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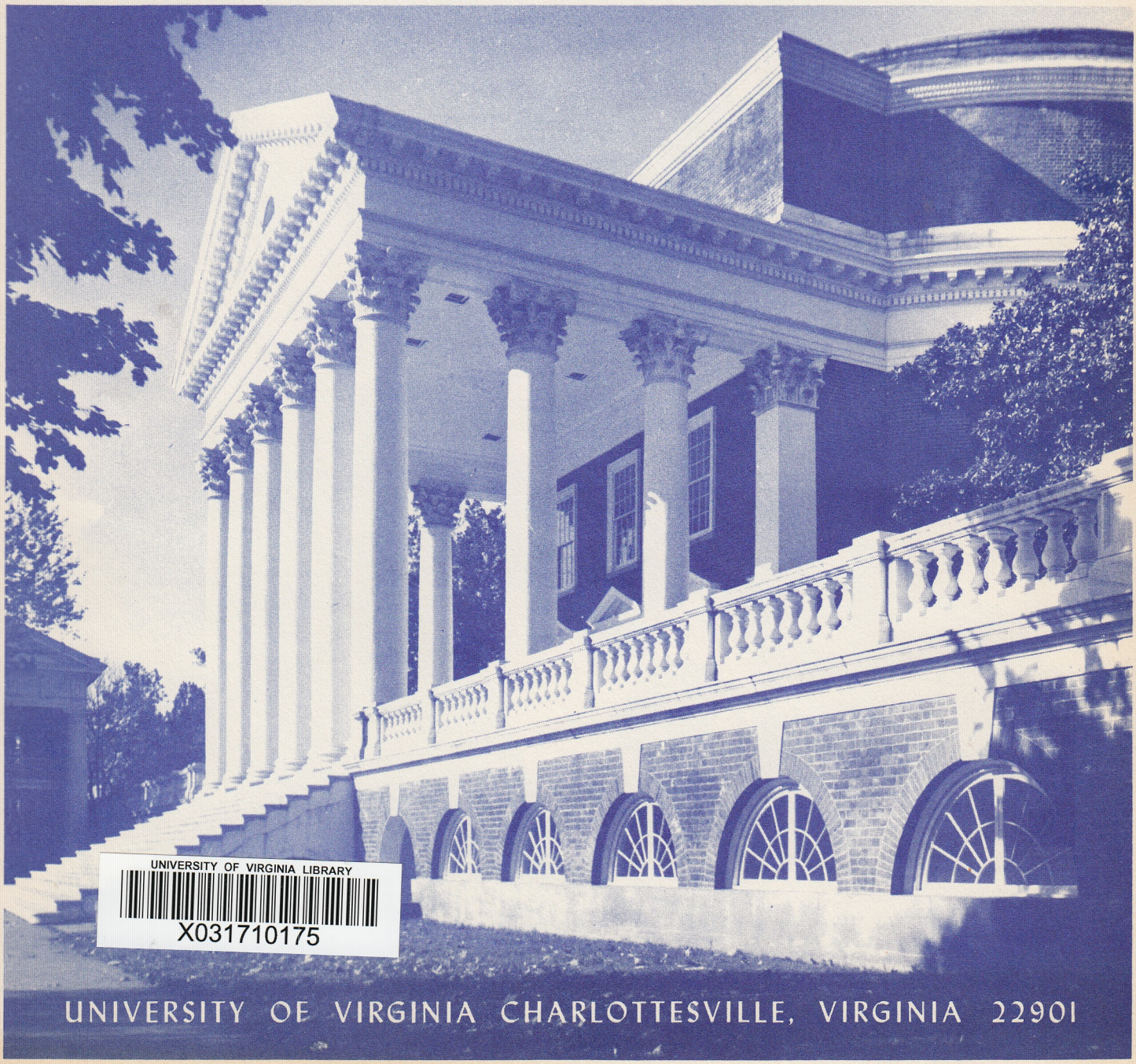
By Jesse W. Beams

DEPARTMENT OF PHYSICS and SCHOOL OF ENGINEERING AND APPLIED SCIENCE



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These notes on the early history of the gas centrifuge development work in the United States were prepared by J. W. Beams, Emeritus Professor of Physics and Senior Research Scholar at the University of Virginia. They were the basis for seminar talks presented by Professor Beams at the Union Carbide Corporation Nuclear Division in Oak Ridge, Tennessee, on November 18 & 19, 1974, at the AiResearch Manufacturing Division of the Garrett Corporation, Torrance, California, December 12 & 13, 1974, and at the School of Engineering and Applied Science of the University of Virginia on February 3, 1975.

SOME NOTES ON THE EARLY HISTORY OF THE GAS CENTRIFUGE WORK IN THE U.S.A.*

By: Jesse W. Beams

Early studies of the products resulting from radioactive transformations soon after the turn of the century showed that these products could be arranged in groups with identical chemical properties but in some cases with different atomic masses. F. Soddy proposed the name of isotopes for these atomic species, i.e., for atomic species with the same nuclear charge but different atomic weights. Starting in 1907 Sir J. J. Thompson, after inventing the mass spectrometer, succeeded in 1912 in clearly showing that neon consisted of two stable isotopes Ne^{20} and Ne^{22}.(1) F. W. Aston,(1) a student of Thompson's, greatly improved the mass spectrometer and measured the atomic masses of most of the isotopes of the known elements. A. J. Dempster in this country also made parallel and almost simultaneous mass measurements of many of the stable isotopes. This work was followed by the more precise work of A. O. Nier and K. T. Bainbridge. Among the important results emerging from this work was the fact that the abundance ratio of the isotopes in each of the stable elements (non radioactive) found on various places of the earth was constant (within the limits of the accuracy of their mass spectrometers). This is now known not to be strictly true but it requires an accurate modern spectrometer to detect a difference. This constant isotopic ratio found in the stable elements on the earth regardless of their source immediately demonstrated the difficulty of separating the isotopes because if this were not the case the biological, chemical and physical processes which have taken place since the origin of the earth would have produced a separation. The constancy of the isotopic abundance ratios makes possible the important use of separated stable isotopes as biological, industrial and laboratory tracers. Their potential use as tracers and their great need for use in studies in basic science, very early generated

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*Based on lectures given at Union Carbide Co. and A.E.C. (Oak Ridge), AiResearch Co. (California) and at the University of Virginia, 1974-75.

an active interest in possible methods of producing their separation. As a result a number of methods of separation were suggested among which was the centrifuge method.

The centrifuge method of separating isotopes was suggested by Lindemann and Aston in 1919.[1,2] Also they developed the equilibrium theory for the separation in an ideal gas and in an incompressible liquid. Also they observed that isotopic separation should take place in the earth's gravitational field in a convection free atmosphere. They estimated that at an altitude of 100,000 ft. in neon the heavy isotope should be reduced from 10 to 8.15 percent. Soon after the above work the theory was further discussed by Chapman[3], Mulliken[4], Harkins[5] and others[6] and several attempts were made to obtain a separation experimentally.[1,4,5,6] Unfortunately in each case the separation experiments were unsuccessful and the method was abandoned as impractical. However, the high rotational speed as well as the convection-free sedimentation, attained by the self balancing vacuum type ultracentrifuge which had been developed at the University of Virginia starting in 1934,[7-11] encouraged us to give the method another trial. This seemed worthwhile, because according to the theory the separation factor should depend principally upon the differences in the masses of the isotopes, rather than their absolute values, so that, if successful the method could separate the isotopes of the heavier as well as the lighter isotopes. The first experiments were successful and the results were in good agreement with the existing theory provided the process was carried out slow enough for quasi-equilibrium to be established.[7,8]

In the first successful experiments the "evaporative centrifuge" method suggested by Mulliken[4] was employed. It consisted in drawing out vapor from the axis of a hollow spinning rotor containing a volitable liquid near its periphery. If this process takes place slowly enough for an isothermal equilibrium to be established between sedimentation and diffusion and if the vapor approximately obeys the ideal gas laws then the separation factor α for the case of a mixture of two isotopes of masses M_1 and M_2 is

$$\alpha = \frac{K_0}{K} = \exp \frac{(M_2 - M_1)\,\omega^2 r^2}{2RT} \qquad (1)$$

where K_0 is the ratio of the quantities of (light) M_1 to (heavy) M_2 isotopes at the axis and K is the same ratio at the periphery, $\omega = 2\pi N$ is the angular velocity of the centrifuge, r is the radius of the inside of the rotor, T is the absolute temperature and R the gas constant. The change in the average atomic weight $\Delta\bar{A}$ of the element in the residue in the rotor from that in the original material with the evaporative centrifuge method according to Mulliken is for a mixture of two isotopes

$$\Delta\bar{A} = \frac{(M_2 - M_1)^2 \, N_1(1 - N_1) \, \omega^2 r^2}{2RT} \times \log_e C \qquad (2)$$

where N_1 represents the mole fraction of the light isotope in the material and C is the cut which is the ratio of the amount of material present initially to that remaining in the centrifuge. Mulliken had further shown that the above relations (1) and (2) are independent of the state of combination of the element. While this is true for the separation factor α, it is not correct for the separative power $\Delta U/\Delta T$ of a centrifuge which is also a function of $\rho D \approx \eta$, where ρ is the density, D the diffusion constant and η the coefficient of viscosity of the gas as will be shown later (see Appendix I).

In view of the fact that the expected separation was independent of the chemical compound in which the element occurred, it was decided to carry out the initial developmental work with carbon tetrachloride because of its availability, non-inflammability, suitable vapor pressure and diffusion constant at room temperature as well as easy condensability at dry ice temperature. Furthermore, we wished to experiment with the separated chlorine isotopes. Also the isotopic analysis was simplified by its suitability for density measurements as well as use in the mass spectrometer which had been developed and built in our laboratory by H. E. Carr.[12] Since the abundance of C^{13} is small there are five principal molecular species namely $C^{12}(Cl^{35})_4$; $C^{12}(Cl^{35})_3(Cl^{37})$; $C^{12}(Cl^{35})_2(Cl^{37})_2$; $C^{12}Cl^{35}(Cl^{37})_3$ and $C^{12}(Cl^{37})_4$ in ordinary CCl_4. The concentrations of the above species if they follow the laws of chance are of course given by the terms of the binomial expansion of $(X_1 + X_2)^4$.

Figure 1 shows a schematic drawing of the centrifuge. The rotating parts of the centrifuge consisted of the rotor R, a hollow stainless steel hypodermic needle shaft A, and an air supported air driven turbine T. The shaft passes through oil sealed vacuum tight glands G_1 and G_2. The rotor spun in a vacuum and was surrounded by a coil of copper tubing which carried circulating water which could be used for heating or cooling the rotor when desired. The rotor itself could be evacuated through a connection at D_2 not shown in the figure. The experimental procedure consisted in distilling or injecting the material to be centrifuged into the rotor through the hollow shaft. Next, the chamber surrounding the rotor was evacuated and the rotor was spun to operating speed. In some experiments when it was necessary to conduct large amounts of heat to or from the rotor, the vacuum chamber was filled with hydrogen at a pressure of about 5 Torr. At this pressure of hydrogen the ratio of heat conducted to the rotor to the gaseous friction on the rotor is roughly a maximum. The rotor itself was then evacuated. Since the rotor was only partially filled, the liquid material to be centrifuged evaporated near the periphery, diffused through the pressure gradient produced by the centrifugal potential, passed out of the hollow shaft and was collected in cold traps at a continuous uniform rate. It was found that the first sample collected had the lighter isotope concentrated while the heavier isotope was concentrated in the residue remaining in the rotor. It can be shown[4,8,11] that the fractional change in the isotopic abundance ratio in the residue f_r and the fractional change in the abundance ratio f_t in the total sample drawn off along the axis are given by the relations

$$f_r = (\alpha - 1) \log_e C \text{ and } f_t = \frac{(1 - \alpha) C \log_e C}{1 - C}$$

where C is the cut. In the first experiments[7,8,9] the rotor of Figure 1 had a hollow flat cylindrical chamber 8.9 cm inside diameter 0.64 cm high and the rotor was spun in a vacuum at 1550 rps (peripheral speed 440 m/sec). The CCl_4 vapor was drawn out at the rate of 2cc of

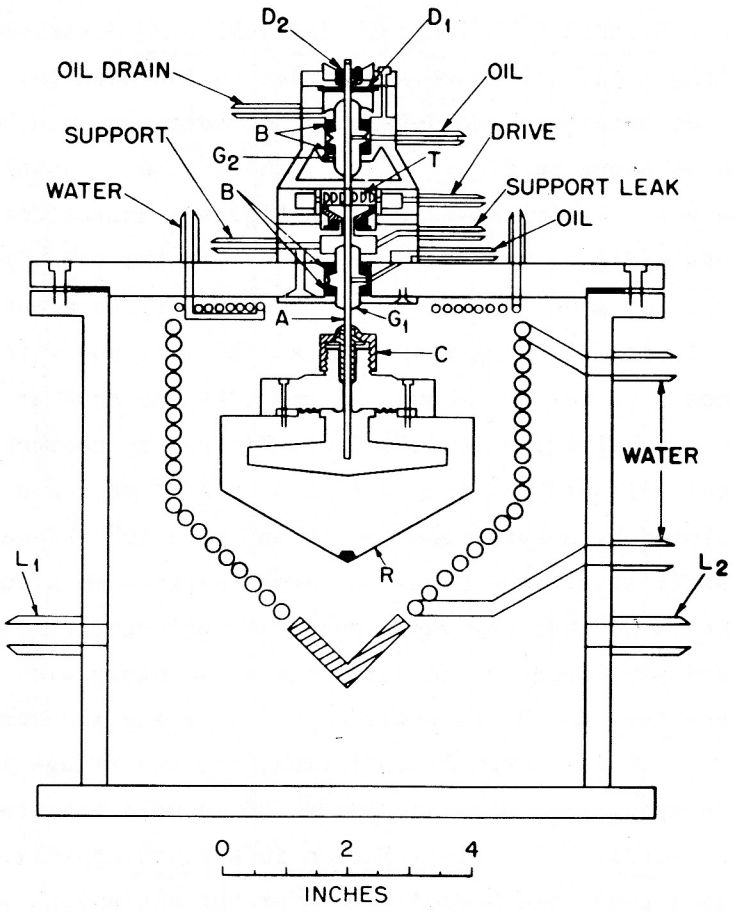

Figure 1 Schematic Drawing of the Centrifuge

liquid per hour. Under these conditions the observed separation was only about 40 to 50 percent of that to be expected from equations (1) and (2) above. When the rate of drawing out vapor was reduced the separation factor α increased to 1.1 and approached the equilibrium theory prediction (1) and (2). When the ratio of drawing off vapor became too large we concluded from rough theoretical considerations, that equilibrium conditions did not exist. In order to increase the rate of drawing out vapor an apparatus was developed for spinning long hollow tubes in a vacuum as shown in Figure 2.[11] The method of spinning the tubular rotor was essentially the same as in Figure 1 except that a lower damper G_4 was added to help damp and stabalize the rotation.[11] The tubular rotor C was made of a steel alloy with a yield point of 159,000 lbs/in^2. It was 11" long x 4" O.D. x 1/2" wall and with its Duralumin ST-14 end caps weighed about 25 lbs.* It was spun at 1060 rps which was 100 rps below its yield point. The vacuum chamber V was a cold rolled steel cylinder 18" long x 7" O.D. x 1/2" wall and the apparatus was enclosed by a barricade consisting of a 10" thickness of soft wood. The small steel tube M was uniformly perforated with a large number of small holes so that the vapor was drawn off uniformly along the axis of the spinning tube. Thin Duralumin ST-14 discs with holes on their axis brace the tube M and prevent it from going into critical flexure vibrations. To carry out a separation, the centrifuge tube was accelerated to operating speed and about 100 cc were injected or distilled into the rotor. This distillation took place rapidly, because the rotor acted as a pump, and heated the rotor but not enough to give serious trouble. The cut varied from about 6 to over 300. Figure 3 shows a curve represented by the circles of the separation obtained to that expected as a function of the rate of removal in cc of liquid CCl_4 per minute. It will be observed that the separation falls off rapidly at rates above 0.22 cc/min which indicated that in addition to the absence of approximate equilibrium there was some disturbing hydrodynamic effect taking place in the rotor. At first sight it was difficult to see how

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*See Appendix II for early development of a magnetic support, magnetic damper and an electric motor drive for the vacuum type centrifuge.

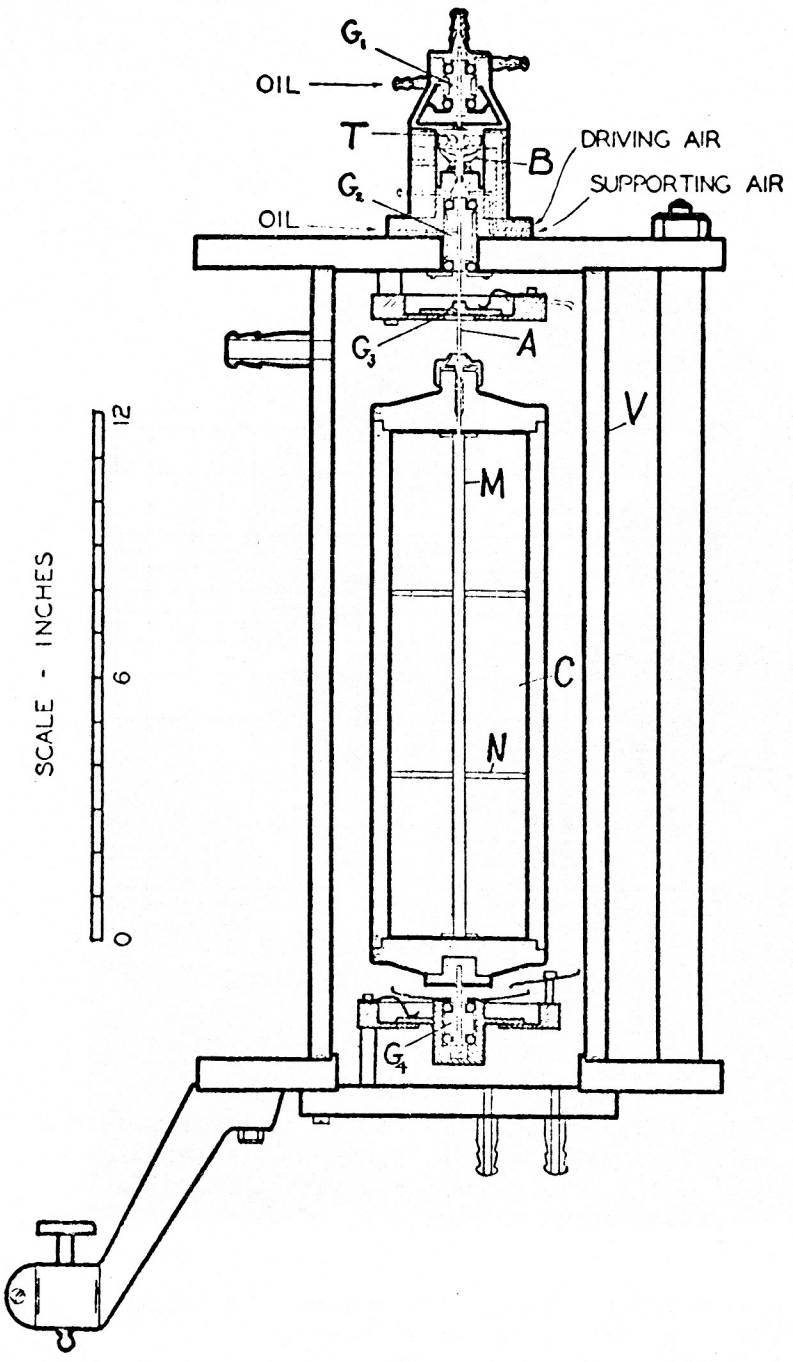

Figure 2 Schematic Vertical Section of the Centrifuge

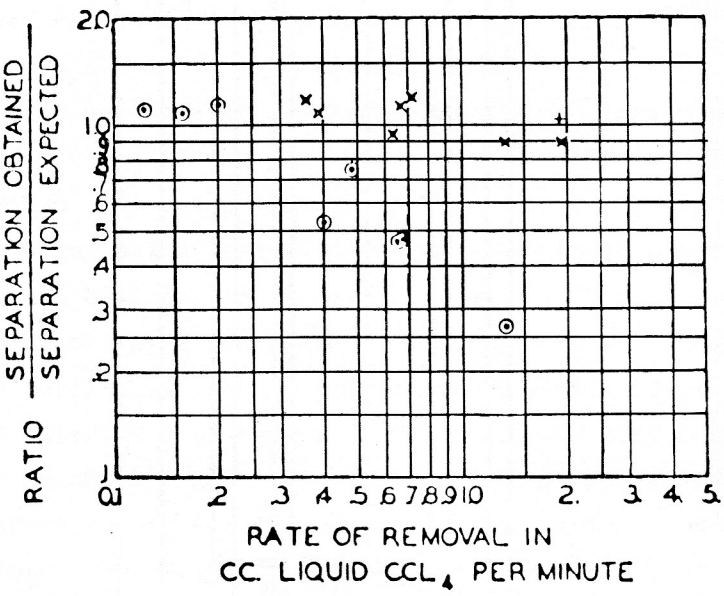

Figure 3 The circles give the results obtained with the hollow tube shown in Figure 1. The crosses give the results obtained with the "spider" baffles shown in Figure 4.

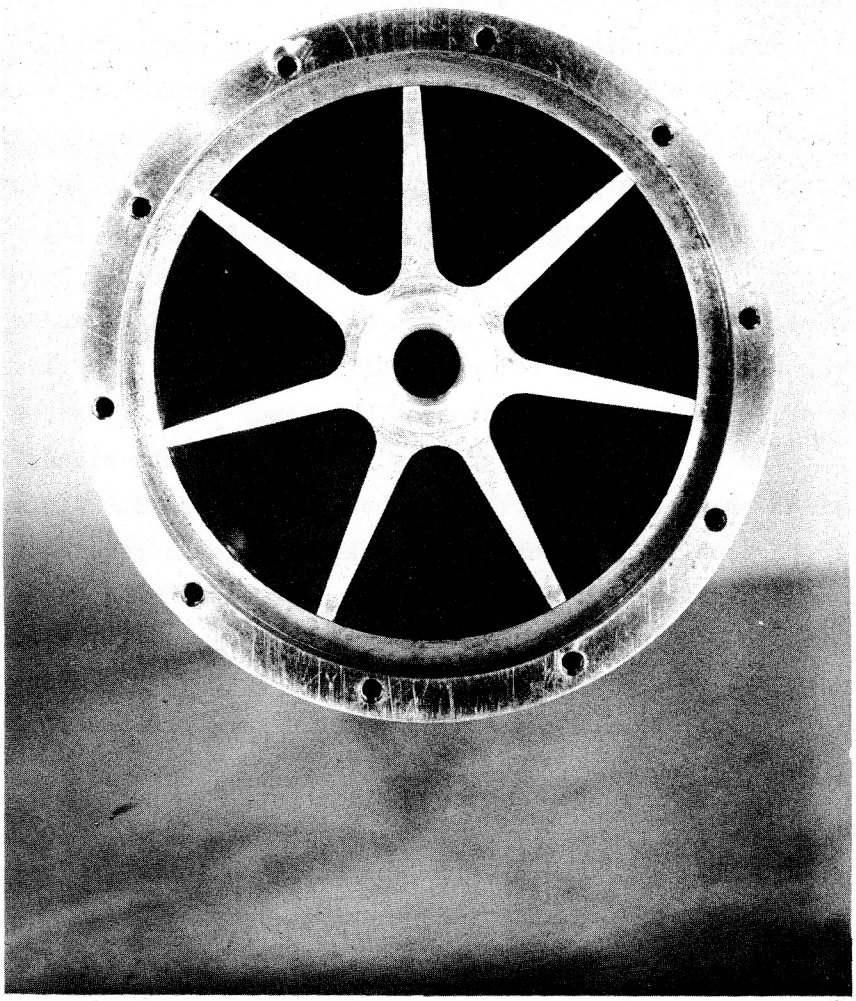

Figure 4 Photograph of tubular rotor with end cap removed, showing the "spider" baffle in place. The vapor enters the axial channel through a large number of uniformly spaced small holes (not shown) connecting the sector-shaped compartments with the axial channel.

radial turbulence could cause the trouble. For example if a small mass of the vapor moved inward along the radius it would expand and cool which would increase its density. This would cause it to move back outward along the radius. Furthermore as it moves inward its angular momentum is conserved and the coriolis and other forces cause its angular velocity to increase which in turn would increase the centrifugal forces also causing the small mass to move back outward, i.e., there is great resistance to radial hydrodynamic flow in a centrifuge.* Nonetheless we were led to believe that a whirlwind in the direction of rotation was taking place inside the centrifuge with some accompanying radial turbulence. Such a whirlwind without radial turbulence would probably increase the separation rather than decreasing it. In order to reduce this whirlwind and turbulence of CCl_4 vapor, a star shaped spider made of Duralumin ST-14 was introduced into the rotor as shown in Figure 4. This spider formed long sector shaped cells in which the diffusion could take place and which would transfer the angular momentum of the vapor to that of the rotor. As a result the separation was greatly increased at the higher rates of withdrawal as shown by the curve of crosses in Figure 3. Since the tubular rotor spun in a vacuum and on air bearings it required only a small amount of power to drive it. Consequently at the higher rates of withdrawal of the CCl_4 vapor the air drive could be turned off. Actually it was necessary in some instances to "brake" the rotor to prevent overspeeding and a resultant rotor explosion. Shortly after the above results were obtained, R. F. Humphreys[13] carried out the separation of the bromine isotopes with a centrifuge similar to that shown in Figure 1. Humphreys[13] and K. Cohen[14] also modified the theory to take into account the case in the evaporative centrifuge method where the vapor is drawn off so rapidly that equilibrium conditions are not obtained. They showed that equation (1) should be replaced by the following:

$$\alpha = \exp \frac{g(M_2 - M_1)\,\omega^2 r^2}{2RT} \qquad (3)$$

$$g = \frac{2K}{2K + Q} \qquad \text{where } K = \frac{2\pi Z \rho D}{\bar{M}}$$

*See Appendix III

where Q is the rate of drawing off vapor, ρ is the vapor density, D is the diffusion constant, Z is the height or length of the centrifuge chamber and $\bar{M}$ the average molecular weight of the vapor or gas being centrifuged. Later Cohen[14] also found that the separative power of an evaporative centrifuge δU is given by the relation

$$\delta U = \frac{1}{2} Z \left(\frac{2K}{2K + Q}\right)^2 (\log_e \alpha_0)^2 \qquad (4)$$

where Z is the length of the centrifuge bowl and α_0 is the value of α in equation (1). The separative work performed by a single separating unit per unit of time or separative power is a measure of the capability of any isotope separation unit. Cohen[14] was further shown that for any gas centrifuge process

$$\delta U = \frac{\pi \rho D Z}{2} \left[\frac{(M_2 - M_1)\, \omega^2 r^2}{2RT}\right]^2 E \qquad (5)$$

where E is a parameter (less than 1) associated with the flow pattern and magnitude of flow, etc. in the centrifuge.* The maximum possible separative work is performed when E = 1.

Although it was demonstrated experimentally that the evaporative centrifuge method could be cascaded and that the experimental and theoretical results were in excellent agreement, it has two serious drawbacks for the separation of large quantities of isotopes. First, it is a batch process and second, the liquified material which must be maintained in the periphery unnecessarily loads the rotor. Consequently in 1937 the development of the vacuum-type tubular centrifuge was started in which either gases or liquids could be circulated through the long centrifuge while it was spinning.[9,10,15,16] Although some of these early tubular ultracentrifuges were used for the purification of substances of interest in biology and medicine,[17,18] the major effort was concentrated on gas centrifuging.[10] Two general types of flow patterns of the gas in the centrifuge were used. The first was the so called "flow through" or concurrent flow method in which the gas or vapor entered the spinning tube at a continuous rate at one end and emerged continuously at the other

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*See Appendix I

end in a light fraction collected near the axis and in a heavy fraction collected near the periphery. The second method known as the countercurrent method in which the gas or vapor enters the spinning tube continuously usually at one end or near the middle and is collected continuously in a heavy fraction at one end and a light fraction at the other end. Various means were used for producing a countercurrent flow inside the spinning tube in such a way that the light fractions could be collected at one end and the heavier isotope fractions at the other.[10] Figure 5 shows a schematic diagram of the first vacuum-type tubular centrifuge developed for the concurrent and countercurrent flow experiments.[10] The rotating parts consist of the air driven air supported turbine T, the tubular rotor R and the flexible stainless steel tubular shafts S_1, S_2 and S_3. S_2 and S_3 pass through the vacuum tight oil glands G_2 and G_3 while S_2 passes through the vacuum tight oil gland G_4. G_3 and G_4 make it possible to separate the fraction of the material flowing out through S_2 from that flowing out between the coaxial tubular shafts S_2 and S_3. The first tubular rotor was made of alloy steel and was 7.62 in I.D. and 12 in long with Duralumin ST-14 end caps. It was spun at 1060 rps. Figure 5 shows the arrangement for the concurrent flow method of separation. For the early countercurrent separation experiments an arrangement of shafts almost identical with that at the bottom was added at the top so that material from the axis and from the periphery could be withdrawn or material could be forced into each end of the spinning tube continuously. The flow through or concurrent flow pattern was used first. Because of its higher vapor pressure ethyl chloride vapor was used instead of CCl_4. Also N_2 and CO_2 mixtures were used initially to check the method. The vapor entered through A and was drawn out in a light fraction at C and a heavy fraction at B. Later Cohen[14] and E. V. Murphree[14] independently worked out the theory for the concurrent flow and the experimental results obtained were found to be in good agreement with theory. During the course of these experiments (1938),

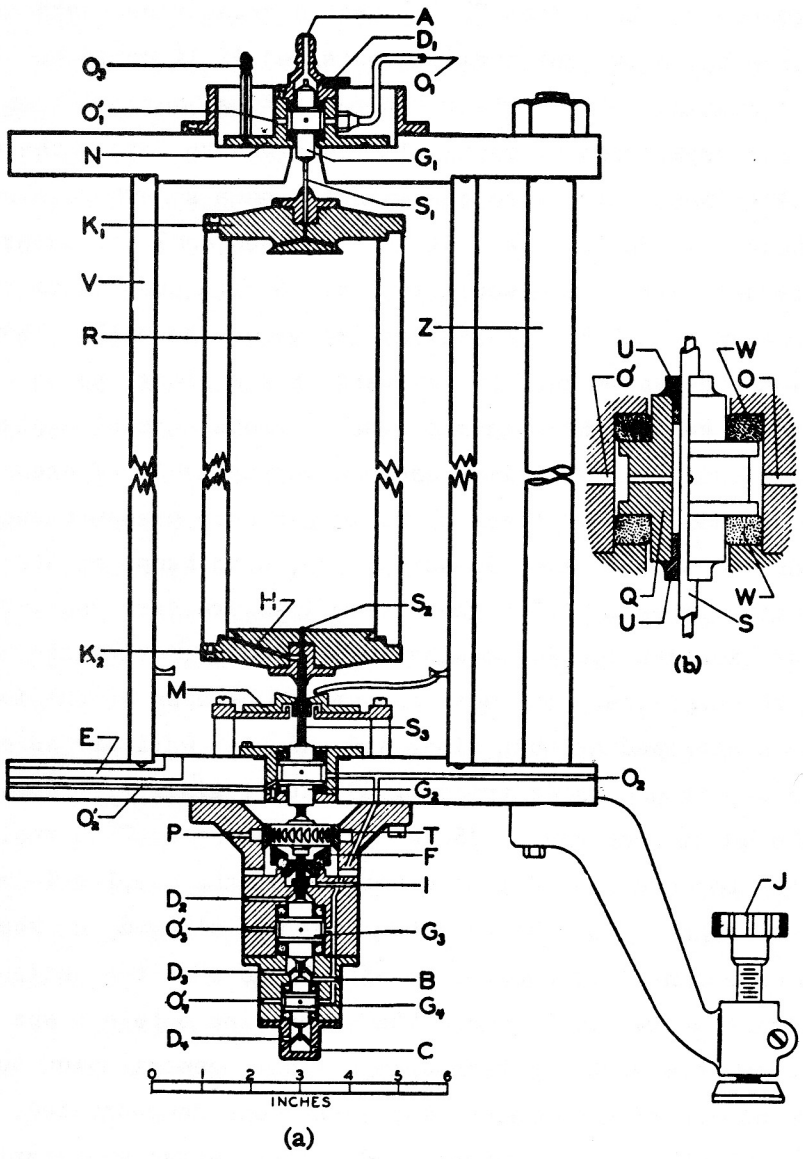

Figure 5 (a) Drawing of Tubular Vacuum-Type Centrifuge Used to Centrifuge Gases and Vapors

(b) Enlarged Drawing of Vacuum-Tight Oil Gland

H. C. Urey suggested to the writer[10,19] that a greater separation per unit length of the spinning tube could be obtained if it could be used as a "fractionating column" where the centrifugal potential or field would serve as the separation element. The idea was to inject the unseparated material (feed CCl_4) into the spinning tube somewhere along its length and to heat say the lower end of the tube and cool the upper end. In this way internal circulation would take place with one stream rising near the spinning axis and the other decending along the wall. The upward stream would be vapor and the downward stream along the wall could be either liquid or vapor. He suggested that "scoops" could be placed at the ends for withdrawing the light and heavy fractions. These scoops convert the mechanical energy of the whirling gas into pressure energy and were in wide use at that time in aeroplanes, wind tunnels, etc. for converting the kinetic energy of a gas stream into pressure energy. They were designed at Virginia for increasing the ease of removing the heavy gas (UF_6) from the centrifuge but were not needed because of the low peripheral speeds attained at that time, and the high wall pressure employed. This scheme was later tried (1939) at Virginia with CCl_4 in a Duralumin tube 50 cm long and 1 1/8 in I.D. spinning at 2300 rps. It was cooled at the top and heated at the bottom and the samples (~1cc) were withdrawn through hollow shafts after time was allowed for equilibrium to occur. The tube contained about 6 cc of CCl_4 so that the peripheral stream was at least partially liquid. The separation obtained was between four and five times that possible in a single centrifuging and the countercurrent centrifuge process apparently was demonstrated.
H. C. Pollock and H. H. Kingdon (1939) used a circulating evaporative-centrifuge method for the concentration of the tin isotopes, but the method was not developed further due to the urgency of other work at the time.[20] In 1940 A. Bramley and K. Brewer[21] and Martin and Kuhn[22] in important papers proposed methods of circulating the gas in a spinning centrifuge tube by thermal means. However it remained for H. C. Urey, K. Cohen, C. Skarstrom and their collaborators[14] and P. A. M. Dirac[23] in 1941 to work out the general mathematical theory and to indicate theoretically the most efficient way to go about separating the isotopes by centrifuging.

Soon after the announcement of uranium fission by neutrons in March 1939, the writer and L. B. Snoddy at the University of Virginia, like many other workers, became interested in the separation of U^{235} and U^{238} isotopes. The centrifuge method appeared to be well suited for the separation of these heavy isotopes since the separation factor depended upon the difference of the masses ΔM in the exponent of e rather than their absolute values. (i.e., in the diffusion method the separation depends upon $\sim\Delta M/2M$ and for distillation and chemical exchange it depends upon $\sim\Delta M/M^2$). Also the separation factor did not depend upon the state of combination of the elements or did it decrease with increase or decrease in concentration of the rare isotope. Furthermore the elementary separation process is quasi reversible and should require a relatively small amount of energy, i.e., most of the energy requirements are for spinning the centrifuges, maintaining the vacuum in the jacket and circulating the gas both inside and outside the centrifuge, etc. which could be reduced by proper design. A search of the literature at that time showed that of all of the uranium compounds, UF_6 was the best adapted for the gas centrifuge process. It was liquid at 69°C with a vapor pressure of 2 atmospheres and hence could be used in any of the centrifuge methods which had been proposed. On the other hand apparently only a small amount had ever been made and it was not commercially available. The literature further revealed that UF_6 attacked water violently as well as all metals except gold and nickel. In addition it was extremely poisonous. The process of making it was not clear and the estimated yields were small. Consequently we were faced with the task of producing our own UF_6 if the work was to go forward. Fortunately my colleague Professor L. B. Snoddy was an excellent chemist as well as a physicist and he undertook to make a small amount of UF_6 in order to study its physical and chemical properties. From these early studies he concluded that, if the UF_6 were pure it not only did not attack gold and nickel but it did not appreciably attack stainless steel and duraluminum, the metals used in the centrifuge, when they were dry and at room temperature. Snoddy also confirmed that fluorination of the metals before use was helpful. While this work was getting under way (March 1940) Dr. M. A. Tuve of the Department of Terrestrial Magnetism of the Carnegie Institution of

Washington became interested in the separation of the uranium isotopes and very kindly obtained for us a generous grant in aid from the Carnegie Institution to get the work started. It will be recalled that Dr. Tuve and his colleagues were among the first workers in this country to experimentally confirm the fission of uranium by neutrons. In the meantime, Dr. R. Gunn, Director of the Naval Research Laboratory, also offered assistance and secured funds to help finance our work for a year (August 2, 1940-May 31, 1941 total amount $6,353.57). Dr. Gunn also was most helpful in searching for a supply of UF_6. Dr. Gunn was most anxious as early as 1940 to obtain enriched U^{235} in order to construct a "nuclear reactor with which to drive a submarine." He actually had some advanced practical ideas on how to do it and together with his colleague P. Abelson, later received one of the Navy's highest awards in part for this work. The generous early financial support allowed us to expand our research efforts. By the end of 1940 the support of our work was taken over first by the NDRC and later by the OSRD. The work was supervised by Dr. L. J. Briggs, Chairman of the Uranium Committee, who took an active interest in the experiments and who personally not only helped us to get special materials, but made many useful suggestions. Dr. Briggs and his committee set up four groups to investigate the feasibility of separating the uranium isotopes by centrifuging as follows: (1) A group at Columbia University under the direction of Dr. H. C. Urey whose purpose was to work out the theory in detail and to coordinate the overall program; (2) A group at Westinghouse Electric Company under L. B. Chubb whose objective was to develop and build the centrifuges which were to be used in a pilot plant; (3) A group at the Standard Oil Development Company (now a part of EXXON) under Dr. E. V. Murphree whose function was to build and operate the pilot plant and to plan a larger separation plant; (4) A group at the University of Virginia whose purpose was to test the theory of separation by experiment. Also it was to make such tests as the other groups might require. The four groups worked together most harmoniously especially considering the stresses of war. This spirit of cooperating was due in a large measure to the kindliness and administrative genius of Dr. Briggs.

EXPERIMENTAL RESULTS

The first attempts to concentrate the U^{235} by centrifuging were held up almost a year because enough UF_6 could not be obtained for the experiments. Late in 1940, Ross Gunn of the Naval Research Laboratory supplied 6 g of UF_6, and the first experiments were made immediately. These experiments demonstrated that the material could be put through the centrifuging process without loss or deterioration; however, the samples collected were not measured. The mass spectrometer available at the University of Virginia at that time was not suitable for determining the isotopic ratios in uranium, so the centrifuged product was sent away for analysis and was apparently lost before being analyzed. After these first experiments were completed, the method was set aside until a sufficient supply of UF_6 became available. At this time, emphasis was shifted to developing long tubular centrifuges for use with the flow-through and countercurrent methods to be described later.

Fortunately in 1941 Drs. Briggs and Gunn made an ample supply of UF_6 available, and at the suggestion of Briggs the experiments using the evaporative centrifuge were resumed. Also Alfred O. C. Nier not only offered to measure the changes in the isotopic-abundance ratios with his mass spectrometer but later loaned one of these instruments. E. P. Ney, one of Nier's assistants, also assisted in the experiment. In addition, L. F. Curtis and his group at the Bureau of Standards used the alpha-counting technique to analyze some of the centrifuged samples. It would have been impossible to have reached any conclusions at all at that time without the measurements of Nier and Curtis.

Tables 1 and 2 show the results obtained with the samples analyzed. Table 1 gives the results obtained with a single centrifuging, while Table 2 gives the data obtained for cascade-centrifuging experiments, i.e., recentrifuging. In Table 1, the first column gives the date of the experiment. The second column gives the temperature of the rotor, which is precise to ± 20°C. The third column gives the weight of the sample in grams or the amount of product in which the isotopic ratio was changed.

Table 1 - Analysis of Samples Produced by Single-centrifuging Method

Date	Rotor temp. °C	Weight of Product, g	Rate of pumping (Q), g/sec (h = 0.72 cm)	Increase in isotopic-abundance ratio of U^{235}/U^{238} Calculated on Mulliken-Harkins Theory, %	Calculated on Humphreys-Cohen Theory, %	Measured values of Nier, %	Measured Values of Curtis, %
9/26/41	82	1.2	3.2×10^{-4}	5.0	4.3	4.0	3.0
10/7/41	81	0.6	1×10^{-4}	5.7	5.4	5.5	
10/9/41	81-85	0.5	0.37×10^{-4}	5.3	5.2	4.6	
10/17/41	81-83	1.0	0.7×10^{-4}	5.5	5.3	5.6	5.5
10/23/41	83	0.8	2.8×10^{-4}	5.0	4.4	5.3	
11/11/41	81	1.1	3.1×10^{-4}	5.6	4.8	4.8	4.6
12/31/42	90	3.6	5.6×10^{-3}	3.0	0.8	0.7	

Table 2 - Analysis of Samples Produced by Cascade-centrifuging Method

Date	Rotor temp., °C	Weight of product, g	Number of cascades	Change in abundance ratio of U^{235}/U^{238} Calculated on Humphreys-Cohen theory, %	Measured values of Nier, %	Measured values of Curtis, %
12/22/41	81 - 87	1	1	+14	+12.4	11-12.7
12/22/41	81 - 87	1	1	-12	-9.8	-7.7
12/22/41	90 - 95	1	2	+18	+17.5	
12/22/41	88 - 93	1	2	-19	-20 to -28	

The fourth column gives the rate of pumping Q. The fifth column gives the increase in the abundance ratio of U^{235}/U^{238} in percent, as calculated by the Mulliken-Harkins theory for no flow. This is the value of $\alpha - 1 = \varepsilon$ (equation 3 when g = 1) times 100 to give the separation in percent. The sixth column gives the separation calculated by the more exact theory of Humphreys and Cohen which allows for the flow, i.e., $\alpha - 1 = \varepsilon$, equation 3. This is also multiplied by 100 to give the percent separation. The seventh column gives the values measured by Nier with his mass spectrometer, and the eighth column gives the values measured by Curtis with the alpha-counting method. The alpha-particle counting method also was used at the University of Virginia by J. H. Reisner for measuring the isotopic separation in Tables 1 and 2. His results were in good agreement with those of Nier and Curtis. Both the calculated and measured values are in excellent agreement.

In Table 2 the values for the first and second cascade processes are given. Column five gives the calculated value for the percent separation (ε in equation 3) times 100, as calculated by the Humphreys-Cohen theory. The sixth column gives the measured values of Nier, and the seventh column gives the measured values of Curtis. In these experiments some of the product samples had U^{235} enriched and others had U^{238} enriched. The experimental values are in good agreement with theory considering the necessarily complicated weighing of products needed for the calculations. In the last sample in Table 2, the measured values are somewhat inaccurate, but if the smallest values are taken, a measured change in isotopic ratio between the last two 1 g samples of about 40 percent is obtained. These two samples were sent to Briggs for use in other experiments immediately after analysis. The fact that the results of the method of evaporative centrifuge were in good agreement with the Cohen theory showed that the theory could be depended upon to predict the performance of a centrifuge provided the experimental conditions were the same as those assumed in the theory.

Another result of interest is the rather close agreement between Nier's mass-spectrometer measurements and those of Curtis' alpha-counting method. The agreement seems to be best where the separation is small. Incidentally, this agreement is an independent check on the fact that the centrifuge separation depends upon the difference of the masses in the exponent of e since this was assumed in the alpha-counting method.

The last experiment recorded in Table 1 shows that the theory holds remarkably well even at a high flow rate. This is important because it might be expected that, when the vapor is drawn out rapidly along the axis, some stirring of the vapor should take place. For example, when the vapor moves along the radius toward the axis, its angular momentum is conserved and hence tends to rotate in the same direction as the centrifuge. However, the height of the chamber was only 0.72 cm so that the friction on the upper and lower ends of the rotor chamber apparently prevents excessive rotation. In previous experiments[5] this stirring was prevented by radial baffles in the cases of the separation of isotopes in long rotors, so that it presented no problem in the method.

The evaporative centrifuge method as applied to the separation of the uranium isotopes has been outlined above in some detail because it clearly established for the first time that the observed elementary separation was in excellent agreement with the theory when UF_6 was used as the process gas. The problem remaining was to determine if the hydrodynamic flow inside the centrifuge in both the concurrent flow and in the counter flow processes could be produced in such a way as to give the separative work expected. This, of course, required the development of long tubular centrifuges spinning at as high peripheral velocities as possible. Consequently while the evaporative centrifuge experiments were being carried out and while it was necessary to wait until enough UF_6 would become available for tubular operation the effort was directed to the development of long tubular rotors and testing them on the separation of gaseous mixtures and isotopes other than uranium. As a result as soon as ample supplies of UF_6 became available the tubular centrifuges were ready for the experimental separation tests. Two tubular centrifuges

were used for most of the separation work - one 32 in. long with an I.D. of 3 in. and a second 11 ft. 4 in. in length, 8.35 in. O.D., and 7.35 in. I.D. They were both made of forged Duralumin ST-14. The latter was obtained for us by Dr. E. V. Murphree and was as long as could be accurately machined at that time. These tubes were spun by steam or air turbines and were operated above their second critical. The first tube operated at 1020 ± 0.3 rps and the second tube at 350 ± 0.3 rps. They both operated stably and without appreciable wear for many days. The centrifuges have been described previously and reference should be made to this book for details.[24] The above centrifuges were used in both concurrent and countercurrent operation. However, it soon became evident that the countercurrent operation in practice gave the highest separative work per unit length of tube. Also this was in agreement with the theory of Cohen for the practical case. For this reason only counter flow operation will be discussed here and reference made to the detailed previous account of the work.[24]

Figure 6 shows a schematic diagram of a typical countercurrent operation. UF_6 vapor flows at a rate of L mg/sec into the upper end of the spinning tube and is directed downward near the periphery at AA in a thin cylindrical stream. It flows out of the centrifuge at BB into a centrifugal pump located on the shaft of the centrifuge.

This external pump was used for circulating the gas instead of such devices as scoops located inside the spinning tube in order to avoid all possible disturbance of the flow pattern inside the spinning tube, i.e., the flow was externally instead of internally driven, in order to insure that the flow pattern was the same as assumed by the Cohen theory. Dr. Murphree, Dr. Urey, Dr. Briggs and others strongly emphasized to us that the principal objective of our experiments was to test the Cohen theory so that it could be used to design a pilot plant. The actual magnitude of the separation was secondary. The gas leaving the centrifugal pump passes through a flow meter and is divided into two streams. One stream is collected in a cold trap as the product P mg/sec. At the same time, P mg/sec of ordinary UF_6 (called the make up) is introduced into the second stream which in turn

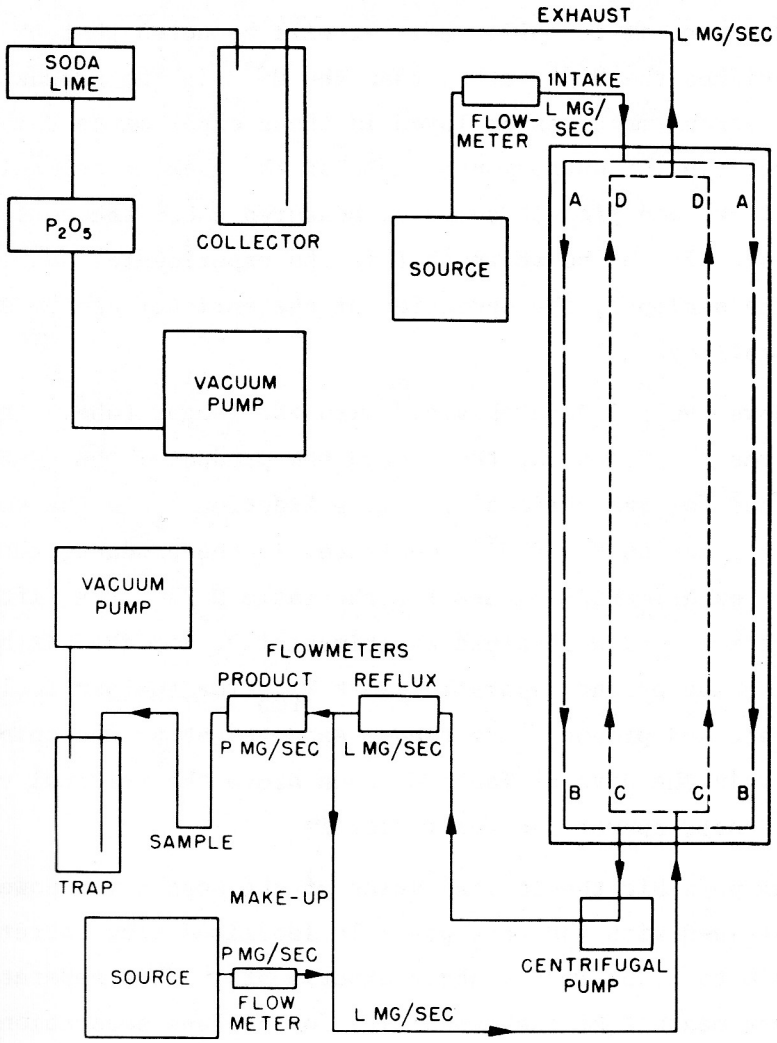

Figure 6 Diagram of Type II Operation for a Single Machine

is returned to the lower end of the spinning tube through the divided shaft and directed upward by the end cap in a thin, cylindrical stream CC to DD where it passes out of the end cap and divided centrifuge shaft into a cold trap where it is collected. It will be noted that this type of operation enriches the U^{238} rather than the U^{235} in the product. This "stripper" arrangement was employed in these experiments rather than the enricher type of operation because of the greater reliability with which the flows and product could be measured under the conditions of the experiment. It can be shown that if the experimental data are available for the stripper, the operation of the enricher can be determined with reliability.

Table 3 shows typical data obtained with the longer tube. The first column gives the flows, L, the second the products, the fourth the theoretical values for the ratio of the mole fraction N_o in the source of U^{235} to the mole fraction N_z of U^{235} collected in the product, the third column gives the experimental values for the ratio N_o/N_z, the fifth column gives the separative work W obtained experimentally, and the sixth column the theoretical value of the separative work W_{fmp} for the particular flow pattern, flow rate and product rate used. As noted above the spinning tube used to obtain the data of Table 3 spun above the critical vibration frequency and, hence, operated supercritically.

The maximum possible theoretical value of the separative power W_{max} which can be obtained with the best possible idealized flow pattern varied from 0.050 to 0.051 in the above experiments. This separative power is a direct measure of the capability for isotope separation. The maximum theoretical ratio of W_{fmp} to W_{max} is 81 percent, assuming an idealized thin stream flow pattern, as shown in Figure 6.

It will be observed that the experimental results are in good agreement with the Cohen theory. Over certain ranges of flow rates and product rates there are some differences but this is to be expected. In any case, the data shows that if the flow pattern inside the centrifuge is

Table 3

L mg/sec	P mg/sec	N_o/N_z Obtained	N_o/N_z Theoretical	W mg/sec Obtained	W_{fmp} mg/sec Theoretical
80.7	25.65	1.039	1.0394	0.038	0.0383
79.5	19.52	1.0475	1.048	0.042	0.0429
79.3	15.06	1.054	1.057	0.041	0.0469
79.7	10.64	1.061	1.071	0.037	0.0506
79.3	5.35	1.073	1.101	0.026	0.0496
64.3	19.80	1.047	1.0424	0.042	0.0341
65.4	14.90	1.054	1.054	0.041	0.0403
65.2	9.70	1.070	0.072	0.044	0.0472
64.9	24.83	1.042	0.036	0.041	0.0302
65.2	15.11	1.053	1.053	0.040	0.040
65.3	10.26	1.067	1.069	0.043	0.0463
50.0	19.72	1.044	1.036	0.036	0.0245
49.8	15.03	1.052	1.045	0.038	0.0285
50.3	10.12	1.066	1.063	0.042	0.0373

known, the Cohen theory[14] can be used with reliability to calculate the amount of isotope separation in the centrifuge or the amount of separative work.

While the work just described was being carried out, the Westinghouse Company was designing and building the centrifuges which were to be used in the pilot plant to be operated by the Standard Oil Development Company in Bay Way, New Jersey. They started designs in 1941 and delivered two machines in Bay Way; one in May and one in September, 1943. The cylindrical centrifuge bowls of these machines were 42 in. long, 7.2 in. I. D. and 8.2 in. O.D. with a rated speed of 28,200 rpm. They operated at subcritical frequencies and were driven by electrical motors. Figure 7 shows a sketch of these short bowl machines. In addition to developing and constructing two of the above centrifuges, Westinghouse developed a dynamic model of a 132 in. long centrifuge with the same bowl diameter. This was assembled and tested at 28,200 rpm in December 1943. The over-all length of the machine was 188 in. and the total weight 3,200 lbs. This machine was finally wrecked in tests; however, the experience indicated that successful design and operation of a production centrifuge of this length was feasible. Since the over-all project was terminated in January, 1944, the tests were not continued.

Originally, the Standard Oil Development Company had planned to operate a pilot plant of 24 identical centrifuges, but the delivery of the machines was much delayed and only one centrifuge was used for any length of time. This centrifuge was operated a total of 99 days of which 93 days were with UF_6 feed. During this period, the longest time of uninterrupted UF_6 operation was 69 days. The operation was finally interrupted by a leakage of UF_6 from the bowl seals into the casing space. Since this occurred on January 23, 1944, and the work was terminated on January 31, 1944, it was not repaired. The single centrifuge operated amply long to give valuable information. A total of 1,000 lbs. of UF_6 was passed through the centrifuge. 800 lbs. of this had its average composition in U^{235} reduced by about 1 percent while 200 lbs. of the product had the U^{235} isotope enriched by an average of about 5 percent. Figure 8 shows the separative efficiency plotted against the product rate.

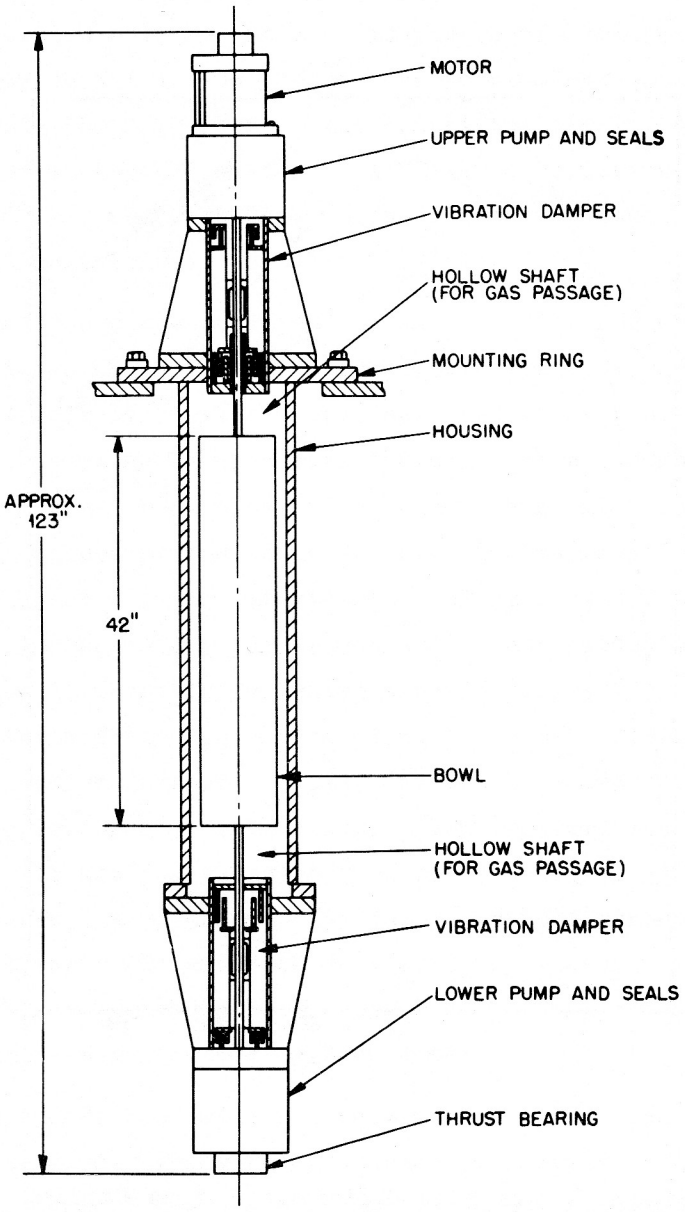

Figure 7 Short-Bowl Gas Separator

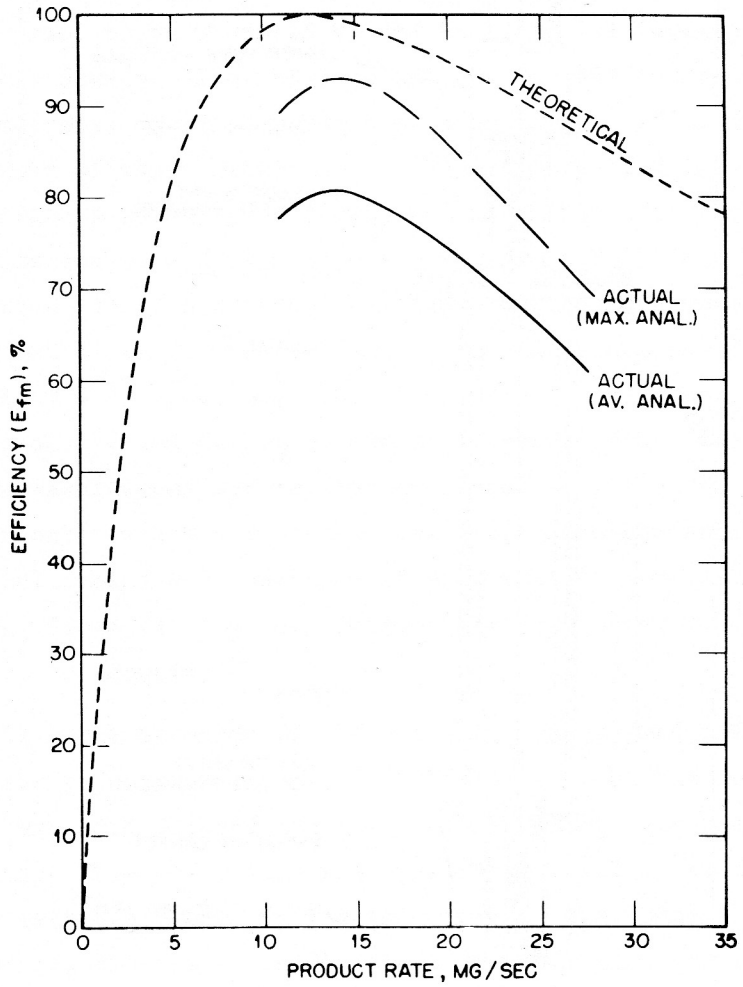

Figure 8 Graph of Efficiency Versus Product Rate for Type II Operation

It will be noted that this efficiency falls between 80 and 90 percent which uncertainty was due to the possible errors in the analysis. To quote their report, "The operating experience with the single-centrifuge unit led to the conclusion that the unit is adaptable to commercial plant operation of long duration. No insurmountable problems were recognized in connection with the operation of the centrifuge itself, or the various auxiliary systems or the UF_6 system." A fuller description, not only of this work, but of all the work done on the project during the war is contained in an AEC unclassified publication.(24)

As mentioned above the decision was made by OSRD and the Manhattan District U. S. Army to proceed with the diffusion method and to terminate all work on the gas centrifuge project on January 31, 1944. Although the work was halted abruptly, the work had proceeded far enough to allow the following conclusions to be drawn concerning the gas centriguge method: (1) The method proved to be an effective method of separating the uranium isotopes; (2) The experimental values obtained were in close agreement with the separation theory and sufficient operational data were collected to make it possible to design a pilot plant; (3) Centrifuges could be built and made to operate for long periods of time while separating the uranium isotopes in UF_6; (4) Tubular centrifuges could be satisfactorily operated both subcritically and supercritically while separating UF_6. It was demonstrated that rotors over 11 ft. long could be satisfactorily operated. (5) The limiting factor in the gas centrifuge was clearly the strength divided by the density of the rotor material. If this ratio could be increased the effectiveness of the method would be increased accordingly.

During the decade following the termination of the gas centrifuge project in the U. S. A., the work lay dormant primarily because of the strict classification of the project. On the other hand, the work in Germany and Russia at least continued. In Germany, W. E. Groth and K. Beyerle improved their war time centrifuges and by 1958 had made significant advances especially in the study of the internally driven counter flow patterns. They also had developed their centrifuges to the

point where they could offer them for "sale" on a commercial basis. However the cost of separative work with their centrifuges was too large to compete with the U. S. diffusion plants.

Work in Russia on the centrifuge method of isotope separation was carried out by a team of Germans and Russians under the scientific direction of Professor Max Steenbeck, Dr. Zippe, Dr. Scheffel, and Dr. Steudel.[19] In the period 1946 to 1953, work was first done by this group on a long tube approach using short elements of tubes connected by sylphons so as to give an over-all length of 3 meters and a ratio of length to diameter of 50 to 1. The operational peripheral speed of the units was 250 meters/sec. Life-time tests of over 3,000 hours were successfully completed. Separation tests using an axial thermal gradient to produce the countercurrent flow demonstrated an efficiency of about 65 percent of the theoretical separation ability (the basis of the calculations are not known). This device apparently was developed for use as the final enrichment stages of a diffusion plant, but the German group was informed in 1953 that this application was no longer required. As a result, the group turned their attention to the development of a simple inexpensive short bowl centrifuge which would be suitable for a complete plant starting with natural uranium.*

In the late 1940's, it became clear that a careful reappraisal should be made of the gas centrifuge project in this country. As a result of "spin off" from other projects stronger rotor material became available, etc. which should enhance the effectiveness of the method. Fortunately, Dr. P. W. McDaniel and Dr. G. A. Kolstad of the AEC Research Division became interested in the project and modest funds were provided to reactivate the project on a small scale at the University of Virginia. Dr. A. R. Kuhlthau who had worked on the wartime project, was given the responsibility of assembling the personnel and getting the work under way. Since so much could be gained by increasing the peripheral velocity and length of the centrifuge tube, Dr. Kuhlthau and his colleagues started out to maximize both of these factors. In addition to this developmental work, Dr. Kuhlthau and his colleagues undertook to evaluate the

*NOTE: Quoted from memory of conversations and reports.

gas centrifuge reports coming out of Europe on a continuing basis. In connection with this latter work they became interested in the Russian work described in an interview with Dr. G. Zippe who had been allowed to return to Germany from Russia. The interview had been carried out by Dr. M. Schutte who reported to Dr. K. Brewer at the Navy. Dr. Brewer had long been interested in the gas centrifuge project and had made an important contribution to the theory.[21] Through the most effective efforts of Dr. Kuhlthau, Dr. McDaniel and Dr. Kolstad of the AEC, and Dr. Brewer and Dr. Schutte, Dr. Zippe was invited to come to Virginia and substantially repeat the short bowl experiments which he had carried out in Russia. This work was started in August 1958 and completed in June 1960 when Dr. Zippe returned to Germany and/or his native Austria.

A schematic diagram of the Zippe short bowl centrifuge is shown in Figure 9. The centrifuge rotor, 0, spins about its inertial axis on a thin, flexible steel needle, A. The rotors are made of 7075-T6 aluminum alloy, the tubes being taken directly from the extrusion benches and, hence, requiring no machining other than that necessary for adapting the end caps. The rotor is 3 inches O.D. and 13 inches long. This provides a subcritical operation; i.e., below the fundamental vibration frequency of the tube, at the maximum permissible operating speed of 350 m/sec peripheral velocity obtainable with this material.

The shaft needle itself is centered on a depression of a hard metallic plate, J, which, in turn, is centered elastically and whose lateral motion is damped by oil friction. A closer look at the construction of the support bearing is in order as it represents one of the major achievements of the design. A schematic of this assembly is shown in Figure 10. The only continuous contact in the entire rotor takes place between the tip end of the flexible needle, 2, and the bearing point, 9. The flexible needle itself is a steel piano wire whose tip has been heated to glass hardness. It is soldered onto a steel shaft, 3, which is, in turn, fastened to the steel plate, 12. This plate is attached to the bottom of the rotor and is actually the armature of the drive motor.

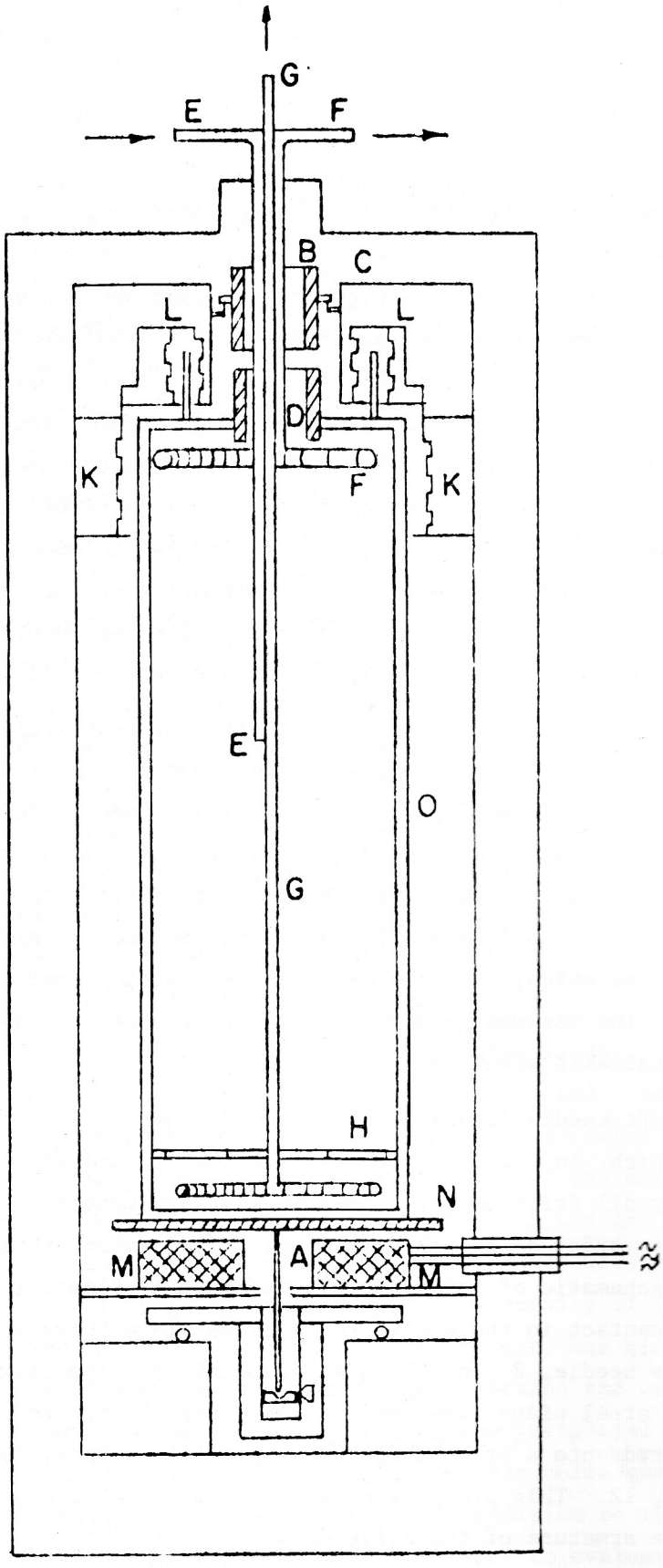

Figure 9 Short Bowl Centrifuge

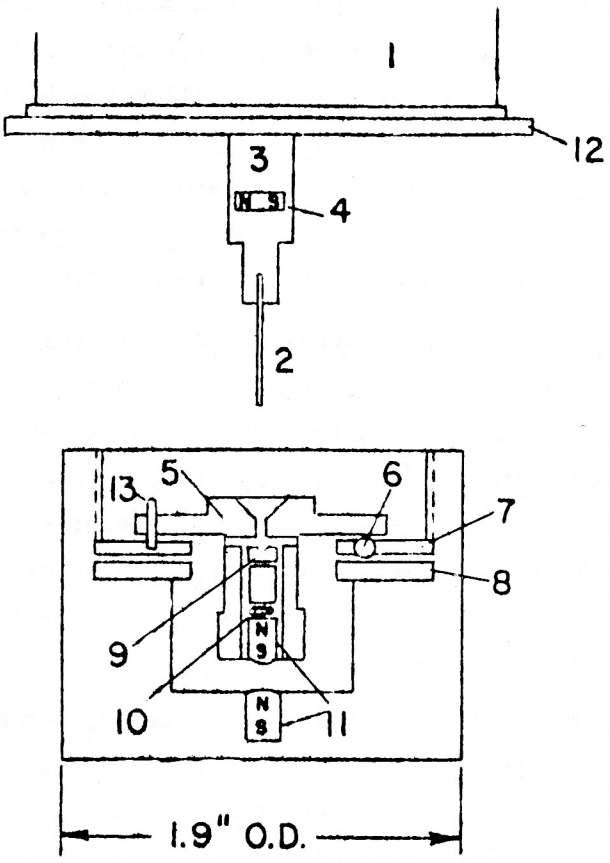

Figure 10 Lower Bearing Assembly

The bearing point, 9, is made of tungsten carbide into which a small groove has been ground to receive the needle tip, 2. To overcome vibrations, the bearing is made flexible by mounting the bearing point on a housing, 5, which is damped in oil. To regulate the damping forces, the clearance between the disk, 7, and the housing can be varied. The three steel balls, 6, standing on the steel plate, 8, allow the housing to move in a horizontal plane, but complete spinning of the housing is prevented by the three pins, 13. Vertical damping is provided by means of the spring, 10, which holds the bearing point in place. The magnets, 11, serve to center the housing with regard to the entire mounting block.

Most of the desirable attributes of good mechanical operation are inherent in this design. It is simple and cheap to build. The only lubrication system required is a pool of oil in the block cavity, no circulation system being needed. The power loss is small. The power loss in the bearing determined with a 1-pound rotor at 1500 rps (approximately full speed) was 1/2 watt. Finally, the bearing is quite reliable as evidenced by the fact that two units were in mechanical operation for about a year without appreciable wear.

Returning now to Figure 9, the upper bearing, B, consists of a hollow, cylindrical, permanent magnet mounted in a damped cardanic suspension, C, which attracts a steel tube, D, mounted rigidly on the rotor. There is no mechanical contact between these parts, nor does the magnet support a major portion of the weight of the rotor. Its purpose is to provide a damping and a centering force on the top axis.

The rotor is driven by an electric motor, M, the armature of which is a flattened steel plate, N, which is identical with part 12 of Figure 10, and is attached rigidly to the bottom of the rotor*. The field winding, M, is located in the vacuum and is fed by an alternator at a frequency near synchronous with the speed of the rotor.

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*NOTE: Mr. W. Dancy who worked with Dr. Zippe at Virginia informs me that in the original Zippe model the rotor itself acted as the armature and that Dancy suggested and helped design the arrangement used at Virginia and shown in the Figure.

The rotor is operated in a vacuum within a protective jacket. The pressure is initially reduced to a low value through an external mechanical pump. The pump connection is then sealed off by means of a vacuum valve and the pump no longer required. The vacuum in the jacket is subsequently maintained by means of the molecular pumping action caused by the screw-type grooves in the wall of the vacuum cylinder in combination with the rotating tube augmented by a second similar pump, L, located on the end of the rotor. The main pump grooves, K, can be seen in Figure 11.

Next, we turn our attention to the method in which the process gas is handled in the unit. Three tubes are inserted in the axial opening of the top bearing assembly. The feed tube, E, ends in the middle of the rotor, while the two other tubes, F and G, terminate at opposite ends of the rotor, and have their extremities bent into a hook-shaped curve in a plane perpendicular to the axis of rotation. These end sections, known as scoops, make use of the high impact pressure in the vicinity of the periphery for the transport of the heavy and light fractions. Simultaneously, one of the tubes, F, by means of the reduction of the angular velocity of the gas, induces the desired countercurrent flow, while the other, G, is shielded by a perforated plate, H, so as not to disturb the gas flow.

An exploded view of the rotor, showing a typical scoop system, is presented in the photograph of Figure 12. The top of the feed pipe system is also visible in Figure 13.

The centrifuge tube which was operated at Virginia by Dr. Zippe had the following operating characteristics:

1. Over-all length of rotor: 33.2 cm.
2. Effective length of rotor: 30.2 cm.
3. Inside diameter of rotor: 7.41 cm.
4. Peripheral speed of rotor: 3.5×10^4 cm/sec
5. Power consumption (measured by deceleration tests): 10 watts
6. Temperature: 350°K
7. Maximum theoretical spearative power: 4.75×10^{-5} gm/sec
8. Experimental separative power obtained was: 30 percent of the theoretical.

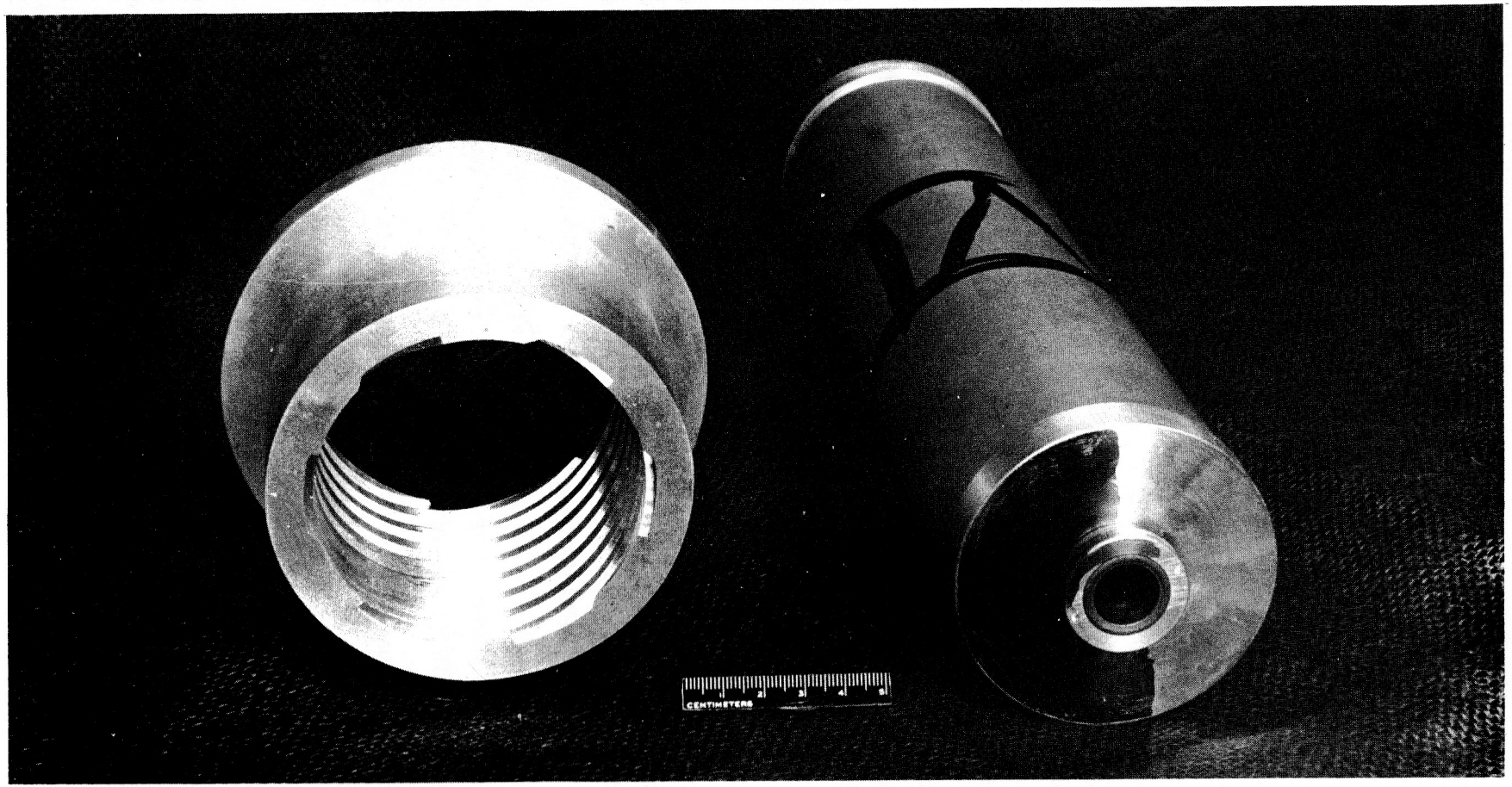

Figure 11 Molecular Pump and Rotor

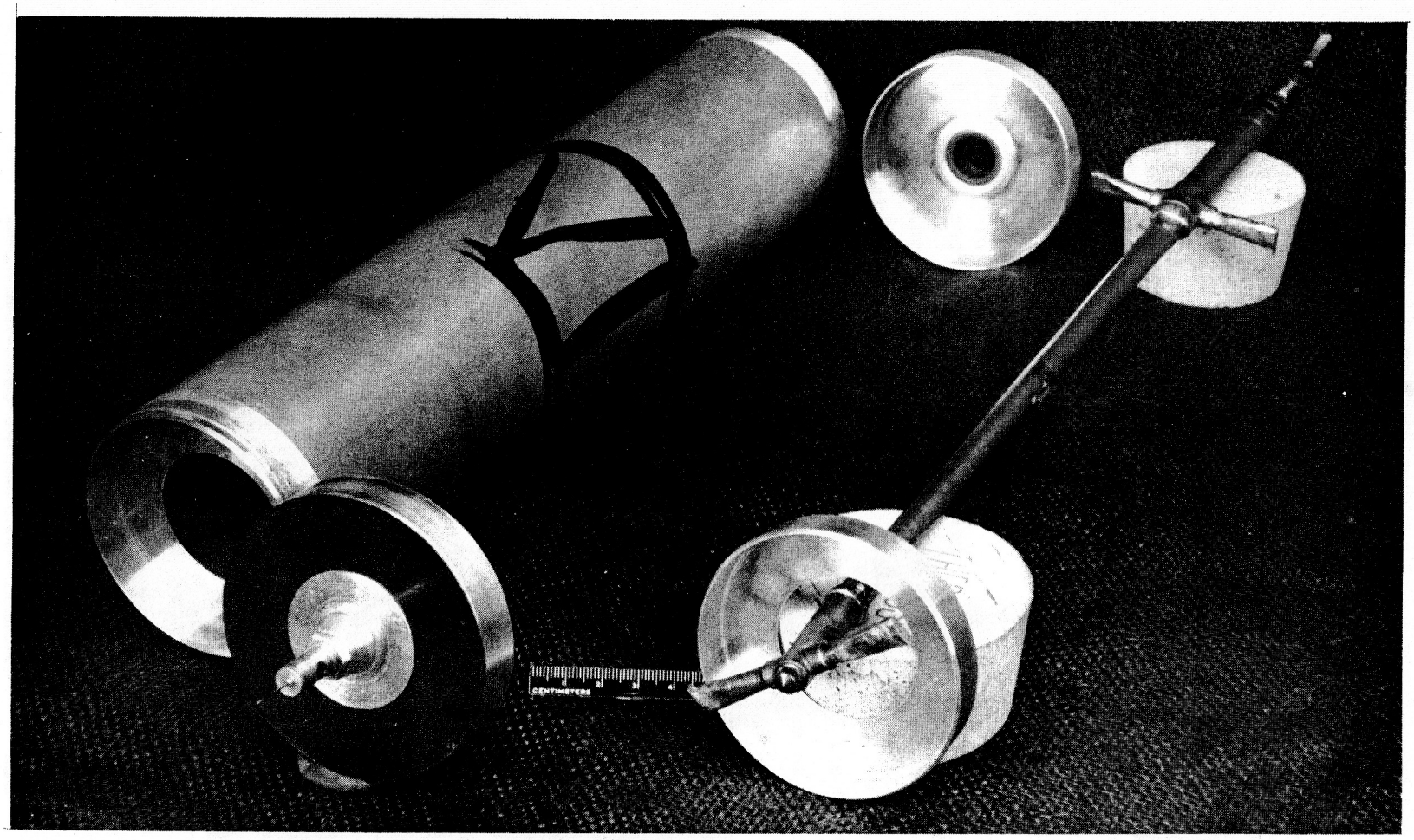

Figure 12 Scoop Assembly for Handling Gas Inside the Rotor and Parts of the Rotor

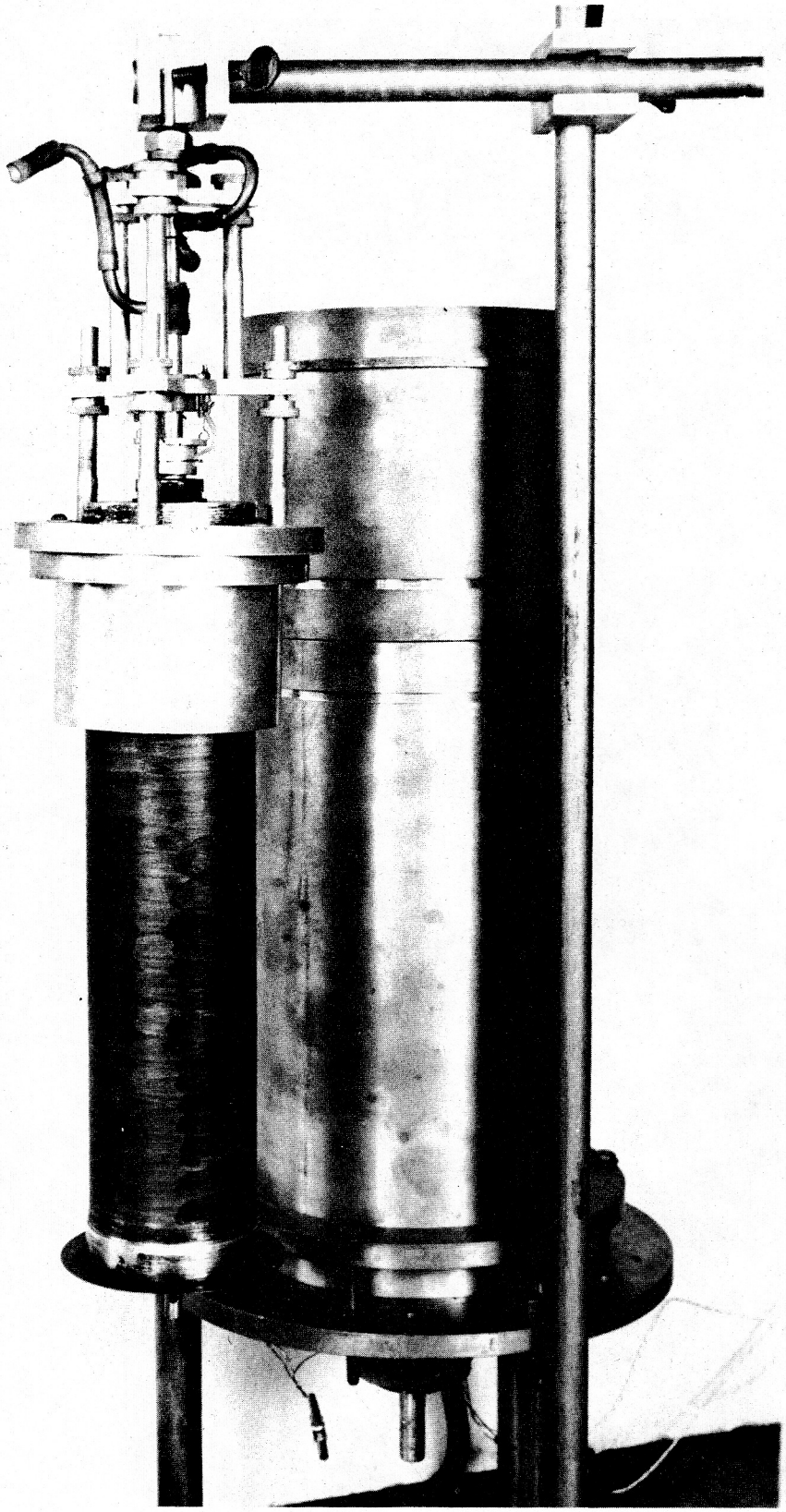

Figure 13 Rotor with Molecular Pump and Scoops Assembled

While Dr. Zippe was still at Virginia, Dr. Ralph Lowry, who was soon to follow Kuhlthau as Director when the latter became Associate Provost of the University, and Dr. Alwyn Lapsley joined the Virginia group and together they set about to assemble and utilize all of the advantages of their own, the Zippe and all other known techniques. As a result it soon became clear (to a number of optimists) that the gas centrifuge might possibly eventually become a competitor with the diffusion method. The progress made at Virginia soon persuaded the AEC to add a group at Oak Ridge and one at the AiResearch Company in California to the project. Also to shift the responsibility for the project from the Division of Research to the Production Division. The whole hearted cooperation of the three contractors together with the amazing developments in the method since that time is striking testimony not only to the wisdom of this action but to the administrative skill and devotion to excellence on the part of the directors and staffs of the three projects as well as the AEC staff that has had the AEC administrative responsibility.

Since the formation of the above three groups the progress has been so rapid and well documented that this is a logical point to terminate these rather rambling notes on the early work and leave it to others better qualified at some future time to give a more complete account of the gas centrifuge development in this country. Whether or not the gas centrifuge eventually will be adapted for isotope separation obviously will depend upon whether or not it is more economical in terms of dollars and/or energy than other competing methods. The fact that the fundamental separation unit is quasi-reversible, places the major responsibility for cost on the ingenuity of the experimenter and secondarily on nature's limitations. It is this type of challenge that makes work on the project not only interesting and rewarding but gives the worker a chance to exercise the full range of his genius. Under such circumstances there is reason for optimism concerning the ultimate success of the method.

This photograph was taken on the entrance to the Rouss Physics Laboratory soon after the termination of the Gas Centrifuge Project at the University of Virginia during World War II (~1944).

From Left to Right. First Row: L. B. Snoddy, L. G. Hoxton, A. Lipscomb, J. W. Beams, S. Coleman, Jr.; Second Row: J. S. Miller, E. P. Ney, S. J. Diggs, J. L. Young, III, F. L. Hereford, Jr.,: Third Row: J. Carter, P. Sommer, V. Chambers, F. Linke, F. Austin; Back Row: M. L. Randolph, W. A. Hogan, D. Payne, M. Irby, W. D. Whitehead, A. C. Lapsley, A. R. Kuhlthau, A. K. Mann.

Other collaborators during all or part of the project and not shown in the photograph were F. Bader, A. K. Cocke, F. T. Holmes, J. H. McQueen, C. S. Simmons, C. Henderson, B. C. Belden, C. A. Hutchison, Jr., J. W. Moore, and L. R. Quarles.

This photograph was taken at the Central Data Processing Facility.
From Left to Right: E. C. Evans, G. R. H. Geoghegan, P. R. Vanstrum,
E. F. Babelay, E. J. Grabowski, S. Whitley, A. P. Huber, N. L. Franklin,
H. M. Parker, S. A. Levin, D. M. Lang, E. C. W. Perryman, R. A. Lowry,
L. G. Williams, A. R. Kuhlthau, R. E. Leed, E. Von Halle.


CENTRAL DATA P CESSING FACILITY
OPERATED B UNION CARBIDE CORPORATION FOR THE U.S. ATOMIC ENERGY COMMISSION

Photograph taken at the University of Virginia (~1964) during a joint exchange of information meeting of the U. S. and British Workers in Gas Centrifugation. By this time the Project in the U. S. was taking shape and the three Contractors; University of Virginia, Union Carbide Corporation and the AiResearch Company were underway. The close and most effective cooperation of the three above Contractors together with the wise, efficient and understanding administration by Dr. Russel E. Leed and his colleagues in the A.E.C. has had an enormous influence for excellence in the work since that time.

(Shown from Left to Right) First Row: L. G. Williams, J. Stephenson, C. Allday, R. E. Leed, P. R. Vanstrum, S. Whitley, G. R. F. Geoghegan; Second Row: J. LaGraff, E. C. Evans, S. A. Levin, J. A. Friedericy, E. F. Babelay, H. Zimmer, F. D. Mikelson; Third Row: W. E. McMahon, W. J. Wilcox, Jr., A. C. Lapsley, E. M. Forsythe, R. A. Maurus, Jr., E. Von Halle; Fourth Row: L. Brenner, R. B. Nelson, H. M. Parker, G. I. Coulbourn, R. A. Lowry.

BIBLIOGRAPHY

1. F. W. Aston, Mass Spectra and Isotopes, Longmans, Green and Co., 2nd Ed. 1941.

2. F. A. Lindemann and F. W. Aston, Phil. Mag. [6] 37 523 (1919).

3. S. Chapman; Phil. Mag. [6] 38, 182 (1919) Phil. Trans. A217, 115 (1918).

4. R. S. Mulliken; J. Am. Chem. Soc. 44, 1033, 1729 (1922); 45 1592, (1923).

5. W. D. Harkins, J. Frank. Inst. 194 783 (1922).

6. J. Joly and J. H. J. Pool, Phil. Mag. [ ] 39 372 (1920); 41 818 (1921).

7. J. W. Beams and F. B. Haynes, Phys. Rev. 50 491 (1936).

8. J. W. Beams and A. V. Masket, Phys. Rev. 51 384 (1937).

9. J. W. Beams, J. Appl. Phys. 8 795, (1937); Phys. Rev. 55, 591 (1939).

10. J. W. Beams, Rev. Mod. Phys. 10 245, (1938).

11. J. W. Beams and C. Skarstom, Phys. Rev. 56 266 (1939).

12. H. E. Carr, Dissertation University of Virginia (1938).

13. R. F. Humphreys, Phys. Rev. 56 684 (1939).

14. K. Cohen, Theory of Isotope Separation, McGraw Hill Book Co., Inc. New York, (1951).

15. J. W. Beams, F. Linke and P. Sommer, Rev. Sci. 9 248 (1938).

16. J. W. Beams, Rev. Sci. Instr. 9, 413 (1938).

17. J. W. Beams and L. B. Snoddy, J. Chem. Phys. 5, 993 (1937).

18. J. W. Beams, J. Wash. Acad. Sciences 37 221, (1947).

19. H. C. Urey, Report on Progress in Physics, 6, 48 (1939).

20. H. C. Pollock and H. H. Kingdon, unpublished material (1939).

21. A. Bramley and K. Brewer, Science 92 427 (1940).

22. Martin and Kuhn, Z. Physik, Chem, A 189 219 (1940).

23. P. A. M. Dirac, British M. S. (1941).

24. J. W. Beams, A. C. Hagg and E. V. Murphree, Developments in Centrifuge Separation, T. I. D. - 5230 AEC Washington (1951).

APPENDIX I

Any process used for the separation of isotopes may be envisioned as one which removes the disorder produced by the mixing of the isotopes. In thermodynamic terms this reversible mixing (or separation) is given by the change in entrophy ΔS. To a first approximation the change in free energy $\Delta F = T\Delta S$ produced by the reversible mixing of the uranium isotopes in UF_6 is equivalent to that of the mixing of the same number of ideal gases at the same absolute temperature T where $S = R\Sigma N_k \ell n X_k$. R is the gas constant, N_k is the number of moles of the kth isotope and $X_k = N_k/\Sigma N$ or the mole fraction or concentration of the kth isotope in the mixture. However ΔF is not the work actually done by an isotope separator working at a constant rate because this depends upon the composition of the mixture at any time. For the case of a mixture of two isotopes with mole fractions of X and 1 - X respectively the change in entrophy ΔS produced by the isotope separator is proportional to $X(1 - X)$. This results from the fact that it is only upon the unlike molecules that the separator can do useful work, i.e., upon the fraction $X(1 - X)$. For any isotope separator processing L moles/sec the separative power δU can be written as

$$\delta U = L/R \left(\frac{\Delta S}{X(1 - X)}\right)$$

[See D. G. Avery and E. Davies Uranium Enrichment by Gas Centrifuge, Mills and Boon, Ltd., London (1973) for an excellent discussion of the concept of separative power]

For a separator producing P_{moles} of product of mole fraction X_p and W_{moles} of waste with mole fraction X_w from a feed of F_{moles} of mole fraction X_F

$$\Delta F = -RT\{X_r \ell n X_p + (1 - X_p)\ \ell n\ (1 - X_p)] + W\ [X_w \ell n X_w + (1 - X_w)\ \ell n(1 - X_w)]$$

$$-F[X_F \ell n X_F + (1 - X_F)\ \ell n\ (1 - X_F)]\}$$

while

$$\Delta U = P[2X_p - 1)\ \ell n \frac{X_p}{1 - X_p} + W[(2X_w - 1)\ \ell n \frac{X_w}{1 - X_w}] - F[(2X_F - 1)\ell n \frac{X_F}{1 - X_F}]$$

APPENDIX II

Early Magnetically Supported Electrically-Driven Vacuum Type Ultracentrifuges

Soon after the development of the air-driven, air-supported self-balancing vacuum type Ultracentrifuge at Virginia in 1934, work was started on magnetic methods of supporting most or all of the weight of the rotor as well as high speed electrical motors for spinning them. The task of making a suitable magnetic support "bearing" was much simplified because the rotor spun about a vertical axis. Consequently it was only necessary to attach a cylindrical rod or tube of ferromagnetic material (plunger) to the upper end of the rotor and attract it upward with an electromagnet or a permanent magnet. In some of the experiments at Virginia the magnetic lift on the rotor was adjusted to support almost but not quite all of the weight of the ultracentrifuge rotor [see Beams and Snoddy, Science 85, 185 (1937); Scarstrom and Beams, Rev. Sci. Instr. 11 398 (1940); and References 9, 10, 18]. In other experiments at Virginia the rotor was freely suspended inside of a vacuum chamber by a servo system [Holmes, Rev. Sci. Instr. 8, 444 (1937); Holmes and Beams, Nature 140, 30 (1937) and References 9, 10, 18]. Since the magnetic suspension field is approximately symmetrical around the axis of rotation of the rotor no flux is cut when the rotor spins and hence eddy currents are absent so that the friction due to the magnetic suspension per se is vanishingly small, i.e., the Q of these bearings often exceeds 10^8.

Various kinds of electrical motors were developed for successfully spinning the ultracentrifuge rotors up to their bursting speeds [see above references]. Rotor speeds of 1.5×10^6 rev. per sec and centrifugal fields of 10^9 times that of gravity have been obtained. In all cases the limiting factor in the above is the strenght to density ratio of the rotor. With specially shaped steel rotors peripheral speeds of between 1000 meters/sec and 1500 meters/sec were obtained. Recently, Professor Philip Moon of the University of Birmingham in England has reported reaching a peripheral speed of 2,200 meters/sec. His rotor consisted of a specially tapered rod made of carbon filaments attached to ferromagnetic material located at the center.

It should be noted that it was found in the early experiments that by damping the magnetic support the rotor vibrations could be effectively damped.

APPENDIX III

The maximum thermodynamically stable temperature gradient which can be maintained in a gas in a centrifugal field is the adiabatic gradient. [See W. J. Humphreys, Physics of the Air, McGraw-Hill, New York, 1929 and Reference 9]. This can be evaluated as follows: for an adiabatic process. (See for example, Zemansky, Heat and Thermodynamics, 3rd ed., Page 254).

$$\left(\frac{dT}{dp}\right)_s = \frac{T}{C}\left(\frac{ds}{dT}\right)_p \qquad (1)$$

where p is the pressure, T the Kelvin Temperature, V the volume, S the entrophy and C_p the specific heat at constant pressure. For an ideal gas in the centrifuge by substitution, the radial temperature gradient becomes

$$\frac{dT}{dr} = \frac{M\ r\ w^2}{C_p} \qquad (2)$$

where M is the mean molecular weight of the gas, C_p is the molar specific heat which is practically the same for all of the isotopic molecules of the gas. For example, in a centrifugal field of 10^5 times gravity g in air at 20°C $\frac{dT}{dr} \approx 10°C/cm$ and in liquid water at 20°C, $\frac{dT}{dr} \approx 0.18°C/cm$.

In condensable gases the saturation curve is given by $d\ \ell n\ p/dT = \Delta S/RT$ where ΔS is the entrophy of vaporization. Since $d(\ell n)p = \frac{M}{RT}\ r\ w^2\ dr$ then

$$\left.\frac{dT}{dr}\right]_{Sat} = \frac{M\ r\ w^2}{\Delta S} \qquad (3)$$

In general $C_p < \Delta S$ so that if at any radius of a centrifuge the saturation point is reached the maximum stable temperature gradient possible at smaller radii is that of the saturated atmosphere (equation 3) above. Scarstrom and Cohen have suggested to me that some of the experimental values larger than the equilibrium isothermal α shown in Figure 3 may be due to this phenomenon.