HW-39695

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ADDITIONAL INFORMATION, HWIR-560 [redacted]
AEC CASE S-12,485

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ADDITIONAL INFORMATION, HWIR-560, AEC CASE S-12,485

The following information is being furnished to supplement Invention Report HWIR-560, Document HW-33435 (AEC Case S-12,485).

1. All aluminum nitrate solutions used in the laboratory experimental work were 2.5 M with 1600 milliliters of solution used to dissolve 2.2 M of aluminum jacket material. Other concentrations and volumes were not considered due to final solution volume and freezing point limitations of the facilities for which this process was developed; other concentrations and volumes have been shown by other workers to give satisfactory reaction rates. All development work with the procedures presented in this work was done at the same catalyst concentration; this concentration is expressed as 5 per cent by weight of aluminum metal with the addition calculated as $Hg(NO_3)_2 \cdot H_2O$ (for 58 grams of aluminum jacket, 2.9 grams of $Hg(NO_3)_2 \cdot H_2O$ would be used). In the laboratory and on a semi-works scale similar media to this (HNO_3, etc.) have shown that catalyst concentrations may vary from 0.5% to 10% expressed on the above basis.

2. The uranium metal was best dissolved in the laboratory equipment used by starting with 45% nitric acid, and adding 60% nitric acid as the reaction progressed, thus maintaining a 45% nitric acid concentration over the initial dissolving. The stoichiometrical requirements for the nitric acid (or nitrate ion) reaction with metals varies according to conditions; with air sparge rate, condenser arrangement, etc., influencing the stoichiometry; reflux conditions were used in this work. A range of concentrations have been used; initial acid concentrations of 50% or 60% nitric acid, with no subsequent additions, have been used for very rapid dissolving, as have flowsheets using from 30% to 60% nitric acid with subsequent addition of 60% acid. Using fluoride ion catalyst, thorium may be dissolved in a manner similar to uranium over a range of acid concentrations; catalyst concentrations from 0.0[illegible] to 0.1 M fluoride ion may be used, but 0.03 M is suitable to give a reaction for thorium similar to that of uranium.

3. The dibasic aluminum nitrate was converted to aluminum nitrate by addition of 28% nitric acid; in practice this is usually accomplished by adding 60% nitric acid, and then diluting with water.

4. All the reactions were conducted at the boiling temperature. The boiling temperature is a function of the free nitric acid present,

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and may vary over the duration of the run. Dissolving aluminum in aluminum nitrate, or in water with slow nitric acid addition, gave temperatures from 100° to 104°C. Dissolving uranium or thorium in 45% acid gave initial temperatures of about 110°C and these decreased to about 100°C as the reaction progressed. Dissolving uranium or thorium in 60% nitric acid would increase the starting temperature to about 115°C at the start of the reaction, but the temperature would drop to near 100°C when all acid had been consumed.

5. Detailed flowsheets are attached showing the preferred conditions. Ranges are as indicated in the foregoing discussion.

6. The attached flowsheets give details of the essential slug dimensions, as well as further details on the conditions used and results obtained.

7. The following reports may be considered to give pertinent information relative to the invention:

a. Details of the development work leading to and demonstrating the invention are given in "Progress Report - Chemical Development Unit", by F. W. Woodfield. HW-32339 for June, 1954; HW-32663 for July, 1954; and HW-32964 for August, 1954.

b. A report summarizing the material in the above progress reports, plus giving the details of uranium dissolving flowsheets found in the previously mentioned HW-32823 has been prepared but not published. It will probably be a secret HW document entitled, "Application of the Mercury-Catalyzed Aluminum Jacket Dissolving Technique to the Redox Process - II" by M. H. Curtis-J. L. Bradford.

c. The utilization of solutions obtained by this process is illustrated in flowsheets presented in HW-30838, "Application of the Mercury-Catalyzed Aluminum Jacket Dissolving Technique to the Redox Process", M. H. Curtis et al.

8. The uranium dissolving techniques were especially developed for the Redox process; it is also applicable to any uranium separation processes which may tolerate or need aluminum nitrate in the feed solutions. The thorium solutions obtained by this process (Flowsheet D) may also be used with any thorium separation process which may tolerate or need aluminum nitrate in the feed solutions, with Thorex and Thorox processes being specific examples; the basic aluminum nitrate produced as a separations process chemical is used in some Thorex separations processes.

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FIGURE 1

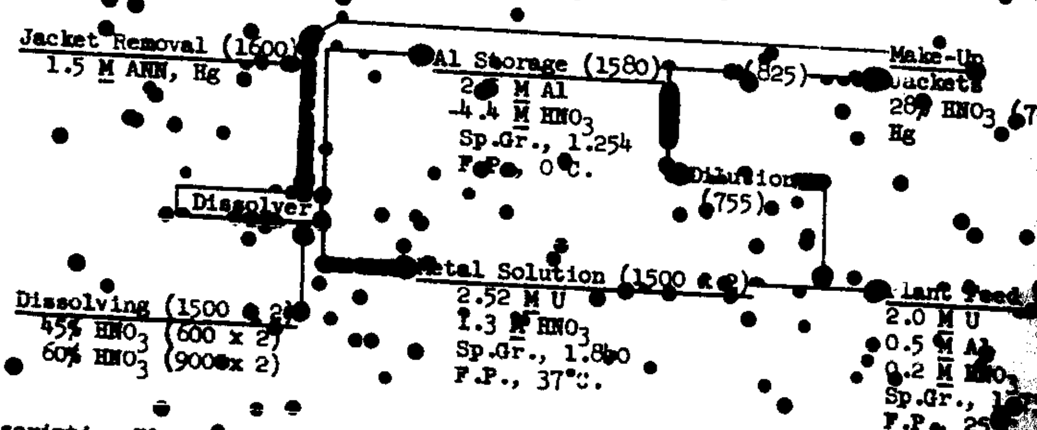
URANIUM DISSOLVING FLOWSHEET "A"

Basis: Dissolving aluminum metal jacket (59 grams) and the uranium equivalent to that of one slug (1778 grams) from a dissolver charge of one jacketed slug and one unjacketed slug. Uranium core of slugs is 4 inches long and 1.385 inches in diameter. The jackets are 25 mils thick around the slug axis, and have end caps 0.185 inches thick.

All volumes are in milliliters, and are enclosed in parentheses.

Catalyst: $Hg(NO_3)_2 \cdot H_2O$, 5% by weight of initial aluminum metal present.

Schematic Flowsheet:



Descriptive Flowsheet:

a. Coating Removal - The aluminum jackets are dissolved by the catalytic action of 5% mercury in aluminum nitrate solution at boiling temperature for three hours resulting in a solution of basic aluminum nitrate. This coating removal solution is then used to 1) neutralize and dilute the subsequent uranium cuts, and 2) provide aluminum nitrate solution for the next jacket removal.

b. Dissolving Cuts - Uranium dissolving is begun by covering the bare slugs with 45% nitric acid, heating to boiling, and making incremental addition of 60% nitric acid to maintain a constant reaction rate. The reaction is terminated at a higher free-acid concentration (ca. 1.5 M) resulting in a shorter dissolving time than previous flowsheets. Two uranium cuts are required, six and one-half hours per cut.

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FIGURE 2

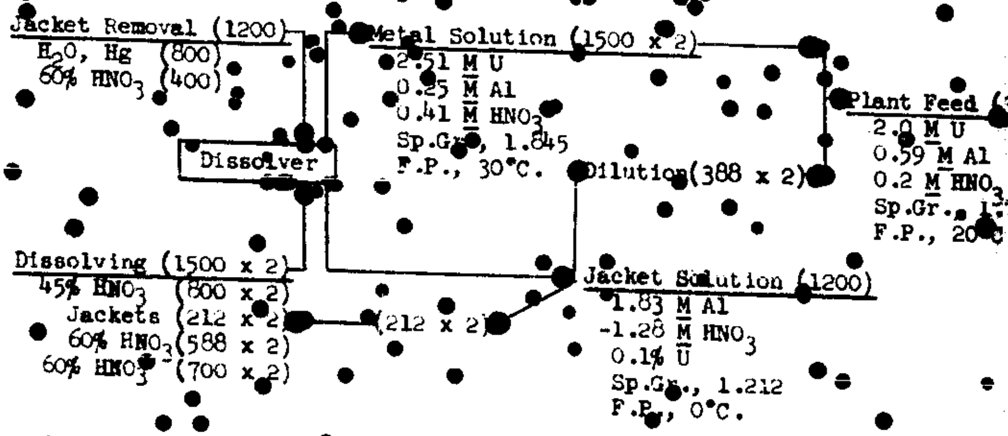
URANIUM DISSOLVING FLOWSHEET "B"

Basis: Dissolving aluminum metal jacket (59 grams) and the uranium equivalent to that of one slug (1776 grams) from a dissolver charge of one jacketed slug and one unjacketed slug. Uranium core of slugs is 4 inches long and 1.385 inches in diameter. The jackets are 25 mils thick around the slug axis, and have end caps of 0.185 inches thick.

All volumes are in milliliters, and are enclosed in parentheses.

Catalyst: $Hg(NO_3)_2 \cdot H_2O$ 5% by weight of initial aluminum metal present.

Schematic Flowsheet:



Descriptive Flowsheet:

a. Coating Removal - Aluminum jackets are removed by slowly adding 60% HNO_3 to the boiling water (containing 5% mercury catalyst) in which the slugs are immersed. The reaction takes three hours. The coating removal solution is used for dilution of the dissolving cuts, and for dilution of dissolving acid.

b. Dissolving Cuts - Uranium dissolving is begun by covering the bare slugs with 45% nitric acid, heating to boiling, and making incremental addition of 60% nitric acid to maintain a constant reaction rate. Dilute nitric acid is made up by using the coating removal solution. Dissolving time is eight hours per cut (2 cuts).

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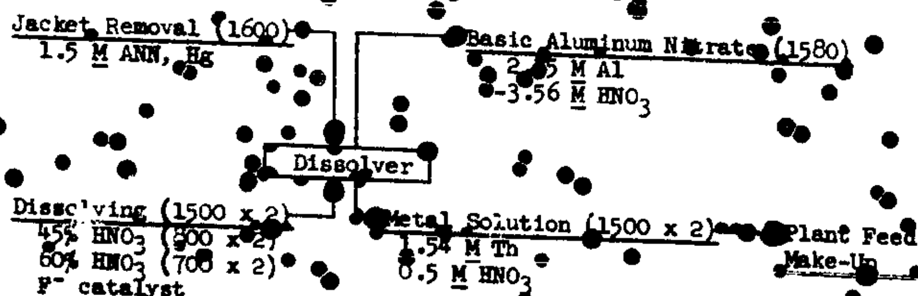
FIGURE 3

THORIUM DISSOLVING FLOWSHEET "C"

Basis: Dissolving aluminum metal jacket (59 grams) and the thorium equivalent to that of one slug (1075 grams) from a dissolver charge of one jacketed slug and one unjacketed slug. Thorium core of slugs is 4 inches long and 1.385 inches in diameter. The jacket is about 25 mils thick around the slug axis and has end caps about 0.185 inches thick.
All volumes are in milliliters, and are enclosed in parentheses.

Catalyst: $Hg(NO_3)_2 \cdot H_2O$, 5% by weight of initial aluminum metal present (for jacket removal, only)
NaF, 0.03 M in acid used for dissolving.

Schematic Flowsheet:



Descriptive Flowsheet:

a. Coating Removal - The aluminum jackets are dissolved by the catalytic action of 5% mercury in aluminum nitrate solution at boiling temperature for three hours, resulting in a solution of basic aluminum nitrate, which is used as a process chemical.

b. Dissolving Cuts - Two dissolving cuts are made, using fluoride ion as a catalyst. Thorium dissolving is begun by covering the bare slugs with 45% nitric acid, heating to boiling, and making incremental addition of 60% nitric acid to maintain a constant reaction rate. Dissolving is eight hours per cut (2 cuts).

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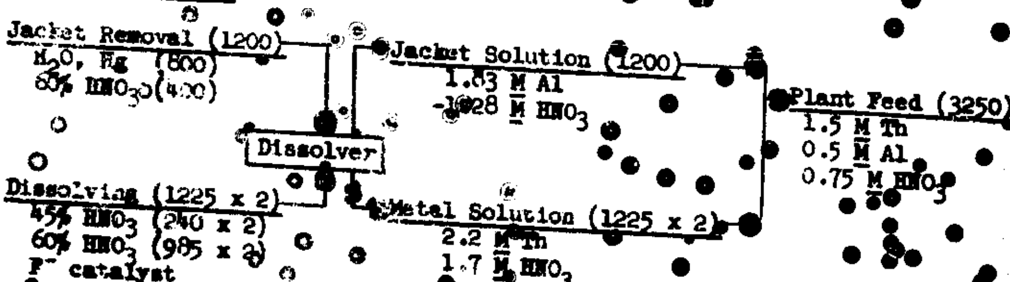
FIGURE 4

THORIUM DISSOLVING FLOWSHEET "D"

Basis. Dissolving aluminum metal jacket (59 grams) and the thorium equivalent to that of one slug (1075 grams) from a dissolver charge of one jacketed slug and one unjacketed slug. Thorium core of slugs is 4 inches long and 1.385 inches in diameter. The jacket is about 25 mils thick around the slug axis and has end caps about 0.185 inches thick.
All volumes are in milliliters, and are enclosed in parentheses.

Catalyst: $Hg(NO_3)_2 \cdot H_2O$, 5% by weight of initial aluminum metal present (for jacket removal, only).
NaF, 0.03 M in acid used for dissolving.

Schematic Flowsheet:



Descriptive Flowsheet:

a. Coating Removal - Aluminum jackets are removed by slowly adding 60% HNO_3 to the boiling water (containing 5% mercury catalyst) in which the slugs are immersed. This solution is used to dilute the feed.

b. Dissolving Cuts - Thorium dissolving is aided by the use of fluoride catalyst. Thorium dissolving is begun by covering the bare slugs with 45% nitric acid, heating to boiling, and making incremental addition of 60% nitric acid to maintain a constant reaction rate. Dissolving time is six and one-half hours per cut (2 cuts).

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