PLUTONIUM ISOLATION FLOWSHEETS

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To: F. L. Steahly

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The attached flowsheet for plutonium isolation from the Purex IIBP stream by means of ion exchange represents the most complete outline possible at the present time. It includes several points which are doubtful and which must be clarified by further experimental work.

Plutonium Reduction

The IIBP stream will be taken as it comes from the Purex process, without boildown. The chief requirement here is that the acidity be preferably 0.3M HNO₃ or less, and certainly not over 0.5M.

It is proposed that Pu(IV) and Fe(III) be reduced by making the IIBP 0.03M in hydroxylamine, and allowing three hours for the reduction. Detailed study of the rate of this reduction is needed; the specified conditions were arbitrarily selected as probably being sufficient for complete plutonium reduction.

Coupling

It is proposed to use a three-fold excess of resin, based on the plutonium. Uranium, iron, nickel, and chromium, as well as plutonium, will be retained on the resin. It is expected that UX₁ and UX₂ and fission products will also be largely retained on the resin.

The most serious lack of information on the coupling step, and probably on the whole process, is with regard to flow rate of the feed, and the resulting size of the resin column, or columns, that will be needed. With
regard to flow rate, there are two opposing requirements. The resin bed
should have a large cross-sectional area, so that the flow/unit area/minute
will be sufficiently low. A commonly accepted upper limit for flow through
resin beds is 10 ml/cm²/m.; on the basis of 2000 liters of IIBP per eight
hours, a bed area of 420 cm² would be required to give that flow rate. On
the other hand, criticality requirements on plutonium preclude the use of
columns larger than ca. 11 cm diameter; such a column has a cross sectional
area of 95 cm². Thus five columns in parallel would correspond to a relatively
high flow of 10 ml/cm²/m.

Information is therefore needed on maximum permissible flow rates per
unit area, since the larger the flow rate that can be tolerated, the fewer
the columns that will be needed.

Another point on which information is needed is on plutonium distribution
through the resin bed after the coupling, U elution, and Pu elution steps.
Such information would determine the minimum bed height which is necessary;
it would also determine whether elution of sections of the bed is feasible.
The latter point is based on the observation that plutonium forms a sharp band
at the top of the resin column. It is possible that the bed should therefore
be made in two sections; the upper one to contain plutonium, the lower con-
taining the remainder of the cations and very little plutonium. Uranium could
then be eluted from the lower section, plutonium from the upper.

Uranium Elution

The important point is that plutonium be entirely in the trivalent state,
because Pu(III) is complexed by sulfate ion much less than is Pu(IV).

**Plutonium Elution**

The eluting agent is specified as 6M nitric acid; a study should be made of the effect of acid concentration on the elution; specifically, the effect on gassing due to plutonium oxidation, and on the plutonium concentration attainable. The figure of 6M was selected somewhat arbitrarily; both lower and higher acid strengths should be tried. Elution with sodium or potassium nitrate is also a possibility.

**Re-use of Column**

It is planned to elute about 90% of the plutonium from the resin bed, leaving the remaining 10% on the bed during the next coupling step. The plutonium is probably oxidized to the tetravalent state during the nitric acid elution; it is not known whether plutonium remaining on the resin will be reduced to the trivalent state during the next coupling step. As yet no data have been obtained on re-use of resin columns containing residual plutonium; such information is urgently needed.

**Peroxide Precipitation**

The flowsheet shows addition of 30% H₂O₂ directly to the 6M nitric acid solution of plutonium. A report from Hanford (HW-19827) indicates that plutonium loss in the supernates is not excessive if the final acidity is 2.6M or less. More information on this point is needed.
Recycle of the peroxide supernates to the reduction step should cause no difficulty. The nitric acid in the supernates amounts to 72 moles; in 530 gallons of 0.3M HIBP solution there are 600 moles of HNO₃. Thus the increase in acidity of the column feed due to recycle of the supernates would be only about 12%, or 0.04M.
Ion Exchange Program

1. Reduction of Pu and Fe.
   Determine rate of reduction as a function of acid and hydroxylamine concentration.

2. Feed flow rate.
   Using a wide range of flows, determine (a) Pu loss in effluent,
   (b) Pu distribution in column.

   In connection with (2) determine what bed heights are necessary to avoid excessive Pu loss. Perhaps higher flow rates could be tolerated by using longer beds. Also study effect of variation in resin size.

4. Sectional columns.
   In connection with (2) determine whether it is feasible to divide column into two sections, with Pu retained chiefly by the first one. More efficient U and Pu elution might be possible.

5. Uranium elution.
   Determine behavior of stainless steel corrosion products, \( \text{UX}_1 \) and \( \text{UX}_2 \), and fission products. Determine flow rate necessary for equilibrium.

   Find out why we are using 6M HNO\(_3\); i.e., vary acidity. Also try sodium and potassium nitrate solutions for eluting (object - to prevent gassing). Determine flow rate necessary for equilibrium.
7. Re-use of column.

Carry out a number of coupling-elution cycles, using the same resin bed. Check plutonium loss during both coupling and uranium elution steps.

8. Peroxide precipitation.

Study precipitation from high-acid solutions from standpoint of plutonium purification and losses. Determine D.F.'s for UX₁ and UX₂, fission products, uranium and stainless steel corrosion products.

9. In conjunction with (7), evaluate the whole process, including as a part of each cycle the peroxide precipitation and recycle of supernates to the resin column.

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Plutonium Isolation
ORNL Ion Exchange Flowchart (Tentative)

Base: 1 Ton Uranium; 400 g Pu/Ton U

II BP 530 gal

Pu & Fa Reduction
(3 Hours Mixing)

Pu 80 g
Pu2O3 82.3 lb
U 0.05 lb
Fe 0.05
Ni 0.003
Co 0.006
TBP 4
Ammonia 8
H2O, HCl, HNO3 10.8

Uranium Elution

H2O 60 gal
H2SO4 2.4 lb

Resin Column

Effluent 530 gal

Pu 8 mg
(0.025 lb/m3)
U 0.05 lb
Fe 0.05
Ni 0.003
Co 0.006
TBP 4
Ammonia 8

Eluate 60 gal

Pu 0.8 mg
(0.003 lb/m3)
U 0.8 lb
Fe 0.8
Ni 0.003
Co 0.006
TBP 4
Ammonia 8

Notes:
1) Void space ca 0.8 gal
2) Plutonium crude ca 20 g/l.

Resin Column

Pu 0.8 gal
U 70 g

Pu Reduction

Pu3+ 3 g
Pu 2.2 g
U 10.2 lb
H2SO4 60 lb
H2O 10 lb

Notes:
1) Urea
2) TBP
3) Ammonia

Pu 367 g
H2O 2.2 lb
H2SO4 10.2 lb
H2O 10 lb

Pu 2.3 g

Pu Precipitate

Pu 2.3 g
H2O 10 lb
H2SO4 10 lb

Pu Precipitate To AM Line (Pu 367 g)