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COMPARISON OF PUREX PROCESS RESULTS IN MIXER SETTLERS AND PULSE COLUMNS

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1.0 Introduction

A feasibility study of the Purex flowsheet in pulse column equipment has been completed in the ORNL Purex Pilot Plant. Eleven runs [REDACTED] [REDACTED] have been made in the pilot plant at a throughput of 75 kg. to 125 kg. of uranium per day. The comparison between the ORNL pulse column and the SPRU mixer settler is obviously preliminary since KAPL has made only one run in the pilot plant SPRU equipment which was not given the head-end treatment. All data were obtained using feed solutions prepared from slugs having similar history of irradiation and cooling. No head-end treatment was made on any of the runs reported in this memo.

There are three differences in the way results are measured at ORNL and at KAPL. (1) ORNL data for the uranium product in both cycles and for the plutonium product in the first cycle are based upon the composite sample, i.e. the entire product is caught in a vessel and sampled. Since the plutonium product goes continuously to a resin bed following the second plutonium cycle, no composite is possible. KAPL reports the average of the flowing stream samples for losses and decontamination. It is believed that both the flowing stream average and the composite sample results should be published so that an index of the performance of the system under optimum conditions might be obtained from the flowing stream samples and an index of the actual performance of the system under conditions simulating plant operation might be realized from the composite samples.

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(2) KAPL does not analyze for niobium as a separate fission product and one can not trace the separation of niobium in the system. Since niobium may limit uranium gamma decontamination it would be fortunate if these results were available.

(3) KAPL has developed a system of radiochemical counting based upon adsorber techniques while ORNL prefers the individual fission product separation analysis. It is not known whether these differences markedly affect the validity of the comparisons made in this memo. However, a standardization of methods of analysis between the two sites would simplify the comparison of contactors.

The comparisons are made on the basis of uranium and plutonium losses, the decontamination of uranium and plutonium from fission products, and the separation of uranium and plutonium from each other.

2.0 Summary

2.1 Uranium and Plutonium Losses

The loss of uranium through two solvent extraction cycles is 0.20 percent (composite) for pulse columns and 0.04 percent for the SPRU mixer settlers. The plutonium loss through the first cycle is 0.14 percent (composite) for the pulse columns and 0.03 percent for the mixer settlers.

These losses for both contactors are not to be regarded as final. Recent pilot plant pulse column runs with a more optimum pulse amplitude and frequency has decreased the IBU plutonium loss from 0.13 to 0.04 percent

2.2 Decontamination Factors for Pulse Column and Mixer Settler Two-Cycle Runs Compared With Specifications

The two cycle uranium and plutonium decontamination factors for pulse columns and mixer settlers are compared with process specifications in the

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following table:

	<u>Pulse Column</u>	<u>Specification</u>	<u>Mixer Settler</u>
<u>Uranium</u>			
Gross Beta D.F.	5.8×10^6	4.0×10^6	6.3×10^6
Gross Gamma D.F.	6.1×10^5	6.3×10^6	2.0×10^6
Pu D.F.	1.1×10^5	4.0×10^4	4.2×10^4
<u>Plutonium</u>			
Gross Beta D.F.	1.7×10^6	1.0×10^7	1.0×10^7
Gross Gamma D.F.	3.2×10^6	1.0×10^7	5.0×10^6
Uranium D.F.	6.1×10^5	2.5×10^5	2.5×10^5

From the data presented above, it is seen that ORNL and KAPL both fail to meet the uranium gamma specification probably due to niobium limitations. There is no cause for concern on plutonium specifications because the plutonium decontamination factors listed above are the most pessimistic possible in that UX is considered as a fission product contamination in the plutonium stream. Further, the ion exchange resin step would enable the plutonium stream to meet specification even counting UX as fission product.

2.3 Performance of Pulse Column Using Purex Flowsheet

Pulse columns have exceeded expectations based upon laboratory countercurrent batch extraction runs and have compared favorably with mixer settlers in demonstrating the Purex flowsheet.

The pulse columns have operated as smoothly as packed columns, requiring almost no maintenance and very little operational attention.

3.0 First Cycle Purex Performance of ORNL Pulse Columns, Laboratory Batch Countercurrent Extractor and SPRU Mixer Settlers

The runs used for this comparison are ORNL Run No. 8 and SPRU Run No. 7 both without head-end treatment. ORNL Run No. IHP-8 is considered a typical run. SPRU Run No. 7 is the only run made under these

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conditions at KAPL using the pilot plant mixer settlers. KAPL has kindly supplied ORNL with unreported information on this run and the results may be considered tentative. It is considered necessary at this time however to evaluate pulse column performance using the data which are available. It is particularly advantageous to have the data from the SPRU mixer settler run since the mixer settler with sufficient stages should define the ultimate chemical limit of a given flowsheet. Detailed first cycle results are given in Table I. The results for the second plutonium cycle at ORNL in which packed columns are in operation can not be used for this comparison although these results are given for general interest. The results are significant, however, in that pulse columns have shown marked superiority over the packed columns in the first cycle.

3.1 First Cycle

3.11 Uranium and Plutonium Losses

The loss of uranium and plutonium in all three contactors is less than 0.05 percent uranium and less than 0.17 percent plutonium (Table I). Although the pulse column exhibits lower uranium loss values than the counter-current extractor, the mixer settler gives still lower values. Lower plutonium losses in the IA contactor are given by the pulse column but the IB plutonium loss is 0.13 percent compared with 0.004 percent for the mixer settler. Greater pulse amplitude is necessary for pulse column IB.

The uranium losses through the first cycle are 0.02 percent for pulse columns and 0.005 percent for mixer settlers. The plutonium losses are 0.14 for pulse columns and 0.03 percent for mixer settlers.

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3.12 Uranium Decontamination

The decontamination factors for the three contactors in the first cycle uranium product are listed below:

Uranium Decontamination Factors in the First Cycle

<u>ICU</u>	<u>Pulse Column</u>	<u>Batch Countercurrent</u>	<u>Mixer Settler</u>
Gross Beta	2.9×10^4	4.9×10^3	3.2×10^5
Gross Gamma	6.1×10^3	-	1.6×10^4
Ru Beta	1.6×10^3	8.5×10^2	8.0×10^3
Zr Beta	1.9×10^4	6.3×10^4	4.0×10^4
TRE Beta	2.4×10^7	2.3×10^6	6.3×10^6
Nb Beta	1.2×10^4	-	-

The following conclusions may be drawn with reference to the decontamination of uranium in the first cycle. (1) The KAPL mixer settlers give the best decontamination of the uranium stream through the first cycle, followed by the pulse column and the laboratory batch countercurrent extractor in that order. (2) The fundamental difference between the performance of the contactors in the first cycle lies in the distribution of ruthenium in the A contactor. (3) The cause of the increased ruthenium decontamination in the A contactor of the mixer settler is not known but it has been suggested by KAPL as possibly due to a change in ruthenium extractibility brought about by greater aging of feed solutions before entrance to the mixer settler (Page 94, KAPL 523).

It is seen that the gross beta decontamination factor for the first cycle uranium product is a factor of eleven greater, the gross gamma a factor of three greater and ruthenium beta a factor of five greater in the mixer settler run than in the pulse column run. The ruthenium decontamination in the first cycle of the pulse column runs is aided greatly by a good ruthenium decontamination in column C.

The pulse column exceeds the performance predicted in batch countercurrent throughout the first cycle and exceeds the total rare earth decontamination factor of the mixer settler.

Ruthenium Distribution The Fundamental Difference in Results Between Contactors

From the table above it is seen that there is a factor of five greater ruthenium decontamination in the mixer settler than in the pulse column for the uranium stream. Ruthenium decontamination for the first cycle plutonium stream is a factor of 35 greater in the mixer settler than in the pulse column. This means that the mixer settler must be achieving a decontamination factor for ruthenium in the A contactor six times as great as the pulse column and twelve times as great as the batch countercurrent extractor. It is difficult to understand how any contactor could perform markedly better than the countercurrent batch extractor which consists of eight stages at nearly 100 percent efficiency. There is very little additional decontamination after the sixth stage in the A contactor. This means that the system is chemically limited to countercurrent ruthenium results unless there exist in the SPRU feed preparation procedure conditions which are not comparable to the feed preparation of the material used for the countercurrent runs.

Probable Cause of Increased Ruthenium Decontamination in the A Contactor of the Mixer Settler Over the Pulse Column

A trip was made to KAPL to check its method of feed preparation. ORNL and KAPL were in good agreement on methods except for this fact: KAPL feeds may be stored for days at various points before being fed to the A bank of mixer settlers (Page 68, KAPL-523). KAPL further noted that the gross

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beta, gross gamma and ruthenium beta decontamination improved steadily in the course of a five day run. This indicates that a possible cause of the increased ruthenium decontamination in the A contactor of the mixer settler is due to a chemical difference in feed preparation brought about by the aging of feed solution rather than a contactor characteristic.

3.13 Plutonium Decontamination

Plutonium decontamination data for the first cycle are summarized in the following table:

Plutonium Decontamination Factors in the First Cycle

<u>IBP</u>	<u>Pulse Column</u>	<u>Batch Countercurrent</u>	<u>Mixer Settler</u>
Gross Beta	6.7×10^3	9.2×10^3	2.0×10^5
Gross Gamma	2.2×10^3	-	1.3×10^4
Ru Beta	7.2×10^2	2.0×10^3	2.5×10^4
Zr Beta	2.0×10^3	5.8×10^3	1.3×10^4
TRE Beta	1.3×10^5	1.1×10^5	3.2×10^6
Nb Beta	3.2×10^3	-	-

The following conclusions may be drawn from this data:

- (1) Ruthenium decontamination in the A contactor dictates the difference in results between the pulse column and the mixer settler. Because of volume relationships involved in the B contactor, the six fold better ruthenium decontamination recorded for the A contactor makes a very large effect in the B column plutonium stream.
- (2) Total rare earth beta decontamination is less in the plutonium stream for the pulse column because this fission product showed greater uranium stream decontamination than did the mixer settlers, and a given quantity of fission products in the AP stream must split either into the uranium stream or into the plutonium stream.

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- (3) The results from the pulse column run closely approximate the results from the batch countercurrent run.

4.0 Second Uranium Cycle Purex Performance of ORNL Pulse Columns and

SPRU Mixer Settlers

4.1 Uranium Losses

The mixer settlers show a second cycle uranium loss of 0.001 percent compared with 0.17 percent for the pulse column. The uranium loss through the two cycles is 0.04 percent for the mixer settlers and 0.20 percent for the pulse columns. The greatest uranium loss in the pulse columns is to the IEW stream (0.12 percent) and is due to an undersized pulse pump which is being replaced.

4.2 Decontamination of Uranium

The decontamination factors for the second uranium cycle are listed in the table below:

Second Uranium Cycle Decontamination Factors

	<u>Pulse Column</u>	<u>Mixer Settlers</u>
Gross Beta	210	20
Gross Gamma	76	125
Ru Beta	420	125
Zr Beta	78	62
TRE Beta	13	-
Nb Beta	78	-

It will be noted that the principal difference in the results is a ten fold better gross beta and a three-fold better ruthenium beta decontamination factor in the pulse column than in the mixer settler.

The two cycle uranium decontamination factors for the pulse column and mixer settler runs are listed below.

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Two Cycle Uranium Decontamination Factors

	<u>Pulse Columns</u>	<u>Mixer Settlers</u>
Gross Beta	5.8×10^6	6.3×10^6
Gross Gamma	6.1×10^5	2.0×10^6
Ru Beta	7.9×10^5	1.0×10^6
Zr Beta	1.5×10^6	2.5×10^6
TRE Beta	1.9×10^7	"Too high to measure"
Nb Beta	5.4×10^5	-

The conclusions to be drawn here are these:

- (1) The results of the individual fission product decontamination of uranium in two cycles solvent extraction in pulse columns and mixer settlers are almost identical.
- (2) Gamma decontamination in the uranium product is limited by niobium, which contributes over half the residual gamma in the uranium product.
- (3) The pulse column is producing uranium product that is equivalent to that produced in the mixer settler through two cycles of solvent extraction.

4.3 Plutonium in the Uranium Product

The uranium in two cycles of solvent extraction has been decontaminated from plutonium by a factor of 1.1×10^5 in the pulse column run and by a factor of 4.2×10^4 in the mixer settler.

4.4 Specification for the Uranium Product

The specification for the final uranium product is that it must not contain fission product beta activity of greater than 10 percent of natural uranium background nor gamma activity greater than 100 percent of natural uranium gamma activity. This corresponds to a beta decontamination factor of 4.0×10^6 and a gamma factor of 6.3×10^6 for [REDACTED] material cooled 90 days. The uranium product must be decontaminated from plutonium by a factor of 4×10^4 .

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Both the ORNL and the KAPL uranium products meet beta specifications and plutonium in uranium product specifications. Neither ORNL nor KAPL meet the gamma specification for the uranium stream due to insufficient decontamination for niobium. A flowsheet modification which should correct the niobium distribution in the second uranium cycle has been developed by the ORNL laboratory section and is being demonstrated in the pilot plant.

5.0 Second Plutonium Cycle Purex Performance of ORNL Packed Columns and SPRU Mixer Settlers.

This section is not intended as a comparison between packed columns and mixer settlers but is presented as the remainder of the data for the comparison between ORNL Run No. 8 and SPRU Run No. 7.

5.1 Plutonium Losses

The plutonium loss in the second cycle in the ORNL packed columns is 0.12 percent to the IIAW stream and 0.02 percent to the IIBW stream for a total of 0.14 percent in the second cycle and a plutonium loss through two cycles of 0.28 percent.

The plutonium loss in the second cycle in the SPRU mixer settler is 0.01 percent to the IIAW stream and 0.05 percent to the IIBW stream to give a two cycle plutonium loss of 0.09 percent.

5.2 Plutonium Decontamination in the Second Cycle

The decontamination factors for plutonium in the second cycle in the ORNL packed columns and in the SPRU mixer settlers are listed in the table below:

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Second Plutonium Cycle Decontamination Factors

	<u>ORNL Packed Columns</u>	<u>Mixer Settlers</u>
Gross Beta	540	50
Gross Gamma	1000	380
Ru Beta	410	64
Zr Beta	1800	100
TRE Beta	680	8
Nb Beta	280	-

It is seen that the ORNL second plutonium cycle decontamination is better than the KAPL second plutonium cycle by an average factor of ten.

The table below lists the plutonium decontamination factors for two cycles at ORNL and at KAPL.

Plutonium Decontamination Through Two Cycles Purex

For ORNL and KAPL

<u>Decontamination Factor</u>	<u>ORNL</u>	<u>KAPL</u>
Gross Beta	1.7×10^6	1.0×10^7
Gross Gamma	3.2×10^6	5.0×10^6
Ru Beta	3.2×10^5	1.6×10^6
Zr Beta	3.6×10^6	1.3×10^6
TRE Beta	5.6×10^7	2.5×10^7
Nb Beta	2.3×10^6	-

The conclusion to be drawn from these data is that the KAPL advantage in the first cycle ruthenium decontamination is not entirely overcome in the second cycle packed columns at ORNL. It is interesting to note that the ORNL zirconium and total rare earth decontamination is superior to that of KAPL. It should be noted further that these ORNL results were achieved with packed columns which were limited to only ten feet of scrub section.

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5.3 Uranium in Plutonium Product

The ORNL plutonium product through two cycles was decontaminated from uranium by a factor of 6.1×10^5 compared to 2.5×10^5 for the KAPL product.

5.4 Plutonium Specifications

The specifications for plutonium are that the beta and gamma decontamination factors be at least 1.0×10^7 each for material cooled 90 days.

It will be noted that ORNL does not meet the beta or gamma specification for plutonium and KAPL barely meets the beta specification and fails to meet the gamma specification. Failure to meet the plutonium specification is of little concern at ORNL due to these facts. (1) The factors listed above are the most pessimistic possible in that UX activity is counted as fission product activity in the plutonium stream. This correction increases the gross beta decontamination factor to 4.5×10^6 .

(2) The ion exchange resin bed plutonium isolation step increases the gross beta decontamination factor to 4.7×10^7 and the gross gamma decontamination factor to 1.1×10^7 . (3) It is believed that better decontamination will be achieved in solvent extraction when pulse columns are installed to give adequate scrub height to the IIA column.

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Table I

Comparison of Pulse Column Results with Countercurrent Results and with
SPRU Mixer Settler Results Using Purex Process, First Cycle

Conditions:

Pulse Column: Throughput - 75 kg metal/day

Column	Diameter (in.)	Contacting Height	Gals/hr/ft. ²	Pulse Ampl. (in.)	Pulse Frequency (cpm)
IA (Extraction)	2	12	555	0.92	58
IA (Scrub)	2	18	460	0.92	58
IB (Partitioning)	2.5	18	390	0.75	58
IB (Scrub)	2	8	190	1.08	58
IC	4	12	310	0.56	73

All pulse plates on 2" spacing with 1/8" diameter holes, 23% free area per plate.

Mixer Settler: Throughput - 7.4 kg metal/day

Bank	Actual Stages
IA (Extraction)	12
IA (Scrub)	8
IB (Partitioning)	10
IB (Scrub)	10
IC	10

Batch Countercurrent Extractor

Bank	Actual Stages
IA (Extraction)	12
IA (Scrub)	8
IB (Partitioning)	10
IB (Scrub)	10
IC	10

Three minute equilibrations, five volume changes.

Results:

1st Cycle Losses

% U Loss

	Pulse Column	Countercurrent	Mixer Settler
IA	0.002	0.002	0.003
IB	0.001	0.0001	0.0009
IC	0.020	0.053	0.0007
Total	0.023	0.055	0.0046

% Pu Loss

IA	0.006	0.015	0.023
IB	0.13	0.057	0.004
Total	0.14	0.072	0.027

1st Cycle Decontamination Factors

IAP

Gross Beta	4.2×10^3	3.1×10^3	-
Gross Gamma	5.3×10^2	1.1×10^3	-
Ru Beta	4.6×10^2	2.3×10^2	2.6×10^3 (calculated)
Zr Beta	1.5×10^3	5.2×10^3	-
TRE Beta	1.1×10^5	2.5×10^6	-

ICU

Gross Beta	2.9×10^4	4.9×10^3	3.2×10^5
Gross Gamma	6.1×10^3	-	1.6×10^4
Ru Beta	1.6×10^3	8.5×10^2	8.0×10^3
Zr Beta	1.9×10^4	6.3×10^4	4.0×10^4
TRE Beta	2.4×10^7	2.3×10^6	6.3×10^6
Nb Beta	1.2×10^4	-	-

IBP

Gross Beta	6.7×10^3	9.2×10^3	2.0×10^5
Gross Gamma	2.2×10^3	-	1.3×10^4
Ru Beta	7.2×10^2	2.0×10^3	2.5×10^4
Zr Beta	2.0×10^3	5.8×10^3	1.3×10^4
TRE Beta	1.3×10^5	1.1×10^5	3.2×10^6
Nb Beta	3.2×10^3	-	-

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Table II

Comparison of Two Cycle Uranium Results in ORNL

Pulse Columns and SPU Mixer Settlers

Conditions for Second Uranium Cycle

Pulse Column: Throughput - 75 kg metal/day					
Column	Diameter (in.)	Contacting Height	Gal/hr/ft. ²	Pulse Ampl. (in.)	Pulse Freq. (cpm)
ID (Extraction)	2	8.5	588	0.92	58
ID (Scrub)	2	13	469	0.92	58
IE	4	15	308	0.56	73

All pulse plates on 2" spacing, 1/8" diameter holes, 23% free area per plate.

Mixer Settler - 7.4 kg metal/day

Bank	Actual Stages
ID (Extraction)	12
ID (Scrub)	8
IE	10

Two Cycle Uranium Results:

<u>% U Loss</u>	Pulse Column	Mixer Settler
ID	0.05	0.0008
IE	0.12	0.009
Total	0.17	0.0098
Two Cycle Total	0.20	0.038

Two Cycle Uranium Decontamination Factors:

Gross Beta	5.8x10 ⁶	6.3x10 ⁶
Gross Gamma	6.1x10 ⁵	2.0x10 ⁶
Ru Beta	7.9x10 ⁵	1.0x10 ⁶
Zr Beta	1.5x10 ⁶	2.5x10 ⁶
TRE Beta	1.9x10 ⁷	"Too high to measure"
Nb Beta	5.4x10 ⁵	-
Pu	1.1x10 ⁵	4.2x10 ⁴

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Table III

Comparison of Two Cycle Plutonium Results at ORNL
and at KAPL

<u>% Plutonium Loss</u>	<u>ORNL</u>	<u>KAPL</u>
IIA	0.12	0.011
IIB	0.02	0.05
Total	0.14	0.06
Two Cycle Total	0.28	0.09

Two Cycle Plutonium Decontamination Factors:

Gross Beta	1.7×10^6	1.0×10^7
Gross Gamma	3.2×10^6	5.0×10^6
Ru Beta	3.2×10^5	1.6×10^6
Zr Beta	3.6×10^6	1.3×10^6
TRE Beta	5.6×10^7	2.5×10^7
Nb Beta	2.3×10^6	-
U	6.1×10^5	2.5×10^5

Packed columns used for second plutonium cycle at ORNL.

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