UNCLASSIFIED

COVER SE

X-426 (Rev. 8-55)

OAK RIDGE NATIONAL LABORATORY

Operated By
UNION CARBIDE NUCLEAR COMPANY

UCC

POST OFFICE BOX P
OAK RIDGE, TENNESSEE

ORNL
CENTRAL FILES NUMBER

55-9-51

DATE: September 14, 1955
SUBJECT: Preliminary Economic Evaluation
of the Fluorox Process For Manu-
facturing UF₄ and UF₆
TO: F. L. Culler
FROM: J. E. Moore

Photostat Charge \$ 2.72 for
Access Permittees

Available from
Technical Information Service Extension
P. O. Box 1001, Oak Ridge, Tennessee

~~RESTRICTED DATA~~

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

~~This document is a Secret-Re-
stricted Data release to civilian
employment of the energy.~~

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.

Classification cancelled (or changed to UNCLASSIFIED)
by authority of *James S. Galt (Chief Branch, AEC)*
date *April 28, 1960*
TIT, date *5-13-60*

950-0011

2185

UNCLASSIFIED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

An interim technical evaluation showing the feasibility of the Fluorox Process for manufacturing UF_4 and UF_6 in moving-bed reactors has been presented in an earlier paper¹. The development program has progressed far enough to provide firm plant design criteria for the pellet feed preparation facility and the reduction-hydrofluorination (UF_4) reactor. Additional tests are required to provide firm design criteria for the oxidation (UF_6) reactor.

The present report presents a process flow diagram, descriptions of the process equipment, and a preliminary economic evaluation of the Fluorox Process. The Fluorox Process promises substantial savings not only in new plants but also in operating and maintenance costs if installed in existing feed manufacturing plants. Therefore, for the purpose of this estimate, it was assumed that the Fluorox Process replaces existing equipment in plants that process 10, 20 and 40 tons of uranium per day and that the cost of replacement is paid for from the resultant savings. These potential cost savings are attributed to (1) increased HF utilization efficiency, (2) reduction in number and complexity of reduction-hydrofluorination units, (3) reduction in number of fluorination reactors, and (4) elimination of large-scale production of higher-cost fluorine.

PROCESS EQUIPMENT

A flow diagram of the Fluorox Process is presented in Figure 1. The major equipment needed for materials handling, pellet feed preparation, and UF_4 and UF_6 production is shown. Necessary items that are not shown are assumed to be already available, including facilities for supplying HF gas, for filtering and scrubbing the waste gas, for filtering and cold-trapping the UF_6 product gas, and for treating the small blow-down stream of UO_2F_2 or UF_4 ². The description, function, and estimated purchase price of each unit shown in Figure 1 follows:

- (1) Hopper-Feeder Assemblies. Four hoppers and feeders store and proportion the incoming UO_3 and starch and the recycled UO_2F_2 and fines. Another hopper stores a reserve supply of pellets. Each hopper has a 12-hr storage capacity. Standard commercial feeders are used. The combined cost of these units is estimated to be:

10 tons of uranium/day plant - \$10,000

20 tons of uranium/day plant - 13,000

40 tons of uranium/day plant - 16,000

1) See CF-55-5-176 (ORNL-1985)

2) An existing fluorine clean-up reactor could be used to convert the blow-down stream to UF_6 by utilizing waste fluorine from the diffusion plant.

DECLASSIFIED

- (2) Dry Mixer. A continuous mixer blends the four streams of dry solids prior to the addition of water. Several suitable types are commercially available. A ribbon (double-helical) mixer, estimated to cost \$3,000, is assumed for each plant size.
- (3) Wet Mixer-Extruder. A continuous mixer-extruder combines the blended solids and water and extrudes the resultant paste onto a belt drier. An automatically-controlled hammermill, developed at ORNL, has proven satisfactory for this purpose on a variety of feed materials. The small ORNL unit, which costs \$3,850, has a minimum capacity of 1/2 ton of material/hr. The maximum capacity has not been determined, but the unit has been operated for brief periods at an intermediate rate of 3 tons/hr. The following costs are allowed in the estimate for this unit:

10 tons of uranium/day plant - \$5,000
20 tons of uranium/day plant - 7,000
40 tons of uranium/day plant - 10,000

- (4) Belt Drier. The extruded material is spread onto a conveyor belt, where it is allowed to harden during the rapid hydration reactions, which are initiated in the mixer-extruder. Although the exothermic hydration reactions provide sufficient heat to drive off the excess water, steam heat is applied as required to accelerate the reaction. A retention time of two or three minutes on this belt is adequate for the material to "set" and loose its stickiness. The following are estimated for this unit:

<u>Plant Size</u>	<u>Belt Size</u>	<u>Cost</u>
10 tons of uranium/day	18" x 12'	\$10,000
20 tons of uranium/day	2' x 18'	15,000
40 tons of uranium/day	3' x 24'	20,000

- (5) Scrubber-Ejector. The housings enclosing the belt conveyors are kept under negative pressure by an ejector-scrubber which removes water vapor and entrained dust from the exhaust gases. The incoming water, condensate, and collected dust are automatically-metered to the mixer-extruder. The estimated cost of this unit for each plant is \$3,000.
- (6) Curing Belt. The hot material from the drying belt is retained on a curing belt for 15 to 20 minutes, during which time the hydration reactions are completed. The excess water evaporates, and the material becomes hard enough for the subsequent milling operations. This belt is not heated, but is totally enclosed and exhausted. The following are estimated for this unit:

<u>Plant Size</u>	<u>Belt Size</u>	<u>Cost</u>
10 tons of uranium/day	18" x 12'	\$3,000
20 tons of uranium/day	2' x 18'	5,000
40 tons of uranium/day	3' x 24'	7,000

- (7) Granulator. The hardened material is crushed and granulated in a combination roll crusher and hammermill, similar to the equipment in satisfactory use for this purpose at the FMPC. The cost of this small, commercially-available equipment is estimated at \$6,000 for each plant.
- (8) Elevators. Vertical conveyors are used in the process to minimize building height. One is used to elevate the granulated feed material and one to recycle the UO_2F_2 intermediate. The combined cost of these elevators is estimated to be \$10,000 for each plant.
- (9) Screen. The fines in the granulated feed, estimated to be 10-20% of the total, are removed by a standard, commercial screen, which is estimated to cost:

10 tons of uranium/day plant	- \$1,000
20 tons of uranium/day plant	- 2,000
40 tons of uranium/day plant	- 3,000

- (10) Hydrofluorination Reactor. The UO_3 - UO_2F_2 -starch pellets are converted to UF_4 by reaction with HF gas in a moving-bed shaft reactor (see Figure II). This reactor consists of an insulated, electrically-traced shell, gas distribution tubes, and a perforated solids distribution plate. In moving-bed tests, UO_3 -starch pellets have been continuously converted to 99% UF_4 , using 10% excess HF, in a 4-in.-dia. x 5-ft-long reactor at a rate of 50 lbs of uranium/hr/cu. ft of reactor volume¹. For the purpose of this estimate, it is assumed that the production rate is only 20 lbs of uranium/hr/cu. ft. The reactor size and costs are estimated:

<u>Plant Size</u>	<u>UF₄ Required</u>	<u>Reaction Zone Size</u>		<u>Cost</u>
		<u>Volume</u>	<u>Dimensions</u>	
10 tons U/day	20 tons U/day	85 cu. ft	2.5'-dia.x17' high	\$12,000
20 tons U/day	40 tons U/day	170 cu. ft	3.5'-dia.x18' high	17,000
40 tons U/day	80 tons U/day	340 cu. ft	4.5'-dia.x21' high	21,000

- (11) HF Gas Heater. Approximately 50% of the HF gas for the hydrofluorinator is preheated in a gas-fired or electrically-heated Inconel heat exchanger. The cost of this unit is estimated:

1) See CF-55-5-176.

FIGURE II
MOVING-BED REACTOR FOR
 $\text{UO}_3 \rightarrow \text{UF}_4$

MATERIAL ----- INCONEL
SIZE ----- 4.5' DIA. X 21' HIGH
CAPACITY ----- 80 TU/D (UF_4)

10 tons of uranium/day plant - \$4,000
20 tons of uranium/day plant - 6,000
40 tons of uranium/day plant - 8,000

- (12) Oxidation Reactor. The UF_4 pellets are converted to UF_6 gas and UO_2F_2 pellets with oxygen or air in a second shaft reactor (see Figure III). This reactor consists of an Inconel shell, a lined reaction zone, and a cooling zone. The liner, consisting of high-fired U_3O_8 or UO_2F_2 bricks, serves as a refractory and insulator and protects the Inconel shell from excessive temperatures. Preliminary studies indicate that such a liner will be feasible. Internal coolant tubes are provided for cooling the products. Laboratory studies¹ indicate that the reaction between UF_4 and O_2 is complete in 2-5 minutes at 900°C . Tests in a 1-in.-dia. moving-bed reactor² showed promise that a larger moving-bed reactor should be feasible. Larger-scale studies are required to obtain complete design criteria for this reactor. It is probably conservative to assume, however, that the cost of the oxidation reactor is the same as the cost of hydrofluorination reactor, excluding the value of the uranium content of the lining.
- (13) Solids Meters. Two rotary-vane meters regulate the flow of solids from the reactors. The bulk of the UO_2F_2 intermediate is withdrawn and recycled to the pelleting operation, and a small (approximately 1%) blow-down stream is removed for treatment to remove impurities from the recycle stream. The solids meters are estimated at \$2,000 each, or \$4,000 total for each plant.
- (14) Pulverizer. The UO_2F_2 pellets are pulverized in a hammermill, similar to the ones that are presently used for pulverizing UO_3 , before being recycled to the pelleting equipment. The cost of this unit is estimated to be \$4,000 for each plant.

ESTIMATED INSTALLATION COST

The estimated costs of replacing existing equipment with Fluorox units are tabulated (see Table I) for the three plant sizes (10, 20 and 40 tons of uranium/day). It is assumed that initially only one set of Fluorox equipment is installed and that the unused existing equipment would be kept in standby operating condition. The installed cost of the equipment is estimated to be 140% of its purchase price. Because of the simplicity of the equipment, the auxiliary services are estimated to be fairly low: piping, 30%; instrumentation, 10%; and electrical, 15% of the equipment installed cost. Allowances are included for minor modification of the existing equipment that is utilized, such as heat exchangers, filters, scrubbers, and

-
- 1) Mandleberg, C. J. and Davies, D., "A Study of the Reaction Between UF_4 and O_2 ", AERE-CR-863, February 13, 1952.
2) See CF-55-5-176.

FIGURE III
MOVING BED
OXIDATION REACTOR

Table I

Preliminary Cost Estimate - Fluorox Process

Cost in thousands of dollars for the installation of one line of equipment in an existing plant per flow diagram, Figure I.

<u>Cost Item</u>		<u>Equipment Capacity, T U/D</u>		
		<u>10</u>	<u>20</u>	<u>40</u>
A	New Equipment			
	1. Misc. hopper-feeder assemblies	10	13	16
	2. Dry mixer	3	3	3
	3. Wet mixer-extruder	5	7	10
	4. Belt drier	10	15	20
	5. Scrubber-ejector	3	3	3
	6. Curing belt	3	5	7
	7. Granulator	6	6	6
	8. Elevators	10	10	10
	9. Screen	1	2	3
	10. Hydrofluorination reactor	12	17	21
	11. HF gas heater	4	6	8
	12. Oxidation reactor	12	17	21
	13. Solids meters	4	4	4
	14. Pulverizer	4	4	4
		<hr/>	<hr/>	<hr/>
B	Sub-total: New equipment cost	87	112	136
	Installation, 40%	35	45	54
		<hr/>	<hr/>	<hr/>
C	Sub-total: Equipment installed cost	122	157	190
	Piping and connections, 30%	37	47	57
	Instrumentation, 10%	12	16	19
	Electrical, 15%	18	24	29
		<hr/>	<hr/>	<hr/>
D	Total physical cost, new equipment	189	244	295
E	Modification, existing equipment*	30	50	90
F	Modification, existing building	30	40	50
		<hr/>	<hr/>	<hr/>
G	Total physical cost	249	334	435
H	Engineering and construction supervision, 40%	100	134	174
I	Contingency, 50%	125	167	218
		<hr/>	<hr/>	<hr/>
J	Total investment cost	474	635	827

*Existing heat exchangers, filters, scrubbers, conveyors, etc.

DECLASSIFIED

9509

conveyors: 10 tons of uranium/day plant, \$30,000; 20 tons of uranium/day plant, \$50,000; and 40 tons of uranium/day plant, \$90,000.

The equipment is compact and therefore requires very little building space. The volume required for the 40 tons of uranium/day plant is estimated to be 40' x 40' x 80', average height, or 128,000 cu. ft. Assuming that most of the required space and building services are available in the existing plant, \$50,000 is allowed for building modification. Thirty and forty thousand dollars are included, respectively, for building modifications in the 10 and 20 tons of uranium/day plants.

The costs of engineering design and construction supervision are estimated to be 40% of the total physical cost, and a contingency factor of 50% of the total physical cost is added. This high contingency is allowed in the estimate to compensate for unexpected costs in modification of the existing building and equipment, purchase of minor auxiliary items not included, and for lack of firm cost estimates on some of the items.

The total estimated investment costs for installing Fluorox equipment in the existing plants are:

10 tons of uranium/day plant - \$474,000
20 tons of uranium/day plant - 635,000
40 tons of uranium/day plant - 827,000

ECONOMIC EVALUATION

In a new plant or plant expansion for producing UF_4 or UF_6 , a perfected Fluorox Process obviously would be more economical than the present processes. In considering new processing techniques, the moving-bed reactors that are employed in the Fluorox Process should be competitive with any other known equipment, because of their inherent simplicity and high chemical utilization efficiency. In improving costs in present plants, the potential savings to be realized by replacing existing equipment with moving-bed units are of such magnitude that it should be possible to completely and quickly pay for the new equipment out of the savings in operating and maintenance costs. Indicated advantages of the Fluorox Process, leading to reduced investment, operating, and maintenance cost, are:

- (1) High Capacity: One set of moving-bed reactors has the capacity to replace as many of the present reactors as desired. Such reactors, if sized to process all of the anticipated uranium production, would be small in comparison with similar industrial units now in use. In comparison, one of the existing plants has 40 reactors for converting UO_3 to UF_4 .
- (2) Few Processing Steps. Only two reactors (plus appropriate spares) are required, one for producing UF_4 and one for UF_6 . The feed (pellet) pre-

DECLASSIFIED

950-10

paration step has been demonstrated to be easily carried out on a plant scale. Present plants for producing UF_6 have separate, complicated processing steps for reduction, hydrofluorination, fluorination, and fluorine manufacturing.

- (3) Few Auxiliary Units. No recycle of process gases is required. The number of auxiliary units such as heat exchangers, entrainment separators, filters, powder feeding mechanisms, etc., is minimized. Special gas blowers and compressors are eliminated. Extremely-low-temperature refrigeration is eliminated.
- (4) Safety. The simpler, moving-bed process, which eliminates hydrogen and fluorine manufacture and use and reduces the complexity of the HF processing equipment, should be much safer to operate and maintain than present plants.
- (5) Mechanical Simplicity. No moving parts and few metal surfaces are required to be in direct contact with the corrosive process gases. This should result in a lower maintenance load on items such as screw feeders, reactor screws, drive mechanisms, packing glands, bellows connectors, etc., which contribute greatly to maintenance costs.
- (6) High Chemical Efficiency. Initial tests in the 5-ft-high moving-bed reactor have demonstrated that no more than 10% excess HF is required in the production of 99% UF_4 . Even greater efficiency should be possible with experience in a taller, plant-scale unit.
- (7) Cheaper Raw Materials. HF, starch, and oxygen or air replace fluorine in converting UF_4 to UF_6 . The need for operating a large fluorine generating plant, containing as many as 90 fluorine generators plus appertenances, would be eliminated.

Because of the considerable reduction in the amount and complexity of the process equipment that would result if Fluorox units replaced existing equipment, the manpower and materials required in the operation and maintenance of the plant would be considerably reduced. An estimate of the amount of this potential reduction on the basis of current plant operating costs will be delayed until a later date, since less involved calculations show that the savings in chemical costs alone would more than justify installation of Fluorox equipment in the present plants.

For example, the present reactors employ at least 100% excess HF in the hydrofluorination step. This is returned to the vendor in the form of 71% aqueous HF, at a loss of 5.5¢ per lb of HF. In a 40 tons of uranium/day plant, this loss amounts to \$540,000/yr. If this entire amount were saved by installing Fluorox equipment, the savings would repay the installation costs in two years. The small HF loss in the Fluorox Process probably would be no greater than the unrecovered miscellaneous losses in the present feed and fluorine plants.

750-11
DECLASSIFIED

In the production of UF_6 , an even greater cost saving would result from the elimination of fluorine by substituting HF , starch, and oxygen or air as the reactants. The amount of these savings (see Figure IV) will vary with fluctuations in the operating costs of the fluorine plants (see Figure V). However, a comparison between the potential savings and the recent fluorine plant operating costs indicates that the savings should be great enough to amortize the Fluorox equipment in a 40 tons of uranium/day plant, for example, in about one year (see Figure VI).

RECOMMENDATIONS

Since the potential cost savings of the Fluorox Process are so great, it is recommended that the development program be increased as outlined in report CF-55-5-176. The proposed program would permit carrying out the following studies, which are required before the process can be employed to its fullest advantage:

- (1) Conduct tests of the oxidation step in a 4-in.-i.d. refractory-lined moving-bed reactor to study:
 - (a) Techniques for preventing corrosion.
 - (b) Reactor capacity and efficiency.
 - (c) Intermediate formation and dust entrainment.
- (2) Operate an integrated system, consisting of the pelleting, reduction-hydrofluorination, and oxidation steps, at a rate of about 10 lbs of UF_6 /hr for a period long enough to determine whether problems will be encountered from build-up of non-volatile impurities during continuous operation.
- (3) Continue studies of promising techniques for the production of carbon-free metal-grade UF_4 .
- (4) Investigate the direct conversion of ore concentrates to UF_6 by the Fluorox Process. Figure VII shows that there is a good possibility that impure ore concentrates could be treated in the Fluorox Process. If this becomes feasible, additional worthwhile savings could be realized.

The pelleting and moving-bed equipment on hand in the Unit Operations Section is adequate for studying each step of the process; however, an allowance of \$15,000 is made in the proposed budget¹ for integration of the equipment into a unit for demonstrating the feasibility of continuous operation with UO_2F_2 recycle.

1) See CF-55-5-176.

JEM/mcj

950-12
DECLASSIFIED

FIGURE IV
ESTIMATED ANNUAL SAVINGS IN CHEMICAL COSTS
FROM THE FLUOROX PROCESS,
VERSUS OPERATING COSTS
IN MANUFACTURING FLUORINE

FIGURE V
CHEMICAL COSTS

FIGURE VI
TIME REQUIRED FOR AMORTIZATION
OF THE FLUOROX PROCESS IN EXISTING PLANTS
FROM OPERATING COSTS FOR
ELIMINATED FLUORINE PLANTS

FIGURE VII
CALCULATED EFFECT OF URANIUM
FEED ASSAY ON URANIUM TO BE
REPROCESSED IN BLOW-DOWN EQUIPMENT