

Gas Centrifuge Theory and Development: A Review of U.S. Programs

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This article gives a historical and technical review of the U.S. gas centrifuge efforts between 1934 and 1985. The first section tells of how the United States initially led in centrifuge design, only to abandon the still-immature technology in the midst of the Manhattan Project. While interest in the technology continued in Europe, the United States decided that centrifuges were not a viable alternative to existing gaseous-diffusion plants. Five years later, U.S. spies learned of important Soviet achievements in centrifuge design, which, it appeared, might place centrifuges in direct competition with gaseous diffusion. When combined with European enthusiasm for the centrifuge, the United States faced the prospect of losing its control on the enrichment market in the West. Accordingly, the United States organized a program to rebuild its centrifuge dominance. Over the next 25 years, it led the world in machine performance but ultimately failed to commercialize its designs for a variety of reasons. The basic specifications and performance data for several of the U.S. centrifuges designed and tested during these years are reported here for the first time in the public domain.

The United States also made a number of contributions in the theoretical domain that were openly shared over the course of the U.S. program. Most of these fell into the field of fluid dynamics, developed to guide the design and optimization of centrifuges. The most important elements, especially those relating to the calculation of separative power, are described in Theoretical Developments. These are of interest to policy-makers because they can be used to predict the rate at which centrifuges produce fissile material for nuclear weapons, an important factor for nuclear nonproliferation. They are also used in calculations related to multi-isotope separation, which is important for nuclear forensics. Examples of both kinds of policy analysis are given in articles by Glaser¹ and Wood² appearing in an earlier issue of *Science and Global Security*. The U.S. theory is used here to derive a simple equation that enables policy-makers to make good estimates for the separative power of a real centrifuge by knowing only its length and speed.

Received 15 April 2008; accepted 23 June 2008.

The author thanks Houston G. Wood and A. Robert Kuhlthau for their assistance with centrifuge flow theory and documenting the history of the U.S. centrifuge program. Address correspondence to R. Scott Kemp, Program on Science & Global Security, 221 Nassau St., 2nd Floor, Princeton University, Princeton, NJ 08542, USA. E-mail: rskemp@princeton.edu

MACHINE DEVELOPMENT IN THE UNITED STATES

Early History

The use of centrifuges for isotope separation was first proposed by British scientists Lindeman and Aston in 1919, shortly after the existence of isotopes had been experimentally confirmed. In the following years, primitive centrifuges were tested by a number of groups, but their experiments proved unsuccessful; excessive heat transfer across the wall of the centrifuge rotor created convective currents that mixed the gas, negating any separative effect. In 1934, Jesse Beams at the University of Virginia had the insight to thermally isolate the centrifuge rotor by placing it inside a vacuum, and in doing so successfully separated the isotopes of chlorine. Beams' early designs consisted of tubular rotors connected to co-axial driveshafts mounted in thrust bearings outside the vacuum casing. A heavy-oil gland sealed the gap between the driveshaft and the casing. The design was basically functional, but friction in the bearing and around the oil gland consumed kilowatts of energy, thus heating the gas, lowering the separative power, raising the operating cost, and shortening the life of the device.

When the Manhattan Project began, centrifuges of this kind were considered as a candidate technology for concentrating uranium-235 for the first nuclear weapon. Beams was asked to oversee the centrifuge project and led a design team at the University of Virginia. The Westinghouse Research Laboratory was charged with building the first production machines, and testing was done at the Standard Oil Development Company in Bayway, New Jersey. The team at the University of Virginia designed a supercritical machine, 7.35 in diameter and 136 in long, which operated at 270 m/s in a four-stream mode. Westinghouse built a slightly smaller version, 7.2 in diameter by 132 in long, as well as a subcritical machine, 7.2 in diameter by 42 in long. Separation tests began with the subcritical unit in August 1943 and continued for 93 of 99 days before the oil gland developed a leak and caused the centrifuge to crash at 215 m/s in December of 1943. The program was terminated by enrichment-project manager Harold Urey a few weeks later in favor of gaseous diffusion.

After WWII, various groups around the world worked to reduce the friction problem in an effort to make centrifuges competitive with gaseous diffusion. Progress with thrust bearings was made in Germany and at the Franklin Institute in Philadelphia, and Beams briefly experimented with magnetic suspension as an alternative. However, the U.S. Atomic Energy Commission (AEC) ultimately decided not to pursue centrifuge technology. In December 1951, the Committee on Isotope Separation of the Division of Research strongly advised against a post-war centrifuge revival, noting that centrifuges could not compete economically with existing gaseous diffusion technology. That would change in 1953 when German groups led by Wilhelm Groth and Konrad

Beyerle, and a Dutch group led by Jacob Kistemaker, began asserting that their centrifuges would soon be more economical than American-style diffusion. Fearing a loss of their uranium-enrichment monopoly, the U.S. Atomic Energy Commission decided to revive centrifuge research in September 1954. But the revival was desultory: No machines were built because the AEC was confident that it would have full access to German technology, which was at that time the most advanced in the West. The most significant task done at home was a study of novel, high-strength materials and supercritical rotors commissioned to Arthur R. Kuhlthau at the University of Virginia in August 1956.

Zippe at the University of Virginia

In the summer of 1956, the Soviet Union began repatriating captive German scientists who had been working on uranium enrichment at Institutes A and G at Sukhumi on the Black Sea. Oswald Francis (“Mike”) Schuette of the U.S. Office of Naval Intelligence had the task of interrogating the returning Germans. By chance, one of his subjects was the Austrian scientist Gernot Zippe, who wound up in Germany only because he could not afford rail fare back to his native Austria. Zippe had become a flight instructor for the Luftwaffe after he finished his Ph.D. He was captured by Soviet forces and sent to the Krasnogorsk detention camp, but when the Soviets learned of his technical background he was transferred to Institute A, where he was put in charge of mechanical developments for the Soviet centrifuge program.

Schuette’s report of Zippe’s activities attracted the interest of the Intelligence Division of the U.S. Atomic Energy Commission. They arranged to bring Zippe to the United States in 1957 under a false passport and the assumed name of “Dr. Schubert.” AEC scientists and intelligence officials interrogated Zippe in an all-day session at the Shoreham Hotel in Washington, D.C. This was followed by a more collegial debriefing several days later at the University of Virginia. Zippe revealed that he and the Soviets had worked out a completely novel design that did away with the thrust bearings and oil glands that had been so problematic in the U.S. centrifuge. The AEC arranged to have Zippe return to the United States on contract to the University of Virginia, where he would replicate the Soviet machine in exchange for a \$10,000 salary. In August 1958, Zippe returned to Virginia, and less than a year later he produced a working replica of the Soviet machine.

The U.S. Program Expanded

Karl Cohen, whose theoretical contributions appear later in this article, was asked to assess Zippe’s work for the Atomic Energy Commission. He was so impressed that he advised the commission to expand its centrifuge efforts in the direction of the Soviet design. By April 1960 the Division of Research

approved a program to be housed at the Oak Ridge Gaseous Diffusion Plant under the management of Union Carbide Corporation, Nuclear Division. Work was begun 1 November 1960, and included the construction of a cascade based on the Soviet design, as well as improvements to centrifuge theory, and a continuation of the study on novel materials. The original machines were 3-inch diameter aluminum rotors copied from the Zippe replica. The program moved quickly to stronger materials, including fiberglass-overwrapped aluminum and composites. The program also moved to 6 in rotors and then to 10-, 14-, 19-, 20-, and 24-in diameters. The timing of these developments are reflected in Table 1.

The focus throughout the 1960s was on improving the performance of individual machines. Work was split between Union Carbide at Oak Ridge and the AirResearch Manufacturing Company in Torrance, California. It took only two years for the program to move from the original Zippe model to a machine with more than 2 SWU/year capacity, and seven years to achieve a machine with 30 SWU/year capacity. By the end of the 1960s, the technology was considered mature enough to consider long-term reliability testing. So-called *Set I* centrifuges began reliability testing at the newly constructed Equipment Test Facility in 1972 and continued through at least 1977. Machine production techniques were studied at the Component Preparation Laboratories at both Oak Ridge and Torrance starting in 1974. In addition to Set I testing, basic machine design continued in parallel, leading to the development of additional sets. Reliability testing for Set II designs commenced in 1974, and Set III in 1977. This final set was tested on a large scale at the 50,000 SWU/yr Component Test Facility at Oak Ridge.

Table 1: Select U.S. centrifuges operated between 1961 and 2008. Values in parenthesis estimated by author. Blank areas denotes missing data. "X" in machine name indicates omitted name segment. Sources: AC-100 data from Glaser. All other notes of R. A. Lowry.

Name	Date	Diameter (in)	Length (m)	Velocity (m/s)	Material	δU actual (kg-SWU/yr)	Temp. (K)
Zippe UVA	Jan. 1961	3	0.305	350	Aluminum	0.39	321
Cascade 3	May 1961	3	0.305	370	Aluminum	0.46	(318)
Ti-6	March 1962	6	0.61	450	Titanium	1.53	(318)
Al-Fg-6	June 1962	6	0.75	450	Al-Fiberglass	1.75	(318)
Thermal-6	Oct. 1962	6		450		1.95	(318)
Cascade-6	March 1963	6		500		2.4	(318)
DMS-1	Oct. 1962	4.5	1.5	350	Aluminum		(318)
DMS-2	Jan. 1964	4.5	1.5	425	Al-Fiberglass	3.5	(321)
10-X-X	July 1965	10		550			
20-X-X		20					
14-X-X	1965	14	(2.16)			11	
14-X-X	1967	14	(4.04)	600		30	(315)
AC-100	2008	24	12	900	Composite	330	

In the late 1970s, the U.S. Department of Energy predicted that the demand for nuclear power, and thus enriched uranium, would grow dramatically in the coming decade. Saddled with only aging gaseous-diffusion plants, the DOE proposed a commercial-scale centrifuge facility to meet future enrichment demand. In 1977, Congress authorized an 8.8 million SWU/yr plant at Portsmouth, Ohio. The first stages of the plant were based on Set III centrifuges, 24 in diameter, 40 feet long, at 200 SWU/yr per machine. In time, however, it became apparent that the demand for enriched uranium had not increased as predicted; competing lifetime-extension programs on sunk-cost gaseous diffusion plants obviated the need for a new plant; and claims of economic mismanagement plagued the centrifuge program. The entire centrifuge effort was cancelled on June 5, 1985. Only 3000 machines had been installed at the Portsmouth facility at a total cost of \$2.6 billion.

THEORETICAL DEVELOPMENTS

Theoretical work can be broken into two categories: early efforts to estimate the performance of machines, and later efforts to understand the internal gas flows more precisely so as to guide optimization and the design of internal components. The former has its origins in the United Kingdom during the months just prior to the creation of the Manhattan Project. British physicists Franz Simon, Rudolf Peierls, Karl Fuchs and Nicholas Kurti worked out the general theory of isotope separation.¹ Paul Dirac used this theory to derive what is perhaps the first and most well-known expression for the separative performance for a gas centrifuge:

$$\delta U_{\max} = \rho D_{AB} \left(\frac{\pi Z}{2} \right) \left(\frac{\Delta M}{2\mathcal{R}T} \right)^2 (v_a)^4 \quad (1)$$

where ρD_{AB} is the density multiplied the coefficient of self-diffusion, the product of which is constant for a given gas; Z is the length, ΔM is the mass difference between the two isotopes being separated; \mathcal{R} is the gas-law constant; T is the temperature; and v_a is the peripheral velocity of the inner surface of the rotor.

Dirac's equation treats the centrifuge in a way that is independent of the internal operation of the machine. It is useful in that it correctly guides the experimentalist to seek foremost high peripheral velocities (v_a), second low temperatures (T), and third long rotors (Z)—but while useful and elegant, it overestimates the separative work for a machine. In particular, the fourth-power dependency of the peripheral velocity tends in practice to approach the second power, owing mainly to the generation of a vacuum core along the axis of the rotor. Thus, it remained a task to develop a theory that more accurately characterized physical machines.

Separative Performance

The distinctly U.S. theoretical strain begins with the Manhattan Project's uranium-enrichment effort, under the leadership of Harold Urey. Improvements to Dirac's formulation were developed as part of work done at Columbia University by Karl Cohen. He showed that the performance of a centrifuge depends on the shape of the countercurrent flow in the rotor and on the balance between the axial circulation of the gas and the radial diffusion of the isotopes.⁴ Cohen's solution follows in approach earlier work by Furry, Jones, and Onsager⁵ on thermal-diffusion columns, which have a similar internal flow. Cohen's solution assumes that (1) the countercurrent flow is uniform along the length of the rotor, (2) the radial change in the fraction of the desired isotope is small compared to the radial change in the integrated countercurrent mass flow, and (3) the gas is always in the continuum (viscous) regime. The following summary of Cohen's model is based on an improved presentation by Høglund, Schacter, and von Halle.⁶ The complete derivation is given in Appendix B.

Cohen's Model

The gas centrifuge works on the principle of pressure diffusion. The rotational motion of the centrifuge accelerates gas molecules toward the wall of the rotor. A counteracting diffusive force created by the thermal motion of the molecules seeks to distribute the gas evenly throughout the rotor volume. The two forces balance to create a dynamic equilibrium, which gives rise to a pressure distribution in the rotor that is a function of the molecular mass of the gas:

$$p(r) = p(0) e^{M\Omega^2 r^2 / 2RT} \quad (2)$$

When the rotor contains a mixture of gases, the distribution holds independently for each species. Thus, we have for two isotopes *A* and *B* the partial pressures:

$$p_A(r) = p_A(0) e^{M_A \Omega^2 r^2 / 2RT} \quad (3)$$

$$p_B(r) = p_B(0) e^{M_B \Omega^2 r^2 / 2RT} \quad (4)$$

Dividing the two equations gives a simple separation factor for the centrifuge, which is a function of the mass difference and the peripheral velocity:

$$\alpha_0 = \frac{p_A(0)}{p_B(0)} \bigg/ \frac{p_A(r)}{p_B(r)} = e^{[(M_B - M_A)\Omega^2 r^2 / 2RT]} \quad (5)$$

Equation (5) is valid in the equilibrium condition where the gas is stationary in the reference frame of the rotor and enough time has passed that the molecules have diffused into their final distribution. However, the performance of a centrifuge can be enhanced by introducing a countercurrent flow, in which the gas flows up the center axis of the rotor and back down along the rotor wall. This countercurrent motion exploits the radial diffusive process

repeatedly along the length of the centrifuge, multiplying the separation effect much as a fractional distillation column would. Because the gas never achieves the dynamic equilibrium mentioned above, the radial separation at any position along the axis is never as large as in Eq. 5, but the multiplying effect of the countercurrent flow produces an axial separation that is many times greater.

Cohen derives an expression for the overall axial separation in a centrifuge based on the individual transport phenomena inside the centrifuge. Consider the axisymmetric cylindrical volume element, shown in Figure 1, which contains a binary gas mixture with components *A* and *B*:

The isotopes diffuse through the volume element differently in each direction. In the radial direction there is a concentration gradient and a pressure gradient, both of which give rise to diffusive currents Eq. 6.² In the axial direction, there is a concentration gradient but no pressure gradient Eq. 7. In the azimuthal direction there is no net diffusion because the centrifuge is axisymmetric Eq. 8. Thus, for the desired isotope *A*, we have:

$$\mathbf{J}_A \cdot \hat{r} = -cD_{AB} \left[\frac{\partial N}{\partial r} + \frac{(M_B - M_A)N(1 - N)}{\mathcal{R}T} \Omega^2 r \right] \quad (6)$$

$$\mathbf{J}_A \cdot \hat{z} = -cD_{AB} \left[\frac{\partial N}{\partial z} \right] \quad (7)$$

$$\mathbf{J}_A \cdot \hat{\theta} = 0 \quad (8)$$

where (r, θ, z) are the cylindrical polar coordinates with the origin fixed at the bottom of the centrifuge on the axis of rotation; c , D_{AB} , and T are respectively the molar density, mass density, binary self-diffusion coefficient, and temperature of the gas entire; \mathcal{R} is the universal gas constant; Ω the angular velocity; and M and N the molar mass and mole fraction of isotope *A*, respectively.

The non-diffusive countercurrent transport is characterized by a velocity field $\mathbf{v} = v(r) \cdot \hat{z}$, illustrated in Figure 2. Cohen solves the problem without specifying a function explicitly but requires that the magnitude of the velocity is a function of r only. The effect of different velocity profiles is discussed in Flow Profile Efficiency.

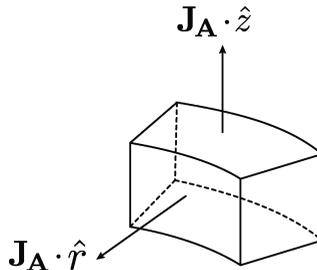


Figure 1: Diffusive fluxes in a volume element.

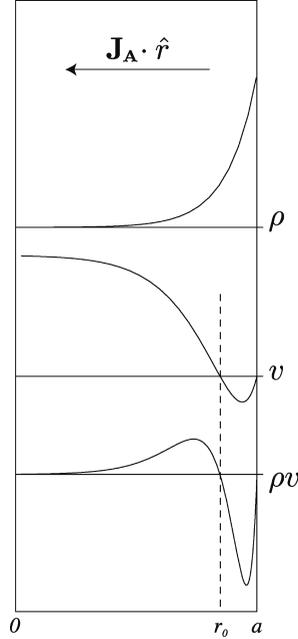


Figure 2: Schematic profiles in a centrifuge. ρ : the density; v : the countercurrent velocity; and ρv : the countercurrent mass flow.

Having defined the basic transport phenomena, Cohen proceeds to write an expression that joins the diffusive and nondiffusive components together. The task is simplified if we consider the centrifuge as two parts: the segment above the feed point that emits product enriched in the light isotope (called the *rectifier*), and the segment below the feed point that emits tails depleted in the light isotope (the *stripper*). Only the rectifier is treated here, but a parallel analysis applies for the stripper.

The net transport of component A being withdrawn as product at the top of the centrifuge is the sum of the countercurrent transports up and down the rectifier, plus the axial back-diffusion of the isotope (Eq. 7), which is nonzero and negative as a consequence of the axial separation gradient:

$$\frac{d(PN_P)}{dz} = 2\pi \int_0^a cvN_r dr - 2\pi \int_0^a cD_{AB} \frac{\partial N}{\partial z} r dr \quad (9)$$

where P is the molar flow rate on the product side, and N_P is the mole fraction of isotope A in the product.

The net transport of both components is:

$$P = 2\pi \int_0^a cv_r dr \quad (10)$$

As evident from Eq. A4, Eqs. 9 and 10 are sufficient to determine the performance of the rectifier. If a similar analysis is applied to the stripper, we can sum the two parts to obtain the performance for the entire machine. Nearly all of the parameters are known. The molar density c is known from Eq. 2. The value cD_{AB} is a constant according to the kinetic theory of gasses. The velocity profile $v(r)$ is determined by the design of the machine. The only remaining task is to determine an expression for $N(r, z)$. The solution, given in Appendix B, takes the form of an expression for $dN(z)/dz$, which can be integrated along the length of the centrifuge to find the molar concentration of the desired isotope in the product stream, N_P , and accordingly used to compute the separative work by Eq. A4. With this solution in hand, the problem is reduced to determining the appropriate function for the countercurrent profile $v(r)$. If the profile is known, then we can proceed directly using analytical or numerical integration. However, often we do not know the profile and we must assume a function. Some common assumptions are discussed in Flow Profile Efficiency.

The Development of Correction Factors

If we compare Cohen's expression for separative work to Dirac's original formulation given by Eq. 1, we can develop a correction term for Dirac's formula. In this section, we derive this and other correction terms and separate them into a series of "efficiency factors" according to the physical phenomenon each represents. The separative power can then be expressed as:

$$\delta U = \delta U_{\max} e_I e_C e_F e_E \dots \quad (11)$$

This approach is useful because the parameters that govern each phenomenon can be considered individually. If we then estimate these parameters in an intelligent way, we can produce a good estimate for the performance of a generic machine with only a minimum of information.

Ideality Efficiency

The result given in Appendix B is an expression with a form analogous to the gradient equation of a square cascade or fractional distillation column:

$$SdN/dz = (\alpha - 1)N(1 - N) - (P/L)(N_P - N) \quad (12)$$

where S is the virtual stage length inside the centrifuge, α is the stage separation factor, and L is the interstage mass up-flow inside the centrifuge. Assumption (1) of Cohen's solution was that the countercurrent flow was non-decaying. Accordingly, the interstage up-flow L is constant along the length of the centrifuge, which is analogous to a square cascade. Dirac's estimate, by contrast, assumed that the internal workings of the centrifuge approximated an ideal cascade. It can be shown from general separation theory that a square

cascade performs to only $\sim 81\%$ of an ideal cascade, giving us the *ideality efficiency*:

$$e_l = 0.81 \quad (13)$$

In very long or very sophisticated centrifuges, damping can cause the counter-current flow to decay along the length of the rotor. This profile more closely approximates an ideal cascade and can lead to ideality efficiencies of greater than 81%. However, such advanced designs are rare and difficult to construct.

Circulation Efficiency

The *circulation efficiency* takes into account the effect of axial back-diffusion. Recall that the transport of isotope A (Eq. 7) contained a positive contribution from the internal countercurrent flow and a negative contribution from axial back-diffusion. Since Dirac's estimate does not consider axial back-diffusion, it overestimates the performance and the circulation efficiency corrects for this.

The circulation efficiency is part of a larger efficiency term that arises when comparing Cohen's expression to Dirac's expression. The separative work for separating element whose separation factor is very small, $(\alpha - 1) \ll 1$, can be expressed as:

$$\delta U_S = L(\alpha - 1)^2 \quad (14)$$

This condition is usually met for each virtual stage inside the centrifuge. Thus, by dividing by the stage length S , we derive an approximate expression for the separative work per unit length of the centrifuge:

$$\frac{\partial(\delta U)}{\partial z} = \frac{L(\alpha - 1)^2}{S} \quad (15)$$

where L , $(\alpha - 1)$, and S are all determined from Cohen's theory and are given in Appendix B. When divided by the derivative of Dirac's expression, $\partial(\delta U_{\max})/\partial z$, we produce a correction term. The portion of that term arising from the axial back-diffusion is the circulation efficiency:

$$e_c = \frac{m^2}{1 + m^2} \quad (16)$$

where $m \equiv L/L_0$ is the ratio of the internal interstage mass up-flow L to the up-flow which gives the greatest separation factor L_0 , and both of which are defined in Appendix B. Conceptually, m is a dimensionless measure of how fast the mass is circulated within the machine. It is worth noting that e_c converges to 1 rapidly. Typical values of m in an optimized centrifuge may be around 3 to 4, giving 90 and 94% efficiency, respectively.

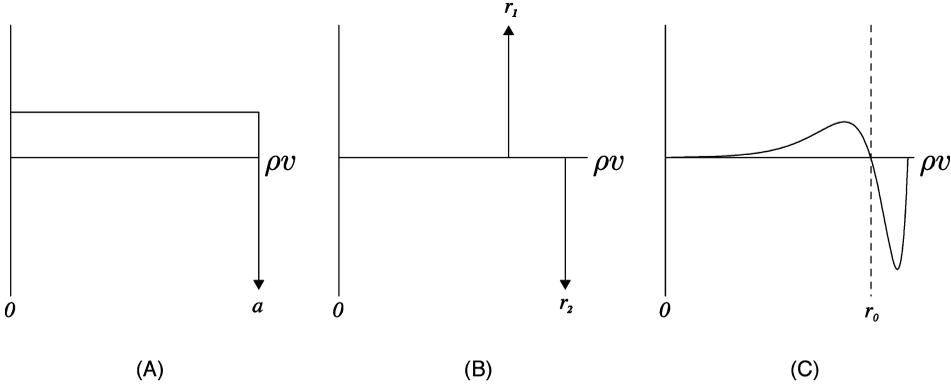


Figure 3: Profiles commonly used in estimating the flow-profile efficiency. (A) The idealized profile. (B) The two-shell profile. (C) The thermal profile.

Flow Profile Efficiency

The *flow-profile efficiency* takes into account the impact of the shape of the countercurrent velocity profile on the separative performance. By following the same procedure described for the circulation efficiency above, it can be shown that the portion of the overall efficiency factor arising from the countercurrent flow term is

$$e_F = \frac{4 \left[\int_0^a \left(\int_0^r cvr' dr' \right) r dr \right]^2}{\alpha^4 \int_0^a \frac{1}{r} \left(\int_0^r cvr' dr' \right)^2 dr} \quad (17)$$

Since the countercurrent-velocity profile v is often unknown, it is useful to consider several generic profiles.

Idealized profile. The idealized profile assumes that mass flows upward equally over the entire volume of the centrifuge, except for a returning delta function at the wall of the centrifuge, as shown in Figure 3(A). This profile, while not physically possible, yields a flow-profile efficiency of unity and corresponds to the assumed profile in Dirac's expression for the maximum possible separative work.

Two-shell profile. A simple velocity profile used in theories like that of Rätz assumes two thin streams, one situated at r_1 flowing upward and one at r_2 flowing downward. The profile, shown in Figure 3(B), can be shown to have an efficiency of:

$$e_F = \left[1 - \left(\frac{r_1}{r_2} \right)^2 \right]^2 / \ln \left(\frac{r_2}{r_1} \right) \quad (18)$$

The two-shell profile is also not physically possible, but it simplifies the mathematics significantly and can provide a reasonable estimate for the efficiency

if r_1 and r_2 are chosen carefully. For more on the uses of the two-shell profile, see Glaser.¹

Thermal profile. A physically realistic profile is that excited by a linear temperature gradient along the wall of the centrifuge. The profile, shown in Figure 3(C), was derived in Wood and Morton⁸ using Onsager’s pancake approximation (see Flow Theory), and corresponds to the “universal high-speed pattern” derived by Parker and Kelly of the University of Virginia in 1967. It turns out that the mathematical expression for the profile can be integrated analytically and is also usefully representative of the profiles in a real machine. For this reason, it is widely used for computing the performance of a generic machine. It has a flow-profile efficiency of:

$$e_f = \frac{14.4\mathcal{R}T}{Mv_a^2} \quad (19)$$

This flow-profile efficiency is inversely proportional to the square of the peripheral velocity. Thus, when multiplied by Dirac’s expression, it reduces the overall velocity dependence of the separative work from a fourth to second power, which fits experimental data better.

Experimental Efficiency

The *experimental efficiency* lumps together all the inefficiencies not addressed by the forgoing, such as turbulent mixing or disturbances to the countercurrent flow. The factor is unique to every machine, but the efficiency typically increases as designs become more cultivated. Figure 4 shows estimates for the experimental efficiencies of 17 machines calculated by assuming that 100% performance corresponds to that given by the formulas above. Note that some of the most advanced U.S. machines have “efficiencies” greater than 100%. These machines are so highly tuned that the slightly beneficial effects of feed and baffles become visible, effects that were not included in the expressions for the flow-profile efficiency in Eq. (17).

Simple Formulation for Policy-Makers

By combining the forgoing efficiency factors, assuming a linear-thermal-gradient profile, taking the typical values $m = 3$, $T = 310$ K, and using the diffusion constant for UF_6 given in appendix C, we arrive at a simplified equation for the separative work of an generic centrifuge:

$$\delta U = \frac{V^2 Z}{33,000} e_e \quad (20)$$

where V is the peripheral velocity in meters per second, Z is the length of the centrifuge in meters, e_e is the experimental efficiency described above, and δU is in the usual units of kg-SWU per year. From Figure 4, it is reasonable to

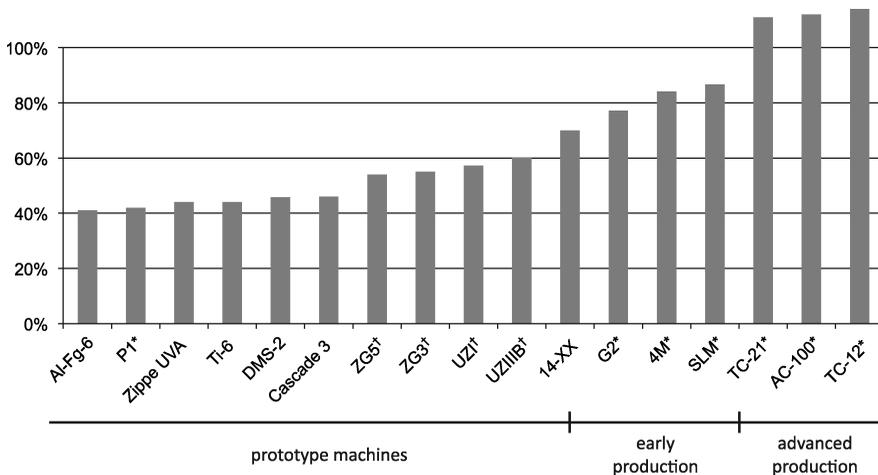


Figure 4: Experimental efficiencies for a selection of centrifuges. Centrifuges marked with an asterisk (*) were built by the Urenco consortium and are based on performance estimates reported by Glaser.¹ Centrifuges marked with a dagger (†) are pre-Urenco German models based on data reported by Groth⁹ and have uncertainties of $\pm 4\%$. All other values are based on experimental data from the U.S. centrifuge program and have uncertainties of less than 1%. See main text for an explanation of why some models exceeded 100% efficiency.

assume that e_e is in the range of 0.35 to 0.45 for new designs; in the range 0.50 to 0.60 for established production machines; and in the range of 0.8 to 1.14 for the most advanced centrifuge designs.

Flow Theory

Cohen's theory is a one-dimensional approximation of the centrifuge in which an assumed behavior of the gas is imposed for the purpose of estimating separative performance. In particular, it ignores the effect of caps and baffles at the end of the rotor, which interfere with and turn the countercurrent flow through the generation of an Ekman layer. The estimate is reasonable for very long centrifuges, but in most practical machines the Ekman layer extends over a significant fraction of the rotor's length, weakening the magnitude of the countercurrent velocity as the gas moves from the center to the end regions, and changing the machine's separative performance. Additionally, the Cohen theory requires that we assume a velocity profile for the countercurrent flow, whereas a more complete theory would determine the shape of the flow according to prescribed boundary conditions. The Navier-Stokes equations can describe the gas dynamics exactly, but as with most fluid-dynamic systems, there is no closed-form solution.

The development of a fully two-dimensional closed-form analytical theory thus became the focus of a U.S. effort known as the Centrifuge Flow Theory Study Group founded in the early months of 1961. Lars Onsager of Yale

University was selected to chair the group. Other members included George F. Carrier of Harvard University, Stirling A. Colgate of the Lawrence Radiation Laboratory, Wendell C. DeMarcus of the University of Kentucky, Carl Eckart of the University of California, Harold Grad of New York University, and Stephen A. Maslen of the Martin Company. Maria Goeppert-Mayer of the University of California was added in October 1963.

Within a year of its formation, Onsager had led the group in developing an extension of the thin-shell approximation that he originally proposed for the thermal-diffusion column 23 years earlier.⁵ Onsager noted that at high speed most of the gas was compressed in a thin annulus against the wall of the centrifuge. Starting with the linear Navier-Stokes equations, he retained only those terms most highly differentiated in the radial direction and treated the gas as being locally flat by substituting the radius of the centrifuge for the radial coordinate wherever it appeared algebraically (i.e., $r \rightarrow a$), a step known today as the “pancake approximation.” The result was a sixth-order, anisotropic linear elliptic partial differential equation in two dimensions. Carrier and Maslen developed the mathematics and worked out appropriate boundary conditions for the Ekman layer at the end of the centrifuge. Onsager showed that the homogenous part could be solved by eigenfunctions and linked the solutions to various physical phenomena. The first numerical results were obtained in 1976. Houston Wood and Jeffrey B. Morton added the non-homogenous solutions, which took into account feed effects and scoop drag, and published solutions for the homogenous case in 1980.¹⁰ The complete pancake model is able to predict the separative work of a real centrifuge to within 1–2%.

It became possible in the mid to late 1970s to model internal flows using computer simulations, but this did not lead to the demise of the analytical model. Numerical solutions were originally expensive and time consuming. Even with today’s computing capabilities, the problem is difficult to set up because of the extreme range of length scales and the stiffness of the equations involved. But the main advantage of the pancake model was that it could give analytical insight into the workings of the centrifuge that were previously unobtainable with one-dimensional models and without the many iterations required to do phenomenology using numerical methods.

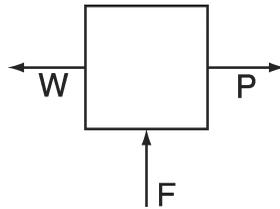
NOTES AND REFERENCES

1. A. Glaser, “Characteristics of the Gas Centrifuge for Uranium Enrichment and Their Relevance for Nuclear Weapon Proliferation,” *Science and Global Security* 16(1–2) (2008): 1–25.
2. H. G. Wood, “Effects of Separation Processes on Minor Uranium Isotopes in Enrichment Cascades,” *Science and Global Security* 16(1–2) (2008): 26–36.
3. R. DeWitt, *UF₆: A Survey of the Physico-Chemical Properties*, (Goodyear Atomic Corp.), (1960).

4. K. Cohen, *The Theory of Isotope Separation as Applied to the Large-Scale Production of U235*, (McGraw-Hill, 1951).
5. W. H. Furry, R. Clark Jones, and L. Onsager, "On the Theory of Isotope Separation by Thermal Diffusion," *Physical Review*, 55(1939), 1083–1095.
6. R. L. Høglund, J. Shacter, and E. von Halle, "Diffusion Separation Methods," in *Encyclopedia of Chemical Technology*, pp. 639–723 (John Wiley and Sons, 1979).
7. W. E. Groth, K. Beyerle, E. Nann, and K. H. Welge, "Enrichment of Uranium Isotopes by the Gas Centrifuge Method," In *Proc. Second International Conference on the Peaceful Uses of Atomic Energy*, Geneva pp. 439–446, (1958).
8. H. G. Wood and J. B. Morton, "Onsager's Pancake Approximation for the Fluid Dynamics of a Gas Centrifuge," *Journal of Fluid Mechanics*, 101(1980), 1–31.
9. S. Villani, ed., *Uranium Enrichment* (Springer-Verlag, 1979).
10. L. Zarkova and U. Hohm, *Journal of Physical Chemistry Ref. Data*, 31 (2002), 1.

APPENDIX A: AN INTRODUCTION TO ISOTOPE SEPARATION

For a three-stream separating element, like the centrifuge, we have feed, product, and waste flows. The magnitude of the flows (in moles per unit time) are designated F , P , and W ; the mole fraction of the desired isotope in any stream is designated N with the appropriate subscript. Inside the separating element (centrifuge rotor), N becomes a function of position.



$$N \equiv \frac{n_A}{n} = \frac{p_A}{p} = \frac{\rho_A}{\rho} \quad (\text{A1})$$

We also define the abundance R :

$$R \equiv \frac{N}{1-N} \quad \text{giving} \quad N = \frac{R}{1+R} \quad (\text{A2})$$

A measure of the separation between any two streams is the ratio of their respective abundances. Thus, we have individual separation factors for the feed-to-product stream and the waste-to-feed stream:

$$\alpha \equiv \frac{R_P}{R_F} \quad \beta \equiv \frac{R_F}{R_W} \quad (\text{A3})$$

Definition of Separative Work

The performance metric of a separating element must be a function of both the total separation it can produce ($\alpha\beta$) and the amount of material that the separating element can process per unit time. Thus, separative work is defined as:

$$\delta U = PV(N_P) + WV(N_W) - FV(N_F) \quad (\text{A4})$$

where V is a value function for a binary mixture contrived to operate such that (1) the lowest value is (arbitrarily) set to be a 50:50 mixture, (2) the value increases as the mixture becomes purified in either component, and (3) scales in such a way that the work of n elements (arranged in a non-mixing cascade) will produce n times the work of a single element given the product and waste values $R_P = \alpha^n R_F$ and $R_W = \beta^n R_F$. It can be shown that the value function is:

$$V(N) \equiv (2N - 1) \ln \left(\frac{N}{1 - N} \right) \quad (\text{A5})$$

A mathematical discussion of its origins, and a more complete introduction to separation theory, can be found in Villani.⁹

APPENDIX B: SOLUTION FOR VON HALLE'S FORMULATION OF COHEN'S NET DESIRED MATERIAL TRANSPORT

The axisymmetric continuity equation for the desired component is:

$$\nabla \cdot (\rho \mathbf{v} + \mathbf{J}_A) = 0 \quad (\text{B1})$$

$$\frac{1}{r} \frac{\partial}{\partial r} r (\mathbf{J}_A \cdot \hat{r}) + \frac{\partial}{\partial z} (cvN + \mathbf{J}_A \cdot \hat{z}) = 0 \quad (\text{B2})$$

Cohen asserts that the isotopic gradient, and thus axial back-diffusion, is constant along the length of the centrifuge:

$$\frac{\partial}{\partial z} (\mathbf{J}_A \cdot \hat{z}) = -cD_{AB} \frac{\partial}{\partial z} \left[\frac{\partial N}{\partial z} \right] = 0 \quad (\text{B3})$$

Substituting (B3)→(B2), and integrating with respect to r :

$$\mathbf{J}_A \cdot \hat{r} = -\frac{1}{r} \int_0^r cv \frac{\partial N}{\partial z} r' dr' \quad (\text{B4})$$

Setting Eq. (6) equal to Eq. (B4) and solving for $\partial N / \partial r$:

$$\frac{\partial N}{\partial r} = -\frac{\Delta M \Omega^2 r}{\mathcal{R}T} N(1 - N) + \frac{1}{rcD_{AB}} \int_0^r cv \frac{\partial N}{\partial z} r' dr' \quad (\text{B5})$$

Now we define the function:

$$G(r) \equiv \int_0^r 2\pi c v r' dr' \quad (\text{B6})$$

$$\frac{dG}{dr} = 2\pi r c v \quad \text{and} \quad G(0) = 0 \quad (\text{B7})$$

Thus, we have:

$$\begin{aligned} \int_0^a 2\pi c v N r dr &= \int_0^a \frac{dG}{dr} N dr & (\text{B8}) \\ &= \int_0^a N dG \\ &= GN \Big|_0^a - \int_0^a G dN \\ &= G(a)N|_a - G(0)N|_0 - \int_0^a G \frac{dN}{dr} dr \end{aligned}$$

and by (B7), (B6), and (10):

$$\begin{aligned} &= N|_a P - \int_0^a G \frac{dN}{dr} dr \\ &= N|_a P - \int_0^a G \frac{\partial N}{\partial r} dr \end{aligned}$$

The net upward transport of the desired component (Eq. 9) can then be written:

$$PN_P = N|_a P - \int_0^a G \frac{\partial N}{\partial r} dr - \int_0^a 2\pi c D_{AB} \frac{\partial N}{\partial z} r dr \quad (\text{B9})$$

Cohen assumes that the axial isotopic gradient is the same at any radius, and that the isotopic concentration changes only slowly with r such that $\partial N/\partial z$ and $N(1-N)$ can be treated as constants. While this is not exactly true, full three-dimensional solutions of the Navier-Stokes equations reportedly show that it is very nearly true. Recall also that cD_{AB} is a constant. Substituting in Eq. B5 and collecting the terms with $\partial N/\partial z$, we have:

$$\left(\int_0^a \frac{2\pi}{cD_{AB}} \frac{G^2}{r} dr + \pi \alpha^2 c D_{AB} \right) \frac{\partial N}{\partial z} = \left(\frac{\Delta M \Omega^2}{RT} \int_0^a G r dr \right) N(1-N) - P(N_p - N|_a) \quad (\text{B10})$$

which, if we divide through by L , would have the form of the gradient equation analogous to that of a fractionating column given in Eq. (12):

$$S dN/dz = (\alpha - 1)N(1-N) - (P/L)(N_p - N)$$

where L is a measure of the upflowing mass in the countercurrent flow, which for the gas centrifuge is:

$$L \equiv \int_0^{r_0} 2\pi c v r dr \quad (\text{B11})$$

where r_0 is the inflection point in the counter-current profile shown in Figure 2. From this and Eq. (B10) we can solve for the stage length:

$$S = \frac{1}{L} \left(\int_0^a \frac{2\pi}{c D_{AB}} \frac{G^2}{r} dr + \pi \alpha^2 c D_{AB} \right) \quad (\text{B12})$$

where the first term in parentheses is the contribution from the countercurrent flow, and the second term is the contribution for the axial back-diffusion. Similarly, the stage separation factor is:

$$\alpha - 1 = \frac{\Delta M \Omega^2}{\mathcal{R} T} \frac{1}{L} \int_0^a G r dr \quad (\text{B13})$$

Both L and G are functions of c and v only, and both of which are known. If one notes that both L and G depend linearly on the magnitude of v , then it is evident that the countercurrent portion of the stage length varies directly with the magnitude of L , whereas the diffusive term varies inversely with the magnitude of L . Thus, there exists a value for the circulation rate L where the stage S length is minimized, and therefore the number of stages in any given length of rotor is maximized. These are:

$$S_0 = \frac{\alpha \left[2 \int_0^a \frac{1}{r} G^2 dr \right]^{1/2}}{\int_0^{r_0} c v r dr} \quad (\text{B14})$$

$$L_0 = \pi c D_{AB} \left[\frac{2}{\int_0^a \frac{1}{r} G^2 dr} \right]^{1/2} \int_0^{r_0} c v r dr \quad (\text{B15})$$

These let us rewrite Eq. (B10) as:

$$S \frac{dN}{dz} = \left(\frac{1 + m^2}{2m} \right) S_0 \frac{dN}{dz} = (\alpha - 1) N (1 - N) - \left(\frac{P}{m L_0} \right) (N_P - N) \quad (\text{B16})$$

Since the overall separation factor for the centrifuge is inversely proportional to the stage length, the overall separation factor scales as:

$$\left(\frac{2m}{1 + m^2} \right) \quad (\text{B17})$$

APPENDIX C: PROPERTIES OF UF₆

The following are from references 3 and 10 in Kelvin-centimeter-gram-second units.

Equation of state:

$$\rho(p, T) = (4.235 \times 10^{-6})p \left/ \left(T - \frac{1.359p}{T^2} \right) \right.$$

Dynamic viscosity:

$$\eta(T) = (-2.099 \times 10^{-13})T^3 + (2.783 \times 10^{-10})T^2 + (3.83 \times 10^{-7})T + 3.907 \times 10^{-5}$$

Coefficient of self-diffusion times density:

$$D\rho(T) = (-1.725 \times 10^{-14})T^3 + (1.33 \times 10^{-11})T^2 + (6.349 \times 10^{-8})T + 2.756$$

Vapor pressure:

$$p_v = \begin{cases} \exp \left[\frac{0.01736(T-195.4)(T+1094)}{T-89.73} \right] & T < 337.2 \\ \exp \left[23.30 + \frac{4668}{92.137-1.8T} \right] & 337.2 \leq T \leq 389.3 \\ \exp \left[24.90 - \frac{6976}{52.20+1.8T} \right] & T > 389.3 \end{cases}$$