STATUS AND FUTURE REQUIREMENTS FOR THE
URANIUM-233 POWER REACTOR PROGRAM

ASAE-S-4

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SUMMARY

This study reviews the status of thorium and uranium-233 technology for power reactor applications. In the areas of fuel cycle, reactor design and reactor operational problems, information was developed on:

- (1) Current status of technology
- (2) Current and planned research and development programs
- (3) Need for additional research and development, and
- (4) Time schedule of required kilograms of U-233 over the next five years to carry out
  - (a) The research and development now being planned, and
  - (b) The further work believed desirable in the longer term.

It is found that the additional programs needed to permit a reliable evaluation of the U-233 cycle for power reactors are as follows:

FUEL CYCLE

Substantial effort has gone into certain phases of the U-233 fuel cycle, but this effort has been primarily on a small scale for the production of experimental quantities of U-233. At present the production technology of thorium and thorium oxide has reached a stage where its effect on fuel-cycle cost can be estimated fairly well. However, more development is needed in the following aspects of the cycle:

1. Fuel element technology must be extended to ascertain permissible exposure levels for both U-233 and thorium elements. The exposure level has a major effect on U-233 reactor economics.
2. The reprocessing and refabrication of U-233 and of irradiated thorium is probably the area of greatest uncertainty in the development of realistic cost information. Fabrication problems of U-233 and thorium elements need to be studied in relation to the available reprocessing methods to determine the lowest cost cycle. It appears probable that at least semi-remote fabrication must be employed for both U-233 and recycle thorium. Hence, promising low-decontamination schemes such as pyroprocessing need more examination.
3. Reliable cost information should be developed through extended runs of reprocessing pilot plants. The Thorex process has reached a greater state of development than other reprocessing methods. Hence,

the needed technical and economic information should be obtainable for the Thorex process more readily than for other proposed methods.

Operation of the MTR on U-233 for a complete cycle would be an immediately available method of obtaining firm information on U-233 fabrication and reprocessing. There is present capability at Hanford to fabricate U-233 elements for the MTR, and the spent elements can be processed at Oak Ridge or possibly at Idaho Falls. This program is recommended.

REACTOR DESIGN

Several AEC and industrial reactor programs are planned to use the U-233 fuel cycle. For the design of cores for these reactors, data on U-233 cross sections and on η (number of neutrons released per neutron absorbed in the fuel) as a function of energy are believed reliable in the thermal range. However, additional data are needed in the intermediate-energy range (1-10,000 ev).

Because of the lack of experience with U-233 reactors and the difficulty of measuring U-233 cross sections in the intermediate-energy range, a number of critical experiments will probably be necessary in the next five years to verify core calculations.

In addition, there is need for an actual core loading with U-233 to obtain information on the change in gross reactivity as a function of exposure and on the build-up of fission products and U-232. This might be done by operating the MTR on a U-233 loading.

REACTOR OPERATIONAL PROBLEMS

The fission of U-233 (and also of plutonium) results in a markedly lower fraction of delayed neutrons than the fission of U-235. The values most recently published are: U-235 - 0.0064; U-233 - 0.0026; Pu-239 - 0.0021. The low fraction of delayed neutrons for U-233 and Pu-239 will cause control and safety problems, but the seriousness of these problems has not yet been evaluated. Again, operation of the MTR on U-233 would provide valuable information in this respect.

The U-233 requirements necessary to support development (including critical experiments) for reactor programs which have been formally proposed to date are summarized in Table I. The over-all longer-range foreseeable requirements for development of the U-233 power reactor program are tabulated in Table II. These requirements are shown graphically in Figures 1 and 2.

TABLE I
FORECAST OF U²³³ REQUIREMENTS FOR MINIMUM PROGRAM

a. Currently Planned Reactor Projects

<u>Project</u>	<u>Purpose of Requirement</u>	<u>Location</u>	<u>Form</u>	<u>Fiscal Year Requirement (kg)</u>					
				<u>1958</u>	<u>1959</u>	<u>1960</u>	<u>1961</u>	<u>1962</u>	<u>1963</u>
HRE-II	Nuclear Stability Experiment	ORNL	Uranyl sulfate Withdrawals Returns		7.5		5.0		
ATR	Critical Experiment	AI	Uranium Oxide Withdrawals Returns	.050		50.0	50.0		
Con-Ed	Critical Experiment (including .01 kg for basic studies)	B & W	Uranium Oxide (0.01 kg metal foils) Withdrawals Returns	8.0	8.0				
FSR	Critical Experiment	ORNL	Metal Foil Withdrawals Returns			20-70 ^b	20-70		
LMFR	Critical Experiment	BNL	Metal Withdrawals	1 ^a					
ZPR-VI	Critical Experiment	ANL	Metal Withdrawals				25		
MTR	Core Loading (tentative)	IDO	Metal Withdrawals Returns	7.5	6.5				
			Subtotal	17	-7	70	-50	0	0

TABLE I (continued)

b. Current and Planned Basic Physics and Other Research Projects

<u>Location</u>	<u>Purpose of Requirement</u>	<u>Form</u>	<u>Fiscal Year Requirement (kg)</u>					
			<u>1958</u>	<u>1959</u>	<u>1960</u>	<u>1961</u>	<u>1962</u>	<u>1963</u>
BNL	Cross sections and resonance measurements	Metal Withdrawals	0.1 ^a					
MTR	Measurements of η and σ_f and RMF standards	Metal Withdrawals	2	2	2	2	2	2
		Returns		2	2	2	2	2
ORNL	Fuel hazards evaluation (criticality studies)	Uranyl Fluoride Withdrawals	10					
		Returns		10				
ANL	Basic quota, cross section studies	Metal Withdrawals	0.3	1	1	1	1	1
		Returns		1	1	1	1	1
KAPL	Radiation chemistry and cross-section studies	Metal Withdrawals	0.380	0.100	0.100	0.100	0.100	0.100
		Returns	<u>0.080</u>	<u>0.025</u>	<u>0.100</u>	<u>0.100</u>	<u>0.100</u>	<u>0.100</u>
		Subtotal	13	-10	0	0	0	0
		TOTAL	30	-17	70	-50	0	0
		Cumulative	30	13	83	33	33	33

^aCurrently on hand.^bWhere ranges are shown for an entry, the minimum value has been used in computing totals.

TABLE II

FORECAST OF U^{233} REQUIREMENTS FOR LONG-TERM REACTOR PROJECTS

Project	Purpose of Requirement	Location	Form	Fiscal Year Requirements (kg)					
				1958	1959	1960	1961	1962	1963
Advanced gas-cooled, graphite high-temperature reactor	Exponential experiment	UCRL	U Oxide Withdrawals Returns			20			
D ₂ O-U Oxide Reactor	Critical experiment	ANL	U Oxide in ThO ₂ Withdrawals Returns		20-75 ^a				
EBWR	Reactor operations	ANL	U Oxide in ThO ₂ Withdrawals Returns			20		14	
HRE-III	Breeding experiment	ORNL	Uranyl Sulfate Withdrawals Returns					36	
LMFR	Reactor operation	BNL	Metal Withdrawals Returns						50
PAR	Reactor operation	Westinghouse	Uranyl sulfate Withdrawals Returns						385-642
EBR-II	Reactor operation	ANL	Metal Withdrawals Returns			100-200		70-140	
ZPR-III	Fast critical experiment	ANL	Metal Withdrawals Returns			100-200			
Con-Ed	Critical experiment for third core	B&W	U Oxide Withdrawals Returns				100		100

TABLE II (continued)

<u>Project</u>	<u>Purpose of Requirement</u>	<u>Location</u>	<u>Form</u>	<u>Fiscal Year Requirements (kg)</u>					
				<u>1958</u>	<u>1959</u>	<u>1960</u>	<u>1961</u>	<u>1962</u>	<u>1963</u>
FSR	Critical experiment and initial reactor operation	ORNL	U tetrafluoride Withdrawals				100-200		
			Returns						
			Total (net withdrawal)	0	20	240	200	-48	335
			GRAND TOTAL (Tables I and II, net withdrawal)	30	3	310	150	-48	335
			Cumulative outstanding (Tables I and II)	30	33	343	493	445	780

^aWhere ranges are shown for an entry, the minimum value has been used in computing totals.

Figure 1

FORECAST OF U-233 DEMAND TO SUPPORT THE MINIMUM
REACTOR DEVELOPMENT PROGRAM

Figure 2

TOTAL FORESEEABLE U-233 DEMAND TO SUPPORT
THE REACTOR DEVELOPMENT PROGRAM

I. INTRODUCTION

A. STATEMENT OF THE PROBLEM

American-Standard, Atomic Energy Division, has been requested by the Atomic Energy Commission to review the current U-233 program for civilian power reactors in order to develop the following information:

1. Status of the Th - U-233 research and development program, current and planned activities.
2. Evaluation of the program to indicate aspects not sufficiently covered to permit an ensuing critical appraisal of U-233 as a power reactor fuel.
3. Quantities of U-233 and time schedule of requirements necessary to implement the research and development program over the next five years.

B. SCOPE OF THE STUDY

This report is primarily a compilation of up-to-date information on the points listed above. It is intended to provide a basis for predicting U-233 demand for the Power Reactor Program over the next five years and thus to help the scheduling of U-233 production. An important aim is to define the types of information necessary for the critical appraisal of U-233 as an economic reactor fuel and to evaluate the current status of Th - U-233 technology in this light. A review of current and planned research and development programs permits the indication of areas of investigation not adequately covered for purposes of a future comprehensive appraisal.

During the course of this study, a number of detailed technical problems became apparent in connection with various stages of the over-all fuel cycle. These potential areas of investigation are included under the appropriate headings. Any comprehensive research and development program would include a consideration of such problems, especially since it is often difficult to predict their individual and collective effects on the total economic picture.

Information relative to current or planned U-233 production capabilities or quantities of U-233 presently on hand is not included in this report since such data are not necessary for present purposes.

In addition, it should be understood that a critical evaluation of individual projects making up the current research and development program is beyond the scope of this report.

C. APPROACH

The use of U-233 as an economic power reactor fuel can be evaluated only after certain experience has been developed through a well conceived and executed research and development program. The ultimate objectives of this study are to indicate technical areas not sufficiently covered by current programs and to project U-233 requirements on a realistic basis for the next five years. To meet these objectives, criteria were established against which current effort could be compared in the following general areas:

1. Th - U-233 fuel cycle
2. Reactor design
3. Reactor operational problems and limitations

Each area was reviewed with the following questions in mind:

1. What is the status of development in each area with regard to technical and economic information?
2. Are additional improvements clearly needed for economic power reactor applications?
3. What aspects of each area are not being investigated currently?
4. What are the planned research and development programs?
5. What is the U-233 requirement and time schedule over the next five years for current and planned programs as well as necessary additional programs?
6. What are the practical problems associated with the Atomic Energy Commission's furnishing U-233 for research and development to meet near-term demands?

Considerable information has been obtained through direct contact with both Atomic Energy Commission and private industrial groups engaged in reactor design and in various aspects of the Th - U-233 fuel cycle. A review of the current literature including progress reports and proposals for future work by the various groups active in this field has also contributed greatly to this report.

D. INTEREST IN TH - U-233 CYCLE

Power reactors based upon the Th - U-233 fuel cycle have been receiving increased attention over the last few years. There are a number of reasons for this:

1. Thorium supplements uranium as a source material and, therefore, its use will extend the world's supply of potential nuclear fuel. This is expected to become an important consideration within a period of 30 years at the anticipated growth rate of nuclear power capacity.
2. From the standpoint of reactor design, U-233 possesses certain advantages:
  - a. U-233 will provide breeding in thermal and epithermal neutron energy regions, as well as the fast range. U-238-Pu as a potential breeding cycle is restricted to the fast range. Thus, a U-233 breeder offers considerably more flexibility to the designer as to choice of concept. For example, U-233 is the only breeder possibility for an aqueous homogeneous reactor.
  - b. A thermal breeder has an advantage over a fast breeder from the standpoint of reactor safety in that the short neutron lifetime of the fast reactor could lead to a more severe accident than would occur in a thermal reactor under prompt critical conditions.
  - c. The use of thorium metal in fuel elements may permit higher operating temperatures and longer exposures than are possible with uranium metal.
3. U-233 may be a desirable material in dual-purpose reactors if weapons application is found for U-233.
4. Certain foreign countries possess an abundant supply of thorium; for example, India, Canada, and Brazil. It appears to be inevitable that such countries will look to thorium as a means of meeting their future power requirements.

5. Countries with abundant thorium reserves can reduce their foreign exchange drain for purchase of raw materials by using thorium. The potential for the Th - U-233 fuel cycle was discussed at the 1955 Geneva Conference (Volume 1 and Volume 3). It was pointed out by A. M. Weinberg¹ that: "Once a reactor is constructed with natural uranium fuel rods, there will be strong nuclear incentive, after the first few loadings with ordinary uranium, to switch to the U-233 - Th cycle. The strong possibility, even in the heterogeneous system, of making the U-233 - Th thermal system self-sustaining will always be an attractive goal, no matter how plentiful natural uranium may be. Thus, it may not really be relevant to argue whether breeders ought to be developed as soon as possible. It seems to me entirely likely that most heterogeneous power reactors which begin their lives as thermal, natural uranium converters will gradually convert to the U-233 - Th cycle simply because operation with this fuel is probably independent of raw material."

E. SPECIAL CONSIDERATIONS FOR TH - U-233 FUEL CYCLE

In contrast to the U-238 - Pu fuel cycle, the Th - U-233 fuel cycle is complicated by undesirable nuclear side reactions that occur during the irradiation. The most important of these side chains is the result of an (n, 2n) reaction on Th-232:

Th-228 decays through the well-known thorium decay scheme, giving two gamma emitters of greater than 2 mev (see Figure 3). Some portion of the U-232 will remain with the U-233 during reprocessing and will give rise to increasing gamma activity in the recovered U-233. A mixture of U-233 and U-232 which leaves the reprocessing unit with essentially no activity, soon (in about two weeks) possesses sufficient activity as a result of the U-232 decay chain to require a shielded fabrication facility.

By very prompt processing of discharged thorium from a reactor, it may be possible to minimize U-232 decay and hence keep the Th-228 content of the recovered thorium sufficiently low to permit nonremote fabrication

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¹ Geneva Conference Proceedings, Vol. III, page 862.

Figure 3

NUCLEAR REACTIONS OCCURRING IN THE NEUTRON IRRADIATION OF THORIUM

of recycled thorium. However, this complicates the fuel cycle considerably. This will be discussed in the section on processing.

In addition, the $(n, 2n)$ reaction on the U-233 fuel itself must be considered: $\text{U-233} \xrightarrow{n, 2n} \text{U-232}$. This produces a buildup of U-232 within the fuel elements (as distinct from the thorium fertile material) which will proceed until the rate of formation of U-232 becomes equal to the rate of removal by decay and by the $\text{U-232} \xrightarrow{n, \gamma} \text{U-233}$ reaction. The U-232 formed will complicate the processing and refabrication of U-233 fuel elements as described above.

Another disadvantage of the Th - U-233 cycle is the relatively long half-life of Pa-233, the precursor of U-233. The Pa-233 has a half-life of 27 days, whereas Np-239, the precursor of Pu-239, has a half-life of only 2.3 days. This means that the protactinium must be held up for about a year in order to give the same percent loss of product (less than 0.1%) as obtained with about one month holdup of Np-239.

Finally, the Th-234 resulting from the $\text{Th-232} \xrightarrow{n, \gamma} \text{Th-233} \xrightarrow{n, \gamma} \text{Th-234}$ reactions leads to an additional reprocessing problem. The activity due to the $\text{Th-234} \xrightarrow{\beta, \gamma} \text{Pa-234} \xrightarrow{\beta, \gamma} \text{U-234}$ chain can be accommodated by permitting the thorium product to cool for eight months to a year. This obviously increases the inventory and, in addition, the β, γ activity complicates the control of the purification process.

II. TH - U-233 FUEL CYCLE

A. THORIUM AND ITS COMPOUNDS

1. Sources and Availability

The availability of thorium ore is open to considerable question, since no exhaustive evaluation of potential reserves has been made to date as has been done for uranium. However, it is believed by many observers that sufficient thorium is available for a full-scale power program based on a Th - U-233 breeding cycle.

There are a number of known reserves of monazite and other ores. For example, the Blind River, Canada, uranium mines are valued at probably 40-100,000 tons. The Davison Chemical Company of Erwin, Tennessee, feels that native American reserves are adequate to permit them to supply 100,000 tons of thorium over a 20-year period.

E. D. Arnold of Oak Ridge has estimated that if, by the year 2000, about 37% of all reactor fuel requirements were made up through the use of thorium (25% homogeneous reactors, 12% heterogeneous), there would be a total thorium demand of 60,000 tons. This requirement is considerably less than known reserves, and there is little concern about availability for the next 50 years at least. As in the past, it is to be expected that additional reserves will be uncovered as economics and interest dictate.

It appears that a survey of known and potential thorium ore reserves should be carried out to clarify the extent of availability.

2. Production of Nitrate and Oxide

Thorium is extracted primarily from monazite sands at present by an acid leach or an alkaline leach. The thorium is then separated from rare earths and uranium, either by fractional crystallization or by conversion to the nitrate followed by a solvent extraction purification.

Extensive research is being done on the extraction of thorium from the ore and purification of the product. It would not seem that additional work in this field needs to be encouraged at this time.

Following production of the purified thorium nitrate, thorium oxide is produced by precipitating the oxalate and then calcining to the oxide. The thorium oxide may then be used to produce a ceramic element or a slurry or it may be reduced to the metal.

3. Reduction to Metal

At present, only one process has been used for relatively large-scale reduction to the metal. This process involves the conversion of thorium oxide to the fluoride followed by reduction of the fluoride by calcium in the presence of zinc. Other methods show promise, however.

A number of reduction methods for thorium have been taken to various stages of development over the past few years. These are enumerated as follows:

a. Calcium Reduction of the Tetrafluoride - (Ames Process) - Fernald and Ames

This process has been employed on a pilot plant basis and has produced all of the thorium metal used to date for U-233 production. Fernald is currently inactive, all reduction equipment having been diverted to other work.

b. Electrolysis of Thorium Chloride in a Fused Salt Bath of Sodium and Potassium Chloride - Horizons, Inc.

This process was taken to pilot plant stage. Acceptable metal was produced in the form of an electrode which is crushed to give all size particles including some fines. This project is currently inactive.

c. Oxide Reduction with Calcium - Sylvania-Corning and Davison Chemical Company.

This is a potentially good long-range method. A bench top operation was carried out at Sylvania-Corning. A production scale operation is being carried out at Davison. Fine powder of high purity is produced. The fines are compressed into an electrode and melted by vacuum arc melting. This project is no longer active at Sylvania. Davison, on the other hand now produces metal commercially by this method.

d. Tetrachloride Reduction by Magnesium - Ames

This is a very small-scale laboratory process. It is not very far along, but is still in progress.

e. Metallex Process - Oak Ridge

This process involves direct thermal decomposition of the nitrate to the oxide, followed by conversion of the oxide to the tetrachloride. ThCl_4 is then reduced to the metal by sodium amalgam. This pilot plant scale operation is scheduled for termination in the near future.

Continued work on thorium metal and oxide production aimed at lower cost and uniform quality is definitely required.

4. Cost of Thorium

Thorium ore is generally quoted at \$300 per short ton. At an average concentration of about 6% thorium, the cost of contained thorium is about \$3/lb. This assumes that no value is given to rare earth and uranium content of the tailings.

The cost of producing thorium metal or oxide by a commercial process is not very well established at present. Factors such as profits from rare earth content, process used, methods of cost accounting, and the like greatly affect cost estimates. Davison Chemical Company and the Institute for Atomic Studies at Ames, Iowa, provided the best available cost information on processes capable of producing ton quantities.

The production of thorium metal was developed for the Commission at Ames during the early years of the program. For a time Ames was the only producer of thorium, supplying the metal at the rate of about 72 tons per year. The following cost estimates from Ames are based on the process involving reduction of the ThF_4 by calcium (at present, the reduction of ThCl_4 by magnesium is under investigation):

	<u>Dollars per Pound of Thorium</u>
Purchase mantle-grade $\text{Th}(\text{NO}_3)_4 \cdot 4\text{H}_2\text{O}$ from Lindsay Chemical Company	3.80
Form oxalate and calcine to oxide	0.80
Oxide $\longrightarrow$ Fluoride	0.60
Fluoride $\longrightarrow$ Derby metal (based on calcium at \$2.10/lb of Calcium)	2.80
Derby $\longrightarrow$ metal ingots (induction melted)	<u>2.70</u>
Total cost	10.70

The cost of purified nitrate represents a major uncertainty in the above estimate. Starting with ore at \$3/lb of contained thorium and making no allowance for by-product sales, the cost of producing the purified nitrate using a TBP solvent extraction scheme would be about \$9/lb of thorium. On this basis, Ames cost estimate for thorium metal would be about \$16/lb.

The Davison Chemical Company provided a cost estimate which includes \$3/lb of contained thorium for concentrate obtained from their rare earth extraction plant. This concentrate is purified by a solvent extraction process using TBP in a hydrocarbon diluent. The oxalate is precipitated and calcined to the oxide, giving reactor grade ThO_2 which sells for about \$7.50/lb. The oxide is reduced directly by calcium and metal ingots are produced by arc melting. The metal sells for about \$20/lb.

If there were no profits from rare-earth sales, the Davison concentrates would cost \$4.75/lb of contained thorium instead of the \$3/lb of thorium assumed above. However, it is expected that a large-scale demand for thorium would lower this cost to \$4/lb of contained thorium even without rare-earth profits.

An accurate estimation of the future cost trend for thorium would require a careful analysis of such factors as by-product sales, market, and plant amortization. The complexity of the picture is demonstrated by the information on rare-earth sales obtained from the Ames staff. For example, there are about 45 pounds of rare-earth nitrate tetrahydrate produced per pound of thorium. These sell for 75 cents per pound with essentially no extra handling resulting in a rare-earth profit of about \$35 per pound of thorium. Therefore, even if it costs \$20 per pound to produce the thorium metal, there should be a profit of about \$15 per pound of thorium as a result of the rare-earth sales. The importance of the future market for rare-earths to the thorium cost trend is apparent.

Work in progress at Oak Ridge, Ames, Batelle, and the Davison Chemical Company on more efficient means of extracting thorium from the ore and purifying it should also provide more definite cost data. In addition, metal reduction processes such as Metallex are being studied in order to find a cheaper means of producing the metal.

Recent estimates by Oak Ridge indicate that thorium metal can be produced by the Metallex process - sodium amalgam reduction of ThCl_4 - for a cost of \$3-5/lb for the steps from the purified nitrate to the final arc-melted metal. These costs include plant amortization at 20% annual depreciation.

Assuming an adequate ore availability, it is difficult to become concerned over thorium costs. Certainly \$10-20/lb of thorium metal would be considered acceptable since the effect on over-all fuel-cycle economics appears to be negligible when compared to other potential cost factors.

5. Physical Metallurgy of Thorium and Its Alloys

The metal thorium has not been investigated as intensively as uranium. The current status of thorium metallurgy is well covered in papers given at the 1955 Geneva Conference¹ and subsequent articles based upon these papers. Little additional information has been developed since that time. Physical properties of thorium are not accurately known, especially those properties of engineering interest.

The metal has never been prepared in the high-purity form; thus, the value of any property subject to alteration by impurities is somewhat questionable. This accounts for the poor state of knowledge of the strength of thorium and its alloys. For example, reported values of room temperature tensile strength vary from 22,000 to 40,000 psi depending upon the method of fabrication and upon the purity. Since mechanical strength is an important factor in high-temperature irradiations, considerable effort is indicated in this direction.

An excellent survey of the present state of knowledge of the binary constitution diagrams of thorium has been published by Saller and Rough.² Only three systems (zirconium, calcium, and cerium) have been reported to exhibit extensive mutual solubilities. The transformation kinetics of these systems are not reported. The oxygen, carbon, and uranium systems are under investigation at present.

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¹ Geneva Conference Proceedings, Vol. 9, page 556.

² Saller, H. A. and Rough, F. A., Compilation of U. S. and U. K. Uranium and Thorium Constitution Diagrams, BMI-1000, June 1955

The ternary systems of thorium-uranium have received only cursory attention. One-region breeder fuels utilizing thorium will be based upon a low uranium composition; therefore, this area is deserving of a thorough metallurgical study.

6. Thorium - Fabrication of Fertile Elements

Fabrication of fertile elements has been very limited as compared to uranium, the chief activity having been in the production of metal slugs for Hanford and Savannah River. Most of the current experience in metal fabrication has been summarized by J. Cunningham of Oak Ridge.¹ Indications are that, provided material of adequate purity has been produced, thorium can be fabricated by most conventional processes. However, due to the experimental nature of the work to date, no reliable cost estimates are available. A number of areas appear to require effort from the technical and economic standpoints.

- a. It is well known that certain impurities or minor constituents can markedly affect the mechanical behavior of thorium. It is important to ascertain the effect of these trace elements on fabricability as a function of concentration to develop a realistic specification for maximum permissible impurity content.
- b. Impurities arise chiefly in processing, metal reduction, and melting steps. Accordingly, these production steps should be carefully studied with a view toward minimizing impurity pickup.
- c. The melting and casting of thorium is the source of internal and surface defects such as laps, seams, shrinkage cavities, and hot tears. These undesirable ingot characteristics directly influence fabricability. Therefore, the melting and casting operation should be investigated so as to develop methods that minimize such defects.
- d. Extrusion appears to be a particularly attractive method of fabricating thorium shapes. However, past experience has shown that die life is very short because of seizure and abrasion. Die materials and thorium purity (from abrasive constituents) should be investigated to minimize fabrication costs and improve product quality.

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¹Thorium, ASM, to be published.

- e. A correlation of the effects of various alloying elements on the mechanical working properties of thorium is very much needed.

The specifications for thorium metal to be used for fabrication are based on both activity content and chemical analysis. Effects of impurities on the fabricability of thorium are known to some extent.¹ The oxide content has a marked effect on fabricability and the tendency to corrode. Carbon and nitrogen increase the hardness markedly and, therefore, interfere with fabrication. Nickel, iron, chromium, and copper, all of which come from process equipment, form low melting eutectics which in a cast ingot segregate at grain boundaries. Specifications for reactor grade thorium are shown in Table III.

Detailed specifications for the permitted activity of various isotopes or chains may be found in ORNL 2056.² The over-all activity specification for direct refabrication of thorium metal is that the activity shall be no greater than 0.332 r/h at the surface of a thorium slug (1.36 in. x 6 in.).

Oxide pellets for fuel element applications have been produced at Argonne and Davison. The technology appears to be in relatively good shape, and cost estimation probably can be done satisfactorily.

Very little large-scale effort has been required to date; thus the economics of thorium fuel element fabrication are difficult to estimate accurately. In the case of the oxide, some of this information will be developed as current projects advance.

7. Thorium Fabrication Facilities - Present and Planned

The fabrication of thorium metal into various shapes for reactor use has been carried out by several industrial plants in the past. No apparent problem exists in obtaining a limited quantity of thorium blanket elements or fuel elements for a near-term irradiation program similar to that carried out at Savannah River. However, no production-scale thorium fabrication plant exists, and cost data obtained through any short-term program with present facilities would provide little information relative to fabrication economics.

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¹TH - U-233 Information Meeting - Wilmington, August 3, 1955, Sponsored by the AEC Production Division, TID-7521.

²Thorex Thorium Nitrate Product Specifications, Arnold, E. D., and Wischow, R. P., ORNL 2056, May, 1956.

TABLE III

Chemical and Physical Specifications for Thorium¹

Oxide	Desired (max)	Typical Ames Metal	Fernald Metal
	1%	1.25% (av)	<1.5% (av)
C	400 ppm	300-800 ppm	<300 ppm
N	150 ppm	85-155 ppm	<300 ppm
Al	25 ppm	<25 ppm	<200 ppm
Ca	70 ppm	<70 ppm	<100 ppm
Mg	20 ppm	<20 ppm	< 20 ppm
Si	50 ppm	<50 ppm	< 50 ppm
Fe	100 ppm	35-115 ppm	<500 ppm
Ni	50 ppm	--	<500 ppm
Cr	50 ppm	--	<200 ppm
Cu	50 ppm	--	<200 ppm
Zn	100 ppm	< 20 ppm	<500 ppm
B	1 ppm	< 1 ppm	< 1 ppm
Cd	0.5 ppm	< 0.5 ppm	<0.5 ppm
Gd	0.2 ppm		< 5 ppm
Sm	0.1 ppm		< 2 ppm
Dy	0.5 ppm		< 2 ppm
Eu	0.5 ppm		< 2 ppm
U	25 ppm		< 10 ppm

Density 11.60 min

Hardness 60 BHN (500 kg load) max for ingots.

Soundness - Free from casting and fabrication defects.

The Ames material was induction melted.

The Fernald material was double-arc melted.

¹TH - U-233 Information Meeting - Wilmington, August 3, 1955, Sponsored by the AEC Production Division, TID-7521.

An extensive fabrication plant for thorium is not planned, nor would one be justified until the Th - U-233 fuel cycle is on a sounder footing.

Some current possibilities for thorium metal fabrication based upon past experience are as follows:

- a. Rolled sheets - Simonds Saw and Steel Company
- b. Extruded rods - Bridgeport Brass Company,
Nuclear Metals, Inc.
- c. Co-extruded tubes (Al clad) - Nuclear Metals, Inc.
- d. Canned Slugs (Al can) - Sylvania-Corning Nuclear Corporation

In addition, Oak Ridge has fabricated zirconium-clad thorium plates in the past. Argonne possesses extensive facilities for fabricating Zircaloy-clad uranium plates, and these facilities could probably be used to produce a similar product out of thorium. Castings could be produced at a number of installations including Davison Chemical Company and Ames.

Some developmental effort on the fabrication of thorium as metal and oxide is currently in progress at Babcock & Wilcox's Alliance, Ohio, plant. In addition, Babcock & Wilcox is supporting some work at Battelle on ThO_2 - UO_2 (U-235) fuel elements and plans to irradiate samples in the Babcock & Wilcox pressurized water loop in the MTR.

Although facilities exist to fabricate thorium elements, it should be pointed out that the technology has not reached a level comparable to that of uranium and, therefore, a development program will generally be required for all but the simplest elements, namely, canned slugs.

Oxide fabrication is probably in better shape currently than metal fabrication. This stems from the great current interest in ceramic-type fuels. Argonne has recently produced a loading of $\text{ThO}_2 + \text{UO}_2$ for the Borax IV reactor, and facilities there should be available. Oak Ridge has produced ThO_2 to nearly 100% of theoretical density. Commercially, Davison Chemical is accepting orders for thorium pellets and American Lava Company is producing thorium-uranium compacts of various types. The similarity of thorium fuel elements to currently conceived or produced uranium fuel elements indicates that little difficulty should be encountered in finding facilities to assemble pellets into some useful clad form.

8. Thorium Irradiation Stability

Experience concerning the behavior of thorium metal under irradiation has been accumulated chiefly through the AEC's U-233 production program. Some additional exposures have been carried out in the MTR.

Thorium metal slugs, produced at Fernald, have been irradiated at Savannah River on a two-year experimental program which has recently been completed. No radiation damage problems or slug failures were encountered with the target elements under conditions at Savannah River. The thorium slugs were bonded to aluminum cans at Sylvania by the hot pressing method and the aluminum caps were then welded at Savannah River.

Hanford has irradiated thorium metal and has experienced several slug failures, presumably as a result of water entering at defective welds. Hanford employed an unbonded aluminum canned slug in its irradiations.

Thorium metal slugs have been irradiated at the MTR under a program initiated during the first year of MTR operation and continued until June, 1954. Maximum exposure appears to be about 7000 g/T with no radiation damage observed. Thorium metal has been reported to be very resistant to dimensional change under irradiation exposures of greater than 10^{21} nvt.

More thorium is currently being irradiated in the MTR for physics studies in which 150-200 gram capsules of thorium will be irradiated for long periods up to five years and reactivity gains measured.

Knolls Atomic Power measured one Hanford-size thorium slug after a 4000 g/T irradiation at the MTR and found only a 0.2% change in the diameter.

Atomics International is planning the use of Th - U alloys as a second core for the SRE and is currently carrying out some irradiation tests in the MTR. Preliminary tests on 3/8-inch diameter rods were reported to have resulted in 4% swelling after 3000 MWD/T exposure at 1200°F central temperature.

The chief experience with ThO_2 as a potential reactor fuel is currently being accumulated at Argonne's Borax IV reactor, which employs approximately 6.5% UO_2 - ThO_2 cold pressed and sintered pellets

clad in a 1% nickel-aluminum alloy. Preliminary irradiation experiments carried out in the MTR on compacts of this type have been described.¹ Specimens were irradiated from 0.34 to 1.25% total atom burnup. Surface heat flux was estimated to be 27×10^4 to 102×10^4 BTU/hr-sq ft, and central temperatures were estimated to range from 1060-3700°C (melting). Dimensional changes were slight where central temperatures were estimated to be 2000°C or less, although some fragmentation had occurred. Specimens irradiated to 1.25% burnup appeared to fail in two ways:

- a. In the tests in which the annulus was filled with lead for thermal bonding, the failure appeared to be due to gas pressure with subsequent hoop tensile stress fracture.
- b. In the tests in which the annulus was filled with gas, the failure appeared to result from melting the cladding.

It appears, therefore, that an extensive program of irradiation experimentation on both metal and oxide fuel element types will be necessary before maximum permissible exposures can be estimated for given power reactor conditions. Most of the available experience with thorium metal has been obtained through the U-233 production program where fuel burnup is very small and temperatures are low. It would be desirable to obtain considerably more information on the behavior of thorium under power reactor conditions. Dimensional instability unaccompanied by density changes is generally not expected to be an important part of radiation damage for thorium, which is isotropic and well behaved metallurgically. The same cannot be said for instability with accompanying density change (swelling) resulting from fission product buildup in the metal. In addition, if thorium is to be employed at relatively high temperatures (around 800°C) in a reactor, considerably more effort aimed at improving high temperature mechanical properties through suitable alloying is required. The effect of irradiation on such alloys containing fissionable material must be thoroughly investigated so that upper limits of temperature and exposure can be ascertained.

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¹Chemical Engineering Progress, Vol. 53, No. 2.

The same situation holds for oxide-type elements. The many variables associated with employing such fuels in a power reactor need to be carefully investigated at an early date if the adequacy of this type of fuel is to be established. In the light of experimental work carried out to date, it is clear that oxide elements are not without limitations; such limitations must be studied and measured for satisfactory prediction of fuel element life under given reactor conditions. In addition, the effect of irradiation upon important physical properties such as thermal conductivity as a function of temperature should be determined for more accurate analyses of reactor kinetics.

Slurries of ThO_2 have proven to be excellent for use as fertile blanket material. There is no phase transformation of the ThO_2 at elevated temperatures. ThO_2 slurries show no crystal growth and they pump easily. They are readily dispersible, although they do have a tendency to cake in the presence of some impurities.

B. PROCESSING OF TH - U-233 MIXTURES

The mode of production of U-233 from thorium, together with the fact that the U-232 decay chain passes initially through the Th-228 daughter, greatly complicates the processing of the Th - U-233 mixture.

About 95% of the U-232 formed in the thorium blanket is the product of the Th-232 ($n, 2n$) Th-231 reaction. The bulk of the remaining U-232 is formed by the U-233 ($n, 2n$) U-232 reaction and a very small portion arises from the Pa-233 ($n, 2n$) Pa-232 step. Since these ($n, 2n$) reactions are threshold reactions (about 6-1/2 mev), it is obvious that the reactor and reactor loading will influence the ratio of U-232 to U-233 formed. In addition, the fraction of U-232 increases with time. Thus, both the ratio of fast to thermal flux and the extent of irradiation will affect the ratio of U-232 to U-233.

Processing is further complicated by the decay of Th-234 through Pa-234 to give U-234. The Pa-234 decay yields high energy β and γ activity. The Th-234 chain dies out rapidly ($T_{1/2} = 24$ days); however, some delay is required before the thorium can be fabricated (about 240 days). The β, γ activity of the Th-234 and Pa-234 makes it difficult to monitor the purification process for quality control.

It is generally assumed that metal which contains up to 10^9 dis/min/kg may be handled without shielding for radiation protection. The decontamination factor for irradiated thorium required to reduce the fission product activity to this level after ten days' cooling may be calculated from the following equation:¹

$$D.F. = \frac{0.25 (GT)^{1.893}}{\left[\frac{1.5 \times 10^{13}}{\phi} \right]^{1/2} \text{ or } \left[\frac{\phi}{1.5 \times 10^{13}} \right]^{1/2}}$$

$\phi < 1.5 \times 10^{13}$ $\phi > 1.5 \times 10^{13}$

$\phi = n/\text{cm}^2\text{-sec}$

GT = irradiation level in gms U-233 + Pa-233 per ton of thorium

1. Separations Processes

a. Thorex (see Figure 4)

The Thorex process involves liquid-liquid extraction in pulsed columns, starting with a nitrate aqueous phase and using TBP (tributyl phosphate) in a hydrocarbon diluent as the organic extractant. In the first cycle, the thorium and uranium are extracted into the organic phase while the protoactinium remains with the fission products in the aqueous stream. The thorium and uranium are then stripped back into an aqueous stream and thence go to an evaporator for concentration. In the second cycle the thorium-uranium aqueous feed goes to another extracting column where the thorium and uranium are again extracted into the organic phase. This phase is then sent to a partitioning column where the thorium is taken into the aqueous phase while the uranium leaves in the organic phase. The aqueous stream is evaporated to recover thorium. The organic phase carries the uranium to a stripping column where the uranium is taken into the aqueous phase. From there, the uranium goes to an ion exchange column which gives further purification and concentration.

This process has received the most consideration of the potential Th - U-233 processing schemes but is still in the developmental stage.

¹Progress in Nuclear Energy, Vol. III, page 175.

Figure 4

THOREX CO-DECONTAMINATION FLOWSHEET

Either a long or short cooling period may precede Thorex processing. These two routes are demonstrated graphically in Figure 5. In the first case (long cooled process), the irradiated thorium is allowed to cool for eight months to a year, depending upon the U-233 recovery desired. During this time, essentially all of the Pa-233 decays to U-233. The material is then passed through the Thorex process. The U-233 product obtained is relatively free of fission products and if fabricated within two weeks, presents no major handling problem from U-232 decay. Beyond this time limit, the activity growing in from U-232 presents a serious problem. The thorium recovered from the long-cooling process is quite active because it contains the Th-228 chain as a result of U-232 decay. Remote fabrication of the thorium is required.

In order that the activity of the thorium product received from processing shall be low enough to permit direct fabrication, it is necessary to prevent the buildup of Th-228 from the decay of U-232. This requires that the irradiated material be processed after a holdup time of only 30 to 60 days. This short holdup creates processing problems arising from high activity level contributed by Pa-233, Th-234, Pa-234, and fission products. Severe radiation damage to the extracting vehicle may make the process uneconomical.

In the short-cooled Thorex process, thorium and uranium are extracted, leaving the protactinium in the waste stream. It is then necessary to reprocess the waste stream after an appropriate holdup time in order to recover the resultant U-233, which can amount to about 15% of the total U-233 product.

The added cost of processing the short-cooled material is estimated to be about 25%, but this is offset by advantages such as less activity in the thorium metal and smaller U-233 inventories.

The thorium which has been recovered in this manner could be fabricated without shielding after appropriate holdup time for the decay of the Th-234 chain. The gamma activity of recycled thorium metal from the Thorex process would be 0.083 r/hr at the surface of a 1.36 by 6-inch slug when a 60-day pre-Thorex period is used for 2000 g/T material. Infinite recycle of thorium metal through fabri-

FIGURE 5

ALTERNATIVE THOREX ROUTES

A. Long cool scheme - 8 months to one year pre-Thorex cooling

B. Short cool scheme - less than 60 days (preferably 30 days) pre-Thorex cooling

cation, irradiation, and processing would increase the activity by a factor of two or three.

The primary U-233 product recovered from the Thorex process could be handled in a plutonium-type facility if fabrication were carried out within a few weeks after processing. However, the activity due to the U-232 decay chain grows in quite rapidly. Beyond this period of time, the gamma activity becomes severe enough to require shielding. Alternately, an additional purification cycle to remove Th-228 and other decay products is necessary for nonremote fabrication of U-233.

The Thorex process removes the protactinium with the fission products in the first extraction step. Hence, it should be possible to produce essentially isotopically pure U-233 by holding out the fission product fraction which contains the Pa-233, permitting the Pa-233 to decay to U-233, and then processing for U-233. An eight to nine months' holdup time of this stream is required to reduce the U-233 loss to less than 0.1% of the total potential U-233.

No production scale cost information is available for processing thorium blanket material to yield U-233. Oak Ridge estimates that the cost of processing 2000 g/T material to the purified uranyl nitrate is approximately \$8/gm U-233. The cost for 4000 g/T material should be about \$4/gm U-233. Little significance can be attached to these figures since they are based on pilot-plant production. About 20 tons of normal material (3000-4000 g/T) has been processed at Oak Ridge.

The Thorex pilot plant appears to be as big as can be built from criticality considerations. The critical concentration is about 15 gms U-233 per liter in the 12-inch diameter portions of the columns, depending upon impurities present. Oak Ridge is looking into the possibility of increasing the column size by the use of neutron absorbers in solution.

To date, there have been insufficient runs on material of only 30-day holdup time. As a result, there is uncertainty concerning activities to be expected in the products. It is imperative that this problem be resolved, and Oak Ridge presently plans

to carry out two runs on 30-day material. These should be sufficient to demonstrate feasibility. This program would require about 15 tons of irradiated thorium (about 4000 g/T) through the first half of fiscal 1959. Oak Ridge also hopes to study means of improving the process, especially with respect to ruthenium decontamination.

The Thorex group effort may be continued beyond the end of fiscal 1958 as a part of the power reactor processing program now indicated in the AEC Production Division budget. Oak Ridge believes that their present facility is capable of handling any proposed program short of a major weapons effort.

b. Fused Salt and Volatile Fluoride Methods

Brookhaven has done some work in the past on the processing of liquid bismuth alloys by means of fused salts. This system is particularly applicable to processing LMFR fuel for the separation of fission products from U-233. Presumably, the system may be applied also to the separation of U-233 from a thorium blanket; in fact, the possibility exists that this system would lend itself well to a continuous processing scheme for the blanket.

The fused salt processing scheme is based on the preferential oxidation of some of the metals present in the liquid metal fuel or blanket by the addition of a fused salt. The metal or metals which are oxidized to salts are easily extractable into the fused salt bath while the unoxidized metals remain in the liquid metal bath. For example, when a mixture of uranium and fission products in molten bismuth is equilibrated with salt mixtures such as LiCl - KCl, the fission product chlorides are extracted into the fused salt while the uranium remains in the molten metal.

The volatile fluoride method is based on the possibility of the continuous separation of protactinium and uranium from thorium by taking advantage of the large differences in the volatility of the fluorides. Work done at Brookhaven has not been sufficient to provide a good evaluation of this scheme.

The Brookhaven group is doing no work at present on Th - U-233 processing, and have no program planned for such studies.

c. Pyrometallurgical Processing

Pyrometallurgical processing, in general, results in decontamination factors which are considerably lower than those obtained by wet chemical processing. As a consequence, a pyroprocessing scheme usually assumes that the products will be fabricated remotely. Holdup times in such systems would not be dictated by radiation stability of the solvent, but rather by the path taken by the protactinium.

In making cost comparisons between pyroprocessing and wet chemical processing, it is necessary to take into account the fact that pyroprocessing necessitates remote fabrication. However, if the assumption that wet chemical processing can permit non-remote fabrication is invalid, then pyroprocessing becomes much more competitive. This situation could occur as the result of inability to fabricate U-233 rapidly enough after processing or inability of the Thorex system to handle short-cooled material.

The Ames group has been studying ternary systems such as Mg-Th-U, visualizing a liquid-metal blanket which could be continuously processed. To date, this work has been carried out using unirradiated material. The indications are that excellent separations of uranium and thorium can be achieved. The courses of the protactinium and fission products in the system have not been studied yet and would eventually have to be determined.

Atomics International plans to develop the SRE as a high-temperature fuel irradiation facility. This ties in with their proposal for a reprocessing-refabrication experiment to be sponsored by the AEC at Canoga Park. The Processing Refabrication Experiment is based upon Atomics International's desire to operate at a level sufficient to verify cost estimates derived from laboratory scale work. These cost estimates are available in this organization's proposal to the AEC.

Since the uranium pyroprocessing schemes under development at Atomics International are aimed at only 90% decontamination of metallic fuels and necessarily are remote operations, the introduction of thorium activities in a former uranium processing

stream does not pose any radical change in the over-all plant design. Atomics International feels that the minor nature of the modifications involved is justification for continuation of development of the process along generalized lines until a better definition of the composition of reactor fuels on the long-term basis is obtained.

The thorium pyroprocessing development programs at Atomics International will involve little or no U-233 in the foreseeable future. The thorium metal (irradiated plus natural) requirements are estimated to be about 100 kilograms per year beginning fiscal 1959. The research and reactor development divisions of the AEC are presently supporting limited efforts at Atomics International on reprocessing chemistry.

C. U-233 AND ITS COMPOUNDS

1. Special Problems in Fuel Element Fabrication

The nonradiation aspects of the fabrication of metal or oxide fuel elements using U-233 do not differ from those associated with any other uranium isotope.

Attention has already been called to the severe handling problems associated with U-233 as a result of the accompanying U-232, which decays through a chain yielding severe gamma-ray activity. Theoretical considerations indicate that the gamma activity (above 40 kev) due to the U-232 chain in the U-233 product will be given, in r/hr, at the surface of a 3 cm radius sphere of U-233, by the following expression:

$$R = 15 \left[\text{ppm U-232} \right] \text{TF}$$

TF is a time factor which describes the growth of Th-228 into equilibrium with U-232 and is given by,

$$\text{TF} = (e^{-0.0099t} - e^{-0.3647t})$$

where t is in years. The equilibrium (10 years) value of $\text{TF} = 0.88$.

Some values of TF and R at different times are given below for a U-232 concentration of 20 ppm:

t, years	TF	R (r/hr)
1	3	90
2	5	150
5	8	240
10	9	270

Of particular interest is the fact that when the important 2.6 mev Tl-208 gamma activity in the Th-228 decay chain is plotted against time and extrapolated to zero activity for processed and fabricated U-233, the intercept is the date of metal reduction, not the earlier date of chemical processing. (The decay-chain isotopes are removed in the metal reduction, melting, and casting steps.)

U-233 handling problems are closely associated with the processing scheme. It is generally agreed the U-233 product of the present Thorex process could be fabricated in a plutonium-type facility if fabrication were carried out almost immediately (within two weeks). The most convenient arrangement for such a process would be to have the fabricating facility very close to the processing facility. If fabrication immediately after processing were not possible, then it would be necessary to build a highly shielded facility for remote processing or to recycle the U-233 just prior to fabrication.

Pyroprocessing appears to lend itself only to remote fabrication. This scheme of processing and fabricating has not been developed to the stage where cost data are available for the purpose of comparing pyroprocessing cycles with wet chemical cycles.

A serious U-232 problem may also exist in irradiated U-233 fuel elements. This is a consequence of the U-233 (n, 2n) reaction. The cross section for this reaction is estimated to be of the same order of magnitude as the cross section for the thorium (n, 2n) reaction. (It is estimated that about 5% of the U-232 formed in the blanket occurs as a result of the U-233 (n, 2n) reaction.) The equilibrium U-232 concentration in the U-233 fuel elements and the attendant handling problems have not been studied. This is a problem which requires attention.

2. U-233 Fabrication Capabilities

A major problem in implementing a U-233 research and development program has to do with facilities capable of processing U-233 nitrate from the Oak Ridge Thorex separations plant to the various forms needed.

Chemical compounds such as the sulfate, fluoride, and oxide, as well as the metal, will be required. In addition, the oxide and metal must be fabricated to the desired forms; oxide will be required as pellets or as high-fired powder for dispersions, and metal may be required as foil or as metal for alloying purposes.

An extensive review of existing and adaptable facilities within the AEC capable of meeting the above requirements has been carried out. No industrial fabricators were consulted since it is believed that the relatively small quantities of U-233 involved and the need for some rather specialized facilities would make participation in the short-range program unattractive to them. In addition, only AEC installations with some background or capabilities in this field were considered since the short-range nature of the research and development program would not justify large capital expenditures for construction of a facility and the building up of a competent staff. Until sufficient information is developed through the short-range research and development program to permit evaluation of U-233 as a potentially economic reactor fuel, a minimum capital investment within the AEC is indicated; maximum use should be made of currently available facilities.

As a longer range possibility, the Argonne 350 facility for plutonium fabrication could be modified to permit the preparation of the oxide from fresh U-233 nitrate, the reduction to metal, and fabrication. This facility will be completed in 12 to 15 months and will be contained but not shielded.

The status of the AEC capability for U-233 fabrication is as follows:

a. U-233 Chemical Compounds

Figure 6 shows a flow sheet for the preparation of U_3O_8 and metal from U-233 nitrate. The production of uranyl sulfate and fluoride compounds should present no problem for existing facilities at Oak Ridge, assuming that fresh U-233 is available and modifications can be made to contain the alpha activity. Development of this capability in the Chemical Technology Division, Oak Ridge, appears to be most convenient from all standpoints since it would be adjacent to the Thorex separations plant operated by the same division. In addition, there is considerable experience available resulting from the production of uranyl sulfate for the

FIGURE 6

U-233 PROCESSING FLOW SHEET

* About 50 gms of U-233 are tied up in resin, so the resin must be treated by elution with HCl and recycled to ion exchanger.

** Shielded equipment used before this point; unshielded equipment used after this point.

HRE program. A capital expenditure of about \$100,000 would provide facilities adjacent to the Thorex plant for producing the necessary salts, in addition to the oxides. The U_3O_8 shown in Figure 6 could probably be employed for thorium-uranium compacts or could be further reduced to UO_2 for compacting into pellets.

b. Reduction to Metal

Production of U-233 metal from UF_4 is shown in Figure 6. Oak Ridge does not possess a reduction facility; however, as has been mentioned earlier, the possibility exists that for a reasonable investment a pilot plant could be established adjacent to the Thorex plant for processing fresh uranyl (233) nitrate to the fluoride. Equipment for the subsequent reduction to the metal could be provided for approximately \$100,000.

Hanford possesses a plutonium reduction facility operating on a production basis. It is doubtful that they would permit contamination of the facility with U-233 or interference with plutonium production scheduling. However, there is a research group at Hanford with small-scale reduction facilities for plutonium which could be adapted to U-233 processing.

c. Fabrication of Metal Shapes

A long-term possibility for U-233 metal fabrication at Argonne is the EBR-II fuel element fabrication facility which is to be fully shielded. This facility is scheduled for completion in March, 1960; however, first priority will be given to the EBR-II program. Thus, U-233 fabrication would not be possible until 1963.

Oak Ridge currently has no U-233 fabrication capabilities but does possess considerable background both in U-233 handling problems and in fabrication techniques for uranium. A capital investment of about \$500,000 would probably be required to provide a versatile U-233 metal production and fabrication facility at Oak Ridge.

Hanford currently possesses a glove-box operation for fabricating plutonium which could readily be adapted to U-233. All necessary equipment for melting, casting, rolling, annealing, brazing, and machining is on hand. In addition, a versatile plutonium metal and ceramic fabrication pilot plant is scheduled for completion.

by April, 1959, and can be considered a longer range possibility for fresh U-233 fabrication.

d. Production of MTR-Type Fuel Elements

If there is to be a U-233 loading in the MTR in the near future, a fuel element fabrication facility must be developed with minimum delay. Maximum use of existing facilities is clearly indicated for this application.

Hanford currently possesses equipment suitable for producing MTR-type elements and has been assigned the task of producing a plutonium core loading for the MTR. The schedule called for completion of this work by February 15, 1958. With minor modifications, it was believed that Hanford should be able to produce a U-233 loading shortly after that time.

Oak Ridge can be considered as another possibility for MTR elements based upon their experience in this area, as well as the existence of applicable equipment. However, a capital expenditure of approximately \$200,000, would be involved in developing U-233 fabrication capability at Oak Ridge.

If the scheduling of an MTR U-233 loading is delayed by approximately one year, the Argonne 350 facility could be considered as a third possibility for the production of the fuel loading.

e. Production of Oxide Pellets or Thoria-Urania Pellets

There are no facilities at present for the pelletizing of U-233 oxides. To meet the requirements of the minimum U-233 reactor programs it will be necessary to develop such facilities. In this connection both Hanford and Argonne will have versatile fuel element fabrication facilities in operation within one to two years. Fabrication of U-233 oxides should be possible in these plutonium facilities with minor modifications. For shorter range applications, current research facilities at Oak Ridge, Argonne, Hanford, and Los Alamos may be adapted to produce relatively small quantities.

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III. REACTOR DESIGN

A. PHYSICS DATA

Basic physics data for U-233 appear to be about as well developed as for U-235 except that fewer resonance parameters have been measured for U-233. The microscopic data needed for U-233 criticality calculations are considered to be as reliable as for U-235 at thermal energies. However, there is a great lack of critical experiments using U-233, and therefore, information from this source is not available.

Neutron-yield data from U-233 fission are not as well known as for U-235, although they are known to be different. The average cross section of U-233 fission products appears to be lower than in the case of U-235; however, more data are necessary before this can be firmly established.

Data on cross sections and η (number of neutrons/neutron captured in the fuel) as a function of energy are being obtained at Brookhaven, MTR, Oak Ridge, Argonne, Los Alamos, and Livermore. It is well known that η for U-233 is essentially independent of energy in the thermal and fast neutron ranges. However, values are lacking for η in the resonance region. Many of the above sites are also studying the delayed neutron fraction from U-233. Appendix A should be consulted for details concerning programs at each site.

The spatial distribution of U-233 buildup in thorium has been studied at MTR. The results of this study were presented at the October 1957 meeting of the American Nuclear Society.

Los Alamos has recently published new data on the delayed neutron fractions for thermal fission of Pu-239, U-233 and U-235¹. The values were found to be 0.0021, 0.0026, and 0.0064, respectively. This reverses the order of the older measurements of U-233 and Pu-239.

UCRL, Livermore, is presently studying the criticality conditions of aqueous solutions of U-233. Five kg of uranyl fluoride are on hand for this purpose.

The MTR physics group is interested in measuring cross sections of Pa-231 and Pa-233. Pa-231 is important because of the Pa-231 (n, γ) Pa-232

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¹LA 2118 and Phys. Rev. Vol. 107 #4

reaction leading to U-232. The cross section of Pa-233 is of importance because it affects the breeding ratio. Plans have been made to determine the fast effect, ϵ , and the effective resonance integral, $\int \sigma_{a\text{eff}} \frac{dE}{E}$, for various shapes of thorium lumps. In addition, a study of the transient behavior of fission product poisoning in U-233 after irradiation is being planned.

Studies on cross sections will continue at most of the facilities. Although the work mentioned above is quite extensive, U-233 requirements are small (on the order of a few kg).

Extensive measurements on high neutron energy cross sections have been carried out at Los Alamos.

B. CURRENT AND PROPOSED PROGRAMS USING TH - U-233

A variety of experiments are of interest to reactor designers in order to obtain the following information:

1. Critical mass
2. Resonance escape probability
3. Breeding gain
4. Effective fraction of delayed neutrons
5. Reactivity changes as a function of flux and time.

Most of the data related to specific reactor designs can best be determined by means of exponential and critical experiments. These experiments will require large amounts of U-233. The amounts desired are given here in conjunction with each project.

Table IV summarizes current and proposed programs contemplating the use of U-233 in the next five years. The reactors are described in Appendix B. Initial requirements for U-233 associated with each project were given in Tables I and II.

1. Brookhaven National Laboratory

The only reactor concept being considered at Brookhaven that may use Th - U-233 is the Liquid Metal Fuel Reactor (LMFR). At present

TABLE IV

CURRENT AND PROPOSED U-233 REACTOR PROGRAMS

<u>INDUSTRIAL PROJECTS</u>		
<u>Reactor</u>	<u>Description</u>	<u>Designer</u>
1. Advanced Thorium Reactor	Epithermal breeder, sodium cooled	Atomics International
2. Consolidated Edison	Thermal, pressurized water	Babcock & Wilcox
<u>ATOMIC ENERGY COMMISSION PROJECTS</u>		
1. LMFR	Liquid metal fuel. Thermal-breeding experiments may be tried.	BNL
2. FSR	Fused Salt Reactor - potential burner or breeder - thermal, intermediate or fast	ORNL
3. Homogeneous Reactors (HRE-II & HRE-III)	Aqueous Homogeneous Potential thermal breeder	ORNL
4. EBWR	Heavy water - thermal	ANL
5. EBR-II	Fast breeder, sodium cooled	ANL

no work has been done along these lines. Brookhaven has 1 kg of U-233 which is to be alloyed with aluminum for elements to be used as a central 1-kg loading for a two-region study.

A very recent development is the possibility that Brookhaven will want 15 kg of U-233 within two years for carrying out a general study of breeding in liquid metal reactors. The U-233 would be alloyed with aluminum and a critical facility core would be mocked up using a metal, such as bismuth, in the solid rather than the liquid form.

2. Oak Ridge National Laboratory

a. Fused Salt Reactor Program

Oak Ridge is considering a fused salt civilian power reactor to operate in the intermediate range using the Th - U-233 cycle. It is estimated that the critical mass for U-233 in such a system would be about one-half that for U-235 because parasitic capture should be about one-half that found with U-235. Therefore, even at twice the cost of U-235, the U-233 system would still be competitive and, at the same time, would have a 0.2 breeding ratio advantage over the U-235 reactor. Oak Ridge requires 20-70 kg of U-233 in the form of metal foils for critical studies in the resonance region (1-100 ev), and hopes to initiate this work by fiscal 1959.

In addition, 100-200 kg of U-233 would be required for critical experiments and an initial reactor loading about 1961.

b. The Homogeneous Reactor Program

It would be desirable to operate the HRE-II with a U-233 loading for about a year to test the safety of the concept. In particular, it is desirable to see if the reactor is stable with the small fraction of delayed neutrons associated with U-233. No trouble is anticipated from this source since HRE-I was taken prompt critical with no problems. Calculations have shown that delayed neutrons may not be necessary for the control of circulating homogeneous reactors. About 715 kg of U-233 would be needed for the HRE-II by July, 1958. It would be used as the uranyl sulfate. Isotopic purity would not be too important as

long as there is no large fraction of U-235 (80% U-233 with less than 8% U-235). About 5 kg of U-233 would be returned after two years.

It is assumed that the HRE-II, a Th - U-233 breeder reactor, will be critical by the first quarter of fiscal 1963. The purpose of this project is to demonstrate the possibility of obtaining a conversion ratio greater than one.

This reactor will require 36 kg of U-233 as uranyl sulfate by the last quarter of 1962 or the first quarter of 1963. Of this amount, 16 kg will be for inventory, 12 kg will be burned (200 days full power operation), and 8 kg will be in chemical processing. At the end of the experiment, the entire reactor contents will be processed. This program should be completed within two years of the starting date; however, Oak Ridge would like to retain the processed material for further operations.

3. Argonne National Laboratory

a. EBWR

Argonne would like a full loading of U-233 for the EBWR after it has been changed over to D_2O and the U-235 data are complete. This would require about 20 kg of U-233 as the oxide to be available in about two years. In addition, about 20 kg of U-233, as the oxide, will be required for critical experiments to be run in conjunction with the EBWR experiments and as part of a general study of large D_2O oxide reactors. The U-233 would be needed in about 18 months. It may be helpful to have as much as 75 kg of U-233 as the oxide for such a program if critical experiments in moderate sized reactors become necessary.

b. EBR-II

U-233 is required for a study of fast breeding in the EBR-II. This work will require 100-200 kg of U-233 for a complete loading of EBR-II, around 1960. In addition, 100-200 kg of U-233 will be required for the fast critical assembly (ZPR-III) to be built at NRTS, Idaho. It should be noted that a requirement for 25 kg of U-233 is shown in Table I for Argonne's ZPR-VI facility. The ZPR-VI is planned as a minimal facility for fast critical studies to be

constructed at the Du Page site, while ZPR-III is to be a rather extensive facility. It is possible that if the data from the experiments at ZPR-III were forthcoming soon enough, it would not be necessary to proceed with ZPR-VI plans.

4. University of California Radiation Laboratory, Livermore

a. Critical Assembly Group

The critical assembly research group desires to measure critical masses for U-233 and will work with the metal and metal hydrides. Spherical time-dependent studies and critical geometry studies will be carried out.

At present, UCRL has 5 kg of U-233 on hand as uranyl fluoride which is intended for use in the study of physical constants. The experiments will be carried out with the uranyl fluoride in aqueous solution. The data should be available by the end of 1957.

b. Gas-cooled Reactor

A group at Livermore is thinking in terms of a large, gas-cooled, graphite-moderated reactor. This reactor would preferably use U-233 and thorium oxides impregnated in graphite. There would be no fuel cladding; power densities would be on the order of 1 MW/cu ft. The reactor would operate at 1200-2200°C. These high temperatures would cause the fission fragments, including the normally solid members, to diffuse out of the graphite and be carried away by the cooling gas. Such a reactor would have a high burnup and long operating time.

This plan would require a large research program. Information would be required on resonance escape probability in the various possible systems of U, Th, C, Pa, and fission products. Several kg of U-233 for studies of resonance escape probability would be required. In about two years, 20 kg would be necessary for exponential experiments.

5. Atomics International

a. Advanced Thorium Reactor

Atomics International would like 50 kg of U-233 for a critical experiment in the 100-10,000 ev region. It is felt that the available physics constants are not sufficient to predict the control and critical mass variables. U-233 oxide canned in stainless steel would be employed, and the array would be completed with 12,000 kg of thorium.

b. Babcock and Wilcox

The Consolidated Edison project visualizes a thermal, one-region, pressurized-water reactor utilizing the Th - U-233 fuel cycle.

Approximately 10 grams of U-233 in the form of metallic foils are required for:

- (1) Comparison of the fission resonance integrals of U-233 and U-235 in the Consolidated Edison neutron flux spectrum.
- (2) Comparison of the total fission rate of U-233 and U-235 in the Consolidated Edison neutron flux spectrum.

Also, 7-8 kg of U-233 in the form of oxide will be required for comparison of the reactivity difference between U-235 and U-233 in a critical experiment. This can be carried out by replacing U-235 with U-233 in the central portion of the experiment and measuring reactivity differences with a calibrated control rod. This material would be needed for a nine-month period if it can be received by March, 1958.

IV. REACTOR OPERATIONAL PROBLEMS AND LIMITATIONS

To date, no reactor has been operated on U-233, and therefore, the unique problems, known and unknown, associated with its utilization will not be brought into proper perspective until the firm requirement to produce and operate a reactor loading exists. In this connection, MTR operation using U-233 for a complete cycle would be of general interest to the over-all reactor program. Valuable data relative to reactor design and operation could be obtained for

comparison with U-235 and plutonium under similar reactor conditions. In addition, the problems inherent in a complete U-233 fuel cycle would become better appreciated.

Pertinent information obtainable from an MTR loading would include the following:

1. Critical mass
2. Excess reactivity of 3 x 9 and 5 x 5 loadings
3. Reactivity lifetime of the loading
4. Xenon and Sm poisoning
5. Additional fission product poisons resulting from U-233 fission
6. Control characteristics
7. Activity level of spent fuel elements at time of discharge
8. "Cooling" behavior of Fuel elements
9. U-232 content of spent fuel elements
10. Problems involved in the reprocessing of spent fuel elements
11. Problems associated with fuel element fabrication using reprocessed U-233
12. Flux plots of thermal and above-thermal flux over the reactor core.

There has been some hesitation expressed about running a load of U-233 assemblies in the MTR because of the small fraction of fission neutrons that are delayed. The data which had been in use for many years and which were first published in 1948¹ gave the fraction of delayed fission neutrons as 0.0024, 0.0073, and 0.0036 for U-233, U-235, and Pu-239, respectively. However, as noted previously, the latest values are 0.0026, 0.0064, and 0.0021, respectively. Thus, these fissionable isotopes have a lower delayed neutron fraction than was heretofore believed and U-233 is actually safer than plutonium. With β established as 0.0021 instead of 0.0036 for Pu-239, extreme care will have to be taken in the planned MTR plutonium loading to ensure that no inadvertent behavior in any experiments can suddenly inject 0.21% excess reactivity (a rather small amount of reactivity).

The feasibility of running the MTR on a load of plutonium assemblies was reviewed by D. R. de Boisblanc². In this report it is assumed $\beta = 0.0036$ for

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¹Physics Review 73, 111 (1948).

²Phillips Petroleum Company Report - 224

Pu-239. (The new data were published after this study was completed.) It was concluded that operation on plutonium is feasible, but with regard to safety, the report states, "The shorter lifetime and lower values of the delayed neutron fraction have a bearing on the ultimate safety of the reactor in the event of a large reactivity excursion, but the magnitude of the difference between Pu-239 and U-235 cannot be estimated because there are no kinetic data on systems such as the MTR under full hydraulic flow conditions. About all that could be said with certainty is that the plutonium reactor would be less safe."

It is possible that with the new information on β , the advisability of the Pu loading should be reviewed. If it is considered acceptable to use a Pu loading then, from the safety standpoint, it will be acceptable to use a U-233 loading.

V. REVIEW OF PROPOSED RESEARCH AND DEVELOPMENT PROGRAM

The preceding discussion has indicated many areas where future research and development are required before a realistic appraisal of the economics and technical feasibility of U-233 as a power reactor fuel can be undertaken. A brief review of these areas is given in the following summary:

A. REACTOR DESIGN

1. Basic physics research on nuclear constants is in progress at various installations and such work should be continued. Additional effort is needed in the intermediate neutron energy range.
2. There is a general need for a variety of critical experiments to be carried out in connection with individual reactor projects.
3. A review of the problems associated with control and safety of U-233 fueled reactors is greatly needed. This is especially important in the light of recent information on the fraction of delayed neutrons for U-233. This review should be supplemented by an experimental program to verify the current value of delayed neutron fraction assigned to U-233.

B. REACTOR OPERATION AND LIMITATIONS

1. Change in gross reactivity as a function of flux and time should be measured in order to compare the effects of fission product poisoning of U-233 with those for U-235.

2. No reactor has ever operated on U-233. The unique problems associated with the use of this fuel will not be brought into proper perspective until a comparison can be made with other fuels under similar conditions.
3. In connection with No. 2, above, a U-233 loading for MTR has been suggested by others and is believed to offer several attractive features, including:
  - a. The firm requirement to produce at least one core loading of U-233 will provide considerable information concerning problems encountered in fuel element fabrication. No U-233 irradiated as a fuel has been reprocessed, so problems associated with this phase of the fuel cycle are not well known. Recycling a core loading from the MTR would provide this type of information.
  - b. Plots (of thermal and above-thermal flux) can be obtained over the reactor core, and the thermal flux rise in the Be reflector can be measured.
  - c. Reactivity lifetime of the fuel can be obtained for comparison with U-235 and plutonium.
  - d. Control aspects of reactor operation can be studied.

C. FUEL CYCLE

1. A large area of economic uncertainty is centered around the reprocessing phase of the fuel cycle. Various alternatives available in the recycling of U-233 and thorium for power reactor applications should be studied more extensively from an economic standpoint.
  - a. Cooling period should be studied in relation to inventory cost, reprocessing costs, and fuel element fabrication costs.
  - b. Wet chemical separations and pyroprocessing methods require more technical development before realistic relative cost data can be determined.
  - c. Reliable cost information should be developed through extended runs of processing pilot plants. The Thorex process has reached a greater state of development than other processing methods and, consequently, less additional effort will be required to obtain information on economic and technical feasibility.

- d. It is necessary to evaluate the economics of a given processing scheme in terms of the associated fabrication scheme which is dictated by the activity levels of the products. Information relative to the saving in fabrication costs per dollar of processing cost must be developed for the various processing schemes to achieve minimum over-all costs.
2. The problems inherent in fabricating recycled thorium and U-233 blanket and fuel elements require further study supplemented by actual operating experience before an adequate economic evaluation can be carried out. In the short run, a laboratory scale reduction and fabrication facility should be developed to handle small quantities of U-233 for reactor development programs.
3. Extensive data relative to permissible reactor exposure levels from the metallurgical standpoint for both solid fuel and fertile elements under anticipated power reactor conditions are required before this important economic factor can be satisfactorily evaluated. Thorium and Th-U mixtures should be studied as metal, alloys, and ceramics.

D. OTHER CONSIDERATIONS

In addition to the above factors which strongly affect economics and technical feasibility, a number of others should be explored which could contribute greatly to technical progress in the thorium - U-233 field; These include:

1. Potential metal reduction processes should be reviewed and the most economically attractive ones further developed.
2. Product purity requirements need extensive study, especially with regard to their effects on fabricability of thorium metal and reactor performance. The effect of type and concentration of impurities is not well known at the present time.
3. Melting and casting methods for thorium should be further investigated with a view toward economics and improved metal quality.
4. Considerable effort is needed on fabrication technology of thorium and its alloys. To date, most of the available experience has been accumulated on thorium metal.

5. Considerable work on the physical metallurgy of thorium is required. Although many of the thorium binary phase diagrams are known, practically no effort has been expended on achieving improved mechanical properties at elevated temperatures through alloying nor has a careful study of potential heat treatable alloys been made. This is quite a large area for research, especially since one of the attractive features of thorium is the possibility of elevated temperature operation of thorium fuel and fertile elements.
6. Physical property data on both irradiated and unirradiated systems are required for thoria and urania or mixtures of the two for improved reactor design.

APPENDIX A
TECHNICAL STATUS, FUTURE PLANS,
AND
U-233 REQUIREMENTS OF INDIVIDUAL SITES

NOTE: Appendix A is primarily a compilation of pertinent information developed through discussions with the representatives of various AEC and industrial installations currently carrying out or planning activities which are related to the Th - U-233 reactor program. Although every effort has been made to prevent errors in recording the substance of these discussions, omissions or inaccuracies are possible. The conclusions and statements regarding program plans are informal and neither represent the opinions of the Atomic Energy Commission or American-Standard, nor even the formal requirements of the installations concerned.

I. ARGONNE NATIONAL LABORATORY

A. STATUS

1. Some basic physics measurements are proceeding at present. These experiments require only very small amounts of U-233. It is felt that the available data are good, but are not sufficient.
2. The Metallurgy Department is studying the metallic systems; Th-U and Th-Pu. It is felt that the system Th-U is best suited for fuel elements for liquid-metal cooled fast reactors. Since Th is an excellent alloying agent for Pu, the Th-Pu system is also being considered for fuel elements.
3. The ANL #350 facility, which is designed for handling plutonium, should be ready in 12 to 15 months. It should be possible to use this facility for the fabrication of freshly processed U-233. The #350 facility is an unshielded glove box train which is designed for the containment of α active material. The cost of adding shielding to permit the handling of strong β , γ activity would be quite high.
4. The facility for fabricating EBR-II fuel elements is to be fully shielded and should be capable of handling high activity U-233. However, this facility will not be completed until about March of 1960, and will not be able to handle U-233 until 1963.

B. FUTURE PLANS AND REQUIREMENTS

1. In about two years ANL would like to place a full U-233 core in the EBWR. This would require about 20 kg of U-233 in the form of the oxide.
2. Twenty kg of U-233, as the oxide, will be required in about eighteen months for critical experiments to be run in conjunction with the EBWR and to be used as part of a general study of large D_2O - oxide reactors. It may be helpful to have as much as 75 kg of U-233, as the oxide, for such a program if critical experiments in moderate sized reactors become necessary.

3. It is possible that around 1960, the Reactor Engineering Division would like a complete loading of U-233 for the EBR-II for a study of fast breeding. This would require 100-200 kg of U-233 in the EBR-II loading and 100-200 kg of U-233 for the fast critical assembly (ZPR-III).
4. In addition to the requests for U-233 in (3) above, which are long-range requests, there is a request by ANL for 25 kg of U-233 for the short-range program (see Table I). This request for 25 kg is for the ZPR-VI which is proposed for the DuPage site. The ZPR-VI is to be a minimal facility for the study of fast breeders. It is possible that if the information from the large-scale ZPR-III facility were forthcoming soon enough, it would not be necessary to construct the ZPR-VI facility.
5. In addition to the above requests, ANL would like 3 kg of U-233 on hand to be used for miscellaneous experiments as they arise.
6. If U-233 were common, it would be desirable for use in research reactors. This would permit higher flux levels for the same power levels obtained with U-235.

II. ATOMICS INTERNATIONAL, CANOGA PARK, CALIFORNIA

A. STATUS

1. The Advanced Thorium Reactor (ATR) is in the conceptual design phase at present.
2. The Sodium Reactor at Santa Susana is presently undergoing physics experiments.
3. Pyroprocessing Refabrication Experiment (PRE) has been proposed to the AEC on the following basis:

The costs of remotely reprocessing a metallic fuel by pyrometallurgical methods and then refabricating by remote casting appear to be competitive with chemical methods such as Thorex. The PRE proposal is based on Atomic International's desire to operate at a scale sufficient to firm up cost estimates which have been derived from laboratory scale work. These numbers have been quoted to the Commission in the PRE facility proposal. The process to be used for thorium fuels will be substantially different from the uranium process proposed.

Since the pyroprocessing schemes under development by Atomics International are aimed at only 90% decontamination of metallic fuels, and necessarily are remote operations, the introduction of thorium activities in a uranium processing stream and vice-versa does not pose any radical change in over-all plant design. This is felt to be a justification for continuation of development of the process along generalized lines until better definition of the composition of reactor fuels is obtained.

The research and reactor development divisions of the AEC are presently supporting limited efforts at Atomics International on thorium-uranium alloy phase diagram work and reprocessing chemistry. This work was initiated during the current calendar year.

B. FUTURE PLANS AND REQUIREMENTS

1. Advanced Thorium Reactor

Atomics International would like 50 kg of U-233 for a critical experiment to be operated in the region of 100 to 10,000 ev where available physics constants are not sufficient to predict control and critical mass variables of such an array. U-233 would be used as oxide canned in stainless steel tubes. Due to the activity considerations during fabrication, Atomics International would like to receive this material evenly distributed over a six-month period beginning July, 1959. This would be followed by the receipt of 12,000 kg of thorium (metal or oxide) which will be used to complete the critical array.

2. The Sodium Reactor

This reactor will shortly begin operation under full power. The slightly enriched loadings in this reactor are due for replacement

April, 1958. A thorium-uranium loading will be introduced at this time. This loading will consist of 1.6 metric tons of thorium obtained from SRP. It is not known how much exposure this material will stand, but preliminary tests (3/8-inch diameter thorium-uranium pins irradiated in the MTR) indicate that 6000 MWD/T is a possibility. The total U-233 production at this exposure will be negligible and no plans have been made to reprocess this loading.

3. The Pyroprocessing Development Programs

These programs at AI will involve little, or no, U-233 as such in the foreseeable future. The thorium metal (irradiated plus natural) requirements are estimated to be about 100 kg per year beginning fiscal 1959.

4. The Thorium Metal Requirements

The thorium metal requirements were totaled for the entire development at AI up to January, 1960, as follows: March, 1959 - 2400 kg; September, 1959 - 500 kg. It is thought that this material will be used in chemistry and metallurgy programs in support of SRE and PRE.

III. BABCOCK & WILCOX COMPANY, LYNCHBURG, VIRGINIA

A. STATUS

1. The chief interest in Th - U-233 is in connection with the Consolidated Edison program.
2. Some work on the fabrication of thorium as metal and oxide is currently in progress at Alliance, Ohio.
3. Babcock & Wilcox currently has on hand about 9000 kg of thorium metal in the form of 3-inch plates, 56 mils thick. This material was obtained from Fernald (rolled at Simonds) for critical work on the earlier Consolidated Edison reference design.
4. Babcock & Wilcox is supporting some development work at Battelle on ThO_2 - UO_2 fuel elements. Samples will be irradiated in the MTR in the Babcock & Wilcox pressurized water loop.

B. FUTURE PLANS AND REQUIREMENTS

1. The Consolidated Edison time schedule looks something like this:
By June, 1959, they will need to decide what the second core will be.

There will be no U-233 available for this loading. In June, 1963, the second core goes in, and in June, 1965, the third core, which could be U-233. If so, physics work should begin by June, 1962, and delivery of U-233 should be made by the second quarter of 1961 to allow fabrication time. About 100 kg of U-233 would be required for the critical experiment. It is not recommended to consider this requirement now, since the above schedule is quite tentative.

2. Ten grams of U-233 are needed as metallic foil for comparison of fission resonance integrals of U-233 and U-235 in the Consolidated Edison neutron flux spectrum and for the comparison of total fission rate of U-233 and U-235.
3. Seven or eight kg of U-233 oxide are needed for comparison of the reactivity difference between U-235 and U-233 in the critical experiment. This can be done by replacing U-235 by U-233 in the central portion of the experiment and measuring reactivity differences with a calibrated control rod. This material would be needed for a nine-month period if it can be received by March 1, 1958.
4. It is felt that fission product poisoning should be studied as a function of time. This work might be done at a national laboratory with Babcock & Wilcox perhaps contributing man power. This work would require 0.5-1 kg of U-233. Change in reactivity as a function of flux and time would be measured. A reactivity-measuring reactor would be necessary for this work. Data on gross reactivity effects only would be obtained.

IV. BROOKHAVEN NATIONAL LABORATORY

A. STATUS

1. Fundamental physics investigations are proceeding at BNL on the study of cross sections and analysis of resonances over a wide energy range. Results of this work to date can be found in BNL-325 and its supplements.
2. Funds are not available for the operation of the LMFR with a breeding blanket.

B. FUTURE PLANS AND REQUIREMENTS

1. It is planned to operate LMFR on U-235 without a breeding blanket because of lack of funds.
2. One kg of U-233 has been allocated. It will be used to replace a 1 kg loading for a two-region study to yield data necessary for design of a U-233 core in the LMFR.
3. A recent development involves the possible expansion of the above program to a 15 kg loading of a critical facility.

V. DAVISON CHEMICAL COMPANY, ERWIN, TENNESSEE

A. STATUS

1. Davison is presently equipped to produce a purified thorium nitrate by a solvent extraction process using an aqueous nitrate feed and extraction by means of TBP in kerosene. The starting material is obtained from a sulfuric acid leach of the ore; the sulfate concentrate is converted to the nitrate prior to processing. This material is a by-product of their eastern rare-earth plants.
2. Some studies are being done on nitric acid leaching of the ore, but facilities are not set up to use this process as yet.
3. A reactor grade ThO_2 which sells for \$7.50/lb is produced.
4. Davison converts the oxide to the metal by reduction of the oxide with calcium.
5. The final product (reactor-grade thorium metal) is formed by a double arc melting process.
6. The reactor grade metal sells for \$20/lb in tonnage quantities. Other prices are as follows:

<u>Quantity</u> <u>(lb)</u>	<u>Price</u> <u>(\$/lb)</u>
to 100	50
500	40
1000	30
5000	25
large tonnage quantities	20

7. All prices quoted above are based on \$3/lb contained thorium for concentrate from the rare-earth plants. If there were no rare-earth

profit, the concentrates would cost \$4.75/lb; however, the expectation is that this would drop to \$4/lb for a large-scale program.

8. Estimates have been made of thorium ores in this country which are potentially available to Davison. On this basis, Davison could supply 100,000 tons of thorium metal over a period of 20 years.
9. As of now, Davison can produce thorium metal at the rate of 50 tons/yr. If given a lead time of four months, this could be increased to a rate of 100 tons/yr.

B. FUTURE PLANS AND REQUIREMENTS

There are no future requirements for U-233.

VI. INSTITUTE FOR ATOMIC STUDIES, AMES, IOWA

A. STATUS

1. Methods of extracting thorium from the ore and producing purified thorium nitrate are being studied.
2. A production method for metallic thorium is under investigation. The method involves conversion of the nitrate to the anhydrous chloride followed by reduction of the chloride by magnesium to give metallic thorium.
3. Pyrometallurgical methods of processing thorium-uranium mixtures are being studied. For example, low melting Th-Mg eutectic is formed in which uranium has only very small solubility. Good separations of thorium and uranium have been obtained; however, the paths of protactinium and the fission products are not yet known.
4. There are no studies under way involving U-233.

B. FUTURE PLANS AND REQUIREMENTS

1. The projects now in progress will be continued.
2. The path of protactinium and fission products in pyrometallurgical processing will be followed. This will require only small amounts of irradiated material which can be obtained by irradiation of small samples of thorium as they are needed.
3. There is no requirement for U-233.

VIII. PHILLIPS PETROLEUM COMPANY, NRTS

A. STATUS

1. Thorium slugs have been irradiated in the MTR under a program initiated during the first year of MTR operation and continued until June, 1954.
2. The metallurgists at the MTR found no apparent radiation damage to the thorium slugs. It is felt that thorium would be a much better material than uranium from the radiation damage standpoint.
3. More thorium is being irradiated in the MTR at present, and consideration is being given to intermittent irradiation of slugs to diminish the destruction of intermediate isotopes.
4. The fast chopper technique is being used to measure η and σ_f for U-233. These measurements will extend over energies from 0.02 to 10 or 11 ev.
5. The Applied Physics Section is studying the delayed neutrons from U-233 fission. A paper was presented on the spatial distribution of U-233 buildup in thorium at the October, 1957, meeting of the ANS.

B. FUTURE PLANS AND REQUIREMENTS

1. Plans are being made for a program in which 150-200 gm capsules of thorium will be irradiated for long periods up to five years and reactivity gains measured.
2. Measurement of the cross sections of Pa-231 and Pa-233 in the chopper are of interest. Pa-231 is important because of the Pa-231 (n, γ) reaction which leads to U-232. The cross section of Pa-233 is of importance because it affects the breeding ratio.
3. The Applied Physics Section has plans to study the transient behavior of fission-product poisoning in U-233 after irradiation.
4. Plans are being made to determine the fast effect, ϵ , and the effective resonance integral $\int \sigma_{a\text{eff}} \frac{dE}{E}$ for various shapes of thorium lumps.
5. The use of thorium as a flux monitor for high nvt's has been suggested. At the completion of irradiation, the monitor would be compared with standards in the Reactivity Measuring Facility to determine its U-233 content, the U-233 content being an index of the nvt.

- 6 At the MTR, a probable need exists for approximately 1 kg of U-233 for cross section studies and 1 kg for the RMF measurements over the next five years.
7. There has been some hesitation expressed about running a load of U-233 assemblies in the MTR because of the small fraction of delayed neutrons. Previous data that had been in use for many years¹ indicated that the delayed neutron fractions for U-233, U-235 and Pu-239 were 0.0024, 0.0073, and 0.0036, respectively. However, the latest data² are 0.0026, 0.0064, and 0.0021 for U-233, U-235, and Pu-239, respectively. Thus, if a plutonium loading is considered to be safe, a U-233 loading will probably be permissible.

VIII. OAK RIDGE NATIONAL LABORATORY

A. STATUS

1. Processing

- a. ORNL has extensively studied the Thorex process for separating purified U-233 and thorium from irradiated thorium.
- b. The stability of the solvent in the presence of short-cooled material (30 days) is not yet fully established.
- c. The Thorex effort may be continued beyond the end of fiscal 1958 as part of the power reactor processing program now indicated in the production division budget.
- d. ORNL is also engaged in the study of new methods for processing thorium ores. In particular, processes using amines as the extractant are being studied.

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¹Phys. Review 73, 111, (1948).

²Phys. Review, 107, No. 4, 1044, (1957).

2. Metal Production

- a. Continuing studies on Metallex processes which involve the reduction of ThCl_4 by sodium amalgam are being carried out.
- b. It is felt that metal can be produced from the nitrate for \$3-5/lb of thorium. This includes the cost of arc melting.

3. Metallurgy and Fabrication

- a. ORNL has produced U-233 metallic foils for physics research at ANL and BNL. The metal was melted, cast, and rolled to a slab. It was then put in a Ti-enamel jacket and rolled to a thin foil.
- b. Most fabrication work at ORNL of interest to the present study has been concerned with thorium metal. Castings have been handled like uranium.

4. Ceramics

- a. It has been possible to fabricate ThO_2 to nearly 100% of the theoretical density by using CaO .
- b. ThO_2 and UO_2 crystals have been produced that show excellent behavior in slurry systems.
- c. Almost no work is being done now on ThO_2 ceramic elements.

5. Homogeneous Reactor Program

- a. HRE-I was operated successfully.
- b. The slurry system for uranium or thorium oxides has been proven to be satisfactory.
- c. The HRE-II is almost ready for operation.

6. Molten Salt Power Reactor

- a. ORNL has made studies of molten salt technology as applied to power reactors.
- b. Most of the information required to design a practical power reactor is felt to be available; however, long-term tests of materials and components are lacking.
- c. Considerable information from the ANP program is applicable.
- d. Physics data on U-233 is not sufficient in the intermediate range where the Th - U-233 breeder is most attractive.
- e. None of the reactor physics experiments necessary for utilizing U-233 in such a reactor have been performed.

B. FUTURE PLANS AND REQUIREMENTS

1. Processing

- a. More definite information concerning future purity specifications for thorium and U-233 are required.
- b. Studies will continue on improving the Thorex process. Improvement of the ruthenium decontamination, in particular, is required.
- c. At least two short cool runs (30-day pre-Thorex cooling) are required to demonstrate the process.
- d. About 15 tons of irradiated thorium are required by the first half of the fiscal year 1959.

2. Metal Production

- a. ORNL plans to shut down the Metallex thorium study and go into the study of production of metallic uranium.

3. Metallurgy and Fabrication

- a. There are no future plans for this program.

4. Ceramics

- a. There are no future plans for this program.

5. Homogeneous Reactor Program

- a. It is planned to operate the HRE-II with U-233 fuel for about one year to demonstrate that the reactor is stable with a small fraction of delayed neutrons. No trouble is anticipated, since the HRE-I went prompt critical with no problems. In addition, it has been calculated that delayed neutrons may not be necessary for the control of circulating homogeneous reactors. For the above purpose, a requirement of about 7.5 kg of U-233 by July, 1958, is foreseen. The isotopic purity is not too important as long as there is no appreciable amount of U-235 present. At least 80% U-233 with less than 8% U-235 is needed. Five kg could be returned after two years.
- b. It is assumed that the HRE-III, which is to be a thorium breeder, will be critical by the first quarter of the fiscal year 1963. For this purpose, 36 kg of U-233 will be needed, as uranyl sulfate, by the last quarter of the fiscal year 1962 or the first quarter of the fiscal year 1963. There will be 16 kg in the inventory - 12 kg burned and 8 kg in chemical processing.

It is important to demonstrate that this reactor has a conversion ratio greater than unity. It is desirable to show also that the reactor is capable of operating without an auxiliary system; e.g., there will be a facility for the removal of xenon, but if this went out, the reactor should continue to operate safely. At the completion of the experiments, the entire system will be processed. This should be completed within two years, but ORNL would like to keep the U-233 for further operations.

6. Molten Salt Power Reactor

- a. It is proposed to use a fuel composed of U-233 tetrafluoride in a molten salt bath of BeF_2 and Li^7F . The blanket will be ThF_4 in molten $\text{BeF}_2 - \text{Li}^7\text{F}$.
- b. Twenty to seventy kg in the form of metal foils are needed for critical studies in the resonance region (1-100 ev) by fiscal year 1959. This will be a one-year program and the material will be returned.
- c. By about 1961, 100-200 kg of U-233 as the tetrafluoride are needed for critical experiments and an initial reactor loading. (It is actually desirable to have 500 kg available to build a power reactor with good breeding.)

X. SAVANNAH RIVER PROJECT (DuPONT)

A. STATUS

1. Thorium metal slugs, produced at Fernald, have been irradiated at Savannah River on a two-year experimental program which is about completed. The slugs were canned and bonded at Sylvania. No radiation damage problems were encountered with target elements at Savannah River.
2. Savannah River has had no experience with irradiation of thorium oxide elements.
3. The Savannah River solvent extraction equipment could be modified to a short cool Thorex process, but this would require additional equipment.
4. No work has been done on U-233 as a reactor fuel.
5. ThO_2 has never been considered seriously by Savannah River. Careful consideration and more background would be necessary before the oxide could be used. There are no plans to pursue this aspect.

6. It is felt that physics constants for U-233 are sufficiently well known to permit calculations for a thermal reactor.
7. Slugs are irradiated at Savannah River on the following basis:
The slugs are placed in high flux zones and irradiated for an arbitrary time. A small number are placed in the middle of the reactor. There is no attempt at optimization.

B. FUTURE PLANS AND REQUIREMENTS

There are no future plans expect to irradiate slugs as requested.

X. UNIVERSITY OF CALIFORNIA, RADIATION LABORATORY

A. STATUS

1. Five kg of U-233 are on hand in the form of a uranyl fluoride solution. This is being used to determine criticality conditions of aqueous solutions of U-233.
2. Results of the experiments are expected by early December.

B. FUTURE PLANS AND REQUIREMENTS

1. Experiments with solid U-233 hydrides are desirable.
2. Fifteen kg of U-233 are required for study of critical masses.
UCRL would like this as soon as possible.
3. The possibilities of a large gas-cooled, graphite-moderated reactor are being studied. This reactor would preferably use U-233 and thorium oxides impregnated in graphite. There would be no fuel cladding. Power densities would be on the order of 1 MW/cu ft or less. The reactor would operate at 1200-2200°C and at these high temperatures, the fission fragments would be expected to diffuse out of the graphite and be carried away by the cooling gas. Such a reactor should have a high burnup and a long operating time. This plan would require a long-term research program. Information is needed on resonance escape probabilities in the various systems of U, Th, Pa, C, and fission products.
4. The program outlined in No. 3 above would require several kg of U-233 for studies of resonance escape probabilities. If the information obtained was encouraging, then about 20 kg of U-233 would be needed within two years for an exponential experiment.

5. The impregnation of the graphite could be carried out by the chemistry group at UCRL.

APPENDIX B

REACTOR DESCRIPTIONS

NOTE: Appendix B is primarily a compilation of pertinent information developed through discussions with the representatives of various AEC and industrial installations currently carrying out or planning activities which are related to the Th - U-233 reactor program. Although every effort has been made to prevent errors in recording the substance of these discussions, omissions or inaccuracies are possible. The conclusions and statements regarding program plans are informal and neither represent the opinions of the Atomic Energy Commission or American-Standard, nor even the formal requirements of the installations concerned.

ADVANCED THORIUM REACTOR

The Advanced Thorium Reactor is an epithermal breeder concept for power development purposes. The present plan calls for constructing this plant with private capital and obtaining fuels cost-free from the Government.

This plant has the advantage of high breeding gain in a neutron energy range where parasitic capture by fission products and constructional materials is low. Since a low moderating ratio is desired, the fuel concentration in the core can be higher than with natural uranium - Pu-239 reactors. The ATR design is thought to be capable of true breeding; i.e., it should produce sufficient U-233 to be a self-sustaining unit. Also, since thorium fuel temperatures may be allowed to reach much higher temperatures than uranium metal fuels, the over-all thermal-to-electrical conversion efficiency looks very promising with a liquid metal heat transfer media.

CONSOLIDATED EDISON

This is a thermal thorium-converter-type reactor. It is fueled with enriched uranium and moderated and cooled with pressurized light water. Babcock & Wilcox is the designer and constructor.

The initial loading of the one-region oxide core will contain no U-233. Conversion of Th-232 to U-233 will occur and a conversion ratio for the duration of core lifetime is expected to be 0.47.

A feature of this reactor is the demonstration of the feasibility of adding an oil fired super heater to deliver the design steam temperature. The nuclear fuel provides energy at the rate of 500 MW and the oil fired unit provides 200 MW.

LIQUID METAL FUEL REACTOR CONCEPT

The basic long-range concept of this reactor is that it is to be a thermal breeder using a solution of U-233 in liquid bismuth as fuel and a fertile blanket consisting of a suspension of Th_3Bi_5 in liquid bismuth. Heat removal is accomplished by circulation of fuel and blanket through heat exchangers.

The fission neutrons are thermalized by a graphite moderator which is in the form of a cylinder drilled with holes through which the fuel circulates.

Funds are not available at present to permit BNL to carry out this concept to the breeding stage. At present, the reactor is to be operated on a U-235 fuel to obtain metallurgical, chemical, and engineering data. There is to be no blanket initially.

THE HOMOGENEOUS REACTORS

The various homogeneous reactors being studied at Oak Ridge are to be fueled with solutions of uranyl sulfate in D_2O . Depending on which reactor is being considered and the phase of the study, the fuel may be U-235 or U-233 and the blanket may be a D_2O solution of depleted uranyl sulfate or a D_2O slurry of depleted UO_2 or ThO_2 . Heat removal is accomplished by circulating the core and blanket through external heat exchangers.

These reactors are designed to test the basic concepts of homogeneous reactors. The HRE-III is designed to be a thorium breeder reactor and as such, it is important to demonstrate that it has a conversion ratio greater than unity. HRE-III shall be fueled with a D_2O solution of U-233 uranyl sulfate and shall have a blanket of a ThO_2 slurry in D_2O .

MOLTEN SALT POWER REACTOR CONCEPT (FUSED SALT REACTOR)

The Molten Salt Reactor concept is based on the use of a solution of a uranium or plutonium salt in a molten salt or mixture of salts for the fuel. If a blanket is used, then a thorium or U-238 salt is dissolved in molten salt or mixture of salts to form the blanket. The concept may be applied to the following systems: U-235 or Pu burner; Th - U or U - Pu thermal converter (or breeder in the case of Th - U); Th - U intermediate breeder; Th - U or U - Pu fast converter or breeder.

In the case of the Th - U-233 breeding system, the fuel might be a solution of U-233 fluoride in a molten mixture of BeF_2 and Li^7F or NaF. The blanket could be a solution of ThF_4 in molten Li^7F . Various combinations of BeF_2 and Li^7F or NaF are possible. In the case of a thermal breeder, a graphite moderator can be used in a heterogeneous arrangement with a base salt of BeF_2 and Li^7F .

The chief advantage of this system is that it is the only one in which a soluble thorium compound may be used, and thus, the problem of slurry handling is avoided. In addition, high temperatures (about $1000^\circ C$) are obtainable at low pressures.

Studies indicate that the Molten Salt Power Reactor can produce economical power provided longevity of components can be assured. This reactor complex using the Th - U-233 breeding cycle is most attractive in the intermediate range. The critical mass of U-233 should be about one-half that of U-235 and the breeding ratio should be about 0.2 units better than that obtainable with U-235. Therefore, it is assumed that it should be possible to pay twice as much for U-233 as for U-235 and still be competitive.

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