

~~SECRET RD~~

~~SECRET~~

#00027252
D00033209

ORNL-321

This document consists of 38 pages.

REDACTED COPY

No. 18 of 60 copies, Series A.

Department of Energy Declassification Review	Determination: (Circle Number(s))
1st Review Date: 7-5-74	Classification Retained
Authority: <i>g/b/br/</i>	Classification Changed To:
Derived From: <input type="checkbox"/> DC <input type="checkbox"/> DD	Contains No DOE Classified Info
Declassify On:	Coordinate With:
2nd Review Date: 07/12/2012	5. Declassified
Authority: <i>DD</i>	7. Other (Specify)

Contract No. W-7405-eng-26

ANALYTICAL CHEMISTRY DIVISION

(C) ASSESSMENT OF A CONTROL SYSTEM

Detection of Hidden Isotope Separation Facilities

A. E. Cameron



2012000093

DEPARTMENT OF ENERGY DECLASSIFICATION REVIEW	
1st Review Date: 10/22/2012	Determination (Circle Number(s))
Authority: <i>ADD</i>	Classification Retained
Name: <i>g/b/br/</i>	Classification Changed To:
2nd Review Date: 2/1/12	Contains No DOE Classified Information
Authority: <i>ADD</i>	Coordinate With:
Name: <i>g/b/br/</i>	Declassified Information Bracketed
Guidance: <i>CG-2005-12/05</i>	7. Other (Specify) <i>ORNL/br/</i>

DATE ISSUED

APR 10 1961

DOE OC: CG-15C-1, 6/2002, DOE OC: Y/CG-100, R1, ch1,
 2/2001, DOE OC: CG-NMP-2, 8/2000, 200505
 4 TG-NMP-1, 8/1984, DOE OC
 38 pgs

OAK RIDGE NATIONAL LABORATORY
 Oak Ridge, Tennessee
 operated by
 UNION CARBIDE CORPORATION
 for the
 U.S. ATOMIC ENERGY COMMISSION

~~RESTRICTED DATA~~
 This document contains Restricted Data as defined in the Atomic Energy Act of 1954. Unauthorized disclosure subject to administrative and criminal sanctions.

~~RESTRICTED DATA~~
 This document contains restricted data as defined in the Atomic Energy Act of 1954. Its transmission or the disclosure of its contents in any manner to an unauthorized person is prohibited.

~~SECRET~~

~~SECRET RD~~

2014007761

20130006589

~~SECRET RD~~~~SECRET~~Distribution

- 1-50. S. G. English, USAEC, Washington
- 51. S. R. Sapirie, USAEC, Oak Ridge
- 52. C. E. Center, UCNC, ORGDP
- 53. J. A. Swartout, ORNL
- 54. G. E. Boyd, ORNL
- 55. A. de la Garza, ORGDP
- 56. R. J. Brouns, HAPO, Hanford
- 57. W. Singlevich, AFTAC, Washington
- 58. D. R. Miller, USAEC, Washington
- 59-60. A. E. Cameron, ORNL

~~SECRET~~~~SECRET RD~~

TABLE OF CONTENTS

	<u>Page</u>
Introduction	3
Statement of Problem	3
Assumptions	4
Basis for Detection and Location	5
Uranium Isotope Separation Plants	6
Gaseous Diffusion	6
1. Feed Preparation	6
2. The Diffusion Cascade	9
Characteristics of the Process	9
The Cascade Operation	13
Gas Centrifuge	18
Electromagnetic Process	23
Other Processes	24
Metal Production Plants	24
1. Fernald Operation	24
2. Weapons Parts Fabrication	27
Non-Fissionable Nuclides	28
Deuterium Production	29
Lithium Isotope Separation	30
Detection Capability	33
Bibliography	37

Introduction

The Atomic Energy Commission, on December 16, 1959, requested a special study and report on the feasibility of detecting and locating uranium and lithium isotope separation facilities which might be undeclared under a disarmament agreement or which could have been constructed and operated clandestinely to avoid detection. A preliminary report was prepared and issued (ORNL 59-12-76). Dr. R. J. Brouns of Hanford Atomic Products Operation and the writer also prepared a condensed report that included the detection of reactors, irradiated fuel processing sites and metal fabrication plants, which appeared as an attachment to an AEC staff paper issued February 29, 1960. A revision of this attachment was submitted March 30, 1960, as ORNL 60-3-115. This present report is intended to be a final report on this subject and to include the best estimate of capability that can be made on the basis of present techniques of sample collection and laboratory determinations.

The techniques that have been developed have, in general, been aimed directly at gathering information through clandestine sample collection. While they appear to be merely extensions of rather conventional health physics environmental studies, the detection capabilities are, in most cases, some orders of magnitude greater. The objective has been not only detection of an operating facility but the acquisition of diagnostic information.

Because this report reveals the precise location and interrelationship of USAEC installations and some details of processes, it is classified "Restricted Data". Because it reveals techniques which can be applied to clandestine acquisition of intelligence data, it is not suitable for release to Foreign Nationals.

Statement of Problem

Under the presumed international agreement, there would be a cessation of production of fissionable or fusible material for weapons fabrication. Existing stockpiles would be retained and reworking of weapons would be permitted for modernization, modification, etc. The problem is then, briefly, to monitor allowed facilities for production of enriched uranium, the fabrication of fuel elements for reactor usage, the reprocessing of irradiated enriched fuel elements, and plutonium production from reactors to prevent diversion of material to increase the weapons stockpile. The portion of this problem to which this report applies is the detection of any production facilities which were not declared under the agreement at the cutoff date and to detect such plants as might be built clandestinely either before or after the agreement. The procedures developed would apply equally well to detection of production facilities in an Nth country which might undertake the production of weapons on a clandestine basis.

The following definitions are given for the purpose of the discussion:

UNDECLARED PLANTS: This category is assumed to consist of production facilities of a normal type of construction which had been built prior to a disarmament agreement and that had been operated in a normal manner not intended to avoid detection. They are assumed to be of conventional size and type and to have relatively high production capability.

CLANDESTINE PLANTS: These are plants or production facilities constructed on a covert basis and purposely designed to avoid detection by any of the inspection systems agreed to in the treaty. It appears more likely that such facilities would have been constructed between the time of disarmament negotiations and the actual institution of inspection or control. They might thus have been built, tested, and held ready for operation when needed later. The construction of a complex isotope separation facility would be susceptible to detection by the conventional intelligence techniques and thus would become more expensive in the presence of an inspectorate.

Assumptions

The problem of detection of the two classes of facilities - undeclared and clandestine - will be discussed against the background of the following assumptions:

1. It will be assumed that certain rights will be granted under a disarmament treaty which will permit environmental surveys to be made. Exclusion areas will be defined and located. The presumption is that these will be military reservations, regions in which stockpiles of weapons exist, and probably other areas in which it will be stated that no operations of nuclear energy nature are conducted. Since the closeness of access cannot be defined at the present time, it will be possible in the discussion only to indicate the detection possibilities at favorable and unfavorable access distances.
2. It is assumed that the details of the inspection methods will be known. One must, therefore, assume that any clandestine or undeclared operation will be conducted in a fashion carefully designed to evade detection.
3. It is assumed that information from conventional intelligence operations will not be available to the inspection operation. The rights of over-flight for aerial photography will be carefully defined in any agreement. The results of such survey would be of particular use to a ground survey system and the availability of this information upon at least a limited basis is assumed.
4. The magnitude and complexity of a nuclear energy operation, aimed at the production of even a limited number of weapons, are sufficiently great that the complicity of at least a large portion of the foreign government must be assumed. Thus, the resources and facilities for support of evasion may be great.

5. It will be assumed that there is no inspection of mining operations or of primary chemical concentration of ores. Feed material of normal isotopic composition for a clandestine operation would thus be available.

Basis for Detection and Location

All atomic energy establishments emit effluents which are to a great extent characteristic of the operation. Considerations of health hazards and the unit cost of the materials being handled are the factors which determine the degree to which these effluents are controlled. Normal uranium, for instance, offers a comparatively minor health hazard and is not a particularly valuable material when compared to enriched uranium of product level. The control of uranium losses from a plant handling normal or near normal material will thus, under normal operating conditions, be less stringent than would be the case in an operation involving product level uranium. Plutonium, the transuranics in general, and U-233 will be much more stringently contained because of their extreme health hazard.

The effluents may be gaseous or fine particulate materials and will be air-transported for great distances, or may be water soluble or carried in liquid effluent in finely divided form. Much more is known at the present time about air-borne materials than about liquid effluents. The fate of uranium in a river system has not been thoroughly investigated. If, as is the case with some materials, it is possible to find a type of vegetation or other environmental sample in which the effluent is concentrated or accumulated, then the detection and characterization of the operation is possible at comparatively great distances.

The detection capabilities are obviously determined by the type of operation, the amount of material being handled - since losses will be expected to be proportional to the size of the operation - the type of sample, and to the sensitivity of the method. The techniques which have been applied are generally extensions of the commonly used methods of health physics environmental surveys but increased in sensitivity by some orders of magnitude in most cases. The detection of uranium may be made exceedingly sensitive by highly specialized mass spectrometric measurements, and to the determination of gross uranium is added the additional tool of measurement of isotopic composition, from which diagnostic information is obtainable about the type of operation that is being detected or monitored.

Containment of iodine from a dissolver operation on irradiated fuel is aimed principally at the short half-life isotope. The long-lived I^{129} will not be detected by a monitoring system aimed at controlling I^{131} . This isotope has been found to cumulate in vegetation and it is readily detected with high sensitivity by neutron activation method. It does not occur in nature and exists because of being a fission product. A plant which is dissolving irradiated fuel elements is thus quite readily detected on the basis of this one effluent. This detection scheme will be covered in detail in a report by R. J. Brouns of HAPO, and will not be further discussed in the following write-up.

Other materials of importance to the operation of a weapons program are lithium-6, deuterium, and tritium. The latter is a reactor produced material and will not be considered in this discussion. The reader is referred to the companion report by R. J. Browns for a discussion of the detection of tritium producing sites. Enrichment of the lithium isotopes and the production of deuterium involve no hazard to health, other than being associated with a chemical process involving normally dangerous materials. Containment will thus not be critical and effluents may be expected to be abundant. The detection capabilities will be discussed under individual plants or processes.

In the discussion which follows, there will be no detailed expose' of individual analytical procedures since it is not believed that a manual of laboratory techniques is germane to the purpose of this report. Individual processes will be outlined in some detail and the source and magnitude of the effluents indicated. In the case of processes that are potentially useful but which have not been constructed and operated, the magnitude of losses can only be guessed at. The centrifuge plant for the separation of uranium isotopes is an example of this situation.

Uranium Isotope Separation Plants

GASEOUS DIFFUSION:

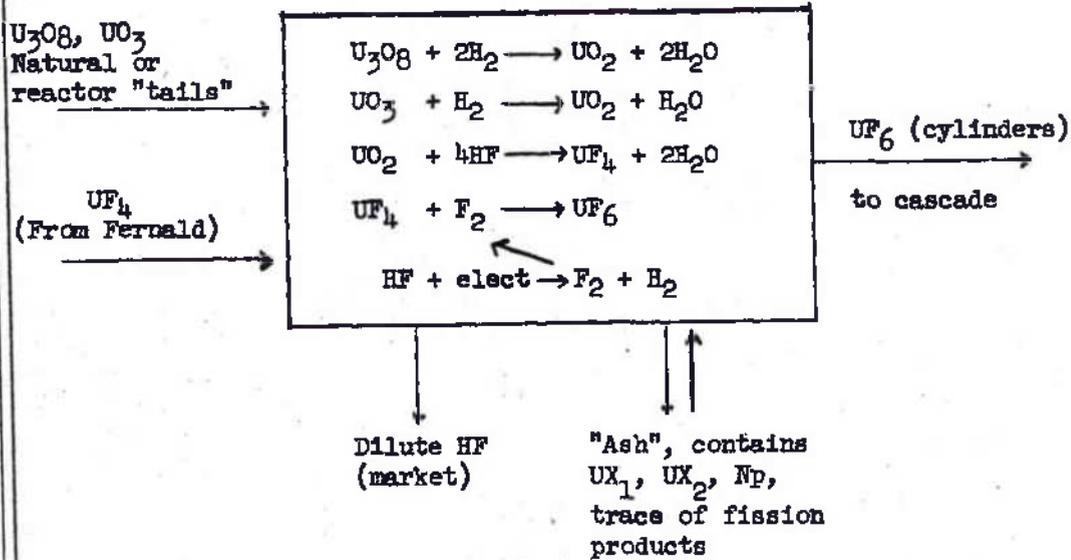
1. Feed Preparation.

The process gas for the diffusion plant is uranium hexafluoride. This is the only uranium compound which is sufficiently stable and which has high enough vapor pressure at ordinary temperatures to be useable in the process. Since fluorine has no stable isotopes, the only mass difference between isotopic molecules is that of the uranium isotopes. The preparation of the feed gas has generally been done at the site of the diffusion plant. There is not, therefore, in the USAEC complex any case of a diffusion plant per se. A portion (approximately 15%) of the feed for the U. S. diffusion plant complex is now being prepared by the Allied Chemical Corporation plant at Metropolis, Illinois. This plant is situated just across the Ohio River from Paducah, Kentucky, and is thus not geographically remote.

Raw uranium feed is generally supplied in the form of UO_3 to the plant producing uranium hexafluoride. It is produced in this form by the ore refinery or the plant processing reactor irradiated uranium by the denitration of uranyl nitrate. If the oxide produced is U_3O_8 , then a further chemical operation is required to prepare UO_3 before the feed plant can process it to hexafluoride. The direct fluorination process is reported to prepare UF_6 from U_3O_8 by reaction with elemental fluorine in a fluidized bed carrying a fluorination catalyst. The general outline of the feed preparation process is given in Figure 1.

The raw oxide is reduced with hydrogen to UO_2 and this is reacted with anhydrous HF to produce the tetrafluoride (green salt). The tetrafluoride is then reacted with elemental fluorine produced by electrolytic generators at the site. The hexafluoride is volatile, is collected in cold traps, and then transferred into cylinders for shipment to the cascade or for storage until needed.

~~SECRET~~
ORNL-LR-DWG. 57208



Characteristic Effluents

Dust: Uranium compounds

Gaseous: UF_6, F_2, HF

Aqueous: Uranium, fluoride

FIGURE 1
Feed Preparation

~~SECRET-RD~~

The major inputs to the processing plant are electric power and anhydrous hydrogen fluoride. The HF is used both for the preparation of "green salt" and for electrolyte in the fluorine cells. Other components of the electrolyte are potassium and lithium fluorides, but these are not consumed in the electrolysis. Materials of construction are generally nickel, monel, or other high nickel content alloy because of the extremely corrosive nature of hydrogen fluoride and elemental fluorine.

The reactors in which the hydrogen reduction and the fluorination are carried out have been vibrating trays or screw reactors, in which contacting of the solid particles with the gaseous phase has been produced by mechanical means. More recently, the "fluidized bed" reactor has been introduced. In this device the powder is fed at the bottom of a vertical cylinder, gas is passed in at the bottom, and the finely divided powder then acts like a "fluid". The reacted powder spills over the top and is fed to the next stage or to another type of reactor.

In all of these devices, there is the problem of removing entrained and finely divided solid from the effluent gas stream. Bag type filters have been used extensively. Containment of micron and submicron particles is not necessarily good, and the bags are reported to be subject to unpredictable failure when the material being trapped is a fluoride and the gas stream contains hydrogen fluoride. Fine pored metallic filters are now available and are being used to contain dust. In use, these filters are periodically "blown-back" with air to remove accumulated solid from the upstream side. Containment of solids by this type of filter is undoubtedly much better than with the bag type.

The entire process involves potentially dusty handling operations. When the uranium is only normal in isotopic concentration or slightly depleted, as in the case of recovered fuel from a plutonium production reactor, the health hazards are comparatively minor. The containment precautions then will be dictated by housekeeping and the cost of the material being handled. Typically, a feed plant is characterized by comparatively significant amounts of material lost to the environment. The feeding of a diffusion plant complex involves preparation of many tons of hexafluoride per day, so the loss of even a very small percentage of the material is sufficient to provide a readily detectable effluent.

Some uranium may potentially be lost as UF_6 from the final fluorination step by transport in the gas stream through the cold trapping. This loss may be negligible from the material accountability or health hazard standpoint, but can provide an indication of the operation by environmental sampling. The molecular UF_6 doubtless immediately hydrolyzes with atmospheric moisture and the resultant UO_2F_2 may remain as a very fine aerosol or attach to dust particles in the atmosphere and be carried for great distances.

Other air-borne effluents are fluoride, either as HF or as fluorine, and its reaction products with water. This has not proven useful as a detection device for a plant because vegetation appears to dispose of excess fluoride and only within one or two miles of a plant is fluoride inorganic material observed to be above the normally rather high background.

~~SECRET-RD~~

Spent hydrogen fluoride that has become diluted with water is, under the U. S. economy, resold to industry. This acid will carry trace uranium which is characteristic of the material handled in the feed plant.

The "ash" from the fluorination reaction is now conventionally recycled and does not appear as an "effluent". This residue will contain UX_1 and UX_2 which are the daughters of U^{238} . It may be necessary to allow these to decay if the ash becomes unduly "hot". When reactor processed uranium is being fed, the ash will contain a significant amount of neptunium and may be processed for the recovery of Np^{237} . The detection of this isotope in the vicinity of a plant would indicate that reactor processed uranium was being handled. However, the observation of U^{235} gives the same indication and its detection is somewhat more sensitive and simpler.

The same effluents which are air-borne will also be water transported as soluble or particulate material. This may be washed from the surroundings or may emerge from the plant through sewerage because of washing and cleaning operations. In this case, again, the significant and diagnostic effluent will be uranium.

Because of the nature of the process, the large tonnage of material handled, and the relatively minor health hazard of normal and near normal uranium, a feed plant is a comparatively dirty and dusty operation. It is readily detected in the environment by isolation of uranium and the establishment of the isotopic composition. The level of enrichment of the uranium can be determined, and the handling of reactor processed uranium is indicated by the presence of U^{236} . Detection of abnormally high fluoride is possible within one or two miles. Np^{237} may possibly be observed to indicate reactor origin of the feed.

2. The Diffusion Cascade.

Characteristics of the Process

The gaseous diffusion cascade is characterized by the large physical size of installation and very high power consumption because of the large number of separation stages necessary. That this must be so is readily seen from the simple relationship that the separation is inversely proportional to the ratio of the square roots of the molecular weights of the isotopic molecules. Thus, the maximum local separation that can be produced by a single stage is - $\sqrt{352} / \sqrt{349} = 1.0043$.

The whole effect rests upon the fact that at the same temperature, the kinetic energies of the molecules are identical, but the slight difference in weight means that the molecule containing the U^{235} isotope will be moving faster, and will pass more rapidly through a porous membrane. The basic

process is illustrated by the following diagram (Figure 2). The feed gas is passed into a chamber that is divided into two sections by a porous membrane or "barrier". The holes in the membrane must be small in diameter compared to the mean-free-path of the gas molecules at that temperature and pressure. Flow through the pores must involve mainly collisions with the walls, rather than intermolecular impacts. The back of the membrane is maintained at some pressure P_2 which is lower than the pressure P_1 at which the feed is introduced. The faster and lighter molecules that contain the wanted isotope U^{235} will pass more rapidly through the membrane, resulting in a thin layer on the high pressure side of the membrane, which is depleted in the light isotope.

DELETED

Two streams thus

emerge from the separation cell, one which is slightly depleted in the light isotope and the other is slightly enriched. The process is, in general, operated to give a "cut" of 0.5, so that the incoming feed is separated into two equal streams.

In order to recreate the pressure differential in the next stage (and the pressure gradient is the driving force in the process), the product stream must be recompressed and combined with the reject stream from the next stage higher in the cascade to maintain material balance. The heat of compression must be removed by a heat exchanger which may follow or precede the compressor. Thus, each stage of the process will involve a "converter" which contains the separative membrane or "barrier", a compressor, and a heat exchanger. The power input to the process appears, then, principally as low-level heat that must eventually be dissipated to atmosphere or to a river.

The cascading arrangement is shown in Figure 3. From stage "A", the feed is split into a product stream going to the compressor for stage A + 1, where it is combined with the reject stream from stage A + 2, and a reject stream which enters the compressor feeding stage A - 1. This staging arrangement is valid only for a "cut" of 50%. Control valves are necessary to equalize flows and to prevent unbalances in the system. These have been omitted from the diagram.

DELETED

~~It is naturally very important that~~

~~no condensation of UF_6 occur, lest criticality be exceeded in the section of the cascade where the enrichment of the U^{235} is high. A balance is made of~~

Doc
6.2(a)

Doc
6.2(a)
UCN1

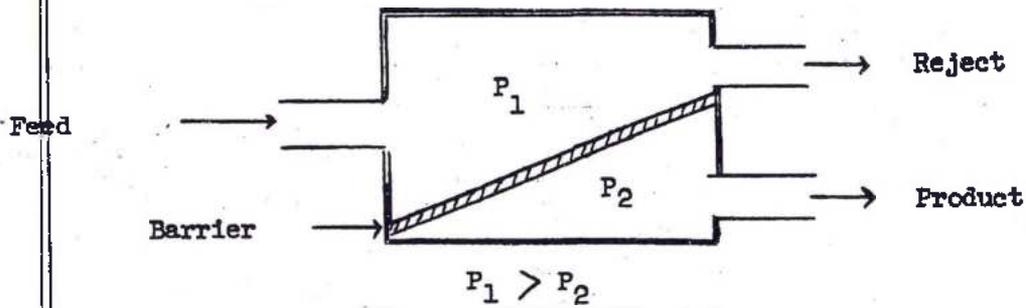


FIGURE 2

Basic Gaseous Diffusion Process

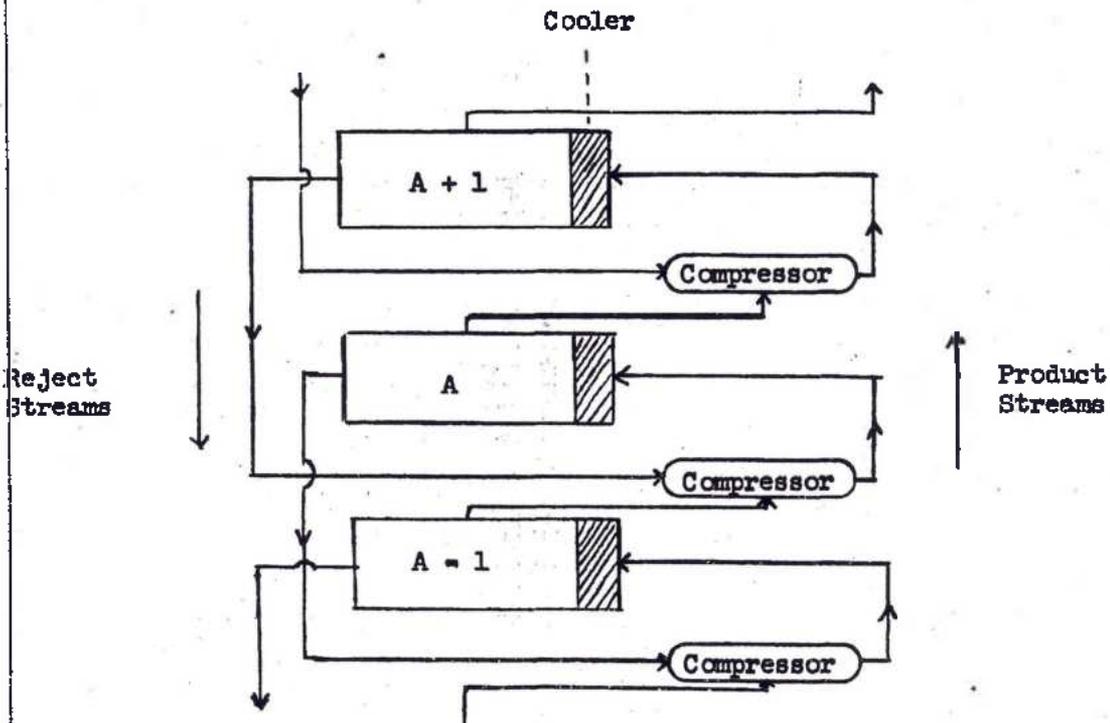


FIGURE 3

Gaseous Diffusion Cascade Staging

the desirable results of raising temperatures against increased reactivity of metallic surfaces and consequent increased corrosion and consumption. The latter term applies to the deposition of solid uranium compounds on surfaces, as contrasted with actual chemical attack, which results in the formation of fluorides of the materials of construction.

The pressure in the plant is generally below atmospheric, so that leakage is inward. Moisture must be avoided because of the reaction with UF_6 which causes the deposition of UO_2F_2 and the formation of HF, which may have severe corrosive effects. Compressor shaft seals are maintained in an atmosphere of dry nitrogen so that there is always an appreciable amount of this gas that must be purged from the cascade. There are portions of the cascade whose function is to separate UF_6 from lighter gases and to vent these to atmosphere.

DOE
62a

DELETED

United States technology has employed two different types of fluorocarbons. These compounds are hydrocarbons in which the hydrogen has been replaced by fluorine. They are extremely inert chemically and so produce no undesirable reactions if they leak into the cascade. Originally, a high boiling perfluorinated compound of the general formula C_8F_{16} was used. This was circulated through the heat exchangers to remove the heat of compression and was then used to transfer heat to water. This system avoids the possibility of a water leak developing inside the cascade. More recently, a lighter fluorochloro-compound - trichloro-heptafluoro butane (Freon 114) - is used as an evaporative coolant. This compound boils in the heat exchanger and yields its heat to water when it is recondensed. Other technology is reported to be to use heat exchange directly to water without the fluorocarbon intermediate.

Materials of construction for the gaseous diffusion process must be inert to active fluorinating agents and particularly to UF_6 . Nickel is the material that best resists the attack of fluorine and fluorides; thus, nickel and alloys containing a high percentage of this element are used in large amounts. Aluminum has proven to be remarkably resistant to attack and has been used to some extent in compressors and piping. For large equipment, it may be economical to use steel or other appropriate base material and to nickel-plate it.

DELETED

DOE
62(a)

Fluorocarbons in solid form - e.g., Teflon, Fluorothene - are useful for valve seats, gaskets, etc. Fluorocarbon oils have been used for lubrication of moving parts and pumps. The use of fluorocarbons for heat transfer media has been mentioned above. Thus, a well-developed fluorine technology is necessary to support a diffusion plant.

The entire plant must be very tight against atmospheric in-leakage so advanced "vacuum" technology is required. Precision machine shop skills for fabrication of shaft seals and other specialized parts are a must for success. Thorough instrumentation of the entire process is required for operation. The ORGDP complex is operated entirely from a central control room, even though it spreads over many acres of ground. The operating force can be comparatively small, with the bulk of the personnel required being for maintenance of equipment. Construction, on the other hand, could require a large labor force.

All in all, construction of a clandestine plant under an inspection system would appear to be an undertaking of such magnitude that it would not be seriously considered. It would be too readily susceptible to detection by conventional intelligence methods because of the large construction forces, the unusual and specialized materials or machinery, and the type of personnel required.

One can summarize the characteristics of the process as involving the following:

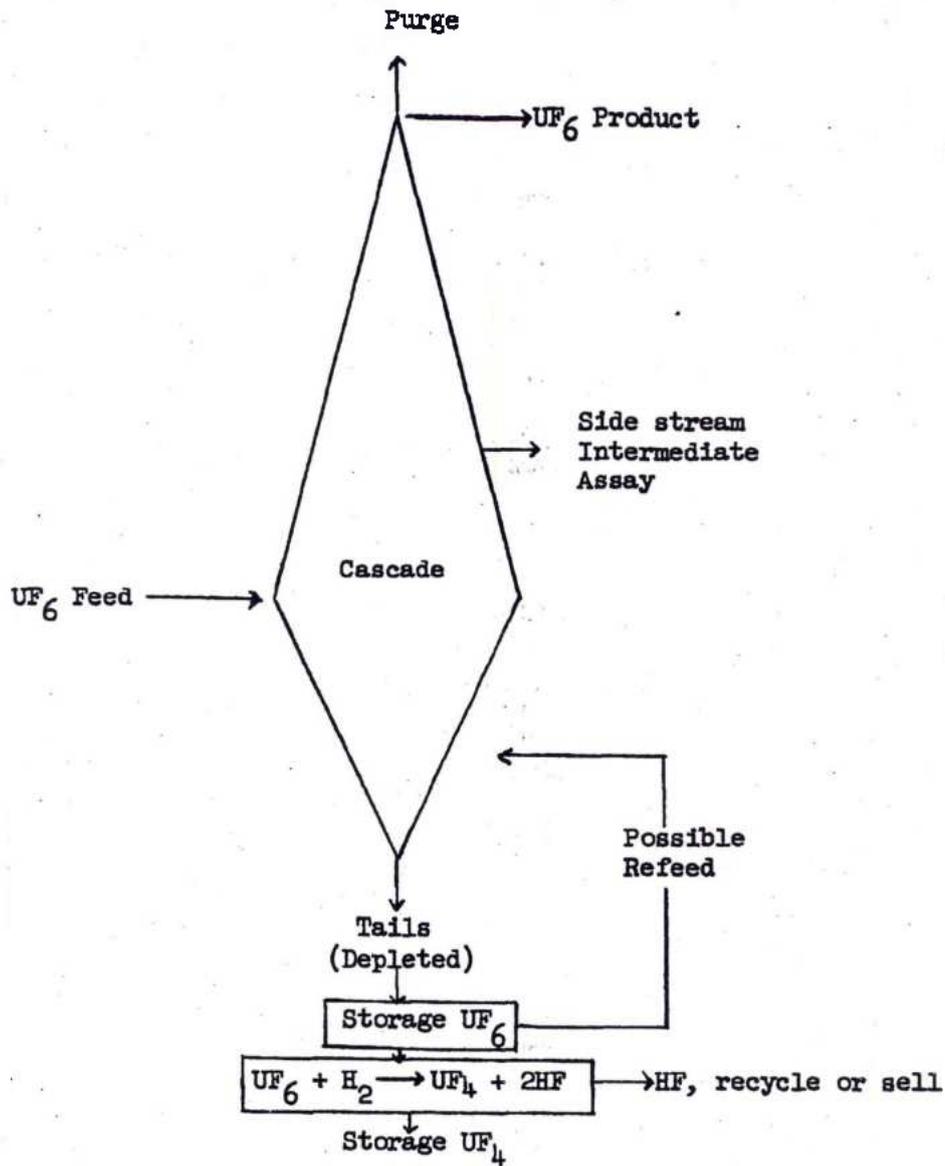
1. Very high power consumption, released as low-level heat.
2. Large physical size.
3. Specialized equipment and instrumentation.
4. Extensive use of nickel or high nickel alloys,
5. Well developed fluorine technology.
6. Highly specialized personnel.

DoS
6.2(a)

DELETED

The Cascade Operation

An outline of the various inputs and withdrawals that may occur in a typical cascade is given in Figure 4. The tapering of the cascade is indicated, both toward product and reject. The input UF₆ is introduced at the feed point, which is the "widest" portion of the cascade. It is usually handled in steel cylinders which may typically weigh ten tons. The cylinders are heated in water baths to melt and volatilize the UF₆. There is the possibility of some occasional loss at this point. At various places in the cascade there are purges to atmosphere by which light gases are separated from UF₆. Effluents from these will be fluorine, hydrogen fluoride, fluorocarbons (if used), and probably a very small amount of UF₆. Because of the value of the product, the purge at the upper end of the cascade is probably more carefully monitored than one lower down.



Characteristic Effluents:

Particulate: Uranium (depleted) from reduction

Gaseous: UF₆, F₂, HF, fluorocarbons

FIGURE 4

Gaseous Diffusion Cascade

Product is withdrawn by freezing into a trap or by pumping into a cylinder in which the UF_6 is liquefied under pressure and subsequently solidifies on cooling. "Always safe" geometry of container must be used at high enrichments. Withdrawal may be continuous, or intermittent. Sidestreams may be withdrawn at intermediate enrichment for use in reactors or in some types of weapons.

The waste withdrawal must be on a large scale. This is more likely to be done by compressing the UF_6 and filling the cylinders with liquid. The bulk is, of course, that of the feed material since the volume or weight of U^{235} removed as product is a small portion of the feed. Waste, depleted in the light isotope to a greater or lesser extent, is stored in cylinders. If not depleted to the limit possible in the cascade, then under conditions where normal feed is scarce and product is needed even at increased unit cost, it may be economical to re-feed. If this is done at the normal feed point, then there will be a re-establishment of the cascade gradient.

Waste that has been depleted to the economical maximum is reacted with hydrogen to reduce it to solid UF_4 , which is stored as a dry powder, and the recovered HF is used in feed preparation or is resold on the open market. This operation will be somewhat dusty and losses will tend to be high because of the large amount of material handled and its low economic value.

Feed to the cascade may be natural uranium or uranium that has been somewhat depleted by irradiation in a plutonium producing reactor. Effluent from a cascade, if detectable, will carry U^{236} if irradiated uranium is being used as feed. Slightly enriched uranium which was used as fuel in a reactor, e.g., in the 1-2% range, may be re-fed; as may be recovered uranium from salvage operations of any kind. High level irradiated uranium recovered from spent reactor cores from high Flux reactors or power reactors (e.g., from submarines) is not at present likely to be re-fed to a cascade, but will be blended with product produced at enough higher assay to make blending to specification possible. This will also typically carry comparatively large amounts of the 2% isotope. Losses from re-feeding of enriched materials can be expected to be very slight.

The auxiliary processes that are necessary for a cascade operation are shown schematically in Figure 5.

~~DELETED~~

Doc
6.4(a)

The UF_6 thus produced is recovered. Equipment may also be decontaminated by chemical means, usually by nitric acid spray or baths.

~~SECRET~~
ORNL-IR-DWG. 57204

DELETED

Del
6.2(a)

FIGURE 5

Gaseous Diffusion Auxiliary Processes



Uranium of various enrichment levels is recovered by both of these processes and will eventually be refluorinated and fed again to the cascade at an appropriate point. Junked equipment or scrap may be sufficiently decontaminated to be sold on the open market, or may be buried if the economics of the market are not favorable. From these operations, there is the possibility of liquid effluents which will typically contain isotopically altered uranium at the level of parts per million and nickel, aluminum, and fluoride which may, to some extent, be typical of the process.

In United States technology, the three existing cascades are operated essentially as a single unit. Paducah serves as a "feed" plant for Portsmouth and for the Oak Ridge Gaseous Diffusion Plant. While variations occur, depending upon economics and the type of feed material available, the general operation is that all uranium hexafluoride is prepared at Paducah. Then, whether normal or reactor tails (slightly impoverished), it is fed to the Paducah cascade. Product - at perhaps 2-3% U^{235} - is then shipped to Oak Ridge or to Portsmouth where it is fed to the cascades there. Tails from these two cascades are returned to Paducah for further enrichment. The final waste from Paducah is stored as UF_6 or, since it is very highly depleted, may be reduced with hydrogen and stored as dry UF_4 .

Such an arrangement is quite feasible in an economy where rail, water, or truck transport is common and where weather does not interfere with shipments. It permits spreading the extremely high power load over different portions of the country, as well as providing the advantage of dispersal of important targets. Under the United States system, the processing of product to metal and the fabrication of weapons parts or reactor cores takes place at a site other than the diffusion plant. This is a very likely arrangement since the final product is not bulky and no great amount of shipping space is required. Shipment by air would be very feasible.

Summarizing, then: Detection of an undeclared diffusion plant would be very likely by conventional intelligence means which could well include aerial survey. The large size of the plant, the extremely large power requirements, and the evolution of great amounts of waste low level heat would be readily detectable. Construction of a clandestine diffusion plant would appear to be most unattractive. A diffusion plant must be large to be useful.

No criterion exists for the estimation of the capability of detection of a diffusion plant per se. USAEC plants have, or have had, feed plants associated with them and, as pointed out above, the preparation of UF_6 from uranium oxide is an operation that is high in uranium loss compared to operations of other types. Surveys around the Portsmouth plant in the early days of operation showed that the environmental contamination was very low. There was no feed plant operating there at that time.

Because of the small losses from purge, some water-borne losses from salvage and decontamination operations, and occasional losses at feed and waste withdrawal points, one would expect to be able to prove the presence of isotopically altered uranium in the environment of a diffusion plant that had been operating for some time. The detection capability would be much lower than for a feed preparation operation or - as will be seen later in this report - for an enriched metal fabrication shop.

Nickel may be found to be above background in the immediate vicinity of a plant, particularly if barrier manufacture is carried on there. High fluoride is typical, but detectable only within two miles at the most. No good method now exists for the detection of fluorocarbon vapors, and the use of these compounds in large quantity is not necessary for cascade operation.

GAS CENTRIFUGE:

The gas centrifuge has, from the theoretical standpoint, been a very appealing method of isotope separation because the separative effect is a function of the mass difference, rather than the percentage mass difference. Thus, separation of the principal uranium isotopes with three mass units difference in weight at mass 350 would be as effective as separation of, for instance, something of mass 7 and 10. The lack of adequate technology for building centrifuge drums capable of withstanding the forces generated by peripheral speeds on the order of 300 meters per second has, until recently, prevented application of this separation scheme. Development work has proceeded in Russia, in Germany, and in the United States and recent newspaper publicity has led the public to believe that here is, indeed, a cheap and relatively simple method of isotope separation which would enable even small nations to become nuclear powers.

The fact is that, at the present time, there is no known cascade operating with this process and applications to uranium hexafluoride have been on a limited experimental basis. For the purposes of this report and the clandestine plant potentialities of this method, it must be given very serious consideration. Calculations indicate that a cascade is feasible, that no elaborate controls would be necessary, that the power consumption would be meager compared to that required for a gaseous diffusion plant, and the physical size of the plant and its uranium inventory would both be small. These factors would all make detection and location of such a plant difficult.

The gas centrifuge is typically a length of high strength tubing with end closures to form a drum which may conveniently be 10 cm. radius with a length of 100 cm. By proper choice of materials of construction, a peripheral speed of 300 meters per second is possible and this can probably be increased as the technology of high strength materials progresses.

A bearing at the bottom must support the weight of the drum, unless a portion of the load is removed by magnetic suspension at the top. The drum spins in an evacuated chamber and is driven by a synchronous motor attached at the top or bottom. These design points are crudely indicated in Figure 6. The method of introduction of feed gas and of removal of product and waste will vary with design of the equipment. The feed, however, is introduced on the axis and the waste and product may be removed from the top and bottom at the periphery by "scoops", as indicated in the sketch.

DOB
6.2(a)

DELETED

A circulation is produced in the drum as indicated by the flow arrows and enhances the effect of the centrifugal separation.

DOB
6.2(a)

DELETED

According to the simplified theory the separative work depends upon the fourth power of the peripheral speed, the inverse square of the absolute temperature, and the first power of the axial length. The radius and the pressure do not appear explicitly in the mathematics. There is obvious advantage in increasing the peripheral speed.

DOB
6.2(a)

DELETED

The following table gives the relative estimated characteristics of plants constructed for two suggested types of centrifuges. The 300 meter per second is assumed to exist (sub-critical) and to have been demonstrated as feasible.

DELETED

Both plants are calculated to produce 50 kg. of weapons grade (U^{235} 90%) uranium per year. This would require the feeding of 25 tons of uranium hexafluoride per year.*

2(a)

Type	Number of Centrifuges	Power Input	Number of Stages
300 m./sec.	2400	2400 kw	100

DOB

DELETED

6.2(a)

centrifuges would be necessary in parallel in the plant operating at the higher peripheral velocity, and controls for gas flow would thereby be reduced in complexity.

*KOA-662 (Secret), R. A. Levin, D. E. Hatch, and E. Van Halle, February 26, 1960.



~~SECRET~~
ORNL-LR-DWG. 57203

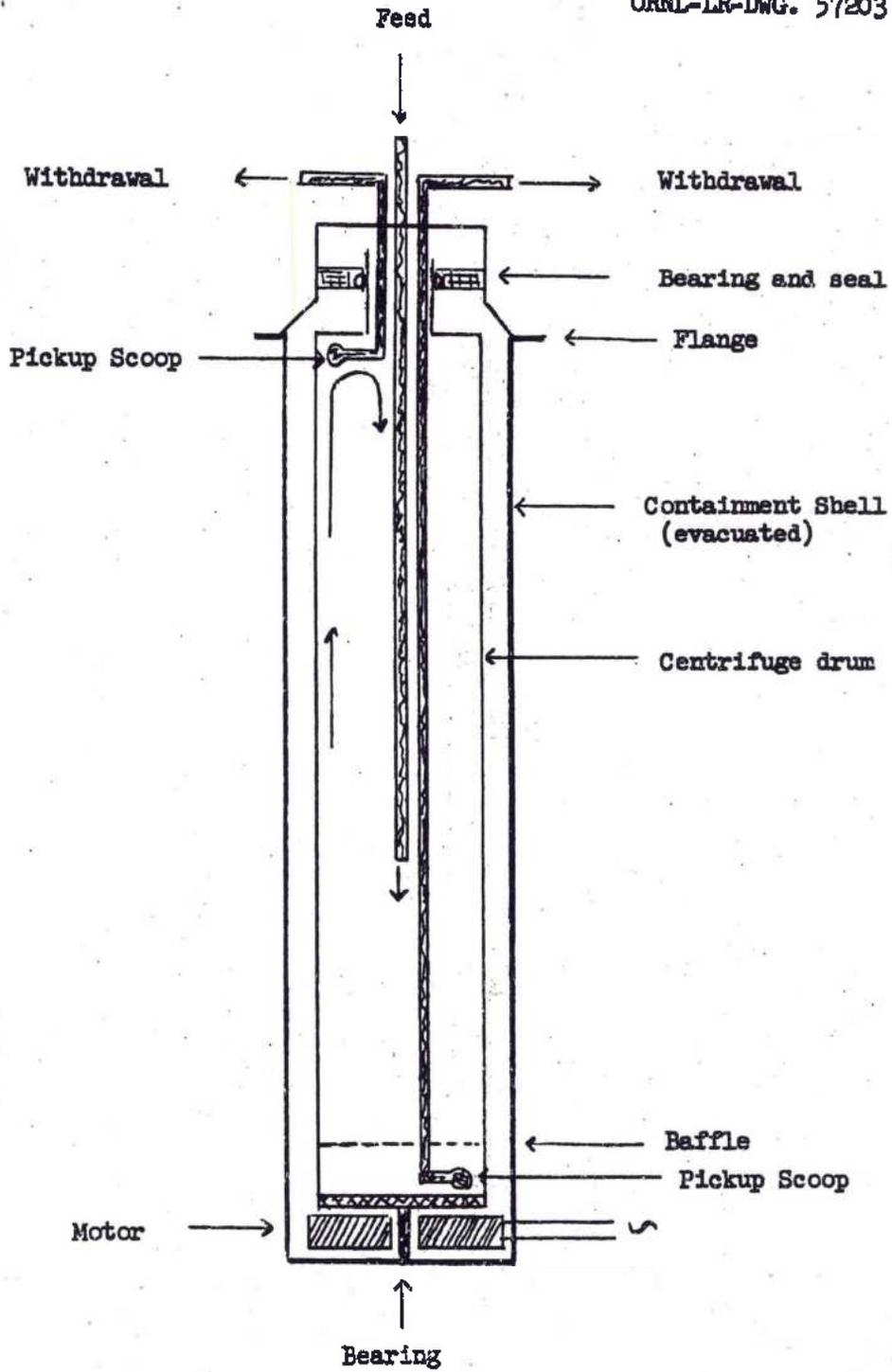


FIGURE 6

Gas Ultracentrifuge

One suggested type of structure for the plants would be a four-story building with a ground area of 24,000 feet² for the slow speed plant and a three-story building with 12,000 feet² on grade for the high speed plant. A single, or two-story building, with basement, could equally well be used, but would call for somewhat more process piping. It is apparent, however, from these suggested sizes that a building of quite modest size would serve to contain the centrifuge cascade.

The relatively modest power requirements could be supplied by diesel generators. Furthermore, the cascade could be broken into sections which could be operated in different buildings or even in different parts of a country if clandestine operation were being achieved.

A plant of this type, if built before a disarmament agreement was reached and if constructed with a view to clandestine operation, would be very difficult to detect and locate. Construction in the presence of an inspectorate under conditions where conventional intelligence techniques might lead to the knowledge of construction of centrifuge components could be more difficult. Detection under these conditions would depend to a large extent upon the complexity of the centrifuges being built, particularly as concerns the bearings which may require machine work of high quality.

The requirements for unusual equipment would be the centrifuges and bearings, and the power source which must be a variable frequency generator that will drive the synchronous motors on the individual centrifuges. The drums, themselves, would be of a high strength alloy but it is of the general type which is developed for other uses, such as fuel tanks in rockets. The manufacture of all of these components could be concealed in a country having a well developed industrial potential.

It is not known how complex a system of controls would be needed to operate a centrifuge cascade. Some opinions are that such a system would be relatively well self-adjusting and would not require any extensive system of gas flow control valves to prevent surging and to permit parallel operation of centrifuges.

The outstanding characteristics of this type of plant then, in marked contrast to the gaseous diffusion, would be the following:

1. High separation per stage.
2. Low power consumption.
3. Small inventory.
4. Small physical plant.
5. Capable of being operated in dispersed units.
6. No large scale heat dissipation.

Like the diffusion cascade, the centrifuge system would operate under reduced pressure and leakage would be into the cascade. Purging, because of the smaller volume of gas being handled, would be much simpler. Cold-trapping to remove hexafluoride from vented gases would probably be all that would be necessary. The centrifuge drums would have to be contained mechanically for safety reasons and the container would be evacuated during operation. This system would provide perfect protection against loss of uranium to atmosphere in case of a drum failure. The only loss to surroundings would come about as a result of severe malfunction or accident.

DELETED

At these temperatures and in the absence of barrier whose fine pores are plugged by corrosion products, the problems of corrosion would essentially disappear and much less stringent requirements would be placed upon materials of construction. Aluminum and plated steel would replace nickel and alloys containing high percentages of that metal.

DELETED

The cascade auxiliary system would be similar to that outlined for the gaseous diffusion process (Cf. Figure 5). Decontamination of equipment would be necessary before repair, particularly if the centrifuges were from the upper part of the cascade where the alpha activity of the uranium is high. This cleaning operation would be on a comparatively small scale, the surfaces being cleaned would be relatively small and the solutions would be entirely recovered by evaporative concentration or ion exchange to prevent any detectable loss to sewer. The comparatively small amounts of enriched uranium recovered could be stored and periodically refluorinated for re-feeding if warranted.

For maximum concealment of this type of plant it would seem desirable to divert feed from some other operation, rather than to have a process for the preparation of uranium hexafluoride in the same location. Diversion of hexafluoride in these comparatively small amounts would appear feasible. Alternatively, diversion of green salt (UF_4) and its fluorination at the site in a "flame reactor" whose effluent could be comparatively easily contained would be attractive. This problem and the problem of complete concealment of the plant would best be accomplished by locating the plant in the immediate vicinity of a facility which was engaged in the large scale production of normal uranium metal slugs for reactors. A plant of the type of that operated by National Lead Company at Fernald, Ohio would, because of its comparatively high loss of uranium to the environment, provide perfect and complete masking of the centrifuge operation.

The bulk of the final product would be small and it could readily be transported on a clandestine basis to another location for reduction to metal and fabrication of weapons parts. It must be remembered, however, that in

DOE
6.2(a)

DOE
6.2(a)

SECRET

the amounts considered for this plant described, this operation can be carried on in a comparatively small laboratory. Concealment of effluents is readily possible by application of the type of techniques used in the production of metallic plutonium. Glove boxes and stringent control of all effluents make the detection of plutonium fabrication operations practically impossible.

It is apparent that the chances of locating a centrifuge plant are very poor if the facility was constructed with the intention of clandestine operation.

ELECTROMAGNETIC PROCESS:

The electromagnetic process was used to produce the first highly enriched uranium in quantity, and the first bomb used in warfare was loaded with weapons grade U^{235} produced by this process. The method is not appealing enough to be seriously considered as a means of producing appreciable amounts of U^{235} for weapons purposes under present technology. The number of people required, the size of the equipment, and the very low process efficiency of the method all are against its application. The chief utility at present is in the production of research quantities of separated isotopes of most of the stable elements and for the preparation of highly purified separated isotopes of uranium, plutonium, and eventually of other transuranic elements.

The method requires the preparation of a volatile "charge" material which can be heated in the ion source to provide a vapor which is then ionized by an arc discharge. The ions are accelerated to a final energy of 35 KV, separated according to mass in a magnetic field, and collected in water-cooled pockets. Arc ionization seldom reaches an efficiency greater than 10% so that 90% of the charge material must be recovered from the interior of the vacuum chamber if it is valuable or scarce. Since the process must be operated in two stages in order that the final enrichment be greater than 90% an enriched "recycle" is required. The Y-12 operation during the war used "Alpha" stage units with 48-inch radius to attain an enrichment to 12-15% and this material was then used in "Beta" stage units with 24-inch radius to attain the final product.

The production rate is determined by the current in the wanted ion beam. Roughly, 1 milliamperere of current per day is equivalent to 1 millimole. If a current of 138 milliampere total is obtained with normal uranium, then 1 milliamperere is the current from the U^{235} beam. One millimole per day is 0.235 grams and even if this were of the wanted enrichment, the production rate is very small.

This process will not be considered any further in this report.

Other Processes

Some small enrichments of uranium 235 were obtained early in the Manhattan project by a thermal diffusion process in liquid uranium hexafluoride. The increase in concentration was small and the power consumption was very high. As soon as the gaseous diffusion process became operable, this thermal diffusion plant was shut down.

No successful separation of uranium isotopes has been demonstrated by processes of ion exchange, distillation, fractional crystallization, or chemical exchange.

Metal Production Plants

A somewhat arbitrary distinction has been drawn between metal production plants, which in the USAEC complex are typified by the Fernald operation near Cincinnati and the Y-12 weapons fabrication plant, and plants which produce feed for a gaseous diffusion or centrifuge cascade. Many of the same manufacturing operations are conducted in the two categories of plants and feed preparation could well be carried on in the same location where metal is produced. The distinction between Fernald and Y-12 is made principally because of the difference in level of enrichment of the uranium handled.

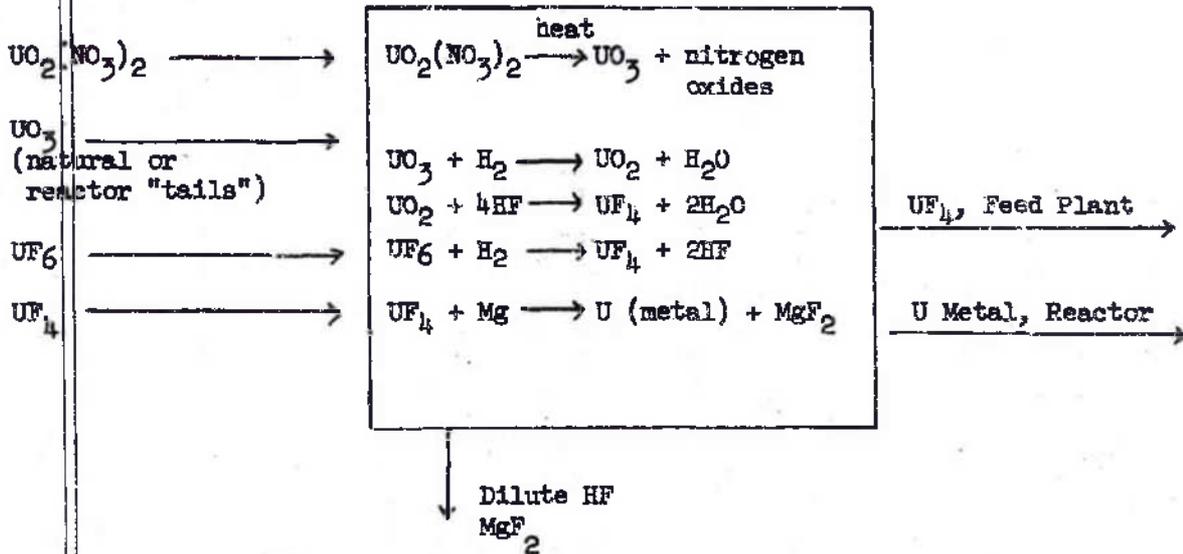
1. Fernald Operation.

The Fernald plant, operated by the National Lead Company of Ohio, handles principally normal or near normal uranium. The source of raw material may be uranyl nitrate or uranium oxide from ore refining and from the recovered uranium from plutonium production sites. In the latter case, the U^{235} content will be somewhat below normal concentration and the intent will be to prepare UF_4 (green salt) or uranium hexafluoride for shipment to a gaseous diffusion cascade for enrichment.

The various chemical processes which will be conducted in the plant are outlined in Figure 7. If the raw material is in the form of uranyl nitrate as a result of extraction of uranium by a solvent process, an initial denitration step is required. The salt is roasted and uranium trioxide is produced. Large amounts of oxides of nitrogen are released. The effluent gases may be scrubbed to reduce atmospheric contamination but, in any case, nitrate will appear either in the air or in aqueous effluent.

Uranium trioxide from this operation or from a denitration step at source is then reduced with hydrogen to UO_2 . If the form in which the uranium was received was U_3O_8 , the same hydrogen reduction is required to

~~SECRET~~
ORNL-LR-DWG. 57207



Effluents: Airborne, Particulate Uranium
Oxides of nitrogen
Fluorides

Water	Uranium
	Nitrate
	Fluoride

FIGURE 7

"Fernald" Type Metal Plant

secure the desired form for fluorination. Reactions which involve the contacting of a solid with a gas are dusty and filtration of the effluent gases is required. Where the health hazards are small, as they are with uranium of near normal concentration in U^{235} , the economics of the operation will dictate the containment.

The uranium dioxide is fluorinated with hydrogen fluoride gas (HF). This may be done in screw reactors, vibrating trays, or in "fluidized" bed reactors, but in each case the operation is dusty. The gaseous reactant must be in excess and trapping of the effluent mixture of gas and powder is required. Bag filters are subject to failure and to rupture, particularly when the effluent gas is HF, and losses to the environment are comparatively high. Losses of very fine particles will, in any case, occur and these may be airborne for considerable distances and be detectable in environmental samples.

DELETED

005
6.2p)

The primary purpose of the metal production is to manufacture slugs for fueling plutonium producing reactors or for power reactors of the gas cycle type (EGCR) which utilize normal or near normal uranium. Various extrusion and machining operations may be conducted to prepare the shapes needed and "canning" of the slugs may be done at the same plant. Currently, the slugs for Hanford Operations are clad with aluminum at the site, rather than at Fernald. Machining and extruding result in chips and wastes of various forms which ultimately have to be reworked or discarded. Economic considerations will dictate the degree to which salvage of this type of waste is conducted.

If metal somewhat enriched in U^{235} is desired, the feed will come from a separations plant and may be in the form of UF_6 . In this case it is reacted with water to produce UO_2F_2 or reduced with hydrogen to give UF_4 directly, as outlined in the discussion of waste handling under the gaseous diffusion process. At low enrichments, circa 2% U^{235} , criticality considerations still permit comparatively large scale operations and the same remarks made above about losses will apply. Containment would be somewhat more stringent because of the higher economic value of the material.

The characteristics of such a plant which are of interest for the present discussion are as follows:

- a. The operation is "dirty". Losses of uranium to the environment are high.
- b. Effluents, other than uranium, are nitrate or nitrogen oxides and fluoride.
- c. An operation of this kind can mask very effectively effluents from another type of uranium handling installation in the vicinity.
- d. The amount of uranium handled is large and control of diversion, as for feed for a centrifuge plant of modest productivity, may be difficult.

2. Weapons Parts Fabrication.

The Y-12 metal production plant is an example of this type of operation. Most of the reactions discussed under the Fernald plant are conducted in this facility. The feed will, in general, be received in the form of uranium hexafluoride (UF_6) in "always safe" containers from the diffusion cascades. The U^{235} content is 93% or better and criticality requirements make it necessary to conduct the operations under a batch basis. The UF_6 is "knocked out" in water in the presence of a sufficient concentration of aluminum salt to complex the fluoride, the uranium precipitated, filtered, and processed to UF_4 . This salt is then reduced to metal in bombs with metallic magnesium or calcium. The waste MgF_2 or CaF_2 are leached to recover uranium and sent to the burial ground.

The enriched metal is cast, machined, rolled, or extruded as necessary for forming weapons parts. Recovery of the scrap is a necessary part of the process since the intrinsic value of the material is high at this enrichment. A further consideration is that the U^{234} isotope has been enriched along with the U^{235} and the alpha activity is much higher than normal uranium. The health hazard is accordingly much greater. Containment precautions are not as stringent as for plutonium and losses are readily observed in the environment. The high quality filters on exhausts still permit the passage of very small particles which accumulate and are detectable on vegetation.

The Y-12 weapons plant also handles large amounts of depleted uranium which is used in metallic form for the fabrication of "tamper" for weapons. This material is comparatively carelessly handled and it tends to cover the enriched material in environmental samples. This provides an example of the "masking" possible by an effluent which would override one present in smaller amount.

A plant handling highly enriched uranium for manufacture of power reactor fuel elements would not differ in effluents from a weapons parts shop. Such a plant would, as does the Y-12 installation, receive recovered uranium from power reactor fuel elements and blend it with more highly enriched material before reprocessing into new cores. If, in addition, reprocessing of weapons parts for modernization were permitted under the disarmament agreement, the problem of "in-plant" inspection and control to prevent diversion would be complex. This problem is outside the scope of this report and there exist reports written to deal specifically with this phase of the inspection problem.

A further complication of the picture around a plant which makes highly enriched U^{235} into reactor fuel elements could be the presence of thorium and of U^{233} recovered from thorium containing fuels and from thorium blankets on breeder reactors. The latter isotope is highly radioactive, being an alpha particle emitter with a half life on the order of that of U^{234} . Handling precautions would have to be stringent. The presence of this isotope in environmental samples is, however, quite readily detected and it should not interfere with the primary diagnosis of the presence of uranium enriched in U^{235} .

The characteristics of a weapons parts shop, or one manufacturing power reactor fuel elements are:

- a. Moderate but detectable loss of enriched uranium. This is enriched in U^{234} and will carry U^{236} if diffusion plant feed has been reactor processed uranium.
- b. Highly depleted uranium will be observed readily if uranium tampers have been manufactured.
- c. No way of distinguishing, by means of effluents, between weapons parts manufacture and fabrication of reactor cores.

Non-Fissionable Nuclides

A number of isotopes of other elements are of importance to a weapons program. Dr. R. J. Brouns has discussed those which are reactor produced and which are used in initiators.* These nuclides are radioactive and the

*"Studies of Inspection and Control Systems, Number II", HW-68000 (Secret Limited), R. J. Brouns, June 1, 1960.

possibility of detection at least exists from that standpoint, although tritium may, because of high background from heavy water-moderated reactors, present difficulties. Two other materials which are non-radioactive deserve consideration from the standpoint of inspection and control.

Highly enriched Li^6 is used in thermonuclear weapons, usually as the deuteride (LiD). This isotope captures a neutron, and tritium and helium are produced; ($\text{Li}^6 (n, \alpha \text{C})$). The capture cross section of the other isotope of lithium is very small and highly enriched Li^7 is of potential utility as a liquid metal heat transfer medium for reactors. Lithium fluoride is a component of the fuel melt for proposed fused salt reactors. For both uses the purity of the isotope would have to be very high to reduce neutron loss. Thus, there would be legitimate reason for the operation of a plant to separate Li^7 and it would be necessary to insure that it was not operated to produce Li^6 in high purity.

Plants to produce heavy water are a normal part of an atomic energy complex because of the importance of deuterium oxide (D_2O) as a moderator. The Savannah River reactors, for instance, are heavy water-moderated. The large tonnage of heavy water required for such operations makes the diversion of sufficient deuterium for production of thermonuclear weapons very easy.

Deuterium Production

The chemical and physical properties of hydrogen and deuterium differ sufficiently because of the large mass difference that a variety of separation methods have been used to produce heavy water. The natural abundance of the heavy isotope is about 0.015% in water. Multistage electrolysis of water was conducted in Norway and at Trails, B. C. during the war. This process is appealing only when there is an excess of cheap power available. The electrolytic process was supplanted by large scale distillation of water carried out under somewhat reduced pressure. The difference in the vapor pressures of H_2O and D_2O is sufficient to make this separation process effective. The deuterium containing molecule is the less volatile. The initial concentration by distillation was followed by electrolysis as a "tepping" process to give D_2O with purity greater than 99%.

Distillation has, in turn, been supplanted by a cheaper and more efficient separation scheme which involves the "dual temperature" exchange between water and hydrogen sulfide, H_2S . If these two materials are in contact, an equilibrium exists

and the deuterium is shared between the two compounds. The equilibrium is shifted to the right at low temperatures and to the left at higher temperatures.

The feed water is contacted cold with hydrogen sulfide gas and the water transfers part of its deuterium content to the gas. The slightly depleted water is then rejected and the hydrogen sulfide pumped to a second column reactor in which the water is heated. In this column, the deuterium is stripped from the gas and the water leaving this stage is somewhat enriched in deuterium. This process is continued by pumping the enriched water to the next stage where it is cooled before contacting H_2S . The hydrogen sulfide gas serves only as a transfer medium for the deuterium and is reused. After a sufficient number of stages, the deuterium content has been raised to about 2% in the water. Further concentration is then accomplished by electrolysis or by distillation. The latter procedure is now favored and is used at Savannah River for preparation and for repurification of moderator.

Hydrogen sulfide would not appear as an effluent from such a plant, but would be burned to sulfur dioxide before release. Hydrogen sulfide is so toxic that this precaution would be necessary. Sulfur dioxide is a common industrial air pollutant and its presence would not be characteristic of a heavy water production plant. The depletion of deuterium is so slight in the exit water that there would be no hope of using this as a tool to identify such a plant. The deuterium content of natural waters is so variable that such a method would have very low sensitivity.

Furthermore, there would seem to be no reason for not declaring a heavy water plant. The production of such a plant for legitimate purposes is so great that the diversion of the relatively small amounts required for a thermonuclear weapons program would offer no difficulty. A specific gravity measurement, refractive index, or some other physical test would be required to prove that the container held heavy water and not ordinary water.

Lithium Isotope Separation

Numerous methods of enriching Li^6 have been investigated. The most successful process and the one which is used in the United States involves an isotopic exchange between lithium hydroxide in aqueous solution and lithium metal dissolved in mercury. The equipment used is not highly specialized and the process can be operated on a small or large scale, as desired. It can be operated to produce very high enrichment of Li^6 , starting with 7.5% in normal material, or may be adapted also to production of very pure Li^7 .

The process is outlined in Figure 8. Lithium feed is supplied in the form of $\text{LiOH}\cdot\text{H}_2\text{O}$. The initial purification step is to electrolyze the lithium into mercury in a tray. At this point the small amounts of other alkali metals and impurities are wasted to sewer. The amalgam is then decomposed by contacting with water and an appropriate catalyst to provide very pure lithium hydroxide feed for the cascade. This feed is introduced in the column at an appropriate point, the portion above the feed point being the "enricher" and below it the "stripper".

DOB
6.2 (1)

DELETED

At the product end, the amalgam is decomposed in the "desorber" by contacting with water in the presence of carbon as a catalyst. Some of the hydroxide is recycled and another portion is removed, neutralized with hydrochloric acid to form lithium chloride which is concentrated by evaporation, dried, and fed to the electrolysis cells. Fused lithium chloride is decomposed by electric current to produce chlorine and lithium metal highly enriched in the light isotope. The metal is reacted with deuterium produced by the electrolytic decomposition of heavy water to prepare lithium deuteride.

DELETED

DOB
6.2 (2)

A portion of the reject stream is withdrawn, evaporated, and $\text{LiOH}\cdot\text{H}_2\text{O}$ filtered off. This "tails" material is sold on the market and for a time, at least, commanded a premium in the lithium chemical trade because of its very high chemical purity. It typically contains about half the normal amount of Li^6 or about 3.5%.

Losses of material occur at several places in the operation. The wastes from the initial purification step will contain the other alkali metals--sodium, potassium, rubidium, and cesium. Effluent water may be unusually high in the range of these elements. Waste from the mercury scrubber may carry enriched Li^6 and will certainly be high in mercury. The major loss is at the tails handling point where filter blankets and filter presses are washed and where a large scale packaging operation is carried on.

The handling of lithium salts offers no more than the normal hazards of a chemical operation and only the economics of the operation will dictate the degree of housekeeping and of recovery of material. The enriched material is, of course, handled with greater care because of its increased intrinsic value.

DELETED

Doc
6-2a

FIGURE 8

Lithium Isotope Separation Cascade

The airborne effluents are dust from lithium hydroxide handling and mercury vapor. Lithium salts are water soluble and accumulation in the environment would be significant only in an arid region. Mercury vapor may be detected by accumulation in vegetation for some miles around a large operation. This in itself is not diagnostic because mercury may be used in large amounts in other processes, such as the production of sodium and potassium hydroxides where the "mercury cell" for the electrolysis is a standard item.

Aqueous effluents provide a ready method of detecting such a plant. Effluent from the U. S. plant is typically lithium which is depleted in the light isotope and, as such, may be detected many miles down river from the installation. A plant like that of Foote Mineral Company at Sunbright, Virginia, which processes ore to produce lithium salts, contributes lithium to the river in that vicinity which, although normal in isotopic composition, is significantly higher in concentration than normal river water.

Detection Capability

In the discussion above of individual types of facilities which handle uranium for processing to a different chemical form, for preparation of metal or which enrich U^{235} , it has been indicated that the effluent common to these operations is uranium. This may be airborne as particles or may be emitted in aqueous wastes. Other effluents will be of a less characteristic nature and might well be those which would be found in the vicinity of various chemical manufacturing operations.

The utilization of this characteristic effluent to locate and possibly to identify the type of facility requires a comprehensive scheme of sampling. This may well be the use of environmental samples in which uranium has accumulated or been preferentially concentrated. Soil, grass, deciduous or evergreen leaves, tree bark, silt, etc., may be sample sources. In addition, an inspection scheme would necessarily make use of deliberately taken air filter samples. Highly specialized laboratory facilities are required to make successful implementation of such a program possible.

No detailed discussion of the proposed detection scheme will be given. The reader is referred to pages 70-116 of the report by R. J. Brouns (op cit.) which gives a detailed discussion of the proposed sampling scheme, the personnel required, and the time necessary to implement such a program. The objective in this discussion here will be rather to indicate what such a sampling program can tell about uranium handling facilities and to estimate the possibility of detection and location of undeclared and clandestine plants.

The following pieces of information can be obtained from environmental uranium sampling:

1. Total uranium above some background.
2. Alteration in U^{238}/U^{235} ratio indicating enrichment or impoverishment.
3. Abnormalities in the U^{234} content.
4. Presence or absence of U^{236} .
5. Presence of U^{233} .

U^{236} is not naturally occurring but is produced in reactors by the non-fission capture of a neutron by U^{235} . It is enriched in a separation scheme along with U^{234} and U^{235} . It may be very high--several percent--in uranium recovered from enriched reactor cores which have had high burn-up. U^{233} is produced by irradiation of Th^{232} introduced either as a fuel element component or as a blanket in a "breeder reactor". It is fissionable and has utility, therefore, for weapons components and for power reactor fuel elements. It is an active alpha emitter and must be handled with greater care than enriched uranium, although probably not with the same containment required for plutonium.

The following cases are likely to be observed in the environment of plants handling uranium:

- Case 1. Normal uranium, no U^{236} , concentration higher than background: Indicates ore processing, separations plant feed preparation, reactor fuel element preparation.
- Case 2. U^{235} (and U^{234}) enriched, no U^{236} : Indicates separations plant not using reactor irradiated feed, or metal (weapons) preparation from enriched material.
- Case 3. U^{235} (and U^{234}) depleted, no U^{236} : Indicates separations plant "tails" and may be separations plant or weapons "tamper" fabrication.
- Case 4. U^{236} present: Indicates handling of reactor processed uranium. Applies to Cases 2 and 3. May indicate, if U^{235} is depleted (case 3), preparation of separations plant feed from reactor "tails".
- Case 5. U^{235} normal, U^{234} enriched, U^{236} present: Indicates mixture of enriched and impoverished material, from separations plant or weapons plant.
- Case 6. U^{233} detected: Indicates processing of thorium bearing fuel elements, of thorium blanket from breeder or fabrication of weapons parts or reactor fuel elements.

It is evident that it is possible to determine the general class of uranium being handled, but there are, in every case, alternative interpretations if evidence is based solely upon the detection of uranium and determination of its isotopic composition. Additional evidence from other effluents may narrow the choices somewhat. High fluoride in the immediate vicinity of a facility might indicate a separations cascade, a feed preparation operation, or it might equally well indicate an aluminum plant and the fluoride would be from cryolite dust (Na AlF_4). Similarly, high nitrate would tend to indicate a denitration of uranyl nitrate, but it might well have come from some other chemical facility located in the same industrial complex. Unless unlimited right of entry is granted, then recourse must be had to other means of collecting information. Materials entering and leaving the plant must be monitored, power input determined, etc.

Two methods of evading inspection and detection are evident. A facility could be constructed or modified with the objective of attaining such complete containment that no uranium could be detected in gaseous or liquid effluent. It is a corollary to this that the amount of material handled would have to be small. Large scale filtration of air will not be perfect and containment of very small particles will not be possible. Any losses of uranium as hexafluoride in trace amounts would be difficult to prevent on large scale operations. Aqueous wastes would be more readily controlled. Such wastes could be pumped into underground storage or into underground water channels. Ion exchange resin treatment of wastes which has first been somewhat concentrated would also probably be very effective.

It would probably be more feasible to make a separations plant using centrifuges effluent free, than to prevent detectable losses from feed preparation or metal fabrication on anything but a very small scale. By far, the most attractive and more easily attained situation would be to put the clandestine facility in or near to a large scale operation handling uranium. This would be the more difficult case to detect. There is no apparent means of detecting the difference between a weapons parts fabrication and the construction of reactor cores from highly enriched uranium by environmental studies.

The following table will give the best estimate which can presently be made for detecting and locating undeclared facilities operated in what would be considered a normal manner and clandestine facilities operated with maximum attention given to concealment. The distance limitation is arbitrary. The closer one can get to a facility, the better the chance of positive detection.

DETECTION CAPABILITIES FOR HIDDEN PLANTS

Type of Plant	Normal (1)		Clandestine (2)		Means of Detection
	Close 0-5 mi.	Distant > 5 mi.	Close 0-5 mi.	Distant > 5 mi.	
Diffusion Cascade	Positive	Probable	Probable	Dubious	Uranium isotopic comp.
Centrifuge Cascade	Probable	Dubious	Dubious	Very unlikely	" " "
Feed (UF ₆) Preparation	Positive	Probable	Probable	Dubious	" " "
Metal Prep. (Near Normal Conc.)	Positive	Probable	Probable	Dubious	" " "
Metal Prep. (High Enrichment)	Positive	Probable	Dubious	Very unlikely	" " "
Deuterium Enrichment	Dubious	Impossible	Impossible	Impossible	Deuterium
Lithium Isotope Separation	Positive	Positive	Probable	Very unlikely	Altered isotopic comp. of lithium in aqueous effluent.

(1) A normal facility is defined as having been constructed and operated in ordinary fashion for at least a year before inspection begins. It is assumed to be of large size.

(2) A clandestine facility is one which was constructed or modified to avoid detection. It is anticipated that it will be of smaller size and may be located in the vicinity of a "normal" operation, which would provide masking for effluents if containment is not perfect.

~~SECRET~~
~~TITLES~~

BIBLIOGRAPHY

The reports cited here are referred to in the text, or are pertinent to the subjects discussed.

Production of Enriched Uranium for Nuclear Weapons by Nations X, Y and Z by Means of the Gas Centrifuge Process, R. A. Levin, D. E. Hatch, and E. Von Halle, KOA-662, February 26, 1960, SECRET.

A Study of Isotope Separation with Sub-Critical Centrifuges, F. J. Neugebauer, D. H. Marquis, R. Plunkett, E. Sternlicht, TIS 60GL15, January 15, 1960, SECRET.

Studies of Inspection and Control Systems, Number II, The Detection of Hidden Facilities, R. J. Brouns, HW-68000, June 1, 1960, SECRET.

Capability Estimate, Detection of Uranium and Lithium Isotope Separation Facilities, A. E. Cameron, December 29, 1959, ORNL 59-12-76, SECRET.

Capabilities and Requirements for Detecting Hidden Plants, R. J. Brouns and A. E. Cameron, March 30, 1960, ORNL 60-3-115, SECRET.

Conversion of Uranium Trioxide to Uranium Hexafluoride, C. S. Cronan, March 23, 1959, Chemical Engineering, Pages 140-143, UNCLASSIFIED.