APPENDIX VI-C

ELECTROMAGNETIC SEPARATION OF ISOTOPES

by

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Abstract

Since World War II there have been many advances in technology which are relevant to the development of electromagnetic separation of uranium on a large scale. These include magnets, pumps, controls and apparatus for carrying out the related chemical operations.

A significant contribution may be the techniques and hardware which **have** been developed for ion propulsion of spacecraft. It is necessary, however, to modify the systems to provide very intense focused beams of singly charged uranium ions instead of broad diffuse beams of elements such as cesium. Some progress has been made in the development of electrohydrodynamic sources in which ions are extracted directly from the surface of a liquid metal. A low accelerating potential may permit the use of smaller intensity magnetic fields of limited size.

If the many scientific and engineering problems can be solved, it seems possible that an electromagnetic isotope separator based on this new technology can efficiently produce enriched uranium. Because individual units are small and are able to effect a rather high degree of separation of isotopes this process may be suitable for the production of kilogram quantities of weapons grade uranium.

<u>Introduction</u>

The electromagnetic method for enrichment of uranium was found during World War II to be uneconomical as compared with gaseous diffusion. All of its components have been declassified and many are now available from commercial sources. Many advances have been made during the past thirty years which are relevant to this process. In this paper the possible effect of these advances on the efficiency of the system is assessed. Also, the difficulty of assembling and operating such a system by a small country with modest technical resources is estimated.

<u>History</u>

The electromagnetic method for separation of uranium isotopes on a large scale was developed by the United States during World War II. Nearly 500 million dollars were spent for equipment and operation. (This is nearly one-quarter the total cost of the Manhattan project and is about the same amount as was used for each of the other two major efforts, gaseous diffusion and reactors for the production of plutonium.) The Y-12 plant at Oak Ridge included 850 first-stage "Alpha" units (Calutrons) and 72 units in the second "Beta" stage. When it was shut down in December 1945 because of its low efficiency, 7000 persons were needed to keep it in operation. **1**/

After the war, scientists in the United States, the United Kingdom, and the Soviet Union used electromagnetic separators developed in wartime for the protection of highly enriched samples of practically all of the elements. Electromagnetic separators for scientific research have also been developed in Switzerland, the Netherlands, Germany, Denmark, Sweden and Austria. They are now available commercially and are widely used as sources of both stable and radioactive isotopes. For these applications, milligram or gram quantities are usually sufficient.

Development Objectives

In order to make this method suitable for large-scale isotope separation

it was necessary:

- 1. To develop focusing magnetic fields with small aberrations for large angle ion beams.
- 2. To design large well regulated power supplies for the source, accelerating electrodes and the magnet.
- 3. To develop large pumping systems to maintain a high vacuum in large volumes in which a considerable quantity of gas is released.
- 4. The determination of the effect of space-charge repulsion on the ion trajectories in dense ion beams.
- 5. The production of relatively large ion currents (about one hundred milliamperes).
- 6. The development of methods for the efficient collection of the enriched material.
- 7. The training of personnel and the development of techniques necessary for the operation of the system. The steps include:
 - a. Preparation of charge material
 - b. Assembly of sources and receivers
 - c. Operation of the separator
 - d. Extraction of the separated material
 - e. Chemical refining
 - f. Measurement of isotopic abundance
 - q. Cleaning of source and liner

Output

<u>Faraday's Law</u> tells us that a 100 milliampere current of singly ionized uranium atoms corresponds to a flow from the source of 24 grams during 24 hours of operation. In case of uranium, this corresponds to 0.17 grams of U-235 per day or 0.06 kg. per year at continuous operation. In actual practice many of the ions which leave the source do not reach the collector. This is because: 1) not all of the ions which leave the source are singly charged; 2) many ions are lost from the beam in passage through the separator; 3) not all the ions which reach the collector are collected.

Overall the source-to-receiver efficiency is found to be under 10 percent for well defined foci such as are required for separation of the heaviest masses although figures of around 20 percent have been quoted for the production separators in the Oak Ridge plants. $\underline{2}/$

A 100 milliampere uranium isotope separator with an efficiency of 20 percent will have an output of about 5 grams of uranium a day or 0.035 grams of U-235 per day.

Space Charge Compensation

In the beam itself mutual repulsion of the positive ions would spread out their trajectories were it not for the production by collisions with gas molecules of electrons which neutralize the space-charge forces. This phenomenon has been the subject of intense investigation. It is essential for the operation of the calutron separator.

At the beginning of their trajectory, slow ions and electrons are formed in the beam. The slow moving ions drift from the beam while the electrons concentrate in the potential well at the axis of the beam. With increasing density of negative particles the potential well is gradually smoothed out while electrons and ions of sufficient energy continuously leave the beam. Finally, an "electron gas" of "thermal energies" will be concentrated in the beam. The "temperature" of this gas and the depth of the remaining potential well is defined by equilibrium between the production and loss of charged particles. 3/

Ion-Ion Scattering

1. Alexeff has found a fundamental limit to the throughput of U-235 in an electromagnetic isotope separator. **4**/ Although all ions are extracted

with equal energies the lighter ones move faster than the heavier ones because of the mass difference. In addition to the azimuthal drift there is a slow radial drift in the magnetic field which is responsible for the isotope separation. Both drifts are of the same order of magnitude. When one charged particle drifts very slowly through an environment of other charged particles it is susceptible to multiple scattering. Thus, the directed relative velocity is easily lost.

Reducing the beam voltage reduces this relative velocity and increases the undesired scattering. Increasing the beam current increases the number of scattering centers and also increases the undesired scattering. It can be shown that mass spectrograph isotope separators have a limit on V^2/j , the ratio of the square of the beam voltage to the current density, due to-the scattering of U-235 ions by collisions with U-238 ions. A crude approximation by Alexeff suggests that calutron isotope separators operate within a factor of 100 of this limit.

Enri chment

The operation of an electromagnetic isotope separator is characterized by a very large isotopic separation constant (ratio of the isotopic concentration of the enriched product to that of the feed material).

It depends on the shape of the two beams, their separation and the size of the receiver slits. As beam current is increased) ion-ion scattering reduces the separation achieved, In large-scale electromagnetic separators (Alpha calutrons) the enrichment factor per cycle is usually 20 to 40. With an enrichment factor of 20 the concentration can be increased in one stage from 0.7 percent to about 13 percent U-235. (In March 1944, 200 grams of material enriched to about 12 percent U-235 had been produced by Alpha 2.) **5**/

An Alpha Calutron isotopic separator which processes a total of 5 grams of uranium per day will produce about 0.3 grams enriched to 12 percent U-235 per day. (This is consistent with the estimate made in the fall of 1942 that 2000 sources and collectors were expected to be required to separate 100 grams of U-235 per day.) $\underline{6}/$

Tails

An advantage of the electromagnetic separator is the very low concentration of U-235 in the waste, or tails. This results in a significant saving in the quantity of uranium required as feed as compared to gas diffusion or the centrifuge which normally operate with a tails concentration of 0.2 percent U-235. Stated another way, approximately 30 percent of the U-235 contained in the feed to a gaseous diffusion plant comes out in the tails.

<u>Apparatus</u>

A single calutron-type separator requires a one hundred ton electromagnet with a rated power of 45 kilowatts. Two large capacity multiple stage oil diffusion pumps (rated power about 5 kilowatts each) are required to maintain the vacuum of 10⁻⁵ Torr. A 100 milliampere uranium tetrachloride arc ion source and a 40 kilovolt power supply are used to provide the beam which is deflected 180° in a semi-circle with a radius of 120 centimeters by the magnetic field before it enters the collector. The cost of each such separator is several hundred thousand dollars. More than a thousand of these units would be required to produce enough highly enriched uranium for one explosive per year. Even with the installation of more than one source and receiver per unit and the assembly of many units in a single magnet (Racetrack), a system based on the, calutron is simultaneously capital, labor, and energy intensive. In the form described, it would not only be the process chosen

by a country seeking to produce material for a nuclear weapon.

Prospects for Development

Advances have been made during the past 30 years in many aspects of the technology relevant to electromagnetic separators. These include ion sources, magnets, vacuum pumps, power supplies and controls. Although quantitative improvements have been made in the performance of all the components, additional development is required to make the process attractive to a country wishing to produce material for an explosive.

Ion Sources

An increase in the beam current will result in a corresponding increase in the rate of production of separated material.

1. Arc Source

After several years of intensive wartime development the uranium tetrachloride arc source was selected as superior to other candidates. It produces a large current with a relatively large percentage of singly charged uranium ions. Independent studies of sideband efficiencies from uranium tetrachloride arc sources in 180° separators at 0ak Ridge and at Amsterdam show that the singly ionized uranium ions constitute 60 to 70 percent of all uranium containing ions. 7/ This compound seems better in this respect than the other uranium tetrahalides.

The beam current for these sources has up to now been limited to about 200 milliamperes because of instabilities which develop at greater values. Of course, an increase in beam current will result in an increase in the generation of chlorine which in turn requires vacuum pumps of greater capacity. High speed turbine pumps are available which may be suitable. Another problem is the increased spread of each of the separated beams which may increase losses and decrease the degree of enrichment achieved. Some form of space charge compensation or other technique for neutralization of the beam will be necessary.

2. El ectrohydrodynami c Source

During the past twenty years an intensive research and development effort has been" carried out on systems intended for ion propulsion of spacecraft. One goal of this research has been the production of large currents of metal ions.

These sources are now used in microprobe for analysis. One variation uses a hypodermic needle filled with liquid metal. A meniscus at the tip is formed into a cone by an applied electric field ("<u>Taylor Cone"</u> with a theoretically predicted half angle of 49°). A very large local field is developed a the tip which extracts ions from the surface. The maximum ion current from a 0.005 inch diameter needle is about one hundred microampere into a large solid angle. Metals used have included cesium and gallium. Nearly all the ions produced from these sources are singly charged. ~/

R. Clampitt, Culham Laboratory, United Kingdom, has described an Electrodynamics Ion Source which uses cesium in a tube with an axial wire, at a recent conference on electric propulsion. **9**/ If these sources could be developed to function as a line source (from a slit) rather than as a point large currents might be achieved, perhaps many hundreds of milliamperes.

Such sources if developed for uranium metal have several advantages as compared to the halide arc. These include:

1. Source feed would be uranium metal.

2. No filaments are needed (a continuous problem in the arc source).

- 3. Very small down time is required to add feed material or to remove product.
- 4. pumping requirement would be relatively small because no gas is evolved from source.
- 5. If uranium behaves in a manner similar to cesium or gallium all of the ions will be singly charged (no side band losses).
- 6. Since a lower pressure could be maintained in the separator, there may be less loss of ions from the beam. A higher source-to-collector efficiency might result.

There are also problems to be solved:

- 1. Achievement of a large uranium ion beam current in a high vacuum chamber will result in a spread of the beam due to mutual repulsion of the ions. Some technique for neutralization must be developed. Also the maximum current will be limited by ion-ion scattering.
- 2. Suitable materials for fabrication of the source must be found. It will be necessary to have molten uranium (1300°C) maintained at **a** constant temperature.
- 3. Techniques for maintaining stability of the beam must be developed.
- 4. Extracting and accelerating electrode structures must be designed. In order to have a smaller magnet it would be desirable to have lower accelerating voltages. (The 40 kilowatt accelerating potential used in the Calutron is necessary in part to optimize space charge compensation.)

Magnet

The hundred-ton forty-kilowatt magnets were required for the 180°, 120 centimeter radius Calutron in order to give an adequate spacing between the U-235 beams and U-238 beams. This resulted in a system with a separation factor of 20 to 40.

A modern approach to this problem might use a half-toroid tank with the field provided by small permanent magnets or electromagnets.

If a system were designed for lower velocity ions (accelerated by 10 kilovolts instead of 40 kilovolts) a much less intense magnetic field would

suffice. For a constant radius of deflection, reducing the voltage by a factor of four will result in a reduction in the required field by a factor of two.

Another approach which has been successfully used in mass separators for scientific applications is to use a 90° deflection. A 90° sector machine is arranged with the source, the center of curvature of the ion path, and the collector on a straight line. In this geometry both the source and the collector are one beam radius from the edge of the magnetic field. This arrangement permits the use of a smaller magnet.

Manpower Requirements

The procedures required for operation of a systems such as this which involves a "batch" process is intrinsically labor intensive. An essential part of the development will be the training of technicians to service the sources and receivers, to operate the separator and to carry out the necessary chemical procedures on the output. In order to provide one person per unit in three shift operation, about three trained personnel will be required for each unit. Approximately one half the work force will supervise the actual operation of the separators, the rest will provide the other essential services.

Possibility of Proliferation

Two scenarios will be considered, the first to assemble an electromagnetic isotope separation plant large enough to **produce material** for a single explosive each year (15 kilograms of fully enriched material) with a minimum of development and a maximum use of off-the-shelf items. The second is to develop the necessary components and to build a plant of the source capacity which might

produce enriched uranium at a cost comparable to that of the centrifuge or gaseous diffusion processes.

1. "State-of-the-art" Plant

In order to initiate the program several 90° sector electromagnetic separators would be obtained through commercial channels. Possible suppliers include companies in the United States, United Kingdom, Switzerland, the Netherlands, Denmark, Sweden and Germany. These units which cost about two hundred and fifty thousand dollars apiece should have a sector radius of about 60 centimeters, more than 10 centimeter gap, and a magnetic field of more than 8000 Oersted. Ion sources, pump collectors and power supplies would be purchased with each unit. Several high capacity turbine vacuum pumps should be ordered at the same time.

Prototype uranium tetrachloride arc courses and receivers must be fabricated. Published designs are available and would be the basis for this essential development. The first models would be designed to be used with the commercial isotope separators. These tasks must be carried out in a well equipped machine shop.

At the same time that the research isotope separators are ordered, design and construction of a prototype production unit must be started. Nearly all of the features have been described in the scientific literature. Some of the components can be obtained commercially. Others can be copied from the purchased units. For example, it is possible that suitable electromagnets could be fabricated in a plant which manufactures large transformers. The ion source, receiver and the tank might be produced by a factory which produces major electrical appliances. At least two years will be required to fabricate the sources and receivers and develop techniques for their operation with the purchased separators. It is likely-that the prototype of the production unit designed during this period will also be a 90° sector machine. Another year of development will be required before the performance of this unit can be evaluated. It seems probable that this unit would have a few hundred milliampere beam and an efficiency of about 20 percent.

A cost estimate for the Calutron Process was a part of a review of methods for uranium isotope separation which was made by an ad hoc committee in 1972. <u>10</u>/ Using up-to-date costs and incorporating known improvements they predicted costs per gram of U-235 ranging from \$160 (for a beam of 600 milliamperes reached with minimum development) to \$9 for a unit with beam current of the maximum value permitted by ion-ion scattering (40 amperes). The lower cost is more than that for enrichment to weapons grade material by gaseous diffusion. It is likely at the largest beam currents that the enrichment would, in fact, be very low. Also the development costs to achieve the high currents were estimated to be very high.

A minimum total cost of 100 million dollars is estimated for the construction of a plant based on calutron separator technology. It could not be attempted by a country which does not have considerable scientific and industrial resources. The size of the country and the large number of persons involved would make it very difficult to conceal.

2. Advanced Design

In order to make this isotope separation process more attractive than gaseous diffusion or centrifuge it is necessary to develop relatively cheap,

small units which can handle large ion currents. It also will be essential to reduce the cost and complexity of the auxillary operations.

In order to get away from the corrosion, chemical processing and pumping associated with chlorine it may be desirable to develop a substitute for the uranium tetrachloride arc source. A possibility is an electrohydrodynamic ion source which would operate with liquid uranium metal. A singly ionized uranium beam of more than one ampere is needed. It is also important to develop a system with low accelerating voltage so the required magnetic field will be significantly reduced. This may make feasible the use of permanent magnets in a small 90° sector machine. Improved vacuum and geometry of the receiver may result in a rather high source to receiver efficiency. The commercial development of a unit of this type has been proposed by PHRASOR. Although they have apparently done little laboratory work, they estimate a four-year development at a cost of 30 million dollars of a plant to produce annually 30,000 kg of uranium enriched to 3%. $n_/$

In order to attempt this development a staff of at least twenty research physicists and chemists and an equal number of electrical, mechanical and chemical engineers with design experience will be needed. It will be essential to recruit at least one person who has been working on the relevant technology in ion propulsion.

If the goals described above are met, a unit would result which has a one-half ampere beam, efficiency of as much as eighty percent and a separation factor of about 4. It would produce approximately 15 grams per day of a product containing 3 percent U-235. The cost of such a unit might be as low as \$50,000, not counting the research and development costs. A plant based on units with this performance would require 33,000 separator days to produce 15 kilograms a year of U-235 in 3 percent material. (This is 500 kilograms of enriched uranium.) Assuming 333 days of operation per year about 100 Alpha separators will be needed. An additional 50 units would be required for additional stages needed to produce weapons grade material.

The development program will require at least five years and might cost in the neighborhood of several tens of millions of dollars. A minimum cost for a two hundred unit plant might be about fifteen million dollars. It could be built in about two years after development of the prototype enrichment unit.

This development could only be accomplished by industrialized countries with an established scientific and engineering infrastructure.

References

- 1. Hewlett, R. G. and Anderson, O. E., <u>The New World 1939/1946</u>, Pennsylvania State University Press 1962. p. 646.
- 2. Harmatz, B., **Googin,** J. M., Livingston, R. S. and Sprague, T. P., <u>Beta Calutron Performance and the Effects of Recovery Methods</u>, Union Carbide Corporation, Nuclear Division, Y-12 Plant, July 1, 1948 (Y-720).
- Koch, J. (Editor), <u>Electromagnetic Isotope Separators and Applications</u> of <u>Electromagnetically</u> Enriched Isotopes, North Holland Publishing Company, Amsterdam, 1"958, p. 287.
- 4. Alexeff, I., <u>Throughput Limit in Mass Spectrograph-type Isotope</u> <u>Separators</u>, Journal of **Applied** Physics <u>44</u>, 4592 (1973).
- 5. Reference 1, p. 166.
- 6. Reference 1, p. 147.
- 7. Reference 3, p. 137, also <u>Sources and Collectors for Use in Calutrons</u>, USAE T. I **.D.** 5218 (1949).
- 8. Krohn, V. E. and Ringo, G. R., <u>Ion Source of High Brightness Using</u> <u>Liquid Metal</u>, Applied Physics Letters <u>27</u>, 479 (1975).
- 9. Clampitt, R., Harrison, M. F. A., Hotson, E. S., Aitken, K. L., and Jefferies, D. K., <u>Mechanisms of Electrostatic Spraying Colloids and</u> <u>Liquid Caesium</u>, European Electric Propulsion Conference, 3rd., Hinterzarten, West Germany, October 14-18, 1974, Proceedings, p. 118 Deutsche Gesellschaff fur Luft and Raumfahrt, Cologne, 1975.
- 10. Benedict, Manson; Berman, Abraham S.; Biegeleisen, Jacob; Powell, Jack L.; Shacter, John and Vanstram, Paul R., <u>Report of Uranium</u> <u>Isotope Separation Review Ad Hoc Committee</u>, Oak Ridge Operations Office, U. S. Atomic Energy Commission 1972 (ORO-694).
- 11. PHRASOR TECHNOLOGY INCORPORATED, 110 South Euclid Avenue, Pasadena, California, 91101.