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TITLE

INTERIM REPORT
A CLOSED URANIUM CYCLE AT HAPO
PHASE I

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INTERIM REPORT
A CLOSED URANIUM CYCLE AT HAPO
PHASE I

BY

W. R. HAMILTON
ADVANCED ENGINEERING
ENGINEERING OPERATION
FUELS PREPARATION DEPARTMENT

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INTERIM REPORT
A CLOSED URANIUM CYCLE AT HAPO
PHASE I

DECLASSIFIEDINTRODUCTION:

This study was undertaken to determine the most feasible and economical process or path for closing the uranium fuel cycle at HAPO, and to establish what benefits, other than improvement in FPD's competitive position, would result from the selected closed fuel cycle. The study was separated into four phases; Phase I includes the selection and organization of plausible processes and the establishment of a realistic and effective evaluation procedure, Phase II will include evaluation and selection of an optimum path based on a medium range and a short range approach (5 to 10 years), Phase III will include evaluation and selection of an optimum path based on a long range approach (>10 yrs.), and Phase IV will include refinement of previous work and issuance of a final report summarizing the study and the conclusions or recommendations which develop from the study.

Phase I of the study has been completed. The purpose of this paper is to document the assembled data and the completed work.

SUMMARY:

The general problems of a closed fuel cycle must be clearly defined and some definite limits must be established, either by necessity or by arbitrary selection if a study of this magnitude is to have any direction or logic. Once the problems and limitations are clearly understood, a method of evaluation may be established which will permit a realistic selection of an optimum path from the many alternatives.

Among the many complex facets of the closed fuel cycle which were considered were past recycle experience, isotope accumulation, selection of anchor points in the present process, limitations imposed by this anchor point selection, uranium flow rates, selection of a typical stream, and characteristics common to any closed cycle such as basic categories, basic sequences, and re-enrichment. Enrichment is by far the most important single problem of closing the fuel cycle, since it has great influence on the recycle material balance, irradiation reactivity controls, total investment and operating costs and criticality control.

All processes which could logically be a part of the closed fuel cycle would have to be considered individually and in order to keep these processes in their proper perspective, a method of classifying and organizing them was essential. Three categories based on the physical form of uranium entering the purification process were established: Category 1 based on an aqueous solution of uranium salts, covered three different separation processes; extraction, precipitation, and ion exchange. Category 2, representing processes starting with liquid molten uranium was labeled "Pyrometallurgy". Category 3, based on starting with liquid UF_6 , was the sequence commonly called the fluoride volatility process. The presentation of individual processes under appropriate operations in each of these three categories constitutes a major portion of this report.

A method of evaluating the individual steps or processes was developed and expressed as a list of questions (p. 8) which when answered would define the process or step so that its relative merit could be realistically established. This should permit effective comparison and final selection of steps or processes.

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DECLASSIFIEDDISCUSSION:Effects of Recycle:

A study of past recycling of uranium at HAPO (1) revealed that although variations in pile reactivity were observed with recycled fuel, no significant problems resulted from its use. Isotope accumulation resulting from repeated recycle has been evaluated based on isotope distribution calculations⁽²⁾⁽³⁾. Results indicate that complete loading of K-pile with 10-times recycled metal would result in a reactivity loss of 150 inhours due to U-236 buildup, and cooling time after reactor discharge would have to be increased by about two weeks due to U-237 buildup. The same data indicate that operation would be feasible after twenty recycles. (A cycle is roughly equivalent to one year).

The Basic Premise:

In order to maintain a consistent philosophy or frame of mind in a study of this magnitude, it is well to have a stated premise or basis to which all the actions at HAPO, including this study, can be related. The premise for the initial study consists of three statements.

1. The ultimate goal at HAPO is the maximum plutonium production available commensurate with supplying all market needs for transuranic elements and fission product "source" material.
2. A constant composition uranium compound or alloy can be used (as well as the pure metal) in the graphite moderated reactors at HAPO.
3. An optimum number of U-235 enrichment levels exists that will result in a maximum plutonium production at a lowest cost under conditions of Item 1.

Anchor Point Selection:

One of the first considerations relating to a closed fuel cycle must be establishing what part of the existing process will be left intact. There must be a start and a finish, that is, two anchor points. The more of the existing process left intact between the two anchor points, the more restraint is imposed on the section remaining to be closed.

Three fairly obvious choices of anchor points are immediately apparent. These are illustrated in Figure 1. The most obvious choice is from door-to-door at HAPO. The second choice is door-to-door at the reactors. The third choice would be a single point, i.e., no restrictions. All three must eventually be considered if this study is to be inclusive and yet remain open-minded. However, a selection must be made to form a basis for initial study.

The present thinking at HAPO involves predictions that reactors as much as 14 years old can reasonably be expected to last ten more years. In view of the fact that some reactors are only two years old, a need for reactor replacement cannot be used to support the third "no restrictions" approach except on an extremely long term basis. However, an effort to make our most advanced separation plant, Purex, obsolete in five years is being seriously considered at HAPO, and this would tend to support the second or door-to-door reactor approach.

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Since the door-to-door reactor (DTDR) approach will also undoubtedly lead to investigation of paths that will satisfy the door-to-door HAPO (DTDH) approach, the DTDR approach is the logical selection for initial study.

Restrictions Imposed by Anchor Point Selection:

With the reactor entrance and exit established as anchor points, certain definite restrictions have automatically been imposed on the portion of the cycle remaining to be closed. The limitations imposed by the present irradiation process are listed below:

1. The uranium form for fuel elements must be a solid, that is, either uranium metal, uranium alloy, or a uranium compound.
2. There is a definite limit on the impurities permissible in the uranium form for fuel elements.
 - (a) Some impurities change the physical properties of the uranium causing difficulty during fuel element forming and during exposure in the reactor.
 - (b) Certain impurities have large neutron capture cross sections and will seriously poison a reactor when present even in very small amounts.
 - (c) Gases dissolved in the uranium can create great problems in maintaining a good bond between the fuel and the cladding.

An example of impurity effects on the physical properties of metallic uranium is the cracking of uranium metal under working when iron is present in concentrations above several hundred parts per million. Carbon, on the other hand, is a grain refiner and should be beneficial (within limits, of course) in avoiding warpage during irradiation which is caused by large (statistically) oriented grains. Hydrogen absorbed in uranium upon being evolved after or during cladding can separate the cladding-to-element bond.

The effect of foreign elements on pile reactivity is such that if an element has a neutron capture cross section of 1 barn or less (O, C, He, Be, F, Bi, Mg, Si, Pb, Zr, P, Al, H, Ca, Na, S, A, Sn, Ce, Rb) the effect is essentially one of dilution, that is, a decrease in density. Density may be decreased 1 to 5 per cent without decreasing reactivity any more than an estimated 3.5 per cent.⁽⁴⁾ For elements with cross sections much greater than 1 or 2 barns this simple dilution effect no longer holds and very minor amounts (less than 0.5%) can result in significant reactivity decreases.

Stream Selection:

Having established a fairly inclusive set of ground rules for closing the uranium cycle, it would be well to determine what specific materials will make up the total stream and what flow rates can be anticipated. Present forecasts⁽⁵⁾⁽⁶⁾ indicate that within ten years the material flow will have leveled out at values as shown below:

<u>Type Fuel</u>	<u>Abbr.</u>	<u>%U-235</u>	<u>Flow, %</u>
Natural Uranium	N	0.71	77.6
Enriched Uranium	E	0.94	10.0
NPR Fuel	NPR	0.80	9.3
Off-site Power Fuels	OS	0.2 - 3.0	3.1

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The consumption or burn-out of U-235 predicted for each of these fuels is summarized in the following table.

<u>Type Fuel</u>	<u>Initial % U-235</u>	<u>Final % U-235</u>
N	0.71	0.61
E	0.94	0.80
NPR	0.80	0.61
OS (7)		
Yankee	2.6	1.9
Dresden	1.5	0.7
CPPD	3.5	2.7
Piqua	2.5	1.4
Chugach	1.6	1.3
PRDC	0.71	0.51
		0.21
Northern States	1.6	0.9

The final or depleted U-235 concentration will be representative of the uranium stream until it reaches the re-enrichment step, and so will be of major concern in the closed cycle. Due to high enrichment costs, it is desirable to avoid diluting enriched uranium (if all depleted streams were combined, the final enrichment would be about 0.64% U-235). This means that depleted streams of different U-235 concentrations must be kept separated during processing. Presently action and funds are directed toward establishing processes and equipment to keep enriched uranium (E) and off-site uranium (OS) separate from each other and from the major stream. Off-site uranium will have to be kept in separate batches since power fuel burnout charges by the AEC will be based on uranium isotopic analyses obtained at HAPO. In view of these considerations, it was assumed that NPR and normal fuels would be depleted to the same final U-235 content, 0.61%, and would be combined during processing. This total stream would represent 87% of the total uranium flow and was taken as the characteristic or representative stream for this first analysis of the closed cycle.

The Eight Basic Steps That Close the Cycle:

The path from reactor door to reactor door can be completed in eight basic steps.

1. Decladding and dissolution.
2. Decontamination.
3. Separation.
4. Conversion to a "salt".
5. Conversion to metal.
6. Core formation.
7. Cladding core.
8. Re-enrichment.

The re-enrichment step, 8, can be inserted anywhere prior to core formation and will involve different physical operations in each case, depending on the uranium form at the selected point. The two conversion steps, 4 and 5, conceivably could be eliminated, but

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the others are common to all paths. The purpose of the decontamination and separation steps, 2 and 3, is the separation of plutonium and uranium from each other and the fission products. The separation of three materials by two steps or processes leads to three possible basic purification sequences.

Basic Purification Sequences:

Three purification sequences are illustrated in Figure 2 and have been arbitrarily numbered one through three as shown, based on the material which is first removed from the stream.

Sequence 1: (Immediate fission product removal)

This sequence is in use at HAPO and would be considered during any short term (DTDH) approach to closing the cycle. It includes separation by extraction, precipitation and ion exchange.

Sequence 2: (Immediate plutonium removal)

This order of operation is basic to most pyrometallurgical processes, and has the advantage of isolating criticality problems early in the process. However, the inherent maintenance and operating problems that result from remote handling of the highly radioactive fission products will remain to the last sequence step.

Sequence 3: (Immediate uranium removal)

The simplest uranium flow yields the most complex plutonium purification. The high fission product radioactivity combined with the high toxicity of plutonium and the criticality problem will require carefully engineered facilities, equipment, and procedures for safe operation and maintenance. The fluoride volatility process falls into this category.

PROCESS ORGANIZATION:

Three process categories were established based on the liquid form of uranium entering the purification sequence, i.e., an aqueous solution of a uranium salt such as uranyl nitrate hexahydrate (UNH), molten uranium, and liquid uranium hexafluoride (UF₆). By coincidence these included all three of the purification sequences described above, and each category was given a number corresponding with the appropriate sequence number.

<u>Category Number</u>	<u>Uranium Liquid Form</u>	<u>Separation Process</u>	<u>Sequence Number</u>	<u>First Material Separated</u>
1	Aqueous Salt Solution	Extraction, Precipitation, Ion exchange	1	Fission Products
2	Molten Metal	Pyrometallurgy	2	Plutonium
3	Liquid UF ₆	Fluoride Volatility	3	Uranium

The known processes were organized on large block diagram "work sheets" for each category, since a block diagram is one of the easiest ways to visualize a sequence of operations. Each category was divided into a series of vertical columns, each of which represented a

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general operation such as decladding, dissolution, decontamination, separation, etc. Similar specific operations were then shown within appropriate columns as a block. The uranium form in and out of each operation was indicated on the left and right sides, respectively, of each block. Typical examples of the resultant block diagrams are illustrated in Figures 3, 4, and 5. Since operations such as decladding, dissolution, core fabrication, cladding, and enrichment are common to all three categories, the major difference between categories is the process series required between purification and enrichment and return of the uranium to metallic form.

With processes thus categorized, the many special procedures which have been developed can be kept in proper perspective for valid comparison. Procedures which eliminate steps, actually shortening the cycle, become more obvious. A proposed shortest path is indicated on each diagram (Figures 3, 4, and 5).

Re-enrichment:

Re-enrichment is the most important single problem of closing the fuel cycle. It has significant influence on the recycle material balance, reactivity control, operating and capital costs, and criticality control.

Enrichment can be obtained by isotope separation or by blending high and low enrichment uranium. At first glance isotope enrichment in a facility based on isotope separation appears to be the only way to completely close the cycle. However, a material balance around a diffusion plant reveals that even if the waste is stripped completely of U-235 and only natural concentration uranium is produced, the total product will only be 85% of the feed. In other words, even with a perfect diffusion plant, at least 15% of the cycle flow would have to be purchased from off-site. (There would also be 15% of flow accumulating as waste).

This is at best a partial recycle.

Since off-site uranium purchase is inevitable even with a special enrichment facility at HAPO, blending to desired enrichments using high enrichment material (greater than natural concentration) from the cascades was investigated. Financial studies(8)(9) based on the AEC U-235 price curve (cost per Kg versus enrichment) indicate that it is cheapest to blend enrichments which are nearly equal, that is, as the difference between the two enrichments being blended increases, costs increase. However, this cost increase is gradual enough to permit blending of low difference enrichments for only slightly greater costs than buying natural uranium.

The blending operation, although simple in principle, is complex in practice because of the strict requirements on the final blend or product. Specifications of concentration variation permitted on the final U-235 concentration are as low as $\pm 0.0005\%$ (2). Problems of analytical accuracy, obtaining representative samples, maintaining large inventories of nearly acceptable materials, obtaining thorough mixing of large volumes, and avoiding a critical mass are directly involved in blending. A more indirect, although extremely important problem is reactivity control, since the fuel reliability must eventually be evaluated by pile behavior.

Selecting the material form of uranium to be blended has great influence on blending equipment and procedures required. It can be blended as a solid powder (UO_3 , UO_2 , UF_4), as a liquid (Aqueous salt solution, UF_6 , molten uranium), as a gas (UF_6), or as a solid (metal) to a liquid (molten metal). Blending in liquid form permits greater operational flexibility.

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THE TEST:

Having organized the many processes that can logically be a part of the closed fuel cycle, a procedure or test for comparing and evaluating individual equivalent processes is mandatory. A test has been established in the form of a series of questions, some of which are very general in order to encompass all processes. The specific questions composing the "test" are listed below:

1. Can this step be eliminated?
2. Is this step technically feasible?
 - a. Is this an efficient step?
 - b. What are the yields and losses?
 - c. Is equipment size (production) limited by criticality?
 - d. What are the power requirements?
 - e. Are materials of construction a problem?
 - f. What special equipment or raw material is required?
 - g. Has it been done on a plant scale?
 - h. How much development will be required?
 - i. Is the process amenable to instrumentation for control?
3. Can it be done in existing plants and/or equipment?
 - a. What are the personnel hazards?
 - b. Is there danger of geographical contamination?
 - c. Is remote handling necessary?
 - d. What are the restrictions imposed by criticality?
 - e. What type of building is required?
 - f. Is such a building presently available?
 - g. What change in present buildings and equipment is needed?
 - h. What are personnel requirements? (See 2 i above).
 - i. Are personnel available?
4. How do process costs compare with other methods?
 - a. Estimated capital expenditures?
 - b. Will operating costs be reasonable? (1) labor cost
(2) raw material cost
(3) utility cost
 - c. What depreciation rates appear reasonable?
5. What is the fission product build-up, removal, or effect?
 - a. Should f.p.'s be removed? decayed? or handled hot?
 - b. What are the effects of these fission products.
6. Will the product meet specifications?
 - a. If not, what steps will make it okay?
 - b. Does this step add significantly to impurities?

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7. What is the isotopic buildup, removal, or effect of this step?
8. Should the U-235 be added here? Can reactivity control be established here?
9. Should the natural uranium be added here?

FUTURE WORK - PHASE II:

Using the work sheets shown in Figures 3, 4, and 5, a shortest path will be selected in each category based mainly on the minimum number of steps, with appropriate consideration of the order of magnitude of individual steps. The selected paths will then be compared step by step in an effort to establish which path is the optimum path. This sequence will be approached twice; once with a short term (approximately 5 years) approach, and once with a long term (>10 years) approach. Another interim report will be issued following completion of Phase II.

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* Recorded in HWN-2247.

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THREE BASIC APPROACHES TO CLOSING THE FUEL CYCLE AT HAPO

FIGURE 1

SEQUENCE 1. (Immediate F.P. Removal)

SEQUENCE 2. (Immediate Pu removal)

SEQUENCE 3. (Immediate U removal)

Circles = Materials in stream.

Rectangles = Operation performed on entering material.

F.P. = Fission Products

Pu = Plutonium

U = Uranium

- - - - - = Criticallity Problem

FIGURE 2

THE THREE BASIC PURIFICATION SEQUENCES

CATEGORY I - LIQUID EXTRACTION - PRECIPITATION - ION

ION EXCHANGE

1013
2

HYDROFLUORINATION
TO UF_4

FLUORINATION
TO UF_6

CTOR UO_2 UO_2 HF TRAY REACTOR UF_4
BED UO_2 UO_2 HF FLUIDIZED BED UF_4
BED UO_2 UO_2 HF MOVING BED UF_4

W/TH BIFLUORIDE OR NH_3 & HF
700°C UF_4

EDN 114 ($CCl_2Cl_2F_4$)
700°C UF_4

COUANE 9 FLAME UO_2 UO_2 $C + Cl_2$ UCl_4

UO_2 UO_2 CCl_4 UCl_4

UO_2 UO_2 S_2Cl_2 UCl_4

UO_2 UO_2 PHOSGENE UCl_4

CHLORIDE $\Rightarrow$ FUSED SALT CHLORINATION TO UCl_3 UCl_3

LYSIC NH_4UF_5 or $UF_4 \cdot xH_2O$ DECOMPOSITION BY HEATING $\sim 300^\circ C$ UF_4

DEHYDRATION $\sim 500^\circ C$ UF_4

PELLETS UF_4 (CO + H_2O) HYDROFLUORINATION (MOVING BED) UF_4

PRECIPITATION OF "GREEN SALT" WITH AQUEOUS HF FILTRATION, ADD Na_2SO_4 HF $NaUF_5$

ELECTROLYTIC REDUCTION U^{IV} $U(50\pm 2)$

UO_2SO_4 UO_2

UF_4 F_2 TRAY REACTOR UF_6

UF_4 F_2 FLUIDIZED BED UF_6

UF_4 F_2 MOVING BED UF_6

UF_4 O_2 TRAY REACTOR UF_6

UF_4 O_2 FLUIDIZED BED UF_6

UF_4 O_2 MOVING BED UF_6

UF_4 F_2 (ALSO WITH C) UF_6

UF_4 UF_6

UF_4 UF_6

UF_4 UF_6

UF_4 UF_6

UF_4 UF_6

UF_4 UF_6

UF_4 UF_6

UF_4 UF_6

UF_4 UF_6

UF_6 + (UO_2F_2) RECYCLE

PRECIPITATION WITH AQUEOUS HF UF_6

REDUCTIONS
TO UF₄

ISOTOPE
ADJUSTMENTS

REDUCTIONS

UF₆ (GAS)
UF₆ (LIQ)

UF₆ (GAS)
UF₆ (LIQ)

UF₆ (GAS)
UF₆ (LIQ)

UF₆ (GAS)
UF₆ (LIQ)

UF₆ (GAS)
UF₆ (LIQ)

UF₆ (GAS)
UF₆ (LIQ)

UF₆ H₂ UF₄

UF₆ HBr UF₄

UF₆ HCl UF₄

UF₆ SOCl₂ UF₄

UF₄ ENRICHED UF₄ ADDITION

UF₄ NORMAL UF₄ ADDITION

UF₄ CATIONIC ELECTROMIGRATION (FUSED SALT)

DOUBLE
SALTS

UO₂U₃O₈
UF₄ UCl₄
UF₄

UO₂U₃O₈
UF₄ UCl₄
UF₄

UO₂U₃O₈
UF₄ UCl₄
UF₄

UCl₄ UF₄

UCl₃

MgF₂ BOMB

Al

ALKALINE METALS
Na, K

ELECTROLYTIC
REDUCTION

DRUM
(NA VAPOR)

ELECTROLYTIC
REDUCTION TO
METAL

RE WITH LI

BOMB
USING

FUNCTIONS
C, I₂

FIGURE 3 - CATEGORY 1

LIQUID-LIQUID EXTRACTION:
 PRECIPITATION:
 ION EXCHANGE:

<u>DECLADDING</u>		<u>DISSOLUTION</u>	<u>SEPARATION</u>
Al/U	NaOH	U Melting 1135°C	U Distillation
Al/U	Melting 700°C		U Molten Ag Extraction
Zr/U	Zirflex		U Molten Mg Extraction
Zr/U	Zircex		U Fused Salt Extraction
Zr/U	Aqueous HF		
SS/U	Sulphex		
Al/U Zr/U SS/U	Mechanical		

FIGURE 4 - PART 1

CATEGORY 2 - PYROMETALLURGICAL PROCESSES

<u>DECONTAMINATION</u>		<u>ISOTOPE ADJUSTMENT</u>	<u>CORE FABRICATION</u>		
U	Molten Chloride Extraction	U	Liquid Thermal Diffusion	U	Cast
U	Slagging	U	Enriched U Addition	U	Forge-Roll- Machine
U	Drossing	U	Normal U Addition		
U	Filtration	U	Distillation Diffusion		
U	Chloride Conversion				
U	Recrystalli- zation From Hg				
U	Electrorefining				
U	Zone Melting				

FIGURE 4 - PART 2

CATEGORY 2 - PYROMETALLURGICAL PROCESSES

<u>DECLADDING</u>		<u>DISSOLUTION</u>	<u>SEPARATION</u>
Al/U	NaOH U	U BrF ₃ UF ₆	UF ₆ Distillation- Bubble Cap UF ₆
Al/U	Melting U	U BrF ₅ UF ₆	UF ₆ Distillation- Packing UF ₆
Zr/U	Zirflex U	U ClF ₃ = HF UO ₂ UF ₆	UF ₆ Distillation- Sieve Plate UF ₆
Zr/U	Zircex U	U HF-Molten UO ₂ Salt UF ₄ F ₂ Molten Salt UF ₆	UF ₆ Absorption- Desorption UF ₆
Zr/U	Aqueous HF U	U F ₂ UO ₂ UF ₆	
SS/U	Sulphex U		
Al/U Zr/U SS/U	Mechanical U		
Zr/U SS/U	Chopping and Leaching UF ₆		

FIGURE 5 - PART 1

CATEGORY 3 - FLUORIDE VOLATILITY

FIGURE 5 - PART 2

CATEGORY 3 - FLUORIDE VOLATILITY

FIGURE 5 - PART 3

CATEGORY 3 - FLUORIDE VOLATILITY

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