economic potential of the process by increasing the angle through which the streamlines turn with the aid of a curved wall [8] met with little success. In the case of a mixture of argon isotopes alone, the mechanical deflection of the jet exhibited no clear economic advantages. This could be explained by the fact that, in this case, the increase in the angle of deflection of the streamlines which could be realized was relatively small, while, on the other hand, the decrease in the Mach number, caused by the entropy-producing effects associated with deflecting the jet, was relatively large.

An attempt to increase the Mach number of the isotopic mixture being separated by adding a light auxiliary gas in large molar excess was more successful [9]. The experimental arrangement used was that shown in Figure 1a. In the case of the argon isotopes, both the separation effect and the absolute pressure could be increased considerably, while the expansion ratio required could be decreased considerably. In spite of the necessity of compressing the light auxiliary gas along with the isotopic mixture, an appreciable improvement in the economic efficiency of the process was obtained.

However, when it came to the separation of the isotopes of uranium, the use of a light auxiliary gas with the experimental arrangement of Figure 1a appeared to be of questionable merit. Although the specific cost factors (defined below) which reflect the investment costs of the process could be reduced by more than a factor of four by the addition of 400 mole per cent of helium to the UF_6, the energy requirement increased by about the same factor [10]. This result could be explained by the fact that the large separation that occurred between the isotopic mixture and the light auxiliary gas caused a decrease in the angle of deflection of the streamlines of the isotopic mixture.

Thus, in the case of deflecting the jet with a curved wall, the advantage of the greater angle of deflection could only be obtained along with the accompanying disadvantage of having a smaller Mach number; while, in the case of adding a light auxiliary gas without deflecting the jet, the advantage of the larger

[3] *E. W. Becker*, *K. Bier*, and *H. Burghoff*, Z. Naturforschg. *10a*, 565 (1955).
[4] *E. W. Becker*, *W. Beyrich*, *K. Bier*, *H. Burghoff*, and *F. Zigan*, Z. Naturforschg. *12a*, 609 (1957).
[5] *E. W. Becker* and *R. Schütte*, Z. Naturforschg. *15a*, 336 (1960)
[6] *K. Bier*, Z. Naturforschg. *15a*, 714 (1960).
[7] *F. Zigan*, Z. Naturforschg. *17a*, 772 (1962).
[8] *E. W. Becker*, *H. Burghoff*, and *J. Gspann*, Z. Naturforschg. *16a*, 955 (1961).
[9] *E. W. Becker*, *K. Bier*, and *W. Bier*, Z. Naturforschg. *17a*, 778 (1962).
[10] *E. W. Becker*, *K. Bier*, *W. Bier*, and *R. Schütte*, Z. Naturforschg. *18a*, 246 (1963).

Mach number of the isotopic mixture was counteracted by the accompanying disadvantage of a smaller angle of deflection. It therefore appeared promising to combine the two types of operation. With the type of operation shown in Figure 1b, it was, in fact, possible to achieve a considerable decrease in the investment costs for separating the uranium isotopes without an appreciable increase in the operating costs [10]. Further development work was therefore concentrated on the method combining the use of a light auxiliary gas with the mechanical deflection of the jet.

In order to obtain criteria for the optimum shape of the separating system without performing time consuming separation experiments, Schlieren investigations were carried out using an experimental arrangement corresponding to that shown in Figure 1b [11]. It was assumed that a deflection of the jet as free as possible from disturbances would be the optimum for the separation process. Figure 2 shows examples of the formation of a jet, using nitrogen as a test gas, produced in one case by a convergent nozzle and in the other case by a curved Laval nozzle. It is evident from the figure that in the case of the convergent nozzle both shock waves and boundary layer separation occurred, while the curved Laval nozzle, on the other hand, produced an essentially disturbance-free, curved jet.

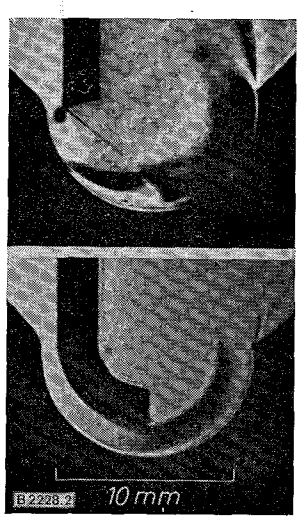

Fig. 2. Schlieren photographs of a nitrogen jet deflected through 180° produced by a convergent nozzle (above) and by a curved Laval nozzle (below). Inlet pressure, $p_0 = 258$ mm Hg; Expansion ratio, $p_0/p_M = 12.3$.

1. Determination of the Optimum Operating Conditions

On the basis of the Schlieren investigations and preliminary experiments with $He-UF_6$ mixtures, the separating elements shown in Figure 3 were selected for further experimentation. The two elements have essentially the same shape, but differ by a factor of two in the characteristic dimensions of the nozzle and of the deflecting surface (see Table 1).

[11] *K. Bier, V. Lippig,* and *K. Schlegel,* to be published.

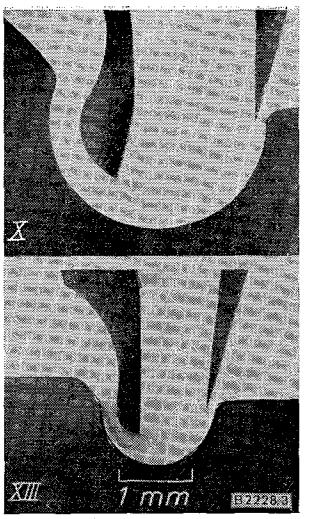

Fig. 3. Side views of the separation nozzle arrangements, X and XIII, used in the experiments for evaluating the optimum operating conditions (see Table 1).

Table 1. The characteristic dimensions of the separating systems investigated. The effective length of the nozzle slit (perpendicular to the plane shown in Figure 3) was 10 mm for both systems.

System	Width of nozzle at throat, a (mm)	Width of nozzle at mouth (mm)	Radius of curvature of the deflecting surface (mm)
X	0.40	0.60	1.5
XIII	0.20	0.30	0.75

1.1. Apparatus and Method of Evaluation

The separation experiments for the determination of the most favorable operating conditions were carried out with the apparatus shown schematically in Figure 4.

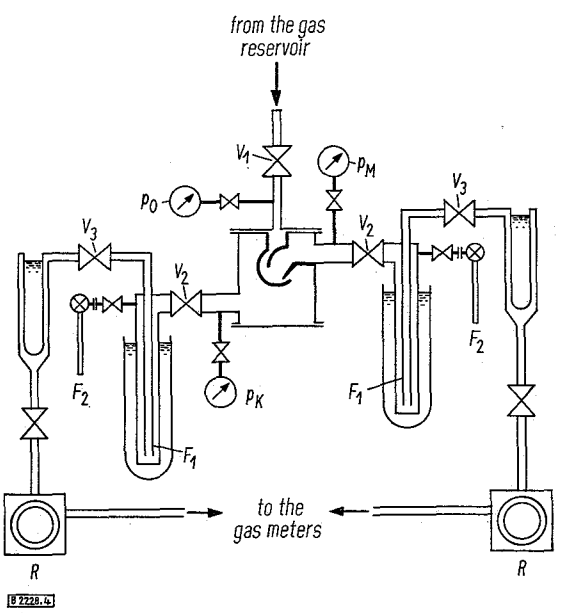

Fig. 4. Schematic diagram of the separation apparatus. F, cold traps; R, rotary pumps; V, valves.

The mixture of UF_6 and auxiliary gas which had been charged to the gas reservoir flowed through the regulating valve, V_1, to the nozzle of the separating system. The pressures, p_M and p_K, of the light and

heavy fractions, respectively, were adjusted to the desired values by means of the valves, V_2. Three diaphragm-type manometers were provided for the measurement of the pressures, p_0, p_M, and p_K. The UF_6 portions of the light and heavy gas fractions were precipitated out in the cold traps, F_1, which were cooled with liquid nitrogen. The cold traps could be isolated from the rotary pumps, R, by means of the valves, V_3.

The cut of the auxiliary gas (of the helium, for example), defined by

$$\theta_{He} = \frac{\text{flow rate of He in the light fraction}}{\text{flow rate of He in the feed gas}}, \quad (1)$$

was determined by means of gas meters installed on the high pressure side of the pumps. The cut of the UF_6 gas,

$$\theta_{UF_6} = \frac{\text{flow rate of } UF_6 \text{ in the light fraction}}{\text{flow rate of } UF_6 \text{ in the feed gas}}, \quad (2)$$

was determined by weighing the two UF_6 fractions which had been collected in the cold traps, F_1, and then reprecipitated in the pipettes, F_2.

The separation factor between the helium and the UF_6, A, was obtained from the cuts defined above by means of the relationship:

$$A = \theta_{He}(1-\theta_{UF_6})/\theta_{UF_6}(1-\theta_{He}) \quad (3)$$

The elementary separation effect for the uranium isotope separation, ε_A, is defined by

$$\varepsilon_A = [n_M(1-n_K)/n_K(1-n_M)]-1, \quad (4)$$

where n_M and n_K are the mole fractions of the light uranium isotope in the UF_6 isotopic mixture in the light and heavy fractions, respectively. The concentrations, n_M and n_K were determined mass spectrometrically [*].

1.2. Bases for the Optimization

In order to make practical use of the process, the elementary separation effect must be multiplied by connecting a large number of separating elements together to form a separation cascade. The elements should be connected in such a way that only streams with the same isotopic abundance ratio are mixed together in the cascade [12]. A separation of the auxiliary gas, accomplished, for example, by first enriching it with several stages of the separation nozzle process itself and then by freezing out the remaining UF_6, will be required only where the gas throughput changes, that is, only at the end of each square section in the cascade. The auxiliary gas which is separated at the top of a cascade section can be recycled and added to the UF_6 at the bottom of the cascade section.

Since a square cascade section consists, in practice, of many identical separating elements connected in series, the cost of separating the auxiliary gas, which needs to be done only at the top of the section, can be neglected in comparison with the other cascade costs. The economic efficiency of the process, using a light auxiliary gas, can therefore also be characterized by the previously defined "specific cost factors" [4,9].

Since, in the experimental work described in this report, the suction pressures, p_M and p_K, of the light and heavy fractions were always equal, the following simplified expressions could be used:

1) the specific ideal isothermal work of compression,

$$E_s = (1/\delta U)\, LRT \ln(p_o/p_M), \quad (5)$$

2) the specific suction volume,

$$V_s = (1/\delta U)\, LRT/p_M, \quad (6)$$

3) and the specific slit length,

$$l_s = (1/\delta U)\, l, \quad (7)$$

where δU is the separative power [12], given by

$$\delta U = \frac{L^*}{2}\varepsilon_A^2\, \theta_{UF_6}\, (1-\theta_{UF_6}), \quad (8)$$

L is the throughput of the gas mixture, L^* is the UF_6 throughput, and l is the slit length of the separating element.

The quantity, E_s, is a measure of the power requirement of the process; the quantity, V_s, is a measure of the investment costs of the compressors, piping, and valves; and the quantity, l_s, is a measure of the investment costs of the separating elements. From an investigation of the dependence of these three cost factors on the experimental parameters, the most favorable operating conditions for the process can be determined to within reasonable limits.

1.3. Experimental Results

The following parameters were varied during the experiments: the type of auxiliary gas, the mole fraction of the UF_6 in the mixture, the inlet pressure, the size of the separation system, the expansion ratio, and the distance between the knife edge and the curved wall (see Figure 1b or 3).

1.3.1. Type of Auxiliary Gas

Nitrogen, neon, and helium were tried as the auxiliary gas using separation system X (see Table 1). The UF_6 concentration was 10 mole per cent in these experiments. First, by varying the inlet pressure, p_0, with the expansion ratio, $p_0/p_M = 4$, held constant, the inlet

[*] Mass Spectrometer, Type 21–220 A, manufactured by CEC, Pasadena, California (U.S.A.).

[12] *K. Cohen*: The Theory of Isotope Separation as Applied to the Large Scale Production of U^{235}. McGraw-Hill Book Co., New York 1951.

pressure which yielded the minimum specific suction volume was determined for each of the three mixtures. The optimum values obtained for p_0 were: 16 mm Hg for the nitrogen-UF_6 mixture, 24 mm Hg for the neon-UF_6 mixture, and 32 mm Hg for the helium-UF_6 mixture. Next, the expansion ratio was varied with the inlet pressure held constant at its optimum value. From the results of the series of experiments presented in Figure 5, it can be seen that the smallest values of the specific cost factors were obtained with helium as the auxiliary gas. For this reason, only helium was used as the auxiliary gas in the experiments which followed [*].

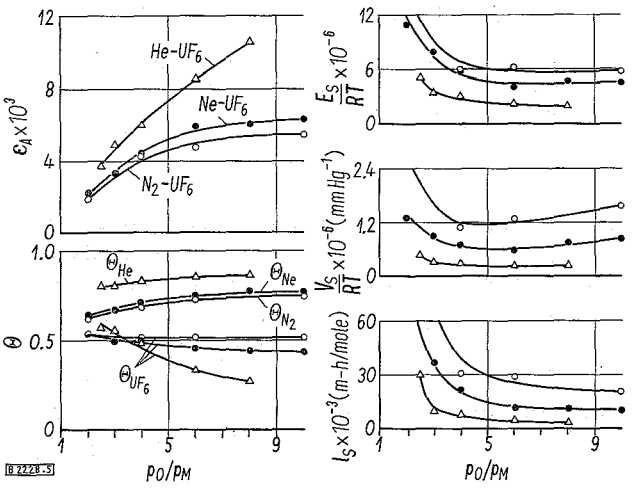

Fig. 5. The effect of different auxiliary gases on the specific cost factors. Inlet pressure: p_0 = 16 mm Hg for the N_2—UF_6 mixture, p_0 = 24 mm Hg for the Ne—UF_6 mixture, and p_0 = 32 mm Hg for the He—UF_6 mixture; Separation system X; Distance between the knife edge and the curved wall: 0.19 mm for the He—UF_6 mixture and 0.29 mm for the N_2—UF_6 and the Ne—UF_6 mixtures; The UF_6 concentration was 10 mole per cent in all cases.

1.3.2. UF_6 Mole Fraction

Mixtures containing 3, 5, and 10 mole per cent UF_6 were investigated using separation system X. The optimum inlet pressure for each of the three mixtures was again determined first by varying the inlet pressure, p_0, with the expansion ratio, p_0/p_M = 4, held constant. The optimum values obtained for p_0 in these cases were: 60 mm Hg for the 3 % UF_6 mixture, 48 mm Hg for the 5 % UF_6 mixture, and 32 mm Hg for the 10 % UF_6 mixture. Next the expansion ratio was again varied with the inlet pressure held constant at its optimum value. It is evident from the results shown in Figure 6 that the specific suction volume, V_s, and the specific slit length, l_s, decrease monotonically with decreasing UF_6 content in the region investigated. However, as can be seen, the gain in going from the 5 % mixture to the 3 % mixture is rather slight. Therefore, since the 5 % mixture exhibits a somewhat smaller specific work of compression, E_s, the subsequent experiments were carried out using a 5 % UF_6 mixture.

[*] More favorable results can be obtained with hydrogen as the auxiliary gas. However, since H_2 and UF_6 can react chemically with each other, the advisability of using hydrogen as the auxiliary gas is questionable.

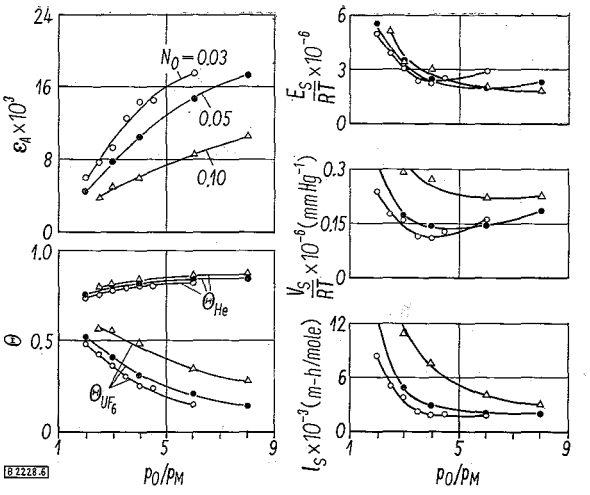

Fig. 6. The effect of the UF_6 mole fraction, N_0, on the specific cost factors.

Helium-UF_6 mixtures; Inlet pressure: p_0 = 60 mm Hg for the 3 % UF_6 mixture, p_0 = 48 mm Hg for the 5 % UF_6 mixture, and p_0 = 32 mm Hg for the 10 % UF_6 mixture; Separation system X; Distance between the knife edge and the curved wall: 0.19 mm.

1.3.3. Inlet Pressure and Size of the Separation System

The effect of the inlet pressure was investigated using separation systems X and XIII with the expansion ratio, p_0/p_M = 4, held constant. It can be seen from Figure 7, where the abscissa is the product of the inlet pressure, p_0, and the nozzle width, a, that both separation systems attain their optimum separating properties at about the same value of the $p_0 a$ product. The minimum values of E_s and l_s for both systems are

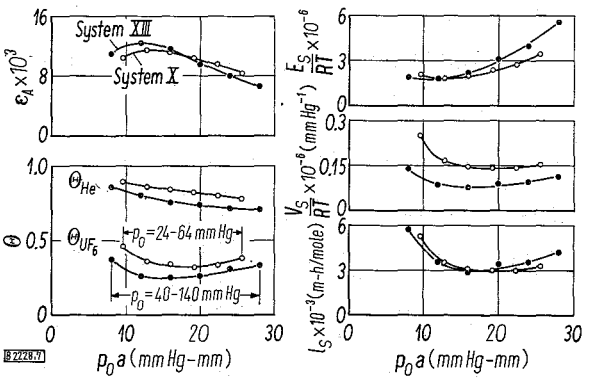

Fig. 7. Effect of the inlet pressure, p_0, and the size of the separation system on the specific cost factors.

Separation system X: width of nozzle at throat, a = 0.4 mm, distance between the knife edge and the curved wall = 0.19 mm.

Separation system XIII: width of nozzle at throat, a = 0.2 mm, distance between the knife edge and the curved wall = 0.11 mm.

Expansion ratio: p_0/p_M = 4; Helium-UF_6 mixture containing 5 mole % UF_6.

essentially the same. However, the minimum specific suction volume for the smaller system, which occurs at a higher value of p_0, is only about one half as large as it is for the larger system. For this reason further work was carried on with the smaller separation system.

1.3.4. Expansion Ratio

The effect of varying the expansion ratio with the inlet pressure held constant at the value which yielded the minimum specific suction volume, $p_0 = 80$ mm Hg, is shown in Figure 8. It can be seen that the minimum

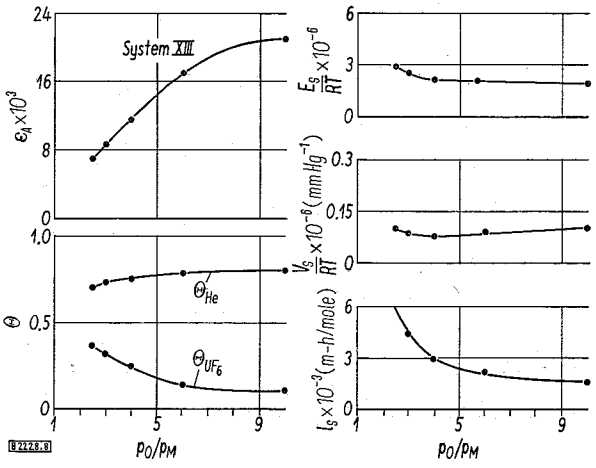

Fig. 8. The effect of the expansion ratio, p_0/p_M, on the specific cost factors.
Inlet pressure: $p_0 = 80$ mm Hg; Separation system XIII; Distance between the knife edge and the curved wall: 0.11 mm; Helium—UF_6 mixture containing 5 mole% UF_6.

values of E_S and V_S are obtained at an expansion ratio equal to about four. The specific slit length, l_S, continues to decrease with further increases in p_0/p_M. However, since the contribution which l_S makes to the cost of product seems to be of less importance than the contributions of the other two specific cost factors, the expansion ratio, $p_0/p_M \approx 4$, can be regarded as an optimum value.

1.3.5. Distance between the Knife Edge and the Curved Wall

It has already been mentioned that in an isotope separation cascade only streams with the same isotopic concentration should mix. In order for this condition to be satisfied, a different type of cascade connection is required for each different value of the UF_6 cut [11]. Economically satisfactory cascade arrangements are to be expected only when θ_{UF_6} can be expressed as the ratio of two small integers. The value, $\theta_{UF_6} = 1/2$, is a particularly good one as it is associated with the simplest type of cascade connection.

In the experiments described above, the minimum values of the specific cost factors were determined without regard to the θ_{UF_6} value which occurred. The cut, $\theta_{UF_6} = 1/2$, was obtained, if at all, only with relatively unsatisfactory values of the specific cost factors. In order to obtain the minimum values of the specific cost factors in conjunction with practical values of the cut, the effect of the expansion ratio was once again investigated, with the inlet pressure, $p_0 = 80$ mm Hg, again held constant, but with the distance between the knife edge and the curved wall a variable. The curves for constant values of θ_{UF_6}, shown in

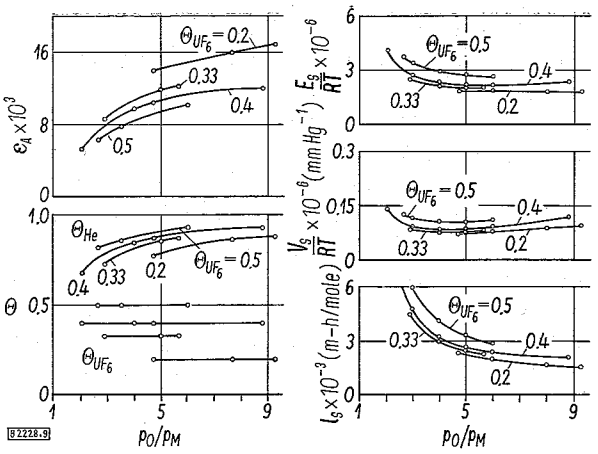

Fig. 9. The effect of the UF_6 cut, θ_{UF_6}, on the specific cost factors. The curves were obtained by varying the distance between the knife edge and the curved wall and interpolating between the measured results. Inlet pressure: $p_0 = 80$ mm Hg; Separation system XIII; Helium—UF_6 mixture containing 5 mole% UF_6.

Figure 9, were obtained by interpolating between the measured values. It can be seen that the smallest values of the specific cost factors were obtained at the smallest value of the cut which was investigated ($\theta_{UF_6} = 0.2$). Between $\theta_{UF_6} = 0.2$ and $\theta_{UF_6} = 0.33$, the cost factors increase only slightly; on the other hand, between $\theta_{UF_6} = 0.33$ and $\theta_{UF_6} = 0.5$, they increase by a factor of about 1.4. Since the cost of piping and valves should be only slightly greater for a cascade operating with the cut, $\theta_{UF_6} = 0.33$, than for a cascade operating with a cut of one half, the value, $\theta_{UF_6} = 0.33$, will be assumed to be the optimum value of the cut for the separation nozzle process.

The optimum values of the specific cost factors resulting from the investigations described above and the associated operating conditions are compared in Table 2 with the corresponding, previously published values [10]. It can be seen that the specific work of compression and the specific suction volume have been reduced by almost a factor of two by the improvement of the separating element. The decrease in the optimum value of the compression ratio from 10 to 4 is also advantageous since it reduces the number of stages required in the compressors. The increase in the optimum suction pressure, $p_M = p_K$, from 12 to 20 mm Hg should also serve to reduce the relative pressure losses in the cascade piping.

Table 2. Comparison of the optimum values of the specific cost factors and associated operating conditions obtained with the improved separating system with the corresponding values from the previous investigation [10]. In both cases the UF_6 content of the He—UF_6 mixture was 5 mole per cent and the radius of the curved deflecting wall 0.75 mm. A temperature of 295 °K was assumed for the calculation of E_S and V_S.

	Present results	Previous results [10]
Specific work of compression, E_S (kWh/mole)	14.4×10^2	26.6×10^2
Specific suction volume, V_S (m³/mole)	1.40×10^6	2.61×10^6
Specific slit length, l_S (m-h/mole)	3.0×10^3	3.2×10^3
Compression ratio, p_0/p_M	4	10
Suction pressure, $p_M = p_K$ (mm Hg)	20	12
Elementary separation effect, ε_A	10.6×10^{-3}	10.6×10^{-3}
UF_6 cut, θ_{UF_6}	0.33	0.31

2. Testing the Process in a Closed Test Loop

In a separation cascade, the process gas containing the UF_6 which is expanded in one separating element must be recompressed by UF_6-resistant compressors before entering the neighboring elements. It must be demonstrated that such a cascade will be gas dynamically stable. Since a theoretical treatment of this problem is not sufficient for the design of large installations, a ten stage pilot plant is currently under construction in our institute. The construction and testing of the prototype unit for this pilot plant are described below.

2.1. Separating Element

Figure 10 shows a cross section of the separation nozzle system used and gives the nominal values of the pressures, p_0, p_M, and p_K, for these experiments. L moles per hour of a gaseous mixture consisting of 5 mole per

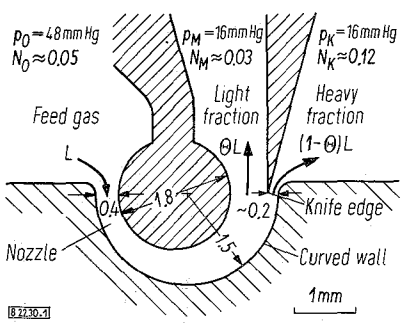

Fig. 10. Cross section of the separation nozzle system used in the closed test loop.

N_0, N_M, and N_K are the mole fractions of the UF_6 in the helium -UF_6 mixture used as the process gas; L is the molar flow rate of the process gas; θ is the process gas cut.

cent UF_6 and 95 mole per cent helium were expanded through the curved Laval nozzle. The nozzle inlet pressure, p_0, was 48 mm Hg. The nozzle consisted of a curved wall with a radius of curvature, r = 1.5 mm, and a solid cylinder, eccentric to the wall, 1.8 mm in diameter. This model of the curved Laval nozzle has the advantage, compared with the nozzles investigated in detail in Section 1, of a simpler fabrication process for producing the longer separating systems. The width of the nozzle at its narrowest point, or throat, was 0.40 mm. The gas mixture expanded in the nozzle to the pressure, p_M = 16 mm Hg. These dimensions and operating conditions were fixed on the basis of the experiments described in Section 1 and from a consideration of the capability of the compressors which were available. The nozzle had a length of 100 mm in the direction perpendicular to the plane shown and was therefore ten times as long as the nozzles used in the earlier investigations.

The details of the assembly of the individual parts can be seen from Figure 11. In order to carry out all the testing desired, a scheme of construction was chosen which offered extensive possibilities for adjusting and changing all of the parameters affecting the geometry of the jet. The plate (b) bearing the nozzle cylinder (c) and the plate (f) terminating in

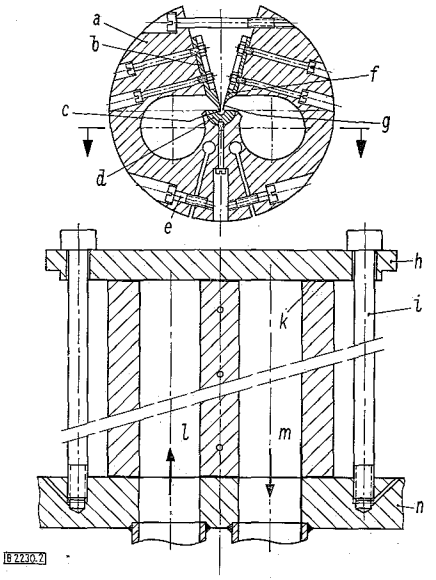

Fig. 11. Details of the separating element assembly. The separation nozzle system consisting of the curved wall (d), the cylinder (c), and the knife edge (g), are shown enlarged in Fig. 10.

(a) mounting block
(b) nozzle cylinder holder
(c) nozzle cylinder
(d) curved wall assembly
(e) adjusting screws
(f) knife edge holder
(g) knife edge
(h) blind flange
(i) bolts
(k) teflon film
(l) feed gas inlet
(m) heavy fraction outlet
(n) separation chamber cover

the knife edge were bolted securely to the machined surfaces of the brass cylinder (a). The plates (b) and (f) were fabricated from hardened tool steel (90 MnV8) and were, as was the brass cylinder (a), nickel-plated after the final machining. The positions of the nozzle cylinder (c) and of the knife edge (g) with respect to the curved wall (d) could be changed either by shifting the plates on their supporting surfaces or by extending the slotted brass cylinder (a) by means of the screws (e) provided for this purpose [*].

The separating element was bolted, by means of a blank flange (h) and the bolts (i), to the cover flange (n) of a cylindrical chamber, not shown, which surrounded the separating element. The sealing between the plane ground surfaces was accomplished with 0.1 mm thick teflon film (k). The light fraction passed between the nozzle plate (b) and the knife edge plate (f) into the chamber surrounding the element and was pumped off from there.

2.2. Compressor

In the construction of a plant utilizing the separation nozzle process, only compressors of the axial or centrifugal type ought to be considered. However, in making the choice of the type of compressor for the pilot plant, it had to be kept in mind that the plant should operate at the lowest practicable throughput in order to keep the cost of the installation low. A reasonable compromise was achieved with the use of Roots compressors. They can be constructed to have the desired, relatively small gas flow rate of about 100 m^3/h without much additional development work being required, and the dependence of their operating characteristics on the composition and physical prop-

[*] In practice, separating systems in which the critical dimensions are fixed at sufficiently close intervals along the length of the system would be used.

erties of the process gas is similar to that for centrifugal and axial compressors.

The Roots compressor used in these investigations has vacuum-tight shaft seals in the form of sealing chambers which are closed off by mechanical, radial-face seals at each end and through which flows a UF_6-resistant sealing oil. The compressor is provided with a built-in gas cooler. Additional internal cooling of the impellers is also possible. The compressor was developed in cooperation with the firm, E. Leybolds Nachfolger, Köln-Bayental, from the commercial Roots compressors designated "Ruvac E 126" [*]. All special elements in the construction were tested in our institute under process conditions and brought step by step to their final form.

The welded compressor housing is closed off on both sides by two face plates which contain four individual bearing assemblies which support the two impellers of the compressor. In each of the four bearing assemblies are a pair of the radial-face seals. They form four sealing chambers through which flows a sealing agent (KEL-F oil) which is maintained at a pressure higher than the surroundings. A water-cooled, spiral-tube cooler is built into the square-cornered gas outlet. If needed, the sealing oil can be conducted from one face plate to the other through the cooling chambers of the impellers to provide additional cooling. The compressor is driven by a three phase electric motor which turns at 2900 rpm. At this speed the compressor has a geometrical suction volume of 380 m^3/h.

The impellers are of stainless steel No. 4571. The face plates, the welded compressor housing, and all of the other parts which come into contact with the process gas were fabricated from stainless steel No. 4541. The sealing between the metal parts exposed to the process gas was accomplished with O-rings of Viton, a fluoroelastomer, which were inserted into slots provided for that purpose.

The radial-face seals have a layer of Stellite on their sliding surfaces. During the life testing of the compressor, values between 0.5 and 1.8 cm^3/d per compressor were obtained for the amount of sealing oil entering the process gas space. This inleakage is drawn off into a water-cooled oil collector. A vapor seal is used to prevent entry of the sealing oil vapor into the process gas region of the compressor.

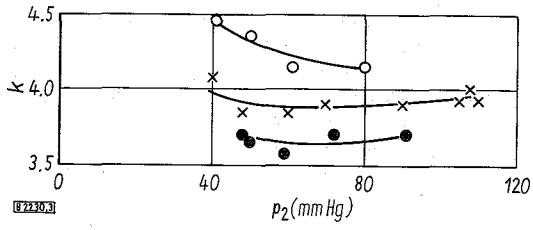

Fig. 12. The dependence of the compression ratio, $k = p_2/p_1$ of the Roots compressor on the discharge pressure, p_2, with the molar throughput, L, as a parameter.

Gas cooler with a cooling water flow rate of 2 l/min;
○ ○ L = 28 mole/h; ✕ ✕ L = 43 mole/h; ● ● L = 60 mole/h;
Helium-UF_6 mixture containing 5 mole% UF_6.

In order to evaluate the performance of the process compressors, the dependence of the throughput, L (moles/h), and of the compression ratio, $k = p_2/p_1$, on the discharge pressure, p_2, was measured using a UF_6-helium mixture containing 5 mole per cent UF_6. The measurements were made after the steady state temperature distribution was attained throughout the compressor. It can be seen from Figure 12 that at the specified operating point, that is, at a throughput of 60 moles/h and at 48 mm Hg discharge pressure, the compression ratio exceeds the required value of 3 by more than 20 per cent.

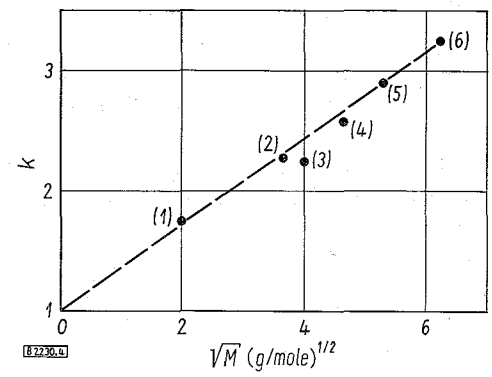

Fig. 13. The dependence of the compression ratio, k, on the square root of the molecular weight, M, of the process gas. Roots compressor without gas cooler;

Discharge pressure: p_2 = 50 mm Hg;
Molar throughput: L = 60 mole/h;
Data points:
(1) He, M = 4
(2) 2.7 mole %UF_6—97.3 mole % He, M = 13.5,
(3) CH_4, M = 16,
(4) 5 mole % UF_6—95 mole % He, M = 21.4,
(5) N_2, M = 28,
(6) 10 mole % UF_6—90 mole % He, M = 38.8

Measurements of the throughput, made with an earlier model compressor without a gas cooler, for various process gases of different densities, showed a linear relationship between the square root of the average molecular weight of the gas and the compression ratio, k, which the compressor achieved at a given molar throughput, L, and discharge pressure, p_2. The measured results for L = 60 moles/h and p_2 = 50 mm Hg are shown in Figure 13.

2.3. Process Test Loop

The construction of the process test loop can be seen from Figure 14. The heavy and light fractions are removed from the separation chamber through separate lines and are pumped off simultaneously through the valves (c) and (b), respectively, by the Roots compressor (h). The compressed gas is returned to the separating element (i) through valve (e). Sample withdrawal reservoirs (k) are connected in parallel with the process gas lines. Their volumes were so determined that they contain, under operating conditions, a sufficient amount of UF_6 (about 0.1 g) for isotopic analysis with a mass spectrometer. During an experiment, they were swept out with process gas and then isolated from the system before the withdrawal of the sample. The samples were pumped from these reservoirs through cold traps (not shown) in which the UF_6 was frozen out. The UF_6, after being weighed, was then subjected to isotopic analysis. The UF_6 concentration in the UF_6-helium mixture was determined from the weight of the UF_6, the pressure, and the volume of the sample withdrawal reservoirs.

In order to facilitate a longer period of operation with frequent sample withdrawal, a 1000 l reservoir (l) can be connected to the system to increase its volume. Without the

[*] We wish to thank *W. Röllinger* and *H. Bode* of the firm, E. Leybolds Nachfolger, Köln-Bayental, for their valuable cooperation.

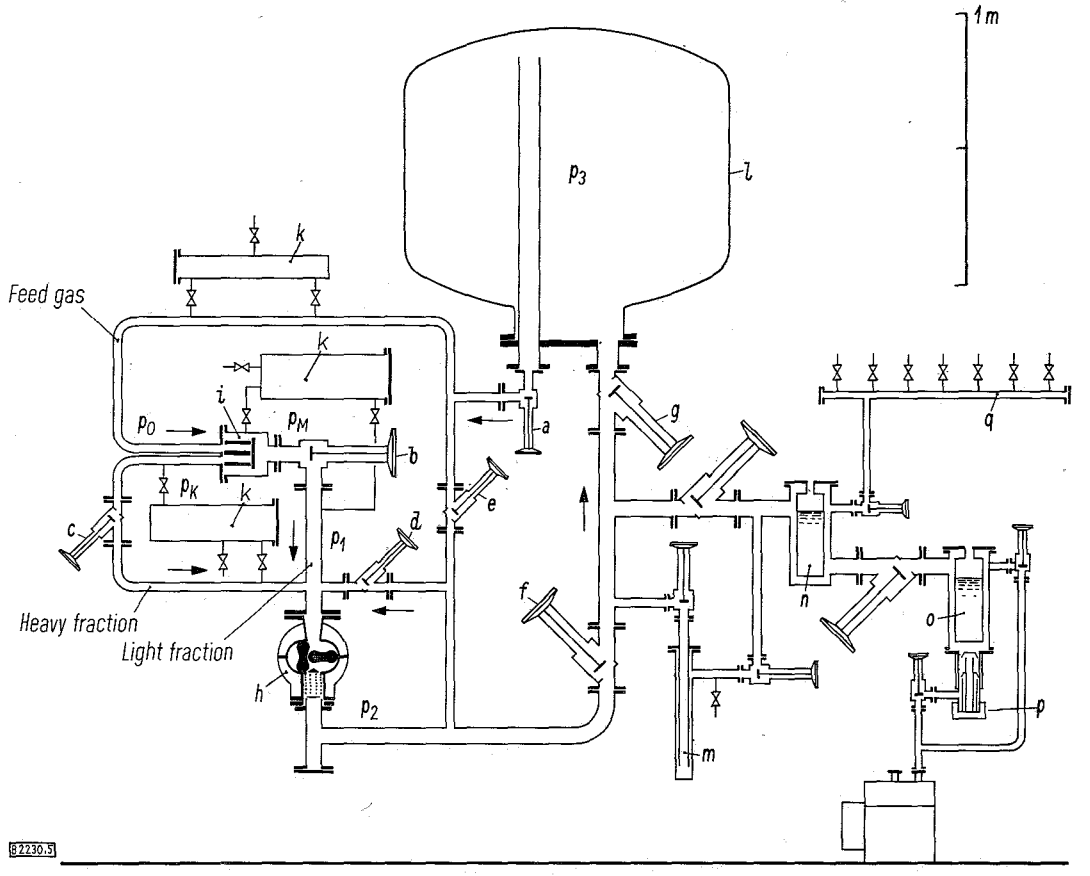

Fig. 14. Schematic drawing of the process test loop.
(a) through (g) valves; (h) Roots compressor; (i) separating element; (k) sample withdrawal reservoirs; (l) gas reservoir; (m) UF_6 trap; (n,o) cold traps; (p) vacuum pumps; (q) vacuum header.

reservoir, the volume of the test loop was 20 l. The reservoir also makes it possible to change the operating pressure without removing or adding process gas. When valve (e) is closed, the compressor pumps gas through the open valves (f) and (g) against the reservoir pressure, p_3. The nozzle fore-pressure, p_0, can then be regulated to values between the reservoir pressure, p_3, and the suction pressure of the compressor, p_1, by throttling the flow with valve (a). The pressures of the light and heavy fractions, p_M and p_K, can be adjusted by means of valves (b) and (c) to values between p_0 and p_1. The suction pressure, p_1, is regulated by means of the by-pass valve (d). The discharge pressure, p_2, of the compressor is normally identical to the reservoir pressure, p_3, and changes only slightly when the operating pressures of the separating element are changed due to the large volume ratio between the reservoir and the test loop. The pressures were measured with direct reading aneroid type and membrane type manometers as well as with electrical pressure transducers[*]. A self-stabilizing electromagnetic pressure balance with a linear response in the region from 0 to 760 mm Hg served as a standard[**]. For measurements in the region below 1 mm Hg, an electrostatic membrane micromanometer with a maximum sensitivity of 10^{-3} mm Hg per scale division was used. All of these instruments are suitable for direct measurements in UF_6 or in other fluorinating gases at room temperature. In addition, thermal conductivity and ionization pressure gauges were used for the control of the evacuation processes and for the measurement of the final vacuum in the different parts of the installation. These instruments were separated from the process gas space during operation with UF_6.

The test loop can be emptied by means of the liquid nitrogen-cooled UF_6 condenser (m). The helium can be pumped off with a rotary pump. A mercury diffusion pump is provided for the production of a high vacuum. It is protected against corrosive gas residues by two liquid nitrogen-cooled traps (n, o).

Austenitic steels, Nos. 4541, 4550, and 4571, were used for the construction of the test loop. All valves have a bellows or membrane type seal around the valve stem. Viton or, in special cases, teflon was used as gasket material. The maximum inleakage rate permitted was 5×10^{-6} mm Hg-l/s for the entire test loop, including the instrumentation and compressor, but excluding the 1000 liter reservoir. Before filling with UF_6, the entire test loop was baked out under high vacuum and passivated with a mixture of chlorine trifluoride (ClF_3) and nitrogen.

2.4. Isotope Separation Experiments

Although, according to the results of the experiments described in Section 1, a value of the UF_6 cut, θ_{UF_6}, equal to 0.33 would be expected to lead to the economically most favorable cascade arrangement, the ten stage pilot plant, because of the simpler interstage connections required, was designed to operate with a value of θ_{UF_6} equal to 0.5.

In practice, the value of θ_{UF_6} can only be set within a certain limit of error by the choice of a particular distance between the knife edge and the curved wall of the Laval nozzle. Since providing a means for

[*] Manufacturer: Appleby & Ireland, Basingstoke (England), Type 101 and 533.
Manufacturer: Heraeus GmbH., Hanau (Germany) Type VM–M.
Manufacturer: CEC, Pasadena, California (U.S.A.), Type 4-353-0001.
Manufacturer: Ateliers de Construction de Bagneux, Bagneux/Seine (France), Type 437 Gd 25121 and 25221.

[**] Manufacturer: CEC, Pasadena, California (U.S.A.), Type 4-336-0102.

adjusting the position of the knife edge during operation would be quite expensive, it seemed reasonable to utilize the dependence of θ_{UF_6} on the expansion ratio as a method for making fine adjustments in the value of the UF_6 cut.

For this reason, several series of isotope separation experiments were carried out with the test loop in which the variation of the separation effect and of the cut as a function of the expansion ratio, p_0/p_M, were of particular interest. At the same time these experiments served the purpose of life-testing the entire installation.

The compressor operated against the reservoir pressure, p_3, which was somewhat greater than the nozzle fore-pressure, p_0 = 48 mm Hg, so that a sufficiently large excess of gas was available in the installation for the withdrawal of samples. The expansion ratio was regulated with the by-pass valve (d) and the nozzle fore-pressure, p_0, with valve (a) (see Figure 14). The pressures of the light and heavy fractions, p_M and p_K, were always equal.

The dependence of the UF_6 mole fractions, N_0, N_M, and N_K in the feed gas, the light fraction, and the heavy fraction, respectively (obtained from the gas analysis), as well as that of the elementary separation effect, ε_A, for the uranium isotopes (defined by equation (4)) on the expansion ratio, p_0/p_M, is shown in Figure 15. The cuts, θ and θ_{UF_6}, were calculated from the relationships:

$$\text{Total cut, } \theta = (N_0 - N_K)/(N_M - N_K) \tag{9}$$

$$UF_6 \text{ cut, } \theta_{UF_6} = N_M\theta/N_0 \tag{10}$$

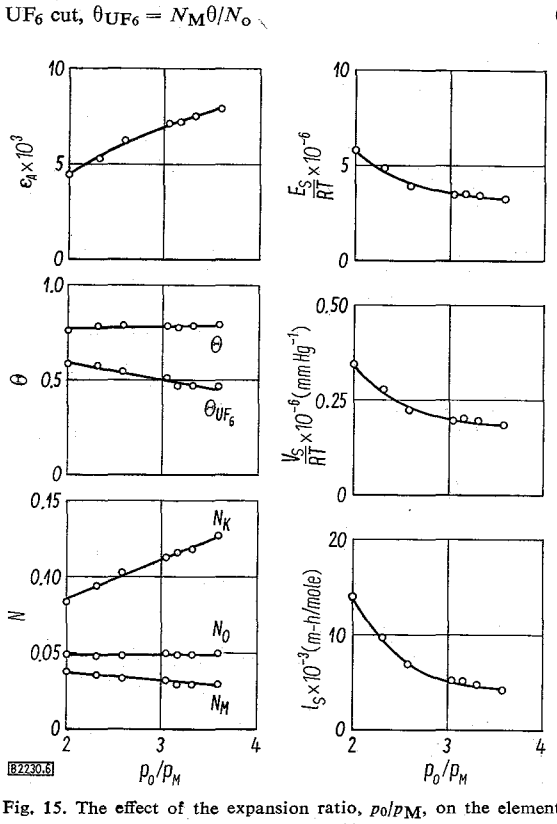

Fig. 15. The effect of the expansion ratio, p_0/p_M, on the elementary separation effect, ε_A; the total cut, θ; the UF_6 cut, θ_{UF_6}; the UF_6 mole fraction in the feed gas, N_0, in the light fraction, N_M, and in the heavy fraction, N_K; the specific work of compression, E_S; the specific suction volume, V_S; and the specific slit length, l_S.
Inlet pressure: p_3 = 48 mm Hg; $p_K = p_M$;
Helium-UF_6 mixture containing 5 mole % UF_6.

The dependence of the specific cost factors, defined by equations (5) through (7), on the expansion ratio is also shown in Figure 15.

The values for the separation effect and for the specific cost factors obtained in the test loop with the separation nozzle system 100 mm in length agree within the limits of error with those obtained in the corresponding earlier experiments with the 10 mm long nozzle system.

All of the experimental results presented in Figure 15 were obtained with the distance between the knife edge and the curved wall of the nozzle equal to 0.20 mm. Within the limit of error of the measurements, the curve for θ_{UF_6} as a function of the expansion ratio passes through the specified operating point (θ_{UF_6} = 0.5 at p_0/p_M = 3). From a different series of measurements, it was observed that, at this value of the expansion ratio, a decrease in the distance between the knife edge and the nozzle wall of 0.01 mm caused the UF_6 cut to increase from 0.50 to 0.54. The θ_{UF_6} curve presented in Figure 15 shows that a deviation from the design point of this magnitude can be compensated for by an increase of about 10 per cent in the expansion ratio.

One of the series of measurements was repeated after an interval of more than four months. In the interim, suction volume measurements were made and the endurance-testing of the Roots compressor was undertaken. During the entire time the installation was filled with the helium-UF_6 mixture, and for about 1100 hours it was maintained under operating conditions. The results from both experiments were in agreement. This signified that the properties of the separating element did not change measurably during the many months of experimental operation.

3. Comparison of the Specific Cost Factors of the Separation Nozzle Process and the Gaseous Diffusion Process

For the purpose of judging the economic potential of the separation nozzle process, one can compare the optimum values obtained for the specific cost factors with the corresponding values for the gaseous diffusion process. Since the operating conditions employed in the gaseous diffusion process and the value of the elementary separation effect which has been attained in practice are not known, the values of the specific cost factors for the diffusion process can only be estimated.

According to theory, the upper limit for the elementary separation effect for the gaseous diffusion process for uranium isotope separation, using UF_6 as the process gas, is given by

$$\varepsilon_A(\max) = 0.00429\,(1-p_M/p_0)\,\frac{1}{\theta}\ln\frac{1}{1-\theta}, \tag{11}$$

where again p_0 is the pressure in the gas entering the separating element and p_M is the pressure in the light fraction. The cut, θ, is the fraction of the feed stream which is pumped from the separating element as the light fraction at the pressure, p_M. One can safely

assume that in the gaseous diffusion process θ is equal to 0.5. The optimum value of the pressure ratio, p_0/p_M, for the gaseous diffusion process, will be assumed to lie in the neighborhood of four, as is the case for the separation nozzle process.

Thus one obtains:

$$\varepsilon_A \text{ (max)} = 4.45\times10^{-3}.$$

This maximum value for the elementary separation effect can only be attained in practice when the mean free path of the UF_6 molecules is large compared to the diameters of the pores in the membrane and when no local depletion of the light isotope occurs on the high pressure side of the membrane. However, in order to decrease the investment costs of the diffusion process, one would choose a gas pressure as high as practicable and thereby accept in the bargain a certain decrease in ε_A due to the smaller ratio of the mean free path to the pore diameter. Moreover, in practice, the mixing in the gas on the high pressure side of the membrane in the direction perpendicular to the membrane surface is not complete. If it is assumed that in practice only 60 per cent of the theoretical maximum value of ε_A is attained, one obtains for the effective elementary separation effect:

$$\varepsilon_A(\text{eff}) = 2.67\times10^{-3}.$$

The specific ideal isothermal work of compression is given by the ratio of the ideal isothermal work of compression performed per unit time to the separative power, δU, defined by equation (8). The resulting expression is

$$E_s = \frac{1}{\delta U}\,\frac{L^*}{2}\,RT\left(\ln\frac{p_0}{p_M} + \ln\frac{p_0}{p_K}\right). \qquad (12)$$

With $\varepsilon_A = 2.67\times10^{-3}$, $p_0/p_M = 4$, an estimated pressure drop for the heavy fraction, $p_0/p_K = 1.2$, and an estimated operating temperature, $T = 330\,^\circ\text{K}$ (see below), it follows that

$$E_s \text{ (diffusion)} = 6.8 \times 10^2 \text{ kWh/mole}.$$

The optimum value, $E_s = 14.4\times10^2$ kWh/mole, obtained for the separation nozzle process with the improved separating system and with $\theta_{UF_6} = 0.33$, is therefore still higher by a factor of 2.1 than that estimated for the diffusion process. In addition, it should be kept in mind that the compressor efficiency to be expected with the He–UF_6 mixture is somewhat lower than that with pure UF_6 because of the smaller molar heat capacity of the mixture.

In the estimate for the diffusion process it was assumed that the heavy fraction is pumped off at an appreciably higher pressure than the light fraction. This implies that each separation stage is provided either with two compressors or with a single compressor of a special type which pumps gas simultaneously at the pressures, p_M and p_K. It could be of advantage, at least when processing the higher ^{235}U concentrations, to avoid this complication by expanding the heavy fraction to the pressure, p_M [13]. In this case the difference between the specific work of compression for the gaseous diffusion process and for the separation nozzle process would almost disappear.

In order to estimate the specific suction volume, one must make an assumption regarding the absolute pressure at which the diffusion process operates. An upper limit, assuming that sufficiently fine-pored membranes are available, is given by the vapor pressure of UF_6. At room temperature the vapor pressure of UF_6 is about 100 mm Hg, and at 53 °C it reaches 1 atm. Since the operating temperature of the plant is probably not much above 50 °C, and since it must be maintained at a safe distance from the condensation point, one can take 0.5 atm to be a reasonable value for the operating pressure. With this value for p_0 and the values for the other variables as given above, one obtains from the equation for the specific suction volume,

$$V_s = \frac{1}{\delta U}\,\frac{L^*}{2}\,RT\left(\frac{1}{p_M} + \frac{1}{p_K}\right), \qquad (13)$$

that for the diffusion process:

$$V_s \text{ (diffusion)} = 1.59\times10^5 \text{ m}^3/\text{mole}.$$

The optimum value, $V_s = 1.40\times10^6$ m³/mole, obtained for the separation nozzle process with the improved separating system, is therefore higher by a factor of 8.8 than that estimated for the gaseous diffusion process.

When judging the economic consequences of the larger specific suction volume, one must bear in mind that the separation nozzle process, operating with a UF_6 cut of 0.33, requires only about 1/2.7 times as many stages as the diffusion process [*]. For a given separation job the suction volume per stage will therefore be greater than that for the gaseous diffusion process by the factor, 8.8 times 2.7, *i.e. ca.* 24. Results obtained in the French gaseous diffusion plant studies indicate that the variation of the investment costs per stage with the size of the stage is given by a constant times $L^{0.38}$, where L represents the throughput of the stage [14]. If one assumes the same dependence and the same constant for the separation nozzle process, and postulates that changes in the volume throughput due to changing the pressure affect the investment costs in exactly the same way as changes in the molar throughput, one obtains the result:

[13] *H. Kronberger* and *G. R. H. Geoghegan*: Proceedings of the International Symposium on Isotope Separation, Amsterdam 1957. North Holland Publ. Co., Amsterdam 1958, p. 517.

[*] The concentration change between adjacent stages in a cascade is equal to 1/3 of the elementary separation effect, ε_A, defined by equation (4), when $\theta_{UF_6} = 0.33$, and is equal to $\varepsilon_A/2$ when $\theta_{UF_6} = 0.5$.

[14] *C. Frejacques* and *R. Galley*, Third International Conference on the Peaceful Uses of Atomic Energy, Geneva 1964, Paper 28/P/89.

$$\left.\begin{matrix}\text{Investment Costs for the}\\ \text{Separation Nozzle Process}\end{matrix}\right\} = \frac{240.38}{2.7} (= 1.2)$$

$$\left\{\begin{matrix}\text{Investment Costs for the}\\ \text{Gaseous Diffusion Process.}\end{matrix}\right.$$

Thus, in spite of the fact that, according to the estimate above, the specific suction volume is greater by a factor of 8.8 one would not expect an appreciably greater investment cost for the separation nozzle process than for the gaseous diffusion process.

It is expected that the separation nozzle process will have a real advantage compared to the gaseous diffusion process with respect to the equilibrium time of a separation cascade. By the term, equilibrium time, one refers to the time required between the start-up of a cascade and the attainment of the product concentration, *i.e.* the time during which no product can be withdrawn. The equilibrium time for cascades which enrich ^{235}U to medium or high concentrations can be so long that they may not be neglected in the economic considerations. For a given separation job, the equilibrium time is directly proportional to the average residence time of a UF_6 molecule in a stage and inversely proportional to the square of the elementary separation effect [12]. Since the average residence time will be at least no greater, and probably appreciably less, in the separation nozzle process than in the diffusion process, one can expect that the equilibrium time will be shorter by at least an order of magnitude in the former owing to the larger elementary separation effect.

We wish to thank G. Frey for his assistance in the development of the Roots compressor and the test loop as well as Miss B. Gebauer and J. Dröge, W. Mannherz, and G. Schüler for their help in carrying out the experiments. We also wish to thank Dr. von Halle for valuable discussions and for the translation of this report.

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