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DATE May 27, 1949

SUBJECT Preliminary Engineer's Flow Sheet and Process Description Redox Feed Preparation from Current Metal

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PRELIMINARY ENGINEER'S FLOW SKETCH AND PROCESS DESCRIPTION

REDOX FEED PREPARATION FROM CURRENT METAL

Project: C-187-D - Redox Production Plant No. 1

Prepared by: H. E. Hanthorn

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PRELIMINARY ENGINEER'S FLOW SKETCH AND PROCESS DESCRIPTION

REDOX FEED PREPARATION FROM CURRENT METAL

INTRODUCTION

The basis feed to the Redox plant is 5,000 lb/day of uranium, supplied both as current irradiated uranium metal slugs in jackets, and as uranium compounds from the existing metal waste storage tanks. The proportions of the two feeds are chosen according to the enrichment level of the irradiated metal, so as to give a plutonium production of about 650 gm/day. The design basis has been taken as 0 to 1500 lb/day U from the waste storage system and 2000-5000 lb/day of irradiated metal. Only the feed preparation from irradiated metal is discussed in this process description, and a basis of 5000 lb/day is used throughout.

The metal feed has associated with it 172.5 lb. Al and 2.5 lb. Si as slug jackets, and contains about 650 gm. plutonium and 3×10^5 curies of gamma-emitting fission products.

The purpose of the feed preparation section is to remove the aluminum and silicon, and to dissolve the uranium as an aqueous solution for feed to the Redox extraction battery. Secondarily, it is tentatively proposed to provide equipment in this section to achieve a preliminary decontamination from ruthenium, which is poorly handled by the Redox extraction system, and from FP which are adsorbed by the filteraid used in the clarification step.

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The product of the feed preparation section is an aqueous solution of uranyl nitrate hexahydrate of 59.4% (2 M) concentration, with part of the stoichiometric nitrate ion neutralized by NaOH, and containing nearly 2% sodium dichromate as an oxidizing agent. It contains 99.94% of the plutonium originally in the slugs, and perhaps as little as 0.3×10^5 curies of gamma-emitting FP.

PROCESS OUTLINE

1. Removal of jackets and dissolving

Jacketed irradiated slugs are received from the 200-N area in buckets in cask cars as at present. Five buckets, each containing 255 slugs, constitute a basis charge. (Eight such buckets constitute a maximum charge as discussed below.) The slugs are charged to the dissolver as at present. The jackets are then removed by the following procedure: 3208 lb. 26% sodium nitrate is run into the dissolver. Then 892 lb. 50% sodium hydroxide is added at a controlled rate, with sufficient steam on the dissolver coil to cause boiling. Boiling is continued for two hr, then cooling water is run into the coil and the charge cooled to 40°C. for jetting to the waste hold tank. A water wash of 696 lb., followed by a 5% nitric acid wash of 3583 lb., is added and jetted to waste concentration. Dissolving acid is then added in portions of 4083 lb. 60% HNO_3 each; if the nitric acid supplied to the plant is 68% strength, it can be reduced in the weigh tank before adding to the dissolver, or the water for reduction may be used as a tank rinse. Steam is run to the dissolver coil as required to start the reaction, reduced (or cooling water applied) to control the rate, and reapplied to finish the cut, as in present practice.

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The cut is complete when the specific gravity of the solution at the boiling point (about 109°C) reaches 1.82. The solution is cooled to 60°C and jetted to the metal solution tank. Three cuts as described complete the dissolution of a basis charge. No dilution inmthe dissolver is assumed, since any water added at this point would have to be evaporated at a later stage. F. Hollenbach states that it is possible to jet concentrated UNH solutions at 60°C, and freezing point curves show that the solution will not freeze above 35°.

The dissolver off-gases are passed into a scrubber, wherein they are scrubbed with a solution nearly saturated with $NaNO_3$ and $NaNO_2$ and containing NaOH sufficient to absorb all of the oxides of nitrogen entering. It is assumed that a scrubber designed on this basis will give reasonable absorption of radioiodine. The operation of the scrubber is as described in HDC-886, with pH control of the scrubber effluent. The scrubber effluent flows directly to waste storage, since it is extremely unlikely that any U or Pu values could find their way into this stream. It may be noted that if the only activity in the scrubber effluent is I^{131}, this stream could be economically disposed of by routing it to a separate tank cascade, with a holdup of about 150 days minimum, and with overflow from the final tank to crib. The air leaving the scrubber passes through a glass wool capsule and thence to the stack, using a jet as prime mover.

The three cuts constituting a basis charge as described above are all received in one metal solution storage tank and blended therein, in order to maintain a feed stream of as uniform plutonium

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concentration as possible. The metal solution tanks should have capacity for 10,000 lb metal as dissolver solution (two basis charges) in order to provide a reservoir of feed for periods in which the weather is unsatisfactory for dissolving. For the same reason, the actual operation of the dissolvers should be with 8000 lb charges, the maximum of which they are capable, instead of the basis charge as shown above.

2. Oxidation and clarification

Metal solution is withdrawn from the metal solution storage tanks in sufficient quantity for a day's operation of the Redox extraction battery, to the oxidizer, and is there sampled for critical mass control and free acidity. Oxidant solution, 626 lb 50% sodium dichromate dihydrate solution, is added, the solution is heated to 95°C, and held for 6 hr. During this period it is tentatively proposed that 200 cfm of 2% ozonized air be sparged into the oxidizer. A large part of the ruthenium would be oxidized to RuO_4 by this procedure, and volatilized and swept out of the oxidizer by the air. Any I^{131} not removed in the dissolving step would also be swept out at this point. The air leaving the oxidizer would carry about 5531 lb/day of water vapor; this would have to be refluxed by a condenser in a column similar to the arrangement on the dissolver, or the ruthenium scrubber would have to be operated at about 100°C if refluxing should carry back quantities of the volatile FP being evolved

The air, carrying the ruthenium and iodine, passes to a caustic scrubber, which removes the FP. The scrubber is operated at

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room temperature unless refluxing in the oxidizer is not feasible, in which case the temperature will be about 100°C, and a surface condenser will be added following the scrubber, the discharge of which will be routed to the condensate header. The air then passes through a capsule of glass wool, and thence to the stack via a jet as prime mover.

Following the oxidation, 279.5 lbs Superfiltrol is added as a scavenger for the "crud" to be removed by centrifugation. The filteraid is weighed into a bucket, and dumped by crane into a chute, from which it falls into the oxidizer. Water rinsing may be employed if necessary, to transfer the filteraid cleanly. The filteraid is then well suspended in the solution by agitation; the charge is then jetted at a controlled rate to a 40" solid bowl centrifuge, similar to the machines in use in the existing plant. The clarified solution flows to the IAF makeup tank, the filteraid and "crud" being retained in the centrifuge bowl. When the charge is all centrifuged, the bowl holdup is reduced to 35 gal with the skimmer. The cake is then slurry washed three times, first with 50 gal. water, then twice with 50 gal. 1% nitric acid. The dilute acid washes are expected to elute the adsorbed plutonium without removing the adsorbed FP. Finally, the cake is slurried out of the bowl with water or acid to the slurry hold tank, where it is sampled and analyzed for total alpha. If the analysis is low enough for specification, the slurry is jetted to the waste neutralizer; if not, it is returned to the oxidizer for further treatment.

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3. Acidity and concentration adjustment

Final adjustment of the acidity and the concentration of the feed solution is made in the IAF makeup tank. From the results of the free acidity determination on the sample taken in the oxidizer, the weight of nitric acid in the batch is computed. NaOH equivalent to this weight, plus 84.6 lb extra NaOH (shown on the Flow Sketch as 133.2 lb. negative acidity) is added as a 50% solution with agitation. The Flow Sketch shows adjustment of concentration by evaporation of excess water from the IAF makeup tank. If the alternate method of operation of the ruthenium scrubber noted above is chosen, with no reflux from the oxidizer, and with the scrubber temperature 100°C, adjustment would always be made by dilution in the IAF makeup tank, and the reflux column and condenser for distillate shown on the Flow Sketch would be eliminated.

DISCUSSION OF THE DESIGN

The design presented in the Engineer's Flow Sketch SK-2-5311 is based on recommendations of the Technical Divisions as presented by R. E. Smith in a rough draft entitled "Feed Preparation Flowsheet". Minor variations introduced by the writer in the material balances include use of a little less nitric acid for dissolving, in accordance with present plant practice, dissolving to a slightly higher specific gravity solution, also present plant practice, and omission of the dilution water from the dissolver, on the assurance of "S" division personnel that the solution can be jetted at 60°C. The procedure for washing the filteraid in the centrifuge was also varied, since

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displacement washing on a solid bowl centrifuge is not possible.

Materials to be received from Aqueous Makeup (SK-2-5310) are:

1. 40% sodium hydroxide solution. Tankage in the aqueous makeup section can be scheduled for this solution, which is a continuous feed. It can be mixed in the feed tank from 50% NaOH and water, if desired at a later stage in the design, by the addition of an agitator.
2. 26% sodium nitrate solution. This solution is made up by dissolving a solid, hence must be made up in the aqueous makeup section.
3. 50% sodium dichromate dihydrate solution. Same as 2.
4. 50% sodium hydroxide solution. Received on the plant and transmitted to this section via header.
5. 60% (or 68%) nitric acid solution. Same as 4.
6. 61.7% aluminum nitrate nonahydrate solution, or sodium aluminate solution. Shown on the flowsheet in anticipation of a flowsheet change. If used, either will be received from the aqueous makeup section because it is made up by dissolving a solid.

Other concentrations of nitric acid used can be made up in this section from 60% (or 68%) nitric acid and water.

Supply tankage provided includes one small and one large weigh tank for each of two dissolvers, to give desired precision in chemical additions, and to avoid scheduling difficulties and possible errors which might occur if one set of weigh tanks were manifolded to all dissolvers.

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For the same reason, the oxidizer and IAF makeup tanks are provided with separate weigh tanks.

Each dissolver off-gas scrubber is shown as having its own 40% NaOH supply tank. This is not essential, one supply tank and manifolding to the supply pumps being equally applicable if desired. The Ru scrubber is shown as having no supply tank, receiving its caustic supply directly from the headers via meters. The caustic is diluted with water in the scrubber tank. Very little caustic will be consumed by the Ru scrubber, so no weight input has been shown on the Flow Sketch. The batch is jetted to the waste neutralizer and forms a part of the caustic shown entering this vessel on SK-2-5318.

Two dissolvers are shown, each connected to a metal solution storage tank. Analysis of dissolver down time by O. C. Schroeder proves, however, that only approximately half of the total time averaged over a year is available for dissolving if the HI rule of weather conditions giving a dilution factor of 1000 is adhered to. This rule may be relaxed after observation of off-gas decontamination by the caustic scrubbers, but no quantitative information upon which to request an immediate HI ruling is available. Under these circumstances, two dissolvers will barely carry the Redox plant load, even if operated with maximum (8000 lb) charges, with no factor for additional plant down time. It is therefore proposed to provide a third dissolver, not shown on the Engineer's Flow Sketch, manifolded to the weigh tanks of dissolver No. 2, and with off-gas valved to the Ru scrubber. Since the third dissolver would be used only intermittently if at all, it is felt that

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no difficulty would be encountered in scheduling dissolving periods so they would not conflict with the oxidizer.

A dissolver waste holdup tank is provided to receive the jacket removal waste, in order that dissolver operations may be free from the requirement of close scheduling with waste neutralization.

In order to provide a reservoir of feed for the plant in periods of unfavorable dissolving weather, it is proposed to provide three metal solution tanks, each with a capacity of 10,000 lb. uranium as dissolver solution, a total of 6 days basis feed. Each of these will receive from one dissolver, but they will be interconnected by jet lines in order to enable a full inventory of solution to be carried at all times without the necessity of running the third dissolver.

The oxidizer is similar to the dissolvers in design, except that the solids charge hopper is necked down to a pipe and valved, in order to prevent splashing of active solutions up into the part of the hopper contacted by the Superfiltrol charging bucket. Weighing the Superfiltrol into the bucket will in any case be a SWP job, it is feared. The pipe through which the Superfiltrol enters the oxidizer is visualized as of fairly large size (8" or 10") and equipped with a plug cock with hydraulic or electric drive, and with an emergency impact wrench operator. Rinsing down the hopper and pipe line with water may or may not be necessary; allowing for some rinse water is shown on the Flow Sketch. Maintaining an air flow through the charging hopper with the off-gas jet and possibly providing a vibrator on the

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hopper may give a positive enough transfer of the Superfiltrol so that rinsing will be unnecessary.

If ozonation of the solution during oxidation is finally adopted, the effectiveness of a 1% ozone stream should be compared with that of the presently proposed 2% ozone; commercial ozonizers are much less efficient at concentrations above 1% and both larger equipment and higher electrical loads will be required for the 2% ozone stream.

Many serious process feasibility questions center on the use of Superfiltrol (or any insoluble filteraid) in the feed preparation process. The transfer of the slurry from the oxidizer to the centrifuge at a controlled rate offers the opportunity for jet lines to plug; slurrying out the centrifuge cake appears to be very doubtful with such a heavy material; washing the cake in the centrifuge seems of dubious efficiency and offers the possibility of reentraining the "crud" swept down by the filteraid on first centrifugation; performance of the sampler on this heavy slurry seems fraught with possibilities of plugging of lines, etc. The solid bowl centrifuge does not appear to be an entirely practical tool for the purpose of separating and washing the filteraid, yet embarkation at the present time on an equipment development program on a perforate bowl centrifuge seems impractical. It is suggested that further attention be given to the use of manganese dioxide, formed <u>in situ</u>, as the scavenger at this point. After centrifugation is complete, it can be dissolved in the centrifuge bowl with hydrogen peroxide and nitric acid, and readily flushed out to waste.

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A slurry hold tank is provided from which to sample the filteraid slurry and to eliminate the necessity for close scheduling between the oxidizer and the waste neutralizer.

The IAF makeup tank serves as a centrifuge catch tank, an evaporator for adjustment of concentration, and an agitated vessel for final adjustment of pH. Note, however, in the discussion above, that provision for concentration at this point will not be made if ozonation without reflux is a requisite step earlier in the process.

All stream volumes shown are actual daily volumes, on the 5000 lb. uranium basis. The tank working volumes shown are for an 8000 lb. uranium basis for the dissolvers and 10,000 lb. uranium basis for the metal solution tanks, and 5000 lb. uranium basis for the subsequent process steps.

On all tankage volumes, heat transfer surfaces, and the like fixed equipment, with the exception of the dissolvers and their accessories which are already sized at a maximum, factors of 1.2 for down time and 1.1 for freeboard in unagitated tanks, or 1.25 for freeboard in agitated, sparged, or boiled tanks, are applied to reach the final tank working volume, heat transfer surface, or the like.

Materials to be transferred to Waste Treatment and Storage (Sk-2-5318) are:

1. Jacket removal waste; transferred to waste neutralizer at a point in its cycle when the NaOH concentration is not so high that the sodium aluminate will be precipitated.
2. Dissolver rinses; transferred to the waste evaporator.

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3. Off-gas scrubber waste; transferred directly to waste storage.

4. Ru scrubber waste; transferred to the waste neutralizer at any time before the completion of the day's waste batch.

5. Filteraid slurry; transferred to the waste neutralizer at any time before the completion of the day's batch.

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LISTING OF MAJOR EQUIPMENT PIECES

Item	Process Volume	Working Volume	Diameter	Height
Dissolver No. 1		1700	7.0	6.86
Dissolver No. 2		1700	7.0	6.86
Dissolver No. 3		1700	7.0	6.86
Small Dissolver Weigh No. 1	133	146	2.5	4.0
Small Dissolver Weigh No. 2	133	146	2.5	4.0
Large Dissolver Weigh No. 1	667	833	4.5	7.0
Large Dissolver Weigh No. 2	667	833	4.5	7.0
Off-gas Absorber No. 1			4.0	16.0
Off-gas Absorber No. 2			4.0	16.0
Absorber Feed No. 1	968	1065	4.5	9.0
Absorber Feed No. 2	968	1065	4.5	9.0
Waste Holdup	759	835	4.5	7.0
Metal Solution No. 1	1820	2002	6.0	9.5
Metal Solution No. 2	1820	2002	6.0	9.5
Metal Solution No. 3	1820	2002	6.0	9.5
Oxidizer	1039	1700	7.0	6.86
Ru Absorber			4.0	16.0
Oxidant Addition Tank	52.4	69.2	2.0	3.0
Centrifuge				
IAF Make-up Tank	1260	1890	7.0	7.6
Centrifuge Wash Tank	100	150	2.5	4.0
PH Adjustment Tank	25.8	34.1	1.5	2.6
Filteraid Slurry Hold Tank	120	158	2.5	4.3

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