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METHYL ISOBUTYL KETONE AS A SOLVENT FOR U^{233} SEPARATION FROM THORIUM AND FISSION PRODUCTS IN CONTINUOUS COLUMN OPERATIONS

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LABORATORY SECTION

METHYL ISOBUTYL KETONE AS A SOLVENT FOR U^{233} SEPARATION FROM THORIUM AND FISSION PRODUCTS IN CONTINUOUS COLUMN OPERATIONS

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TABLE OF CONTENTS

		Page Number
1.0	Abstract	4
2.0	Introduction	5
3.0	Summary	6
4.0	Extraction	7
5.0	Scrubbing	8
6.0	Stripping	9
7.0	Discussion	9
8.0	Bibliography	10

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1.0 Abstract

The use of methyl isobutyl ketone as a solvent for U^{233} recovery from solutions of irradiated thorium has been evaluated in countercurrent packed columns.

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2.0 Introduction

Uranium-233 is a fissionable material which is produced by the neutron bombardment of Th^{232} as follows:

$$_{90}Th^{232}\ (n,\gamma) \longrightarrow {}_{90}Th^{233} \xrightarrow[23\ min.]{\beta} {}_{91}Pa^{233} \xrightarrow[27.4d]{\beta} {}_{92}U^{233}$$

Uranium-233 may be produced in gram quantities by this reaction in proposed breeder piles. Consequently, a feasible chemical separation process is required for the isolation of this material from bombarded thorium and fission products.

Solvent extraction processes employing diisopropyl ether and dibutyl cellosolve were demonstrated at ORNL (1, 4, 5, 6, 7). Another process using continuous extraction with methyl isobutyl ketone was investigated at the Argonne National Laboratory (8). However, the latter method, which utilized ammonium nitrate as an added salting agent, was developed only for use on a special problem, and did not necessarily represent the optimum conditions for use of methyl isobutyl ketone as a solvent in 23 recovery.

Methyl isobutyl ketone was used successfully in the solvent extraction of uranium-235 from fission products in dissolved MTR fuel assemblies. In this process, aluminum nitrate was employed as a salting agent (2, 3). Since it has been shown that the salting effect of thorium nitrate is almost equivalent, mole for mole, to that of aluminum nitrate (4), the extensive development data obtained in the 25 Process is directly applicable to the solvent extraction recovery of 23 by the substitution of thorium

Introduction (continued)

nitrate for aluminum nitrate in the 25 flowsheet (Figure 2.0-1). However, it was necessary to determine HETS values as a function of flowrates, thorium reduction factors, and the amount of additional decontamination obtainable in a lengthened scrub section.

3.0 Summary

Methyl isobutyl ketone has been found satisfactory as a solvent in a continuous column process for the recovery of uranium 233 from thorium and fission products. Thorium nitrate is employed as salting agent in extraction, aluminum nitrate in scrubbing, and the extraction is performed from an acid deficient system.

The HETS of the system was approximately 2.5 feet and did not vary significantly over the range from 3% to 85% of the flooding rate. Using this HETS value and the loss obtained in a 7.7 foot column (1%), it was calculated that seventeen feet of extraction section would be required to yield 99.99% uranium recovery. Beta decontamination factors of 9×10^3 and thorium reduction factors of 10^4 were demonstrated through one cycle. Each of these values was increased to greater than 10^5 by increasing the scrub section to 14 stages.

This report is concerned with the preliminary development of the process and since this work further improvements, which will be reported, have been made.

4.0 Extraction

The 1-1/2 inch diameter columns used for determining HETS values had 7.7 feet of extraction section and no scrub section, and were packed with 1/4 x 3/8 inch stainless steel split rings. The fission product decontamination studies were made in a 1 inch diameter column, having 14 feet of extraction and 10 feet of scrub section, packed with 3/16 inch Fenske packing.

The optimum conditions for uranium-fission product separation, using methyl isobutyl ketone as a solvent, were established in the development of the 25 process[2,3]. The aqueous phase in extraction was made up of four parts of aqueous feed (1.3 M $Th(NO_3)_4$, 0.05 N acid deficient) and one part of scrub solution (1.3 M $Al(NO_3)_3$, 0.05 N acid deficient).

Under these conditions, a beta decontamination factor of 9×10^3 was demonstrated. The average uranium loss in the 7.7 foot column was 1.0% and the column height calculated for 99.99% 23 recovery was 17 feet.

By batch equilibrations, the effect of temperature on the 23 distribution coefficient was studied for this process. The data obtained were somewhat erratic, but indicated that the 23 distribution coefficient in extraction did not vary greatly from 2.8 in the temperature range of 10-30°C.

The HETS for the methyl isobutyl ketone system did not vary appreciably from 2.5 feet within the range of 3% of 85% of the flooding rate. Thorium

Extraction (continued)

reduction factors did not change significantly from the value of 90 in the same range of flowrates (Table 4.0-1).

5.0 Scrubbing

No continuous column data are available on the added separation gained by scrubbing for the 23 methyl isobutyl ketone system. However, a systematic study of scrubbing was made for the 25 methyl isobutyl ketone process, which is applicable to the 23 process. The scrub solution recommended in early 25 process work was a 1/10 volume of 1.3 M $Al(NO_3)_3$, 0.05 N deficient in HNO_3.

It was found that by increasing the scrub section length from 0 to 14 stages, the gross beta decontamination factor was increased by a factor of 10. This increased decontamination was also obtained by using 6 stages with a 1/5 volume of 0.7 M $Al(NO_3)_3$, 0.2 N HNO_3 deficient scrub. In one run, using 1.3 M $Al(NO_3)_3$, 0.05 N HNO_3 deficient scrub and feed solutions, a gross beta decontamination factor of 10^5 was obtained. In this experiment, there were approximately 14 scrub stages and the solvent, feed, scrub ratio was 3:1:1. Thus, it appears that even better scrubbing is possible by increasing the scrub to solvent ratio.

The recommended scrub conditions for the 23 methyl isobutyl ketone process appear sufficient to insure a fission product decontamination of 10^4 for one cycle. Another promising method of increasing decontamination is

Scrubbing (continued)

the addition of a scrub section to the strip column in which the aqueous strip would be contacted with fresh solvent.

6.0 Stripping

The stripping step of this process consisted of contacting the scrubbed methyl isobutyl ketone, carrying the 23, with demineralized water in a packed column. The 23 distribution coefficient in stripping (aq/org) was found to be ≥ 100.

Less than 0.01% of the 23 was lost when one volume of demineralized water and five volumes of methyl isobutyl ketone were contacted in eleven feet of column packed with 1/4 x 3/8 inch Raschig rings, but essentially no decontamination was gained in this step. No crud or operational difficulties were encountered.

7.0 Discussion

The use of methyl isobutyl ketone as a solvent afforded excellent separation of uranium from thorium and fission products. The low HETS values and general flexibility of the system warrent further investigation of methyl isobutyl ketone as a solvent when facilities are available for continuous column studies at higher activity levels.

In this preliminary work only scouting studies of Pa-U separation with methyl isobutyl ketone were made[7], but subsequent work has shown the separation to be adequate.

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FIGURE 2.0-1

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SCHEMATIC DIAGRAM OF "23" SOLVENT EXTRACTION

FLOWSHEET AND EQUIPMENT

Basic: 50 kg of bombarded thorium metal
75 grams of "23"
Solvent: Methyl isobutyl ketone

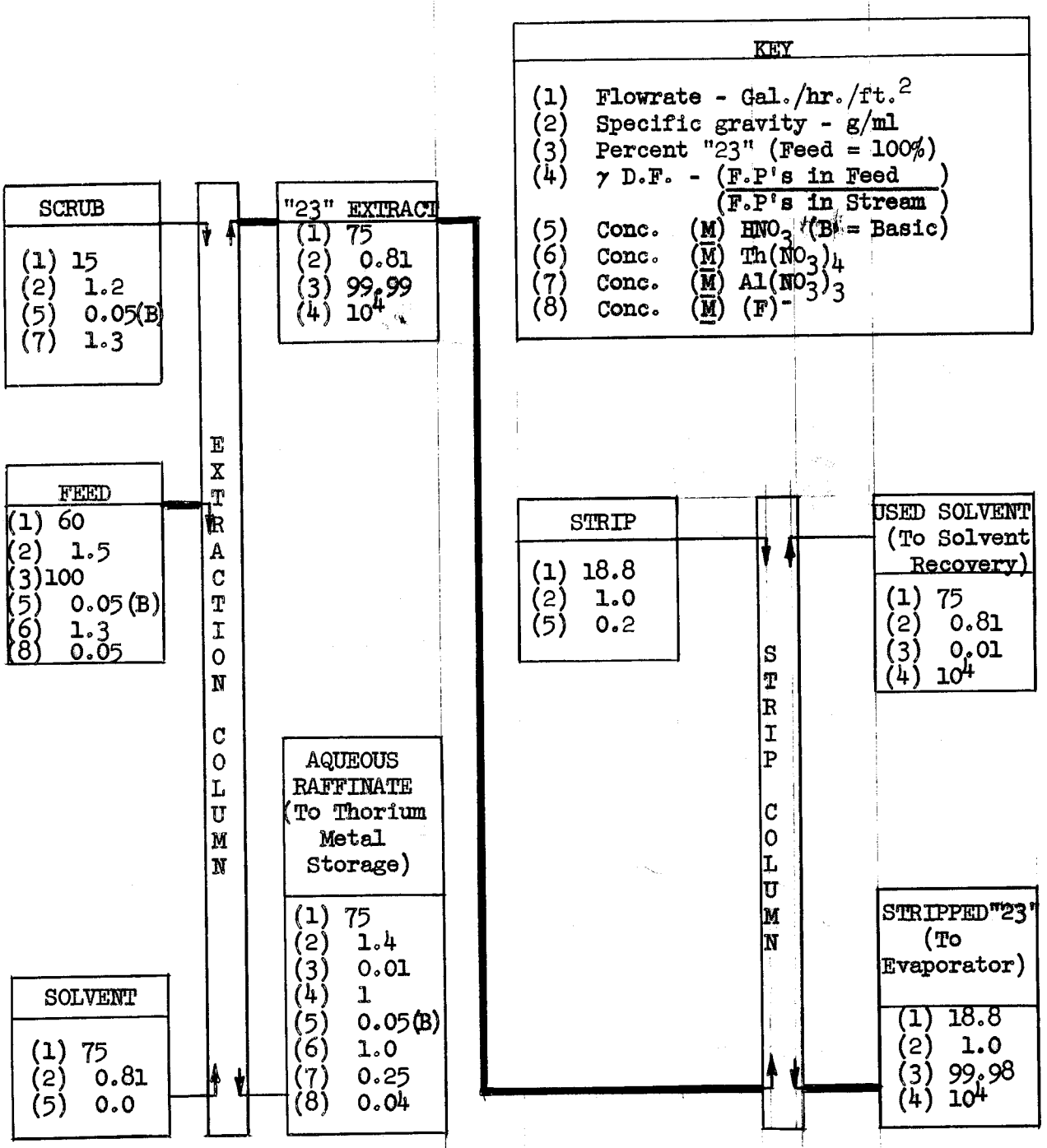

Table 4.0-1

Effect of Flowrate On HETS And Thorium Reduction

Extraction Feed(1 volume): 1.2 M $Th(NO_3)_4$
0.08 N HNO_3 deficiency
5.3×10^4 c/m/ml 23 tracer

Solvent (1 volume): Pretreated methyl isobutyl ketone

Temperature: 23°C

Column: 1-1/2" x 8' pyrex glass column
7.7' of 1/4" x 3/8" Raschig rings
No scrub section - Interface at top.

Flooding rate with equal flowrate of solvent and extraction feed: 1000 gal/hr/ft^2 total through-put.

Flooding %	HETS (Ft.)	Column Height to give 0.01% Loss (Ft.)	Th Reduction Factor
3	2.5	16	105
10	2.6	17	90
20	2.4	15	90
30	2.4	15	-
40	2.4	15	-
55	2.6	17	90
65	2.4	15	90
65	3.1	20	-
75	2.5	16	90
85	2.6	17	-