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URANIUM OXIDE ACTIVATION COST STUDY

By

R. W. McKee

Engineering Development Planning
Chemical Research & Development
HANFORD LABORATORIES OPERATION

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GROUP 1

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URANIUM OXIDE ACTIVATION COST STUDY

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INTRODUCTION

Late in 1956 continuous uranyl nitrate calcining facilities were placed in operation at Hanford. The successful and more economical operation of these facilities has resulted in the recent shutdown of the old batch-type pot calciners. Operating costs have been reduced from 16.6¢ to 11.9¢ per pound of uranium processed.*

A disadvantage for the new process, however, has been a reduction of the so-called reactivity of the uranium oxide powder produced. This results in lower conversion to UF_4 in the hydrofluorination process and subsequent increased consumption of fluorine in the conversion of the UF_4 to UF_6 .

Various processing techniques and additional processing steps have been devised to increase the oxide powder reactivity. For example, addition of sulfate (6000 ppm. U) to the UNH improves the product appreciably and has been adopted as routine procedure.

The most effective treatment so far developed for increasing reactivity consists of a fluidized-bed reduction with hydrogen to UO_2 followed by a fluidized-bed reoxidation with air to U_3O_8 . A possible explanation is that the activation results from separating the crystallites in the individual particles and by the formation of pores. It is claimed that this treatment will increase the reactivity of any commercially prepared UO_3 powder regardless of the method of its manufacture and that the U_3O_8 product reactivity will be the same for all sources of UO_3 . In other words, the process erases the past history of the oxide powder. A several-fold increase in rate of hydrofluorination and 100% conversion to UF_4 is claimed, but large-scale testing that would firmly establish the degree of improvement has not been carried out.

This study was initiated to establish the magnitude of the incentive at Hanford for carrying out pilot-scale development work by determining the cost of a full-scale UO_3 activation unit and the savings that would accrue. The scope of the study included:

- (a) Developing a production-scale process design.
- (b) Obtaining a preliminary construction cost estimate.
- (c) Estimating operating costs.
- (d) Estimating savings and justification for further development work.

SUMMARY

A process design was developed for the activation of uranium oxide powder by fluidized-bed reduction to UO_2 and reoxidation to U_3O_8 . Installation of the process with a capacity equal to the installed continuous calciners is estimated to cost \$700,000 if installed in 224-U Building after removal of the electric

*HW-47786, "Operating Cost Evaluation - Oxide Operation," R. E. Olson, February 13, 1957.

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calcining pots or \$860,000 if installed in a new building. Cost of operating this installation is estimated at \$270,000 per year.

Assuming the full capacity of the equipment would be required, justification for the installation would consist of \$250,000 to \$300,000 for saving the addition of fluorine cells that would otherwise be needed and \$310,000 per year operating cost savings (fluorine consumption). Both savings would accrue at Paducah. Operating cost increases at Hanford and savings at Paducah thus leave a net savings of only \$40,000 per year to amortize approximately \$450,000 of additional capital construction cost (\$700,000 at Hanford less \$250,000 in construction not required at Paducah).

Two intangible benefits can also be credited to the process: (1) A substantial increase in the hydrofluorination reaction rate is predicted and would allow shut down of some hydrofluorination equipment because of increased capacity; (2) product from this process appears to be ideal for ceramic fuel element manufacture. However, the second item could only be credited to a pilot-scale process at Hanford since Hanford oxide would not be used for fuel element manufacture without enrichment.

It was concluded that a full-scale process cannot be justified at this time but that consideration should be given to pilot-scale development as a marginal project.

PROCESS DESIGN

The required process equipment is basically simple and consists of the fluidized-bed reactors for reduction and reoxidation, together with the necessary heating and cooling facilities, powder handling facilities, and process control instrumentation. The process was designed to have a capacity approximately equal to the maximum capacity of the installed continuous calciners or 45 T. U/day instantaneous capacity (36 T. U/day average). This rate exceeds present production forecasts but there is a reasonable possibility that it may be required within the next few years.

The possibility of converting some of the continuous calcining units for this process was considered but was discarded because (1) the full capacity of the calciners will eventually be required for calcination and (2) it would be very difficult to adapt the calciners to a pressurized system which is required for safety reasons.

The details of the process design developed for this study are shown in the attached Figure 1 flowsketch. The design is not claimed to be optimum, necessarily, but is intended to be a reasonable presentation of equipment requirements, sufficiently detailed for a preliminary estimate.

A material balance and heat balance for the process are shown on the attached Figures 2, 3, and 4.

The following basic assumptions were used in developing the process design:

1. Capacity to be 45 T. U/day instantaneous or 36 T. U/day (1080 T. U/month) average (80% service factor).
2. Feed must be milled UO₃ powder since particle size from the continuous calciner is difficult to control within the limits required for fluidized-bed operation.
3. Two sets of reactors should be provided rather than larger single units. This will insure greater process reliability and flexibility, as well as provide for the event that segregated facilities for enriched uranium may be required. In addition, efficient operation at reduced throughputs is possible with the smaller reactors. Other equipment, such as intermediate storage hoppers, would not be provided in duplicate.
4. Assume that the electric calcining pots can be removed and the vacated space utilized for this installation. The gas-fired Luckey pots could be left in standby status for emergency processing requirements or for possible segregated processing of enriched uranium. (The alternate possibility of installing the equipment in a new building was also evaluated.)
5. The existing dust collection and bulk loadout facilities in 224-U Building can be utilized for this process. This would not bar use of this equipment for Luckey pot operation should that become necessary.
6. The basic data for reactor design are presented in Table I.

The reactors themselves turn out to be rather small considering their capacity. The individual reactors for the two 2-stage reduction reactors are 18 inches in diameter with an operating bed depth of 6-1/2 feet. The two reoxidation reactors are each 26 inches in diameter with an operating bed depth of 2-1/2 feet.

A list of the major items of equipment required for this process is shown in Table II.

CONSTRUCTION COST ESTIMATE

Cost estimates were obtained for two alternate installations of the equipment. These were:

1. Removal of the 18 electric calcining pots from Cells E and F in the 224-U Building and installation of the new equipment in this location. The estimated cost for this installation is \$700,000. A detailed breakdown of the estimate is presented in Table III.
2. Construction of a new steel frame and transite building on the southwest end of the 224-U Building to house the new equipment. The estimated cost for this installation is \$860,000. A detailed breakdown of the estimate is presented in Table IV.

These estimates include installation of tubular stainless-steel-screen filters in the reduction reactor off-gas line. There is a good possibility that these could be deleted to reduce the cost estimate by approximately \$15,000. They were included in the design to (1) avoid necessity of recycling the powder recovered in the bag filters and (2) to avoid possible harmful effect on UO₂ powder fines if subjected to the high temperatures of the hydrogen burner.

The estimates were prepared on the basis of current labor and material costs and can be considered good only until January 1958. An escalation factor of 5% per year should be added to the total project cost for starting the project after that date.

OPERATING COST ESTIMATE

The principal items of operating cost are essential materials and labor.

The only essential material required is the reducing agent. This cost estimate is based on using anhydrous ammonia at \$82.50/ton, as it appears to be cheaper than buying hydrogen gas or hydrogen generating equipment.

Direct labor requirements per shift are estimated to be one process operator at the control panel, one utility operator loading five-ton containers of UO₃ into the feed hopper and loading out five-ton containers of U₃O₈ for shipment, and approximately one-half of a process operator being shared with other UO₃ Plant operations to take care of routine equipment inspections, miscellaneous operating chores, and to assist in powder handling at times of peak loads. Indirect labor costs were estimated at 70% of direct labor costs.

No allowance was made for inflation since we are interested primarily in costs relative to savings, not necessarily absolute costs, and both would be equally affected by inflation.

Total yearly operating costs were estimated to be \$270,000 or 1.07¢/pound of uranium. A detailed listing of these costs is presented in Table V.

Adding in a depreciation charge of 10% per year would increase the unit cost to 1.35¢/pound U.

SAVINGS ESTIMATE

The savings that would result from installation of this process at Hanford would all accrue to the Paducah Plant where Hanford's UO₃ is converted to UF₆. (One minor exception is the savings, \$14,000 per year, in shipping cost for shipping U₃O₈ rather than UO₃. This item is included in the operating cost determination.) These savings result from a higher conversion of UO₂ to UF₄ during the hydrofluorination step which reduces consumption of the more costly fluorine during fluorination to UF₆. For example, 100 pounds of UO₂ can be converted to UF₆ with 29.6 pounds of HF and 14 pounds of F₂ or with 42.1 pounds of F₂ and no HF. With HF costing 23¢/pound and F₂ costing 43¢/pound, the cost in this case would be approximately \$18 per 100 pounds for using fluorine only compared to \$13 per 100 pounds for using HF for UF₄ production. Calculation of the actual savings is not quite so simple as this example, but it illustrates the principle involved and a detailed explanation of the calculation is attached as an Appendix.

The estimated savings for Paducah operations should not be interpreted as anything more than rough approximations, since intimate knowledge of Paducah operating costs is not available. The data are considered adequate for the purposes of this study but, if firm justification for a full-scale project is desired, either a thorough study of Paducah operations should be undertaken or Paducah should be requested to supply the justification data.

Cost comparisons based on fluorine and anhydrous HF costs were calculated for five cases. Results of these calculations are presented in Table VI.

Case I represents results of feeding Hanford continuous UO_3 to the Paducah fluid-bed reduction units followed by hydrofluorination in screw and tray reactors.

Case II represents results of feeding Hanford continuous UO_3 to partial reoxidation units after fluid-bed reduction and prior to screw and tray hydrofluorination. This step increases conversion to UF_4 and is current practice at Paducah.

Case III represents results of feeding Hanford pot powder to screw and tray reduction and HF units without fluid-bed reduction. This is presented for comparison purposes and illustrates the operation prior to Hanford continuous calcination. Production rates of this magnitude were never possible from the pots so this is a fictitious case.

Case IV represents results of feeding Hanford pot powder to Paducah fluid-bed reduction units followed by screw and tray hydrofluorination. This is another case presented for comparison purposes and is also fictitious in that the production rate exceeds the capacity of Hanford pots by a factor of two.

Case V represents the predicted result for activated Hanford continuous powder being fed to the Paducah fluid-bed reduction units followed by hydrofluorination in screw and tray reactors.

Estimated chemical costs are shown for two production rates in Table VI. The lower rate, 21 T. U/day, is the five-year production forecast; it is felt to be quite conservative and represents a production floor. The high rate, 36 T. U/day, matches the installed continuous calciner capacity and is within the limits of more optimistic forecasts. The high rate was used as the basis for the savings estimate in this study.

Production capacity of the Paducah equipment is also included in Table VI.

Information on the cost of the partial reoxidation step (Case II) is not available but, for the purpose of this study, is estimated at one-half the materials cost difference for conversion to UF_6 (Case I-Case II), or \$100,000 per year. With powder activation at Hanford, this cost would be eliminated and can be claimed as a savings.

The difference between Case V and Case II at the 36 T. U/day rate is the estimated direct savings for reduced fluorine consumption of \$210,000 per year. Adding in \$100,000 for the reoxidation step gives a total savings of \$310,000 per year. The total operating savings barely offset the operating costs and leave very little to amortize the capital cost. However, the capital cost is

partially offset by the capital cost for the addition of fluorine cells that would otherwise be required without the reactivation process. This is on the order of \$250,000 to \$300,000. The net result is summarized in the following table:

Operating Cost Savings at Paducah	\$310,000/yr.
Additional Operating Cost at Hanford	<u>\$270,000/yr.</u>
Net Savings	\$ 40,000/yr.
Capital Cost at Hanford	\$700,000
Capital Cost Savings at Paducah	<u>\$250,000</u>
Net Capital Cost	\$450,000

The estimated savings are considered optimistic since production rates are higher than firm predictions and since the assumed 100% conversion for activated powder may not be realistic. Inasmuch as this study was undertaken to justify possible development work and since all of the benefits may not have been known or defined, an optimistic approach seemed appropriate.

Two other sources of justification are possible:

1. The increased rate of hydrofluorination predicted for the activated UO₃ would increase hydrofluorination capacity at Paducah and permit shutdown of some equipment. The magnitude of savings should be substantial if this were possible; it would probably be on the order of several hundred thousand dollars per year. A plant-scale test in one of the Paducah units using powder produced in a pilot unit would probably be necessary to define the magnitude of these savings.
2. This same process that improves the reactivity of the oxide powder also imparts ideal properties for ceramic fuel element manufacture. This would not appear to provide any justification for treatment of Hanford's depleted uranium oxides but does provide some incentive for consideration of pilot-scale operations to supply powder for development purposes.

CONCLUSIONS

1. Consideration of this UO₃ activation process on a production scale is not justified at this time.
2. Consideration should be given to pilot-scale development of the ^{process} project as a marginal project. This may be justified on the basis of (a) the potential for shutting down hydrofluorination equipment at Paducah, which

a large-scale demonstration will resolve; (b) the interest in this activated material for fuel element development; and (c) the numerous applications for fluidized-bed processes indicates that some specialized work in this field at Hanford could be valuable.

Robert W. McKee

Engineering Development Planning
Chemical Research & Development
HANFORD LABORATORIES OPERATION

RW McKee:mj

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TABLE I

BASIC DATA FOR REACTOR DESIGN

A. Reduction Reactors

1. Process requires a two-stage fluidized reactor.
2. Minimum superficial gas velocity = 0.8 ft./sec.
3. Fluidized-bed density = 157 lb./ft.³
4. Ratio bed depth/diameter = 4.3.
5. Total holdup time = 1.7 hrs.
6. Operating temperature = 1160°F. ± 50°F.
7. Reaction starts at about 900°F.
8. Pressurized reactor necessary for safety reasons (prevent H₂ inleakage and possible explosion).
9. Reductant may be cracked NH₃ or H₂ diluted with steam or nitrogen. Three volumes H₂ per volume of diluent required.
10. Reaction: $UO_3(s) + H_2(g) \longrightarrow UO_2(s) + H_2O(g) - 28 \text{ Kcal/mol } UO_3.$
11. Design for twice the stoichiometric amount of H₂. *50,500 ft³/hr mol*

B. Reoxidation Reactors

1. Single-stage reactor satisfactory.
2. Maximum superficial gas velocity = 1 ft./sec.
3. Fluidized-bed density = 157 lb./ft.³
4. Reaction is very rapid, not limited by residence time but by UO₂ and air supply rates which in turn are limited by heat removal rates.
5. Reactor size based on heat transfer considerations.
6. Operating temperature = 1020°F. ± 50°F.
7. Reaction starts at about 750°F.
8. Reactor should be pressurized to avoid inleakage of air which might upset reactor control and also to provide more flexibility in control of excess air and fluidizing velocities.
9. Reaction: $UO_2 + 1/3 O_2 \longrightarrow 1/3 U_3O_8 - 23.8 \text{ Kcal/mol } UO_2.$
10. Design for twice the stoichiometric amount of air.

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TABLE II

UO₃ ACTIVATION PROCESS EQUIPMENT LIST

<u>Item</u>	<u>Description</u>	<u>Quantity</u>
1	5-Ton Hoist	1
2	Hoppers	3
3	Screw Conveyors	3
4	Cooling Screw Conveyors	4
5	Rotary Valves with Speed Control	4
6	Rotary Valve with On-Off Control	1
7	Reduction Reactors	4
8	Ratio Flow Controller	1
9	Flow Recorder-Controllers	13
10	Temperature Recorders (8 PT)	8
11	Gas Rotameters	9
12	Steam Orifice Meters	6
13	Control Valves	21
14	Temperature Recorder-Controllers	6
15	Pressure Recorder-Controllers	3
16	Butterfly Control Valves	3
17	Oxidation Reactors	2
18	Reduction Off-Gas Cooler	1
19	Oxidation Off-Gas Cooler	1
20	Off-Gas Filters	5
21	H ₂ Burner	1
22	Pressure Gauges	30
23	Calrod Controls (25 KW)	4
24	Calrod Controls (50 KW)	2
25	Bin Level Alarms	6
26	Piping	
27	H ₂ Supply Facility	1
28	Control Panel	1
29	Compressed Air Facility	1
30	Steam Supply Facility	1

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TABLE III

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STUDY STAGE COST ESTIMATE

UO₃ ACTIVATION PROCESS - ALTERNATE NO. 1

Construction Operation

Temporary Construction	\$ 5,000
Tie-ins	5,000
Start-up	10,000
Remove Electric Calcining Pots	16,000
Equipment	214,000
Piping	19,500
Instrumentation	85,500
Start-up	<u>10,000</u> ?
Sub-Total Direct Costs	\$355,000 ?
Construction Overheads	65,000
Construction Engineering	<u>35,000</u>
Sub-Total Construction Costs	\$455,000
Design and Administrative	90,000
Contingency	<u>155,000</u>
TOTAL COST	<u>\$700,000</u>

Estimating No.
ER-0223

Approved (Estimating)

R. L. Lysher, 4-19-57

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TABLE IV

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STUDY STAGE COST ESTIMATE

UO₃ ACTIVATION PROCESS - ALTERNATE NO. 2

Construction Operation

Temporary Construction	\$ 5,000
Tie-ins	5,000
Start-up	10,000

Fixed Price Contractor

Building, incl. Ventilation & Electrical	85,000
Equipment	255,000
Piping	30,000
Instrumentation	<u>110,000</u>
Sub-Total Direct Costs	\$500,000
Construction Overheads	12,000
Construction Engineering	<u>43,000</u>
Sub-Total Construction Costs	\$555,000
Design and Administrative	110,000
Contingency	<u>195,000</u>
TOTAL COST	<u>\$860,000</u>

Estimating No.
ER-0223

Approved (Estimating)

R. L. Lysher, 4-19-57

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TABLE V

URANIUM OXIDE ACTIVATION YEARLY OPERATING COSTS

Essential Materials		
Anhydrous ammonia		\$102,000
Direct Labor		56,500
Indirect Labor		39,000
Shop Tools and Supplies		18,500
Continuity of Service		8,700
Freight Costs		
Savings for shipping U as U ₃ O ₈ rather than UO ₃		(14,000)
Maintenance		23,000
Electricity		3,500
Laundry		9,500
Other Indirect Expense		2,600
Water, Steam, Sewage		4,600
Radiation Monitoring		3,100
Analytical Control		6,500
Process and Plant Services		5,000
Facilities Engineering		<u>1,500</u>
Total		\$270,000/year
Unit Cost	1.07¢/lb. U	
Depreciation at 10%/Year		<u>70,000/year</u>
Total Cost Including Depreciation		\$340,000/year
Unit Cost Including Depreciation	1.35¢/lb. U	

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TABLE VI

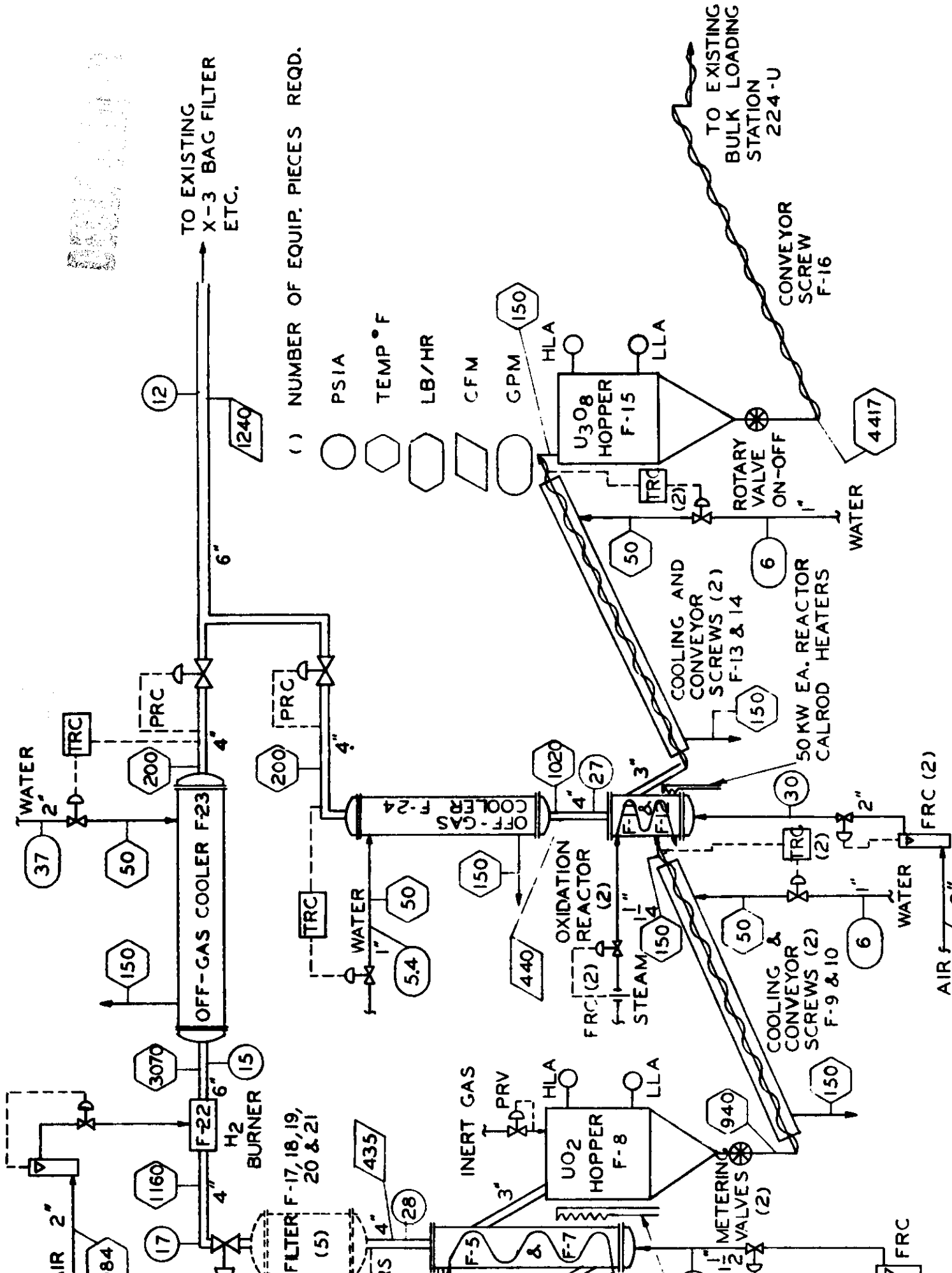
EFFECT OF PROCESSING METHOD ON HF AND F₂ COSTS
FOR UF₆ PRODUCTION

CASE	I	II	III	IV	V
Product from HF Line*					
UF ₄ , %	82	88	92	95	100
UO ₂ , %	15	6	6	3	0
UO ₂ F ₂ , %	3	6	2	2	0
Present Maximum Paducah Capacity, T. U/Day*	20	46	32	46	--
Estimated Cost of HF and F ₂ at 21 T. U/Day Average, M\$/Yr.	3,130	3,030	3,000	2,930	2,910
Estimated Cost of HF and F ₂ at 36 T. U/Day Average, M\$/Yr.	5,590	5,390	5,350	5,270	5,180

- Case I - Hanford continuous UO₃ fed to Paducah fluid-bed reduction followed by screw and tray hydrofluorination without reoxidation.
- Case II - Hanford continuous UO₃ fed to Paducah partial reoxidizer units after fluid-bed reduction and prior to hydrofluorination in screw and tray units.
- Case III - Hanford pot powder fed to Paducah screw and tray lines without fluid-bed reduction.
- Case IV - Hanford pot powder fed to Paducah fluid-bed reduction followed by screw and tray hydrofluorination.
- Case V - Hanford continuous U₃O₈ from activation process fed to fluid-bed reduction followed by screw and tray hydrofluorination.

*HAN-64543-1, "Operation of Green Salt Facilities," L. B. Emlet to S. R. Sapirie (December 11, 1956).

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TO EXISTING
X-3 BAG FILTER
ETC.

() NUMBER OF EQUIP. PIECES REQD.

○ PSIA

⬡ TEMP °F

▭ LB/HR

▭ CFM

⬡ GPM

TO EXISTING
BULK LOADING
STATION
224-U

CONVEYOR
SCREW
F-16

4417

WATER

COOLING AND
CONVEYOR
SCREWS (2)
F-13 & 14

50 KW EA. REACTOR
CALROD HEATERS

COOLING &
CONVEYOR
SCREWS (2)
F-9 & 10

AIR 3"

FRC (2)

WATER

1"

6

2"

3"

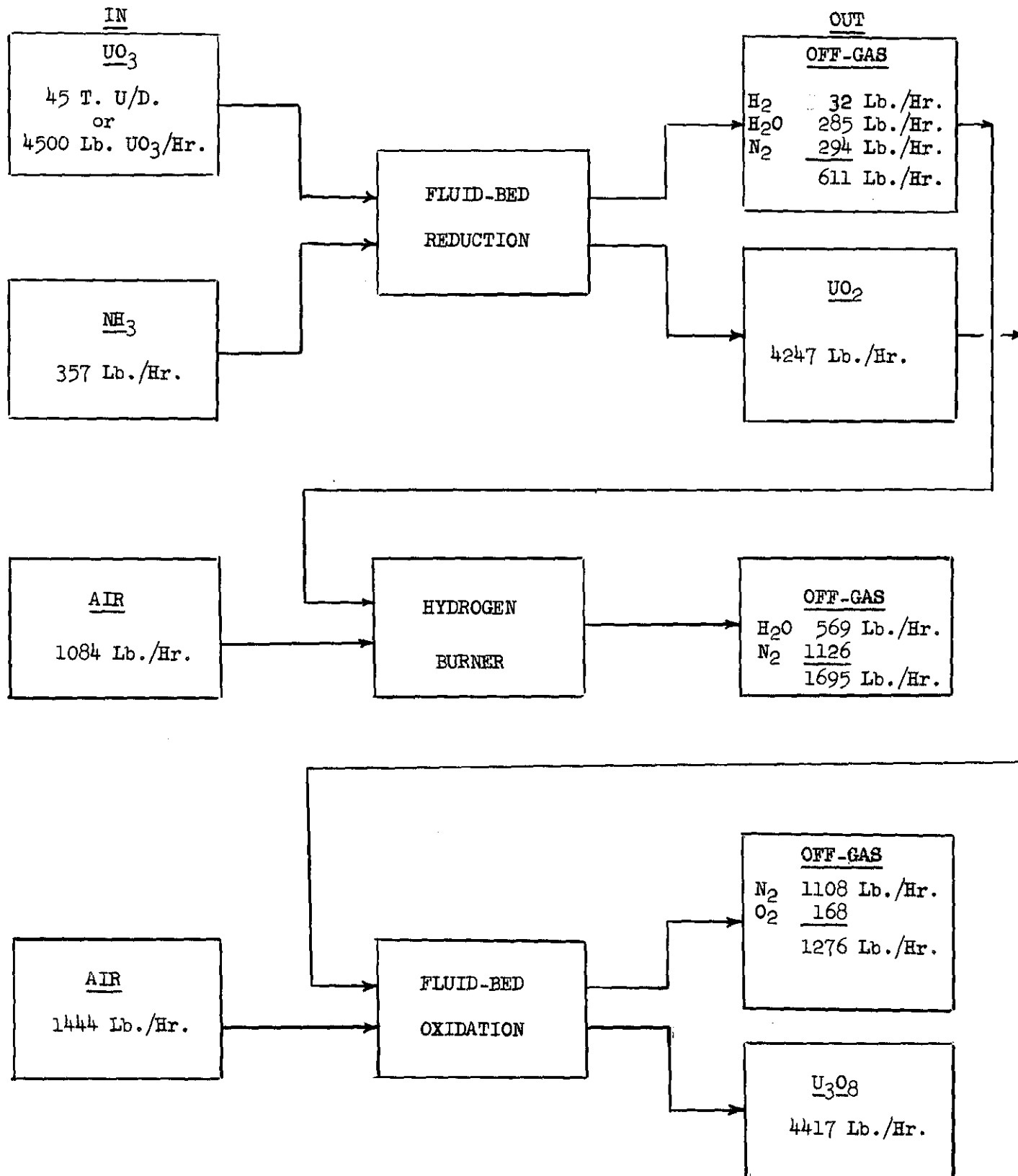
WATER

2"

FRC (2)

FIGURE 2
MATERIAL BALANCE

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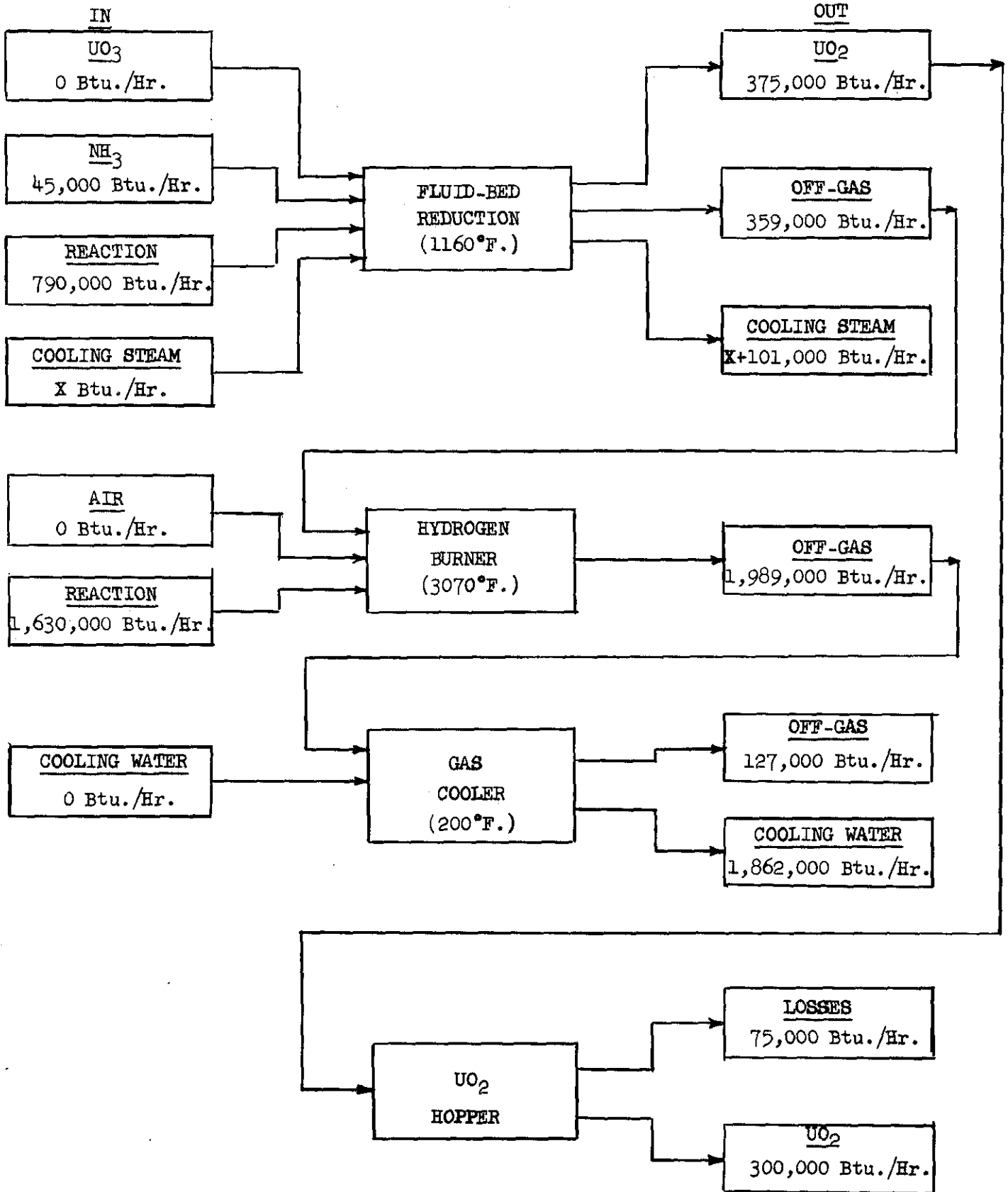


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FIGURE 3

HEAT BALANCE (REDUCTION)

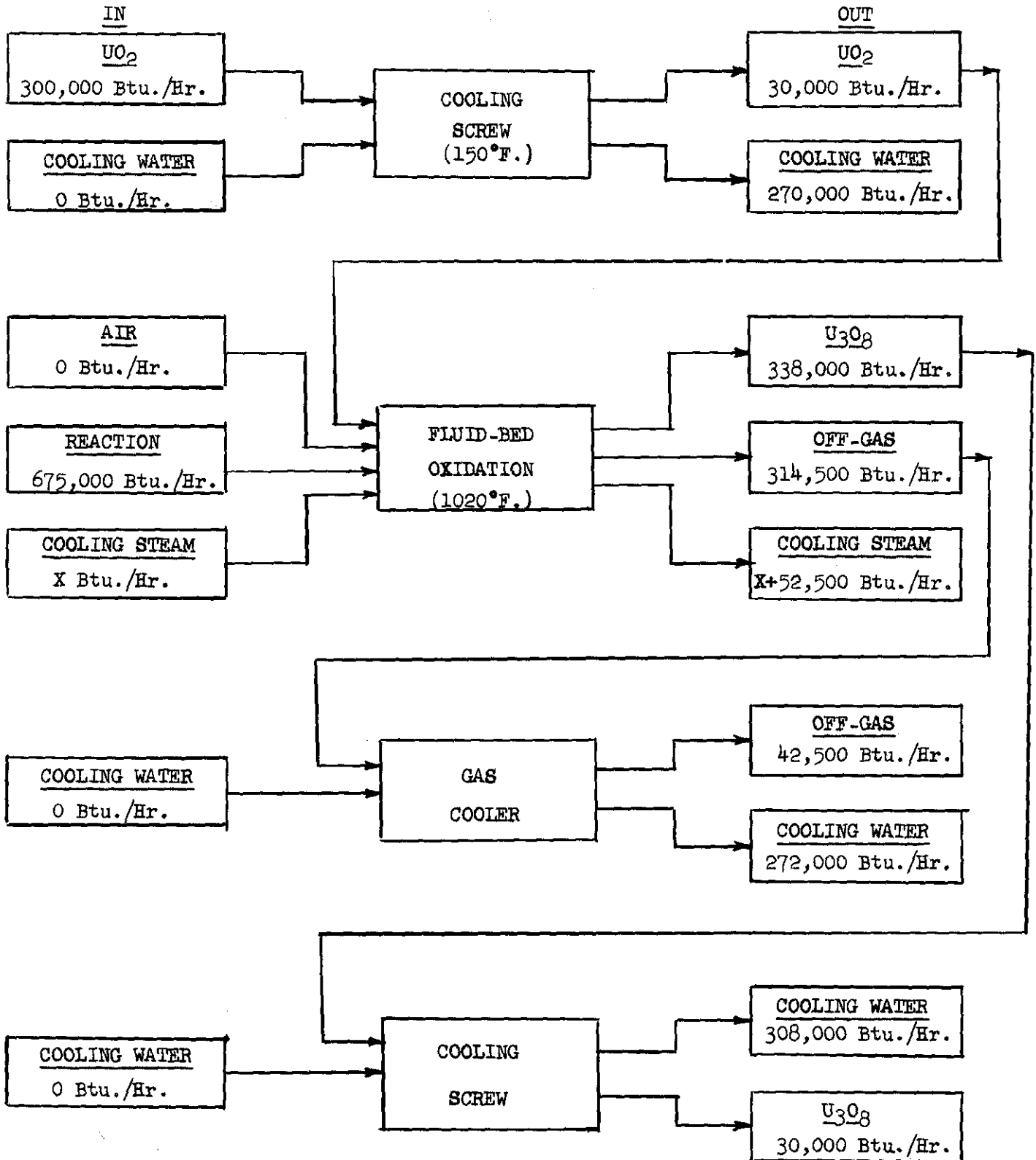


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FIGURE 4

HEAT BALANCE (OXIDATION)

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APPENDIX

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CALCULATION OF ESTIMATED SAVINGS FOR INCREASED UF₄ CONVERSION

Basic data for this calculation were obtained from TID-5295, "Current Commission Methods for Producing UO₃, UF₄ and UF₆" (January 1956).

Anhydrous HF Unit Cost:

(From TID-5295, P. 271)

HF portion of F₂ cost is \$0.23/lb. F₂ produced.

Assuming 5% loss,

$$19/20 \times 0.23/0.95 = \$0.23/\text{lb. HF.}$$

F₂ Unit Cost:

(From TID-5295, P. 271)

F₂ costs \$0.43/lb.

Anhydrous HF Cost to the Process:

(From TID-5295, P. 182)

45% of HF is converted to UF₄.

2% of HF is lost to scrubbers.

53% of HF is converted to 70% aqueous HF.

Basis: 1 lb. HF consumed in reaction.

$$\begin{aligned} 1/0.45 &= 2.22 \text{ lbs. HF feed required/lb. theoretical cost of} \\ &\text{HF} = 2.22 \times 0.23 = \$0.511 \end{aligned}$$

Assuming 70% HF resold at \$0.17/lb:

$$\text{Receipts from 70\% HF sale} = 2.22 \times 0.53 \times 0.17 = \underline{0.200}$$

$$\text{Net Cost of HF} = \$0.311/\text{lb. HF theoretical}$$

F₂ Cost to the Process:

(From TID-5295, P. 271)

F₂ portion of UF₆ cost is \$0.070/lb. UF₆.

F₂ costs \$0.43.

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Appendix, continued

0.070/0.43 = 0.163 lb. F₂ consumed/lb. UF₆ produced.

Theoretical F₂ consumption estimated at 0.121 lb. F₂/lb. UF₆.

0.163/0.121 = 1.35 lb. F₂ required/lb. theoretical.

1.35 x 0.43 = \$0.58/lb. F₂ theoretical.

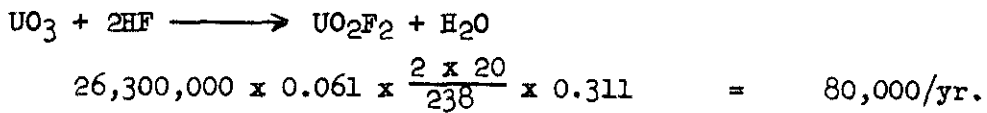
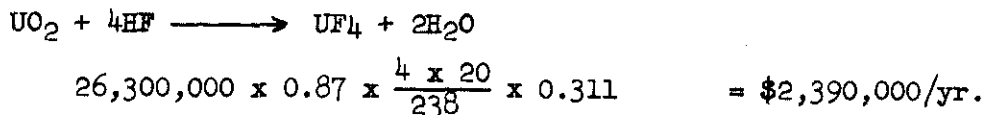
EXAMPLE

Case II Essential Materials Cost:

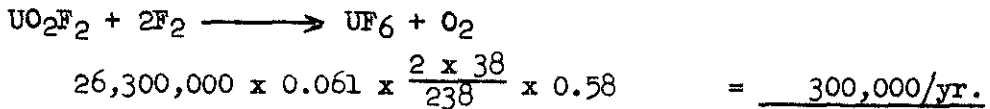
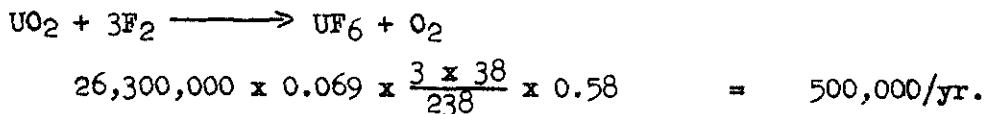
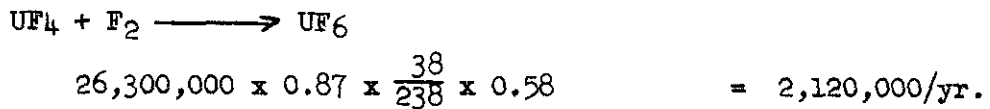
88% UF₄, 6% UO₂, and 6% UO₂F₂ in hydrofluorination product or 87% of the U as UF₄, 6.9% of the U as UO₂, and 6.1% of the U as UO₂F₂.

36 x 365 x 2000 = 26,300,000 lb. U/yr.

HF Cost:



F₂ Cost:



TOTAL \$5,390,000/yr.

DECLASSIFIED