

Appendix VI. Dedicated Facilities

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APPENDIX VI-A

DEDICATED FACILITIES FOR THE PRODUCTION
OF NUCLEAR WEAPONS IN SMALL AND/OR
DEVELOPING NATIONS

by

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NOTE: portions of this report are based on material prepared earlier for the Congressional Research Service of the Library of Congress.

I INTRODUCTION

Preventing the spread of nuclear weapons to other nations has been a major policy objective of the United States Government since the end of World War II. This objective has found expression in domestic legislation restricting the export of nuclear technology and materials, and, on the international front, in the establishment of the International Atomic Energy Agency (IAEA) and in the negotiation of the Treaty on Nonproliferation of Nuclear Weapons (NPT).

With the **increasing** pace in the construction of nuclear power plants abroad, considerable attention has been given to the problem of assuring that none of the plutonium produced in these plants is diverted for use in weapons. Indeed, the NPT requires continuing IAEA inspection of nuclear power plants in signatory nations that currently do not have nuclear weapons.

However, the NPT alone cannot prevent the proliferation of nuclear weapons if these weapons can be produced outside the mainstream of the nuclear industry in facilities specifically dedicated to their manufacture. It appears that this, in fact, is the case. As shown in this report,

many nations in the world today, in particular, many small and/or developing nations, are capable of undertaking nuclear weapons programs that can provide them with a small number of weapons in a period of five to ten years.

It is reasonable to assume that any nation which embarked on a program to procure nuclear weapons would attempt to keep this fact a secret as long as possible.

A nation that can suddenly demonstrate a nuclear capability has an obvious advantage over an unsuspecting adversary. At the same time, a clandestine weapons program avoids the recriminations and international political pressures that the nation might encounter if it pursued the program openly.

Throughout the present report, therefore, it will be assumed that any nuclear weapons program must be kept secret. As will be seen, this severely restricts the kind of weapons producing facilities that can be built, their size, rate of production, and location.

II PRODUCTION OF NUCLEAR WEAPONS MATERIALS

The explosive material in an fission nuclear weapon (as opposed to a hydrogen bomb) can be either plutonium-239 or uranium that has been enriched in the isotope uranium-235, or uranium-233. In order to make a nuclear weapon it is necessary, therefore, to procure one or the other of these substances in the necessary amounts. About 5 to 10 kilograms are required for a plutonium bomb; about 15 to 30 kilograms for a U-235 explosive.

Plutonium-239 is not a naturally-occurring substance. It is manufactured from the isotope uranium-238 in an operating nuclear reactor. The newly-produced plutonium-239 is then separated chemically from the uranium. Virtually all power reactors operating in the world today contain large amounts of uranium-238, so that these reactors are automatic producers of plutonium-239. A typical American light water reactor in a nuclear power plant generating 1000 megawatts of electricity produces about 250 kilograms of plutonium-239 per year.

Plutonium-239 was first obtained for use in nuclear weapons in the United States during the Manhattan Project of World War II from specially-designed plutonium production reactors. It appears entirely possible for many small and/or developing nations to obtain plutonium in a similar way, by

building a small plutonium producing reactor and associated plutonium recovery plant.

These matters are covered in Sections 111 and IV.

The enriching of uranium by such a nation would seem to be a more difficult undertaking than the production of plutonium in a small reactor. There are several methods that might be considered for enriching uranium. To date, the most successful method is the gaseous diffusion process, which was ^{also} developed by the Manhattan Project in World War II. This has remained essentially the only source of enriched uranium for military and civilian nuclear programs since that time, both in the United States and abroad. However, gaseous diffusion plants are inherently large structures that utilize a relatively sophisticated technology, much of which remains classified, they require an enormous investment of capital, and consume large amounts of electric *power*. And, of course, they cannot be concealed. The gaseous diffusion route to nuclear explosives is simply out of the question for all but a handful of the largest and developed countries, and will not be considered further in this report.

The use of high-speed centrifuges to separate the isotopes of uranium, a method that was explored during the Manhattan Project but later abandoned, has re-emerged in

the last few years and appears to be more economical than gaseous diffusion. This method is discussed in Section V of this report.

Another method for enriching uranium is the Becker nozzle process. A variation of this method is being used in the Union of South Africa.

However, this method requires a large number of stages (see discussion of stages in Section V) and consumes two and one-half times as much electric power as gaseous diffusion and about thirty times as much as centrifuges. About the only attractive feature of the Becker method is that it is totally unclassified. In any case, this method does not appear to be a reasonable choice for a small and/or developing nation.

A number of other processes for separating uranium are under development that promise to reduce substantially the cost of enriching uranium. Since these have not been demonstrated in practice, they are not available options for small and/or developing nations in the near future.

III PLUTONIUM-PRODUCING REACTORS

As noted earlier, plutonium-239 is produced in any operating nuclear reactor containing uranium-238. The first problem therefore facing any nation embarking upon an illicit nuclear weapons program based on plutonium is to obtain the necessary nuclear reactor. India received a research reactor from Canada, and by introducing their own uranium-238 into that reactor, the Indians were able to produce enough plutonium to make a bomb. The fact that Indian uranium was used to produce the plutonium presumably circumvented in a legal way the safeguards provisions in the Canadian-Indian reactor agreement. In much the same way, Israel procured a small research reactor from France, and according to some reports this reactor has provided the Israelis over the years with enough plutonium for between 10 and 20 bombs.

No doubt, the Indian and Israeli experience will make it more difficult for other nations to obtain reactors in the future, outside of the provisions of the NPT. The question remains, then, how difficult would it be for a

nation, lacking a major technological base, to construct a plutonium-producing reactor on its own.

The problems which must be solved by such a nation in building a production reactor closely parallel those faced by the United States and her allies in the Manhattan Project - with two important exceptions: (1) the necessary reactor technology is highly developed and readily accessible in the open literature, and (2) important materials unavailable at the beginning of World War II can now be purchased on the free market.

The first issue that the nation's leaders would have to settle is the number of bombs that they felt the nation must acquire and the time scale for getting them. These factors determine the size of the reactor - assuming for the moment that only a single reactor is under consideration. It is shown in Annex A that the rate of plutonium production is proportional to the reactor power level. For example, a reactor operating at 25 megawatts (MW) will produce between 9 and 10 kilograms of plutonium per year, enough for one or two bombs. As outlined below, such a reactor can be built and operated at nominal cost, in a relatively short time, with a small number of personnel, and there is at least a fair chance that its existence could be concealed for several years.

A more ambitious program, one which would yield, say,

between 10 and 20 bombs per year would require a reactor operating at 200 to 500 MW. The construction of a reactor of this size would be a major undertaking. It would necessitate a large investment in capital, take a long time to complete, and involve a large number of engineers and construction workers. There is no chance that the project could be kept secret, either during construction or in operation. What is more, while the 25 MW reactor could be built according to plans of a reactor that was actually operated for many years, a good deal of design and engineering would have to be done on the larger reactor, and there could be no guarantee beforehand that the reactor would operate successfully. Finally, there is the question as to what a small and/or developing nation could possibly do with so many bombs. After 10 years of operation the nation could have as many as 200 weapons, far more than needed to obliterate any other small and/or developing country. Yet, even then, it would be difficult to abandon such an enormous reactor and its associated plutonium recovery plant into which the nation had poured so much wealth.

In short, it does not appear reasonable to assume that the nation in question would attempt to build a large

reactor, and if they elected to do so there is little reason to expect that they would achieve their objective.

One additional factor should be mentioned. If the nation were successful in constructing the smaller reactor and required a higher plutonium production rate, there is nothing to prevent it from building a second or third reactor of the same type. Having built one reactor, it would be an easy matter to build carbon copies.

Since the nation in question would not have access to enriched uranium- if they were able to produce enriched uranium they would never bother to produce plutonium - the reactor would have to be fueled with natural uranium. This automatically places restrictions on the type of reactor that can be built. It either has to be moderated with graphite or with heavy water- these are the only practical moderators that can be used in a natural uranium fueled reactor.

However, heavy water has drawbacks. It is expensive and obtainable from only a handful of countries. Attempts to purchase it would immediately reveal an intention to undertake a nuclear program, since the necessary amounts of heavy water could have no other possible application. The construction of a heavy water production plant requires relatively sophisticated technology, and would be a

difficult task for a small and/or developing nation. At the same time, the technical problems of designing, building, and operating a heavy water reactor are somewhat more formidable than for a graphite reactor with the same plutonium production capacity. For one thing, the reactor vessel, with its many piping connections and instrumentation and control penetrations, must be leak tight to prevent the loss of heavy water. This requires high-(grade manufacturing skills presumably not present in the nation in question. While the Indian and Israeli reactors are of the heavy water type, both were obtained from nations having long experience in fabricating heavy water reactor systems. It may be concluded, therefore, that the plutonium producing reactor would be graphite moderated.

It must be next decided how the reactor would be cooled. Among several possibilities, only air and water are practical choices for the reactor coolant. Other coolants, such as CO₂ or helium require closed cycle operation, an unnecessary complication for a reactor operating at low power levels. Water-cooled reactors are capable of higher plutonium production rates than air-cooled reactors of the same size, because **water** has better cooling properties than air. However, a water-cooled reactor is more difficult to design, construct,

and maintain. Water is more corrosive than air so the fuel elements must be carefully fabricated. The safety analysis of such a reactor is also more involved than for a comparable air-cooled reactor. Both the United States and the U.S.S.R. use water cooling in their production reactors, but these are, after all, major technological nations. On the other hand, Great Britain and France used air, at least initially, in their reactors. It seems reasonable, therefore, to assume that a small and/or developing nation would base its plutonium production program on the construction of at least one natural uranium fueled, graphite-moderated, air-cooled reactor. Another factor favoring this choice of reactor is that the design parameters for successful reactors of this type are freely available in the open literature.

The first natural uranium, graphite reactor was the so-called CP-1 pile, built by Enrico Fermi and his associates in Chicago in 1942. This was also the world's first reactor.* The CP-1 was dismantled after only 4 months of operation and reconstructed as the CP-2 reactor, another experimental system of Fermi design. The first reactor to operate at a significant power level - about 2 MW - was the X-10 reactor at Oak Ridge. The purpose of this reactor was to provide plutonium for the startup phase of the plutonium chemical separation plant at Hanford,

***Except** for the naturally **occurring** Oklo Reactor.

Washington, prior ⁰ to the operation of the Hanford production reactors .

The X-10 reactor also served as the basis for the design of the original Brookhaven Graphite Research Reactor (BGRR), which operated at Brookhaven National Laboratory from 1948 until 1957, when its natural uranium fuel was replaced by enriched uranium. The reactor was shut down in 1969 and decommissioned shortly thereafter. While the BGRR was used primarily for research, about 9 kilograms of plutonium-239 were produced per year in the reactor - sufficient for the fabrication of one or *two bombs* per year, when process losses are taken into consideration.

The construction of either an enlarged X-10 reactor or a simplified version of the BGRR would appear to be the most logical way for a small and/or developing nation to initiate the production of plutonium. Since the BGRR has been more widely discussed in the open literature, only this reactor will be considered in the present report.

Before doing so, however, it should be pointed out that the plutonium produced in a BGRR in the first few years of operation is almost entirely pure plutonium-239. Very little of the plutonium-239 is converted into the heavier isotope plutonium-240. After one year of operation, for example, less than one-half of one percent of the

plutonium in the BGRR is plutonium-240. This is in marked contrast to the plutonium produced in power reactors. Because these reactors operate at much higher neutron fluxes, a substantial amount of the plutonium-239 is converted to plutonium-240. The plutonium in the fuel discharged from a power reactor following a year of operation is typically 10 to 20 percent plutonium-240.

The plutonium-240 content is an important consideration when plutonium is to be used for the manufacturing of nuclear weapons. This is because the spontaneous fission rate of plutonium-240 is so high. The neutrons emitted in spontaneous fissions can lead to the preinitiation of the explosion. In short, the plutonium produced in the BGRR is excellent bomb material; the plutonium produced in an ordinary power reactor is not as good. (See Chapter VI of Vol. I "Nuclear Fission Explosive Weapons".)

Years ago, when the BGRR was in operation at Brookhaven, the AEC was always pleased to learn that one of the reactor's fuel elements had sprung a leak, for this was the only time that the BGRR fuel was removed from the reactor for reprocessing. Leaking fuel meant high grade plutonium for the AEC'S weapons program.

The Brookhaven Graphite Research Reactor

The BGRR consisted of a 25 foot cube of graphite, penetrated by a square, 37 x 37 array of 1368 three inch diameter air channels that contained the fuel assemblies.* For efficiency in cooling the fuel, the graphite cube was split in the middle, and the two halves were separated by a 7 cm air gap. Cool air entered the reactor via this gap, passed through the air channels in both directions to the opposite surfaces of the reactor, and was then exhausted via fans to a 320 foot stack. By introducing the air at the center of the reactor, the pumping power required to move the air was reduced by a factor of eight. This feature of the BGRR was one of the principal design improvements over the X-10 reactor.

The air left the reactor at a temperature of up to 220°C, depending on the reactor power level. Since it is difficult to pump heated, low density air, the air leaving the reactor was passed through a cooler, where its temperature was reduced about a hundred centigrade degrees and its density increased. This saved on the size and operating costs of the fans.

*The central channel contained a removable **plug** for **experimental** purposes. The **number** of channels was therefore **1368**, not 1369.

The fuel for the BGRR was in the form of cylindrical uranium slugs 1.1 inches in diameter and 4 inches long. These slugs were loaded into hollow aluminum cartridges, 33 to a cartridge, to make the fuel assemblies. The cartridges had six aluminum fins running their full length that supported the fuel in the center of the air channels and increased the heat transfer area from the fuel to the air. One fuel assembly was used per air channel on either side of the central air gap.

It was not necessary, however, to load all 1368 fuel channels in order to operate the reactor. The BGRR actually went critical with only about 870 loaded channels. The other channels were available to provide additional reactivity when neutron-absorbing experiments were introduced into the core.

Control of the BGRR was accomplished by the motion of horizontal control rods that entered the reactor from two adjacent corners. Supplementary emergency shutdown control could be obtained by the mechanical dumping of boron shot into holes provided for this purpose at the top of the reactor. The BGRR was equipped with an array of radiation detectors and system monitoring devices that provided operating data to a central control room.

The BGRR was housed in a large and attractive brick

building that also contained a number of offices, laboratories and seminar rooms. Since the BGRR was built primarily for research, heavy platforms were erected at several levels across the face of the reactor to support experimental equipment. The overall cost of the BGRR facility was \$25.5 million when it was built in 1948. Of that total, only \$16.7 million was attributed to the reactor and reactor-related equipment.

A tabulation of the principal characteristics of the BGRR is given in Annex B.

A Simplified BGRR for Plutonium Production

It is not necessary to duplicate the BGRR in detail in order to produce plutonium at the same rate as it was produced in that reactor. Simplifications in the BGRR design would permit the building of a plutonium production reactor that would be cheap and reasonably reliable, and a reactor whose engineering would require the talents of only a small cadre of conventionally trained engineers. The procurement or fabrication of certain key components would be the most difficult problems that a small and/or developing nation would have to solve. These components are as follows:

Fuel. The order of 75 tons of natural uranium metal would be needed to fuel the reactor. The actual amount

of fuel would depend on the design of the reactor and the nature of the materials used to build it. Refined uranium directly suitable for reactor fuel apparently is available only from highly industrialized nations, where it is a controlled substance and not easily obtained on the open market. It cannot be exported from the United States, for example, without a license from the nuclear Regulatory Commission.

Nevertheless, a great many nations in the world possess indigenous sources of uranium ore. A table of 1975 estimates of non-U.S. uranium resources is given in Annex C. As seen in the table, nations such as Algeria and Argentina have estimated resources in excess of 30,000 tons of U_3O_8 recoverable at up to \$30 *per* pound. Only the order of 100 tons of U_3O_8 is needed to obtain 75 tons of uranium metal.

The processing of uranium ore and its reduction to metallic uranium is not a difficult undertaking for a trained metallurgist. The necessary directions are in the open literature. It would require learning experience for a metallurgist who was a novice in uranium metallurgy. The problem would be much simplified if the nation in question were already a producer of U_3O_8 . In any event, a well-trained metallurgist should be capable of designing and setting up a small uranium mill in 12 to

18 months which would produce 75 tons of satisfactory uranium metal in another year. The required equipment and supplies are generally available on the world market.

The cost of producing 75 tons of uranium metal is difficult to estimate, since labor, raw material, and capital costs vary so widely from nation to nation. There is no market in natural uranium in the United States at the present time. The Federal Register price of natural uranium hexafluoride is \$25.39 per kilogram. The cost of uranium metal in this country is therefore about \$25 per kilogram, which is not a free market price. U_3O_8 for future delivery is now being quoted as high as \$40 per pound, which would give the uranium a value of over \$100 per kilogram excluding processing costs. Using the nominal value of \$25 per kilogram, the total cost of 75 tons of uranium comes to about \$1.7 million.

Fuel Assemblies. The fabrication of fuel cartridges similar to those used in the BGRR, with their six aluminum fins running the length of each cartridge, might well pose a serious manufacturing problem to a small and/or developing nation. However, the fins are not entirely necessary for the operation of the reactor. A satisfactory and far simpler fuel assembly could be made by merely inserting uranium metal slugs into a hollow

aluminum tube sealed at both ends. These tubes could then simply be placed along the bottom of the air channels in direct contact with the graphite, a procedure followed in the x-10 reactor, or supported in the center of the channels on suitably machined pieces of graphite (which is easily machined), as was done in the British Windscale plutonium production reactor. The use of this type of fuel assembly would require modifications in the rate of air flow through the reactor, the operating temperature of the fuel, or the reactor power level from the values of these parameters in the BGRR. The necessary adjustments could easily be determined however. The fabrication of the fuel assemblies would require about 6 months, starting with raw uranium metal.

Graphite. The graphite used in nuclear reactors must be of high purity. In particular, the concentration of the impurity boron must be as low as possible. The procurement of reactor-grade graphite was one of the first problems that had to be solved in the Manhattan Project. Although graphite occurs abundantly in nature, all commercial graphite is manufactured artificially from petroleum coke or coal tar pitch.

Graphite of the type used in the BGRR is currently available from a number of companies here and abroad, although the Department of Commerce licenses the export

of reactor grade graphite from the United States. One U.S. manufacturer recently entertained inquiries from Argentina, Brazil, **and** the U.S.S.R.

The Union Carbide Corporation sells reactor grade graphite for approximately \$2 per pound. If the full 700 tons of graphite required to duplicate the BGRR were purchased from this company, the total cost would be \$2.8 million.

However, as already noted, a production reactor does not have to be as large as the BGRR. Instead of building a 25 ft cube, a somewhat smaller cube, say 21 ft on a side, would probably do just as well. The total amount of graphite required in this case could be as little as 415 tons and cost \$1.7 million.

It should be mentioned that the processes for manufacturing reactor-grade graphite and electrode graphite are essentially the same. Facilities used for producing electrode graphite can easily be converted to the production of reactor-grade graphite. To obtain reactor-grade graphite it is most important to start with clean raw materials and to use somewhat higher temperatures. Electrode graphite manufacturing plants are located throughout the world. Union Carbide Corporation, to name but one organization, has subsidiaries manufacturing

electrode graphite in Brazil, Canada, Italy, Japan, Mexico, Puerto Rico, South Africa, Spain, and Sweden.

Graphite is easily machined. Its sizing and fabrication for use in a reactor presents no problems.

Air-Moving Equipment. **blowers of a conventional** type are suitable for moving the air through a BGRR system. This equipment is readily available throughout the world. If purchased from American manufacturers, the fans and motors required to provide a flow of 300,000 cubic feet per minute would cost about \$180,000 at today's prices. The associated ducting, and intake and exhaust structures would present problems. However, all necessary materials are available and could be fitted or fabricated with patience and skill.

Controls. The control of a natural uranium graphite reactor is extremely simple. There is very little excess reactivity in such a reactor so that whatever transients do occur in the reactor have long periods and are easily controlled. The control rods and their drives need not have the short response times required of other types of reactors, e.g., water-cooled power reactors. The **rods** and drives could be fabricated from materials on the open market.

Instrumentation. The elaborate instrumentation found in American reactors would not be necessary for a small production reactor. A few neutron and γ -ray detectors, a large variety of which can be purchased in many different countries, would suffice for the reactor. Monitors for air, graphite, and fuel temperatures are equally available.

Balance of Plant. As noted earlier, the BGRR reactor building was a well-built and attractive structure. The building for a production reactor need not be so ambitious. A simple industrial structure, steel-framed with corrugated siding could be built at no more than \$3 per ft³. A modest cubical building 55 ft on a side would then cost about \$0.5 million. This is the cost if built in the United States. Overseas costs could well be much **less**.

The floor of the building would have to support about 5000 tons over an area of 2000 ft², for a loading on the order of 2 tons per ft². This is not an especially large floor loading and could be satisfied with a slab of reinforced concrete between 2 and 3 feet thick. At \$100 per cubic yard, a square slab 55 ft on a side could be built for less than \$35,000, U.S. prices. Presumably a deep water pool would have to be added along one side of the reactor to receive and store spent fuel until it could be processed for plutonium recovery.

Overall Costs. Estimated costs of the principal materials and structures required for a small production reactor are given in the following table. These costs are based on current U.S. prices, and as such they may have only the roughest applicability to another nation.

Table 1

Costs of Production Reactor Components

<u>Item</u>	<u>Approximate Cost (\$ million)</u>
uranium	1.7
graphite	2.8
air equipment and ducts	0.5
control	0.2
instrumentation	0.2
building and foundation	<u>0.6</u>
Total	6.0

Table 1 does not include the labor costs associated with fabricating the fuel assemblies from the raw uranium metal, Constructing the reactor within the building, connecting the ducts and air-moving equipment, and introducing the control and monitoring systems. Such costs are difficult to estimate since the cost of labor varies so widely from country to country. If 100 workers (not producing uranium

or erecting the building - this labor is included in the components costs) worked for 3 years at \$10,000 per year, their total wages would amount to \$3 million. Using this as a rough estimate, the reactor could be built for about \$10 million - probably correct within a factor of 2.

Personnel Requirements. As pointed out repeatedly in this report, it is not necessary to design the reactor from scratch. All of the essential design parameters are in the open literature. High-level research and development personnel are not required. Only a handful of professional engineers would suffice to design and oversee the construction of the facility. The following is a list of minimum professional personnel requirements.

Table 2

Professional Engineering Requirements

<u>Type of Engineer</u>	<u>Number</u>	<u>Utilization</u>
civil-structural	1	structures, reactor building
electrical	1	<i>control</i> , instrumentation, circuitry
mechanical	2	heat transfer, mechanical devices
metallurgist	1	uranium production
nuclear	3	design theory, nuclear measurements, reactor heat transfer

Time Schedule. In any major project, the proper scheduling of design, procurement, and construction activities permits the simultaneous accomplishment of the required tasks. In the case of the reactor under discussion, the reactor plant parameters could be finalized and purchase orders placed while the uranium mill is being prepared. The reactor could then be erected at the same time as the fuel assemblies are being fabricated. This phase of the project would probably take about 2 or 3 years, depending on the availability and skill of the work force. The reactor could be ready for production operation 4 years from the beginning of the project.

This is probably an overestimate of the time required for the project. The X-10 reactor in Oak Ridge went into operation in November 1943, less than one year after the world's first reactor went critical in December 1942, and it was operated at almost 2 MW in May 1944. The entire BGRR project, which was not a military project, took only about 3 years.

In any case, sufficient plutonium for at least one bomb would be present in the reactor fuel one year later. A simplified scheduling diagram is shown on the next page.

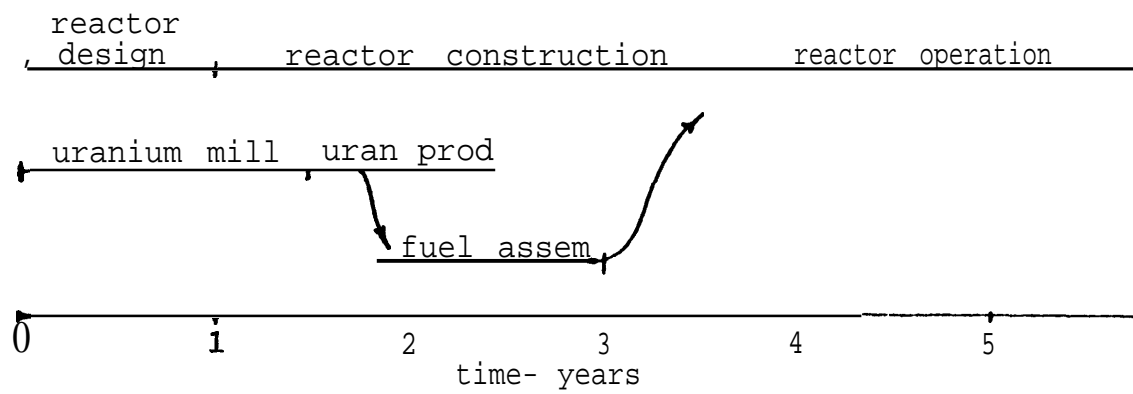


Figure 1. Schedule for design, construction, and operation of simplified BGRR.

IV RECOVERY OF PLUTONIUM FROM REACTOR FUEL

If it is desired to fabricate weapons as quickly as possible, then the fuel from the production reactor would probably be removed for reprocessing after it had been in the reactor for a period of approximately one year. The concentration of plutonium would then be about 9 kilograms in 75 tons of fuel or about 120 grams *per* ton. The problems associated with extracting this plutonium from the fuel and preparing it for fabrication in a weapon are the subject of the present section.

These problems are not insurmountable, even for a small and/or developing nation. Indeed, such a nation could

build a small reprocessing plant and recover essentially all of the plutonium-239 produced in a BGRR-type reactor. The final step of preparing this material for a weapon can also be readily accomplished, as has been amply discussed in the literature.

Some Problems in Fuel Reprocessing

In any case, a plutonium recovery plant must be designed and operated with some care. The raw fuel, when it is first discharged from the reactor, is highly radioactive, largely due to the activity of the fission products. Even if the fuel is allowed to cool for a nominal period of 120 days, during which time the activity decays by a factor of 100 or more, the total radioactivity is still about 45,000 curies per ton or 0.05 curies per gram of fuel. This means

that the chemical processing of the fuel must be carried out remotely, in a shielded cell, at least up to the point where the fission products are removed.

It should be noted, however, that the radioactivity of the BGRR fuel is much smaller than that of a typical power reactor. The activity of power reactor fuel after a cooling-off period of 120 days runs between 2 and 3 million curies per ton, a factor of about 50 times higher than BGRR fuel. Considerably more precautions must therefore be taken in reprocessing power reactor fuel than fuel from a BGRR.

Nevertheless, the chemical methods described below provide almost complete separation of the fission product activity from the plutonium and the uranium remaining in the fuel. It is a remarkable fact that where these methods are used to recover the uranium as well as the plutonium, the activity of the recovered uranium is no greater than that of ordinary, natural uranium, which can safely be held in the bare hands.

The separated plutonium is also free of fission products and it is only mildly radioactive itself, so that it too could be handled like uranium were it not for the possibility of inhaling plutonium-bearing particles. Such airborne particles are extremely dangerous. It is approved practice, therefore, at least in the United States, for all manipulations of plutonium to be carried out in a protected atmosphere. While such an elaborate precaution is not entirely necessary, as

indicated below, it is not difficult to arrange.

Finally, there is the danger of an accidental criticality, that is, the possibility that a critical mass of plutonium may accidentally be assembled. Only a few hundred grams of plutonium can become critical in the proper liquid environment. However, the methods for calculating critical concentrations are given in all nuclear engineering textbooks and these concentrations are widely tabulated (see especially Wick in the References). Procedures for avoiding accidental criticalities can easily be adopted.

Plutonium Recovery Processes

Several processes have been developed over the years to remove the fission products and recover the plutonium and uranium from irradiated fuel. These processes are thoroughly described in journals, textbooks, and in other open literature. The first to be perfected was the so-called bismuth phosphate process, which was the source of plutonium at the beginning of the u.s. nuclear weapons program. This process was later replaced by a solvent extraction process, first using the chemical methyl isobutyl ketone as solvent - this was the so-called Redox process - and somewhat later with the solvent n-tributyl phosphate (TBP), which is the basis of the Purex process. So far as is known, virtually all re-processing plants that have been built in the world

since the 1950's are based on the Purex process.

Solvent extraction processes rely on the following experimental facts. Uranium and plutonium can exist in a number of valence (oxidation) states, and because of differences in their oxidation and reduction potentials it is possible to oxidize or reduce one of these elements without disturbing the other. Furthermore, compounds of these elements in different states have different solubilities in organic solvents. For instance, in their 4^+ and 6^+ states the nitrates of both uranium and plutonium are soluble in certain solvents, while in the 3^+ state these compounds are virtually insoluble in these same solvents.

Solvent extraction therefore involves three critical steps: (1) separating the uranium and plutonium from the fission products by extracting the first two into the appropriate solvent, leaving the latter in aqueous solution; (2) reducing the oxidation state of the plutonium to 3^+ so that it is no longer soluble in the solvent; and (3) back-extracting the plutonium into aqueous solution.

A simplified flow diagram for the Purex process is shown in Figure 2. The batch of fuel to be processed is first dissolved in a concentrated solution of nitric acid. The fission product gases, especially the noble

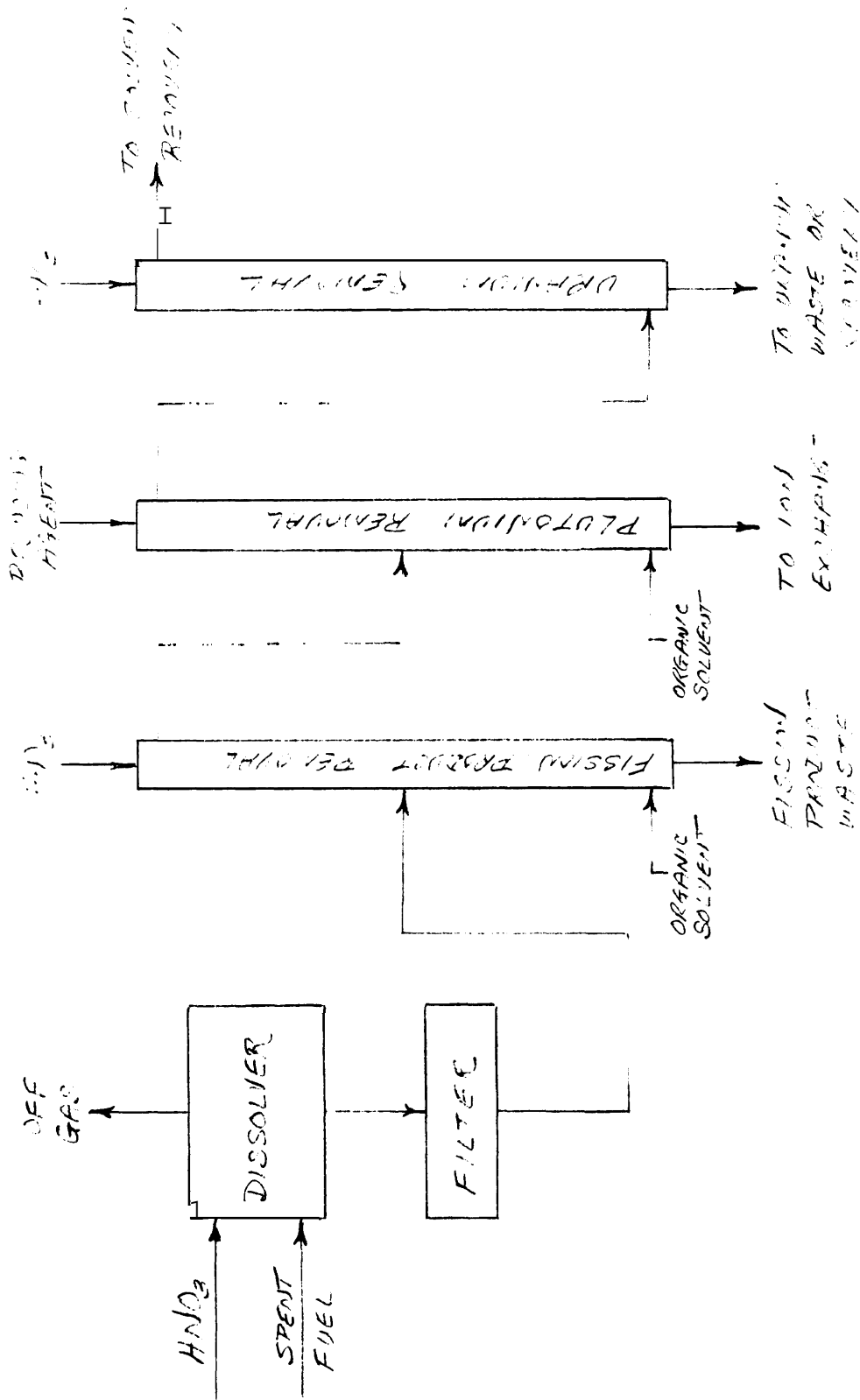


Figure 1. Simplified flow diagram for Purex process.

gases, that had been trapped in the fuel, come out of solution at this point. The release of these gases is the major source of radioactive effluent from a reprocessing plant. The aqueous solution of uranium, plutonium, and fission products, after passing through a filter to remove undissolved remnants of the fuel assemblies, then enters at the middle of the first extraction column. As the organic solvent, TBP diluted in kerosene, flows up the column, it absorbs the uranium and plutonium out of the solution. At the same time, more nitric acid enters from the top of the column to scrub the rising solvent of any fission products it may have picked up. The organic solution which leaves the top of the column contains essentially all of the uranium and plutonium and a trace of fission products, whereas the aqueous solution at the bottom has most of the fission products and very little uranium and plutonium.

The Organic solution passes next into a second column where it counterflows against a dilute solution of chemical reducing agent (a ferrous compound is often used) which reduces the plutonium to the 3^+ state, while leaving the uranium in the 6^+ state. Since the plutonium is now no longer soluble in the TBP, it passes into the aqueous solution before it leaves the column.

The uranium is stripped from the organic solvent in a third column, where the uranium passes into a counter-flowing stream of dilute nitric acid. The solvent leaving the top of the column, from which most of the plutonium, uranium, and fission products have now been removed, is piped to a recovery plant for purification and reuse. The uranium exits the column in aqueous solution.

At this point, the designs of fuel reprocessing plants tend to diverge. If the uranium is enriched in uranium-235, as it is in all American power reactor fuel, then the uranium solution is passed through additional cycles of the Purex process for the purpose of reclaiming the uranium in a highly purified state. With the natural uranium fuel of the BGRR, it is questionable whether recovery of the uranium makes sense, because the spent fuel is somewhat depleted in uranium-235. Whether or not the uranium would be recovered would depend on how long the fuel had been in the reactor and the extent and reliability of the uranium supplies available.

The plutonium solution can either be put through further Purex cycles or, what is preferable, the plutonium can be purified and concentrated through the use of the process of ion exchange. This process involves passing the solution into an ion exchange resin and then

eluting the plutonium with dilute nitric acid. The concentration of the purified Plutonium can then be increased by partially evaporating the solution, care being taken not to approach criticality conditions. This is the usual form of the plutonium output from a fuel reprocessing plant - a highly purified solution of plutonium nitrate.

It is not a difficult problem to reduce the plutonium nitrate solution to a form for making nuclear weapons. Both metallic plutonium and plutonium oxide can be used in a weapon, although the metal is preferable. To obtain the oxide, sodium oxalate, a common chemical, is added to the nitrate solution. This forms plutonium oxalate which is insoluble and precipitates from the solution. The plutonium oxalate, separated from the solution by filtration, is then heated in an oven which yields the oxide PuO_2 .

To produce metallic plutonium, the oxide is heated in the presence of hydrogen fluoride and oxygen which gives plutonium tetrafluoride. This is then reduced by calcium to yield the metal. The procedures for producing the metal and fabricating it into desired forms are fully described in the references (see especially Wick) .

A Small Plutonium Recovery Plant

It is clear from the foregoing remarks that the chemical engineering required for the recovery of plutonium from spent uranium fuel is relatively simple. The facts, mentioned earlier, that the fuel is radioactive, that plutonium in some forms is highly toxic and capable of going critical leads to some, but not insurmountable problems in the design of a recovery plant.

In any event, designs of such plants can be found in the open literature. For example, the plans and specifications for the Allied General Nuclear Services (AGNS) plant in Barnwell, South Carolina, have been widely distributed to the public in connection with the licensing of this plant and are available in NRC Public Document Rooms.* Furthermore, they can also be purchased

*All the plans and specifications for the AGNS plant have been made public except for the details of three devices: the electrochemical plutonium purifiers, the fuel dissolvers, and the mechanical shear. These plans were retained by the AGNS designers as company confidential and provided to the NRC as black box submittals. Nothing about this plant has been classified on the grounds of national security.

from the National Technical Information Service. AGNS is a large plant with a through capacity of 5 tons of fuel per day. Considerable scaling down of this plant would be necessary for the purpose of reprocessing BGRR fuel.

The Phillips Plant. Plans and specifications for a more appropriate, smaller plant are also available, however. In the late 1950's the Phillips Petroleum Company undertook a feasibility study of a small reprocessing plant designed to handle spent fuel from Commonwealth Edison's Dresden-1 plant, then scheduled for operation in 1960. Phillips issued a report on this study in 1961 (see References), and it was later discussed in an article appearing in Nucleonics Magazine. Although some chemical/nuclear engineers have expressed skepticism about the workability of the Phillips plant, because of its compact design and high degree of automation, it nevertheless can be viewed as an excellent starting point for the design of a reprocessing facility in a small and/or developing nation.

The Phillips report contains detailed drawings of every component of this plant. One of the striking features of the plant is its small size. With the exception of storage areas for raw materials and radioactive wastes, the whole plant is enclosed by a 65 ft x 65 ft building of standard construction. The main process

equipment is so small - for instance, the first extraction column is a pipe only 2 1/2 inches in diameter and 12 feet long - that all this equipment can be fitted into a single underground cell 12 ft square. The tail end of the plutonium recovery is carried out in a **simply-constructed** hood 3 1/2 ft wide, 12 ft long, and 12 ft high which contains three ion exchange columns, several small holding tanks, and an area for loading and weighing the product.

The plutonium output from this plant is in the form of plutonium nitrate solution. No provision is made for converting the nitrate to metallic plutonium, since this was not the purpose of the Phillips plant. The plant was designed primarily to recover enriched uranium from the fuel and separate out the fission products for disposal. Equipment to produce the metal would have to be added.

Simplifying the Phillips Plant. A number of simplifications in the Phillips plant are possible when the plant is designed for the sole purpose of recovering plutonium from BGRR fuel. In particular, the so-called head end of the plant, that portion of the plant where the fuel is dissolved into solution, need not be as complicated as in the Phillips plant. Head-end problems, according to the Phillips report, were the most formidable in designing that plant. This is because the

Dresden fuel consists of bundles of fuel rods, each rod being a hollow zircaloy tube filled with uranium dioxide pellets. The pellets fit so tightly in the tubes that they do not fall out on their own when the tube is opened and turned upside down. It was necessary, therefore, to design an elaborate mechanical/chemical procedure for dissolving the uranium - a procedure, incidentally, that is similar to that used in the AGNS plant.

Head-end preparation of BGRR fuel is much less complicated. Since the fuel will be removed so much sooner, in terms of fuel burnup, from the BGRR than it is from Dresden, the uranium will not have had an opportunity to swell within its aluminum cladding. The uranium slugs can simply be dumped (remotely) into the dissolving tank.

As noted earlier, when the fuel dissolves in nitric acid, radioactive fission product gases which had been trapped in the fuel are released and bubble to the top of the dissolving tank. In the Phillips plant, it was proposed to separate out the rare gases and store these permanently underground. With the BGRR fuel, however, the activity of these gases is so small that they can be exhausted directly to the atmosphere - a practice followed in all currently operating reprocessing plants. For example, the total activity of krypton-85 in a full load

of BGRR fuel which has been in the reactor for one year is only 4000 curies, which in the course of reprocessing the fuel would be released from the dissolving tank over a period of a year. This is not an excessively high release rate for this isotope.

There are other simplifications to the Phillips plant. For one thing, as already mentioned, the BGRR fuel is less radioactive by a factor of 50 than typical (e.g., Dresden) power reactor fuel. This means that the concrete shielding wherever it is called for in the Phillips plans can be reduced in thickness. Provision for the storage of fission products need not be as elaborate, since their activity is so much smaller. Finally, all of the process equipment for purifying the uranium can be omitted if the uranium is not recovered and recycled.

Availability of Materials. All of the equipment and supplies required to build and operate a plutonium recovery plant are generally available on world markets. There is no single component which is so exotic that it can only be obtained from a single source. The solvent extraction columns can either be purchased on the open market or fabricated from standard piping. So can the ion exchangers. The resins used in these columns are standard Dow-Chemical type resins that are used for water treatment and other purposes. Automatic valves, ventilation equipment,

flow meters, television monitors, are all standard. None of the necessary chemicals are out of the ordinary. The hood for purifying the plutonium would probably have to be homemade, but this is not a major undertaking. In short, many small and/or developing nations can procure the materials necessary to construct and operate a small plutonium recovery plant.

Economics. The estimated base plant cost (labor and materials) of the Phillips plant was estimated to be \$2,245,200 in 1960 dollars. When fees, taxes, and startup costs were included, the total price tag came to slightly over \$4 million.

This figure cannot be taken seriously, however, based on experience with reprocessing plants that have actually been built. The General Electric plant in Morris, Illinois, a one ton throughput per day plant, was estimated at \$17 million. The cost of the completed plant was \$64 million. When it was found that the plant did not work, another \$120 million was estimated to be required to put it in order. The 5 ton per day AGNS plant was originally costed out at \$70 million. The actual cost (with an output of plutonium nitrate, as planned) was \$200 million, and additions (e.g., nitrate to oxide conversion facility and waste solidification facility) that may be required because of new licensing regulations may add \$300 or \$400 million to the price.

Thus , it is exceedingly difficult to estimate the cost of a reprocessing plant. It is interesting to note, however, that in the breakdown of costs given in the Phillips ~~report~~^{the}, major cost of that plant was in the concrete (and its pouring), which was present in abundance because of the high activity of the Dresden fuel. This fact has been confirmed by officials responsible for the design of the AGNS plant - concrete is the next expensive single item in the plant. Since the BGRR processing plant would have so much less concrete, the cost would be significantly reduced. The other simplifying; features of the plant described earlier also lead to reductions in plant cost.

It would appear, without making a detailed cost analysis, that a plutonium recovery plant of the type discussed in the present report could be built in the United States for a cost of well under \$25 million, 1977 dollars. Cost in other countries would vary and conceivable could be much less. However, whether the actual cost turned out to be \$25 million or twice that amount (there is no chance it would be ten times this amount - that would be the cost of AGNS, the biggest plant in the world) the fact is that this is a relatively low figure, even when combined with the cost of the reactor, compared to the usual military budget of most nations.

Personnel Requirements. As in the construction of the BGRR described earlier, high-level research and development personnel are not required to build a plutonium recovery plant, since what is necessary is largely a matter of following and/or modifying established designs. Many of the same technical personnel involved in the reactor project could be utilized for the plutonium plant. This would make good sense, because the plant would necessarily be located adjacent to the reactor, and would undoubtedly be built during the same time frame. The following is a list of minimum professional personnel requirements.

Table 3

Professional Engineering Requirements

<u>Type of Engineer</u>	<u>Number</u>	<u>Utilization</u>
chemical	2	process design, construction
civil-structural	1	structures
electrical	1	control, instrumentation circuitry
mechanical	1	mechanical devices
metallurgist	1	plutonium preparation
nuclear	1	shielding, criticality

V URANIUM ENRICHMENT BY GAS CENTRIFUGES

A standard method for separating particles of different masses is by centrifuging. This procedure has been used routinely for decades in biology and medicine to fractionate blood and other biological materials. The material to be separated is placed in a suitable vessel and this is rotated at great speed. The rotation creates what, in effect, is a strong gravitational field, and, as a result, the heavier particles tend to move to the periphery of the vessel while the lighter ones remain near the center.

The fact that gravitational or centrifugal fields could be used to separate isotopes was first pointed out by Lindemann and Aston in 1919. It was twenty years, however, before such separation was successfully obtained. This was achieved by J. W. Beams and his coworkers at the University of Virginia, using a specially-designed centrifuge

The possibility of obtaining highly enriched uranium for nuclear weapons by centrifuging was recognized by Beams immediately after the discovery of fission. Indeed, according to the Smyth Report on the Manhattan Project, "for a long time in the early days of **the project, the gaseous** diffusion method and the centrifuge method were

considered the two separation methods most likely to succeed with uranium."

The first attempt **by** Beams to separate the isotopes of uranium, which was held up until late in 1940 because of the unavailability of uranium hexafluoride (UF_6), was an immediate success. Subsequently, it was decided to build a small pilot plant at the Bayway, New Jersey, laboratory of the Standard Oil Development Company using 24 centrifuges designed and built by the Westinghouse Electric Company. However, only two machines were actually delivered to Bayway before the entire centrifuge project was scrapped at the end of January 1944. Nevertheless, one of these machines was operated successfully for a period of 99 days, and it yielded the degree of separation that had been predicted theoretically.

Figure 3 shows a diagram of the Westinghouse short-bowl centrifuge. The bowl or rotor was 42 inches long and 7.2 inches in diameter and rotated at a rate of 28,200 rpm. This was above the critical speed for the vibration of the rotor. A model of a machine with a 132 inch rotor was also built and tested in 1943.

During the decade following World War II, the centrifuge method for separating isotopes was largely

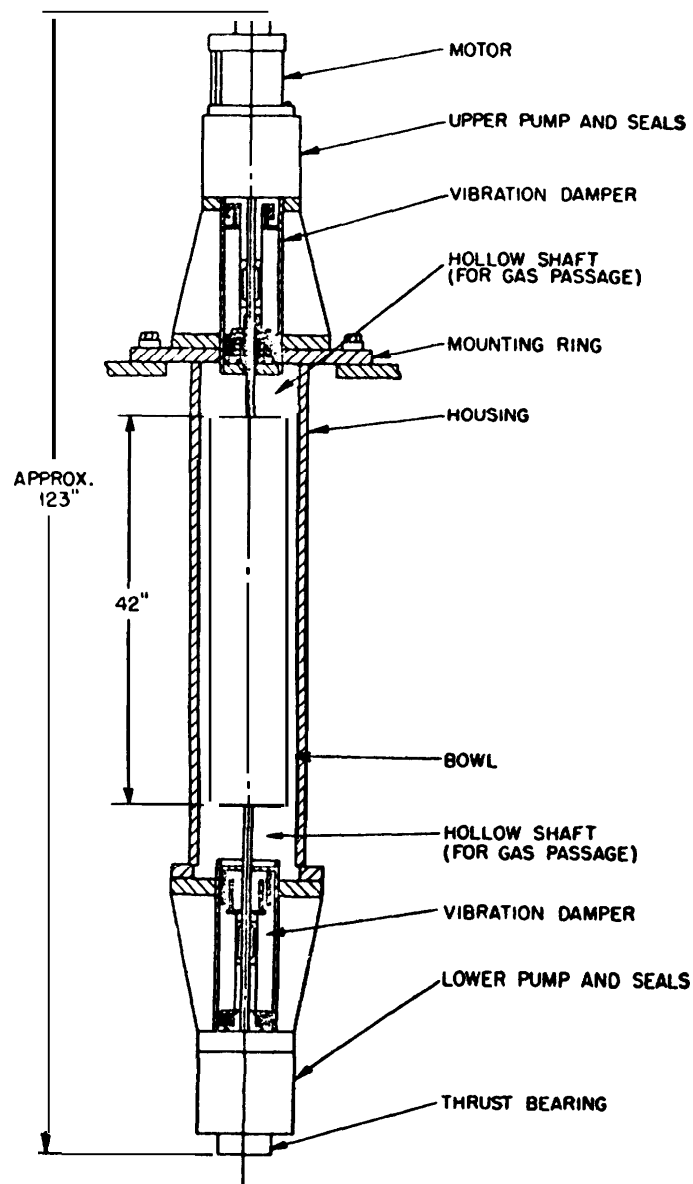


Figure 3. Manhattan Project centrifuge. (From J. W. Beams, see References.)

forgotten in the United States, except for a small project at the University of Virginia. However, work on centrifuges was continued in Germany and also in the Soviet Union. The Soviet team included a number of German scientists that had been captured at the end of the war. Prominent among this group was G. Zippe, who made a number of significant improvements on centrifuge design. Shortly after Zippe had been repatriated from the Soviet Union, he was invited to the University of Virginia to repeat the experiments he had carried out with the Soviets. He began work on a new centrifuge in August 1958 and it was completed in June 1960, when he was repatriated for the second time.

The Zippe machine has evidently been the basis for many of the recent developments in centrifuge technology. A schematic drawing of the Zippe short bowl centrifuge is shown in Figure 4. The centrifuge rotor, O, is 3 inches in diameter and 13 inches long. It spins on a thin, flexible steel needle, which is centered in a depression in a hard metal plate, P, whose lateral motion is damped in oil. Rotational motive power is obtained from the electric motor, M, the armature of which is the steel plate, N, fastened rigidly to the bottom of the rotor. The upper bearing, B, consists of a hollow cylindrical permanent magnet that attracts a

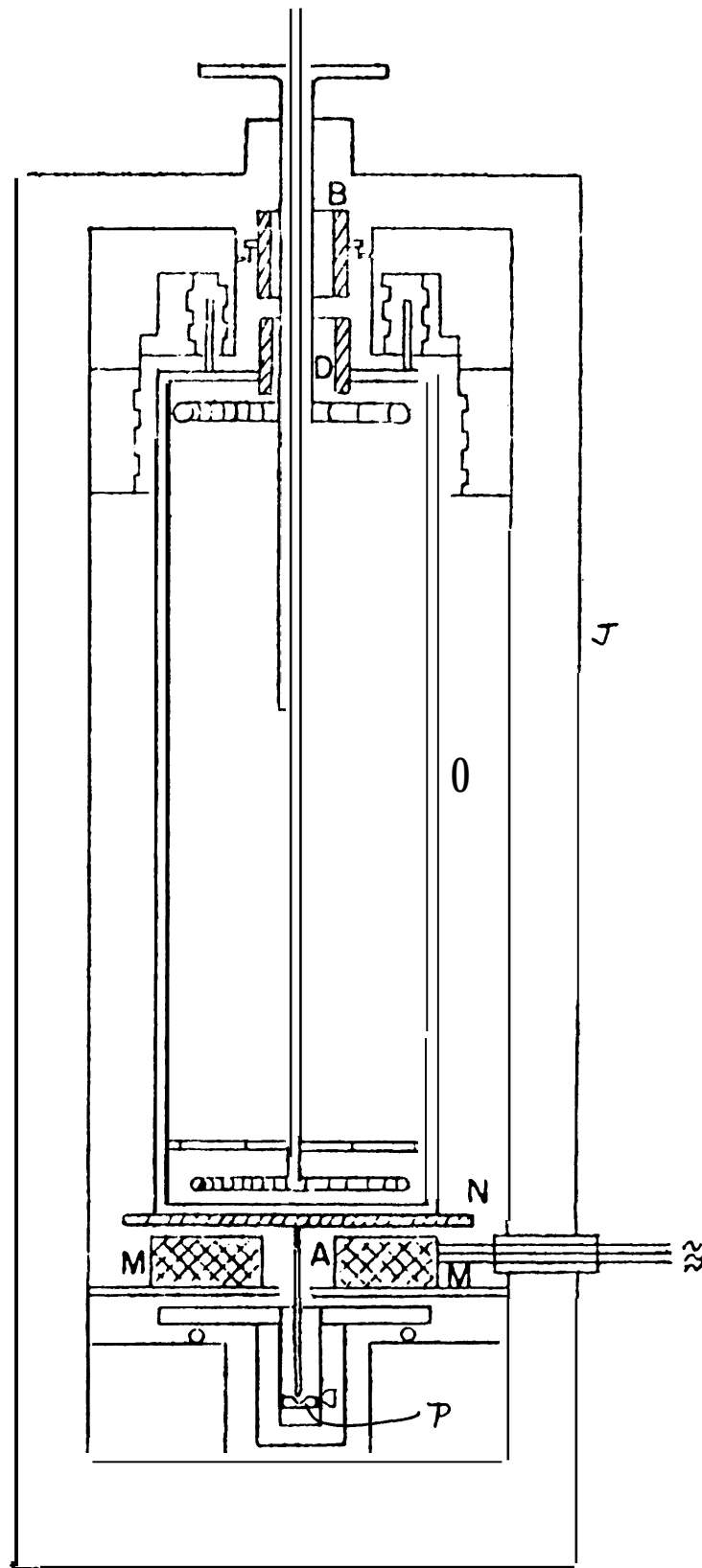


Figure 4. Zippe short bowl centrifuge.

(From J.W. Beams, see References.)

steel tube, D, mounted on the rotor. In this way, there is no mechanical contact between the axis of the rotor and the frame. The volume within the protective jacket, J, is evacuated so that the rotor spins in a vacuum. Even this small machine, which operated at subcritical speeds, was capable of producing separative work (see Annex D) at a rate of 0.45 kg per year.

With the successful demonstrating of the Zippe machine, the U.S. Atomic Energy Commission recognized that the centrifuge might possibly be developed into an economic method of separating the isotopes of uranium. Furthermore, since centrifuges apparently could be fabricated with relatively little difficulty and consumed very little power, the possibility existed that centrifuging might provide a mechanism for many small and/or developing nations to acquire a nuclear weapons capability. Accordingly, in 1960 the AEC declared that all work on centrifuges, which was unclassified at that time, would henceforth be classified.

Thereupon the AEC evidently undertook an accelerated program to develop the centrifuge for isotope separation.

Today the centrifuge process has reached a high level of sophistication, both in this country and abroad. According to authorities in the field, centrifuging is unquestionably cheaper than any other developed (this excludes laser separation) method of enriching uranium. An Anglo-Dutch-German enrichment group, Urenco, has successfully demonstrated the first cascades of two small centrifuge plants each with a planned capacity of about 200,000 kg SWU per year at Capenhurst, England, and Almelo, Holland. A small pilot plant is in operation, or about to go into operation, in Oak Ridge. One American firm has proposed building a major uranium enrichment plant to provide fuel for the nation's nuclear power plants.

Principles of Centrifuge Separation

In an isotope separation plant the basic unit that separates the isotopes is called a separating unit. In a gaseous diffusion plant this is a single diffusion barrier; in a centrifuge plant it is a single centrifuge machine. To provide the necessary material flow through a plant, several separating units are usually connected in parallel, that is, side by side. Such a group of parallel-connected units is referred to as a stage.

Since only a certain amount of separation can be obtained from a single stage, it is always necessary to connect a number of stages in *series*. An arrangement of

this type is called a cascade.

The degree of separation which is possible in one separating unit or one stage is determined by the separation constant α . **Clearly, the larger the value of α , the shorter** the cascade can be. In the gaseous diffusion process, α is given by the square root of the ratio of the masses of the process gases, $^{238}\text{UF}_6$ and $^{235}\text{UF}_6$, and has the value 1.00429. Since this number is only slightly greater than unity, a great many (about 3000) stages are required in a gaseous diffusion cascade to produce weapons grade uranium, about 90 percent ^{235}U . By contrast, in the centrifuge method for isotope separation, α is determined by the difference in mass between the heavy ($^{238}\text{UF}_6$) and light ($^{235}\text{UF}_6$) component, and increases with the length and the peripheral speed of the centrifuge rotor. It is possible, therefore, by operating a centrifuge with a long rotor at a sufficiently high speed to obtain values of α which are substantially larger than for the corresponding case with the diffusion process. The cascade for a separation plant based on the centrifuge process is then shorter than for the equivalent plant using gaseous diffusion. With a separation factor of 2, for example, apparently not an unreasonable value, only about 20 stages would be required to produce 90 percent uranium. This short cascade is one of the attractive features of isotope separation by centrifuge.

The rate at which a given separative unit or a cascade is actually capable of separating isotopes is measured in terms of separative work units, SWU, per unit time. For a single centrifuge, it has been shown that this rate is proportional to the length of the centrifuge and increases rapidly with its peripheral speed. To obtain large amounts of separative work per machine, it is therefore desirable to make the rotors of the machines as long as possible and operate at exceedingly high speed, which, as already noted, also reduces the length of the cascade.

However, this immediately leads to a difficult problem. As the speed of a rotor is increased, the rotor passes through a succession of vibration resonances which place the structure under great stress. At these so-called critical speeds the rotor has a tendency to fly apart, before the centrifuge has had an opportunity to reach its operating speed.

Evidently, the centrifuges used in the European pilot plants do not operate at supercritical speeds. Each is capable of producing somewhere in the neighborhood of 2 to 5 kg of SWU per year. In an unclassified remark, Dixie Lee Ray, former chairman of the U.S. Atomic Energy Commission, was reported to have said that 10,000 centrifuges of American design would do the same job as 100,000

European centrifuges. Since no exotic materials development could possibly account for an improvement by a factor of 10 in the performance of a subcritical centrifuge, it must be concluded that U.S. engineers have solved the problem of substantial supercritical operation. (**It** may be observed that the early machines of the Manhattan Project also operated at supercritical speeds.) If, in fact, Dr. Ray's statement is an accurate account of American centrifuge technology, then each centrifuge must be capable of producing somewhere between 20 and 50 kg of SWU per year.

As shown in Annex D, a plant with a capacity of between 2000 and 2300 kg of SWU per year, depending on tails assay, is necessary to produce 10 kg of weapons grade uranium per year. This means that a total of between 400 and 1200 centrifuges of European design would be required, depending on their individual capacities, or perhaps only 40 to 115 centrifuges of American design.

It should be pointed out that the electrical power required to operate a centrifuge separation plant is estimated to be only one-thirteenth the power for a gaseous diffusion plant. Since a diffusion plant requires an installed capacity of approximately 0.25 kW per kg of SWU per year, the corresponding centrifuge plant would need about 0.020 kW per kg of SWU per year or a total of only

46 kW for a 2300 kg SWU per year facility.

Another striking advantage of the centrifuge method, especially to a small and/or developing nation embarking on a weapons program, is that a small number of units or groups of centrifuges can be placed in operation as soon as they are built and tested. There is no necessity, as there is in the gaseous diffusion process, to wait upon the completion of an enormous facility before beginning separative operations. Production of weapons grade uranium can begin at a small level of SWU per year and gradually be increased as additional centrifuges come off the assembly line.

Problems with Centrifuge Technology

Having discussed the advantages of the **centrifuge** method over other methods of separating isotopes, it is appropriate to ask whether a small and/or developing nation can reasonably be expected to attempt to produce nuclear weapons by this method. For several reasons, it would appear doubtful that centrifuge separation would be the **process of choice** for **obtaining such weapons**.

To begin with, centrifuge separation is a highly sophisticated technology that has only recently been developed by a few of the most advanced nations in the world. The technical problems are formidable. The

centrifuges must spin in a vacuum at high speeds, magnetically supported at one end and on a special bearing at the other. The rotors must be fabricated from special materials of high tensile strength and the interior of the rotors, the process vessel, must be immune to attack by uranium hexafluoride, the process gas, which is extremely corrosive, hygroscopic and dangerous to work with. Arrangements must be made to carry the processed gas into and out of each centrifuge, from stage to stage, and, of course, the entire cascade must be controlled. Finally, unless the critical speed problem is solved, a large number of machines must be used for a comparatively small output.

While the major powers have solved most or all **of these** problems, their technology is classified and likely to remain so. This means that a new nation that elects to pursue centrifuge separation must undertake what can be expected to be a lengthy research and development program with uncertain results. This is in marked contrast to the situation such a nation would face in producing plutonium in small reactors, in which case, as pointed out in Section III, the technology is not only unclassified, but complete facility plans are readily available. **Also, since centrifuge separation** technology is new, the project personnel would have to be trained

from scratch within the nation's borders, whereas nuclear reactor engineering is taught openly around the world. Lastly, the fact that a centrifuge program would take so long to produce results would deter a small and/or developing nation from beginning such a project. Ideally, nuclear weapons should be acquired over a short time span to avert detection, and with a large degree of certainty of success.

It should be added, however, that if a nation were willing to scale down its nuclear weapons program to a level where only one bomb was produced every five or ten years, then in this case the centrifuge method might appear attractive. Nevertheless, it would also seem extremely doubtful that such a long term program could remain secret until a militarily significant number of weapons could be produced.

ANNEX A

PLUTONIUM PRODUCTION RATES

Plutonium-239 is produced in a thermal reactor as the result of the absorption of thermal and resonance neutrons by ^{238}U . The rate of ^{239}Pu production in atoms per second in the entire reactor is given by

$$R = N_{28} \bar{\sigma}_{a28} \phi_T V + (1-p) \epsilon \eta_{25} \bar{\sigma}_{a25} P_f \phi_T V, \quad (1)$$

where N_{28} and N_{25} are, respectively, the number of ^{238}U and ^{235}U atoms per cm^3 and $\bar{\sigma}_{a28}$ and $\bar{\sigma}_{a25}$ are their average thermal absorption cross sections; p is the resonance escape probability; ϵ is the fast fission factor; η_{25} is the average number of neutrons emitted by ^{235}U per neutron absorbed in that nucleus; P_f is the probability that a fission neutron will not escape from the reactor while slowing down; ϕ_T is the average thermal flux in the reactor; and V is the reactor volume. In Eq. (1), fissions in ^{239}Pu and ^{241}Pu have been ignored, since the concentrations of both these nuclides are small in a natural-uranium, graphite-moderated reactor. The first term in Eq. (1) is due to thermal neutron absorption; the second is due to resonance absorption.

The number of atoms of ^{239}Pu produced per atom of ^{235}U consumed in the reactor is called the conversion ratio

or sometimes the breeding ratio and is denoted by the symbol C . Since ^{235}U is consumed at the rate of $N_{25}\bar{\sigma}_{a25}'\phi$ atoms per second, it follows that

$$C = \frac{R}{N_{25}\bar{\sigma}_{a25}'\phi} = \frac{N_{28}\bar{\sigma}_{a28}}{N_{25}\bar{\sigma}_{a25}} + (1-p)\epsilon\eta_{25}P_F. \quad (2)$$

The values of the parameters in Eq. (2) are as follows:

$$N_{28}/N_{25} = 99.27/0.72$$

$$\bar{\sigma}_{a28}/\bar{\sigma}_{a25} = 2.70/680.8$$

$$\eta_{25} = 2.068$$

and for the Brookhaven Graphite Research Reactor,

$$p = 0.8783$$

$$\epsilon = 1.03$$

$$P_F \approx 1.$$

Introducing these parameters into Eq. (2) gives $C = 0.806$. Incidentally, the first term in Eq. (2) is about twice as large as the second term, which means that for reactors of the Brookhaven type twice as much ^{239}Pu is produced by thermal neutron absorption as by resonance neutron capture.

A thermal reactor operating at a power level of one megawatt (MW) consumes 1.23 grams of ^{235}U per day or $1.23 \times 365.25 = 449.26$ grams per year. Such a reactor produces ^{239}Pu at the rate of $449.26 \times 0.806 \times (239/235) = 368.27$ grams per year. If the reactor operates at a power of P MW, it follows that

$$^{239}\text{Pu production rate} = 368.27P \text{ grams per year.} \quad (3)$$

Since ^{239}Pu absorbs neutrons, it is consumed as well as produced in an operating reactor. If n_{49} is the total number of ^{239}Pu atoms in the reactor at any time, then n_{49} is determined by the equation

$$\frac{dn_{49}}{dt} = R - n_{49}\bar{\sigma}_{a49}\phi_T. \quad (4)$$

The solution to this equation is

$$n_{49} = \frac{R}{\bar{\sigma}_{a49}\phi_T} (1 - e^{-\bar{\sigma}_{a49}\phi_T t}). \quad (5)$$

Equation (5) shows that the amount of ^{239}Pu rises with a half-life time constant of $0.693/\bar{\sigma}_{a49}\phi_T$. The value of $\bar{\sigma}_{a49}$ is $0.886 \times 10^{11.3} = 896$ barns $= 8.96 \times 10^{-22} \text{ cm}^2$. In the BGRR the maximum thermal flux was 5×10^{12} neutrons/cm²-sec and so the average thermal flux was approximately $5 \times 10^{12}/3.88 = 1.29 \times 10^{12}$, where the

factor 3.88 is the maximum-to-average flux ratio for a cubical reactor. The time constant in Eq. (5) is then

$$\frac{0.693}{8.96 \times 10^{-22} \times 1.29 \times 10^{12}} = 6.00 \times 10^8 \text{ sec} = 19.0 \text{ years.}$$

This result means that for times short compared with 19 years, the amount of ^{239}Pu in the reactor can be computed by multiplying the production rate in Eq. (3) by the length of time that the fuel is left in the reactor at the power of P MW. In the case of the Brookhaven reactor, P varied between 22 MW and 30 MW. Using the nominal value of 25 MW gives an annual production of $368.27 \times 25 = 9207$ grams or 9.2 kilograms of ^{239}Pu .

This plutonium is not produced uniformly throughout the reactor. Because the neutron flux is highest at the center of the reactor, the ^{239}Pu concentration is also highest in that region. The average concentration of the ^{239}Pu in the fuel is $9207/75 = 123$ grams per ton. Near the center, the concentration is on the order of $3.88 \times 123 = 476$ or about 500 grams per ton. From a practical standpoint, this is the fuel that should be withdrawn from the reactor first, and this is the concentration for which the plutonium extraction facility should be **designed.**

ANNEX B

**PARAMETERS OF THE BROOKHAVEN GRAPHITE
RESEARCH REACTOR**

Power: up to 30 MW.

Neutron flux: 5×10^{12} maximum, 1.3×10^{12} average.

Fuel: natural uranium slugs 4 in. long, 1.1 in. in diameter, in finned aluminum cartridges 11 ft. long; total uranium fully loaded 116 tons, normal loading 75-90 tons.

Fuel arrangement: 37 x 37 square lattice, 8 in. pitch.

Moderator: graphite, 700 tons.

Coolant: Air, 300,000 cubic ft per minute, exit temperature 330°F, fan power 5 MW.

Reflector: graphite, 4.5 ft.

Shielding: iron plate plus 4.25 ft. concrete.

Control: 16-2 in. square by 12.5 ft. long steel rods containing 1.75 percent boron, in 2 banks entering horizontally from **2 corners of reactor.**

Additional features: (1) fuel cartridges pressurized with helium for leak detection; (2) reactor split in middle by 7 cm gap through which air enters.

ANNEX C

FOREIGN, NON-COMMUNIST RESOURCES OF URANIUM*

(in thousands of short tons)

Country	Reasonably assured	Estimated additional	Total
	up to \$15/lb. U₃O₈		
Australia	316	100	416
S and SW Africa	240	8	248
Canada	187	421	608
Niger	52	26	78
France	48	33	81
Algeria	36		36
Gabon	26	6	32
Spain	13	11	24
Argentina	12	18	30
Other	<u>56^{**}</u>	<u>26</u>	<u>82</u>
Total	986	649	1635

Up to \$30/lb. U₃O₈

Sweden	390		390
Australia	316	100	416
S and SW Africa	357	96	453
Canada	216	545	761
Spain	30	55	85
France	71	53	124
Niger	65	39	104

Country	Reasonably assured	Estimated additional	Total
Algeria	36		36
Argentina	27	51	78
Other	<u>152</u> ^{***}	<u>111</u>	<u>263</u>
Total	1660	1050	2710

* 1975 ERDA values

** Includes Central African Republic, Germany, India, Japan, Mexico, Portugal, Turkey, Yugoslavia, Zaire, and Brazil.

*** Includes, in addition to (**), Denmark, Finland, Italy, Korea, and the United Kingdom.

ANNEX D

SEPARATIVE WORK

The overall process of isotope separation is shown schematically in Figure D.1. Over some time period, M_f kg of uranium feed, that is, uranium to be enriched, containing ^{235}U at a concentration of x_f weight percent, enters the separating device or plant and M_p kg of product emerges with the enrichment x_p along with M_t kg of residue (tails) at the depleted enrichment x_t .

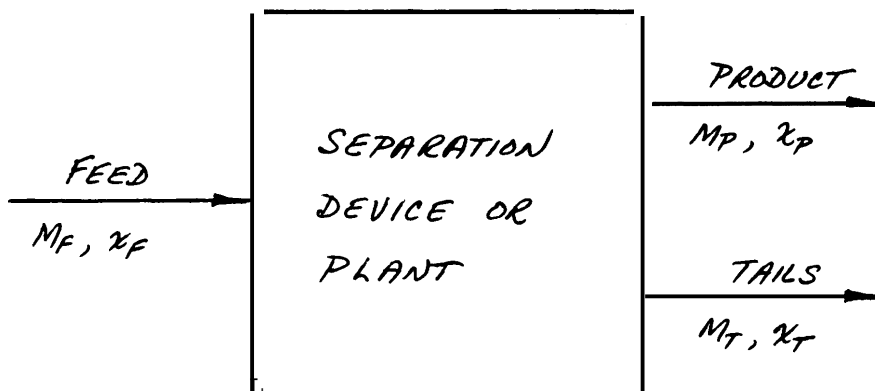


Figure D.1. Schematic representation of isotope separation.

Since the separation of isotopes requires, in effect, an unmixing of two gases, the entropy of the gases decreases in the process. As a result, work must be done on the gases by whatever device is performing the separation. This work is normally measured in Separative Work Units (SWU), which have units of mass (kg). The rate at which a device or an

entire separation plant is separating isotopes is measured in SWU per unit time, e.g., kg of SWU per year.

The separative work can also be expressed as the increase in the value of the enriched product and the depleted tails, taken together, less the value of the feed. Specifically, this is

$$\text{SWU} = M_P V(x_P) + M_T V(x_T) - M_F V(x_F), \quad (1)$$

where $V(x)$ is the value function

$$V(x) = (2x-1) \ln\left(\frac{x}{1-x}\right). \quad (2)$$

In view of the conservation of mass,

$$M_F = M_P + M_T, \quad (3)$$

Eq. (1) can also be written as

$$\text{SWU} = M_P [V(x_P) - V(x_T)] - M_F [V(x_F) - V(x_T)]. \quad (4)$$

From the conservation of ^{235}U , it follows that

$$x_F M_F = x_P M_P + x_T M_T. \quad (5)$$

Combining Eqs. (3) and (5) gives

$$M_F = \left(\frac{x_P - x_T}{x_F - x_T}\right) M_P. \quad (6)$$

Introducing Eq. (6) into Eq. (4) yields finally

$$SWU = M_p \left\{ [V(x_p) - V(x_T)] - \left(\frac{x_p - x_T}{x_F - x_T} \right) [V(x_F) - V(x_T)] \right\} \quad (7)$$

The assay or enrichment of natural uranium feed is fixed at 0.711 weight percent. According to Eq. (7), the amount of separative work required to produce M_p kg of product depends both on the enrichment of the product and on the residual enrichment of the tails.

Suppose it is desired to produce 10 kg of 90 percent uranium (90 w/o ^{235}U) at a tails assay of 0.2 w/o. Then from Eq. (2), $V(0.90) = 1.758$, $V(0.00711) = 4.869$, and $V(0.002) = 6.188$. Equation (7) then gives $SWU = 2274$ kg. On the other hand, if a tails assay of 0.3 w/o is acceptable, then $V(0.003) = 5.771$ and Eq. (7) gives $SWU = 2009$ kg.

It should be noted from Eq. (6) that as the tails assay is increased, the amount of feed material also increases. Thus to produce 10kg of 90w/o at 0.2 w/o tails requires 1757 kg of natural uranium or about 2600 kg of UF_6 . At 0.3 w/o tails the amount of UF_6 increases to 3230 kg.

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