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1986 Summer Meeting of the AIChE
Boston Massachusetts
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Oak Ridge Gaseous Diffusion Plant*
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Oak Ridge, Tennessee

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# INTEGRATION OF THE AVLIS PROCESS INTO THE NUCLEAR FUEL CYCLE

## ABSTRACT

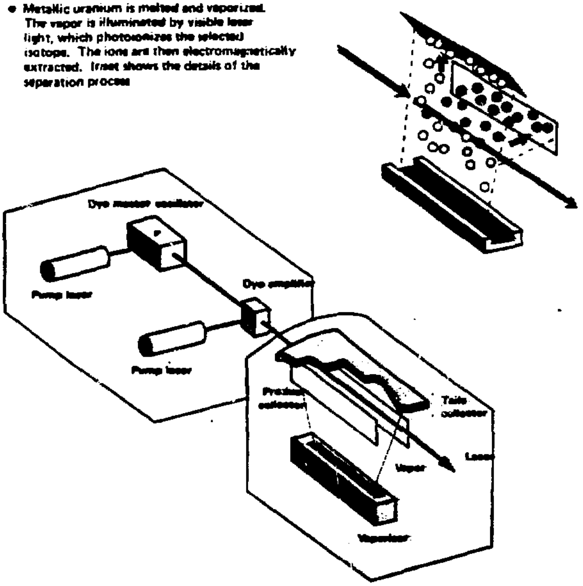
In June 1985, the United States Department of Energy (DOE) completed an extensive evaluation of advanced uranium isotope separation processes and selected the atomic vapor laser isotopic separation (AVLIS) process as its advanced uranium enrichment process. AVLIS utilizes uranium metal feed and produces enriched uranium metal as a product, and because of this does not need to have $UF_6$ as an intermediate material. More cost effective and chemically efficient uranium processing options are available for development to production status. This paper will briefly describe the AVLIS process and present a general overview of alternative chemical processing methods being investigated to ensure a smooth and cost effective integration of AVLIS into the nuclear fuel cycle.

## AVLIS PROCESS DESCRIPTION

The AVLIS process being developed at Lawrence Livermore National Laboratory in Livermore, California with support from Martin Marietta Energy Systems in Oak Ridge, Tennessee exploits the fact that the different electron energies of $U^{235}$ and $U^{238}$ absorb different colors of light. In the AVLIS process, lasers are tuned to emit a combination of colors that will be absorbed only by a $U^{235}$ atom, which subsequently emits an electron, allowing the charged $U^{235}$ ion to be selectively separated using electromagnetic fields. The AVLIS process includes two major component systems: a laser system and a separator system. The schematic in figure 1 illustrates the process.

The Laser System. The laser system uses two types of lasers: dye lasers that generate the light used for photoionization of the uranium, and copper-vapor lasers that pump (energize) the dye lasers. Powerful green-yellow light from electrically driven copper-vapor lasers is converted to red-orange light in the dye laser. This red-orange light is tuned to the precise colors that are absorbed by $U^{235}$ but not by $U^{238}$. This laser-pumped-laser scheme is required because dye lasers cannot be powered directly by electricity.

The Separator System. Uranium metal is melted by means of an electron beam that creates an atomic vapor stream of $U^{235}/U^{238}$. The tuned dye laser beams are passed through the vapor stream where they photoionize the $U^{235}$ atoms. An electromagnetic field deflects the selected photoions to the product collector where they condense. The $U^{238}$ atoms, which are unaffected by the color-selective laser beams, pass through the product collector to condense on the tails-collector. The enriched uranium liquid metal condensate flows out of the separator to be cast and stored in solid metallic form for eventual conversion to uranium oxide for reactor fuel rods.



- Metallic uranium is melted and vaporized. The vapor is illuminated by visible laser light, which photoionizes the selected isotope. The ions are then electromagnetically extracted. Inset shows the details of the separation process

Figure 1

ATOMIC VAPOR LASER ISOTOPE SEPARATION (AVLIS) PROCESS

<u>Basic Advantages of the AVLIS Process.</u> The AVLIS process is inherently more efficient than conventional separation processes. Both gaseous diffusion and gas centrifuge techniques rely on isotopic mass differences (about .01 for uranium), resulting in low enrichment efficiency. In contrast, AVLIS's fundamental atomic selectivity ($U^{235}$ atoms collected per $U^{238}$ atom collected), which uses the isotopic color signature, is greater than 10,000. Because of this,

AVLIS can achieve a high degree of isotopic enrichment, increasing from 0.7% to 3% to 4% $U^{235}$ in a single stage, using relatively little capital equipment. This high enrichment efficiency, together with low energy requirements for vaporization and laser-beam generation, results in the advantageous combination of low capital, low energy, and low operating costs for AVLIS.

Uranium Processing. Currently commercial enrichment methods, and consequently the world's nuclear fuel cycle, are operated within a $UF_6$ in--$UF_6$ out mode. The feed is prepared from the ore concentrate by feed converters under contract to the utility owners, and the enriched product $UF_6$ is processed to nuclear fuel oxide also under contract to the utilities by the fuel fabricators. Since AVLIS is a metal-based process; i.e., uranium metal is required as feed to the separator vessel and an enriched uranium metal product is produced, some changes to integrate AVLIS into the nuclear fuel cycle in both the feed supply and product handling systems are advantageous. For a 14 million separative work unit (SWU/yr) AVLIS enrichment plant, approximately 17,000 metric tons of uranium feed will be required and more than 3000 metric tons of isotopically enriched uranium metal will be produced. During DOE's advanced isotope selection process, part of the basic decision criteria required a $UF_6$ in--$UF_6$ out method of operation. This requirement penalized the AVLIS economics because head end processing was required to convert $UF_6$ to U-metal, and the product metal had to be converted to $UF_6$. It was known some alternative forms other than $UF_6$ for feed and product could provide AVLIS with the opportunity to reduce the overall fuel cycle costs. Soon after AVLIS was selected, a team of uranium processing experts was assembled, and a systems analysis study was initiated. The purpose of this study was to evaluate alternative uranium processing methods and identify the most cost effective feed and product conversion processes, starting with uranium ore concentrate and ending with a qualified ceramic grade uranium oxide for producing fuel pellets. Participants included personnel from Lawrence Livermore National Laboratory, Martin Marietta Energy Systems, Bechtel National, Argonne National Laboratory, Georgia Institute of Technology, Kaiser Aluminum and Chemical Company, and Kaiser Engineers, Inc. The study was constrained only by options that: 1) were possible and sensible technically; 2) could meet the programmatic schedule; and 3) were most able to interface to industry. With regard to schedule, current plans are to deploy AVLIS sometime during the mid 1990's. Since feed and product conversion contracts can easily run 5-10 years, and sometimes even longer, it is important to consider AVLIS uranium processing issues now. This allows not only adequate time for conversion contracts to be negotiated, but also for developing alternative processes, making any capital modifications or building new plants, and obtaining any necessary regulatory approvals that may be required to be able to meet AVLIS uranium processing needs.

A general description of the original baseline processes (those that involved $UF_6$ for feed and product) and the alternative processing methods studied, along with advantages, disadvantages, fuel cycle issues, and general economics will be briefly presented in this paper. Processes studied for feed conversion included: 1) thermite reduction; 2) electrolytic reduction; and 3) pyrometallurgical processes. Product conversion alternatives studied included: 1) fluorination to $UF_6$; 2) dissolution and solvent extraction followed by ADU precipitation and calcination of the uranyl nitrate; 3) direct denitration of the uranyl nitrate; and 4) pyroredox or distillation followed by steam oxidation. Other methods were investigated and eliminated prior to extensive consideration because they did not meet the three constraints imposed on the study. These methods will be briefly mentioned in the paper.

## FEED CONVERSION BASELINE DESIGN

AVLIS enrichment operations are generally insensitive to many impurities, and since product purification is planned, higher impurity levels than have been demonstrated and accepted for reactor metal fuel uses can be accepted for AVLIS feed. This situation provides the opportunity to investigate metal production processes that may not be acceptable for other applications requiring high purity. Table I presents a preliminary metal feed specification to the AVLIS separator. It is the intent of the AVLIS program to continue to evaluate feed specifications to see where other material limits may be relaxed, particularly where it offers the opportunity to further reduce the cost of feed production.

<u>Thermite reduction process--UF6-to-metal.</u> The well-developed industry standard process for preparation of uranium metal is the magnesiothermic reduction process--so called "bomb" by the characteristic ignition that takes place within the reactor when reducing the uranium tetrafluoride ($UF_4$) to uranium metal. (Figure 2) In current practice, metal is produced by first reducing uranium hexafluoride ($UF_6$) to $UF_4$ with hydrogen typically in a flame reactor according to the following equation:

$$UF_6 + H_2 \Rightarrow UF_4 + 2HF$$

Although the thermodynamic equilibrium, even at room temperature, is considerably driven toward the formation of $UF_4$ solid, the minimum wall temperature required for satisfactory initiation of the reaction is 425°C. The exothermicity of the reaction increases the temperature rapidly and the reaction goes to completion essentially as a flame. Extensive off-gas treatment systems are required to recover the anhydrous HF which is a major by-product of this reaction and to eliminate trace quantities from vent gases.

Table I

U-METAL SPECIFICATION

- COMPOSITION: THE MATERIAL SHALL CONFORM TO THE FOLLOWING PERCENTAGES BY WEIGHT, DETERMINED BY METHODS AGREED UPON BY PURCHASER AND VENDOR.

| | |
|---|---|
| URANIUM | 99.00 MINIMUM |
| CARBON | 80 PPM MAXIMUM |
| CHROMIUM | 200 PPM MAXIMUM |
| COPPER | 200 PPM MAXIMUM |
| IRON | 200 PPM MAXIMUM |
| NICKEL | 200 PPM MAXIMUM |
| SILICON | 100 PPM MAXIMUM |
| ALL OTHER METALLICS (EACH) | 50 PPM MAXIMUM |

- CONDITION: DERBIES SHALL BE SURFACE CLEANED AND ACID PICKLED TO REMOVE ALL SURFACE SLAG AND SALTS.
- DENSITY: ≥ 18.0 gms/cc

Magnesium metal in slight excess of the stoichiometric ratio is added to the $UF_4$ in a fully encapsulated steel vessel (reactor) which is lined with magnesium fluoride slag or graphite. The mixture is electrically heated to about 650°C at which point the exothermic reaction which produces magnesium fluoride and uranium metal initiates and the temperature rises rapidly to approximately 1400°C. About 96% metal product yield in the form of a regulus or so-called "derby" is achieved. Product separation of the uranium metal from the magnesium fluoride slag is achieved by gravity flow of the much denser uranium molten metal through the molten slag. The reaction vessel is removed from the furnace, cooled, disassembled, and emptied with the uranium derby breaking away from the slag chunks. Slag is ground for recovery and recycle for use as liner material. Uranium may be recovered from the slag by wet or dry (with fluorine) leaching.

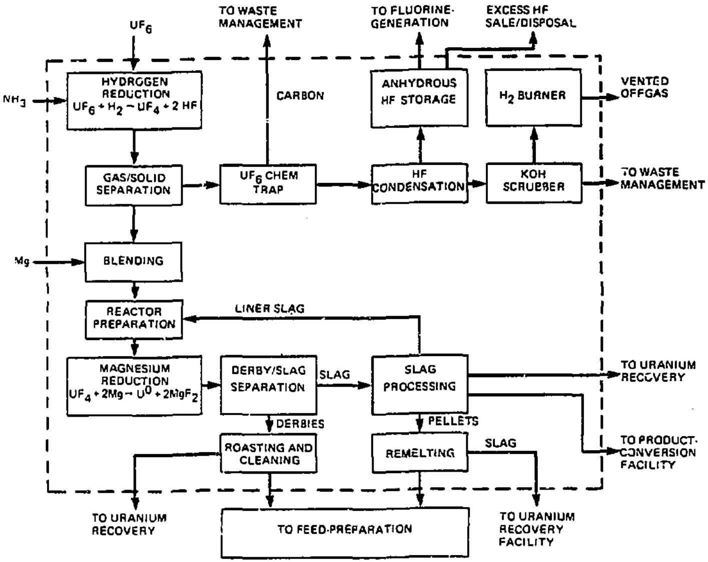


Figure 2
AVLIS BASELINE FEED CONVERSION FLOWSHEET

While this process has been in industrial application for decades, primary disadvantages of the thermite reduction process include: 1) the high value starting materials ($UF_6$, Mg); 2) the large volumes of magnesium fluoride wastes which are generated and the associated escalating waste costs; 3) the batchwise nature of the process, although some R&D work has been conducted on making it a continuous process; and 4) the need for well converted feed material (>96% $UF_4$ content) with satisfactory density and particle size and acceptable amounts of impurities in order to achieve economic yields. It was these primary disadvantages that were considered in the search for a new candidate process that could provide low cost uranium metal to the AVLIS process and meet the required plant development schedules.

<u>Thermite reduction process--$UF_4$-to-metal.</u> Since the standard industrial process for producing $UF_6$ includes hydrofluorination of $UO_2$ to $UF_4$ (figure 3), it seems unnecessary to produce $UF_5$ only then to reduce the material back to $UF_4$ prior to metal reduction. The $UF_4$ produced by hydrogen reduction of $UF_6$ is essentially 100% $UF_4$ with very low impurity levels and with good density. This feed material produces good metal yields of low impurity level. Some industrial methods for $UF_6$ production from ore concentrate include an up-front solvent extraction process which is expected to provide satisfactory metal purity directly from the $UF_4$ intermediate product. This will eliminate the need to produce $UF_6$ and result in measurable cost savings for feed production.

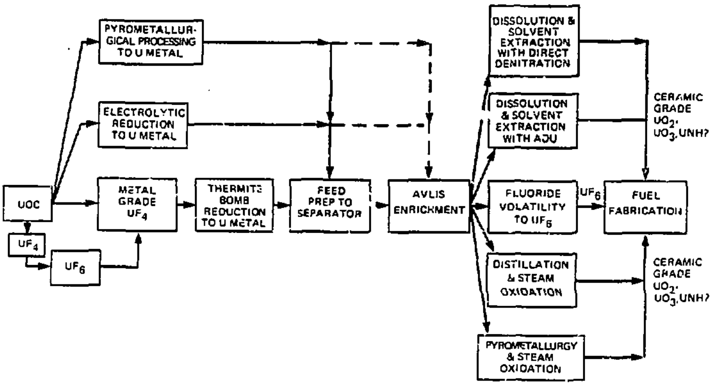


Figure 3

AVLIS URANIUM PROCESSING ALTERNATIVES

## FEED CONVERSION ALTERNATIVES

Shortly after AVLIS was selected for further process development, a systems analysis study was initiated to reevaluate methods for providing uranium processing for AVLIS. It was known with a reasonable level of confidence that $UF_6$ technology did not offer the lowest overall fuel cycle cost for an AVLIS enrichment based fuel cycle. The systems analysis study looked at a broad range of potential technologies for both feed and product processing. The better technologies, based on technical maturity, demonstrated performance, industrial interface, and required RD&D resources were selected to develop scoping level, plant-scale flowsheets, hardware designs, facility layouts, and operating requirements. Production plant capital and operating costs were roughly estimated from these designs. Based on this study, those uranium processing technologies which provided the lowest overall cost, most mature technology and most easily met plant deployment schedules were identified. Potential advanced AVLIS uranium processing feed conversion methods were evaluated by starting with ore concentrate (primarily $U_3O_8$) as an initial feedstock rather than $UF_6$ to provide metal feed to an AVLIS production plant.

Electrolytic Reduction. Electrolytic reduction has long been an established industrial process for producing metals. Electrolytic reduction of uranium in fact, has been reported in the literature for many years with by far the most significant work being performed by the Mallinckrodt Chemical Company for the Atomic Energy Commission (the predecessor of the DOE) during the late 1950's and early 1960's[1,2,3]. The basic chemical reactions in the Mallinckrodt cell are as follows:

$$UO_2 + 2C \Rightarrow 2CO + U^{+4} + 4e^- \quad \text{(At anode)}$$

and

$$UF_4 + C \Rightarrow CF_4 + U^{+4} + 4e^-$$

$$U^{+4} + 4e^- \Rightarrow U^0 \quad \text{(At cathode)}$$

or

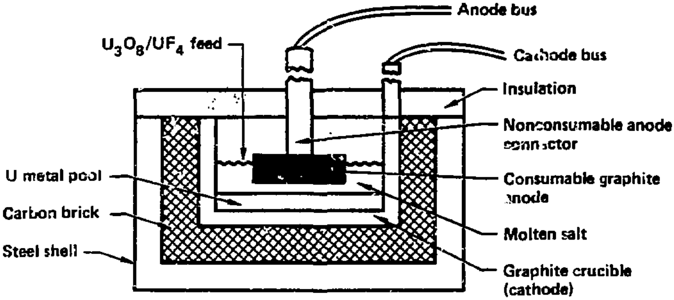
$$UO_2 + 2C \Rightarrow 2CO + U^0 \quad \text{(Overall)}$$

$$UF_4 + C \Rightarrow CF_4 + U^0$$

The operating temperature may be selected above 1133°C to allow recovery of U as a molten metal or at a lower temperature to produce a dendritic U-solid. For this latter case, the temperature must be kept above the electrolyte freezing point. The Mallinckrodt effort, which was operated at a cell temperature of about 1200°C, was successful in producing more than 50 tons of uranium metal by electrolytically reducing a mixture of partially converted uranium dioxide ($UO_2$) and $UF_4$ in a molten salt electrolyte bath consisting of $UF_4$, lithium fluoride (LiF), and barium fluoride ($BaF_2$). The $UF_4$ is needed to provide sufficient solubility of the uranium oxide in the molten salt bath. The Mallinckrodt cell was operated such that significant quantities of the U-metal were produced from $UF_4$. This is a disadvantage of the Mallinckrodt operation since one objective of the advanced processes for supplying feed was to minimize the required uranium processing prior to metal reduction. For large requirements of $UF_4$ in the cell feed, electrolytic reduction then loses part of its advantage over thermite reduction of $UF_4$.

During operation of the Mallinckrodt cell, low current efficiencies of about 20-40% were obtained due to several factors:

1. Parasitic current flow to the graphite side walls;
2. A substantial cyclic valence interchange from $U^{+4} + e^- \rightleftharpoons U^{+3}$
3. A significant back reaction of the uranium with the anode gas.



$UO_2 + 2C \rightarrow 2\,CO + U^{+4} + 4e^-$ At anode

$U^{+4} + 4e^- \rightarrow U^0$ At cathode

$UO_2 + 2C \rightarrow 2\,CO + U^0$ Overall

Figure 4

SCHEMATIC OF ELECTROLYTIC CELL FOR PREPARATION OF U-METAL

A cell schematic and a flowsheet for the electrolytic production of uranium metal are shown in figures 4 & 5, respectively. The electrolytic cell consists of a graphite container surrounded by low density insulation. Between the graphite and the low density insulation is a lining of high density refractory. The graphite provides nominal containment of the bath while the dense refractory provides ultimate containment of the bath by allowing establishment of a freeze line in the refractory itself. Uranium oxide mixed with $UF_4$ is fed to the cell continuously. While the solubility of the oxide in the bath is relatively low, studies indicate the solubility may be improved by as much as a factor of 2-5 by using $CaF_2$ instead of $BaF_2$ in the electrolyte salt[1,4]. Uranium is formed at the cathode and coalesces in the cell bottom and is recovered from each cell by vacuum tapping. The cell and plant were designed for total containment of the uranium as required by current standards. While the Mallinckrodt work served as a valuable experience base, many advancements in electrolytic technology have taken place since the Mallinckrodt efforts, primarily in the aluminum industry. It is expected that by incorporating many of these technology advancements in the uranium application, current efficiencies may improve to 40-80%.

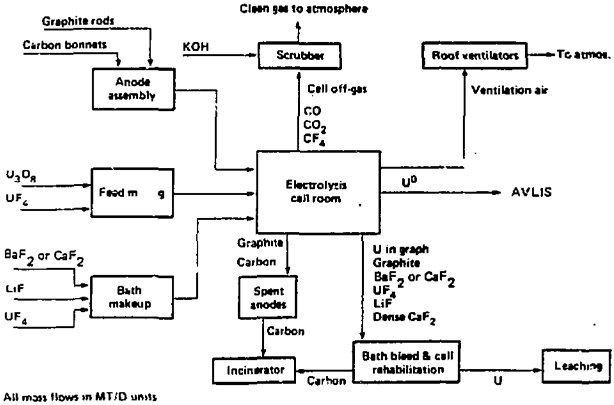


Figure 5

ELECTROLYTIC REDUCTION OF $U_3O_8$ TO U-METAL

As each cell reaches a predetermined time period, it will be shutdown and refurbished. During refurbishment all materials are recovered and recycled to the maximum extent possible to minimize waste disposal requirements. The recycled materials include bath chemicals and the graphite container which should have a lifetime greater than the planned operating period between shutdown. Waste streams from the process include CO and $CO_2$ from the cell off-gas and graphite, LiF, $UF_4$, and U from the refurbishment step.

The electrolytic process has several advantages over the thermite reduction process. First, it is a one step continuous process which has a large portion of the feed as a lower value added material ($UO_2$ or UOC where UOC stands for uranium ore concentrate). Next, the process is continuous rather than batch resulting in lower operating costs. Also the product is in a liquid form which may have advantages in processing it further in the AVLIS system. A further advantage of the electrolytic process is that it generates significantly less quantities of waste materials than the thermite process, particularly solid waste. This means the process has a low sensitivity to increasing waste disposal costs. The fact that the electrolytic process is based on an industrialized process for making aluminum is also a significant advantage. Disadvantages of the process are that it requires: 1) a high operating temperature (1170-1200°C); 2) careful control of the oxide feed rate, 3) requires some quantity of $UF_4$ in the feed, and results in additional impurities in the AVLIS feed. This latter item will only be a disadvantage if it proves to exceed AVLIS feed specifications.

In the current AVLIS study, the cost impact of starting with different feed materials was evaluated. To provide the lowest overall cost, the process should start with the lowest value added material which is UOC or $U_3O_8$. An evaluation was made which indicates that UOC may be a feasible starting material for the electrolytic process. However, UOC has a higher impurity level than $UO_2$ which won't affect the electrolytic process but potentially impacts the AVLIS process itself. Although preparation of uranium metal by electrolytic reduction appears promising, a significant development program would be required to demonstrate the process and the improved flowsheet.

<u>Pyrometallurgical processes.</u> As defined in this study, pyrometallurgical reduction processes are characterized by reduction of the uranium compound using a metal agent in the presence of a molten salt. Possible reductants are in the alkaline earth (Mg, Ca) or alkali metal (Li, Na) families. Pyrometallurgical process have been applied commonly in the Kroll process for titanium and zirconium production. For the uranium systems analysis study, two methods were considered: 1) reduction of uranium trichloride, and 2) direct reduction of uranium oxide. Flowsheets for both are presented in figures 6 & 7. The process is operated batchwise and follows a cycle of charging, reduction, distillation, melting and casting. Feed mixtures are prepared by combining either $UCl_3$ or $U_3O_8$ with molten anhydrous $MgCl_2$. This mixture enters the reducer/melter along with a magnesium-zinc alloy. Mg metal is then added which is supplied with Mg from the $MgCl_2$ electrolysis step. Two phases are formed; a molten salt phase which contains the uranium compound, $MgCl_2$ and Mg and a molten Mg-Zn phase at the bottom of the vessel. The ingredients are then heated to 800°C to maintain a molten state. Uranium formed from the reduction process enters the Zn-Mg alloy

phase and is only soluble to about 1% in this alloy phase. Any uranium in excess of this solubility limit precipitates as metallic uranium to form a third phase in the vessel. Other alloy compositions may be suitable. The alloy phase separates the molten salt from the solid uranium to protect from back reaction of the uranium metal.

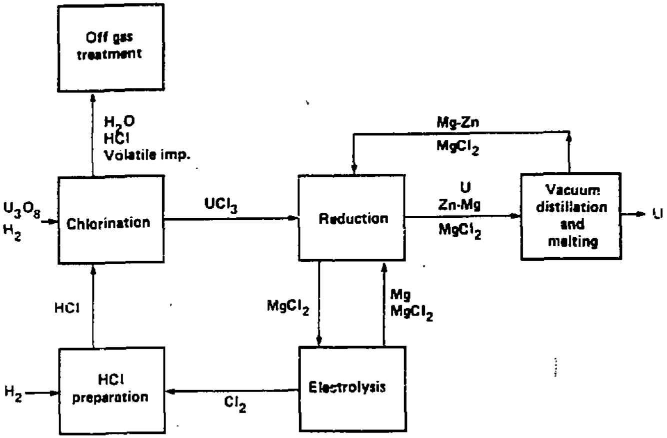


Figure 6

PRYOMETALLURGICAL Mg-Zn REDUCTION TO METAL

Once the reaction has reached completion, the $MgCl_2$ salt phase is transferred out via pressuring the vessel with an inert gas; e.g., argon. The Zn-Mg alloy and any residual $MgCl_2$ are vacuum distilled and condensed in the overhead condenser of the reducer-melter vessel. After vacuum distillation is complete, pure uranium solids remain in the reducer-melter which is then heated to 1200°C so that the uranium is molten and flows easily for casting.

While both processes are similar in nature, each produces some very unique problems. Chlorination of uranium oxide is not a straight forward task. Uranium tetrachloride has been produced in significant quantities by reacting the oxide with carbon tetrachloride with the resulting byproduct being a mixture of $CO_2$ and phosgene. Other organic reagents such as hexachloropropene may be used. Another possible method to chlorinate uranium oxide is by reaction of a mixture of carbon and the $UO_2$ with $Cl_2$. Preparation by crystallization from aqueous solution and dehydration is not straight forward because of the strong tendency of the hydrate to convert to oxychlorides during decomposition. If desired, the $UCl_4$ may be reduced to $UCl_3$ with $H_2$. Other ways have been demonstrated but all either use expensive reagents, are difficult to achieve or produce hazardous intermediate or final waste products. For this reason, the direct reduction of $U_3O_8$ with magnesium was investigated.

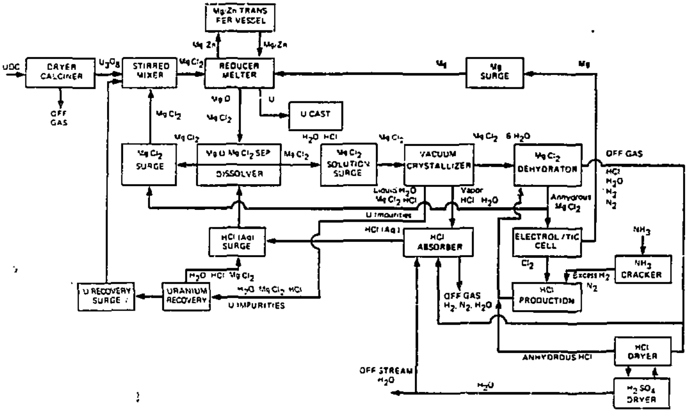


Figure 7

DIRECT REDUCTION OF $U_3O_8$ WITH MgO RECYCLE

For the direct reduction process, significant quantities of magnesium oxide are formed and remain with the $MgCl_2$ molten salt. It is desirable to separate the MgO from the $MgCl_2$, and then to convert the MgO to $MgCl_2$ for subsequent reduction by electrolysis to magnesium metal and chlorine gas. Separation of the MgO from the $MgCl_2$ presents the first obstacle. Once this is accomplished, industrial processes do exist[5,6] to convert and dehydrate the MgO to anhydrous $MgCl_2$, although most industrial processes require some additional reagents and the formation of intermediate compounds. Because of stoichiometric relationships, this reaction requires 1.75 times the magnesium to produce a unit of uranium metal than does the chlorination process. There are also several uncertainties regarding how well the MgO will flow in molten $MgCl_2$. Furthermore, if it proves impractical to separate MgO from the $MgCl_2$, the total $MgO/MgCl_2$ mixture will have to be cooled, dissolved, then later made anhydrous again.

Pyrometallurgical processes, either chlorination or direct reduction, are further complicated by the fact that process demonstrations have only been performed on bench scale equipment and the design of the reducer/melter is conceptual.

Other feed conversion alternatives. In the investigation for viable alternatives, other possible metal reduction methods were identified. These included carbothermic hydrogen reduction and hydrogen plasma reduction. These processes were initially screened out on the basis of technical feasibility. While carbothermic reduction may not be thermodynamically possible, plasma reduction appears to have some potential merit, though certainly a lengthy development effort would be required.

## PRODUCT CONVERSION BASELINE DESIGN

Enriched material is received from the separator in one of two forms--either as metal uranium or as a mixture of oxides and graphite from the refurbishment of separator components. The eventual material for pellet fabrication, ceramic grade $UO_2$, has some very unique characteristics and stringent performance requirements before it can be accepted. It is widely reported that the properties of the $UO_2$ powder are profoundly influenced by the powder's method of production, and the powder properties have a strong impact on its compressibility and sinterability to pellet form. Factors expected to affect sinterability are grain and particle size, shape and density, surface area, oxygen-uranium ratio, fabrication procedures, sintering atmospheres, and impurities. In the selection of product conversion alternatives for AVLIS, the method that is finally implemented must ultimately provide fully acceptable fuel pellets.

Fluorination to $UF_6$. The enriched metal and metal oxides can be converted to a nuclear grade $UF_6$ as shown on the product conversion process flow diagram (Figure 8). After conversion the enriched $UF_6$ would follow the same path as enriched product from diffusion and centrifuge plants. It would be transported to the various fuel fabricators for conversion to ceramic grade uranium oxide and ultimately manufactured into fuel pellets and placed into fuel rods. No difficulties are expected with the introduction of AVLIS-produced $UF_6$ into the fuel cycle.

The principal steps in the $UF_6$ conversion process are oxidation, fluorination, product collection, purification, and blending. The heart of the process is centered around the oxidation and fluorination steps. For the enriched metal stream, oxidation to $U_3O_8$ can be accomplished in an electrically heated vibrating tray reactor with wall temperature of 400°C. For the oxide stream, the oxidation step serves to burn out residual carbon left from the graphite components used in the separator vessel. This material must be heated to about 800°C in a rotary kiln to effect the carbon removal. Oxidized material must contain >95% oxide to meet process fluorination feed specifications.

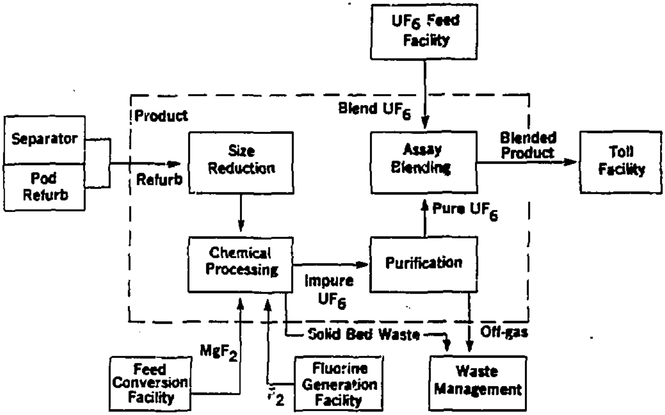


Figure 8
PRODUCT CONVERSION FLOW SHEET

The oxidized material is converted directly to $UF_6$ by reaction with fluorine in a fluidized bed. A magnesium fluoride diluent is used in the reactor to provide distribution of the reaction through the bed volume and help carry the heat of the very exothermic reaction to the walls for heat removal. Once the $U_3O_8$ feed is introduced into the reactor, the reaction temperature is brought to 450 to 550°C by appropriately adjusting the reactor gas flow, heaters and cooling air flow, and the feed rate of $U_3O_8$. The fluidized beds with a nominal bed depth of one meter (m) are designed to process 450 kg of uranium per day.

## PRODUCT CONVERSION ALTERNATIVES

Just as for feed conversion, product conversion was given a broader evaluation in the systems analysis study. If the constraint to supply $UF_6$ is removed, cost reduction can be achieved in the AVLIS plant, and another less expensive material suitable for conversion to ceramic grade oxide and fuel pellets may be used as the interface material between the enrichment and the fuel fabrication steps. For cost comparisons, it is not sufficient to compare only the production cost to $UF_6$; the cost required to convert the uranium hexafluoride gas to ceramic grade uranium oxide must also be included. Since $UF_6 \rightarrow UO_2$ conversion is done routinely throughout the fuel fabrication industry (through either dry or wet chemical processing methods) this paper will not delve into the different proven processes used.

Because the AVLIS product will contain some impurities, a purification step is first required prior to converting the material into a form suitable for further fuel fabrication processing. For some advanced processing alternatives, this purification step can be handled with a solvent extraction cycle of the dissolved metal. For other processes considered, pyrometallurgical processes (similar to those proposed for feed conversion), distillation and electrorefining are methods to purify the metal. For the purposes of this paper, the purification methods other than dissolution/solvent extraction will not be discussed in great detail. Instead, emphasis will be given to the subsequent processing steps being considered for providing ceramic grade $UO_2$ prior to conventional fabrication processing to fuel rods. The primary oxide preparation methods considered include 1) ammonium diuranate (ADU) precipitation followed by calcination and reduction, 2) direct denitration of uranyl nitrate (UNH) followed by reduction, and 3) steam oxidation of uranium metal to uranium dioxide. A variation of this latter method is to provide additional steps of oxidation and reduction to the uranium dioxide to improve the ceramic qualities of the final uranium dioxide material produced. Each of these methods will be presented below.

_Dissolution/solvent extraction with ADU precipitation and calcination._ This process utilizes process steps that are already used to some degree and in some manner on a commercial scale by the nuclear industry, although not in an integrated form for uranium metal. Figure 9 provides a general flowsheet of the process as envisioned for this application. The enriched uranium metal casting is dissolved in a nitric acid solution. Uranium is separated from the impurities by solvent extraction into a solvent composed of an organic; such as tributyl phosphate (TBP) dissolved in dodecane. Impurities remain in the aqueous or raffinate phase. The extraction raffinate, containing trace amounts of uranium in addition to the contaminants, is calcined to form a mixed metal oxide before disposal at a low level waste disposal site. The uranium loaded solvent is fed to a stripping column where the purified uranium is removed from the organic solvent with dilute nitric acid. The resulting uranyl nitrate solution is concentrated by evaporation and blended to the correct uranium assay by the addition of normal assay uranyl nitrate. Ammonia is then added to precipitate ammonium diuranate (ADU), and the ADU precipitate is removed by centrifugation. Uranium dioxide is formed by calcination and reduction of the ADU material. Gaseous effluents from all process steps are scrubbed and filtered prior to release.

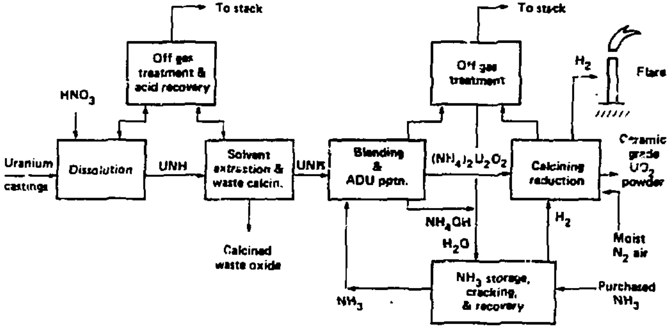


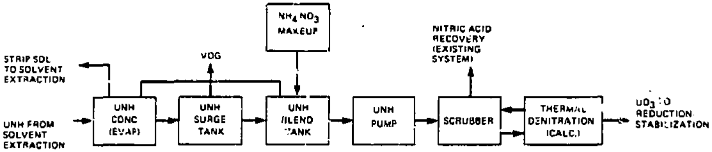
Figure 9

SOLVENT EXTRACTION/ADU—PRODUCT PURIFICATION OVERVIEW

Although the major process steps in the solvent extraction/ADU precipitation process for AVLIS product conversion are common in the nuclear industry, process demonstration efforts will be required to adapt these processes to the AVLIS uranium product. Due to criticality/throughput concerns, a slab type dissolver may be best. The dissolver design for the uranium oxide separator refurbishment stream may be different than the dissolver for the uranium metal stream because of expected differences in dissolution rates. The extraction system may employ pulse columns or centrifugal contactors. Once the material is purified, the ADU precipitation/calcination equipment is based on that typically used in the nuclear industry for wet processing and scrap recovery lines. Some development work will be required in tuning specific extraction/precipitation designs and in waste treatment areas.

One potential disadvantage of this process is shared with all of the advanced options investigated. Fuel pellets produced with starting materials other than $UF_6$ will require extensive characterization and testing to assure pellet quality and performance. ADU precipitation/calcination also has some disadvantages in terms of controlling precipitation to give the correct morphology, yet yielding a slurry which may be readily filtered or centrifuged. Problems with processing substantial ammonium nitrate wastes are also present.

Direct denitration of UNH. Direct denitration of the uranyl nitrate solution to $UO_3$ and subsequent hydrogen reduction to $UO_2$ is an alternative that has received much attention primarily in mixed oxide fuels (MOX) studies and also appears to have distinct advantages for AVLIS uranium product. Typically, thermal denitration of UNH has the disadvantages of "waste" or "dough" stages and of giving a low surface area product with poor sinterability characteristics. Two modified denitration processes, one developed by the Oak Ridge National Laboratory and one developed by Comurhex of France, eliminate these disadvantages and appear to be attractive candidates in this alternative route. These processes both avoid the molten stage in dehydration-denitration which result in low surface area, non-ceramic grade powder



THERMAL DENITRATION

Figure 10

SCHEMATIC FLOWSHEET FOR DIRECT DENITRATION URANIUM OXIDE PRODUCTION

In the ORNL process (figure 10), powders possessing precipitation-like characteristics, can be produced by the simple and inexpensive addition of ammonium nitrate to the heavy metal solution followed by the thermal decomposition in the temperature range of about 300 to 800°C. The advantage of this process is the derivation of good quality ceramic powders without actual precipitation or the handling of the associated waste from a precipitation process. Powders produced have excellent physical characteristics with B.E.T. surface area of about 5-15 $m^2/g$, a predominant particle size of less than 10 microns, and result in a uniform pellet having a favorable density potential after sintering at 1450-1700°C. Typically, ammonium nitrate additions of 1.0 to 2.0 $NH_4^+$/metal molar compositions are sufficient to produce the good powder characteristics[7].

At a recent conference in Vienna, Austria[8], the French company Comurhex (a subsidiary of Pechiney) reported successful development of a process to obtain sinterable oxides by direct denitration. This process, called NITROX, was developed for MOX fuels but there is also direct application to denitration of uranyl nitrate solutions. Comurhex reports nitrate solutions are concentrated to about 1200 g/l and then crystallized by cooling. The nitrate powder is dehydrated and then denitrated at reduced pressure. The oxide is finally calcined and reduced to $UO_2$. The main step in the process is denitration and dehydration which is conducted using a pressure and temperature combination that obviates melting of the nitrates. Dehydration and denitration is carried out in discrete steps holding the material at 20, 260, and 400°C for periods of about 1.5 hours each. Calcination and reduction of the $UO_3$ is accomplished at about 600°C and requires about 1 hour for each step. $UO_2$ with B.E.T. surface area of 6.2 $m^2/g$ and density of 1.7g/cc is typically produced. Sintered oxide density up to 96% of theoretical is commonly achieved.

Steam oxidation. Direct oxidation of pure uranium metal to uranium dioxide was examined as a possible alternative for product processing primarily because it also provides a dry method for producing $UO_2$. Many fuel fabricators are turning away from wet chemical methods in favor of dry processes which produce less wastes. Process steps for steam oxidation are presented in figure 11.

The equation:

$$U + 2H_2O \Rightarrow UO_2 + 2H_2$$

is commonly reported as the resultant equation of reaction, although the reaction mechanisms are much more complicated.[9] Many experiments have been performed to gather kinetic data and identify the form of the oxide produced. Wilson, et al.[10] reported the resulting form was dramatically affected by the temperature at which the oxidation was performed. At 400°C, the $UO_2$ powdered continuously and was nearly colloidal in character. At about 600°C, the $UO_2$ formed a hard layer on the uranium with no tendency to flake off. As the sample cooled below 300°C, the dioxide layer popped off and basically retained the hard, smooth shape of the original metal.

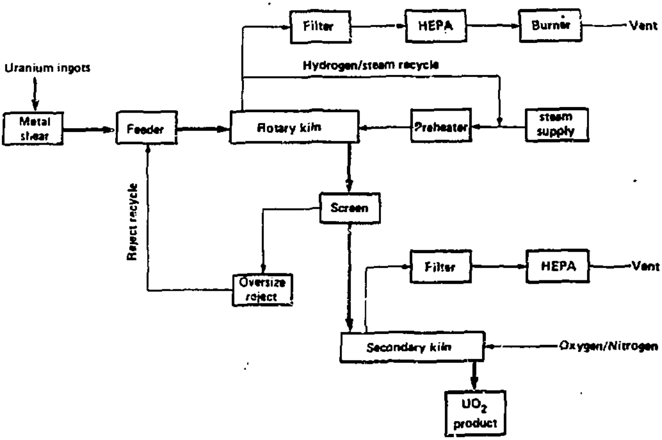


Figure 11
URANIUM STEAM OXIDATION TO $UO_2$

Specifically, for the design envisioned for AVLIS uranium processing, uranium ingots produced from the purification process, e.g., pyroredox, distillation, electrorefining, etc., must first be reduced in size in an inert atmosphere shear to enhance the reactivity of the uranium metal before being fed to the oxidizer units through some type mechanical feeder. Two reactors; e.g., rotary kilns or vibrating trays, in series then can serve the purpose of oxidizing the metal to the required oxide form. Critically safe geometry or the use of fixed poisons are included in the reactor design. The sheared uranium segments are fed in parallel with superheated steam in an approximate 145 percent stoichiometric quantity. As the pieces of uranium and superheated steam pass through the reactor, the steam reacts with the uranium producing hydrogen and uranium dioxide which flakes off as a powder. One third of the exiting steam/hydrogen is recycled and combined with the incoming superheated steam in order to keep the hydrogen concentration in the steam at a level that optimizes the oxidation of the uranium.

The product (uranium/$UO_2$) exiting the primary oxidizer is passed through a screen, and the oversized material is collected and recycled as feed material to the primary oxidizer. The product passing through the vibrating screen is fed to a secondary oxidizer in parallel with a stream of nitrogen with a small amount of heated air. The powdered $UO_2$/uranium and nitrogen/air mix pass through the secondary oxidizer to adjust the O/U ratio.

Major uncertainties of the process are the current state of development and the ceramic quality of oxide that will be produced. While, steam oxidation of uranium metal may not produce a powder with acceptable physical characteristics, it is conceivable to expect that subsequent oxidation to $U_3O_8$ followed by hydrogen reduction to $UO_2$ will improve the morphology of the dioxide powder produced. The number of oxidation/reduction cycles which would be required to produce acceptable fuel pellets is unknown at this time. Cost evaluations for the impact of each additional oxidation/reduction cycle that would be required are underway. Steam oxidation certainly needs considerable experimental development and demonstration testing if it is to become a viable alternative to wet processing technology.

## ECONOMIC SUMMARY

Uranium processing for a $UF_6$ in--$UF_6$ out AVLIS scenario amounts to about $15/separative work unit, or almost one-third the total enrichment SWU cost for an AVLIS enrichment plant, according to studies completed in DOE's advanced isotope process selection. When added to the cost to produce the $UF_6$ from uranium ore concentrate and to convert the enriched $UF_6$ to uranium dioxide, the total uranium processing cost for AVLIS is approximately $140/kg U product or equivalent to about $30/SWU. For a 14 million SWU per year plant, this would amount to more than $400 million per year in cost to the nuclear fuel cycle customers. The systems analysis study has identified alternatives which provide the option to substantially reduce the cost of uranium processing for AVLIS. These alternatives are ranked according to estimated production costs in Table II.

Detailed capital and operating cost estimates were prepared for the thermite reduction and product fluorination to $UF_6$ processes, since they were the baseline design for AVLIS during the advanced isotope selection process. These cost estimates were developed with support from Martin Marietta Energy Systems, LLNL, Stone & Webster Engineering Corporation, Westinghouse Electric Corporation, and Bechtel National, Inc. and were based on a government operated, 12 million SWU/year production plant, located at the existing Oak Ridge Gaseous Plant site. Cost estimates indicate a unit cost of approximately $8-9/kg U feed to reduce natural $UF_6$ to uranium metal, excluding the cost of the uranium hexafluoride feed material. Elimination of the $UF_4 \rightarrow UF_6 \rightarrow UF_4$ process steps can result in approximately \$2.50/kg savings to the feed conversion process (avoiding the $UF_4 \rightarrow UF_6$ step) and \$1/kg savings to the thermite reduction process (avoiding the $UF_6 \rightarrow UF_4$ reduction step) for an overall fuel cycle savings on the order of \$3.50/kg feed.

Table II

ECONOMIC RANKING OF VARIOUS URANIUM PROCESSING ALTERNATIVES

| | |
|---|---|
| FEED CONVERSION (PROJECTED $U_3O_8 \rightarrow$ U-metal) | |
| • ELECTROLYTIC REDUCTION | INCREASING COSTS ↓ |
| • PYROMETALLURGICAL PROCESS | |
| • THERMITE BOMB REDUCTION ($UF_4$) | |
| • THERMITE BOMB REDUCTION ($UF_6$) | |
| PRODUCT FABRICATION (U-metal $\rightarrow UO_2$) | |
| • DISSOLUTION/SOLVENT EXTRACTION--ADU PRECIPITATION | INCREASING COSTS ↓ |
| • DISTILLATION/STEAM OXIDATION (1 CYCLE) | |
| • DISTILLATION/STEAM OXIDATION (MULTIPLE CYCLES) | (EXPECTED) |
| • PYROMETALLURGICAL/STEAM OXIDATION | |
| • FLUORINATION TO $UF_6$, ADU OR DRY PROCESSING TO $UO_2$ | |
| ALSO UNDER STUDY | |
| • DISSOLUTION/SOLVENT EXTRACTION--DIRECT DENITRATION | |

Cost estimates were performed on the oxidation-fluorination product processing in the same manner as for the thermite bomb reduction process for feed conversion. These cost studies indicated the cost to produce pure $UF_6$ from the enriched AVLIS metal is on the order of \$21/kg of uranium (U) product. Since about 6 kg of feed are required to produce one kilogram of product, the cost of product conversion on a feed basis is approximately \$3.50/kg.

Capital and annual operating costs for the alternative designs were scoped in a manner patterned after the baselined design cost estimates but not to the same level of detail. The alternative designs for feed, electrolytic reduction and pyrometallurgy both appear to offer significant cost reduction opportunities for supplying AVLIS with metal feed. While there were not significant cost differences between those two technologies, the pyrometallurgical process has not reached the state of developmental maturity characteristic of electrolytic reduction, and would be expected to require a more costly and time consuming RD&D program to permit a production commitment.

Metal dissolution/solvent extraction with ADU precipitation provides several advantages when compared to the other possible product technologies. There is a large economic cost advantage to this process. The oxide product should not require further processing to allow use as a ceramic oxide. Furthermore, since each of the unit processes is well-established operationally, it is expected that RD&D costs will be lower than for any of the other possible AVLIS product conversion alternatives. In addition, this approach avoids the hazards and fluoride waste disposal problems associated with the $UF_6$ based process and allows highly precise liquid phase isotopic blending. Finally, similar solvent extraction/ADU precipitation processes have demonstrated high levels of reliability, availability, and maintainability during long-term operations. Cost estimates indicate the direct steam oxidation process, when coupled with distillation for metal product material purification, can produce $UO_2$ on the order of the estimate for dissolution/solvent extraction with ADU precipitation, though it lacks the development maturity of the ADU process.

At the time of this writing, economic evaluation of the ORNL direct denitration process has yet to be completed. As can be seen in figure 10, however, a comparison to ADU precipitation/calcination indicates that the direct denitration process eliminates approximately half the required unit operations. On the other hand, pellet characterization and testing has not been performed to fully qualify the oxide, although some sinterability testing at Hanford indicated very positive results. If the expected cost savings are achieved, direct denitration would be a viable method worth more serious consideration.

Economic analysis requires many assumptions such as those relating to plant size, price/cost relationships, return on investment, capital recovery factors, siting, waste disposal cost, market conditions, age of facilities and plant capacity. We have placed all estimates on an equal basis and have assumed enrichment enterprise/government contractor operations. Even accounting for expected cost estimate uncertainties, it is believed with a high level of confidence that for a metal-based fuel cycle, moving away from a $UF_6$ intermediate product offers substantial savings and gives electric utility customers the opportunity to realize the full savings AVLIS enrichment can offer. It is hoped that industry will arrive at the same conclusion and actively seek these lower cost alternatives.

## CONCLUSIONS

AVLIS RD&D efforts are currently proceeding toward full-scale integrated enrichment demonstrations in the late 1980's and potential plant deployment in the mid 1990's. Since AVLIS requires a uranium metal feed and produces an enriched uranium metal product, some change in current uranium processing practices are necessitated. AVLIS could operate with a $UF_6$-in $UF_6$-out interface with little effect to the remainder of the fuel cycle. This path, however, does not allow electric utility customers to realize the full potential for low cost AVLIS enrichment. Several alternative processing methods have been identified and evaluated which appear to provide opportunities to make substantial cost savings. These alternatives involve varying levels of RD&D resources, calendar time, and technical risk to implement and provide these cost reduction opportunities. Both feed conversion contracts and fuel fabricator contracts are long term entities. Because of these factors, it is not too early to start planning and making decisions on the most advantageous options so that AVLIS can integrate cost effectively into the fuel cycle. This should offer economic opportunity to all parties involved including DOE, utilities, feed converters, and fuel fabricators.

## ACKNOWLEDGEMENTS

The authors wish to express their appreciation to several people without whose help the systems analysis study would not have been so complete and to special people who had a significant part in pulling this paper together. Major contributions were made by Ted Breitmeyer, Lee Thomas, Rich Tosetti, Don Crandall, and Les Jardine (Bechtel National); Alfred Schneider (Georgia Tech); John Payne (Kaiser Aluminum and Chemical Corporation); Larry Huston and Marty Mesner (Kaiser Engineers, Inc.); William Miller (ANL); and Paul Haas (ORNL). Thanks also go to Deborah Andrews, Earl Munday and Jeff Patton (Martin Marietta Energy Systems) for providing information and assisting in writing and preparing the paper. Many individuals throughout several organizations provided critical technical and economic review during the process and are given much thanks.

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DWG. NO. K/G-86-2094 R1

# INTEGRATION OF AVLIS INTO THE FUEL CYCLE

| | |
|---|---|
| R. S. EBY | MARTIN MARIETTA ENERGY SYSTEMS |
| J. H. PASHLEY | MARTIN MARIETTA ENERGY SYSTEMS |
| R. E. NORMAN | MARTIN MARIETTA ENERGY SYSTEMS |
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| J. B. KNIGHTON | LAWRENCE LIVERMORE NATIONAL LABORATORY |

AICHE SUMMER NATIONAL MEETING

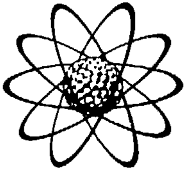
August 27, 1986

MARTIN MARIETTA

# THE AVLIS PHOTOIONIZATION PROCESS

DWG. NO. K/G-85-5293

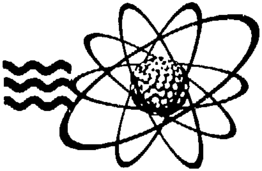
1



## ATOMIC STRUCTURE

The electron energy states of the uranium atom are very precisely defined and depend on the mass of the nucleus. These energies give rise to light absorption characteristics that are unique to each isotope.

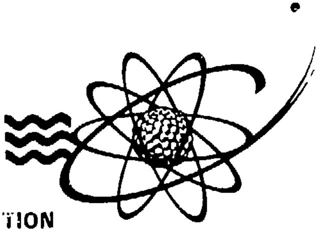
2



## LASER LIGHT

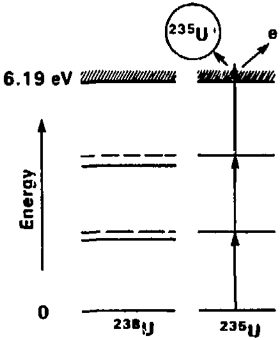
As an isotope absorbs light precisely color-tuned to its discrete energies, its electrons are excited to higher states.

3



## SELECTION

With sufficient energy, the electron can leave the atom. This selected isotope is now tagged as a positive ion ready for separation. Unselected isotopes remain neutral.



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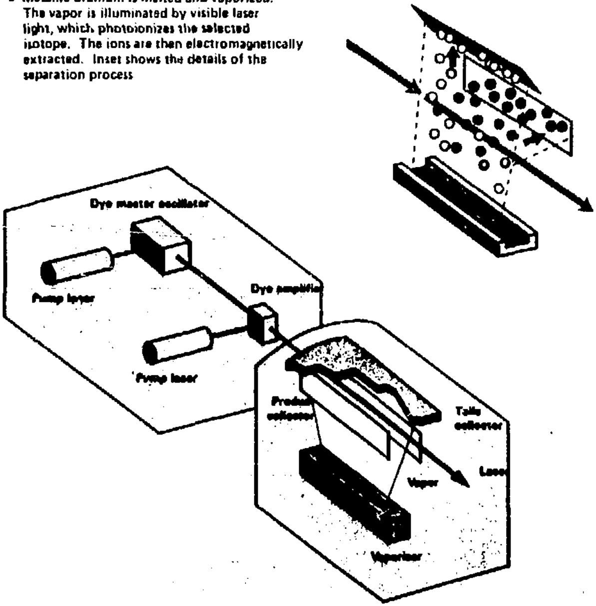
# ATOMIC VAPOR LASER ISOTOPE SEPARATION (AVLIS ) PROCESS

- Metallic uranium is melted and vaporized. The vapor is illuminated by visible laser light, which photoionizes the selected isotope. The ions are then electromagnetically extracted. Inset shows the details of the separation process



DWG. NO. K/G-86-65

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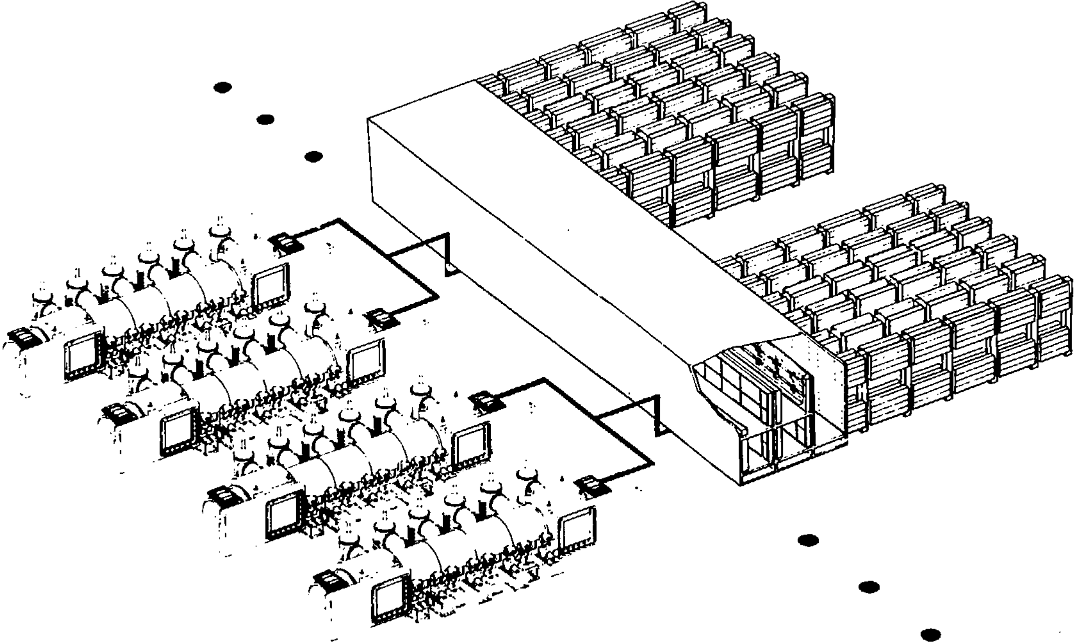
DWG. NO. K/G-85-5298

# BASIC ARCHITECTURE OF AN AVLIS PRODUCTION FACILITY



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# AVLIS PRODUCTION PLANT

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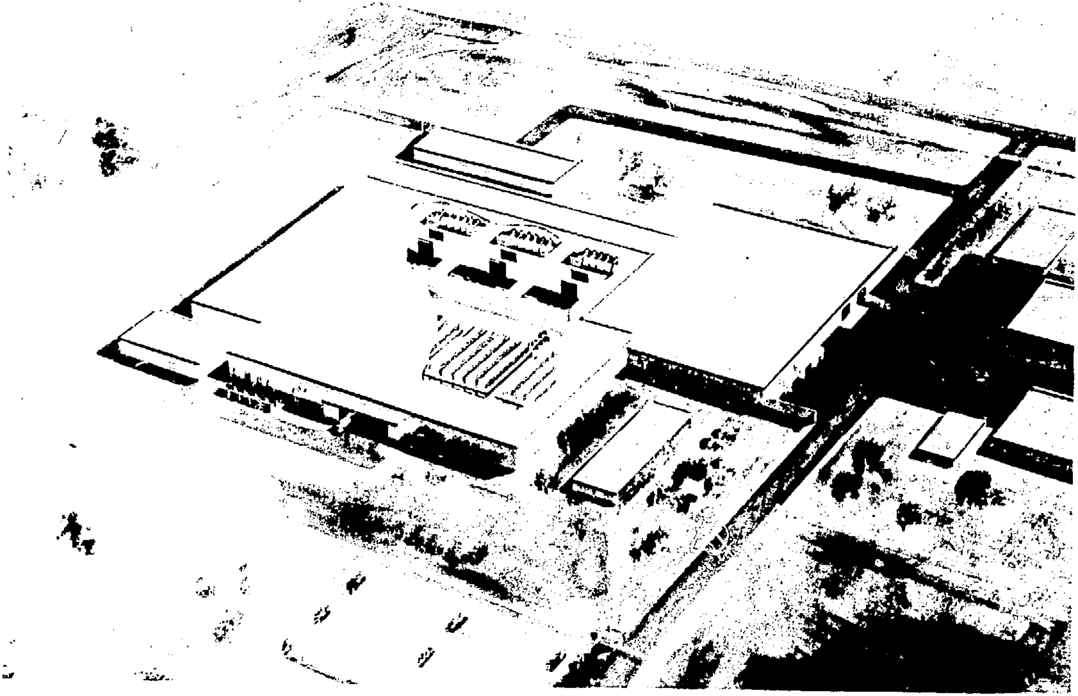

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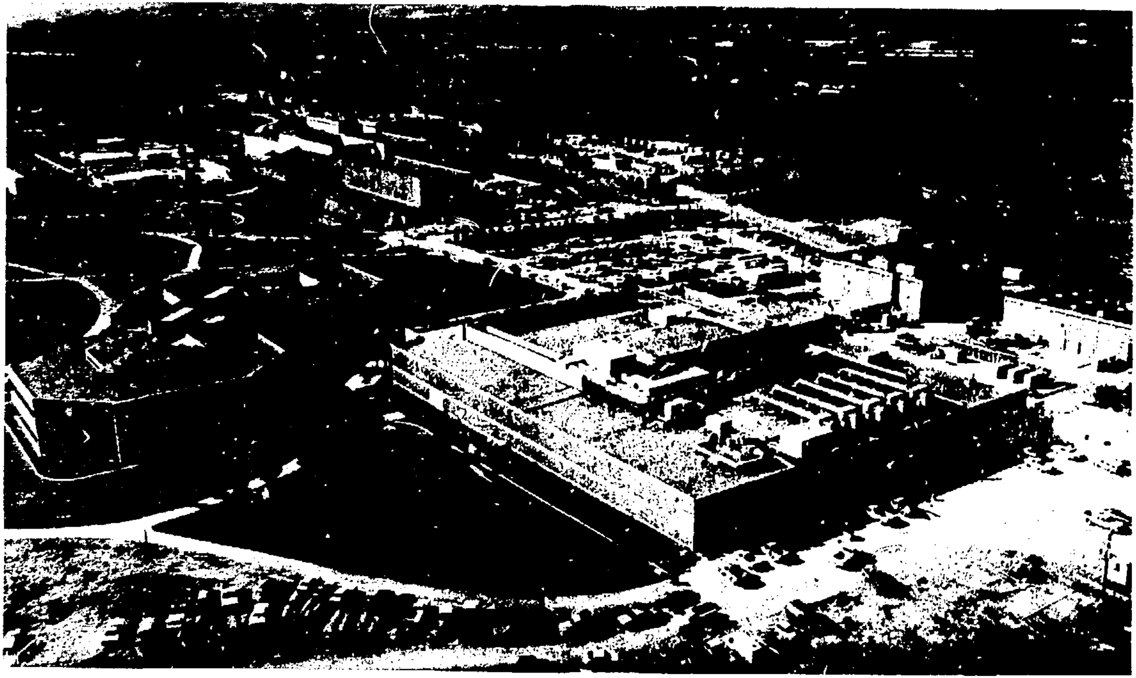
# AERIAL VIEW OF FACILITIES AT LIVERMORE



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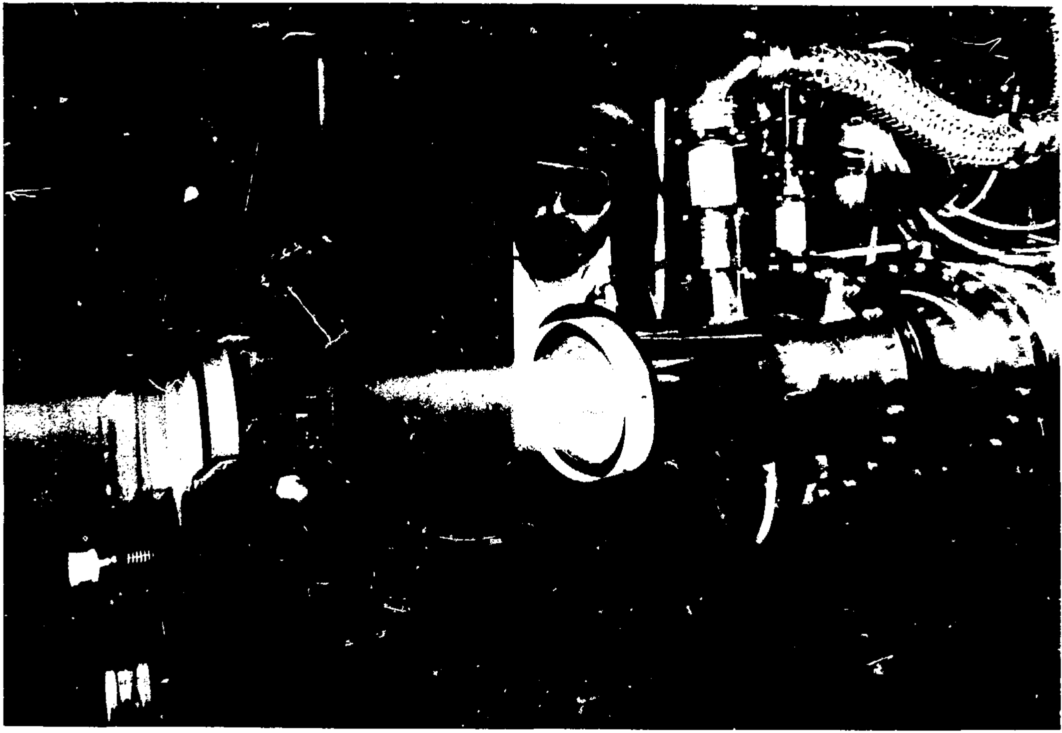
# COPPER VAPOR LASER



DWG. NO. K/G-86-89

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# COPPER VAPOR LASER CORRIDOR

DWG. NO. K/G-88-57

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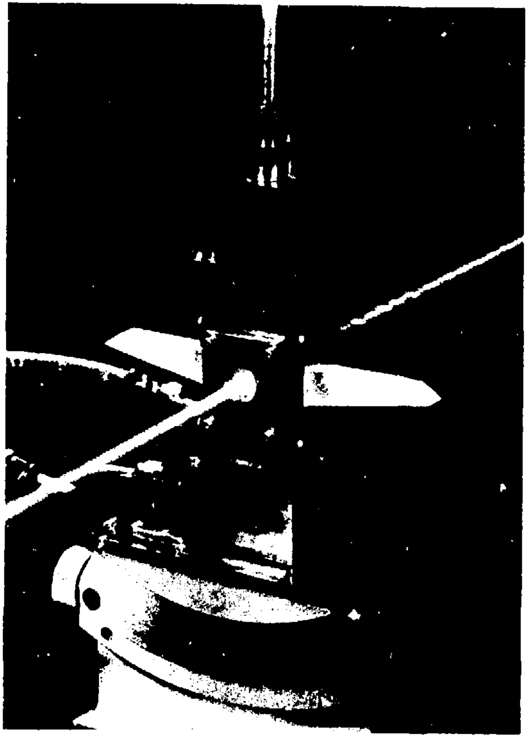
# DYE AMPLIFIER



DWG. NO. K/G-86-2060

MARTIN MARIETTA

DWG. NO. K/G-86-55

# DYE LASER CORRIDOR

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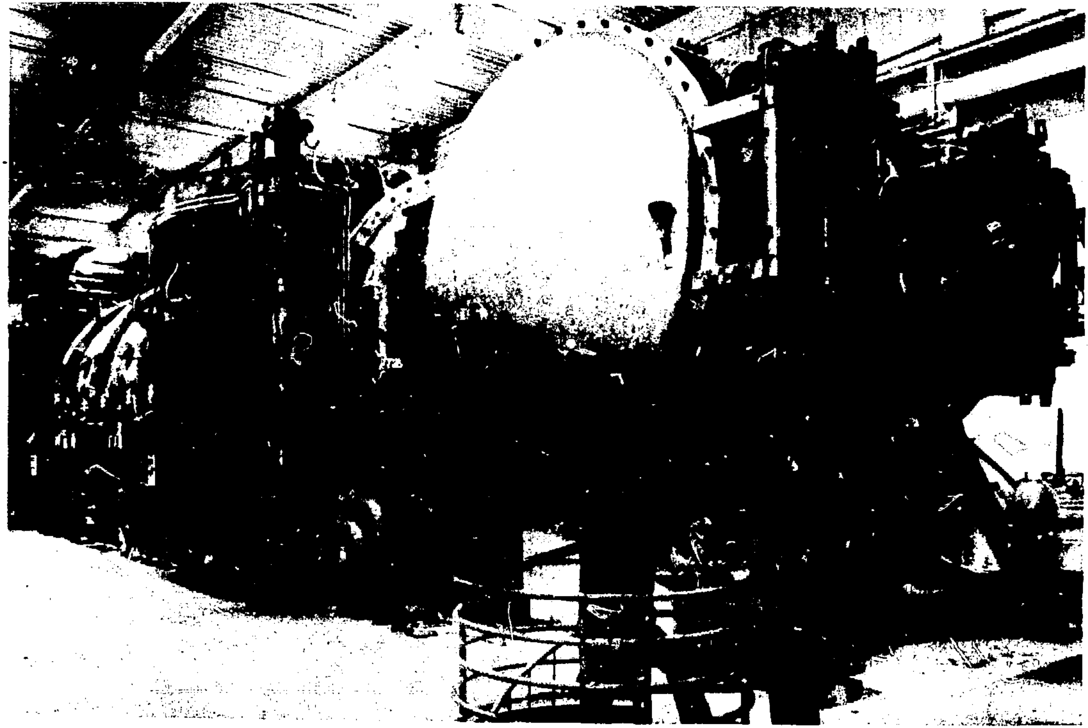
# SEPARATOR DEMONSTRATION FACILITY AT LLNL



DWG. NO. K/G-88-60 R1

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DWG. NO. K/G-86-1906-R1

# AVLIS URANIUM PROCESSING OVERVIEW

- From the viewpoint of process selection
- Today's view

# CRITERIA FOR URANIUM PROCESSING ESTABLISHED FOR PROCESS SELECTION

- $UF_6$ in — AVLIS — $UF_6$ out
  - Preserve existing industrial base and set of interfaces
- Conservative design approach; use of existing technologies
- All uranium processing facilities required for production contained within boundary of AVLIS production plant

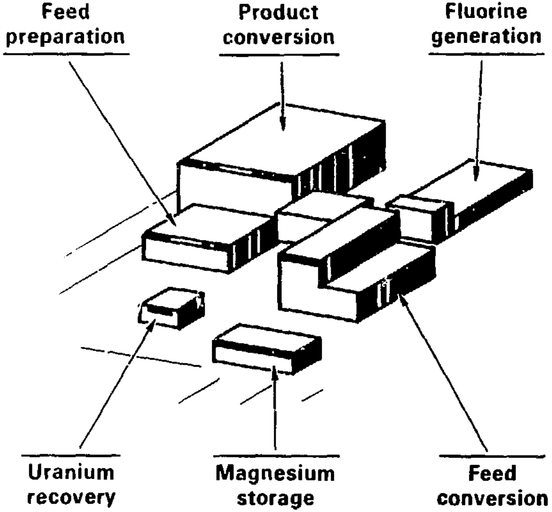
# THE "BASELINE" AVLIS URANIUM PROCESSING FACILITIES PROVIDED FOR A $UF_6$ IN — $UF_6$ OUT OPERATIONAL MODE

THESE FACILITIES INCLUDED:

- FEED CONVERSION – CONVERTS CUSTOMER $UF_6$ TO U METAL DERBIES
  - – $H_2$ REDUCTION OF $UF_6$ TO $UF_4$
  - – Mg REDUCTION OF $UF_4$ TO U
  - – BOTH STEPS BASED ON PADUCAH DESIGN
- FEED PREPARATION – PREPARES SEPARATOR FEED FROM U DERBIES
- PRODUCT CONVERSION – CONVERTS ENRICHED METAL PRODUCT TO $UF_6$
  - – OXIDIZE AND FLUORINATE TO $UF_6$
    - REQUIRES FLUORINE GENERATION PLANT
  - – BLEND TO REQUIRED ASSAY
- URANIUM RECOVERY – CONVERTS NATURAL U DRY SCRAP TO $UF_6$ FOR PROCESS RECYCLE

# "BASELINE" AVLIS U-PROCESSING FACILITIES ASSUMED FOR PEER REVIEW EVALUATION

## "Baseline" facilities convert $UF_6$ feed to metal and enriched metal product to $UF_6$



## Post-selection status of AVLIS U-processing

- PEB evaluation of "baseline" technology:

  "The technology to perform all material processing is widely available"

- PEB evaluation of cost reduction options:

  "The AVLIS plant (assumed for PEB evaluation) includes processing steps not required if feed and product were in forms different than $UF_6$.

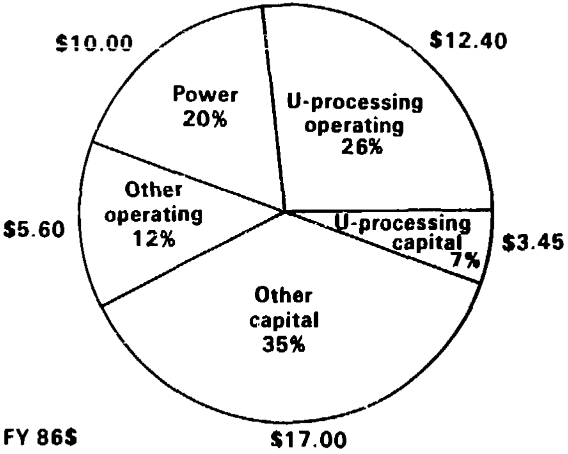
  Several alternatives could reduce the total cost of nuclear fuel."

MARTIN MARIETTA

DWG. NO. K/G-86-1910

# PROCESS EVALUATION BOARD ENRICHMENT COST FOR AVLIS

**PEB projection for total enrichment cost is $48/SWU**



- **Enrichment cost ($/SWU)**

| | |
|---|---|
| — Capital | 17.00 |
| — Operating | 15.60 |
| — Total | 32.60 |

- **Uranium processing cost ($/SWU)**

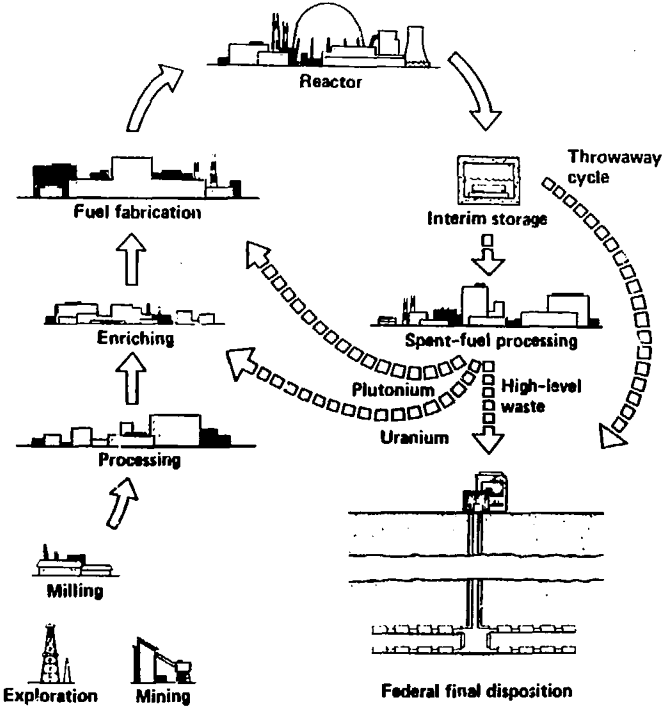
| | |
|---|---|
| — Capital | 3.45 |
| — Operating | 12.40 |
| — Total | 15.85 |

DWG. NO. K/G-86-1908

MARTIN MARIETTA

# TODAY'S CRITERIA FOR URANIUM PROCESSING FOR AVLIS

- To select the lowest cost processing path constrained only by what is possible and sensible
  - Technically
  - Industrial interface
  - Meets plant deployment schedule

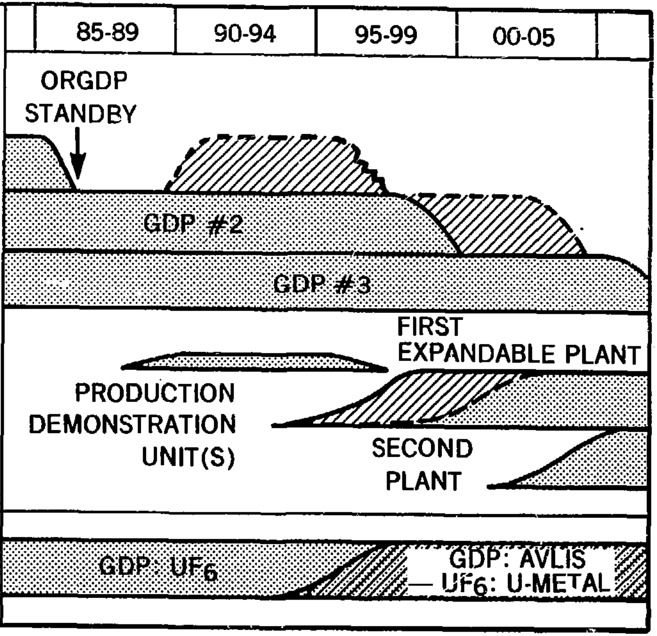


DWG. NO. K/G-86-1911

MARTIN MARIETTA

# EVOLUTION OF ENRICHMENT TECHNOLOGY INTERFACE TO THE FUEL CYCLE

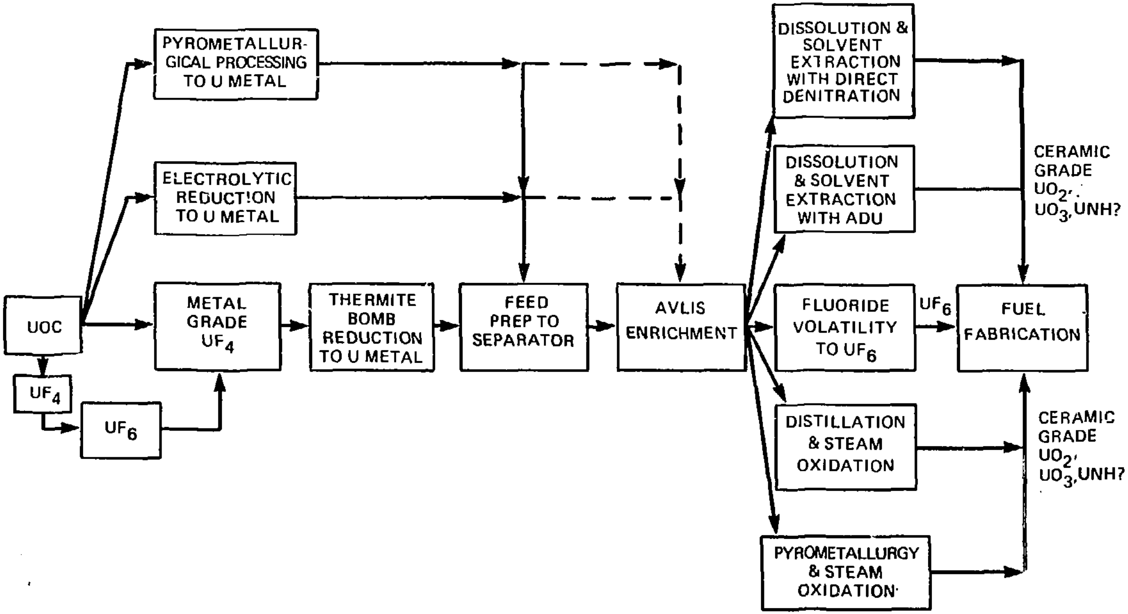
- ENRICHMENT TECHNOLOGY
  - — GASEOUS DIFFUSION PRODUCTION
  - — AVLIS PRODUCTION
- ENRICHMENT INTERFACE TO THE FUEL CYCLE



DWG. NO. K/G-85-5289

MARTIN MARIETTA

# AVLIS URANIUM PROCESSING ALTERNATIVES



UOC

$UF_4$

$UF_6$

PYROMETALLURGICAL PROCESSING TO U METAL

ELECTROLYTIC REDUCTION TO U METAL

METAL GRADE $UF_4$

THERMITE BOMB REDUCTION TO U METAL

FEED PREP TO SEPARATOR

AVLIS ENRICHMENT

DISSOLUTION & SOLVENT EXTRACTION WITH DIRECT DENITRATION

DISSOLUTION & SOLVENT EXTRACTION WITH ADU

FLUORIDE VOLATILITY TO $UF_6$

DISTILLATION & STEAM OXIDATION

PYROMETALLURGY & STEAM OXIDATION

$UF_6$

FUEL FABRICATION

CERAMIC GRADE $UO_2$, $UO_3$, UNH?

CERAMIC GRADE $UO_2$, $UO_3$, UNH?

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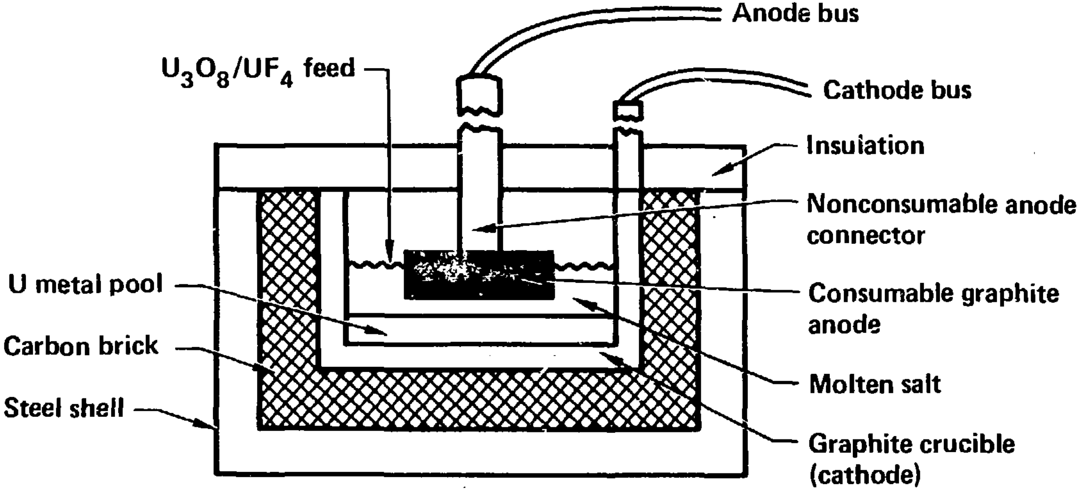
DWG. NO. K/G-85-893-R1

# SCHEMATIC OF ELECTROLYTIC CELL FOR PREPARATION OF U-METAL FROM $U_3O_8$

DWG. NO. K/G-86-1109



| | |
|---|---|
| $UO_2 + 2C \rightarrow 2\,CO + U^{+4} + 4e^-$ | At anode |
| $U^{+4} + 4e^- \rightarrow U^0$ | At cathode |
| $UO_2 + 2C \rightarrow 2\,CO + U^0$ | Overall |

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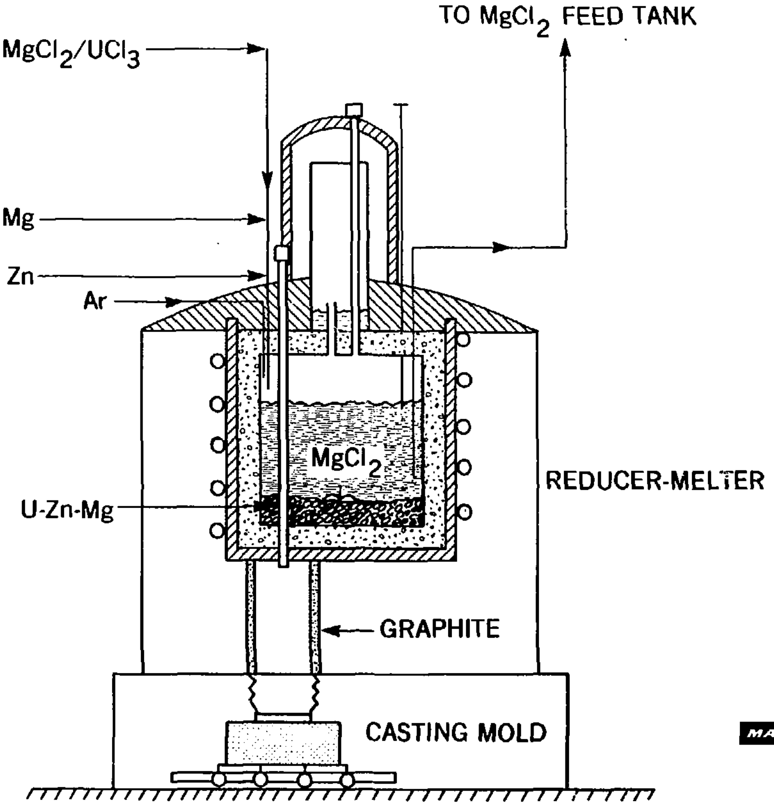
# PYROMETALLURGICAL PROCESSES — REDUCER-MELTING VESSEL



DWG. NO. K/G-86-2084

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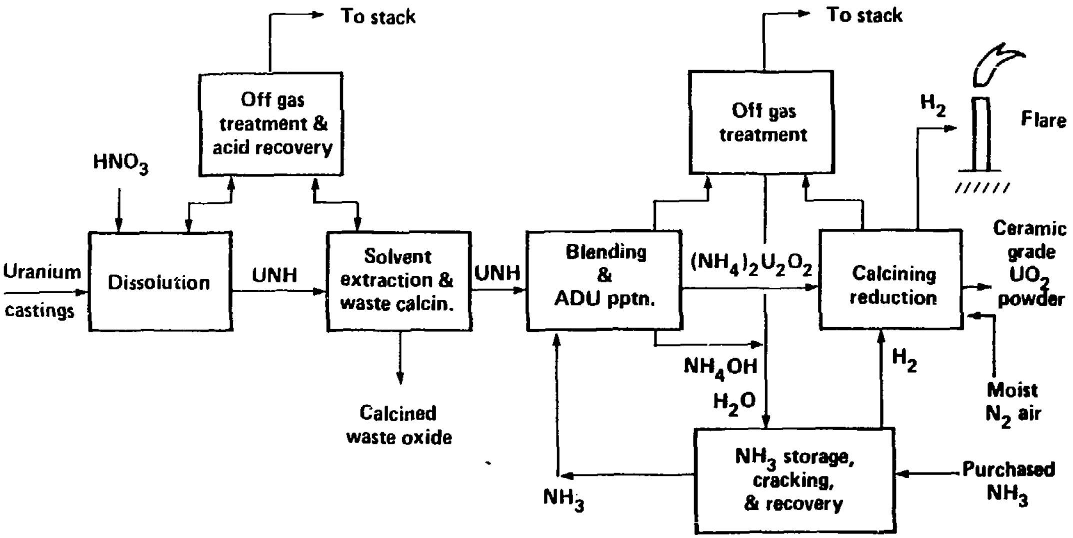
# PRIMARY CANDIDATES FOR AVLIS FEED CONVERSION

- ELECTROLYTIC REDUCTION IS THE PRIMARY TECHNOLOGY AND OFFERS:
  - LOW PRODUCTION COSTS
  - LOW COST SENSITIVITY TO WASTE DISPOSAL COST ESCALATION
  - HIGH LEVELS OF DEMONSTRATED URANIUM PRODUCTION
  - STRONG COUPLING TO MATURE ALUMINUM TECHNOLOGY
- PYROMETALLURGICAL REDUCTION IS A POTENTIAL BACKUP TECHNOLOGY WITH:
  - LOW PRODUCTION COST
  - HIGH COST SENSITIVITY TO WASTE DISPOSAL COSTS FOR $UCl_3$ CONVERSION BUT LOWER SENSITIVITY IS EXPECTED FOR $U_3O_8$ CONVERSION FLOWSHEET
  - LOW LEVELS OF DEMONSTRATED URANIUM PRODUCTION
  - COMMONALITY WITH THE KROLL PROCESS FOR Ti/Zr PRODUCTION

# SOLVENT EXTRACTION/ADU— PRODUCT PURIFICATION OVERVIEW



To stack

Off gas treatment & acid recovery

HNO$_3$

Uranium castings

Dissolution

UNH

Solvent extraction & waste calcin.

Calcined waste oxide

UNH

Blending & ADU pptn.

To stack

Off gas treatment

$(NH_4)_2U_2O_2$

NH$_4$OH

H$_2$O

NH$_3$

NH$_3$ storage, cracking, & recovery

Purchased NH$_3$

Calcining reduction

H$_2$

H$_2$

Flare

Ceramic grade UO$_2$ powder

Moist N$_2$ air

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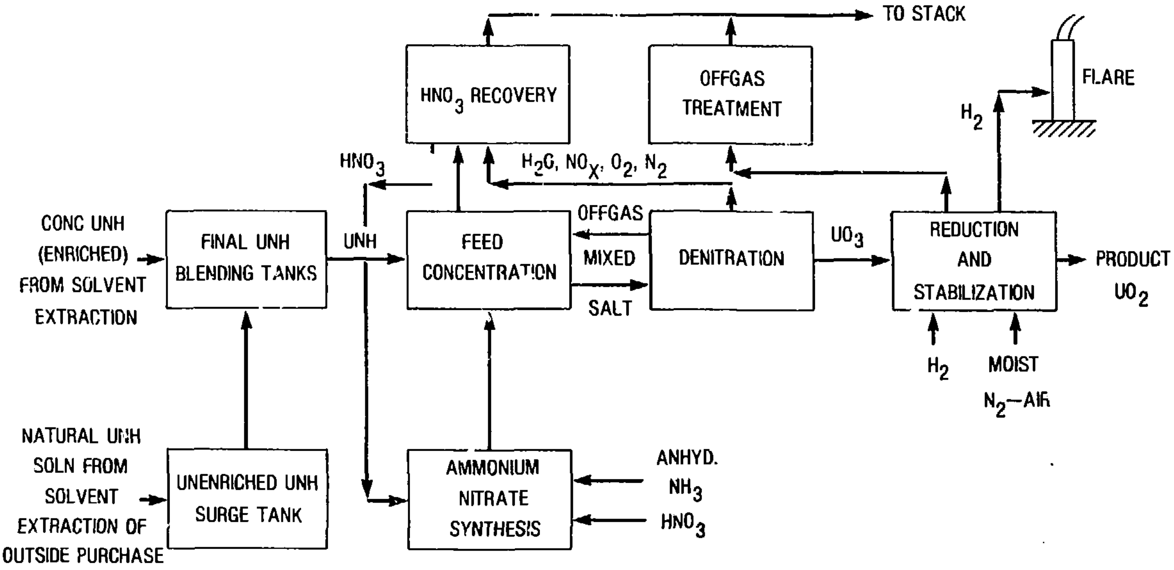
DWG. NO. K/G-86-2110

# BLOCK FLOW DIAGRAM FOR MODIFIED DIRECT DENITRATION PROCESS



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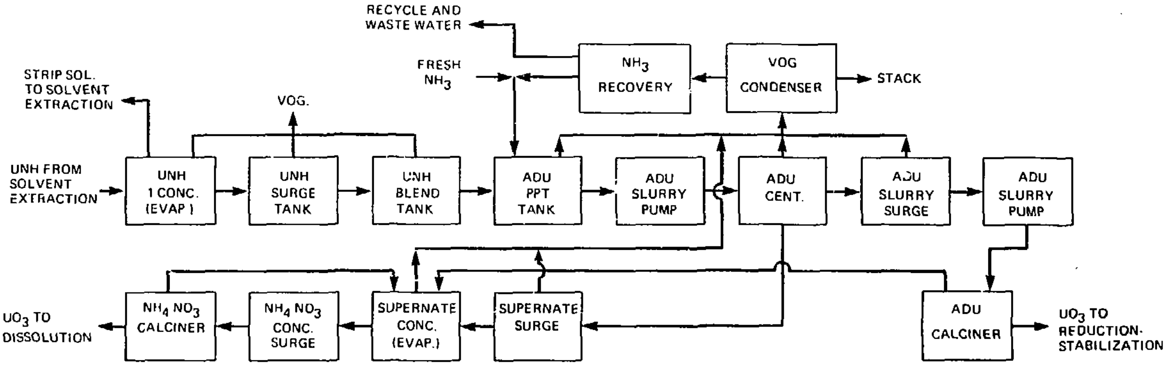
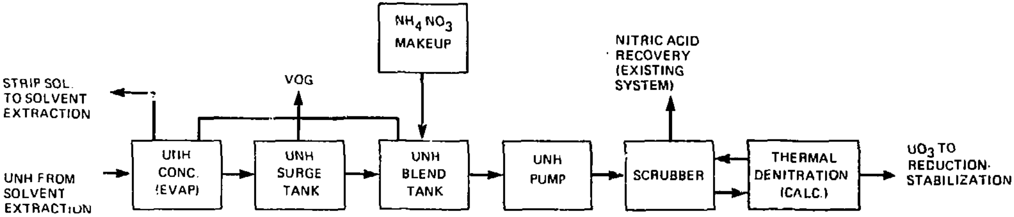
# SCHEMATIC FLOWSHEETS FOR AVLIS PRODUCT CONVERSION (UNH TO $UO_3$)

DWG. NO. K/G-88-2063



A. ADU PRECIPITATION — CALCINATION



B. THERMAL DENITRATION

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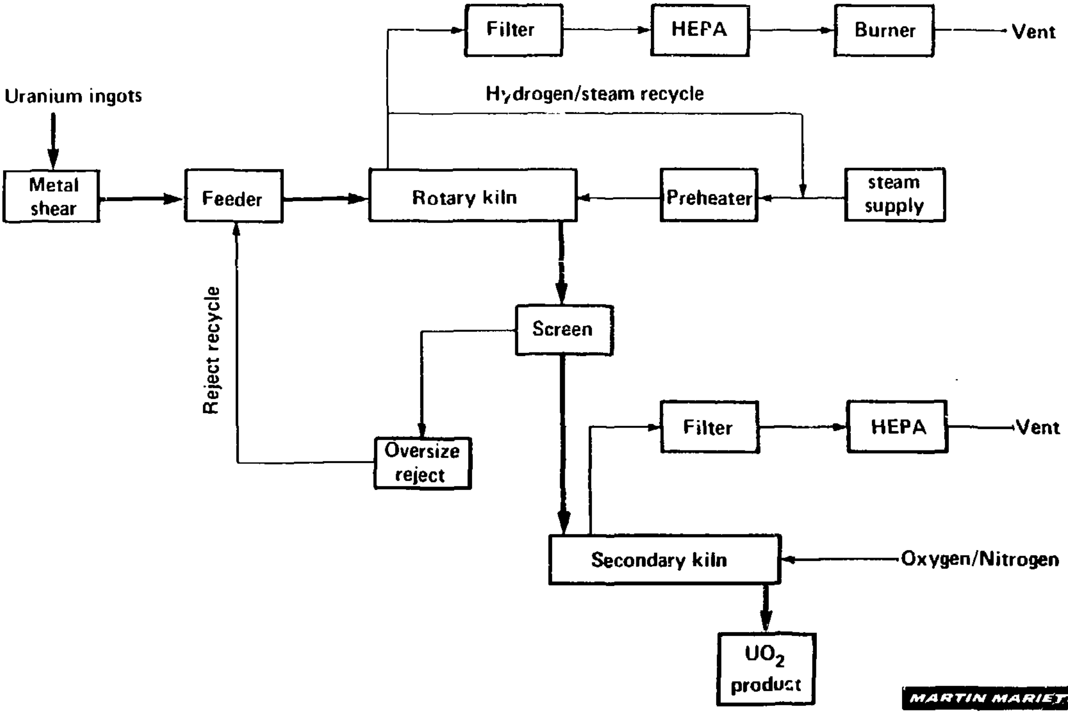
# URANIUM STEAM OXIDATION TO $UO_2$



Filter

HEPA

Burner

Vent

Uranium ingots

Hydrogen/steam recycle

Metal shear

Feeder

Rotary kiln

Preheater

steam supply

Reject recycle

Screen

Oversize reject

Filter

HEPA

Vent

Secondary kiln

Oxygen/Nitrogen

$UO_2$ product

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DWG. NO. K/G-86-1923

# PRIMARY CANDIDATES FOR AVLIS PRODUCT CONVERSION

DWG. NO. K/G-86-1909

- Wet solvent purification — ADU conversion to $UO_2$ is the primary technology and offers:
  - Low production cost
  - Low cost sensitivity to waste disposal costs
  - Conventional, mature technology and production bases

- We are currently evaluating *direct denitration* conversion to $UO_2$ to provide a dry processing alternative to the wet ADU process, because we expect:
  - Low production costs
  - Even less cost sensitivity to waste disposal costs
  - More technically mature dry route than steam oxidation to $UO_2$

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# ECONOMIC RANKING OF VARIOUS URANIUM PROCESSING ALTERNATIVES

DWG. NO. K/G-86-2055

## FEED CONVERSION (PROJECTED $U_3O_8 \rightarrow$ U-metal)

- ELECTROLYTIC REDUCTION
- PYROMETALLURGICAL PROCESS
- THERMITE BOMB REDUCTION ($UF_4$)
- THERMITE BOMB REDUCTION ($UF_6$)

INCREASING COSTS ↓

## PRODUCT FABRICATION (U-metal $\rightarrow UO_2$)

- DISSOLUTION/SOLVENT EXTRACTION--ADU PRECIPITATION
- DISTILLATION/STEAM OXIDATION (1 CYCLE)
- DISTILLATION/STEAM OXIDATION (MULTIPLE CYCLES)
- PYROMETALLURGICAL/STEAM OXIDATION
- FLUORINATION TO $UF_6$, ADU OR DRY PROCESSING TO $UO_2$

INCREASING COSTS (EXPECTED) ↓

## ALSO UNDERSTUDY

OTHER STUDIES UNDERWAY

- DISSOLUTION/SOLVENT EXTRACTION--DIRECT DENITRATION

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# SUMMARY

- $UF_6$ IN — $UF_6$ OUT METHOD OF OPERATION FOR AVLIS DOES NOT PROVIDE THE LOWEST OVERALL FUEL CYCLE COSTS
- A SYSTEMS ANALYSIS APPROACH HAS BEEN PERFORMED WHICH IDENTIFIES LOW COST OPTIONS FOR BOTH NATURAL URANIUM METAL FEED PREPARATION AND ENRICHED METAL PRODUCT CONVERSION
- WHILE THESE ALTERNATIVE PROCESSING PATHS WILL REQUIRE SOME RD&D EFFORT, THERE APPEARS ADEQUATE TIME TO COMPLETE ALL DEVELOPMENT AND DEMONSTRATION NECESSARY TO BE IN PRODUCTION FOR PROCESSING URANIUM WHEN AN AVLIS PRODUCTION PLANT IS DEPLOYED
- BY WORKING TOGETHER, THESE ALTERNATIVE PROCESSING PATHS SHOULD OFFER ECONOMIC OPPORTUNITY TO ALL PARTIES INVOLVED INCLUDING THE DOE, UTILITIES, FEED CONVERTERS, FUEL FABRICATORS AND ULTIMATELY THE ELECTRIC CONSUMERS