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ELEMENTARY INTRODUCTION TO ISOTOPE SEPARATION

Dennis Holliday and Milton S. Plesset

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The RAND Corporation
SANTA MONICA • CALIFORNIA

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PREFACE

The separation of uranium isotopes is an important part of both civilian and military nuclear programs. Slightly enriched uranium is used in power reactors, and very highly enriched uranium is an essential component in certain types of nuclear weapons. The rapid growth of power reactor facilities and the growing possibility that additional nations will undertake nuclear weapons programs makes an elementary understanding of uranium isotope separation essential for anyone who studies the technical and economic aspects of these problems. Unfortunately, a simple discussion of isotope separation devices and their use in separation cascades is not readily found in the nuclear energy literature. This circumstance led to the preparation of notes to provide a brief technical background on isotope separation for the RAND research staff. The resulting notes appear to be of wide enough interest to warrant their publication in the present form.

The material is presented in a concise way which emphasizes the physical principles instead of the mathematical theory. Six different separation processes are discussed: the separation nozzle, sweep diffusion, thermal diffusion, electromagnetic separation, gaseous diffusion, and gas centrifugation. The steady-state theory of isotope separation cascades is also discussed. Numerical examples illustrating the performance of separation devices both individually and in cascades are given. In particular, examples of ideal and "squared-off" gas centrifuge cascades are presented.

Dr. Plesset is a consultant to The RAND Corporation and a professor at the California Institute of Technology.

CONTENTS

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I. INTRODUCTION

Isotopes are nuclei which have the same charge but different mass numbers. For example, 99.28% of the nuclei in naturally occurring uranium have mass number 238 (92 protons and 146 neutrons), 0.71% have mass number 235, and 0.0058% have mass number 234. Since isotopes have the same number of protons, their atoms exhibit identical chemical behavior. But, due to differences in mass and in nuclear structure, differing isotopes have different physical behavior; this results in some isotopes being more valuable than others.

A change in the isotopic concentration of a material without destroying the desired isotope is usually made by separation processes which depend upon the isotopic mass difference. This mass difference results in different accelerations from the same applied force or in different mean velocities at thermal equilibrium. The practical utilization of these physical differences is the subject of these notes. We shall discuss first six separation processes:

a. Separation nozzle
b. Sweep diffusion
c. Thermal diffusion
d. Electromagnetic separation
e. Gaseous diffusion
f. Centrifugation

Then we shall discuss how these processes are used in a cascade.

II. ISOTOPE SEPARATION PROCESSES

A. SEPARATION NOZZLE

Figure 1 illustrates a separation nozzle or separation jet.

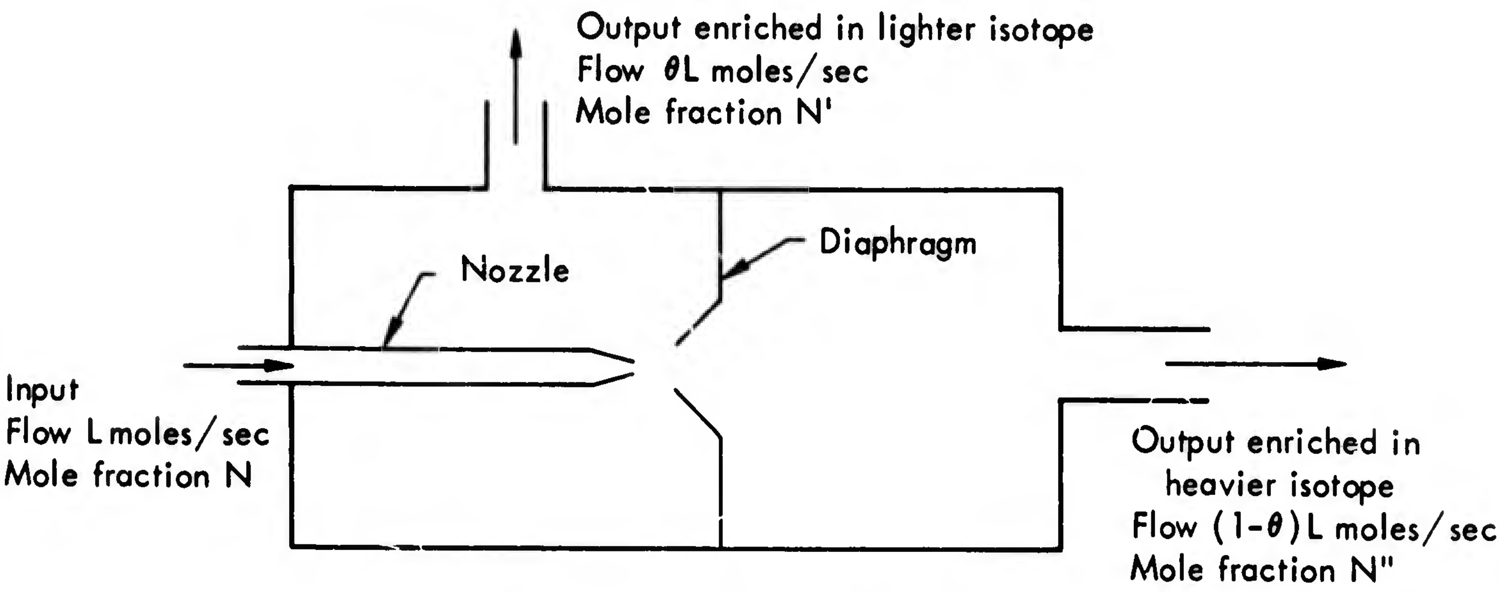

Fig.1 — Separation nozzle

A gaseous molecular mixture of isotopes--two, for example--is fed into the slit type nozzle under pressure (~ 10 mm Hg for UF_6). The mean free path of the molecules must be small compared with the nozzle width which is about 0.1 mm for UF_6. Under this condition the transverse pressure gradient at the mouth of the nozzle deflects the lighter isotope from the stream more easily than the heavier isotope. Consequently, the peripheral portion of the stream will be more highly enriched in the lighter isotope than the core portion. This peripheral portion is separated from the stream by a shaped diaphragm, and the peripheral output is thus enriched in the lighter isotope. The core output is enriched in the heavier isotope or, what amounts to the same thing, is depleted in the lighter isotope. The width of the diaphragm for a UF_6 nozzle is typically about 0.3 mm and is located about 0.1 mm away from the nozzle opening. Separation nozzles can be many meters in length.

The enrichment performed by an isotope separating device such as a separation nozzle is characterized by a <u>simple process factor</u> α. The factor α is the ratio of the abundance of the desired isotope in the output to its abundance in the input. Let us assume that the input to the nozzle in Fig. 1 is L moles/sec of material with a mole fraction N of the desired isotope which we will take to be the lighter isotope in a bicomponent input material. The peripheral output will be θL moles per second with a mole fraction N' of the light isotope. By mass conservation we see that the core output which has a mole fraction N" in the light isotope flows at $(1-\theta)L$ moles per second. The factor θ, which is between 0 and 1, is called the <u>cut</u>. Note that $N' > N > N''$. The abundance of the light isotope in the input stream is

$$R = \frac{N}{1 - N} \, ; \tag{1}$$

the abundance in the peripheral output stream is

$$R' = \frac{N'}{1 - N'} \, . \tag{2}$$

From (1) and (2) we find the simple process factor

$$\alpha = \frac{R'}{R} = \frac{N'/(1 - N')}{N/(1 - N)} \, . \tag{3}$$

For many processes α is only slightly larger than 1, and it is convenient to use the <u>simple process difference</u> ϵ defined by

$$\epsilon \equiv \alpha - 1. \tag{4}$$

Typical separation nozzles using uranium hexafluoride (UF_6) give $\epsilon \cong 0.004$ and $\theta \cong 0.2$.

A measure of how well a separative element performs is not ϵ alone because it does not reflect the rate at which the element can process material. Instead of ϵ, the measure of separative ability is the <u>separative power</u> or <u>separative capacity</u> of the element:

$$\delta U \equiv \frac{\theta}{1-\theta} \frac{G\epsilon^2}{2}, \tag{5}$$

where G is the input flow to the device in some appropriate units. The separative power of a device is an important quantity which we shall discuss and make much use of later. The quantities θ, G, and ϵ are usually related by the nature of the particular separation process under consideration. For the UF_6 separation nozzle one finds that $G \cong 2 - 4$ kg UF_6/hr/meter of nozzle length when $\epsilon \cong 0.004$ and $\theta \cong 0.2$. This gives a separative power for a nozzle one meter long of

$$\delta U = \frac{0.2}{1-0.2} \times 2\text{-}4 \times \frac{(.004)^2}{2}\ \text{kg/hr},$$

$$\cong 4\text{-}8 \times 10^{-6}\ \text{kg/hr} \cong 3\text{-}6 \times 10^{-2}\ \text{kg } UF_6\text{/yr}.$$

Another significant quantity for a separation device is called the <u>average process time per element</u>. It is approximately equal to the average time taken by the device to process under constant operating conditions an amount of material equal to that contained in the device. For example, if the separation nozzle contains Q kg of the UF_6 during constant operating conditions, then the average process time per element is approximately

$$h = Q/G. \tag{6}$$

B. SWEEP DIFFUSION

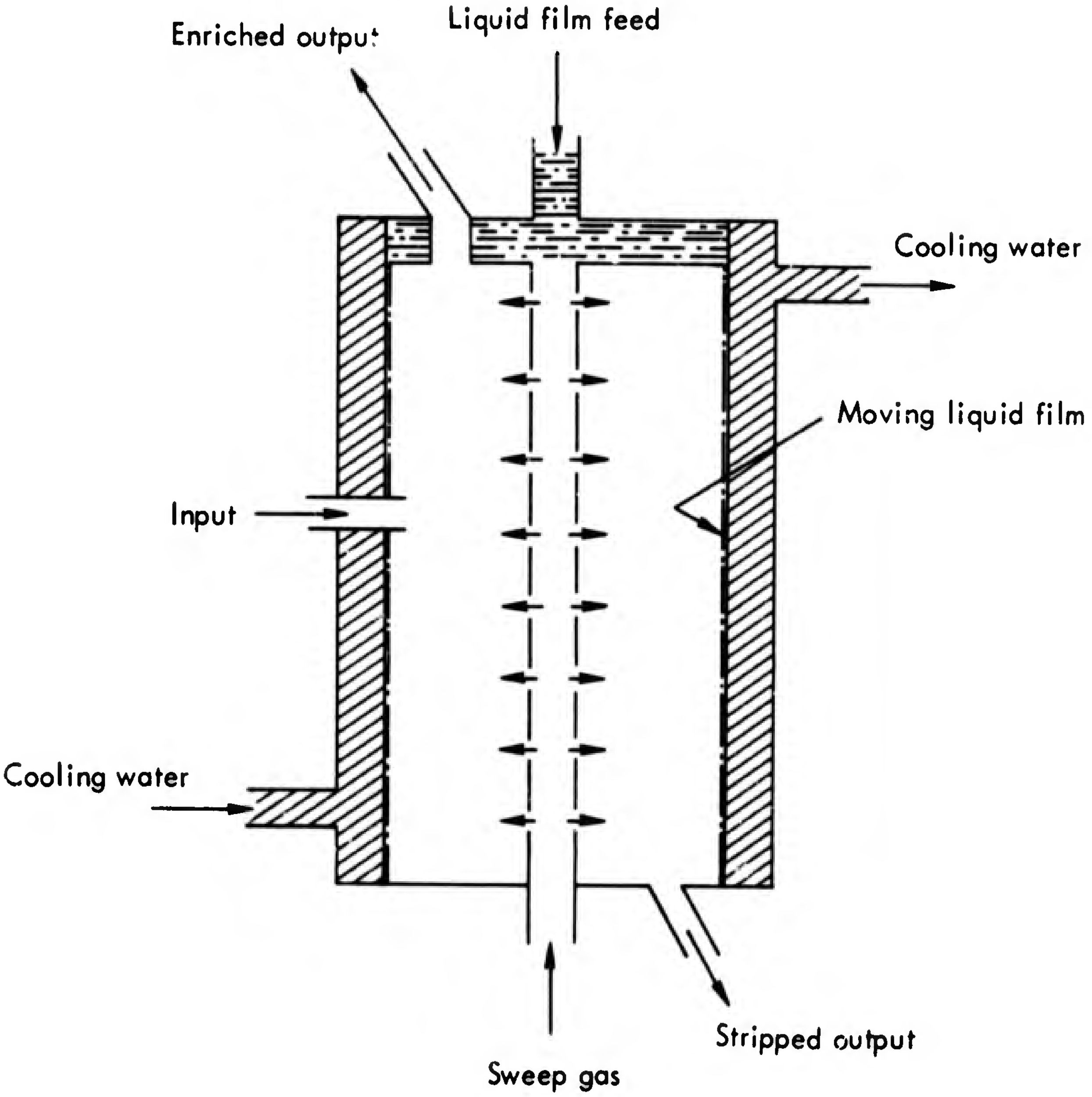

Fig.2 — A sweep diffusion column

The sweep diffusion process is based on the fact that different isotopes in gaseous form have different diffusion rates in another gas. Figure 2 illustrates a sweep diffusion column. The heavier isotopes are swept along with the sweep gas which is condensed on the cool walls of the column and allowed to flow down the sides and be removed. Sweep diffusion requires about the same power as gaseous diffusion for plants having the same inputs and outputs. One of the problems involved in perfecting sweep diffusion is the solubility of the input gas in the condensed sweep vapor. This results in a loss of separative capacity.

C. THERMAL DIFFUSION

Thermal diffusion was used in the Manhattan Project (S-50 plant) to provide slightly enriched UF_6 to the Y-12 electromagnetic separation facility. It is estimated that S-50 accelerated by one week (!) the flow of weapons-grade uranium from the Y-12 plant.(1) The S-50 plant operated only for slightly over a year since the thermal diffusion process was found to be very inefficient compared with gaseous diffusion.

The thermal diffusion of UF_6 occurs in liquid form. (UF_6 is a solid at room temperature and 1 atm. It sublimes at 56.6°C at 1 atm. To liquify the gas one must pressurize it at a temperature above 56.6°C.) A temperature gradient is established in the UF_6 liquid. The lighter $U^{235}F_6$ molecules concentrate in the warmer regions; the heavier $U^{238}F_6$ molecules concentrate in the cooler regions.

The explanation of thermal diffusion is not given by any elementary considerations. A transport theory analysis of interacting molecules is required to understand the effect. Such an analysis appears in The Physical Review paper by W. H. Furry, R. Clark Jones, and L. Onsager.(2) Most molecules interact with a force which goes as $r^{-\mu}$. If $\mu = 5$, there will be no separation by thermal diffusion for these molecules. For $\mu > 5$, the molecules of the lighter isotope concentrate in the warmer regions. If $\mu < 5$, the heavier molecules concentrate in the warmer regions.

A separative apparatus utilizing thermal diffusion consists of two concentric vertical pipes. UF_6 under pressure in liquid form is introduced into the annular space between the pipes. The inner pipe is heated, and the outer pipe is cooled. $U^{235}F_6$ molecules then tend to concentrate near the inner pipe where they are carried to the top by convection. Although the simple process difference in a thermal diffusion apparatus is ~ 0.05, the amount of material a single pipe can process is small. This leads to a very low separative capacity for each pipe. In addition, the time required for a thermal diffusion plant to come to equilibrium is long (~ 100-200 days).

D. ELECTROMAGNETIC SEPARATION

The separation schemes discussed above increase the enrichment in the desired isotope by only a very small amount, and a separation cascade must be used if significant enrichments are desired. In contrast, the electromagnetic separation process can produce relatively pure isotopes in a single stage. Usually, however, it is more efficient to use a small number of cascaded electromagnetic stages for reasons that will become apparent.

Electromagnetic separation is based on the momentum analyzing property of magnetic fields. An ion of mass m, speed v, and charge q is injected into a homogeneous magnetic field with its velocity vector perpendicular to the B field. This situation is illustrated in Fig. 3.

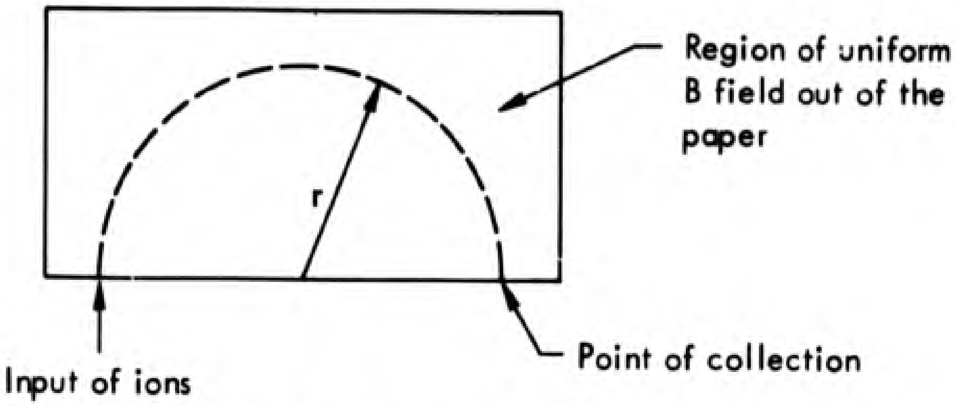

Fig.3 — Basic electromagnetic separator

In the magnetic field the force on the ion is qvB directed perpendicular to both the velocity vector and the magnetic field vector. This force results in a circular path for the ion. To find the radius r of the circle we set the centripetal acceleration equal to the acceleration resulting from the magnetic force:

$$\frac{v^2}{r} = \frac{qvB}{m} \qquad \text{or}$$

$$r = \frac{mv}{qB} \, . \tag{7}$$

If the energy of the incoming ion is expressed in electron volts, then (7) can be rewritten as

$$r = \frac{1}{B}\left(\frac{2mV}{q}\right)^{\frac{1}{2}} , \tag{8}$$

where V is the potential through which the ion has been accelerated from rest and q is the ion's charge. As an example, suppose that an ion of uranium, U^+ is accelerated through a potential of 25,000 volts into a uniform magnetic field of B = 500 gauss = ½ weber/m^2 as illustrated in Fig. 3. From (8) we see that

$$r = (2 \times 238 \times 1.69 \times 10^{-27} \text{ kg} \times 2.5 \times 10^4 \text{ volts} \Big/ 1.6 \times 10^{-19} \text{ coulombs})^{\frac{1}{2}}$$

$$\Big/ \tfrac{1}{2} \text{ weber/m}^2$$

$$= .7 \text{ meters or } 70 \text{ cm.}$$

To see how far apart singly ionized U^{235} and U^{238} atoms will be collected if both ions are accelerated by the same potential, we use a formula found by differentiating (8):

$$\frac{dr}{dm} = \frac{r}{2m} \quad \text{or} \quad \Delta r = \frac{r}{2}\frac{\Delta m}{m} \, . \tag{9}$$

In our example above this distance between collection points is

$$\Delta r = \tfrac{1}{2} \times 70 \text{ cm} \times \frac{\Delta m}{m} = \tfrac{1}{2} \times 70 \text{ cm} \times \frac{3}{238}$$

$$= .45 \text{ cm.}$$

This calculation gives some idea of the collection problem.

The ions entering the magnetic field region must be carefully collimated and quite monoenergetic to avoid defocussing at the collector regions. These conditions place stringent requirements on both the ion source and the accelerating voltage. The ion source must produce monoenergetic ions, and the accelerating voltage must be stable. In addition, the ion source must produce a high density of ions to get a reasonable amount of material separated. (This last requirement conflicts to some extent with having a stable ion source.) But the rate of ion production cannot be too large because space charge effects will smear the beam and result in defocussing. These conflicting requirements usually result in some defocussing being tolerated. Then several stages must be used in the electromagnetic separation of commercial quantities of isotopically pure material.

How well does electromagnetic separation compete with other separation processes in producing fairly large quantities (10-20 kg/yr) of U^{235}? Not so well. In the Manhattan Project the operation of the Y-12 electromagnetic separation facility at Oak Ridge required 20,000 workers by the spring of 1945. During May and June of 1945 S-50 fed K-25, the gaseous diffusion plant, which in turn supplied 7% enriched material to the electromagnetic separation facility for enrichment to weapons-grade uranium. But as K-25 began to meet its design goals and as another gaseous diffusion plant, K-27, started to operate, Y-12 was shut down except for one unit, a Beta track, in December of 1946. Finally this unit ceased operation. The electromagnetic separation process was not as economical as the gaseous diffusion process for producing large quantities of highly enriched U^{235}. Recent cost estimates for electromagnetic separation are about $15,000 per separated gram atom.(3)

Electromagnetic separation is unique in its ability to separate diverse elements. Its technological complexity and associated high costs, however, appear to eliminate it as a means of producing commercial quantities of U^{235}.

E. GASEOUS DIFFUSION

The gaseous diffusion process of isotope separation is based on a phenomenon known as molecular effusion which was first demonstrated by Graham in 1846. Molecular effusion uses the fact that a lighter molecule in a mixture of gas at thermal equilibrium will strike the walls of the confining container more frequently than a heavier molecule. If the walls are perforated by very small holes, the escaping gas will be enriched in the lighter components. Kinetic theory readily explains this effect: The average speed of a gas molecule at temperature T is

$$\bar{v} = (8kT/\pi m)^{\frac{1}{2}}, \qquad (10)$$

where k is the Boltzmann constant and m is the mass of the molecule. Formula (10) shows that a light molecule of mass m_L has an average speed greater than that of a heavier molecule of mass $m_H > m_L$. Greater speed means, of course, more collisions with the walls. If this phenomenon is to be used to separate gaseous isotopes by a porous membrane, the holes in the membrane must be small enough to prevent bulk flow. This implies that the radius of the holes must be much smaller than the mean free path of the gas. Under these circumstances Knudsen's Law gives the flow in moles/unit time/unit area:

$$\bar{n} = \frac{8a}{3\ell} (1/2\pi RTM)^{\frac{1}{2}} (p_F - p_B), \qquad (11)$$

where a = radius of capillary (< < mean free path of gas molecules)
ℓ = length of capillary
p_F = pressure on the input side
p_B = pressure on the output side
M = molecular weight of gas molecules.

Formula (11) follows from straightforward kinetic theory arguments with the assumption that the scattering from the capillary walls is diffuse (Lambert's Law).

Now we can derive the simple process separation factor α for a membrane which obeys Knudsen's Law. Assume that the gas is composed of a light fraction of molar concentration N_o and molecular weight M_L and a heavy fraction of molar concentration $1-N_o$ and molecular weight M_H. Take the back pressure to be negligible. Figure 4 illustrates the situation under consideration.

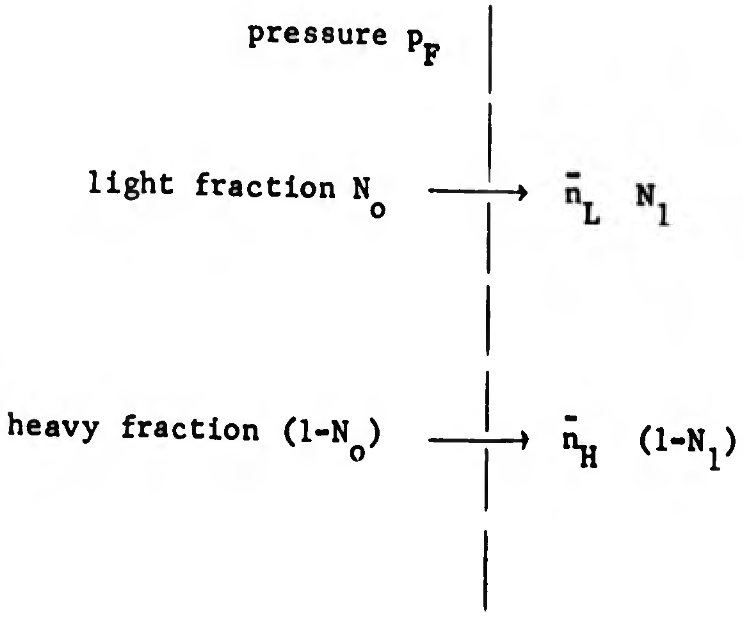

Fig. 4--Separation Membrane

From Knudsen's Law we get

$$\bar{n}_L = \frac{8a}{3\ell} (1/2\pi \, RTM_L)^{\frac{1}{2}} \, p_F^L$$

and

$$\bar{n}_H = \frac{8a}{3\ell} (1/2\pi \, RTM_H)^{\frac{1}{2}} \, p_F^H \, ,$$

where p_F^L and p_F^H are the partial pressures of the light and heavy molecules, respectively. Since $p_F^L \propto N_o$ and $p_F^H \propto 1-N_o$, we obtain

$$\frac{\bar{n}_L}{\bar{n}_H} = \frac{N_o}{1-N_o} \left({}^{M_H}\!/\!_{M_L} \right)^{\frac{1}{2}} .$$

Before significant back pressure builds up, N_1 is proportional to $\bar{n}_L$ and $1-N_1$ to $\bar{n}_H$. This means that

$$\frac{N_1}{1-N_1} = \frac{\bar{n}_L}{\bar{n}_H} = \frac{N_o}{1-N_o}\left({}^{M_H}/{}_{M_L}\right)^{\frac{1}{2}} .$$

Recognizing ${}^{N_1}/{}_{1-N_1}$ as the abundance of the light molecule in the output stream and ${}^{N_o}/{}_{1-N_o}$ as the abundance of the light molecule in the input stream, we obtain from the above equation the simple process factor for the membrane:

$$\alpha = \left({}^{M_H}/{}_{M_L}\right)^{\frac{1}{2}} . \tag{12}$$

For uranium hexafluoride (UF_6), Eq. (12) gives

$$\alpha = \left(\frac{238 + 6 \times 19}{235 + 6 \times 19}\right)^{\frac{1}{2}} = 1.00429 .$$

One can get some idea of the radii of the membrane holes by remembering that they must be much smaller than the mean free path of UF_6 which is 0.02 microns at 20°C and 1 atm pressure. If $M_H \cong M_L$, the simple process difference can be written as

$$\epsilon \equiv \alpha - 1 \cong \frac{M_H - M_L}{2M_H} . \tag{13}$$

In an actual separative element the simple process difference can be somewhat lower than 0.00429 due to a number of physical factors. Since the separative power of an element is $\delta U = \frac{1}{2}\frac{\theta}{1-\theta} G\epsilon^2$, the smallness of ϵ can be somewhat offset by a large G which can be achieved with a large barrier area.

A measure of the energy required by a separative process to enrich material is called the specific electric input, ζ:

$$\zeta = \frac{\text{power consumption of separative element}}{\text{separative capacity of the element.}} \tag{14}$$

For producing enriched uranium by the gaseous diffusion method the specific electric input is estimated to be 9000 Kw/KgU/hr.(4) How this quantity is used to calculate the amount of energy required to produce a given amount of U^{235} having a certain enrichment will be demonstrated in the section on cascade theory.

F. CENTRIFUGATION

Separation of gases of different molecular weights is an old idea. Bredig in 1895 conducted some separation experiments involving centrifuges. Lindemann and Aston in 1919 suggested that isotopes could be enriched by gaseous centrifugation.

Gaseous centrifugation is based on the fact that at equilibrium the centrifugal force on the rotating gas is balanced by an opposing pressure gradient. The pressure gradient results in a density gradient whose magnitude depends on the molecular weight of the gas.

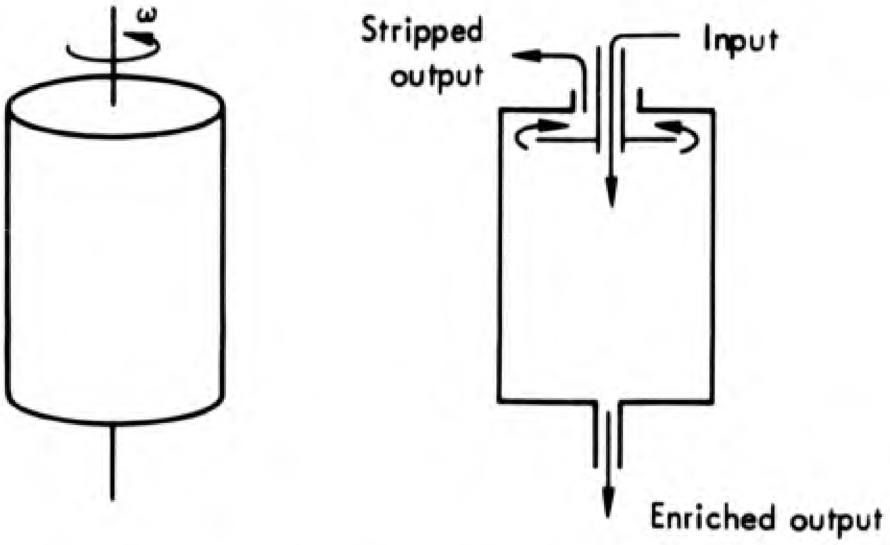

Fig.5 — Simple gaseous centrifuge

To derive the equilibrium distribution of gas in the cylindrical centrifuge shown in Fig. 5 we consider the forces on a rotating mass of gas of height $\Delta \ell$ between the radii r and $r + \Delta r$. The centrifugal force acting on this mass of gas is $[2\pi r \ \Delta r \ \Delta \ell \ \rho(r)] \ \omega^2 \ r$, where ρ is the density of the gas and ω is the angular velocity of the centrifuge

At equilibrium the gas pressure p is a function of r alone and must have a gradient leading to a force equal and opposite to the centrifugal force. This condition is expressed by the equation

$$2\pi r\ \Delta\ell\ [p(r+\Delta r)-p(r)] = [2\pi r\ \Delta r\ \Delta\ell\ \rho(r)]\ \omega^2 r. \qquad (15)$$

If the gas is ideal, this equation leads to a molecular-weight dependent pressure gradient in the gas because of the ideal gas equation $p = \frac{\rho}{M} RT$:

$$\frac{1}{p(r)}\frac{dp(r)}{dr} = \frac{M\omega^2}{RT} r,$$

which is readily integrable to

$$p(r) = p(o)\ e^{\frac{M\omega^2 r^2}{2RT}} . \qquad (16)$$

(We have assumed that there are viscous forces which cause the gas within the centrifuge to rotate.) Taking p_1 to be the partial pressure of the lighter molecules and p_2 to be the partial pressure of the heavier gas molecules, we obtain from (16) that

$$\frac{p_1(r)}{p_2(r)} = \frac{p_1(o)}{p_2(o)}\ e^{-\frac{(M_2-M_1)\omega^2 r^2}{2RT}} ,$$

where M_1 and M_2 are the molecular weights of the lighter and heavier gas molecules, respectively. The mole fraction of the lighter isotope is $N = p_1/(p_1 + p_2)$; that of the heavier isotope is $1-N = p_2/(p_1 + p_2)$. This allows us to write the above equation as

$$\left(\frac{N}{1-N}\right)_r = \left(\frac{N}{1-N}\right)_o e^{-\frac{(M_2-M_1)(\omega r)^2}{2RT}} .$$

If we take as r the radius of the centrifuge, then the equilibrium separation factor for the simple process is

$$\alpha_o = \frac{(N/1-N)_o}{(N/1-N)_r} = e^{\frac{(M_2-M_1)(v_p)^2}{2RT}} , \tag{17}$$

where v_p is the peripheral velocity of the centrifuge. For UF_6 at $v_p = 400$ m/sec, $T = 300^oK$ ($R = 8.3 \times 10^7$ ergs/^oK), we get

$$\frac{(M_2-M_1)\ v_p^2}{2RT} \cong 0.1$$

or

$$\alpha_o = e^{0.1} = 1.1; \quad \text{then}$$

$$\epsilon_o = \alpha_o - 1 = 0.1.$$

For small enough mass differences ($\epsilon_o << 1$), one has

$$\epsilon_o \cong \frac{(M_2-M_1)\ v_p^2}{2RT} . \tag{18}$$

Since ϵ_o varies as v_p^2, the centrifuge should be rotated as rapidly as possible. The tensile strength T of the walls of the centrifuge bowl places an upper limit on v_p:

$$v_p < \sqrt{\frac{T}{\rho_{bowl}}} , \tag{19}$$

where ρ_{bowl} is the density of the bowl material.

The parameter that gives the effectiveness of the centrifuge in isotope separation is the separative capacity

$$\delta U = \frac{1}{2} G\epsilon^2 ,$$

where G is the flow into the centrifuge in some units and the cut θ has been taken to be $\frac{1}{2}$. We can estimate G for the centrifuge by a crude argument. In the centrifuge any flow will tend to disturb the equilibrium concentration. Pressure diffusion will try to restore equilibrium. The approximate time required to establish equilibrium throughout the gas in the centrifuge is $\sim$ radius^2/D, where D is the diffusion coefficient of the gas. In this time an amount of input gas equal approximately to the volume of the bowl will flow into the centrifuge. It then follows that

$$G \sim \frac{\pi(\text{radius})^2 L\rho}{(\text{radius})^2/D} = \pi L\rho D ,$$

where ρ is the density of the input gas, and L is the height of the centrifuge bowl. This result gives

$$\delta U_{\max}^{\text{theor}} = \frac{\pi\rho D}{2} \cdot \left[\frac{(M_2-M_1)v_p^2}{2RT}\right]^2 \cdot L . \tag{20}$$

The qualitative derivation of (20) given here is a basis only for an order of magnitude value for $\delta U_{\max}^{\text{theor}}$.

For UF_6, ρD is equal to 2.35×10^{-4} poises. The ratio of the actual separative capacity to $\delta U_{\max}^{\text{theor}}$ is called the separation efficiency.

The Groth centrifuge UZ111B, which is 63.5 cm long and 6.7 cm in radius, has a separative capacity of about 0.8 kg U/yr with a peripheral speed of 280 m/sec. The flow G of UF_6 is about 1 gm/min. These are 1958 figures. UZ111B consumes energy at the rate of 1.5 Kw, which implies a specific electric input of

$$\zeta = \frac{1.5 \text{ k w}}{0.8\text{Kg U/yr}} \tilde{=} 16{,}000 \text{ Kw/Kg U/hr.}$$

The separation efficiency of UZ111B is about 75%.

III. CASCADE THEORY

All of the separation devices we discussed above except for electromagnetic separation give individually a very small change in enrichment. If large enrichments are desired, many separation devices must be connected together in a cascade. To understand how cascades work, we need to learn some "cascade theory."

A "cascade" is an arrangement of connected separation devices which are referred to as "elements" of the cascade. Elements operating in parallel on material of the same mole fraction form a "stage." Stages connected in series (one feeding another) make up a cascade. In Fig. 6 the flow designations for a single element or a single stage are shown.

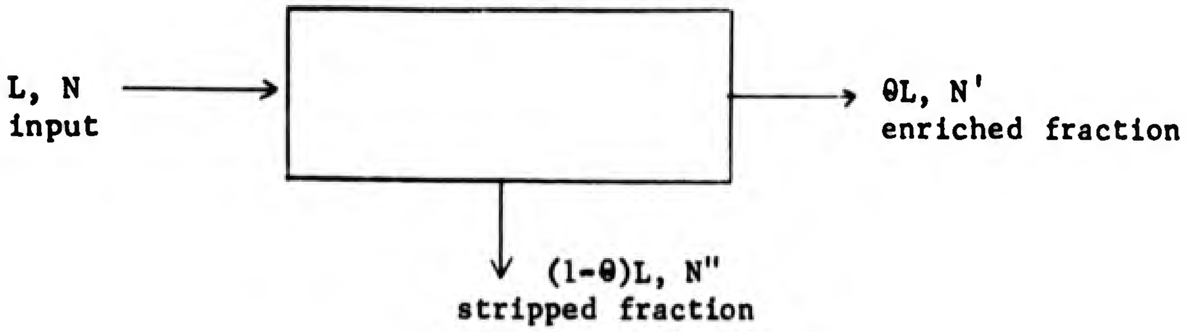

Fig. 6--Flow Designations for an Element or a Stage

L moles per second of material of mole fraction or enrichment N in the desired isotope enter the element or stage. This material undergoes some separation. The enriched output or enriched fraction flows at θL moles per second and has an enrichment N'; the stripped fraction leaves at (1-θ)L moles per second and enrichment N". The process factor α of this element was defined above as the ratio of the enriched abundance to the input abundance:

$$R' = \frac{N'}{1-N'} = \alpha R = \alpha \frac{N}{1-N}. \tag{21}$$

Now let us consider a cascade formed of stages which contain elements operating in parallel. Figure 7 is a diagram of such a cascade along with the designated flows and mole fractions. This cascade has both an enriching section and a stripping section. The need for the latter section in an economically operated cascade will be apparent shortly. Our problem is to choose the cuts θ_s and flows L_s to get the most efficient cascade. What do we mean by "the most efficient cascade"? We mean the cascade with the least number of elements with a given feed enrichment (N_o) for a desired output (P moles/sec and N_p enrichment). One would expect that this cascade is obtained when materials of different mole fractions are not mixed. (A detailed proof of this result can be found in Ref. 5.) The "no-mixing" requirement means that

$$N'_{s-1} = N_s = N''_{s+1} \tag{22}$$

or

$$R'_{s-1} = R_s = R''_{s+1}. \tag{23}$$

In particular, one has

$$N'_{-1} = N_o = N''_1. \tag{24}$$

A cascade obeying the no-mixing conditions (22)-(24) for the configuration of Fig. 7 is called an ideal cascade. It has the fewest number of elements or separation units for a given feed enrichment and output flow and enrichment. From (21) and (23) we see that

$$R_s = \alpha R_{s-1} = \alpha^s R_o. \tag{25}$$

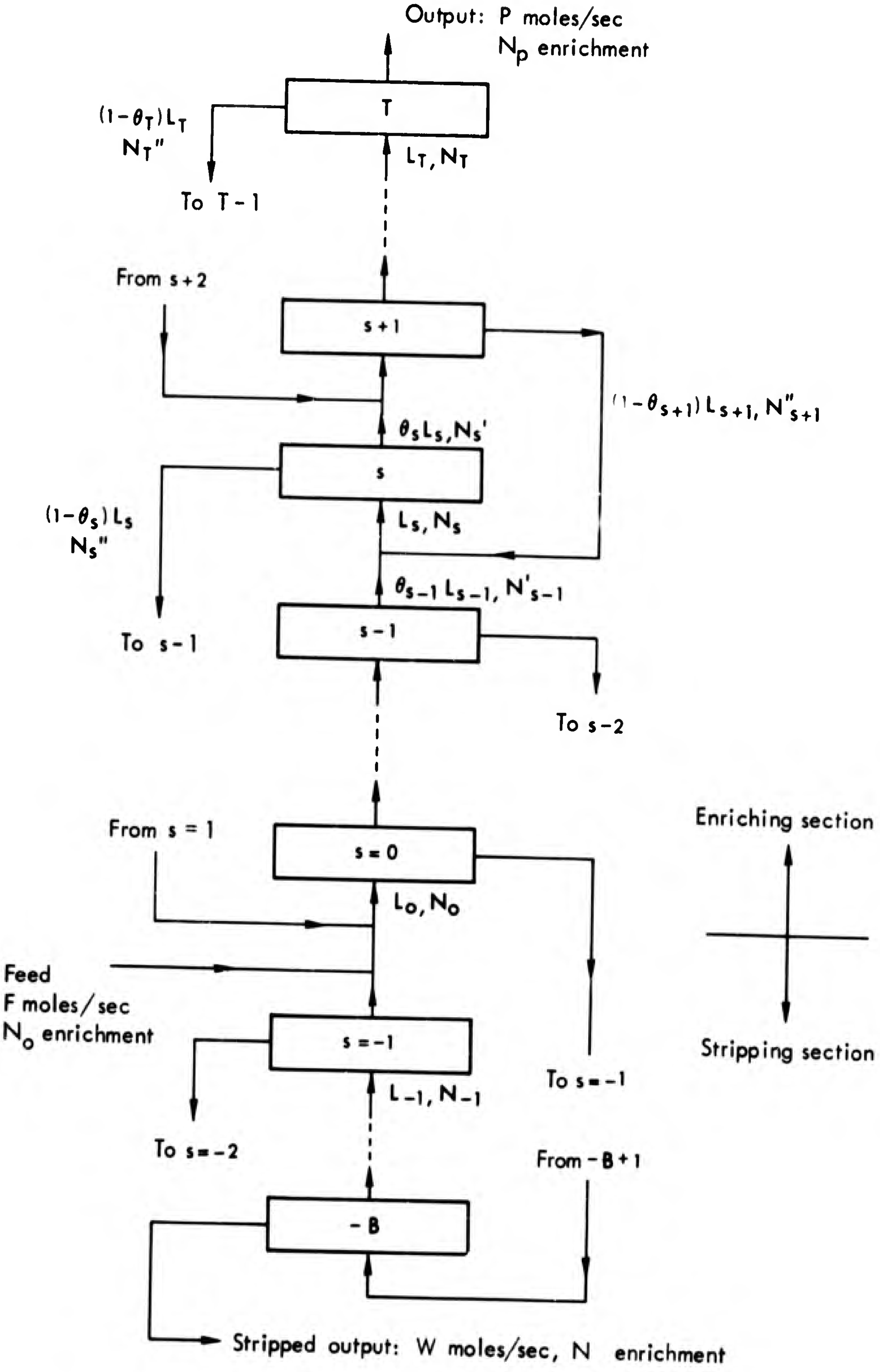

Fig.7 — A cascade

Usually α is only slightly larger than unity, and it is convenient to introduce the process difference

$$\epsilon = \alpha - 1. \tag{26}$$

For all the separation processes we discussed above except for electromagnetic separation $\epsilon << 1$. We shall assume this to be the case for the cascade under study. Since $\epsilon << 1$, we obtain from (21), (25), and (22) the relations

$$N_{s+1} = N'_s = N_s + \epsilon N_s(1-N_s) + 0(\epsilon^2) \tag{27}$$

$$N_{s-1} = N''_s = N_s - \epsilon N_s(1-N_s) + 0(\epsilon^2). \tag{28}$$

Taking N to be a function of a continuous variable s, we can write (27) and (28) as

$$\frac{dN}{ds} = \epsilon N(1-N). \tag{29}$$

This equation can be readily integrated to give the number of stages in the enriching and stripping sections:

$$\# \text{ enriching stages} = \frac{1}{\epsilon} \ln \frac{R_p}{R_o} \tag{30}$$

$$\# \text{ stripping stages} = \frac{1}{\epsilon} \ln \frac{R_o}{R_w} . \tag{31}$$

We could, of course, have derived these two equations from (25), but (29) will prove to be essential for other calculations.

Now we must do some algebra to get expressions for θ_s and L_s. Conservation of the desired isotope at the s-th stage ($L_s N_s = (1-\theta_s)L_s N''_s + \theta_s L_s N'_s$) gives

$$\theta_s = \frac{N_s - N''_s}{N'_s - N''_s} ,$$

which by use of (23), (25), and (26) is seen to be

$$\theta_s = \frac{1}{2} + \frac{\epsilon}{2} (N_s - \frac{1}{2}) + 0(\epsilon^2). \qquad (32)$$

For an ideal cascade of the structure shown in Fig. 7, the cuts are all equal to 1/2 to a very good approximation. If Fig. 7 were different, e.g., the stripped fraction from the s-th stage going to the input of the s-2-th stage, the cut would not be 1/2 (it would be 2/3). To get an expression for L_s, let us consider the conservation of flow and of the desired isotope for all the stages above the s-th:

$$\theta_s L_s = P + (1-\theta_{s+1}) L_{s+1}$$

$$\theta_s L_s N'_s = PN_p + (1 - \theta_{s+1}) L_{s+1} N''_{s+1}.$$

Using (22), (27), and (32), we can solve for L_s from the above two equations:

$$L_s = \frac{2}{\epsilon} \frac{P(N_p - N_s)}{N_s(1-N_s)} . \qquad s \geq 0 \qquad (33)$$

A similar analysis for the stripping stages gives

$$L_s = \frac{2}{\epsilon} \frac{W(N_s - N_w)}{N_s(1-N_s)} . \qquad s < 0 \qquad (34)$$

Since $N_p \approx 1$ and $N_w \approx 0$ in typical operation, (33) and (34) show that the flow through the s-th stage L_s decreases the further the stage is from the feed point. Overall mass and flow conservation for the cascade gives

$$F = \frac{P(N_p - N_w)}{N_o - N_w} \tag{35}$$

$$W = \frac{P(N_p - N_o)}{N_o - N_w} \quad . \tag{36}$$

Since $N_o - N_w = \epsilon N_o(1 - N_o)$ without a stripping section, (35) shows how important a stripping section is in reducing the amount of feed.

Equations (33) - (36) can be used to design an ideal cascade once we have decided on P, N_p, and N_w. (N_o is usually fixed by either nature or by the separation plant feeding the one we are considering.) Since each separation element can handle G moles/sec of material, the number of elements in the s-th enriching stage is

$$n_s = {}^{L_s}/G = \frac{2}{\epsilon G} \, \frac{P(N_p - N_s)}{N_s(1-N_s)} \, , \qquad s \geqslant 0 \tag{37}$$

and the number of elements in the s-th stripping stage is

$$n_s = \frac{2}{\epsilon G} \, \frac{W(N_s - N_w)}{N_s(1-N_s)} \, , \qquad s < 0 \tag{38}$$

From (37) and (38) we can calculate the total number of elements in the cascade:

total # elements

$$= \sum_{s=0}^{\frac{1}{\epsilon}\ln\frac{R_p}{R_o}} \frac{2}{\epsilon G} \, \frac{P(N_p - N_s)}{N_s(1-N_s)} + \sum_{s=-1}^{-\frac{1}{\epsilon}\ln\frac{R_o}{R_w}} \frac{2}{\epsilon G} \, \frac{W(N_s - N_w)}{N_s(1-N_s)} \, .$$

Taking N to be a function of a continuous s and using (29), we can convert this sum into an integral that can be evaluated:

total # elements in cascade

$$= \int_{N_o}^{N_p} \frac{2}{\epsilon G} \frac{P(N_p-N)}{N(1-N)} \frac{1}{\epsilon N(1-N)} dN$$

$$+ \int_{N_w}^{N_o} \frac{2}{\epsilon G} \frac{W(N-N_w)}{N(1-N)} \frac{1}{\epsilon N(1-N)} dN$$

$$= \frac{2}{\epsilon^2 G} P\left[(2N_p-1)\ln\frac{R_p}{R_o} + \frac{1-2N_o}{1-N_o}\,\frac{N_p-N_o}{N_o}\right]$$

$$+ \frac{2}{\epsilon^2 G} W\left[(2N_w-1)\ln\frac{R_w}{R_o} + \frac{1-2N_o}{1-N_o}\,\frac{N_w-N_o}{N_o}\right] . \qquad (39)$$

The first expression on the right-hand side of (39) is the total number of elements in the enriching section, and the second expression is the total number of elements in the stripping section. The term $\frac{1}{2}\epsilon^2 G = \delta U$, which we discussed above and called the separative capacity, depends on the type of separation element used and clearly measures the separative ability of an element when it is used in an ideal cascade.. The other two factors in (39) have nothing to do with the physical properties of an element; they depend only on flows and enrichments. The factors in square brackets are called "value functions":

$$V(N) = \left[(2N-1)\ln\frac{R_N}{R_o} + \frac{1-2N_o}{1-N_o}\,\frac{N-N_o}{N_o}\right] . \qquad (40)$$

V(N) has a minimum at $N = N_o$, where $V(N_o) = 0$. Tables of V(N) for uranium ($N_o = 0.007143$) can be found in many books. Some values of V(N) are listed below in Table 1.

Table 1

V(N) with $N_o = 0.007143$

N	V(N)	N	V(N)
.001	1.115	.8	113.99
.002	.558	.85	121.82
.003	.291	.9	129.81
.004	.142	.91	131.43
.005	.0574	.92	133.08
.006	.0145	.93	134.74
.007143	0	.94	136.42
.01	.0645	.95	138.14
.02	.786	.96	139.90
.03	1.806	.97	141.74
.04	2.951	.98	143.69
.05	4.166	.99	145.95
.1	10.717		
.2	24.677		
.3	39.070		
.4	53.698		
.5	68.504		
.6	83.471		
.7	98.615		
.75	106.27		

Equation (39) is often written in the form

$$\text{total \# elements in cascade} = \frac{\Delta U}{\delta U}\,, \qquad (41)$$

where

$$\Delta U = U_p + U_w \qquad (42)$$

and

$$U_p = PV(N_p) \qquad (43)$$

$$U_w = WV(N_w). \qquad (44)$$

ΔU is called the "change in value" produced by the cascade in unit time. δU is then the "change in value" produced by a single element in unit time. Equation (41) then gives the number of elements required to change the "value" of the feed material by an amount ΔU in unit time. Since ΔU doesn't depend upon what isotope separation scheme is used, it is clearly unrelated to the cost of enriching material or to the market value of the enriched material. But somebody back in the days of the Manhattan Project decided to call V(N) the "value function," and we are stuck with this name today.

How do we design a cascade with the above formulas? Suppose that somebody comes to us and says, for example: "Design me a cascade that will enrich uranium from natural enrichment to 90% U^{235} and will produce 1 kg/day." So we take P = 1kg/day and $N_p = .9$. From Table 1 we find that V(0.9) = 129.81. Equation (35) indicates that the amount of feed is a function of the degree of stripping. Assume that the economics of the situation dictates $N_w = 0.005$; V(.005) = .0574. Then (35) and (36) give

$$F = \frac{1\ (.90-.005)}{.00714-.005}\ {}^{kg}/day = 418\ {}^{kg}/day$$

$$W = 417\ {}^{kg}/day.$$

If we are using separation elements such as the UZ111B centrifuge with a separative capacity of 1 kg/yr, then the number of centrifuges in the enriching section will be

$$\frac{PV(N_p)}{\delta U} = \frac{1\ {}^{kg}/day \times 129.8}{1\ kg/yr} = 129.8 \times 365 = 47{,}400 \text{ centrifuges}$$

and the number of centrifuges in the stripping section will be

$$\frac{WV(N_w)}{\delta U} = \frac{417\ {}^{kg}/day \times .0574}{1\ kg/yr} = 8{,}700 \text{ centrifuges.}$$

From (30) and (31) or (25) we can find the number of enriching stages and stripping stages ($\epsilon = .06$):

$$\#\ \text{enriching stages} = \frac{\ln \frac{9.00}{.0072}}{\ln 1.06} = 122$$

$$\#\ \text{stripping stages} = \frac{\ln \frac{.0072}{.00503}}{\ln 1.06} = 5.18$$

The power required by the enriching and stripping portions of the ideal centrifuge cascade to produce 1 ^{kg}/day of 90% enriched uranium with a stripped output of .5% material is

$$\text{power required} = \zeta\ [PV(N_p) + WV(N_w)]\Big|_{\substack{P = 1\ kg/day,\ N_p = .9 \\ W = 417\ kg/day,\ N_w = .005,}}$$

where ζ is the specific electric input discussed in the previous section. For the centrifuges in our discussion we will take ζ = 547 Kw/KgU/day, which corresponds to 1.5 Kw/centrifuge with a separative capacity of 1 Kg U/yr. We then obtain

$$\text{power required} = 547\ [129.8 + 417 \times .0574]$$

$$= 84{,}000\ \text{Kw.}$$

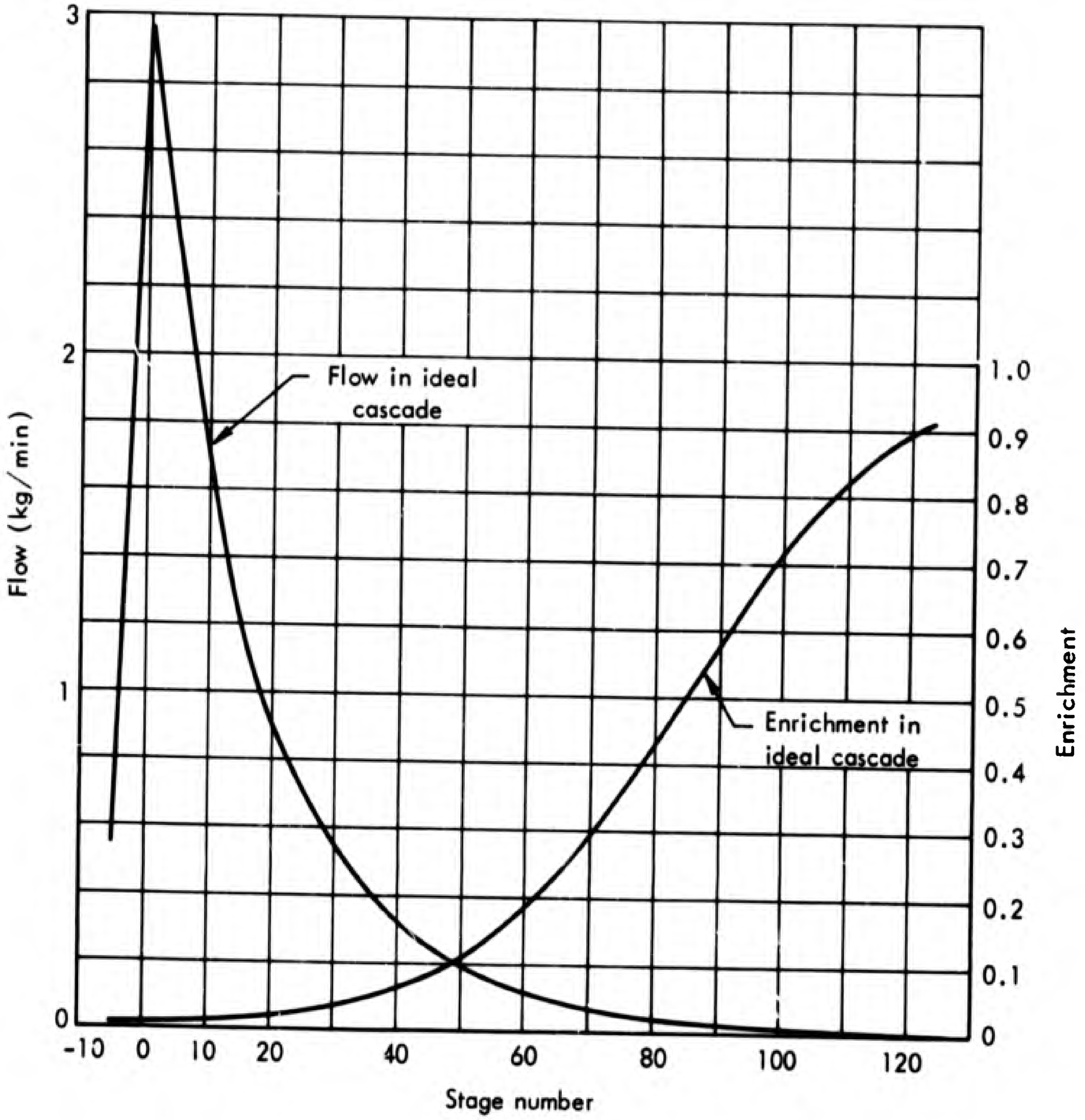

Fig.8 — Flow and enrichment in the centrifuge cascade

Table 2 shows how the ideal flow and the enrichment varies for the above centrifuge cascade. Figure 8 exhibits the information in Table 2 in graphical form. Note that relatively few elements are required at the top of the cascade.

In discussions of isotope separation diagrams similar to that in Fig. 9 often appear. These diagrams are formed by rotating a cascade flow graph such as Fig. 8 and its mirror image through 90°. The width of the diagram is supposed to give the flow and the height the stage number. However, since no scale is shown with these diagrams, they are only useful to remind the reader how actual flow diagrams look.

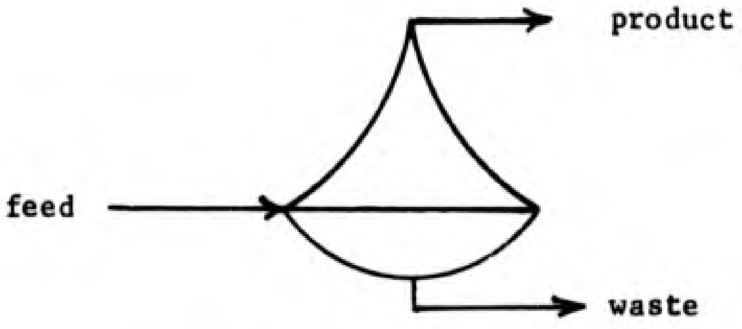

Fig. 9--Typical "isotope separation" figure

Ideal cascades are not employed in practice because their requirement that each stage contain a different number of elements would lead to manufacturing difficulties. Each stage would need different piping, control systems, and, in the case of gaseous diffusion, compressors. What is done in the planning stage is to divide up an ideal cascade into sub-cascades which have the same flow through each stage. Every stage of the same sub-cascade would then be identical and could be manufactured by mass production methods. This process of dividing up an ideal cascade into sub-cascades having constant flow is called "squaring off" an ideal cascade.

Table 2

IDEAL FLOW AND ENRICHMENT IN THE CENTRIFUGE CASCADE

Stage Number	Enrichment	Flow (kg/min)	Stage Number	Enrichment	Flow (kg/min)
- 6	.00504	--	40	.0690	0.301
- 5	.00534	0.548	50	.1097	0.188
- 4	.00566	1.066	60	.192	0.106
- 3	.00600	1.554	70	.298	0.0668
- 2	.00636	2.015	80	.425	0.0453
- 1	.00674	2.451	90	.576	0.0309
0	.00714	2.933	100	.709	0.0218
1	.00756	2.781	110	.815	0.0136
2	.00803	2.60	120	.8868	0.00397
10	.0128	1.64	121	.8925	0.00271
20	.0226	0.921	122	.8980	0.00139
30	.0397	0.525	123	.9032	--

Using Fig. 7 and the mass and flow conservation conditions, we can derive an equation relating the parameters which describe a square sub-cascade. Mass and flow conservation across an imaginary line between the s-th and s-1st stage gives

$$\theta_s L_s - (1-\theta_{s+1}) L_{s+1} = P \tag{45}$$

$$\theta_s L_s N'_s - (1-\theta_{s+1}) L_{s+1} N''_{s+1} = PN_p. \tag{46}$$

[In our previous discussion these two equations plus the no-mixing condition (22) were used to derive the flow (33). For a square cascade there is mixing, and the no-mixing condition doesn't apply.] From (27) and the mass conservation condition for the s-th stage $[N_s = (1-\theta_s)N''_s + \theta_s N'_s]$, we obtain

$$N''_{s+1} = N'_{s+1} - \frac{\epsilon}{1-\theta_{s+1}} N'_{s+1}(1-N'_{s+1}) + O(\epsilon^2). \tag{47}$$

Combining (45) and (46) yields

$$N''_{s+1} = N'_s - \frac{P(N_p-N'_s)}{(1-\theta_{s+1})L_{s+1}} ; \tag{48}$$

substituting this into (47) and rearranging terms results in

$$N'_{s+1} - N'_s = \frac{\epsilon}{1-\theta_{s+1}} N'_{s+1}(1-N'_{s+1}) - \frac{P(N_p-N'_s)}{(1-\theta_{s+1})} L_{s+1}. \tag{49}$$

In a square sub-cascade the flow L is constant and $^P/L \ll 1$. From (45) we see that this implies that $\theta_s = \frac{1}{2} + O(^P/L)$. Substituting these results into (49) and taking N' as a continuous function of s, we obtain the equilibrium differential equation describing the square sub-cascade:

$$\frac{dN'}{ds} = 2\epsilon N'(1-N') - \frac{2P}{L}(N_p-N'). \tag{50}$$

If the square sub-cascade has a length (number of stages) Z and takes material from N_{in} to N_{out}, (50) integrates to

$$Z = \frac{1}{\epsilon\Delta(\psi)} \tanh^{-1}\left[\frac{(N_{out} - N_{in})\Delta(\psi)}{(N_{out} + N_{in})(1+\psi) - 2N_{out}N_{in} - 2N_p\psi}\right], \tag{51}$$

where

$$\psi \equiv {}^P/\epsilon L$$

$$\Delta(\psi) \equiv [1 + 2\psi(1-2N_p) + \psi^2]^{\frac{1}{2}}.$$

For a given P, N_p, N_{in}, and N_{out}, (51) gives Z as a function of ψ. From (50) one can see that the smallest Z will occur when $L \to \infty$ because this gives the largest $\frac{dN'}{ds}$. A simple integration shows that $Z_{min} = \frac{1}{2\epsilon} \ln \frac{R_{out}}{R_{in}}$. The largest Z is clearly ∞ and occurs for $L = \frac{P}{\epsilon} \frac{(N_p - N_{in})}{N_{in}(1-N_{in})}$ since this value of the flow will start the square sub-cascade with $\frac{dN'}{ds} = 0$. Comparing this flow with (33), we see that the flow in a square sub-cascade must exceed 1/2 of the initial flow for the section of ideal cascade being replaced.

In the above discussion we saw that many square sub-cascades having different (Z,L) for given P, N_p, N_{in}, and N_{out} could be used. The most economical sub-cascade will usually be the one with the fewest elements. This sub-cascade can be found from (51) by minimizing the product ZL, or equivalently, the ratio Z/ψ. Since (51) involves an arctanh function, minimizing Z/ψ analytically would be a laborious task. However, an accurate estimate of the length Z which gives the minimum number of elements can be found by estimating the flow L on the basis of the flow in the section of the ideal cascade being replaced. This is possible because of an interesting theorem. The flow in an ideal cascade minimizes integrals of the form [the proof is straightforward and uses (50) with L as a function of s]

$$I = \int_{\text{bottom stage}}^{\text{top stage}} \phi[N(s)]L(s)ds,$$

where $\phi(N) \geq 0$. The total number of elements in a cascade or sub-cascade has this form with $\phi = 1/G$. Since the ideal flow (33) minimizes I, small deviations from the ideal flow will result in even smaller deviations of I from I_{ideal}:(5)

> If the flow L obeys the inequality
>
> $|L - L_{ideal}| \leq y\, L_{ideal}$, $y << 1$, then the
>
> I found from L obeys the inequality
>
> $|I - I_{ideal}| \leq y^2\, I_{ideal}$.

For example, if the flow in a square sub-cascade differs from that in the ideal cascade by no more than 30%, then the number of additional separative units in the square sub-cascade will not exceed 9% of the number in the ideal section being replaced. Using the above theorem, one can make a good guess for the flows in each square sub-cascade if the ideal flow has already been calculated.

Equation (51) described an enriching section where $N_{out} > N_{in}$. The equivalent formula for a stripping section where $N_{in} > N_{out}$ is

$$Z = \frac{1}{\epsilon\Delta(\psi)} \tanh^{-1} \left[\frac{(N_{in} - N_{out})\Delta(\psi)}{(N_{in} + N_{out})(1+\psi) - 2N_{in} N_{out} - 2N_w \psi}\right] , \qquad (52)$$

where

$$\psi \equiv - {}^{W}/_{\epsilon L}$$

$$\Delta(\psi) \equiv [1 + 2\psi(1 - 2N_w) + \psi^2]^{\frac{1}{2}} .$$

Using (51) and (52), the minimum theorem discussed above, and Table 2, we can "square-off" the ideal centrifuge cascade discussed above. Table 3 gives the results of the "squaring-off." Note that there are 56,000 elements in the "squared-off" enriching stages compared with 47,400 in the ideal cascade. But the number of stages were reduced from 122 in the ideal cascade to 87 in the "squared-off" cascade.

Table 3

"SQUARING-OFF" THE IDEAL CENTRIFUGE CASCADE

Substage Number	Number of Stages	Flow (kg/min)	N_{in}	N_{out}	Number of Elements
- 1	5	2.25	.00714	.00504	10,800
1	10	2.25	.00714	.0128	21,400
2	19	1. 25	.0128	.0396	20,400
3	25	.5	.0396	.274	11,900
4	33	.1	.274	.903	3,100

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10. ABSTRACT

A discussion of six isotope separation processes; (1) the separation nozzle, (2) sweep diffusion, (3) thermal diffusion, (4) electromagnetic separation, (5) gaseous diffusion, and (6) centrifugation. The emphasis is on the physical principles involved in each process and on the use of these physical differences. Because five of the processes involve devices that give individually a very small separation, a cascade of these devices is required to effect large separations. The steady-state theory of such cascades is also discussed.

11. KEY WORDS

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34