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for Plutonium and Uranium

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EVALUATION OF THE PENTAETHER AND CHELATE PROCESSES FOR THE
CHEMICAL SEPARATION OF PLUTONIUM AND URANIUM

T. E. Hicks and H. W. Crandall

September 18, 1950

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CHEMICAL SEPARATION OF PLUTONIUM AND URANIUM

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A summary of the work done at the University of California Radiation Laboratory on the recovery and decontamination of uranium using pentaether and the recovery and decontamination of plutonium using ~~TAA~~^{thenoyltrifluoroacetone} is presented and a basic flowsheet proposed. The advantages and disadvantages of the processes are discussed and a program and schedule of future work is presented.

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For the past five years considerable research at the University of California Radiation Laboratory has been directed toward the development of processes designed to recover plutonium and uranium from neutron-irradiated uranium. This report has been prepared in answer to requests for information by interested workers in the Atomic Energy program. The material discussed herein represents the efforts of a sizable research group over several years, and summarizes both the current status of the investigation and the future program now contemplated.

I. INTRODUCTION

The objectives of the chemical separations program assume that the feed material to be processed is neutron-irradiated natural uranium metal and are threefold:

- (1) To separate plutonium in high yield and in sufficiently pure form from uranium and associated fission products,
- (2) To separate uranium in high yield and in sufficiently pure form from plutonium and fission products,
- (3) To concentrate the fission products for economical disposal with little or no non-volatile added materials.

To achieve these objectives, two processes have been considered: The pentaether process which is designed to separate uranium, and the chelte process

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which is designed to separate plutonium. These processes can be used in a variety of combinations and permutations to achieve the above objectives, and it is not yet completely clear what particular combination may be optimum. Therefore a typical flow sheet, representing a major possibility, is described in Sections II and III of this report, following which some alternatives are discussed.

The pentaether process¹ is a solvent extraction process for recovery of uranium in which dibutoxytetraethyleneglycol (pentaether) is the water insoluble extractant. This solvent is a higher homolog of dibutylcarbitol, the solvent used in the British butex process, and has been used by various workers in the AEC program. The generally unfavorable results obtained in the early use of this solvent by others are believed to have originated from impurities in the solvent; for many of its most critical properties, such as stability to acid, and extraction of trace quantities of fission products, are markedly dependent upon its purity. Accordingly, the pentaether process as developed in this study has resulted from a critical re-evaluation of pentaether in highly purified form.² Adequate purity was obtained by vacuum distillation. It has been demonstrated on a milliliter scale, in countercurrent batch studies so designed as to simulate continuous operation, that near-theoretical recovery of uranium with adequate decontamination from fission activity (with the exception of ruthenium) is obtained starting with Hanford dissolver solution.³⁻⁶ Considerable progress has been made in continuous operation of this process in pulse columns on 1-2% full Hanford scale,⁷ and, within the available range of operating variables, it appears quite likely to be adaptable to full scale continuous contacting equipment.

The chelate process⁸⁻¹² is a solvent extraction process which uses thenoyltrifluoroacetone, TTA, as the chelating agent, dissolved in an aromatic

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solvent. This process will selectively extract plutonium from aqueous solutions of irradiated uranium metal or from aqueous fission product solutions. The chemical principles underlying this process are understood rather well, for the same solvent extraction system has seen widespread use throughout the AEC program. In a batch process on a liter scale, it has been demonstrated that nearly theoretical recoveries of plutonium can be obtained along with adequate decontamination from fission products.^{8,9} Considerable progress has been made toward developing a continuous process, and some process features have been demonstrated on 1-2~~5~~ scale in a continuously-operated horizontal extractor.¹⁰⁻¹² Whereas benzene was used as the aromatic solvent in most of this work, heavier aromatic solvents of lower vapor pressure and higher flash points have proved equally satisfactory.³

It is believed that certain combinations of these two processes will accomplish the objectives noted above and have great potential advantages over older processes such as the bismuth phosphate and redox processes; further it is believed that they are potentially competitive with other processes of more recent conception.

II. DESCRIPTION OF THE PROCESSES

A. The Pentaether Process

During the latter part of 1949 a number of solvents for uranium were investigated in an effort to locate one that would possess superior extraction properties in the absence of non-volatile salting agents.^{13*} The project literature abounds in such studies; however, interpretation of the earlier surveys is clouded by the fact that most of them were performed with relatively high salt concentrations present. From our recent survey several promising leads developed, the best of

*The report cited contains references to earlier studies.

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which was pentaether. Equilibria for the system, uranyl nitrate-nitric acid-pentaether-water,¹⁴ studies of pentaether purity and purification,² and other pertinent considerations indicated that this solvent has excellent potentialities for use in a solvent extraction process for purification of uranium.

From a study of the equilibria involved, a schematic process flowsheet as shown in Fig. 1 was adopted for study. A solution of irradiated uranium containing fission products (and plutonium unless it is removed prior to the use of the pentaether process) is center-fed to a continuous countercurrent multistage contactor where the uranium is extracted by pentaether and scrubbed with an aqueous solution. The pentaether-uranium solution from the first contactor is stripped in a second contactor, leaving the pentaether ready to recycle through the process. Although excellent decontamination is obtained in a single cycle, the uranium product could if necessary be treated by a second cycle similar to the one described.

Plutonium would be removed in the aqueous nitric acid raffinate from the first column, in conjunction with the fission products. The final separation of plutonium is not anticipated to cause difficulty, and it is likely that a variety of procedures could be used, e.g., modifications of the purex process plutonium cycle, precipitation of the peroxide, or use of a modified chelate process.

B. The Chelate Process

In 1944 a series of β -diketones were synthesized culminating in the compound thenoyltrifluoroacetone, TTA, which possesses to a high degree the ability to extract metal ions into aromatic solvents.¹⁵ Further study demonstrated that this extraction could be made highly selective, and it was therefore considered for the separation of plutonium from neutron-irradiated uranium.⁸ The following

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two-phase equilibrium was established:

where HK represents TTA and the subscripts indicate the phase. Similar equilibria have been established for most metallic ions in the periodic table,¹⁶ and TTA has found wide application in various AEC laboratories. A batch process using TTA dissolved in benzene was developed^{8,9} and certain features have been tested in continuous operation.¹⁰⁻¹²

The chelate process, as presently conceived, is illustrated in Fig. 1. The solution of irradiated uranium containing plutonium and fission products is treated to obtain and stabilize the +4 oxidation state of plutonium, and is then center-fed into the first of two continuous countercurrent multistage contactors. There it is treated with a solution of TTA in secondary butyl benzene which extracts plutonium into the organic phase. The organic phase is then scrubbed with nitric acid to remove those fission products which are partially extracted with plutonium. The TTA extract from the first contactor is then center-fed into a second contactor, where plutonium is stripped from the organic phase by an aqueous nitric acid phase which in turn is scrubbed free of traces of fission products by a fresh TTA stream.

Uranium and the large bulk of fission products remain in the first aqueous raffinate. The composition of this stream is such that without significant further treatment the uranium can be separated from all fission activities except ruthenium using the pentaether process, or a modified TBP process.

It should be noted that no significant amounts of non-volatile materials have been added in either of the processes considered.

In the following sections further information is presented to substantiate the claims made for these processes described above.

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III. DESCRIPTION OF BASIC FLOWSHEET

The possible permutations and combinations of these processes within the range of variables available are numerous, therefore in the interests of simplicity and clarity, a basic flowsheet is considered. Subsequently, attention is given to the variations possible. Two major permutations of the pentaether and the chelate processes are possible:⁴

(1) Separation of plutonium by the chelate process, followed by separation of uranium by the pentaether process.

(2) Separation of uranium by the pentaether process, followed by separation of plutonium by the chelate process.

The former permutation has been chosen as the basic flowsheet and is diagramed in Fig. 1. The former permutation is preferable in that the concentrations of both TTA and nitric acid are smaller in the chelate section when it is used. It should be noted that the pentaether process is essentially unchanged irrespective of the order of its use, and that in either case it would be necessary to remove ruthenium by either pre-treatment or by post-treatment.²⁻⁴

One thousand kilograms of irradiated uranium would be dissolved to make 480 gallons of solution. This solution would be approximately 2.3M in uranium and 0.1M in nitric acid and would be processed according to the basic flowsheet shown in Fig. 1. The volumes and compositions of the various streams are listed in Table I.

The method by which the volumes and compositions were calculated has been described in detail.¹² The values given below are representative, but subject to variation within certain limits, the product of distribution coefficient and volume ratio being the determining factor. Since the distribution coefficient may be varied, within limits, the volume ratio may likewise vary. This is especially notable in the chelate process.

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Table I

Volumes and Concentrations of Streams of Basic Flowsheet
Based on 1000 kg. of Uranium Metal Feed

Stream	Solution	Vol (gal.)	U(M)	HNO ₃ (M)	TTA(M)	Remarks
IF	Aqueous	480	2.3	0.1	0	(15 lbs. added solids)
IX	Sec-butyl-benzene	120	0	0	0.07	(16 lbs. TTA)
IS	Aqueous	50	0	1.0	0	
IIF	Sec-butyl-benzene	120	0	0	0.07	(Pu, Zr)
IIX	Aqueous	12	0	4.0	0	
IIS	Sec-butyl-benzene	5	0	0	0.07	(0.7 lbs. TTA)
* IIW	Sec-butyl-benzene	125	0	0	0.07	(16.7 lbs. TTA, <0.1% Pu)
* IIP	Aqueous	12	0	3.5	<0.001	(benzene satd., decontaminated Pu)
IIIF	Aqueous	530	2.1	0.2	<0.001	(<0.1% Pu)
IIIX	Pentaether	1060	0	2.0	0	
IIIS	Aqueous	220	0	0	0	
* IIIW	Aqueous	650	<0.01	3.2	0	(pentaether satd., 22 lbs. added solids, F.P.'s, <0.1% Pu)
IVF	Pentaether	1130	1.0	0.1	0	(no Pu)
IVX	Aqueous	900	0	0	0	
* IVW	Pentaether	900	<0.01	0	0	(no Pu)
* IVU	Aqueous	1100	1.0	0.1	0	(pentaether satd., decontaminated uranium)

*Effluent Streams.

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In the chelate section, the valence of the plutonium would be fixed in the +4 state by addition of approximately 15 pounds of solids in the form of ferrous sulfamate and sodium dichromate. The product plutonium stream (IIP) would be 12 gallons of 3.5M aqueous nitric acid saturated with secondary butyl benzene and containing decontaminated plutonium.

In the pentaether section, reducing conditions would be obtained in the uranium feed stream by addition of approximately seven pounds of solids in the form of ferrous sulfamate. The product uranium stream (IVU) would be an aqueous solution saturated with pentaether and containing 0.1M nitric acid.

The TTA raffinate stream (IIW) would be scrubbed with 12.5 gallons of 10% sodium oxalate solution, and the TTA partly recycled and partly recovered by a simple steam or vacuum distillation. The pentaether raffinate stream (IVW) would be washed with 90 gallons of 10% sodium hydroxide solution, partly recycled, and partly vacuum distilled. Both the sodium oxalate and sodium hydroxide solutions would be used for several cycles before concentrating and storing.

The aqueous waste stream (IIIW) would be a pentaether-saturated aqueous solution containing the fission products and 22 pounds of added solids. This stream would have a volume of 650 gallons and would be concentrated to a minimum volume and stored.

The basic flowsheet might be changed for various reasons. For example:

(1) The feed to the pentaether section might be made acid-deficient to obtain better decontamination with respect to ruthenium.

(2) The pentaether and chelate sections might be reversed and the uranium removed before the plutonium should this prove to be economic. If this were done, the stream containing the plutonium could be processed directly or evaporated

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to some smaller volume before processing with TTA. Since the solution containing the plutonium has almost no non-volatile materials in it, although it contains almost all of the fission products, any of several processes might be used for separating the plutonium.

It should also be pointed out that the pentaether process would be ideal for a separation such as the "25" process, in which the recovery of plutonium is not necessary. Its possibilities in the "23" process are as yet unknown, although separation of uranium from UX_1 is achieved in the proposed flow scheme.

Wide variations are possible in the flow scheme for the chelate process. The greatest single limitation is the solubility of the plutonium chelate in the organic phase, which is about 0.01M. This represents a minimum requirement of 50 gallons of solvent (IX stream of basic flowsheet) per 1000 kg. of uranium containing 500 g/ton of plutonium, if it were processed with no reflux of plutonium in the column. If a 1:1 reflux ratio is desired, 100 gallons of solvent per ton of uranium are necessary. The flow ratios and plutonium concentration level may be fixed independently at their economic optimum values, because the TTA and hydrogen ion concentrations may still be varied to give optimum separation and recovery in a chosen number of extraction and scrub stages.

IV. STATUS OF RESEARCH PROGRAM

A. The Pentaether Process

Recovery.-- In a countercurrent batch extractor bank utilizing nine stages of extraction, greater than two stages of scrub, and six stages of re-extraction, uranium recovery of greater than 99.9% has been obtained under the proposed flow conditions. It has been shown that the recovery is independent of the number of scrub stages if there are more than two. The work to date on the recovery of uranium using the pentaether process has been carried out in

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countercurrent batch equipment to simulate continuous flow, and in pulse columns packed with either perforated plates or Raschig rings.

Enough data are available to indicate that several changes may be made to improve the recovery of uranium, with relatively slight effect on the decontamination. These changes have the effect of separating the equilibrium and operating lines on a modified McCabe-Thiele plot by increasing the slope of the equilibrium line and decreasing the slope of the operating line, thus attaining a lower uranium loss with the same number of theoretical stages. The slope of these lines may be changed as follows:

(1) The equilibrium line may be made steeper by increasing the nitric acid in the system. This could be done by increasing the nitric acid concentration in any of the entering streams.

(2) The operating line could be flattened by increasing the extractant flow rate or decreasing the scrub flow rate, thus giving a lower aqueous-to-organic ratio in the extracting section of the column.

Increasing the number of theoretical stages in the extraction section would, of course, increase the recovery while having almost no effect on the decontamination.

The parameters chosen in the basic flowsheet, (1) an extractant-to-feed ratio of 2:1, (2) a scrub-to-feed ratio of 1:2, (3) an extractant nitric acid concentration of $2M$, (4) a feed nitric acid concentration of $0.2M$, and (5) seven extraction stages, are all low enough so that a slight increase in any one will give greater uranium recovery, while having little effect on decontamination.

One change which might be made with advantage is a decrease in scrub flow rate. This would reduce the aqueous-to-organic flow ratio in the extracting section of the column and thus give better recovery for the same number of stages, or else permit a lower pentaether flow rate and a correspondingly higher

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uranium concentration in the organic raffinate. Experimental work indicates that this change would have small effect on the decontamination.

If perforated-plate or packed columns are selected as contacting equipment, both the number of theoretical stages in a given length of column and the throughput for a given diameter can be increased by diluting the pentaether with another organic solvent of low viscosity and of either high or low density. This may be essential if packed column operation is desired, since the density differences between pentaether-uranium and aqueous-uranium solutions are small and the viscosity of pentaether-uranium solutions relatively large. Since the uranium-carrying capacity of the pentaether is much greater than required in the proposed scheme, a suitable diluent would produce little reduction in the distribution coefficient but would increase the density difference of the two phases and lower the viscosity of the organic phase. Specifically, di-n-butyl ether is such a diluent; if it is added to the extent of 20% by volume, the distribution coefficient is essentially unchanged, the viscosity is reduced approximately one-half, and the density difference between the mixture and an equilibrated aqueous uranium-nitric acid solution is increased about five times.

Decontamination.-- Decontamination studies on [REDACTED] dissolver solution have been made in countercurrent batch equipment, simulating continuous flows, on a one milliliter scale.² Ruthenium was the only detectable impurity in the uranium product stream. Samples taken from natural uranium runs indicate that separation from UX_1 and UX_2 is complete within experimental error. Product streams from which the residual ruthenium has been removed by distillation are below natural uranium activity level, with ruthenium, UX_1 , and UX_2 as the only detectable activities other than uranium. All decontaminations were obtained with only one cycle, consisting of seven states of extraction, nine stages of

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scrub, and four stages of re-extraction. No activities other than ruthenium were detected in the fifth or higher scrub stages.

A maximum decontamination in the pentaether process may be obtained by adjusting the number of scrub stages, which has no effect on the recovery when there are more than two. Increasing the scrub-to-extractant flow ratio has only slight effect on decontamination, unless it is changed radically from the proposed ratio. Lowering the acidity of the feed or making the scrub alkaline would help the decontamination for a given number of stages while having small effect on recovery. Since present work indicates that even with water scrub and 0.5M acid feed, five scrub stages are all that is necessary, little would be gained by scrubbing with base even though the quantity of added solids could be made small by having both the concentration and scrub flow rate low.

Re-extraction.-- Because of the shape of the equilibrium curve, an extractant-to-feed ratio of almost 1:1 is necessary for the Number IV column. This means that the concentration of the final aqueous product stream depends upon the concentration of the effluent pentaether stream from the Number III column.

Pentaether Stability and Recovery.-- Pentaether shows no visible change when contacted with 8M nitric acid for a period of several months.² The recovery of pentaether by vacuum distillation has been found to be straightforward. Aqueous scrubbing to remove radioactive and other impurities has presented no problems.

Physical Properties.-- In addition to the phase diagram of the system uranium nitrate-nitric acid-pentaether-water,¹⁴ the viscosity and density of the various solutions and the interfacial tension between the equilibrated solutions has been determined. Some studies of the effect of temperature on these properties has been made, as well as of the effect of certain diluents.

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B. The Chelate Process

Recovery.— The recovery of plutonium, including both chelation and de-chelation, has been demonstrated in two center-fed, countercurrent, horizontal glass extractors using five stages of extraction and five stages of scrub in each column.^{11,12} Recovery was greater than 99.9%. The feed solution was Hanford dissolver solution diluted 1000 fold with natural uranium and enriched with pure plutonium to give a concentration and flow rate of about 1% of present Hanford level.

Decontamination.— Decontamination, using the diluted [REDACTED] dissolver solution and equipment indicated above, was limited only by the rate of growth of niobium from zirconium during the time between the runs in the two columns. This time lag was necessitated by the fact that the same column was used as chelation and de-chelation columns on different days. Experimental evidence indicates that ruthenium was completely removed from the plutonium in the Number I column. This agrees with data from batch experiments, which also indicate that plutonium and ruthenium can be separated quantitatively in a continuous flow scheme.

Batch runs on full-level [REDACTED] dissolver solution indicate that less than 0.5% of the zirconium extracts into 0.5M TTA solution in a 24 hour contact.¹⁷ This would indicate that the scrub section of the Number II column would be quite short.

Recovery of TTA and Solvents.— TTA has been recovered by vacuum distillation, steam distillation, and crystallization.¹⁸ All three methods are simple and direct. Solvent recovery has been by simple distillation. Several solvents have been investigated as a carrier for TTA; and a number with relatively high flash points have been found which are satisfactory, including secondary butyl benzene.³

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The large variations in the extractibility of the various elements makes their separation from plutonium relatively easy, even though the molecules which undergo interphase transfer are quite large, and therefore diffuse rather slowly. The rate of transfer is rapid enough, however, to allow many types of contacting equipment to be used in carrying out this separation. The mechanism of this transfer is now well understood under many conditions.¹⁹ Variation in flow rates may be made within any reasonable limit, since the distribution coefficient and the flow ratios may be varied independently. This extent of control over the operating variables leads to a large volume reduction of the plutonium phase, and is thus an important characteristic of the chelate process.

V. RESEARCH PROGRAM AND SCHEDULE

Plans in this laboratory have been made to carry out the recovery and decontamination of uranium and plutonium from synthetic dissolver solution containing full level uranium and plutonium concentrations and 1% fission product activity at feed rates corresponding to ten kilograms of uranium processed per day. The scheme proposed is the one given in Fig. 1, using perforated plate pulse columns for the uranium recovery and mixer-settlers for the plutonium recovery. The choice of contacting equipment was arbitrary, in that a study of pulse columns was thought timely, and mixer-settlers are a useful laboratory tool. The major portion of the work planned is on the chemical-engineering aspects of the separations although much basic chemistry is to be done.

A. Engineering Problems

The Pentaether Process.-- Although recovery and decontamination have been demonstrated in batch contacting equipment, they must still be demonstrated in continuous-flow contacting equipment. The major effort on the pentaether process will therefore be directed toward its demonstration in perforated-plate

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pulse columns. It will be desirable to feed ozonized Hanford dissolver solution, so that ruthenium will not mask the behavior of other fission products. In preparation for a continuous run, [REDACTED] dissolver solution has been ozonized in this laboratory and will be run through the one milliliter batch equipment to determine the limiting activity other than ruthenium.

The Chelate Process.-- Even though the chelate process has been demonstrated in continuous apparatus, a demonstration of it must be made at full Hanford level plutonium concentrations and at an activity level which is as high as the shielding will permit. Two 20-stage stainless steel mixer-settlers will be used for this purpose. Packed column studies will also be made to determine HTU's for this system.

B. Chemical Problems

The Pentaether Process.-- Further studies are planned on the changes of physical properties, i.e., viscosity, density, distribution coefficients, etc., with temperature and with the addition of diluents.

Although Pu(III) does not follow uranium in the pentaether process, the behavior of Pu(IV) and Pu(VI) in pentaether are unknown. It is possible, therefore, that plutonium can be made to follow uranium as in the redox and purex processes. A study of the distribution coefficient of the various oxidization states of plutonium between pentaether and uranium-nitric acid solutions is under way. This study will also include the effect of temperature and diluents on the plutonium distribution coefficient.

The behavior of certain specific fission products in pentaether will be investigated.

The Chelate Process.-- Although much is known of the rate of plutonium extraction with TTA, work is still continuing along this line. The stability

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of Pu(IV) is being studied and methods of holding it in the +4 state are under investigation. The effects of temperature on the rate of plutonium extraction as well as the stability of Pu(IV) are being studied.

C. Program

The program listed above should be completed in about 85 man-months. With the manpower now contemplated, this will take six to nine months.

VI. SUMMARY

The recovery and decontamination of uranium and plutonium by the flow scheme of Fig. 1 has many potentialities, although a large number of data are still lacking. Losses of both uranium and plutonium have been low, and with the exception of ruthenium in the pentaether process, decontamination has been good. The basic chemistry of the chelate system is well known and data on the distribution of uranium between pentaether and aqueous nitric acid solutions have been obtained.

The proposed base flowsheet has the following advantages:

- (1) There is little solid waste.
- (2) The solution volumes are small; this permits small equipment.
- (3) The plutonium solution can be concentrated to a small volume.
- (4) The solvents are sufficiently stable to both nitric acid and to radiation.
- (5) The solvents have relatively high flash points.
- (6) The fundamental chemistry is well understood.

It has the following disadvantages:

- (1) It is a two-solvent process.
- (2) Ruthenium must be removed in a separate step, so far as is now known.
- (3) The viscosity is high and the density difference is low in the pentaether section. These result in high HTU's.

All sections of the proposed flow scheme have been made to operate in some type of continuous-flow multistage contacting equipment.

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BASIC FLOW DIAGRAM

BASED ON 1000 kg URANIUM METAL FEED

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CHELATE SECTION

FIGURE 1

PENTAETHER SECTION