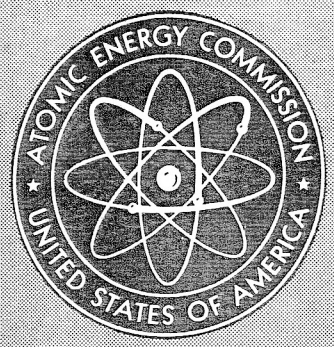

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STABLE ISOTOPE SEPARATION

DEVELOPMENTS IN THE CENTRIFUGE SEPARATION PROJECT

By
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DEVELOPMENTS IN THE CENTRIFUGE SEPARATION PROJECT

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VOLUME PREFACE

Early work on the use of gaseous centrifuges for separating uranium isotopes was initiated by Dr. J. W. Beams at the University of Virginia and Dr. Harold C. Urey at Columbia University. This work was covered by OSRD contracts. Dr. Karl Cohen at Columbia University carried out theoretical studies on the separation of isotopes in a gaseous centrifuge and also considered their use in a cascade. With the formation of the Planning Board it was decided to transfer the responsibility for gaseous centrifuge development to the Standard Oil Development Co. This was done in January 1942.

Shortly after its formation, the Planning Board decided to expand the work on gaseous centrifuge development. Responsibilities for various phases of this development were given to different organizations. Dr. Beams was to carry out separation experiments on a machine built at the University of Virginia. The research laboratory at Westinghouse was given the responsibility of developing a commercial type of gaseous centrifuge suitable for long periods of operation. The Standard Oil Development Co. was to install a number of Westinghouse machines when available and operate them as a cascade. This was essentially a pilot-plant operation. Later plans were changed owing to the time required to develop a satisfactory centrifuge, and the program to be carried out by the Standard Oil Development Co. then involved only the operation of one centrifuge.

In addition to experimental work as outlined, engineering studies were carried out to determine the plant facilities required to produce 1 kg of U^{235} by the gaseous centrifuge operation. These studies indicated the desirability of utilizing centrifuges with longer bowls. The original plan had been based on using a centrifuge with a 36-in. bowl, and a second development was started on centrifuges having a bowl length of 126 in.

The results obtained in the gaseous centrifuge development are outlined in various summary reports submitted by the organizations involved, namely, the University of Virginia, the research laboratory at Westinghouse, and the Standard Oil Development Co. In addition Dr. Karl Cohen (who later transferred to the Standard Oil Development Co.) has prepared a theoretical discussion of separation in gaseous centrifuges and their use as part of a cascade. Dr. W. I. Thompson assisted Dr. Cohen in these later studies.

Dr. Beams and his group at the University of Virginia successfully demonstrated the possibilities of isotope separation in a machine operating with continuous feed. In this work, separation was carried out with both concurrent and countercurrent flow of gases. It was found that the countercurrent flow gave a better separation factor. This early work was carried out on a centrifuge with a bowl length of 36 in. Later the University of Virginia group built a machine with a bowl length of 126 in. and demonstrated its successful mechanical operation. Runs were also made with continuous feed to demonstrate the use of this larger machine for separation of the isotopes. This work showed that the separation efficiency per unit of bowl length was the same for the long machine as for the short machine.

Westinghouse carried out the successful mechanical development of a centrifuge with a bowl length of 36 in. and built a number of these machines. Westinghouse was particularly concerned with the development of a machine suitable for a continuous operation over a long period of time. Later Westinghouse started development of a centrifuge with a bowl length of 126 in. This development was nearing a successful conclusion when it was decided to discontinue the gaseous centrifuge project.

The Standard Oil Development Co. erected pilot-plant facilities for operating a number of centrifuges in a cascade. As pointed out earlier, this program was later modified to operate only one machine which was built by Westinghouse. Very satisfactory operation was obtained on the one-machine basis, continuous operation being maintained for periods as long as 69 days. From the experience obtained it was felt that it would be feasible to operate a large number of machines such as would be required for a full-scale plant and that these machines could be run for long periods of time. In the pilot-plant operation of the centrifuge it was demonstrated that a separation factor of 80 per cent of theoretical for a countercurrent-gas-flow machine could be obtained and maintained for long periods of time.

Original engineering studies were based on the use of a centrifuge with a 36-in. bowl. Owing to the advantages in reduction of number of machines, later studies were based on the use of a centrifuge with a bowl length of 126 in. Approximate estimates and engineering studies were made for a large plant to produce 1 kg of U^{235} from normal uranium per calendar day, the product having 90 per cent purity of U^{235}. This plant was so designed as to recover about half the U^{235} present in the original uranium. These engineering studies indicated that about 30,000 long-bowl centrifuges would be required and that the power needed to run these machines and auxiliary equipment would be around 165,000 kw. The cost of the plant complete with necessary

facilities was roughly estimated at 218 million dollars. At the time the actual work on the gaseous centrifuge project was terminated early in 1944, it was felt that the gaseous centrifuge and gaseous diffusion projects had quite similar problems that would have to be overcome and that their cost was of the same general order; it was felt that, if there was any difference in cost, the gaseous centrifuges would be somewhat cheaper. The two projects reached a point where it was necessary to make a decision as to which should be continued. At the time, it was felt that the gaseous diffusion project was further developed, and for this reason it was decided to terminate the gaseous centrifuge project. At the time of termination it was felt that considerable advance in the gaseous centrifuge technique could be made by using steel bowls for the centrifuges in place of aluminum bowls since the steel bowls would allow higher spinning speeds with consequent more rapid separation.

The results of the work undertaken at the University of Virginia, the research laboratory at Westinghouse, and the Standard Oil Development Co. are reported in Parts I, II, and III, respectively, of this volume.

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Part I

CENTRIFUGE SEPARATION METHODS

By J. W. Beams

INTRODUCTION TO PART I

The possibility of separating isotopes by centrifuging was first suggested by Lindemann and Aston[1,2] in 1919. They also worked out the equilibrium theory for the separation in an ideal gas and in an ideal incompressible liquid. For the case of an ideal gas in a centrifuge under equilibrium conditions

$$dp = \rho\omega^2 \, r \, dr$$

where p = pressure
ρ = density
r = centrifuge radius
ω = angular velocity
dp = pressure change along the radius at r

Since $p = \rho RT/M$ from the gas law,

$$\frac{d\rho}{\rho} = \frac{M\omega^2}{RT} \, r \, dr$$

which upon integration gives

$$\rho = \rho_0 \exp\left(\frac{M\omega^2 r^2}{2RT}\right) = \rho_0 \exp\left(\frac{Mv^2}{2RT}\right) \qquad (1)$$

where M = molecular weight of the gas
T = absolute temperature
ρ_0 = density of the gas at the centrifuge axis
ρ = density at a distance r from the axis
v = peripheral velocity at r

If the gas contains two isotopes of masses M_1 and M_2 and if K_0 is the ratio of the two quantities present at the axis, the ratio at the periphery K is

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

The separation factor α was defined as

$$\alpha = \frac{K}{K_0} = \frac{\frac{(X_1)_p}{(X_2)_p}}{\frac{(X_1)_0}{(X_2)_0}} \qquad (3)$$

where $(X_1)_0$, $(X_1)_p$ = mole fractions of the first isotope at the axis and periphery, respectively
$(X_2)_0$, $(X_2)_p$ = corresponding quantities of the second isotope

The separation factor α depends upon the difference of the masses of the two isotopes rather than upon their absolute values and upon the square of the peripheral speed divided by the absolute temperature in the exponent of e. Following the cited work of Lindemann and Aston, the theory was critically discussed and extended by Mulliken,[3] Chapman,[4] and Harkins.[5] Mulliken showed that the theoretical value of α given in Eq. 3 is independent of the state of combination of the elements provided the isotopes have identical properties. He also investigated centrifugal separation theoretically in ideal liquid isotopic mixtures and suggested the so-called "evaporative-centrifuge" method which consists in drawing out vapor from the axis of a hollow spinning rotor containing the liquid in its periphery. In this method the separation takes place in the vapor as it diffuses from the periphery (where the liquid slowly evaporates) through the centrifugal field to the axis where the vapor is withdrawn. Mulliken showed theoretically that Eq. 3 holds if the process is allowed to take place slowly enough to permit equilibrium to be practically established and if the vapor obeys the ideal gas laws. He also showed that the change in molecular weight of a mixture of two isotopes, ΔM, between the residue remaining in the rotor and the original material is given approximately by

$$\Delta M = \frac{(M_2 - M_1)^2 X_1 X_2 \omega^2 r^2}{2RT} \ln C \qquad (4)$$

where C is the so-called "cut" or the ratio of the total amount of the initial material to the amount of residue remaining in the centrifuge.

Encouraged by Mulliken's theory, which showed that it should be possible to obtain considerable separation of the isotopes, a number of

workers[3,6,7] attempted to obtain the separation of different isotopes in specially constructed centrifuges. Unfortunately, each of these early attempts proved to be unsuccessful, probably because of convection, and the method was abandoned as impracticable. However, with the development of the convection-free vacuum-type ultracentrifuge[8] in 1934, the method was tried again, and the results were successful.[9-12] In most of these first successful experiments, the evaporative-centrifuge method was employed to concentrate the Cl^{37} and Cl^{35} isotopes of chlorine in CCl_4. The bromine isotopes were concentrated also. It was found that considerable separation could be obtained and that the results were in good agreement with the theory when equilibrium conditions were approximately obtained in the centrifuge.[12] The theory was tested further with a special type of centrifuge at dry-ice temperature with different substances at different rotor speeds, and it was found to hold.[13] Humphreys[14] also made use of the same type centrifuge as used in the above experiments[11] to concentrate the isotopes of bromine. He further modified the theory to take into account the case in the evaporative-centrifuge method in which the vapor is drawn off so rapidly that equilibrium conditions are not obtained. Humphreys showed that

$$\alpha = \frac{K}{K_0} = \exp \frac{g(M_2 - M_1)\omega^2 r^2}{2RT} \qquad (5)$$

where

$$g = \frac{2q}{2q + Q}$$

and

$$q = \frac{2\pi h \rho D}{M}$$

where Q = rate of drawing out the vapor at the axis
D = coefficient of diffusion
h = height of the rotor
M = molecular weight

Wilson[15] and Bramley[16] further developed the theory, but it remained for H. C. Urey, Karl Cohen, and their collaborators to work out the general mathematical theory and to indicate theoretically the most efficient way to go about the separation of isotopes by centrifuging.

In addition to the evaporative-centrifuge method, several other schemes were tried. It was clear from elementary theory that the rate at which separation could take place in a centrifuge rotor is proportional to the length or depth of the rotor. Accordingly methods of spinning tubular rotors were developed[11] and used not only in the evaporative-centrifuge method[12] but in several other methods as well.

The so-called "flow-through," "cream-separator," or "concurrent" method utilized the tubular vacuum-type centrifuge.[11] Ethyl chloride vapor at about 300°K flowed through the hollow shaft into the top of an alloy-steel tubular rotor 3 in. I.D. and 14 in. long (containing end baffles) which was spinning at 1060 rps. Two equal fractions were collected at the lower end of the spinning tube, one near the axis of rotation and the other near the periphery. The observed separation factor between the two samples was $\alpha = 1.025$ with a continuous rate of flow of 2 g/min (i.e., 1 g per sample per minute).[18] A large number of observations[18] were also made with the same apparatus on the separation of N_2-O_2 and N_2-CO_2 gaseous mixtures with various rates of flow up to 2 liters per minute. The results were later found to be in agreement with Cohen's theory.[17]

The distillation, or cascade, method first suggested by Urey[19] was also tried[20] using the same type of tubular rotor employed in the above methods.[11,12,18] About 65 cc of liquid CCl_4 was placed in the 3-in.-I.D. and 14-in.-long centrifuge tubular rotor. The tube, which contained radial baffles, was spun to 1060 rps and then evacuated and sealed. The upper end of the spinning tube was surrounded by a copper coil carrying cooling liquid, and H_2 at about 3 mm Hg pressure was introduced into the vacuum chamber surrounding the rotor to conduct heat to it. This arrangement made the liquid evaporate in the lower warm part of the rotor and condense in the upper cool part giving a vapor circulation upward and liquid flow down the periphery. Small samples were collected at the top and bottom of the rotor after 15 hr of centrifuging and were analyzed in the mass spectrometer. Although the results were erratic in some cases, the separations observed were slightly larger than could be accounted for by a simple centrifuging process, showing that evidently some cascading was taking place.

In some further experiments at the University of Virginia,[21] a duralumin tube 50 cm long and 1⅛ in. I.D., spinning at 2300 rps in air at atmospheric pressure, was used. The tube was first evacuated, and then from 5 to 6 cc of CCl_4 was distilled in it. The tube was then sealed and spun to full speed. Tap water at about 20°C was sprayed on the top of the tube to cool it, and at the same time the lower end of the tube was heated by air friction. After the tube had spun for about 2 hr a small sample (about 1 cc) of CCl_4 was pumped out through a

hollow shaft and then condensed. In some of the experiments, the separation obtained was between four and five times that possible in a single centrifuging. In this way the Urey cascading process was shown to exist. However, in actual practice, this method seemed neither so easy to perform nor so promising for large-scale separation of isotopes as the other methods.

Another method of producing circulation in the spinning tubular rotor was tried[11,22] when the material centrifuged was either in the gaseous or in the liquid state. A small rod of resistance material was placed along the axis of the spinning tube and was heated by an electrical current passing through the shafts at the top and bottom. The decrease in density as a result of the increase in temperature near the axis should cause the gas or liquid to rise along the axis and move downward along the cooler rotor wall in a manner similar to the Clusius and Dickel thermal diffusion experiment. This circulation is caused primarily by the gravitational field of the earth and is opposed by the stabilizing influence of the centrifugal field (to be discussed later) where the gas moves radially at the upper and lower ends of the rotor. However, the experimental results were not conclusive. Some difficulty was encountered in supporting the heating rod, or tube, because of its natural vibration. This experimental difficulty was overcome later, but the experiment was dropped, since it seemed at the time to be inferior to other centrifuge methods for production purposes. However, the method should be investigated again experimentally, making use of new experimental techniques.

Somewhat similar experiments were proposed independently by Bramley and Brewer[23] and by Martin and Kuhn.[24] They proposed to produce the proper circulation in the centrifuge by thermal means.

Experiments were also tried[22] in which a "mechanical elevator," or screw, was placed along the axis of a tubular centrifuge to carry the gas or liquid up along the axis of the centrifuge. The material would then flow back down along the wall because of the centrifugal force. Unfortunately, the experiments were never completed.

Pollock and Kingdon[25] used a circulating evaporative-centrifuge method for the concentration of the tin isotopes in $SnCl_4$, but the method was not developed further. In 1940 Gunn[26] suggested a most ingenious method of isotopic separation in which the sedimentation produced by a high centrifugal field was opposed by the mobility of the ions under an electrical field. Gunn also worked out a detailed theory from which he could predict the separation of the U^{235} and U^{238} isotopes and found that samples with large separation should be obtained. The method was tried experimentally at the University of Virginia by Williams[27] and Fox, and a few experiments were carried

through. However, the experiments were dropped to allow greater concentration on other methods.

Immediately after the announcement of uranium fission by neutrons in March 1939, the writer and L. B. Snoddy, at the University of Virginia, became interested in the purification of U^{235} and U^{238} by centrifuging in order to determine the respective part played by each isotope in the process of fission. It will be recalled that the equilibrium separation factor in the centrifuge method depended upon the difference in the masses of the isotopes in the exponent of e rather than upon their absolute values. Also this factor was independent of the state of combination of the element. Furthermore, the separation factor did not decrease with an increase of concentration of the rare isotope. The method seemed well suited to the purification of the heavy U^{235} and U^{238} isotopes.

A study of the literature on the chemical and physical properties of uranium showed that, of the known compounds of uranium, uranium hexafluoride possessed the physical properties which best adapted it for use in the centrifuging process. It was a liquid at 69°C with a vapor pressure of 2 atm. Therefore it could be used in any of the centrifuge methods which had been proposed. On the other hand, UF_6 was not available commercially, and as far as could be found out at that time, only a small amount of the material had ever been made. The literature further revealed that the UF_6 attacked water violently as well as almost all metals except gold and nickel. In addition it was difficult to work with and extremely poisonous. Also the process of making UF_6 was not clear (at least to this research team), and the estimated yields were almost negligibly small. Therefore it was necessary to undertake the production of UF_6 if the work was to go forward. Fortunately, shortly after preliminary work was under way, Gunn, of the Naval Research Laboratory, who was actively interested in the problem because of its military possibilities, agreed to try to procure enough UF_6 to make a test experiment possible. The active help and encouragement that Gunn and the Naval Research Laboratory gave during the entire course of the work on this problem in such vital things as the procurement of UF_6, rotor materials, etc., as well as a generous grant early in the work, is indeed most gratefully acknowledged.

It soon became apparent that the procurement of UF_6 in quantities sufficient for the experiment would take considerable time (it actually took about 1 year). Consequently, Snoddy undertook to make a small amount of the UF_6 at the University of Virginia in order to study its physical and chemical properties. From these early studies he concluded that, if the UF_6 was pure, it not only did not attack gold and

nickel, but it did not appreciably attack, at least at room temperature, the stainless steel and duralumin which were to be used in the centrifuge rotors and rotor shafts. The metals sometimes were fluorinated with F_2 gas before using.

In the meantime, while waiting for enough UF_6 to start the experiments, an effort was made to find, if possible, a better centrifuge method by experimenting with the separation of isotopes of other available materials. Urey had suggested several cascade methods using long tubular rotors. Also previous experiments with the separation of the chlorine isotopes in CCl_4 and the separation of the components of N_2-CO_2 gaseous mixtures had demonstrated the advantage of the long tubular rotor. Therefore work was begun on the problem of spinning tubular rotors longer than any used in the previous work.[11]

The task of spinning tubular rotors many times longer than their diameter to high speed presented several difficult problems. First, such rotors pass through "critical" frequencies before they reach the necessary operating rotor speed. At the first critical speed they vibrate like the fundamental of a vibrating rod, and at the second critical they vibrate like the first harmonic, etc., depending upon how they are mounted. In previous work[11,12] it had been possible to spin alloy-steel tubular rotors 14 in. long and 3 in. I.D. well above their first critical frequency. It had been found that in order to spin them through this first critical frequency special damping in the bearings was required, but when the speed exceeded this critical frequency, the tube spun very stably and smoothly for long periods, over 700 hr, without any signs of fatigue.[12] Tubes having a much greater length-to-diameter ratio must pass through a number of these critical frequencies in order to reach the required rotor speed; however, it was not at all certain that such tubes could be spun through these frequencies. Also it was a debated question whether the tubes would spin stably and without fatigue above the second or higher critical frequencies. About this time F. T. Holmes joined Snoddy and the writer on this problem. It was subsequently found that the long tubes could be spun through as many critical frequencies as were required and that they spun stably at these high speeds. However, considerable research was required on damper bearing, and a description of the schemes used will be given later.

In April 1940, through the kind interest of M. A. Tuve and V. Bush, a grant-in-aid was generously offered by the Carnegie Institution of Washington. This much-needed financial aid allowed an expansion of staff and further intensification of the work on the problem. As a matter of fact, this grant together with a contract with the Naval Research Laboratory made it possible to direct the major effort of the physics staff and graduate students at the University of Virginia into

this problem in the early summer of 1940. In February 1940 Gunn furnished approximately 6 g of UF_6 which was the first sample of this material obtained that was large enough to try out in the centrifuge. The evaporative-centrifuge method was used, and the theoretical separation factor was 1.08 or an 8 per cent change in the isotopic ratio. The samples were successfully centrifuged without appreciable loss or deterioration and were sealed in glass capsules. At that time, February 1940, no means were available for analyzing the samples, and they were sent away for analysis; but apparently they were never analyzed. However, these experiments demonstrated the feasibility of handling the UF_6 in the centrifuge. About this time Urey started active work on the centrifuge method at Columbia University, and his group worked in close cooperation with the writers. C. Skarstrom went to Columbia University from the University of Virginia to assist in this program. Urey had suggested several methods of attack on the problem, and his group, led by K. Cohen, worked out the general centrifuge separation theory and subsequently came to the conclusion that the refluxing counterflow method, using long tubular centrifuges, was the most efficient for large-scale separation.

By the middle of 1940, tubes of various sizes, with lengths many times their diameters, had been spun up to their bursting speeds. Consequently a systematic set of experiments was undertaken to test the efficiencies of the various methods. These methods included the evaporative method; the distillation method; the flow-through, or concurrent, method; and the countercurrent method.

In these methods, compounds of chlorine and bromine and N_2-H_2 and N_2-CO_2 gaseous mixtures were used for separation because of the scarcity of UF_6.

The method using evaporative centrifuge was used to check the centrifuging theory carefully and to make several concentrated samples. In December 1941, two samples of 1 g each were made whose difference in isotopic ratios as measured by A. O. Nier was about 40 per cent. These samples were turned over to Briggs for experimentation.

After the early tests, the distillation method was abandoned as inferior to the other methods, although it was found to give cascading effects.

Data in good agreement with Cohen's theory were also obtained with the flow-through, or concurrent, method using N_2 and CO_2 in 1939 and UF_6 in 1942.

In 1942 data in good agreement with the Cohen theory were obtained with the countercurrent method using N_2 and CO_2. This was the method finally decided upon for the pilot plant; consequently, in 1942 tests

using UF_6 were made with a 4-in.-I.D. tube 3 ft long and with a tube 11 ft long and 8.33 in. O.D.

During the latter part of the work, an effort was made to get the kind of data that would permit an estimate of the performance of a large number of centrifuges in a cascade or in a production plant. This work was carried out in close cooperation with E. V. Murphree and his group at the Standard Oil Development Company. Through the kindness of Murphree, B. C. Belden, of his group, came to the University of Virginia and worked until the experiments were practically completed. Belden's help was invaluable and contributed in a large measure to the success of the experiments. Murphree worked out, independently, a theory for the flow-through experiments and made many valuable suggestions during the course of the work. W. I. Thompson of Murphree's group also gave a great deal of help and made many useful suggestions, especially as to the data necessary for calculating the performance of the cascade from the performance of a single machine.

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Excellent theoretical work which was of utmost importance in guiding the course of our experimental work was given by Urey, Cohen, Skarstrom, and others who were at Columbia University, and it is a pleasure to acknowledge our appreciation to them for their generous help during the whole course of the experiments. Also it is indeed a pleasure to mention the help given by L. J. Briggs who, as chairman of the committee of the NDRC and OSRD, took an active personal interest in the experiments. Briggs' committee took over the support of the work the latter part of 1940, and from that time on, Briggs, serving as our chief adviser, not only saw to it that necessary materials were obtained but personally made many valuable suggestions.

We also wish to express appreciation to Murphree, Belden, Thompson, Kuhl, Deutsch, and others of the Standard Oil Development Co. and to Chubb, Hagg, and others of the Westinghouse Research Laboratories for their most valuable help and cooperation. The work undertaken at the University of Virginia was under the general direction of Beams and Snoddy. Photography of the original drawings was completed by A. C. Lapsley, and the original manuscript was typed by Annie G. Lipscomb.