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E-METAL DISSOLVERS - REDOX

I. Introduction

In December, 1957, the proposal for Project CG-772, "E-Metal Dissolvers - Redox" was returned for reconsideration in order that the units might be designed to accommodate higher enrichment fuels, if practical. Alternate design approaches have been under consideration since that time. It is the purpose of this document to summarize the parameters involved and to present the recommendation developed from the joint thinking of Hanford Laboratories, Research and Engineering, Facilities Engineering, and Redox Operation.

II. Recommendation

A dissolver vessel designed concentrically as illustrated below (plan view) is believed to incorporate best the flexibility required to accommodate future processing requirements.

*Metal crib to have detachable lid and unencumbered access to a large fraction of the crib.

Such a vessel will be critically safe by geometry for up to 0.94 per cent U_{235} and be capable of dissolving about 6 tons of uranium per day. For an enrichment of 1.25 per cent U_{235} dissolving capacity is estimated to be 1.2 tons uranium per day if batch control is used. However, if batch control is augmented by the addition of one gram boron per liter of solution, the rate could be increased to about 2.0 tons per day. The vessel would also be capable of processing long (up to 10 feet) fuel elements originating from an NPR, the PRTR, or other power reactors.

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BY *SW Gordon* DATE 8/6/81

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It has been recommended, therefore, that two such dissolvers be installed in the Redox Plant. With these units, fuels containing up to 0.94 per cent U_{235} could be processed at rates up to 200 tons per month, thereby retaining a large fraction of the plant's operating time for other processing assignments.

III. Production Programs

The following information concerning the proposed programs for HAPD reactors was obtained from Reactor Planning Operation and Reactor studies personnel.

A. E-Spike and Fringe Loadings

Production plans entailing the use of E-metal include:

1. Radial enrichment for pile flattening. E-metal is used in lieu of "C" metal (an aluminum- U_{235} element, reprocessed at Arco).
2. Spike and radial enrichment for bolstering pile reactivity in support of the I and E program.
3. Shield protection. Some of the outermost rows of tubes are loaded with N-metal (lithium-aluminum alloy) to reduce the neutron flux as much as practical. Pile reactivity is retained by loading E-metal in tubes just inside the N-metal tubes, thereby restoring overall pile power values.
4. Fringe poison (blanket load). Similar to Item 3 except that all of the outer lattice tubes are charged with N-metal.
5. E-metal is also required in small quantities to support reactivity during miscellaneous irradiation programs, such as special irradiations for off-site customers, manufacture of plutonium containing high Pu-240 content, etc.

So long as loading patterns are limited to the current type which employs full-tube loadings of E-metal, there is little incentive to increase the U_{235} content of the E-metal above 0.94 per cent. This limitation is set by the ability to remove heat from the process tube, thereby limiting the specific rate of power generation in a given fuel element.

Currently authorized programs call for the use of E-metal at rates increasing to about 50 tons per month being charged to the reactors by fall of 1958, with 100 tons of the metal being accumulated and ready for chemical separation about July, 1958. The 50 ton per month reprocessing rate will be reached in the fall of 1959.

B. The E-N Load

An alternative production plan is currently being recommended by IPD for AEC consideration. This plan involves the loading of the piles with E and N

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elements only, with tritium and plutonium both being prime products. Assuming constant reactor time efficiency, neglecting capital investment requirements, and assuming a value of \$50 per gram for plutonium with tritium having an identical per-atom value, then a \$13,000,000 per year incentive is indicated for the full-scale adoption of the E-N loading pattern in all piles. If the U_{235} enrichment of the E-metal is increased to 1.2 per cent, an additional incentive of \$7,000,000 per year is indicated on a comparable basis. The incentive for further increases of U_{235} concentration decreases rapidly above this point.

In this production pattern, E and N elements would be loaded in the same tube, with an E to N length ratio of about 11 for the 0.94 per cent E-metal and a comparable ratio of about 5 for the 1.2 per cent fuel. I and E elements would be used, and an integrated exposure of about 1200 MWD/T would be scheduled. The economic optimum is estimated at about 1400 and 2000 MWD/T for the 0.94 per cent and 1.2 per cent fuels, respectively, but fuel element integrity cannot yet be assured for such exposures.

By making optimistic assumptions as to future AEC decisions, the following production pattern can be postulated.

Date	Production Change	Ton/Month Loaded		Impact Felt in CPD
		0.94% U_{235}	1.25% U_{235}	
Current	Fringe and Spike Loadings	50	-	100 T available 7/58 50 T/month
July '58	Demonstration 0.94% E-N Load	80-90	-	4/59
Oct. '58	N Blanket Load	130-150	-	7 to 12/59
July '59	1 to 4 Reactors Author- ized for E-N	130-225	-	3 to 9/60
Jan. '60	Demonstration 1.35% E-N Authorized	130-225	30	9/60
July '61	On 1.25% E-N Load Authorized on In- creasing Basis	225-0	30 to 300	2/62 on

During this period, it is quite probable that all three enrichment fuels will be irradiated simultaneously. In estimating total loads, one ton of E-metal (either enrichment) used in the E-N load may be considered as displacing two tons of natural uranium used in the current loading pattern.

C. New Production Reactor

The NPR study currently in progress visualizes a fuel element composed of a cluster of uranium rods (perhaps 0.85 per cent U_{235}) clad in zirconium. Based on this design, an economic incentive exists to make the individual

rods as long as is compatible with reprocessing limitations because of the complications involved in getting end seals in the rod jackets. One estimate places an NPR limit of 35 feet on the length of an individual rod. While this length is obviously incompatible with current design concept, we recognize the desirability of designing for the longest feasible rod length -- perhaps 4 to 8 feet.

D. Power Reactor Fuels

A development program is currently in progress to develop methods of reprocessing low enrichment power fuels in HAPO facilities. The Shippingport reactor is now on stream and we can expect an initial blanket discharge from this reactor as early as 1959, with subsequent deliveries at about 15 tons per year. This material is natural uranium oxide clad in zircalloy in elements having an active length of about 6 feet.

The reactor fuels currently assigned to Hanford for reprocessing are scheduled for initial discharge in 1962 and include materials having U₂₃₅ enrichments of 1.5 to 3 per cent, clad in stainless steel or zircalloy. Many other potential fuels are logically assignable to HAPO for reprocessing, and are planned to be clad in aluminum, stainless steel or zircalloy with U₂₃₅ concentrations up to 3 to 5 per cent. The "active length" of the various fuel elements is generally about 6 to 10 feet.

IV. Nuclear Safety

Existing dissolvers retain the fuel elements in a cylindrical crib during dissolution and can contain from 10 to 25 tons of uranium. This design is safe for natural uranium, but the use of enrichments above natural would require nuclear safety controls based on either operating limitations (batch control) or a new design controlling nuclear safety by geometry.⁽¹⁾ The restrictions imposed by nuclear safety become critical very rapidly as the enrichment level increases. For example, a water-tamped, slab-shaped crib (whether located on the tank bottom or as an annulus) is limited to a thickness as indicated below:

<u>U₂₃₅ Content, %</u>	<u>Calculated Minimum Critical Thickness, In.</u>	<u>Assured Safe* Thickness, In.</u>
0.71 can be made critical, but not in reasonable dissolver designs.		
1.0	12.6	10.2
1.2	9.3	7.4
1.4	7.5	5.9
1.6	6.6	5.1

*Calculated by deducting a 10% confidence factor for application of existing data, 5% for moving from minimum critical to assured safe position, and 1/2" construction tolerance to allow for warped equipment and minor crib penetration by partially dissolved fuel elements.

(1) HW-54759, "Nuclear Safety in Processing of Slightly Enriched Uranium Fuels", by N. Ketzlach, R. D. Cahoon, R. L. Stevenson, January 29, 1958.

Similarly, the critical batch size decreases rapidly as enrichment increases. For example, the minimum tons of uranium required to reach criticality under conditions of optimum H/U ratio are indicated in the table below. It should be noted that a water-tamped pile of the proposed I and E fuel elements in ordered or random array would have a near-optimum H/U ratio.

<u>U₂₃₅ Content, %</u>	<u>Spherical Configuration</u>	<u>Hemispherical Configuration</u>
0.71	50 tons	70 tons
1.0	ca. 2 tons	ca. 3 tons
1.2	ca. 1 ton	ca. 1.2 tons

A single calculation for 1.2 per cent U₂₃₅ uranium loaded into an 11 inch thick annulus indicated that a uniform height of about 2.5 feet or a hemispherical mound (as restricted by the annulus) of about 3.5 feet radius would approximate a minimum critical geometry. These conditions correspond to about 12.5 and 4.0 tons (8.0 tons if two such piles were formed on opposite side of the dissolver), respectively in a 7.5 foot O.D. vessel of an outside annulus design.

The use of a soluble poison such as a boron compound has been suggested to offset the effect of enrichment. Calculations indicate conservative minimum critical thicknesses of a slab or an annulus when the solution contains one gram per liter boron to be:

<u>U₂₃₅ Content, %</u>	<u>Thickness, In.</u>
1.2	21.5
1.4	13.5
1.6	10.9

In other words, a 10.5 inch thick annulus (assured safe by geometry for 0.94 per cent U₂₃₅) would be critically safe for enrichments up to 1.6 per cent U₂₃₅ when one gram boron per liter is in solution. The use of boron as a nuclear safety control necessitates that the required amount be present in all solutions entering the dissolver.

It is the general consensus of personnel responsible for plant safety that this approach might be used on a non-routine basis to increase a working batch size from about 45 per cent to 80 or 90 per cent of a minimum critical batch. This position is based on the premise, that when using batch control only to assure safety, assurance must be given that an accidental double batch would not create a nuclear incident, hence the working batch is chosen as 45 per cent of minimum critical. Since the use of boron compounds would prevent a nuclear incident even though double-batching accumulated more than a minimum critical batch, a working limit of 80 to 90 per cent of minimum critical might be used. A larger batch size is discouraged since the inadvertent absence of boron coincidental with double-batching might permit a nuclear incident. The double-error philosophy is therefore retained in this operating concept. Many process and operational unknowns would need to be defined before the use of boron could be adopted

as a standard plant practice although preliminary data indicate that a reasonable solution can be found.

V. Dissolution Rates

A. Standard Dissolution Procedure

Dissolution experience in the Redox plant during 1957 was reviewed by Redox Technology Operation personnel to provide bases for the estimation of dissolving rates obtainable. The empirical bases established to be consistent with plant performance were:

1. Charging Time

Open and Close Cell - 20 minutes.
Open and Close Cask Car - 25 minutes.
Switch Cars - 20 minutes.
Charge Bucket - 8 minutes.

2. Coating Removal

Time, hrs. = $3.43 + 0.42 T$
T = tons charged

3. Dissolving

First Cut: Time, hrs. = $2.5 + \frac{A}{580 + 55 T}$
A = Pounds dissolved, per cut
T = Average weight of heel, tons

Second Cut: Time, hrs. = $3.2 + \frac{A}{580 + 55 T}$

Utilizing the above bases, dissolution rates for various dissolver capacities have been calculated to be:

Dissolver Capacity, Tons	Charge, Tons	Heel, Tons	Total Time, Hrs.	Inst. Rate per Dissolver, Tcn/Day
8	6	2	28.1	5.1
9	6	3	27.3	5.3
10	9	1	38.2	5.7
12	9	3	34.7	6.2
14	12	2	45.5	6.3
16	12	4	43.1	6.7

B. Heel Removal Each Cycle

The use of batch limitations for criticality control in dissolvers would necessitate that metal heels be removed from the vessel at the end of

each cycle. The following bases were used to estimate dissolver capacity for small charges with complete heel removal.

1. Charging Time - As in A above.

2. Coating Removal

$$\begin{aligned} \text{Time, hrs.} &= 5 + 0.4 T \\ T &= \text{Tons charged} \end{aligned}$$

3. Dissolving

$$\begin{aligned} \text{Time, Hrs.} &= 3.2 + \frac{A}{200 T} \\ A &= \text{Pounds dissolved, per cut} \\ T &= \text{Average weight of heel, tons} \end{aligned}$$

The following table presents the estimated dissolution rate as a function of charge size.

Charge, Tons	Inst. Rate per Dissolver, Ton/Day
0.5	0.4
1.0	0.8
2.0	1.2
4.0	2.0
6.0	2.9

It should also be noted that increased operating costs (chemical and waste storage) accompany the decision to remove the heel each dissolver cycle. The penalty ranges between \$50 and \$100 per ton and increases as the batch size decreases.

VI. Alternative Approaches

The approaches to the E-metal dissolution problem may be categorized as follows:

1. Utilize present dissolvers.
2. Retain 10.5 inch thick annulus and design proposed in CG-772.
3. Design for annular thickness to be critically safe for processing 1.25 per cent U_{235} .
4. Use 10.5 inch thick annulus but redesign to handle long elements.
5. Utilize slabs or multiple small diameter cylinders.

A. Present Dissolvers

Although the present Redox dissolvers are not critically safe for enriched metal

③ dissolution, the vessels could be used for dissolution if batch control were followed. Typical dissolution rates obtainable with batch control are as follows:

<u>Criticality Control</u>	<u>Inst. Rate per Dissolver, Ton/Day</u>	
	<u>0.94% U₂₃₅</u>	<u>1.25% U₂₃₅</u>
Batch	0.8	0.4
Batch plus Boron	1.2	0.8

It is obvious in view of the 50 tons per month of 0.94 per cent U₂₃₅ scheduled for processing that the utility of the Redox plant would be seriously impaired by such an approach.

B. Annular Dissolver - CG-772 Design

The dissolver design proposed for Project CG-772⁽¹⁾ calls for a 10.5 inch thick annular slug crib to be located at the outside of a 7.5 foot O.D. vessel. Such a vessel is critically safe for enrichments up to 0.94 per cent U₂₃₅ and would be subject to batch control for processing metal of higher enrichment. Using a charged uranium density of 250 pounds per cubic foot⁽²⁾ and a 7 foot working depth, estimates of capacity for various processing conditions are:

<u>Criticality Control</u>	<u>Inst. Rate per Dissolver, Ton/Lay</u>	
	<u>0.94% U₂₃₅</u>	<u>1.25% U₂₃₅*</u>
None Required	6.9	-
Batch	-	1.2
Batch plus Boron	-	1.8

*Based on a 3.5 foot high hemispherical configuration with no charging control. Therefore, charge is limited to 4 tons.

It appears that this vessel would be capable of meeting projected needs although the operating requirement of batch control is unattractive for long term operation. The objection is minimized, however, by the thesis that should the production employing 1.25 per cent U₂₃₅ metal be adopted, it is only a matter of time (a few years) before the entire HAPO uranium throughput is of this composition. At that time new dissolvers specifically designed for 1.25 per cent material would be required in the Purex plant, and Redox would no longer be required to process high enrichment production material. Adoption of this approach would provide reasonable answers to the problems of HAPO fuels but would contribute nothing to the problems associated with the NPR fuels or power reactor fuels.

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- (1) HW-52036, "Project Proposal - E-Metal Dissolvers - Redox", August 27, 1957.
 (2) HW-54439, "Slug Distribution in an Annular Dissolver", J. J. Shefcik, January 9, 1958.

C. Design Critically Safe Vessel for 1.25 Per Cent U_{235}

Another approach to geometrical control of nuclear safety is to reduce the annular thickness to 7 inches so that 1.25 per cent U_{235} could be loaded without batch limitation. If the 7 inch thick annulus is assumed to be located around the periphery of a 7.5 foot O.D. vessel, the anticipated capacity would be 6.2 tons/day. However, many engineering problems must be resolved before this approach can be used with confidence. For example, it must be demonstrated that the proposed 6, 7, and 8 inch long fuel elements can be dumped through a chute into an annulus without bridging to such an extent that dissolving capacity is limited by inadequate uranium content in the crib. The bridging and/or jamming may be predicted to be most critical at the point where the chute enters the annulus, thereby preventing the dissolver from being loaded at all. A two to three month development program would be required to demonstrate the feasibility of this approach.

D. Modify 10.5 Inch Thick Annulus Design

Still another approach to this problem lies in the retention of a 10.5 inch thick annulus but a vessel design incorporating a detachable lid and unencumbered access to a large cross-section of the tank bottom. A plan view would be:

In this concept the tank coils, dip legs, tower, etc., would be located at the side of the vessel.

As in approach B, this vessel would be critically safe for metal containing up to 0.94 per cent U_{235} , and batch control would be required for higher enrichments. On the basis of a 7.5 O.D. vessel, a one foot thick solution annulus, and a 10.5 inch thick slug crib annulus, capacities can be postulated to be:

Criticality Control	Inst. Rate Per Dissolver		
	0.94% U_{235}	1.25% U_{235} *	1.25% U_{235} **
None Required	6.0	-	-
Batch	-	1.2	1.8
Batch plus Boron	-	1.8	3.3

*Based on a 3.5 ft. high hemispherical configuration with no charging control. Therefore, charge is limited to 4 tons.

**As above assuming a charging chute design (yet to be provided) which will insure that charge will be equally divided between opposite sides of vessel. Therefore, charge can be 8 tons.

Capacity for 0.94 per cent U_{235} processing is reduced by approximately 15 per cent but is still adequate. No change in the capacity for processing 1.25 per cent U_{235} is foreseen.

With the slug crib unencumbered and the detachable lid, the vessel has the flexibility to handle long (up to 10 ft.) fuel elements, e.g., those anticipated from NPR, PRTR, and several power reactors, without modifications. Engineering problems, such as cell layout, must be resolved, but their resolution can be anticipated if it must be done at the expense of the rinse tank which occupies a portion of each dissolver cell.

E. Slabs or Multiple Cylinders

The use of flat slabs or multiple small diameter cylinders as critically safe dissolver vessels was discarded because of fabrication difficulty and/or cell space limitations.

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