Review of the gas centrifuge until 1962. Part I: Principles of separation physics

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There are two sets of principles involved in the development of the gas centrifuge, the internal separation physics and the external means of spinning a rotor at very high speeds. Only the first aspect is discussed in this part of the review. First, the industrial requirement for the separation of the uranium isotopes is defined so that the separation history can be put in a modern perspective. The history of separation physics itself is then traced back to the theory of centrifugal force by Huygens and the equivalence of this force to that of gravity. The barometric equation giving the variation of atmospheric pressure with height and the law of partial pressures can then be adapted to the centrifuge to give the steady-state theory of separation. This work was completed in the last century but was not confirmed in its application to isotope separation until 1936. The detailed separation physics for non-steady-state conditions required for a production centrifuge was developed during the American wartime Manhattan Project. During this work the theory giving the maximum output of a centrifuge was developed by Dirac, and soon afterwards Cohen and Kaplan showed that the best method of operation for a production centrifuge is in a countercurrent mode of operation. This method gives a large separation factor at relatively small flow rates through the rotor. The theory of how to set up an internal countercurrent was given by Martin during an equivalent wartime German project, and refinements to the theory, showing how the countercurrent persists along a centrifuge rotor, was given by Dirac and Steenbeck, the latter during a postwar Russian project. This theory was extended by Parker, Ging, and Mayo of the University of Virginia, whose work was completed by 1962, the limit of this review.

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Appendix D: Principles of Boundary Layer Theory Applied to

kinetic energy

 $\hat{\mathcal{A}}$

 $\pmb{\alpha}$ enrichment factor of centrifuge

tope there is necessarily a corresponding production of uranium depleted in this isotope, the exact amount being given by simple calculations of mass balance. For example, to make 100 tonnes of uranium enriched by a factor of about 3 requires a plant to convert 400 tonnes of natural uranium into two separate streams, one stream containing the required enriched product and the other containing 300 tonnes of depleted or tailings uranium, which has a residue of about 0.24% of uranium 235. It is not usually economical to reduce the U^{235} content in the tailings stream to a value much lower than this. This size of plant, to produce sufficient enrichment for a 2000 MW power station, is given as an example because two plants of this size are now operating, one in the UK and one in Holland. These plants were built under the auspices of Urenco, a joint Dutch/German/British enterprise set up as a result of an agreement between the three governments in 1970, and further plants are presently under construction. Similarly it has been announced by the Department of Energy that the next US enrichment facility will be a centrifuge plant; this, when complete, will provide sufficient enrichment capacity for 80000 MW of electricity.

For reasons of national security all of the present work in Europe and America is classified, and no details of the centrifuge technology have been released except photographs of the plants. Therefore, this review will be limited mainly to work prior to 1962.' However, this is not too serious, since most of the principles of the centrifuge were worked out before this date. In fact, much of the physics of the process was worked out in the 19th century, with little development in the present century until about 1936, when Beams started his work on isotope separation at the University of Virginia. This work at Virginia rapidly expanded following the discovery of fission of $U^{23\bar{5}}$ and the outbreak of war in Europe in 1939; it was incorporated into the wartime "Manhattan Project" in America. Most of the present technical knowledge of the gas centrifuge used for separating the uranium isotopes derives from this investigation, as well as from similar smaller projects in Germany and Russia during and immediately after the ¹⁹³⁹—⁴⁵ war.

The gas centrifuge described in these three investigations comprised a vertical cylindrical rotor, spinning at high speed with means for both feeding the mixture of isotopes into the rotor and extracting the two streams, one enriched in the light isotope and one depleted in the light isotope. A simplified drawing of a centrifuge rotor is given in Fig. 1; this shows the two important parameters of the centrifuge rotor, so far as its separation performance is concerned, its length Z, and its peripheral speed V.

FIG. 1. Simplified diagram of centrifuge. Feed stream: L mole/sec (abundance ratio R); product flow: θL mole/sec (abundance ratio αR); waste flow: $(1-\theta)L$, mole/sec (abundance ratio R/α). Conservation of mass gives $\theta = [(1+\alpha R)/\alpha]$ $(1+R)$]/(1+ α).

B. Dirac equations

To facilitate the review and to emphasize the importance of these two parameters, length and peripheral speed, it is useful to summarize the two major contributions to separation theory made by Dirac during the Manhattan Project. The first contribution was to give an accurate measure of the usefulness of a separating element which is independent of the concentration of the isotope mixture being used and which is also independent of the size of the element-for example, it must apply equally to a single centrifuge or a large plant. Dirac devised a function, discussed in Appendix A, which correctly determines the value, per mole, of a mixture of isotopes. This value function,² very similar to and related to the entropy function of a perfect gas, is given by

$$
V(J) = [(J-1)/(J+1)]\ln J .
$$
 (1)

When a mixture of isotopes is passed through a separating element, there is a net change of the value of the gas entering and leaving the element; this is used as a measure of the usefulness of the element. For example, in a symmetrical system, in which L moles of feed of abundance ratio J divide into ^a product stream of abundance ratio αJ and a depleted stream of abundance ratio J/α , the change in the Dirac value function reduces to the simple form

$$
\Delta U = L [(\alpha - 1)/(\alpha + 1)] \ln J . \qquad (2)
$$

This change of value is called separative work (SW) and has the same units as the feed flow. Thus, for the plant

There have been several reviews, mainly of the early work on the gas centrifuge, and the author is indebted to them for some of the ideas expressed in this historical review. These reviews are Von Halle et al., 1965; Glander, 1972, 1978; Whitley, 1979a, 1979b.

The value function was derived by Dirac as a mathematical function from cascade theory. At about the same time, Peierls derived the same function by physical considerations of entropy, and it is this version which is given in Appendix A.

described earlier, the feed flow was 400 tonnes/yr and the enrichment factor and depletion factor were both equal to three (in actual plants these factors are rarely equal to each other, but were set equal in this example for simplicity). Substituting these values into Eq. (2) gives the separative work required from the plant as

$$
\Delta U = 220te \, \text{SW/yr}
$$

This amount of separative work per year (sometimes called separative power) is far greater than that given by any individual centrifuge. This latter value can be calculated from the single most important result in centrifuge technology, namely the proof, by Dirac (1941), that irrespective of the method of operation, the maximum output of a centrifuge, in separative work units per unit time, is given by

$$
\delta U_{\text{max}} = \rho D (\Delta M V^2 / 2RT)^2 \pi Z / 2 \ . \tag{3}
$$

This equation will be discussed in more detail later, but a numerical example immediately gives the scale of the problem. Inserting the constants into the equation shows that the maximum output of ¹ m of rotor spinning at 300 m/sec is only 2 kg SW/yr. Thus more than 110000 machines of this length and speed would be required to give the 220 tonnes of separative work for the small plant described above. This requirement for so many machines makes it necessary to pay more attention to detailed physical principles than may be considered usual in the design of most high-speed devices. This will become clear in the following narrative.

C. Gutline of review

The first requirement in the separation of the uranium isotopes is to convert the uranium metal into the gaseous phase. The most convenient gaseous compound of uranium is uranium hexafluoride UF_6 . Some of the physical properties of this compound are listed in Table I, taken from the more comprehensive data of de Witt (1960). As shown, UF_6 has quite a high vapor pressure at modest temperatures and has the added virtue that fluorine exists only with one atomic mass of 19. Therefore, the gaseous compound consists only of the binary mixture $U^{235}F_6$ and $U^{238}F_6$, of molecular weights 349 and 352, respectively.

The partial separation of these gases in the centrifuge is based on the same principle as the partial separation of gases in the terrestrial atmosphere, as discussed by Lindemann and Aston (1919) soon after the discovery of isotopes. From this similarity it follows that the basis of the separation effect can be traced back to Huygens, who first proved that centrifugal force was more or less the same as gravitational force, to Halley, who derived the equation for the pressure distribution in the atmosphere, and to Dalton, who perceived the law of partial pressures —the fact that in ^a mixture of gases, each behaves independently of the others.

These facts were first quantified by Bredig (1895), who also proved them experimentally by partially separating the gases in a binary mixture. The technique was refined and improved by Beams (1936), who partially separated the isotopes of chlorine in his first experiment at the University of Virginia. The uranium isotopes were then separated by using the centrifuge during the last war as part of the Manhattan Project and in similar projects in Germany and Russia during and after the war.

The historical lineage of the separation theory is outlined in Fig. 2, and the present review will follow this plan. The main problem in the review involves the discussion of the early work. Plagiarism appeared rife in the 17th century, and at least two discoveries in the centrifuge story were first transmitted to the Royal Society in the form of anagrams to establish precedence. These were the explanation of centrifugal force by Huygens and the law of elasticity by Hooke. However, within the limitations caused by ibis procedure, it is believed that the various precedents given in this review are correct.

II. HISTORICAL BACKGROUND

A. Centrifugal force and the barometric equation

The most important physical principle involved is that of centrifugal force itself. The effects of the forces asso-

IABLE I. Physical properties of nex.					
Temperature (°C)	0	27	50		
Vapor pressure mm Hg	17.7	128	527		
(Pa)	2360	17000	70200		
Diffusion ρD 10 ⁻⁵ Ns/m ²	2.00	2.19	2.32		
Viscosity η 10 ⁻⁵ Ns/m ²	1.58	1.72	1.84		
$\eta/\rho D$	0.79	0.79	0.79		
Specific-heat					
ratio ν	1.0704	1.0687	1.0666		
Specific heat C_n					
$J/g \,\mathrm{mol}^{\circ}\mathrm{C}$	126	130	133		
J/kg °C	359	370	378		
Thermal conductivity					
10^{-4} W/m °C	61.1	68.0	73.9		
$4\kappa\gamma/(9\gamma-5)\eta C_p$	0.966	0.986	0.983		

 T

FIG. 2. Centrifuge separation history.

ciated with circular motion have been known for at least five thousand years, as evidenced by fragments of potters' wheels dated by Woolley (1930) as 3250 ± 250 B.C. Ancient potters used the forces involved with rotation to practice their art. However, this and other clear experimental evidence of centrifugal forces was of little importance to the early scholastic philosophers, and even Galileo thought that motion in a circle was a perfect and natural form of motion. It was left to Huygens to recognize the existence of centrifugal forces. His main work, De vi Centrifuga (written in 1659), was not published until 1703, after his death, but he had previously published his theories, without proof, in his Horologium Oscillatorium (1673) and had sent a summary of his work to the Royal Society in the form of anagrams in ¹⁶⁶⁹—see Crew (1935).

These theorems of Huygens were included in the published teachings of Keill (1702), the Professor of Astronomy at Oxford in the late 17th century. His early books are in Latin, but later editions of his works were published in English. The work of Keill, who derived the proof of Huygens's theories for himself, is the earliest reference to the teaching of Huygens's theory. The various theorems of Huygens, proved by the complicated geometrical methods of the era, were the equivalent of the modern formula V^2/r for the radial acceleration in a centrifuge. However, the main importance of Huygens's work was his recognition that this centrifugal acceleration and the resulting force were as real as the effects of gravity. Centrifugal force, as defined by Huygens, is the outward force experienced by an observer taking part in the circular motion. It is related to, but has no direct connec-

frame of reference. Centripetal force is the force required to give the inward radial acceleration of an object moving in a circle at a uniform speed, as seen by an observer at rest. The complete works of Huygens, including his treatise on centrifugal force, were translated into French in 1934.

tion with, centripetal force, calculated and discussed by Newton (1687), since this force is defined in a different

According to the translators and other scholars-for example, Taton (1958)—Huygens anticipated Einstein by over 200 years, since, according to modern relativity theory, the forces of inertia, in this case centrifugal force, are not only equivalent to gravitational forces, as averred by Huygens, but identical. This identity is important in the theory of the centrifuge, since much of the theory of the terrestrial atmosphere can be adapted, with a suitable change of geometry, to the centrifuge. In particular, the separation of gases in the centrifuge is based on the same principle as that which gives the variation of gas density in the terrestrial atmosphere, heavy gases tending to concentrate near the ground and lighter gases tending to concentrate in the upper atmosphere.

The pressure distribution in the atmosphere is easily found by balancing the forces on a unit at height h , giving the well-known equation

$$
dp/dh = -\rho g \tag{4}
$$

Assuming isothermal conditions and integrating gives the barometric equation

$$
p = p(o) \exp(-Mgh/RT) . \tag{5}
$$

(6)

This exponential pressure distribution was derived by Halley (1685), who, by using Boyle's law and logarithm tables, correctly gave the pressure distribution for an isothermal atmosphere, showing that the pressure falls off exponentially by a factor of e every 9 km. This height, $H = RT/Mg$, is often called the "height of the homogeneous atmosphere." Sometimes it is more convenient to define the distance H_1 , in which the pressure falls off by a factor of 10. This distance for the atmosphere is about 20 km. Thus at a height of 100 km (5 decades of pressure fall) the pressure is reduced by a factor of $10⁵$ and the atmosphere merges into space, being composed mainly of light molecules moving around in parabolic paths with few intermolecular collisions. This reduction in pressure is indicated in Fig. 3.

An interesting note on this work is recorded in the correspondence of Newton (edited by Turnbull in 1957) to the effect that Halley, when Deputy Controller of the Mint in Chester (1696—98), ascended Mt. Snowdon in North Wales to see if he could determine the height by observing the change of pressure readings on his barometer. It is not recorded whether or not he observed the expected reduction of about 10% in pressure. However, the importance of Halley's work in the present context is that the application of his principles immediately gives the pressure distribution in the centrifuge. The result is given by Eq. (6), the form depending on whether the pressure at the axis or at the periphery is used as reference. Thus

or

$$
p = p(a) \exp[-M\omega^2(a^2 - r^2)/2RT].
$$

 $p = p(i) \exp(M\omega^2 r^2 / 2Rt)$

This barometric-type equation is the key to the separation effect, since it can be shown that in a gas mixture it applies to each gas separately. This in fact is an expression of the law of partial pressures given by Dalton (1801), which states that in a mixture of gases each gas behaves independently of the presence of the others. Thus, for two gases which differ in molecular weights by ΔM , the ratio $J(r)$ of the partial pressures at radius r compared with that at the perimeter would be given by

FIG. 3. Pressure distribution in (a) atmosphere and (b) centrifuge, showing first five decades of pressure. First decade contain 90% of gas, second 9%, third 0.9%, etc.

$$
J = J(a) \exp[\Delta M \omega^2 (a^2 - r^2)/2RT]
$$

= $J(a) \exp[\lambda (a^2 - r^2)]$. (7)

From Avogadro's law (1811), the number of molecules per unit volume of gas is proportional to its partial pressure, so that $J(r)$ is not only the ratio of the partial pressures but also the ratio of the molecular concentrations of the two constituents. This ratio, $N/(1-N)$, is usually called the abundance ratio. Thus the basic equation (7), which defines the separating effect in the centrifuge, derives from the original discoveries of Huygens, Halley, and Dalton.

However, the physical understanding of Dalton's law, Avogadro's law, and the Halley assumption that conditions in the atmosphere are isothermal came only with the advent of the kinetic theory of gases. It is well known from this theory that the pressure of a gas is given by $2ne/3$, where *n* is the number of molecules per unit volume and e is the kinetic energy of each molecule. The demonstration by Maxwell (1860) of the principle of the equipartition of energy then gave the mathematical proof of Avogadro's and Dalton's laws. Actually the demonstration of the equipartition of energy was given much earlier, in 1845, by Waterton in a paper that was rejected by the Royal Society but eventually published in 1892.

The assumption of isothermal conditions in deriving Eqs. (5) and (6) is more difficult to justify without detailed recourse to the kinetic theory of gases and the statistical concepts of the Maxwell-Boltzmann distribution law; the argument is summarized in Appendix B.

From the foregoing it is clear that the semiempirical laws required for the separation of gases were understood by 1811 and the theory by 1860. However, it was Bredig (1895) who finally gave Eqs. (6) and (7) in their present form, although he himself gives credit to some earlier work by des Coudres (1893). Bredig not only derived the separation equation, but attempted to prove it experimentally, achieving a partial separation of a mixture of hydrogen and hydrogen iodide. The tube shown in Fig. 4, with the glass stopper open, was filled with a uniform mixture of gases, clad in cork and brass to minimize frictional heating, and then spun for a few hours at a rotational frequency of 42 Hz. This gave a maximum peripheral speed at point B of 55 m/sec equivalent to a centrifugal force of 1470 times gravity. To do this work Bredig borrowed an analytical-type centrifuge from the Amsterdam Universi-

FIG. 4. The Bredig experiment. Peripheral speed at point $B=55$ m/sec; pressure ratio from B to $A=1.084$ for hydrogen iodide and 1.001 for hydrogen. Thus the separation factor $=1.083$ (from Bredig, 1895).

ty Hygiene Laboratory. After stopping the centrifuge, he closed the stop value as soon as possible. The measured change in concentration between spaces ¹ and 2 was 3%, in approximate agreement with the theoretical value of 4%.

Thus it can be seen that the physical principles of the gas centrifuge and an experimental proof were given even before the discovery of radioactivity in uranium by Becquerel. The work in the 20th century has been in increasing the output; this has, of course, involved significant improvements in the length and peripheral speed of the rotor. For example, the output of the Bredig experimental centrifuge, in modern terms, was about 10^{-6} kg SW/yr. The output of the latest American machine has been given at \sim 100 kg SW/yr, an improvement of 10⁸.

B. Isotope separation

Following the discovery of radioactivity at the turn of the century and the new radiochemistry, the existence of isotopes was soon recognized and the word "isotope" given by Soddy in 1913. However, there was no rush to separate isotopes, and it was not until 1936 that the successful experiments of Bredig were repeated with an isotopic gas mixture. This delay was due in part to the problem of convection, an effect that is more serious for isotopic mixtures of small mass difference than for the gases used by Bredig. In the case of isotope separation, the slightest heating of the rotor can cause mixing by convection, so destroying the small separating effect.

However, in 1934 Beams and others at the University of Virginia undertook the development of a convectionfree centrifuge, a development which is still proceeding 50 years later. Convection-free operation was achieved by operating the rotor in a vacuum or sometimes by introducing hydrogen to establish thermal equilibrium and so reduce unwanted convection to a minimum. After only two years Beams and Haynes (1936) announced the partial separation of the chlorine isotopes. He used a disctype rotor, spinning at 433 m/sec, nearly ten times faster than the speed achieved by Bredig. The centrifuge was operated in what is called the evaporative mode. A small amount of liquid, in this case carbon tetrachloride, was condensed on the periphery of the rotor, forming a large reservoir of nearly constant composition. In equilibrium the light gas in the vapor phase concentrates near the axis and is drawn off via a central hollow shaft which supports the rotor.

Following this early work, Beams (1938) quickly developed his first tubular rotor and Beams and Skarstrom (1939) repeated the experiments with the chlorine isotopes under more controlled conditions. In this new experiment 167 g of liquid carbon tetrachloride were condensed on the periphery and up to 0.05 g/sec withdrawn from the axis. As the vapor moves from the periphery to the axis, it tends to spin faster due to the conservation of its angular momentum, so causing possible stirring and mixing of the separated isotopes. This effect was prevented by using a star-shaped spider to divide the rotor into sector-shaped cells, so forcing all the fluid to spin at the same speed. With this arrangement the experimental results were found to be in good agreement with theory.

These tests using Beams's tubular rotor marked the end of the prewar work. The tests fully confirmed the theory of the separation process, for both steady-state and. dynamic operation.

The rest of the story of centrifuge development is mainly concerned with the separation of the uranium isotopes and efforts to increase the output of the rotor and make it more suitable for production use. The main problem that arises is that, contrary to what is said in most popular expositions of the centrifuge, the separation process depends on the absolute molecular weight and not just on the mass difference of the constituents, as implied by Eq. (1). This dependence of separation on molecular weight (actually on the group $MV^2/2RT$) is particularly important at the higher speeds needed for economic operation. The effect arises because of the extremely large pressure ratios that are set up across the centrifuge rotor. The theoretical pressure ratios can in fact be so large that, if the pressure is set at a reasonable value at the axis, the wall pressure would be so high that it would destroy the centrifuge; actually the gas would condense on the rotor wall with much the same consequence. Conversely, if the wall pressure is set at ^a reasonable value—say, atmospheric pressure —the central region is ^a void and the efficiency is reduced. This effect will become clear from the theoretical analysis of the centrifuge developed during the Manhattan Project.

III. THE MANHATTAN PROJECT

The separation of uranium isotopes on a large scale became a subject of national importance following the discovery of the fission of the uranium nucleus by Otto Hahn and the outbreak of the Second World War in 1939. A major investigation of the gas centrifuge started in America, and the work was eventually unified under the auspices of the Manhattan Engineer District, or, as it came to be known, the Manhattan Project. The development work was done at the University of Virginia, production machines were designed and tested at Westinghouse, and the pilot plant operations were the responsibility of the Standard Qil Development Company. Eventually, in 1944, the project was dropped in favor of gaseous diffusion, but not before almost the whole theory and practice of the process had been established.

The experimental work had a flying start due to the prewar work just described, and the scientists at Virginia and Westinghouse soon developed the mechanical aspects of the three main types of centrifuge—disc, subcritical, and supercritical-and successfully tested three methods of operation of the centrifuge, the evaporative, the concurrent, and the countercurrent methods, respectively. Dirac, as already mentioned, contributed some fundamental studies, and Cohen and others worked out isotope diffusion theory for the centrifuge at the non-steady-state conditions required for a production machine. The compiete theory of cascade design was also established, this being necessary for both the centrifuge and the gaseous diffusion process. Most of the work has been published by Cohen (1951), but in the present context it is the isotope separation theory which is the most important. Cohen divided the work into two parts; first, he derived the steady-state solution following the work of Bredig (1895), and second, he derived the diffusion equation and gave its solution for various modes of operation.

A. Steady-state conditions--the Bredig equations

The basis of the separation effect for isotopes is exactly the same as that derived by Bredig. The mass difference for isotopes, however, is small, so that the pressure ratio across the rotor is only slightly different for the two isotopic species; thus the change of molecular abundance from periphery to axis is also small. At 300 m/sec the pressure ratio for $U^{238}F_6$ is 575, and for $U^{235}F_6$ it is 544. The ratio of these two numbers, 1.056, gives the ratio of isotopic abundance from periphery to axis and is called the radial separation factor, or sometimes the equilibrium simple process factor. This radial separation is the basis of every production mode of operation of the centrifuge. However, the output of a centrifuge is zero at equilibrium conditions, since no isotopes are being withdrawn from the rotor. To determine the solution for the nonequilibrium case it is necessary to consider the equilibrium pressure distribution in the centrifuge as one of dynamic equilibrium between sedimentation and diffusion.

Before examining this concept, however, it may be useful to examine the numerical values of the equilibrium pressure ratio and separation factor given by the Bredig equations (6) and (7). At equilibrium, these values are, for the maximum pressure ratio,

$$
p(a)/p(i) = \exp(MV^2/2RT) , \qquad (8)
$$

and for the maximum radial separation factor,

$$
(\alpha \beta)_r = \exp(\Delta M V^2 / 2RT) \tag{9} \quad \text{where}
$$

The theoretical maximum pressure ratio and maximum separation factor above, together with the Dirac output per meter given by Eq. (3), are summarized in Table II. As can be seen, at the peripheral speeds used during the early projects, the pressure ratio is reasonably small, and it is possible to fill the centrifuge completely with uranium hexafluoride. Thus, if the wall pressure is set at 50000 Pa, the axis pressure at 300 m/sec is still at the relatively high value of 100 Pa. At this pressure the mean free path of hex is still sufficiently small, 0.02 mm, for the gas to behave as a continuum with ρD constant.

However, at higher speeds the theoretical pressure ratios are unrealistic. The real situation is very similar to that in the atmosphere. The pressure of uranium hexafluoride, set at the periphery, reduces exponentially as one moves radially inwards, and falls by a factor of 10 in the small distances given in Table II. After the pressure has fallen by, say, four factors of 10, the mean free path is 0.4 mm, and the hex atmosphere merges into a void near the center, with only a few hex molecules randomly mixed up with the light gas impurities, just as the earth's upper atmosphere merges into space. At these low pressures the central core of the rotor cannot contribute any useful separation, with the result that the output at high speeds increases only as the square of the peripheral speed instead of fourth power as given by the Dirac law. This same conclusion emerges from the hydrodynamic theories discussed later.

A simple way of understanding the effect is to note that the output, in this simplified description, is independent of radius and depends only on peripheral speed. It is therefore permissible to compare the output at different peripheral speeds at radii adjusted to give a constant set value of the centrifugal acceleration at the periphery. For a given peripheral speed this radius equals V^2/g_1 , where g_1 is the set value³ of the centrifugal acceleration. At this constant g_1 value, the pressure distribution inwards from the periphery will be independent of speed and will follow the same barometric law. At high speeds it is permissible to unwrap the centrifuge, as shown in Fig. 5. Then the only effect of speed is to increase the useful area $2\pi aZ$ of the centrifuge, and this, substituting for the radius a , is proportional to the square of the peripheral speed.

The effect can be quantified by setting a pressure p_1 below which useful production is impossible; the limiting pressure could be so low that it is impractical to sustain an axial flow or it could correspond to the transition from laminar to molecular flow, when the relationship $\rho D = \text{const}$ breaks down. Once a limit is set, the maximum possible output can easily be calculated by performing the Dirac integration over the outer annulus, bounded by the radius at which the pressure is p_1 and the outer radius at pressure $p(a)$. This gives an output equal to the Dirac maximum, Eq. (3), reduced by an efficiency factor,

$$
e_1 = (2\nu/A^2)(1 - \nu/2A^2) , \qquad (10)
$$

$$
v=\ln[p(a)/p_1].
$$

At high speeds the second term can be neglected; the equation then shows that the efficiency is proportional to the number of decades of pressure ratio that can be utilized as one moves in from the wall and the efficiency is inversely proportional to the square of the peripheral speed. As mentioned earlier this result has been given by theoreticians using more complicated and more rigorous theories. These are discussed later.

An interesting aside is that if one sets the rotor radii to give $g_1 = 10$ m/sec² and divides the resulting rotor area into the area of the earth, one can calculate the potential separative output of the earth. The earth, if covered with hex, would have an output of 50×10^6 tonnes of SW per year.

		Pressure Ratio				
V		for		$\delta U_{\rm max}$		
(m/sec)	A^2	$U^{235}F_6$	$\alpha\beta$	kg SW/yrm	H_1/a	H_4/a
300	6.35	573	1.056	2.15	0.181	
400	11.29	80×10^3	1.101	6.78	0.102	0.571
500	17.64	46×10^{6}	1.162	16.55	0.0653	0.309
600	25.40	11×10^{10}	1.242	34.32	0.0453	0.202
700	34.57	10×10^{14}	1.343	63.59	0.0333	0.144
800	45.16	41×10^{18}	1.469	108.51	0.0255	0.108
900	57.15	66×10^{23}	1.628	173.81	0.0201	0.084
1000	70.56	44×10^{29}	1.825	264.8	0.0163	0.068

TABLE II. Pressure ratio, enrichment factor, and separative work at various peripheral speeds.

B. Sedimentation and diffusion

Before discussing these theories, it is necessary to review the dynamic equilibrium between sedimentation and diffusion in gases and solve the full diffusion equations for isotope separation. This rather complicated theory of dynamic conditions was worked out by various theoretical physicists during the Manhattan Project and eventually published by Cohen (1951). Full references to the various scientists involved are given by Cohen and therefore are not listed in the following summary.

It is known from the kinetic theory of gases that the molecules in the atmosphere are in a state of continuous motion and that the equations for the pressure distribution represent the statistical average for a large number of molecules moving at random according to the Maxwell-Boltzmann distribution laws. The pressure distribution in a field of force also follows immediately from these laws, and this is discussed in Appendix B. However, apart from this molecular viewpoint using the kinetic theory, it is useful, following Cohen, to consider the pressure distribution as a dynamic equilibrium between the opposing effects of sedimentation and diffusion.

Sedimentation is the effect of gravity trying to pile all the molecules in the atmosphere onto the surface of the

earth, or of centrifugal force trying to pile all the molecules inside the rotor onto its wall. Sedimentation can readily be calculated by various techniques in kinetic theory, but possibly the easiest to understand is that derived using the Nernst-Einstein concept of mobility. Nernst and Einstein showed that the average drift velocity of molecules per unit of force, or "mobility," is related to the diffusion coefficient and equals D/kT . This relation gives the sedimentation rate in a field of force as the product of the mobility, the force on the molecules, and the density. Thus, in the atmosphere, for one molecular species, mass m_2 ,

$$
sedimentation flow = um_2g\rho . \qquad (11)
$$

The balancing flow, diffusion, is the natural tendency of gases to spread out and try to erase pressure or concentration gradients; its value is given by Fick's law,

diffusive flow =
$$
-Dd\rho/dh
$$
. (12)

Equating the sedimentation and diffusive flows at equilibrium, and integrating, immediately gives the standard barometric formula

$$
\rho = \rho_0 \exp(-Mgh/RT) \tag{13}
$$

FIG. S. Centrifuge rotor in planar geometry. At high speeds nearly all the gas is near the periphery, and it is permissible to unwrap the rotor. The useful height for separation is constant at constant acceleration, so the output is proportional to $2\pi aZ$, which is proportional to V^2 .

It is interesting to note that the sedimentation and diffusive flows, given by $D\rho/H$, are constant and independent of height in the atmosphere. The reduction in density with height is just compensated for by the increase in mean free path and hence in the diffusion constant. The result that $D\rho$ is a constant, one of the well-known derivations of the kinetic theory, is of considerable importance in centrifuge theory because in every mode of operation it determines the rate of product removal—see, for example, the basic Dirac equation (3).

C. Equilibrium separation factor

The advantage of considering the atmosphere on the earth or in a centrifuge as one in dynamic equilibrium is that the method can easily be extended to calculate the sedimentation and diffusive flows for a single gas in a mixture of gases and so to determine the separation factor. For example, consider the two isotopic components of a binary gas mixture in the centrifuge, of densities ρ_1 and ρ_2 , respectively. The total density and density gradient in the centrifuge are given by

$$
\rho = pM_1N/RT + pM_2(1-N)/RT \t{,} \t(14)
$$

$$
dp/dr = \omega^2 r(\rho_1 + \rho_2) \tag{15}
$$

Using these equations, we obtain the sedimentation flow and diffusive flow, $D d\rho_1/dr$, of the light isotope:

$$
\text{sedimentation flow} = \rho_1(m_1\omega^2 r) D / kT \tag{16}
$$

diffusive flow =
$$
DM_1(p \, dN/dr + N \, dp/dr)/RT
$$
. (17)

Setting these two flows equal to each other gives the simple differential equation, again for the equilibrium condition,

$$
r dJ/dr + 2\lambda r^2 J = 0 \tag{18}
$$

In this equation the dependent variable has been changed from the concentration to the isotope abundance ratio. Integrating gives the standard steady-state solution, Eq. (7), for the variation of abundance ratio with radius.

In a production centrifuge this dynamic equilibrium is necessarily disturbed as feed gas enters and the enriched product and the depleted waste are withdrawn from the centrifuge rotor. Cohen has set up the diffusion equation, solved it, and optimized it for the three modes of operation investigated during the wartime project. The solutions for these three cases are discussed in the following sections.

D. Evaporative centrifuge

In this model of operation a small amount of liquid is introduced into the centrifuge, forming a layer at the periphery, and thus creating a large reservoir of the isotopic mixture. During the spinning of the rotor, vapor is removed slowly through a shaft along the axis. In practice the reservoir of liquid at the periphery is depleted in its

concentration of the light isotope, but this is neglected in this simplified analysis, originally given by Humphreys (1939). In this mode of operation the optimum rate of removal of the vapor is proportional to the length of the rotor and the diffusion constant and therefore can be written nondimensionally as

$$
q = L/4\pi Z\rho D \tag{19}
$$

The flow L is the inward convective flow of both isotopic species over a total area of $2\pi rZ$. Therefore, the inward convective flow of the light isotope, per unit area, is

$$
convection flow = 2Nq\rho D/r . \qquad (20)
$$

Adding this flow, with the correct algebraic sign, to the sedimentation and diffusion flows gives the modified differential equation

$$
r dJ/dr + 2\lambda r^2 J = -q(1+J)J \tag{21}
$$

The Cohen solution of this equation is

$$
J(r) = J(a) \exp[\lambda(a^2 - r^2)/(1+q)], \qquad (22)
$$

giving

$$
(\alpha \beta)_e = \exp[A_1/(1+q)] \tag{23}
$$

and

$$
\delta U / \delta U_{\text{max}} = 4q / (1 + q)^2 \,. \tag{24}
$$

Equations (23) and (24) are plotted in Fig. 6. As can be seen, the maximum efficiency is 100% and occurs when q is unity and dJ/dr is exactly one-half of the equilibrium value of $-2\lambda rJ$.

Although the evaporative centrifuge achieves the Dirac maximum output, it operates by a batch process not easily adapted to continuous production. It does show, however, that the maximum separative effect occurs when every

FIG. 6. Cohen theory for evaporative centrifuge, plotted from equations in Cohen (1951). The maximum output, of 100% efficiency, occurs at a flowrate of L_0 , when the radial gradient of the abundance ratio is one-half of its equilibrium value.

volume element of the rotor is working at one-half of the equilibrium value. Dirac showed this in a more elegant and general way, and this solution is included by Cohen in his book.

E. The concurrent centrifuge

The next method of operation investigated in the Manhattan Project, the concurrent method, is illustrated in Fig. 7. The gas mixture is introduced at one end of the rotor, and two streams are taken off at the other end, one from near the axis and one from near the periphery. As developed by Beams, the two streams are confined to the thin annuli shown in the figure. During the passage through the rotor, the isotopic concentrations of the two gas streams change, tending towards the radial equilibrium distribution. The diffusion equation, which includes axial as well as radial mass balance, is given by Cohen as

FIG. 7. Countercurrent mode of operation in Beam's centrifuge. Reversing direction of either stream gives the concurrent mode (from Cohen and Kaplan, 1942, and Cohen, 1951).

$$
dJ/dr + 2\lambda r^2 J = (LM/2\pi\rho D)dJ/dz
$$
 (25)

The solution of this equation, including the separation factor and the separative work output, is given by Cohen in terms of a parameter θ , which is an inverse function of the feed flow per unit length of the rotor. This function and the solutions can be written

$$
L/Z = 8\pi\rho D/\theta \ln(a/b) , \qquad (26)
$$

$$
\alpha \beta = \exp[A_1(1 - b^2/a^2)(1 - \exp(-\theta)], \qquad (27)
$$

and

$$
\delta U/\delta U_{\text{max}} = 4FG \tag{28}
$$

where F is a function of the flowrate and G is a function of the geometrical arrangement as defined in the nomenclature. The two functions both have the same maximum value of 0.407 at a θ value of 1.256 and b/a of 0.534, respectively. The fact that these functions have the same maximum value is a mathematical quirk of no significance —the functions can be transformed, one to the other, by setting $\exp - \theta$ equal to $(b/a)^2$ or vice versa. At these optimum values,

$$
L = 32\rho DZ \t{,} \t(29)
$$

$$
\alpha \beta = \exp(0.51 \Delta M V^2 / 2RT) , \qquad (30)
$$

$$
\delta U = 0.66 \delta U_{\text{max}} \tag{31}
$$

These optimal values are shown on the graph of output against flow in Fig. 8.

F. The countercurrent centrifuge

The third mode of operation investigated by Beams and Cohen, which was adopted as the choice for the proposed production machine in 1944, is the system in which an axial countercurrent flow of gas is induced in the rotor. A thin axial flow near the wall becomes enriched in the heavy isotope as it moves along the wall. Similarly an opposing flow in the main body of the rotor carries gas enriched in the light isotope to the opposite end of the rotor. This results in a concentration gradient in the axial direc-

FIG. 8. Cohen theory for concurrent centrifuge (adapted from Cohen, 1951). The maximum output, efficiency 66%, occurs at a feed rate of $32\rho DZ$.

tion similar to that in the radial direction. Then, if the length of the centrifuge is much greater than its diameter, the countercurrent has the property of multiplying the radial separation factor many times in one unit.

There are two variants of the countercurrent system. The first, adopted during the Manhattan Project and shown in Fig. 9(a), uses a forced countercurrent in a "once-through" or "flow-through" system. The second variant, used in the German and Russian projects and shown in Fig. 9(b), uses a continuous internal countercurrent with only partial removal of the product and waste flows at the two ends. The theory is similar for both types, since an internal countercurrent can be considered as two of the ance-through systems joined together, one acting as the rectifier and the other as the stripper. This is indicated in Fig. 10 for two cases, one in which the incoming feed gas enters the outer stream of the internal countercurrent, and one in which it enters the inner stream.

The full diffusion equation for the countercurrent centrifuge is rather difficult to solve analytically, but approximate solutions are possible, following a method originally developed by Furry, Jones, and Onsager (1939) for the thermal diffusion column. Cohen, using this method, has calculated the first integral of the diffusion equation, which is

$$
r dJ/dr + 2\lambda r^2 J = [\psi(r)/2\pi\rho D] dJ/dz . \qquad (32)
$$

The simplifications involved in this integral have to do with the radial concentration gradient and the effect on it of axial back diffusion, as discussed in a more exact theory given by Herman (1962). However, the main prin-

FIG. 9. Flow-through and internal countercurrents. (a) Flowthrough rotor, showing two-shell profile; (b) internal countercurrent, showing three pipes and ideal profile (adapted from Kanagawa and Oyama, 1961, and Von Halle et al., 1965).

FIG. 10. Two flow-through rotors joined to make internal countercurrent rotor. (a) Feed enters outer stream; (b) feed enters inner stream (adapted from Von Halle et al., 1965).

ciples can be followed reasonably well using the Cohen solution, Eq. (32), which shows that the axial concentration gradient, and hence the efficiency, depends only on the function

$$
p(r) = \int_0^r \rho w 2\pi r \, dr \tag{33}
$$

This function, usually called the stream function, defines the mass flow of the countercurrent in the central core of radius r , and its value depends on the magnitude and velocity profile of the countercurrent flow. Cohen solved the differential equation (32) and gave his results in terms of three definite integrals, ϕ_1 , ϕ_2 , and ϕ_3 of the stream function. If these integrals are computed for a given profile, the efficiency can be written as

$$
E = 4\phi_2^2/a^4\phi_3 \tag{34}
$$

The absolute value of the circulation required to achieve this efficiency is

$$
S = \phi_1 S_0 / 2(\phi_3)^{1/2} \tag{35}
$$

G. Optimum profile of countercurrent

Using this solution Cohen was able to examine the properties of several types of axial velocity profile', in particular, he showed that the most efficient profile is one in which the axial velocity w at a given radius is inversely proportional to the pressure (or density) at that radius. This condition, velocity times density equals a constant, means that the axial mass flux in the inner countercurrent is spread out uniformly across the rotor cross section. This "ideal profile" is, of course, a mathematical optimum which is impossible to achieve in a real centrifuge at even moderately high speeds. This is apparent from the theoretical pressure ratios given in Table II—these

pressure ratios become so great that there is no possibility of achieving the inverse relationship with axial velocity. As mentioned earlier, the central core of a high-speed rotor is essentially a void. Nevertheless the ideal profile is easy to calculate and is useful as a reference for comparison with other theoretical or practical profiles. If this ideal internal circulation is set up at a total flow value of mS_0 and no product and waste flows are extracted, i.e., if there is no feed flow, the solution given by Cohen reduces to

$$
\alpha \beta = \exp \left[\frac{\Delta M V^2}{2RT} \frac{\sqrt{2}Z}{d} \frac{2m}{1+m^2} \sqrt{E} \right].
$$
 (36)

In this expression the efficiency term \sqrt{E} is included to allow for less efficient profiles, to be discussed later. For the optimum profile the value of E is unity.

The dependence of the axial separation factor on the circulatory flow factor m is illustrated in Fig. 11. Clearly at zero circulation there is no axial separation (and the radial separation achieves its maximum equilibrium value), while at infinite circulation both the radial and axial separation tend to zero. However, at the optimum flow when the parameter m equals unity, the radial separation gradient is just halved and the axial separation achieves the maximum possible value of

$$
(\alpha \beta)_c = \exp\left[\frac{\Delta MV^2}{2RT} \frac{\sqrt{2}Z}{d} \sqrt{E}\right].
$$
 (37)

This solution for the ideal profile was originally derived by Cohen and Kaplan (1942), but not published until 1960. In the meantime it was derived independently by Los and Kistemaker (1958). Since the length-diameter ratio of a centrifuge rotor is much greater than unity, it is clear from Eq. (37) that it is possible to obtain a separation factor from end to end much bigger than the basic radial separation factor given earlier by Eq. (7).

However, this maximum value cannot be attained in a production centrifuge since, as always, it is necessary to have some feed, product, and waste flow to obtain an out-

FIG. 11. Variation of enrichment with internal circulation. The ordinate is $\ln[2m/(1+m^2)]$. The optimum separation factor for zero throughput occurs at $m =$ unity.

put of separative work. The equations for these conditions are also given by Cohen and are summarized here only for the simple case of machines with one feed stream. Two equations are necessary, one for the length of the rotor in which the light isotope is enriched —the "rectifier"—and one for the other length in which the light isotope is depleted—the "stripper." The equations for these two parts, illustrated in Fig. 10, are

$$
\alpha = \frac{1 + P_1}{P_1 + \exp[A_2 \mu (1 + P_1)]} \tag{38}
$$

$$
\beta = \frac{-W_1 + \exp[A_2\mu(1 + W_1)]}{1 - W_1} \tag{39}
$$

In using these equations care must be taken to conserve the mass balance of both isotopes—see Fig. ¹—and the total length of the centrifuge must be shared between the rectifier and the stripper sections such that the desired enrichment factor and depletion factor are obtained. Normally the enrichment factor and depletion factor are set equal, to avoid mixing losses in cascades in which the enriched and depleted fractions move up or down by only one stage.

For every given feed flow it is necessary, in order to maximize the separation factor, to increase the total value of the internal countercurrent flow to a new optimum value. The new optimum value of this circulation, defined as m_0 times the value for zero feed rate, is readily found by iteration using Eqs. (38) and (39), or, more easily at high flow rates, by the simplified but approximate equations⁴ of Olander (1972), given in his detailed review of the early work on centrifuges:

$$
L = \pi \rho D d^2 (1 + m_0^2) / 0.81 Z , \qquad (40)
$$

$$
\alpha\beta - 1 = [0.81A_2\mu_0/\sqrt{2}] \ . \tag{41}
$$

The separative work from these values, given by $L(\alpha\beta - 1)^2$ /8 and divided by the Dirac maximum, is

$$
\delta U/\delta U_{\text{max}} = 0.81 E m_0^2/(1 + m_0^2) \ . \tag{42}
$$

This final equation for the output of the countercurrent centrifuge shows that the overall efficiency depends on three factors. The factor 0.81 is a loss associated with the use of only one feed stream since the machine of Fig. 9(b) is effectively a square cascade with an inherent loss of 19%. The profile factor E , already defined, is to allow for the more general conditions of less efficient profiles. The final factor expresses the necessity, in a rather roundabout way, of operating with a finite feed rate. The output of the countercurrent centrifuge increases asymptotically with increasing feed rate, providing that at each feed value the internal circulation is reset at its optimum value. Thus, for a feed rate corresponding to $m_0 = 3$, say, the overall efficiency for the optimum profile is 73% .

⁴⁰lander's equations (40) and (41) are effectively the first terms of a power series of Cohen's equations suitably modified by the factors 0.81 and E .

In practice the efficiency factors cannot be separated out so easily because of the effects of axial decay of the countercurrent, a phenomenon to be discussed later. This axial decay of the countercurrent towards the end of the rotor diminishes the overall output by reducing the profile efficiency, an effect partially compensated for by reduced mixing losses in the decaying countercurrent.

However, these effects are neglected in the simplified equations of Cohen and Olander. A comparison of these two solutions is given in Table III for the two main machines developed during the Manhattan Project. As can be seen, the agreement is quite good, even at reasonably high enrichment factors. Thus it is normally sufficient to use the simple Olander equations, and they are particularly useful for comparing the countercurrent and concurrent modes of operation.

H. Comparison of countercurrent and concurrent machines

In the Manhattan Project both the countercurrent and concurrent modes of operation were tested experimentally at 270 m/sec, and both gave more or less the predicted outputs. Normally the machines were operated with a dual feed inlet, but for simplicity they will be compared here for the single-feed design. The comparison given here is taken from the paper by Cohen and Kaplan (1942). As described earlier, the concurrent design has an optimum value of feed rate, giving a maximum efficiency of 66% and a separation factor of about one-half of the basic radial separation factor. The countercurrent machine has an ideal efficiency of about 73%, but uses a much lower feed rate, and has a separation factor much greater than the basic radial separation factor. The effect can be quantified by dividing the equations for the countercurrent machine by the values at optimum conditions for the concurrent design. Thus

ratio of feed rates = $0.12(1+m_0^2)/[(Z/d)^2]$; ratio of feed rates = $0.12(1 + m_0^2) / [(Z/d)^2]$; (43)
ratio of separation factors = $1.6[2m_0/(1 + m_0^2)][(Z/d)]$.

 (44)

These equations confirm that the feed flow is smaller and the separation factor larger the greater the lengthdiameter ratio. Consider the two most important machines developed during the Manhattan Project, one with a subcritical rotor of length-diameter ratio of five, and one supercritical with a rotor three times longer. For the subcritical rotor, Eq. (43) gives the ratio of feed flows at $m = 3$ as twenty; for the longer supercritical rotor, the ratio of the flows is 180. The actual values are listed in Table III.

Thus the first major advantage of the countercurrent centrifuge is that it makes it much easier to extract the gas; not only is the flow very much smaller but, in contradistinction to the concurrent machine, the enrichment is from end to end, so that fractions can be removed from near the periphery where the pressure is higher. An important incidental advantage is noted by Cohen and Kaplan. Removal of the product and waste flows is one of the major causes of power loss in a centrifuge, so the considerable reduction in feed flow has major beneficial consequences for both machine and plant design.

The second major advantage of the countercurrent design is the increased separation factor, making possible great simplification in cascade shape. To appreciate this advantage it is not necessary to discuss cascade theory in detail, since the two most important parameters of the cascade, its size and length, can be determined by elementary algebra. First, using the concept of separative work, one obtains the total number of machines in a cascade by dividing the separative work of the cascade by the separative work of the individual machine. Second, since each stage of a cascade multiplies the abundance ratio by α going up the cascade (the rectifier) and divides it by α going down the cascade⁵ (the stripper), it follows that

$$
s = \ln \alpha_0 / \ln \alpha \tag{45a}
$$

$$
t = \ln \beta_0 / \ln \alpha \tag{45b}
$$

where s is the number of stages in the rectifier, α_0 is the total enrichment achieved by the cascade, t is the number in the rectifier, and β_0 is the total depletion of the cascade. Since the feed stage does one enrichment and one depletion, it counts twice, so the total length of the cascade is $(s+t-1)$.

Cohen and Kaplan discussed this advantage with reference to the wartime plants, but here it is more relevant to consider the 220-tonne plant described earlier. The theoretical separative power of the Manhattan long machine is 4.26 kg SW/yr operated in the concurrent mode and 4.70 kg SW/yr operated in the countercurrent mode. Dividing these values into the 220000 kg SW/yr of the plant shows that 52000 or 47000 machines would be required, respectively, for the two modes of operation. This is not much of a difference, but the difference in shape is considerable. Applying Eq. (45) shows that the concurrent machine would have to be arranged in a long narrow cascade of 157 stages, each effecting a change of concentration of only 1.014, whereas only 11 stages, each effecting a change of 1.20, are required for the countercurrent mode. Cohen and Kaplan decided that it would be difficult to control the long thin cascade of concurrent machines and opted for the countercurrent design in their final report in January 1942 to the Manhattan Project office.

To quote,

"We may say without exaggeration that the problem of running a successful cascade does not exist for countercurrent centrifuges. In general the advantages of the countercurrent centrifuge are its great flexibility, allowing us to determine widths, lengths, temperatures, and peripheral velocities on purely mechanical grounds; the elimination of the cascade problem; and the diminution

⁵This applies for the standard cascade case of $\alpha = \beta$.

			Countercurrent mode			
Length $= 1$ m			Length $=$ 3 m Enrichment factor			
Enrichment factor Feed rate			Feed rate			Optimum circulation
mg/sec	Cohen	Olander	mg/sec	Cohen	Olander	
17.3	1.089	1.088	5.77	1.281	1.263	\overline{c}
34.6	1.077	1.076	11.53	1.207	1.197	3
58.8	1.067	1.066	19.6	1.160	1.155	4
89.9	1.058	1.058	30.0	1.130	1.126	5
			Concurrent mode			
700	1.014		2100	1.014		

TABLE III. Comparison of exact and approximate solutions for rotors of 100 mm radius, $V=300$ m/sec, $T = 300$ K.

of the power loss in the bearings. Since pumping gas and friction are the sole expenditures of energy in the centrifuge plant, the countercurrent centrifuge separation plant will be the most economical to run, by several factors, of all the plants considered. And since operation of a cascade is the chief obstacle to the transition from smallscale to large-scale production, the countercurrent centrifuge is the most suitable apparatus for immediate production of $U^{235}F_6$."

IV. PRACTICAL FLOW PROFILES

A. Two-shell profile

Following the demonstration by Cohen and Kaplan of the great advantage of the countercurrent machine, all of the work in the period under review concentrated on optimizing this mode of operation. The optimum profile derived by Cohen is a mathematical ideal giving the best isotope diffusion between the opposing countercurrent flows. However, the three main experimenters, Beams, Groth, and Zippe, made no attempt to achieve this ideal profile; they used velocity profiles which aimed for the highest possible efficiency within the limitations set by the respective designs of their machines. The difficulty is that the flow patterns which actually occur within a centrifuge rotor are governed by hydrodynamic equations, none of which are concerned with isotope diffusion or even with the fact that the gas is a binary mixture.

The theoretical problem of solving these hydrodynamic equations was avoided in the Manhattan Project by the decision to use external pumps to circulate the gas through gas ports in the end caps, which confined the countercurrent to two opposing thin streams, as shown in Fig. 7. The efficiency of this type of profile, derived from the generalized Cohen integrals and given in Appendix C is

$$
E = \frac{\left[1 - (b^2/a^2)\right]^2}{\ln(a/b)} \ . \tag{46}
$$

Similarly, the internal circulation from the generalized integrals is

$$
S = S_0 / 2[\ln(a/b)]^{1/2} \,. \tag{47}
$$

At low speeds, when the pressure ratio is small, the inner shell can be positioned at the optimum radius ratio of b/a of 0.534, the same as for the two-shell concurrent machine. At this radius the profile efficiency is 0.81 and the optimum integral circulation at zero product rate is $0.61S₀$. The overall efficiency of this optimum two-shell profile is obtained from Eqs. (40)–(42) by setting E equal to 0.81; it is about 59% at $m_0 = 3$.

However, a problem with this two-shell profile is that to obtain the optimum mass flow at the position of the inner shell requires a high gas velocity because of the low pressure. For example, at 300 m/sec the density at the inner shell is only one-hundredth of that at the perimeter, and so the gas in this inner shell has to move at a very high axial velocity. As the speed is increased the pressure at the optimum radius of the inner shell becomes too low to maintain the required mass flow, and it is necessary to move the shell out to a position at a higher pressure. The effect is discussed in the review by Von Halle, Hoglund, and Shacter (1965) and can easily be quantified. Thus, if the pressure at the shell is set at one-hundredth of the peripheral pressure as in the Beam's machine (actually the pressure ratio was 94), the required radius of the inner shell is given from Eq. (6) as

$$
A^2[1-(b^2/a^2)] = \ln 100.
$$
 (48)

Inserting this value of b/a into the efficiency equation (46) gives the values listed in Table IV. The result at high speeds is very similar to that given by the simple equation (10). If the pressure at the inner shell is set at the same limiting pressure p_1 , then the first Taylor expansion of Eq. (48) is

$$
b/a = 1 - (v/2A^2) \tag{49}
$$

Inserting this into Eq. (46) gives the approximation

$$
E = 2v/A^2 \tag{50}
$$

This is the same as the first approximation of Eq. (10). Similarly, Eqs. (47) and (49) give the approximate internal circulation as

		Two-shell profile			Thermal profile		
V	A^2	Radius ratio	Flow ratio	Profile efficiency	Flow ratio	Profile efficiency	
300	6.35	0.534	0.63	81	1.94	70.2	
400	11.29	0.773	0.99	63	2.49	52.6	
500	17.64	0.862	1.30	45	3.06	37.8	
600	25.40	0.906	1.59	32	3.64	27.8	
700	34.57	0.932	1.88	25	4.22	21.2	
800	45.16	0.948	2.17	19	4.81	16.5	
900	57.15	0.959	2.46	15	5.39	13.3	
1000	70.56	0.967	2.74	12	5.98	10.9	

TABLE IV. Profile efficiency of two-shell and thermal profiles.

$$
S/S_0 = (A^2/2\nu)^{1/2} \ . \tag{51}
$$

This work on the two-shell profile, together with theoretical work by Cohen and the experimental work by Beams, virtually marks the end of the centrifuge development which can properly be ascribed to the Manhattan Project. The last theoretical work to be done during the project was that of Dirac on the stability of the axial countercurrent, and this will be described later.

B. Hydrodynamic theory

Most of the theoretical work in the German and Russian projects was concerned with the solution of the hydrodynamic equations which govern the shape and size of the axial countercurrent. There are six equations: the equations of motion for each of the three coordinates, the equations of conservation of mass and of energy, and the equation of state. There is the added complication that for high-speed centrifuges the gas conditions vary from a continuum at high Mach number at the periphery to free molecular flow at the center. These six equations are given and approximate solutions of them are discussed in more detail in American and British work by Parker and Mayo (1961) and by Whipple (1962).

However, important solutions of particular aspects of the theory were given earlier by Martin, Dirac, and Steenbeck. These early solutions were in two parts. The first part was concerned with flow conditions at the ends of the rotor, where there is good heat transfer between the end fittings and the process gas. The good heat transfer enables one to set up radial flows and so calculate the flow profile of the thermal countercurrent, as was shown by Martin (1950). The second part of the theory considers flow in the main body of the rotor, where there is poor heat transfer and radial flow is inhibited. Thus a countercurrent, once set up, will continue along a considerable length of the rotor and will decay away only slowly. The eventual decay is caused by the viscous drag between the various annular layers of the countercurrent, which travel along the tube at different axial velocities.

C. The thermal profile and friction profile

The solution of the hydrodynamic equations at the ends of the rotor involves boundary layer theory, a subject first studied by Prandtl in 1904 and now an established science. By a fortunate coincidence there were two early and exact solutions of boundary layer hydrodynamics of direct relevance to the centrifuge, which aid in understanding how axial countercurrents are set up. These two exact solutions of the Navier-Stokes equations are for the case of an incompressible fluid; the first, given by Cochran (1934), is for the flow of initially stationary fluid in the neighborhood of a rotating disc, and the second solution is for the converse case, solved by Boedewadt (1940), of rotating fluid impinging on a stationary surface. These solutions and the modifications by Martin and Whipple are discussed in more detail in Appendix D and are summarized here only insofar as they are relevant to separation efficiency.

Consider first the thermal countercurrent, which is set up by having one end cap hotter and the other colder than the circulating gas. The mechanism is that the gas near the hot end cap is heated and rises in the centrifugal field—moving radially inwards—and the gas at the cold end cap does the opposite. The net effect of these end flows is to set up an axial countercurrent, with an inner flow from the hot end to the cold end and with an outer opposing flow. The stream function, which is the total mass flow inside a given radius r , must equal the inner radial flow across the hot end cap at this same radius. This equality was used by Martin in his calculations. He first showed that the radial flow at the hot end cap is confined to a layer sufficiently thin for adequate heat exchange between the gas and the end cap. The radial velocity of this flow is of course zero at the cap surface, rising to a maximum within the boundary layer and then falling to zero again at distance δ_2 . Beyond this distance, as is normal in boundary layer theory, there are small decaying oscillations of the radial flow, but these can be safely neglected.

The method used by Steenbeck and Zippe for setting up the countercurrent is similar in principle, but instead of using a temperature perturbation to cause an inward flow at the ends of the rotor and a resulting axial flow, they used a perturbation of the rotational speed. In practice they used a stationary surface such as a disc at the drive end (equivalent to the hot end of a thermal drive), giving the effect indicated in Fig. 12. There is no exact solution for such a drive. However, for the present purpose it can be assumed that the friction drive is set up by having a

FIG. 12. Rotation near stationary disc, showing variation of gas velocities with distance from disc. Radial velocity= $r\omega\hat{F}$; tangential velocity = $r\omega \hat{G}$; axial velocity = $(\eta \omega / \rho)^{1/2} \hat{H}$. Most of the radial flow is within the boundary layer thickness given by Eq. (D3). The upward flow, above the boundary layer, tends to a constant velocity of $1.38(\eta\omega/\rho)^{1/2}$ and carries on indefinitely until dissipated by viscous forces. (From Schlichting, 1951, courtesy of McGraw-Hill.)

disc at each end of the rotor, one spinning slightly faster than the rotor and one slightly slower. Thus if the gas in the body of the rotor is at temperature T and spinning at angular velocity ω and (a) one cap is at temperature $T + \Delta T$ and speed $\omega - \Delta \omega$ and (b) the other cap is at temperature $T - \Delta T$ and speed $\omega + \Delta \omega$ then the stream function will be of the form

$$
h(r) = f(r)(\Delta T/T - 2\Delta\omega/\omega) \tag{52}
$$

The countercurrent in the central region of the rotor is from the hot and/or slow end, with the return flow in the opposite direction confined to a thin layer near the periphery.

If the perturbations are small it is possible to linearize the hydrodynamic equations and obtain approximate solutions for the stream function. At low speeds the solution of Boedewadt (1940) is reasonably accurate, whereas at high speeds that of Martin (1950) is more appropriate. Thus at low speed

$$
\psi_1(r) = f_1(r)(\Delta T/T - 2\Delta\omega/\omega) ,
$$

where

$$
f_1(r) = \pi r^2 (\eta \omega \rho)^{1/2} / 2 \;, \tag{53}
$$

and at high speed

 $\psi_2(r) = f_2(r)(\Delta T/T - 2\Delta\omega/\omega)$,

where

$$
f_2(r) = \pi(\kappa^3 T^3/\eta)^{1/4} (2r\rho)^{1/2}/\omega \ . \tag{54}
$$

With these stream functions, defined by Eqs. (52)—(54), it is easy to calculate the separation efficiency using the Cohen integrals. The calculations for the low-speed approximation are especially easy, and the results are given in Table IV. The calculations for the high-speed approximation involve numerical integration but are almost identical because in both stream functions the dominant variable is the square root of the density. A unified solution for both speeds was given by Whipple (1962).

As shown in Table IV, the profile efficiency is reasonably good at low speeds but falls off rapidly at higher speeds, tending in the limit to $8/A^2$. The reason the efficiency falls off with speed is much the same as that described for the two-shell profile; too much of the useful axial countercurrent is concentrated near the periphery. This is because the velocity profile of the thermal countercurrent, obtained by differentiation of the stream function, is one in which the axial velocity is approximately proportional to the inverse of the square root of the pressure instead of the inverse of the pressure, as required for optimum separation. Thus, moving inward from the periphery, the axial velocity increases less quickly than required for optimum separation. The combination of the low axial velocity and the low pressure near the axis means that the central region of the rotor contributes hardly any useful mass flow to the axial countercurrent, and hence hardly any separation. The effect is much the same as that given earlier using the Bredig solution in Eq. $(10).$

V. STABILITY OF THE COUNTERCURRENT

A. Stabilizing forces

All of the three practical countercurrents described, i.e., the two-shell profile used by Beams, the thermal counter-

current used by Groth, and the friction drive used by Steenbeck and Zippe, are set up at the ends and it is assumed in the calculation of the countercurrent efficiency that the circulatory flow remains of constant shape and magnitude as it progresses along the rotor. In fact, of course, the flow patterns decay as they progress along the rotor, due to viscous drag between the layers. However, the effect is very slow because there are two powerful stabilizing forces sustaining the countercurrent for a reasonable distance along the rotor. One force is based on the conservation of angular momentum, this being more important at low peripheral speeds, and the other force is based on the conservation of energy, and this is the more important at high speeds.

The first stabilizing force is the one used in the early theory of Dirac (1940). Its magnitude is easily calculated from the principle of angular momentum. Thus if a packet of gas of volume Q strays inwards from its correct radius its angular velocity will increase, giving it an enhanced centrifugal force pushing it back again:

(a) Angular momentum $I\omega = (Q\rho)r^2\omega$,

$$
d(I\omega)/dr = (Q\rho)(2\omega r + r^2 d\omega/dr) = 0,
$$

\n
$$
d\omega = -(2\omega/r)dr.
$$

(b) Centrifugal force $F_1 = (Q\rho)r\omega^2$,

$$
dF_1 = 2(Q\rho)r\omega d\omega
$$

= -4(Q\rho)\omega^2 dr . \t(55)

This small restoring force for a small displacement corresponds to the standard equation for simple harmonic motion; it shows that if a packet of gas is displaced from its equilibrium position it will oscillate at an angular frequency equal to twice the angular speed. The particle of fluid thus moves back to its original position twice per revolution. This same result is given by Tritton (1977) using a rather more exact physical picture by consideration of the Coriolis force.

The second stabilizing force, based on the conservation of energy, is due to the change of density caused by adiabatic cooling or heating and is well known in the theory of the atmosphere. If the same packet of gas moves inwards in the main body of the centrifuge, away from the end walls, there is no way it can exchange heat with its new surroundings, so its temperature will fall according to the standard adiabatic equation:

$$
\Delta T/T = [(\gamma - 1)/\gamma] \Delta p / p
$$

= [(\gamma - 1)/\gamma] \delta r / H = \Delta \rho / \rho . \t(56)

This is equivalent to the well-known expression for the "adiabatic lapse rate," which in the terrestrial atmosphere is 12 C per km but in a high-speed centrifuge can be as high as 15 °C per mm. The associated change of density $\Delta \rho$ caused by the cooling gives the packet of gas a considerable force restoring it to its original position. This restoring force is given by the equation

adiabatic force =
$$
Qr\omega^2 \Delta \rho
$$

= $Q\rho r \omega^2 [(\gamma - 1)/\gamma] \Delta r / H$. (57)

The ratio of the adiabatic to the Dirac force is given by

$$
B = (\gamma - 1)M\omega^2 r^2/4\gamma RT \ . \tag{58}
$$

This number relates all the high- and low-speed approximations of boundary layer theory, and equals unity for $UF₆$ at about 700 m/sec, the exact speed depending on the temperature of operation. Thus above 700 m/sec the adiabatic force is larger than the stability force, due to the conservation of angular momentum, and vice versa.

B. Decay lengths

In Dirac's first investigation of this problem he neglected the adiabatic force and made the further simplification of assuming that nearly all of the gas is near the periphery, which is true only at high speeds. This approximation is an artifice for treating the problem as one in planar geometry and writing the equation for the pressure distribution, Eq. (6), in the simpler form

$$
p(h)=p(a)\exp(-h/H), \qquad (59)
$$

where H is the height of the homogeneous atmosphere or the scale height. Making this high-speed approximation and neglecting, slightly inconsistently, adiabatic effects, Dirac searched for flow patterns which stayed the same shape but whose amplitude decayed exponentially with axial distance along the rotor. These natural or eigenflow profiles formed a set of flow patterns, illustrated in Fig. 13, from which other more complicated patterns could be built up.

In his paper Dirac gave the decay length only of the first natural flow pattern. He showed that this flow pattern decreased in magnitude by a factor of e in a distance

$$
Z_D = p a^2 R^2 T^2 / 1.2 \eta V^5 \,. \tag{60}
$$

This is the first equation in centrifuge theory to show a dependence on the absolute pressure p at the periphery of the centrifuge and on the diameter, and so give some information regarding the best values of these parameters. The same equation was also derived independently by Steenbeck (1958) during Soviet work on the centrifuge. Steenbeck quotes his results in the form

FIG. 13. Approximate flow profile of first three eigenmodes (from Steenbeck, 1958).

$$
Z_D = \rho V a^2 / 2\eta C \tag{61}
$$

where the parameter C is a constant for each eigensolution. According to Steenbeck, the constant for the first natural flow pattern tends at high speeds to the value 4.82 $A⁶$. Rearrangement of the parameters shows that this is the Dirac solution. Steenbeck has also given the decay length for lower values of the speed parameter A, summarized in Fig. 14.

Although Steenbeck was unable to calculate the decay lengths for the higher modes for a compressible fluid, he did so for the incompressible case, giving values of C of 20.7, 99.6, 300, and 550 for the first four modes. As shown by these constants, the higher modes decay much more quickly than the fundamental-for example, quoting from Steenbeck a 10-mm-diam tube filled with water and spinning at 5 rev/sec has decay lengths of 94, 19, 6, and 3.5 m for the first four modes. Thus no matter what the initial flow profile it will rapidly decay to the first eigensolution, and then this itself decays exponentially.

Both Dirac and Steenbeck recognized that their calculations underestimated the decay lengths, since they neglected the stability due to adiabatic effects. This omission in the theory was rectified by more exact calculations performed at the University of Virginia, by Ging (1962) using an analytical solution and by Parker and Mayo (1963) using numerical solutions. Ging, still using the high-speed approximations, showed that the Dirac decay length should be increased by a factor $(1+B^2)^{1/2}$ where B is the ratio of the adiabatic and angular momentum forces.

The numerical solutions of Parker and Mayo were far more general than the analytical solutions, in that they required no serious simplifications, particularly the highspeed approximation. In general, however, the calculations confirmed the correctness of the Dirac equation as modified by Ging, which gives the decay length of the first characteristic flow pattern. This numerical work also showed that the higher modes decayed away at approximately the same relative rate as that given by Steenbeck for an incompressible fluid—the numerical solutions give a decay length for the first, second, and fourth modes in the ratio of $1:5.6:-:34$ in fair agreement with the

 $10⁵$ 104 $\mathbf 0$ $\bar{\rm E}_{10^3}$ $\frac{2}{3}$ 10² 10

FIG. 14. Variation of Steenbeck decay factor with speed (from Steenbeck, 1958).

SPEED FACTOR A

7

5

0 2 3

Stennbeck ratio of 1:4.8:14.5:27. (The decay length of the third mode was not given.)

It is perhaps surprising that the decay length of the countercurrent decreases rapidly with peripheral speedas $1/V^3$ —since both of the stabilizing forces increase with speed. At least part of the explanation is that, as the speed is increased, the shape of the characteristic flow pattern changes as it is confined more and more to the outer periphery. Numerous calculations by Parker show that the interface between the opposing flows of the countercurrent is about 1.4 scale heights in from the rotor wall. Since this distance decreases rapidly with speed—as $1/V^2$ —the velocity gradient and dissipative viscous force increase with peripheral speed.

A simplified understanding of this changing shape of the characteristic flow pattern can be given as follows. Consider first a rotor spinning with no axial perturbation. The pressure distribution will then be the same at all axial positions, and there is no driving force for the axial flow. Then partition off each axial section and superpose a linear perturbation of the term $MV^2/2RT$ along the rotor by means of, say, a temperature gradient. The pressure distribution will then change in each partitioned section according to the standard equation (6), the gradient being greater in the colder sections. Differentiation of the pressure equation, at constant hold-up per section, shows that the pressure in each compartment is unaltered at the radial position in the rotor where the pressure has fallen by e. Then, if the partitions are removed, gas will flow up and down with the interface at this position, which is just one scale height in from the wall, a bit closer to the wall than calculated by Parker.

C. Profile efficiency

The most important result of Parker and Mayo in the present context is that given for separation efficiency. They calculated the efficiency, using the Cohen integrals, for a countercurrent equal in profile to that of the first characteristic flow pattern but assuming that it had a constant and optimum value along the length of the rotor. The change of shape of the fIow pattern as it gets pushed to the outer wall results in a change of efficiency with speed similar to that of the thermal profile. The results, given in Fig. 15, are slightly lower than those of the thermal profile and according to Yon Halle (1980), tend at high speed to 7.2/ A^2 , about 10% less than the corresponding equation for the thermal profile given earlier.

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APPENDIX A: SEPARATIVE WORK AND CASCADE THEORY

1. Separative work

Probably the most important theoretical result in centrifuge technology is that of Dirac (1941), who showed

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FIG. 15. Profile efficiencies for various velocity profiles —the thermal profile using the Martin (1950), Eq. (54), the eigensolution of Parker and Mayo (1963), and the low-speed approximation for the thermal profile, Eq. (53). The crosses give the twoshell efficiency given in Table IV (adapted from Von Halle et al., 1965).

that, irrespective of the mode of operation, a centrifuge has a maximum possible value of output given by Eq. (3) in the text. The output of a centrifuge is usually called "separative work," and its rate of output is called "separa-"separative work," and its rate of output is called "separa-
tive power." These functions are not easy to define, but clearly the output is proportional to the throughput and is some function of the change of enrichment achieved.

At first sight the obvious function to use to define the usefulness of the separating process is the decrease in entropy that has been achieved. This, after all, is the main object of the process, to achieve the order of separated isotopes from the disorder of the mixture of isotopes. Unfortunately the decrease of the entropy of mixing cannot be used as a measure of the separative effort because a separating unit costs as much and uses as much energy whether it is fed with a gas containing a mixture of isotopes or with a gas containing only one isotope. The centrifuge can only separate isotopes in proportion to the probability of its being able to find and pair unlike molecules, and this is $N(1 - N)$. It is then possible, following Fuchs and Peierls (1941), to modify the entropy change and define separative work for L moles of gas as

$$
\Delta U = L \Delta S / RN(1 - N) . \tag{A1}
$$

The entropy change is divided by the gas constant R to make it dimensionless. The units of ΔU are then the same as the units of L . This terminology is to some extent misleading, since separative work has nothing to do with the work or energy required to separate the isotopes.

However, rearranging the equation immediately gives the minimum energy required per unit of separation work, as

$$
T\Delta S / \Delta U = RTN(1-N)
$$

= a maximum of $RT/4$. (A2)

This amount of energy, which is very small, assumes that the process is thermodynamically reversible. The actual energy used in real processes is much greater, as will be discussed later, after the application of the Peierls equation to enrichment systems.

2. Entropy and separative work assuming small enrichment factor

Consider first a separation process of small enrichment factor, which converts L moles of feed into $L/2$ moles of product of increased concentration Δ and $L/2$ moles of waste of decreased concentration Δ . The entropy of mixing of one mole of the feed gas is

$$
S = -R [N lnN + (1 - N)ln(1 - N)] .
$$
 (A3)

Therefore, the change in entropy achieved during the separation is given by eved during the
 $\sum S(N - \Delta)$

$$
\Delta S = L S (N) - \frac{1}{2} L S (N + \Delta) - \frac{1}{2} L S (N - \Delta)
$$

= $\frac{1}{2} L R \Delta^2 d^2 S / dN^2$
= $\frac{1}{2} L R \Delta^2 / N (1 - N)$. (A4)

This result is obtained by the Taylor expansion

$$
S (N + \Delta) = S (N) + \Delta d S / dN + (\Delta^2 / 2) d^2 S / dN^2 \cdots
$$

The Peierls equation (Al) then gives

$$
\Delta U = \frac{1}{2} L \Delta^2 / N^2 (1 - N)^2 \ . \tag{A5}
$$

Simple algebraic manipulation shows that the increase in product concentration and the decrease in waste concentration are related to the separation factor by the equation

$$
\Delta = (\alpha - 1)N(1 - N) = \varepsilon N(1 - N) .
$$
 (A6)

This finally gives

$$
\Delta U = \frac{1}{2} L (\alpha - 1)^2 = \frac{1}{2} L \varepsilon^2
$$
 (A7)

3. Entropy and separative work assuming large enrichment factor

These equations are valid only for small changes of concentration, when the concentration N in Eq. (A1) is effectively constant. In the process of achieving a larger separation, the concentrations N and $(1 - N)$ are gradually changing, so to get the total effect it is necessary to divide the differential change of entropy at each stage by $N(1 - N)$ and then reintegrate to get the total separative effort. However, just as Eq. (A4) was derived from the second derivative of the entropy function, it is permissible to consider Eq. (A5) as the second derivative of a new function, to be called the "value function." Thus integrating the function $1/N^2(1-N)^2$ twice with respect to N gives

$$
\mathscr{V}(N) = (2N-1)\ln[N/(1-N)] \tag{A8} \qquad \qquad (A8) \qquad \qquad T\Delta S = LRT \ln f \tag{A14}
$$

Provided one is interested only in changes of this function, the arbitrary constants of integration are unimportant. The separative work in a process can now be defined in a more general way as

$$
\Delta U = P \mathscr{V}(P) + W \mathscr{V}(W) - L \mathscr{V}(L) . \tag{A9}
$$

This equation can be used as it stands, but a useful simplification can be derived for the symmetrical system shown in Fig. 1. Simple algebraic manipulation shows that in this case the product flow, waste flow, and separative work are given by

$$
P = [L/(1+\alpha)][1+(\alpha-1)N], \qquad (A10)
$$

 $W = [\alpha L/(1+\alpha)][1+(1/\alpha-1)N],$ $(A11)$

$$
\delta U = L\left[(\alpha - 1)/(\alpha + 1) \right] \ln \alpha . \tag{A12}
$$

If $(\alpha - 1)$ is small Eq. (A12) tends to the value given by Eq. (A7).

4. Energy usage in separation

The main advantage of the centrifuge plant over the diffusion process is its much lower energy usage. This advantage is so great that the diffusion plant in the UK has now been closed down, even though its capital has been written off. It is important, therefore, to understand the main physical principles involved in the energy usage of these two processes.

First, consider the entropy of mixing, Eq. (A3). This can be derived in a variety of ways, but the proof given by Roberts (1928) is important in the present context. He shows, using an experiment of principle which employs idealized semipermeable membranes, that the energy required to separate isotopes completely is zero if the separated components end up at their original partial pressures. The entropy change given by Eq. (A3) is that required to recompress each gas back to the original total pressure. This energy, for the complete separation of L moles of gas mixture, has a maximum value at $N = 0.5$ of

$$
\Gamma \Delta S = LRT \ln 2 \tag{A13}
$$

In the real cases of the diffusion plant and the centrifuge the energy used per separating element is similar to this value, but the separation of each element is very small, so the energy used per unit of separative work is much greater than the theoretical minimum.

5. Diffusion plant

The diffusion process—more strictly, an effusion process—is intrinsically irreversible because of the free molecular expansion of the gas across the minute oriflces of the separating barrier. The energy consumed per element is given by the standard equation, similar to Eq. (A13), as

$$
T\Delta S = LRT \ln f \tag{A14}
$$

where f is the pressure ratio across the orifice. This gives the power usage. Consider now the separation. Forward effusion will give separation according to the law of equipartition of energy, i.e., Graham's law, and backward effusion will tend to diminish the separation. Thus

 $\varepsilon = \varepsilon_0(1-f)$

where

$$
\varepsilon_0 = \frac{1}{2} (M_2 / M_1)^{1/2} = \Delta M / 4M \; .
$$

Thus, from Eq. (A7), the separative work is

$$
\Delta U = \frac{1}{2}L\,(1-f)^2\varepsilon_0^2\;.
$$

Dividing out and optimizing for the pressure ratio, at $f = 0.285$, gives the minimum power for effusion as

$$
\Gamma \Delta S / \Delta U = 2.5RT / \epsilon_0^2. \tag{A15}
$$

This simple equation is about double the correct value for a properly optimized diffusion plant—for example, see London (1961)—but the difference is unimportant compared with the fact that the value for a diffusion plant is 1.1×10^6 times larger than the minimum value given by Eq. (A2) for a reversible process.

6. Centrifuge process

The separation of isotopes by a centrifuge is often assumed to be a reversible process, but in practice the power consumption is many orders of magnitude greater than the minimum value. In a production centrifuge the gas has to be accelerated to full speed, the countercurrent must be maintained, and the gas must be extracted from the rotor. All of these processes involve energy losses of the order RT per mole. Interestingly, the energy involved in setting up the pressure gradient in the atmosphere (equal to the potential energy of all the molecules) is also RT per mole of gas.

The energy needed to accelerate one mole of gas to full angular speed is easily calculated as

$$
T\Delta S = LRT \ln 2. \qquad (A13) \qquad T\Delta S = (A^2 - 1)RT. \qquad (A16)
$$

This energy of rotation could, in principle, be recovered, but this is not true regarding the countercurrent flow, which dissipates energy in the viscous boundary layers. The magnitude of the flow, given in the text for the ideal, two-shell, and thermal countercurrents, is generally greater than the feed flow, but the change in energy is rather less because, as the gas in the countercurrent proceeds from its outer radius to the inner radius, it does not lose all its kinetic energy (about 50%, 75%, and 67%, respectively, for the ideal, two-shell, and thermal profiles).

It is thus reasonable to accept the value given by Eq. (A16) as a guide to the power usage. Dividing out gives

$$
T\Delta S / \Delta U = 2A^2RT/\varepsilon^2.
$$
 (A17)

Even at low speeds such as 300 m/sec, when ε would be only about 0.1, this equation gives a power usage over 100 times better than the diffusion plant, but still $10⁴$ times higher than the theoretical minimum. In practice the power usage in the centrifuge can be another order of magnitude higher because the above analysis neglects the power dissipated in the bearings.

7. Cascade

The main use of the separative work function is that it gives, by simple calculation, the total number of machines required for a plant. However, it gives no indication of how to connect them together in cascade to give the correct output. The mathematical details of the design of cascades have been given by Cohen (1951), so only the physical principles will be given here. The algebra is particularly easy if both the cascade and the centrifuge are symmetrical, as illustrated in Figs. ¹ and 16. Thus, for the symmetrical machine, the product flow, waste flow, and separative power are given by Eqs. $(A10)$ — $(A12)$. By analogy with these equations, and substituting α^3 for α , one gets the various flows and the separative power of the $1:2:3:2:1$ cascade of Fig. 16 as

$$
feed flow = K mole/yr, abundance J \t(A18)
$$

product flow=K/(1+ α ³) mole/yr, abundance α ³J

 $(A19)$

(A20)

waste flow= $\alpha^3 K(1+\alpha^3)$ mole/yr, abundance J/α^3

separative power =
$$
K[(\alpha^3 - 1)/(\alpha^3 + 1)]\ln \alpha^3
$$
. (A21)

Dividing the separative power of the cascade by that of the machine gives the number of machines required:

No. of machines =
$$
3\frac{\alpha+1}{\alpha-1}\frac{\alpha^3-1}{\alpha^3+1}\frac{K}{L}
$$
. (A22)

The correctness of Eq. (A22) in giving the total number of machines can be confirmed by direct cascade calculation. The sums are very easy if $(\alpha - 1)N$ is small, so that the cut equals $1/(1+\alpha)$ —see Eq. (A10). The method is indicated in Fig. 16. The feed stage C enriches the light iso-

FIG. 16. Simple symmetrical cascade.

tope by a factor of α , stage B by another factor α , and the product stage by another factor, giving a product enriched by α^3 relative to the feed. Similarly stages C, D, and E each deplete the feed by α , giving a total factor α^3 . Notice that the feed stage is part of the enriching section (rectifier) and also part of the depleting section (stripper).

The flows in Fig. 16 are easily derived, starting from the product stage, with the mass balance rule that the flows in the product, feed, and waste of each stage must be in the ratio 1: $(1+\alpha)\alpha$. Using this rule ensures that the centrifuge operates symmetrically, so there is no mixing of flows of different isotopic mixtures. For example, the feed for stage B is the waste of stage A mixed with the product of stage C, and these clearly are of equal isotopic abundance if each stage enriches by α and depletes by α . Inspection of Fig. 16 also shows that the net flow moving to the left across any imaginary line between the stages equals the product flow (unity), and the net flow moving to the right is equal to the waste flow α^3 . At the common feed stage at the junction of the rectifier and stripper, an external feed must be provided equal to the product and waste. Note that the flows are given for a product rate of unity; therefore all the flows must be multiplied by K/L (1+ α ³) to be correct for the present example.

The summation of all the feed flows in the cascade clearly confirms that the total number of machines required in the cascade just equals the total calculated from the separative power calculation, Eq. (A22), given earlier. Simple extrapolation of the numbers given in Fig. 15 and listed in Table V shows that for a symmetrical cascade of $(2n - 1)$ stages, the summation of flows gives the required number of machines as

$$
\Sigma = n \frac{(\alpha+1)(\alpha^n-1)}{(\alpha-1)(\alpha^n+1)} \frac{K}{L} .
$$
 (A23)

Actually Cohen and Dirac followed the logic given in this appendix in reverse. They first calculated the equations for an ideal cascade, then summed up for the total flow, and from this derived the appropriate equation for separative work, making sure that the definition of separative work was independent of the concentration of the feed flow. Full details are given by Cohen (1951).

APPENDIX 8: PROOF OF ISOTHERMAL CONDITIONS IN A CENTRIFUGE

Most modern expositions of the law of the Most modern expositions of the law of the
tmosphere—e.g., that of Jeans (1916)—start with the Maxwell-Boltzmann distribution law. This law states that the number of molecules at a given height (or radius in a centrifuge) is proportional to $exp(-E_1/kT)$, where the potential energy E_1 is m_1gh or $\frac{1}{2}m_1^2r^2$ for a given molecular species in the atmosphere and centrifuge, respectively. This immediately confirms Eq. (5) for the pressure distribution in the atmosphere and Eq. (6) for the pressure distribution in the centrifuge. Moreover, this distribution applies to each molecular species separately,

 $(B1)$

TABLE V. Size of stages in symmetrical cascades.

so confirming Dalton's law and the separation equation (7).

To confirm the isothermal hypothesis it is necessary to apply the Maxwell-Boltzmann law in a more general way. In kinetic theory temperature is a statistical concept with a one-to-one relationship between temperature and the energy of the molecules. Equilibrium conditions occur more rapidly at increased density because of increased collision frequency (Kennard, 1938), but otherwise density is not involved in defining temperature. When the equilibrium state is set up between the translatory and other forms of vibrational and rotational molecular energy, the temperature of the gas is a function only of its translational energy. Thus, considering the translatory and potential energy together, the energy of a molecule can be written as

$$
E_2 = \frac{1}{2} m_1 c^2 + m_1 g h
$$

 α

$$
E_2 = \frac{1}{2} m_1 c^2 - \frac{1}{2} m_1 \omega^2 r^2
$$

in the atmosphere and in the centrifuge, respectively.

If these energy equations are substituted into the Maxwell-Boltzmann distribution law, the two terms separate, showing that at each position in the force field the molecular velocities are unaltered; only the number, and hence density and pressure, falls with height or radius. Thus at each position in the force field the Maxwell distribution law of velocity still holds, and so conditions are by definition isothermal.

There is an apparent paradox in this reasoning, which is discussed by several authors (e.g., Slater, 1939). This is that, as molecules move upwards to points of higher potential energy, they might be expected to lose speed and have a lower kinetic energy and hence a lower temperature than those at low altitude. In fact, individual molecules do lose speed on moving upwards (or gain speed if moving downwards), but detailed calculations show that the arrivals at the new height conform to an overall Maxwellian distribution law of uniform temperature, independent of height, but at a reduced density. This is because the slower molecules at low altitude never reach the high altitude at all. They follow parabolic paths and turn back. Only the fast molecules penetrate to the high regions, and in doing so they are slowed down sufficiently so that their original high velocities are reduced to the correct velocity for isothermal conditions to hold. Thus the average velocity at the high altitudes just equals that at the lower altitudes. For this to be exactly true requires a close interrelationship between the Maxwellian distribution of velocities and the barometric equation, and indeed either can be deduced from the other (Herzfeld, 1929).

APPENDIX C: THE COHEN INTEGRAL

The Cohen equation (34) gives the efficiency for any given flow pattern in a countercurrent centrifuge and Eq. (35) gives the optimum value of the circulatory flow for that flow pattern. Several flow patterns are discussed in the review and in three cases the Cohen integrals can be given analytically. The first two cases, the optimum and two-shell profile, are mathematical specifications and the integrals for these two profiles are listed in Table VI. The third case is the low-speed approximation of the thermal profile. The integrals for this profile are listed in Table

Integrals	Ideal profile	Two-shell profile
$\boldsymbol{\psi}$	kr^2	$r=0$ to b, $\psi=0$
		$r = b$ to a, $\psi = S$
ϕ_1	ka ²	
ϕ_2	$ka^4/4$	$S(a^2-b^2)/2$
ϕ_3	$k^2a^4/4$	S^2 ln(a/b)
E		$(1-b^2/a^2)^2/\ln(a/b)$
S/S_0		$1/2[\ln(a/b)]^{1/2}$

TABLE VI. Mathematical profiles.

VII. Negative exponential terms are neglected in the final two expressions for efficiency and flow.

APPENDIX D: PRINCIPLES OF BOUNDARY LAYER THEORY APPLIED TO THE CENTRIFUGE

1. Low-speed approximation

The complete solution of the hydrodynamic equations in a centrifuge was not obtained in the early work on the centrifuge, but the theories of Martin, Whipple, Ging, and Parker and Mayo clearly showed the importance of boundary layer formation on the various surfaces. The formation of the boundary layer and the resulting axial countercurrent are particularly easy to understand for the two important exact solutions of the Navier-Stokes equations for a rotating disc in stationary fluid and the converse case of rotating fluid hitting a stationary surface. This second case, which is almost a direct analogy with the friction drive mechanism in a centrifuge, is demonstrated by a well-known atmospheric phenomenon, the upward axial draught in a whirlwind being caused by rotating gas being slowed down by a stationary surface, spiralling inwards, and then fiowing axially upwards.

The mathematical solutions are described in several textbooks, e.g., that of Schlichting (1951), which includes the Boedewadt (1940) solution for a rotating gas and the Cochran (1934) solution for the rotating disc. The simplified solutions giveri here are not rigorous and are intended

only to describe the physical principles.

For example, consider the case of the rotating disc. The layer of fluid near the disc is carried by it into a circular motion through the effects of viscous drag and is then thrown outwards by centrifugal force. The fluid is ejected at an angle of about 40° to the circumferential direction, and its average outward velocity v_1 in the boundary layer is therefore much the same as the average circumferential velocity of $\frac{1}{2}\omega r$. The radial flow is replenished by fluid that flows in the axial direction towards the disc, to be, in turn, carried by the disc and ejected outwards, thus setting up an axial countercurrent. The thickness δ_1 of the layer of fluid which can be carried by the disc clearly increases with the viscosity of the fiuid; this can be seen by balancing the viscous force trying to keep the fluid in a circular orbit with the required centripetal force. Thus between r and $r + dr$ one gets

$$
\eta 2\pi r \, dr \, \omega r / \delta_1 \sim 2\pi r \, dr \, \delta_1 \rho r \omega^2 \,,
$$
\n
$$
\delta_1^2 \sim \eta / \rho \omega \, .
$$
\n(D1)

As described above, the mean velocity ν with which fluid is ejected outwards in this boundary layer is $\sim \frac{1}{2}\omega r$. This gives total ejected mass flow up to radius r (the stream function) as

$$
\begin{aligned} \n\psi_1 &= 2\pi r \delta_1 v_1 \rho \\ \n&= \pi r^2 (\eta \omega \rho)^{1/2} \ . \n\end{aligned} \tag{D2}
$$

The exact equations given by Cochran for the spinning disc and Boedewadt for the spinning gas are not much different. For example, in the Boedewadt analysis the gas is stationary on the end cap (or ground), rising to a maximum at distance $\frac{1}{2}\delta_1$ and then falling to zero again at distance δ_1 . Beyond this distance are small decaying oscillations of radial flow as shown in Fig. 11. The constants in the various equations are

$$
\delta_1 = \pi (\eta / \rho \omega)^{1/2} \tag{D3}
$$

$$
v_1 = 1.38 \omega r / 2\pi , \qquad (D4)
$$

$$
\psi_1 = 1.38\pi r^2 (\eta \omega \rho)^{1/2} . \tag{D5}
$$

The Cochran solution is a little more difficult, in that the velocity peaks at the distance given by Eq. (Dl) and decays slowly to zero after four or five boundary layer distances. However, the total integrated flow is not much different and is

$$
b_1 = 0.886 \pi r^2 (\eta \omega \rho)^{1/2} \ . \tag{D6}
$$

Taking the mean of Eqs. (D5) and (D6)—this by chance is approximately equal to Eq. (D2)—and assuming the stream function to be linear with the function $(\Delta T/T - 2\Delta\omega/\omega)$ gives the equation for the stream function for low speeds given in the text.

2. High-speed approximation

The solutions above are applicable to a compressible fluid in a centrifuge only at low speeds. At higher speeds, the change of density as the gas rises or falls in the strong centrifugal field gives rise to heating and cooling effects of the gas and to strong buoyancy forces. These effects, as well as the usual viscous forces, limit the flow in the boundary layers—this is because it is necessary to exchange heat between the end cap and the gas. The gas must be warmed and caused to rise (move inwards) at one end cap and be cooled and caused to fall (move outwards) at the other. Thus as gas comes along from the periphery and moves radially inwards at, say, the hot end, the necessary heat transfer can only extend over a small distance, thereby limiting the thickness of the radial flow, and the flow itself. The first solution of this problem was given by Martin (1950) during the early centrifuge project in Germany and has been discussed by Olander (1972). Martin gave the equation for the flow, which can be written in the form

inward radial velocity= $2\pi v_2 \exp(-\pi z/\delta_2)\sin(\pi z/\delta_2)$.

This is a typical boundary layer equation; the velocity is zero at the end wall, at $z = 0$, rises to a maximum value, falls to zero again at a distance equal to the boundary layer distance δ_2 , and then rapidly decays away. (It actually peaks at about $\frac{1}{4}\delta_2$, at about double the mean velocity v_2 . Martin gives the equations for the boundary layer thickness and the mean velocity is

$$
\delta_2 = \pi (2/r \rho)^{1/2} (\eta \kappa T)^{1/4} / \omega , \qquad (D7)
$$

$$
v_2 = (\kappa T/\eta)^{1/2} (\Delta T/T - 2\Delta\omega/\omega)/2\pi , \qquad (D8)
$$

giving

$$
\psi_2 = 2\pi r \delta_2 \rho v_2 \tag{D9}
$$

which gives the equation for the stream function in the main text.

3. General solution

The low-speed and high-speed solutions above are related to each other by the nondimensional group B defined in the text. Thus, denoting the low-speed and high-speed solutions with subscripts, we have

$$
B = \left[\frac{\delta_1}{\delta_2}\right]^4 = \left[\frac{v_1}{v_2}\right]^2 = \left[\frac{\psi_1}{\psi_2}\right]^{4/3}.
$$
 (D10)

The generalized solution, which includes both the low-

 $\psi_1 = 0.886 \pi r^2 (\eta \omega \rho)^{1/2}$. (D6) and high-speed solutions, has been given by Whipple (1962) and can be written

$$
\frac{1}{\delta^4} = \frac{1}{\delta_1^4} + \frac{1}{\delta_2^4} , \qquad (D11)
$$

$$
\frac{1}{v^2} = \frac{1}{v_1^2} + \frac{1}{v_2^2} ,
$$
 (D12)

$$
\frac{1}{\psi^{4/3}} = \frac{1}{\psi_1^{4/3}} + \frac{1}{\psi_2^{4/3}} , \qquad (D13)
$$

$$
\alpha
$$

$$
\frac{1}{1+B} = \left(\frac{\delta}{\delta_1}\right)^4 = \left(\frac{v}{v_1}\right)^2 = \left(\frac{\psi}{\psi_1}\right)^{4/3}.
$$
 (D14)

As described in the text, the velocity of the radial flow on the end walls must be zero on the surface and zero again some distance away. Nearly all the radial flow occurs in the boundary layer of thickness δ . The same phenomenon also occurs near the perimeter of the rotor because the velocity of the axial countercurrent is zero on the rotor wall. The thickness of the boundary layer on the side walls is a maximum at the rotor midplane and is given by Whipple, for the antisymmetric conditions defined by Eq. (52), as

$$
\delta_W = (Z\eta/2\pi\rho\omega)^{1/3}(1+B)^{1/6} \ . \tag{D15}
$$

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