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TO: L. W. Lang

FROM: A. M. Platt and R. J. Sloat

FEASIBILITY OF PROCESSING IRRADIATED THORIA

INTRODUCTION

In compliance with your request, Chemical Development has investigated the technical feasibility and the economic impact on the separations process of using thorium oxide in fuel elements in lieu of thorium for the production of U^{233}. This memorandum summarizes the implications of such a substitution on the separations process to be employed for processing the irradiated fuel elements.

In addition to the thorium metal (Q) slug, two types of fuel elements have been considered. The first is composed of a sintered thoria core encased in a conventional aluminum alloy jacket. The second uses a heterogeneous mixture of "J" alloy (aluminum - U^{235}) and thoria pellets and is also jacketed with an aluminum alloy.

SUMMARY AND CONCLUSIONS

Since very little experimental data are available on the dissolution of thorium oxide, these conclusions are only tentative. Actual experiments which will be made in the next few months should provide a better basis for "firm" extimates.

1) Based upon an assumed thorium oxide dissolution rate approximately 10-fold slower than that of thorium metal in HNO_3-aluminum-complexed fluoride ion solution, it appears that a 2 ton per day thorium processing capacity could be attained by building a third dissolver cell for the thoria plant, one more than required for a plant processing thorium metal slugs. Each of the three thoria dissolvers would be considerably larger than the dissolvers required in a thorium metal plant (20-ton total charge (slugs plus heel) instead of 8 tons). It is estimated that the additional dissolver cell and the larger dissolver vessels required for thoria dissolution would cost $1,000,000 to $1,500,000 more than the dissolver facilities for a thorium metal plant operating at the same thorium processing rate.

2) If the "J" alloy-thoria pellet slug is adopted as the fuel element for a Thorex Plant instead of a thorium metal or thoria slug, it would be economically feasible to build a "25" processing plant at Hanford either as a part of, or adjacent to, the Thorex Plant instead of transporting the dissolver solution produced from "J" metal to another site for "25" processing. Estimates developed in the discussion below indicate that the $10,000,000 to $15,000,000 annual cost of shipping the dissolver solution would pay for a $40,000,000 "25" facility in approximately three years. In addition, the hazard of shipping large volumes of "hot" solution would be eliminated.

   The above shipping cost could be appreciably reduced if the "J" alloy-ThO_2 pellet slugs were transported off-site for "J" metal dissolution. The ThO_2 pellets could then be shipped back to Hanford for Thorex processing. This approach is not considered attractive since the Thorex Plant would be dependent upon other sites for its feed stock. Strikes either at the other site or in the transport facility could seriously affect Hanford thorium production.

3) The dissolver facilities required for handling the "J" metal-ThO_2 pellet fuel elements would be more complicated and costly than the combined dissolver facilities required for separate dissolution of "J" slugs and thoria slugs. Estimates developed in the discussion below indicate that approximately 30 batch dissolvers would be required to attain a 2 ton/day thorium rate when processing "J" metal-ThO_2 pellet slugs. These dissolvers would require an estimated additional expenditure of $3,000,000 to $4,000,000 over the expenditure required for separate "J" metal slug and ThO_2 slug dissolution facilities. Operating costs for the "J" metal-ThO_2 pellet dissolution would be higher than for

separate dissolution of "J" metal slugs and ThO_2 slugs. It is estimated that six additional men (1 crane operator, 3 dissolver operators, and 2 laboratory people) would be required at an annual cost of $120,000 (4-shift coverage).

4) If the decision were made to build a "25" Plant at Hanford, it would be cheaper to build the plant as a part of the Thorex Building. If such a dual-purpose Thorex-"25" Plant were built, manpower and material could be utilized more efficiently during the construction program resulting in lower capital investment costs than required for separate "25" and Thorex Plants. Similarly, when the plant is operated, overhead costs would probably be reduced (e.g., I.M.E., Plant Protection; General Administration). Operating cost data presented on Table 2 indicte savings of approximately $1,000,000 a year by operating a "25"-Thorex Plant in place of separate "25" and Thorex Plants.

DISCUSSION

Thoria Core Slugs

Processing of thoria-core fuel elements through separations would differ from a conventional Thorex operation using an aluminum-jacketed metallic thorium core for fuel only in the dissolution procedure employed to produce a liquid feed solution from the fuel elements. (However, any radical departure from the nitric acid-trace fluoride dissolution procedure discussed below could affect subsequent processing steps.) Once the thoria is dissolved in nitric acid, the solution may be processed in a manner identical with that used in processing conventional thorium (Q) slugs.

The meager data available indicate that thoria and aluminum mixtures can probably be dissolved in 60% HNO_3 spiked with mercuric and fluoride ion (0.05 to 0.1 M) to catalyze the acid-aluminum and acid-thoria reactions, respectively. These data[1] (on very small samples of 0.5 to 2 grams) indicate that the dissolution rates of thoria (sintering temperature unknown) in fluoride ion-60% HNO_3 solution are the same order of magnitude ($\pm$ 3 fold) as for dissolving uranium metal in 60% HNO_3. However, more recent data[2] show that the rate of dissolution is markedly influenced by the sintering temperature of the thoria. Relative dissolution rates of thorium oxide which has been sintered at temperatures of 700, 1200, and 1630°C. are 1, 0.55, and 0.3 respectively. Although the central core temperature attained during pile irradiation of thoria slugs has not been experimentally verified, it has been postulated to be in the region of 500 to

(1) CC 3[illegible]
(2) ORNL-1431

700°C. It is not known whether dissolution rates of irradiated thoria slugs exposed to such core temperatures would be comparable to the dissolution rates of thoria slugs exposed to such temperatures in sintering operations prior to pile exposure. Also unknown, are any deleterious effects which might result from irradiation of thoria. However, O.R.N.L. experience reveals that a very insoluble, "blue" thorium is formed during the irradiation of thorium metal. This material has been tentatively identified as a thorium oxide and is thought to be soluble in large excesses of acid-fluoride mixtures.

In summary, it might be said that although irradiated thoria dissolution problems do not appear to be insurmountable, they do appear formidable. It is estimated that 15 to 30 man months of research and development work would be necessary to establish conceptual design criteria for the dissolution step in a new separations facility.

The attached Table I summarizes and compares the operating characteristics of uranium, thorium, and thoria dissolvers.(*) These data indicate that one dissolver (a standby spare would also probably be required) would be capable of handling a uranium production rate of 100 tons/month or a thorium production rate of 50 tons/month. Two dissolvers (plus a standby) would be necessary for a thoria production rate of 50 tons/month.

From the above discussion, it appears that an additional dissolver cell would be required if thorium oxide were processed at a 50 ton/month rate instead of thorium. Based upon an estimated canyon cost of $30,000 per foot and the canyon space (approx. 35 ft.) required for a Purex dissolver cell, it appears that an additional capital investment of approximately $1,000,000 would be required to provide the additional dissolver cell required for thorium oxide processing. Each of the dissolvers used for thorium oxide processing would be larger than the dissolvers used in a plant processing thorium metal. However, the additional cost for larger dissolvers would be expected to be relatively small (less than $100,000) in comparison with the million dollar extra cost of the dissolver cell and the equipment in it.

"J" Alloy-Thoria Pellet Slugs

The use of "J" alloy-thoria pellet slugs in the piles would definitely affect the design of a separations plant to process these slugs. In addition, there would be many far reaching effects such as the involvement of "25" separations facilities at other A.E.C. sites. During the processing steps

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(*) Concentrated HNO_3 is assumed used in the uranium dissolver while concentrated HNO_3 and a low fluoride ion concentration are used for the dissolution of thorium and thorium oxide.

which would be required to separate the thorium oxide pellets and U^{233} from the aluminum and U^{235}, a dissolver solution is formed which would be suitable for processing in a "25" plant. This solution could either be processed in a new facility built at Hanford or shipped as dissolver solution to other A.E.C. sites (presumably Arco) for processing.

Shipping problems for this dissolver solution would probably be considerably more complicated than those encountered in shipping slugs. For example, in case of a rail or truck accident, rupture of tanks containing this dissolver solution would be likely to spread gross contamination over a considerable area. In a congested area, such an accident would have serious and expensive consequences.

The cost of shipping the "25" dissolver solution to Arco would be extremely high. Calculations indicate that approximately 70 to 100 shielded "cask type" cars costing approximately $100,000 each and weighing 120,000 lb. each would be required for shipping the dissolver solution resulting from the dissolution of 36 kg. of U^{235} per day. A required capital investment of $7,000,000 to $10,000,000 is estimated for these cars using the following bases.

1) Each car carries 3,000 lb. of dissolver solution in six 9-inch diameter tanks, each 20 ft. long.

2) Each car weighs approximately 12,000 lb. Most of this weight is contributed by the lead shielding required.

3) A one-week round trip is assumed for each car. Ten cars a day would be required to handle the 30,000 lb./day of "25" dissolver solution produced when processing thorium at a 2 ton/day rate.

Order-of-magnitude estimates indicate that shipping charges (amortization of tank cars not included) would be approximately $1,000,000 per month or $.03 per pound of material (lead and solution) transferred to Arco.

From the above discussion, it appears that the cost of shipping "25" dissolver solution to Arco would be $10,000,000 to $15,000,000 per year. Assuming a capital investment of $40,000,000 is required for a new "25" plant here at Hanford, it appears that the plant could be amortized in a 3 year period. Building this plant in conjunction with the Thorex plant it may be possible to better the $40,000,000 figure.

Operating costs for the combined Thorex-"25" Plant would be less than the sum of the operating costs of two separate plants. Table 2 lists estimated operating costs for separate Thorex and "25" plants and similar costs for the combined plant. As noted on Table 2, annual operating cost savings of greater than $1,000,000 may be achieved in the combined Thorex-"25" plant. The major portion of this reduction occurs because of the more efficient utilization of manpower.

"J" Alloy-Thoria Pellet Dissolution Methods

No "J" alloy-thoria pellet slugs have been fabricated either for piles or laboratory testing. Consequently, the dissolution methods developed below are bases upon fragmentary data and are subject to considerable modification as research and development studies are made in the near future. In developing the dissolution methods discussed below, the following assumptions have been made:

1) The composition of the "J" alloy-thoria pellet fuel element has not been specified. The slug is assumed to be a combination of irradiated "J" and thoria slugs having the following average composition:

Compound	Weight %
U^{235}	1.25
U^{233}	0.16
U^{238}	0.10
Fission Products	0.25
ThO_2	73.75
Al	24.49

Processing rates of 2.33 ton/day and 36 kg./day have been assumed for thorium oxide and U^{235}, respectively.

2) Some cross-contamination of the U^{233} and U^{235} is inevitable, depending upon the relative dissolution rates in nitric acid of "J" alloy and thorium oxide (which contains the Pa-233 and U-233). Studies are planned to determine the cross-contamination experimentally. For present purposes, it is assumed that less than 1% of the U^{233} formed will be found in the U^{235} product. Since thorium oxide is very insoluble in nitric acid alone, probably only the Pa^{233} and U^{233} molecules on the surface of the pellets will dissolve along with the J-metal. Calculations indicate that 1% of the volume of a sphere is contained in an outside shell with a thickness 0.33% of the sphere diameter.

3) It is assumed that the Thorex and "25" processing steps subsequent to the dissolution operation would be unaffected by the dissolution method employed.

Batch Dissolution. Batch dissolution, that is, HNO_3 dissolution of the "J" alloy followed by aluminum-complexed fluoride-HNO_3 dissolution of the thorium oxide in the same dissolver, would probably be the most costly method of dissolution both from a capital investment and operating cost standpoint. However, the research, development, and design effort required for this type dissolution would be considerably less than for other schemes discussed later, and hence initial design on the basis of batch dissolution may be dictated by the timing involved.

The [illegible]mum dissolution rates of the "J" alloy and thoria pellets will probably [illegible]fer by a factor of at least ten. Therefore, in a batch-type dissolver the criticality considerations of the "J" alloy will establish the volume of each dissolver and the slow thoria dissolution rate will determine the number of dissolvers required.

Critical mass data have not been obtained for the "J" alloy-pellet slugs. In calculating the number of dissolvers required, it has been assumed that a 9-in.-diameter dissolver would be safe from a slow neutron reaction (safe geometry*) standpoint when processing these slugs. If the dissolver is made 20 ft. long, 1300 lb. of slugs may be charged to each dissolver. Assuming a 5-day dissolver cycle, approximately 30 dissolvers are required for a thorium processing rate of 2 tons/day. Assuming that 8 ft. of canyon length (connector limiting) would be required for each of these dissolvers, its associated receiver-samplers and off-gas handling equipment, approximately 240 ft. of canyon would be required to house the batch dissolvers.

Comparison of this 240 ft. of canyon length with the sum of the 100 ft. of canyon length required for dissolving ThO_2 slugs in a Thorex Plant and the 30 ft. of canyon length required for dissolving the "25" slugs in a "25" Plant, shows approximately 110 to 130 ft. of additional canyon length required for the batch dissolvers in the dual-purpose "J" alloy-thoria pellet plant. The additional 110 to 130 ft. of canyon would necessitate an additional investment of approximately 3 to 4 million dollars.

This 3 to 4 million dollars additonal construction cost would probably be saved by building a "25" plant at the same time the Thorex Plant is built. By combining the construction program of the $50,000,000 Thorex Plant and the $40,000,000 "25" Plant, it should be possible to reduce the total cost $4,000,000; i.e., enough to pay for the additional dissolver facilities required for thorium-oxide pellet-"J" alloy dissolution. The amortization charge shown on Table 2 is based upon a dual-purpose "25" Thoria Pellet Plant cost of $90,000,000.

Alternate Dissolution Techniques

As is indicated in the discussion above, approximately 30 batch dissolvers contained in a 240 ft. length of canyon space are required for processing thoria pellet-"J" alloy slugs at a 2 ton/day thorium processing rate. Appreciable savings ($30,000/ft. of canyon) could be realized by a reduction in the space used for dissolution. A few of the schemes considered are discussed briefly below.

(*)Oak Ridge is currently designing a 9" diameter "25" dissolver. It is considered "safe" because of the dilution of the "25" with aluminum in the slugs and in the resulting solution.

Remote Dumper. The "J" metal is relatively easily dissolved away from the pellets with mercury catalyst and HNO_3. Instead of dissolving the slow-dissolving thorium oxide in the same unit in HNO_3-fluoride over a 5-day period, the thoria pellets might be dumped from the "J" dissolver by the crane into a large thoria pellet dissolver. Before the pellets could be dumped, an accountability check on the "25" would have to be made to prevent discharge of "J" metal into the thoria dissolver. If "J" metal were inadvertently dumped in the thoria dissolver, criticality problems could arise and gross cross-contamination could result.

Continuous or Semi-Continuous Mechanical Dissolver. With this type dissolver, undissolved slugs would be fed to the dissolver on a conveyor belt. After dissolution of the "J" metal, the thoria pellets would be carried from the dissolver by conveyor to a second dissolver. Although designed critically safe, this dissolver would be considerably larger than the "J" metal dissolvers.

Jet Pellet Removal Dissolver. The "J" metal would be dissolved in a critically safe dissolver. Both the "25" dissolver solution and the pellets would be jetted to a centrifuge, from which the "25" solution would be routed to the "25" Plant and the Pellets would be removed to a thoria dissolver.

TABLE I

DISSOLVER SCHEDULE

Operation	Purex Dissolver One Dissolver Operating at Total Processing Capacity of 4.16 Tons U/Day — Material Processed	Time, Hr.	Thorex Dissolver One Dissolver Operating at Total Processing Capacity of 2.05 Tons Th/Day — Material Processed	Time, Hr.	Thorex Dissolver Two Dissolvers Operating at Total Processing Capacity of 2.05 tons Th/Day as ThO_2 Slugs — Material Processed	Time, Hr.
Slug Charging			4.10 (9 buckets)	4.5	10.25 (27 buckets)	13.5
Equilibrium Heel			4.10		10.25	
Jacket Removal and Rinsing				0		0
First Cut			2.05	14	10.25	240
Second Cut			2.05	18		
Slack Time				11.5		10.5
Total Time				48		264
Avg. Dissolution Rate			1.8 %/hr.		0.21 %/hr.	

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TABLE II

ESTIMATED ANNUAL SEPARATIONS COSTS, THOUSANDS OF DOLLARS

Bases: 600 tons Th/yr.; 10,800 Kg. U^{235}/yr.; 1,350 Kg. U^{233}/yr.

	Facility			
	Thorex Plants (1)			
	Thorium Metal Fuel Element	Thorium Oxide Fuel Element	"25" Plant (2) "J" Slugs	Dual-Purpose "25"-Thorex Plant "J" Metal-ThO_2 Pelle Fuel Element
Material, Direct				
Water Storage				
Labor, Direct				
Labor, Indirect				
Maintenance				
Rent, Light, Heat				
Process Steam				
Water, Steam, Air, Sewage				
Radiation Monitoring				
Plant Eng. Service				
Process Sub-Section				
Process				
Analytical				
Process Assistance				
Operating Unit, General				
Manufacturing, General				
Freight, HWRR				
Electricity				
Laundry				
Area Bus Service				
Other IME				
Plant Personnel & Protect				
Direct				
Indirect				
General Administration				
Total Operating Cost				
Amortization (16-2/3%/yr.)				
Total Operating and Amortization Cost				
Unit Cost, $/ton Th				
$/gm. "23"				
$/gm. "25"				

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NOTES

1) Direct material, waste storage, and process steam costs have been taken from the Tentative No. 6 Thorex Flowsheet shown in HW-31131. Labor and indirect costs are for an assumed plant force of 150 people and are therefore quite similar to Redox and Purex costs.

2) Direct material, waste storage, and process steam costs have been calculated from data presented in Reactor Science and Technology, December 1953 (TID-2011). Labor and indirect costs are estimated on the basis of Hanford experience for [redacted] a plant force of 80 people.