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ADDENDUM TO EW-34882, "TECHNICAL APPRAISAL OF
REDOX RUTHENIUM PROBLEMS AND THEIR RESOLUTION"

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HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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INTRODUCTION

In the light of Redox Plant performance since the start-up on 2-3-55, coupled with a review of the technological factors involved in the current flowsheet, a departure from the process program outlined in HW-34882 a) is herewith presented as an addendum to that document which will also serve to detail the corresponding items in the latest Redox Test Program, HW-35069.

OBJECTIVES

The purposes of this document are to:

- 1) Discuss the need for modification of the program given in HW-34882, and
- 2) Present the details of the proposed revisions.

SUMMARY

Recent Redox performance indicates that inadequate plutonium and uranium decontamination would be obtained with the dichromate head end process under current operating conditions. It is therefore recommended that plans (submitted in HW-34882) for the immediate adoption of the dichromate head-end for processing 200 MWD/T metal be suspended, and the following modified approach be adopted instead.

- 1) Continue on permanganate head-end until either adequate tail-end facilities or alternate flowsheet schemes are available to give improved decontamination for plutonium and/or uranium.
- 2) Change the 3A flowsheet to increase the L/V in the 3A Column scrub section.
- 3) Adopt the dual-scrub 1A flowsheet to a) evaluate effect on decontamination (potential D.F. improvement by a factor of about 2), and b) prepare for long-range, complete backcycle flowsheet.
- 4) Test the feasibility of an acid-deficient plutonium (Pu^{+6}) cycle by evaluation of plutonium oxidation states encountered under present and proposed operating conditions. If satisfactory conversion to $Pu(VI)$ can be obtained, and the desired increase in decontamination results, adopt the acid-deficient flowsheet as "standard" prior to conversion to 2-stage backcycle.

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- a) "Technical Appraisal of Redox Ruthenium Problems and their Resolution",
E. R. Irish, 1-21-55

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As recommended in HW-34882, a) the acid-deficient plutonium flowsheet should be incorporated in June or July (after installation of back-cycling), and b) plans for the precycle flowsheet will be held in abeyance until the above steps have been evaluated.

DISCUSSION

It was proposed in HW-34882 that a new low-acid Plutonium cycle flowsheet (previously piloted in the laboratory) would possibly provide adequate decontamination to permit discontinuance of permanganate head-end treatment while processing 200 MWD/T fuel. At the same time it was assumed that decontamination of uranium would be marginal but adequate for its shipment to the calcination plant. However, following the issuance of this document, the proposed low-acid plutonium flowsheet was critically reevaluated with the result that it is not now considered safe enough to assure acceptable plutonium losses. Hence an intermediate flowsheet was adopted which has provided the desired low acid concentration in the final plutonium product, but has had no effect on fission product decontamination. In addition, the final uranium product although adequately decontaminated for the most part, has been neither consistent nor as low (in G ratio) as anticipated, the minimum being 0.7 with the average being approximately 1.2, excluding those batches resulting from definitely off-standard operating conditions.

The net result is thus, that until the originally proposed low-acid plutonium flowsheet is re-examined in the laboratory and pronounced safe for plant use, and until some additional decontamination for uranium is realized (e.g., tail end ozonization), the replacement of permanganate by dichromate for routine feed preparation must be deferred since such replacement would be followed by an approximate 2 to 5-fold decrease in decontamination, sending both products out of specifications.

In view of the foregoing considerations, the following program was discussed and tentatively agreed upon by field personnel for both Separations and Separations Technology on 2-17-55. Its purpose is to provide the additional decontamination for plutonium which will be required to permit the substitution of dichromate for permanganate feed pretreatment, and for this reason should be implemented as soon as possible. It is comprised of the following points, all of which are included (in general terms) in the "Redox Test Program Authorization HW-35609, 2-11-55.

They are:

- a) Continue $KMnO_4$ head-end treatment for the duration of these tests.

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Notes: This will add no further difficulties to the canyon and crane contamination due to 1) the excellent vacuum being maintained in the H-4 oxidizer, 2) elimination of the scavenging step, 3) marked reduction in amount of activity entering the sand filter.

- b) Alter the February 3, start-up flowsheet for the third Plutonium cycle in such a manner as to increase the scrub section aqueous-to-organic flow ratios in the 3A Column.

Notes: This need not change the volume of 3BP solution. An increase of 50% in this L/V for the scrub section may provide a significant increase (factor of 1.5 to 2) in D.F.

- c) Change to dual-scrub operation in the IA Column.

Notes: This will not require a shut-down, and is not expected to affect either the capacity or stability of the column. The scrub section L/V will be decreased from ca. 0.3 to 0.1, and it is estimated that an increase in D.F. by a factor of 2 may result from the decreased salt concentration in the scrub (1.3 M vs. 2.0 M ANN). It also pilots the flowsheet for operation with 2-stage back-cycle.

- d) Attempt an acid-deficient third-cycle plutonium flowsheet to improve D.F.

Notes: This will be based on the success of complete conversion of plutonium in 2AF and 3AF to the Pu(VI) state (to prevent its loss to waste as the Pu(IV) under acid-deficient conditions), and will involve 1) heating the 2AF in E-7 to 50-60°C. with dichromate, 2) the addition of dichromate to 2AS, and 3) the addition of dichromate to 3AF (E-1-A butt). This will be preceded by a study of current plutonium oxidation states during IA dual-scrub operation. The use of acid-deficient feed and scrub, and elimination of HNO₃ from the extractant, will give an acid-deficient 3A Column (similar to IA) which should result in a significant increase in D.F., principally for ruthenium.

If these tests provide the desired margin of D.F., the change to routine dichromate feed treatment will then follow, and if they fail to provide the additional decontamination or prove to be unworkable, it is recommended that a period of test operation with dichromate feed treatment be initiated to determine actual performance under current plant conditions following which Redox

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Plant operation will revert to the schedule given in HW-34882, viz., a) adopt the acid-deficient plutonium flowsheet in June or July (after the installation of the second stage of waste back-cycling), and b) hold plans for the pre-cycle flowsheet in abeyance until the above steps have been evaluated.

Head, Redox Plant Processes
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