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SUBJECT: FUEL ELEMENTS FOR FOREION POOL-TYPE RESEARCH REACTOR (U)

TO: DR. H.M. ROTH

FROM: C.E. LARSON

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OAK RIDGE NATIONAL LABORATORY
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POST OFFICE BOX P
OAK RIDGE, TENNESSEE

September 6, 1955

U. S. Atomic Energy Commission
Post Office Box E
Oak Ridge, Tennessee

Attention: Dr. H. M. Roth, Director, Research and Medicine Division

Subject: FUEL ELEMENTS FOR FOREIGN POOL-TYPE RESEARCH REACTOR

Reference (a): Letter from H. M. Roth to C. E. Larson dated August 19, 1955, (ORA:DCD)

Gentlemen:

In accordance with the requests contained in reference (a), we submit the following comments and proposals on the current status of manufacturing methods for foreign pool-type, research-reactor fuel elements:

Were it not for the specification that the uranium be limited in U-235 enrichment at 20% in foreign reactors, reliable technical information could be provided, since the fuel elements would probably be designed in accordance with MTR-type technology. Complications arise from the fact that a reactor loading, based on an 18-plate, MTR-type fuel element and material enriched at the 20% level, necessitates approximately 48 weight percent of uranium in the fuel-plate core. The metallurgical knowledge of the uranium-aluminum alloy core has been limited to 25 weight-percent uranium. A review of this system indicated that an alloy containing 48 weight-percent uranium would not have acceptable homogeneity, would be quite brittle, and consequently difficult to fabricate. For this reason, the fabrication of the Geneva Conference fuel elements was modified by substituting a powder-metallurgy process involving UO_2 and aluminum powder for the uranium-aluminum alloy.

The decision to utilize UO_2 and powder-metallurgy processing was based on limited development work. However, because of the design of the Geneva Conference fuel element, the lack of complete manufacturing technology with highly loaded aluminum-type fuel elements and the tight time schedule, no other alternative existed. The difficulties encountered in manufacturing the Geneva Conference fuel elements are now a matter of history. It was shortly discovered that the UO_2 reacted with aluminum causing a gross dimensional change in the fuel plate during fabrication. The problem

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was alleviated somewhat by an improvement in the physical characteristics of the UO_2, and fuel elements were processed without gross changes in dimensions. Nevertheless, the best quality of UO_2 showed signs of slight reaction during fabrication, and concern does exist that reaction may continue during irradiation. A Geneva Conference fuel element is presently under test, and apparently is operating satisfactorily in the MTR lattice. At this writing, it has received an estimated 25% burn-up of U-235 atoms. Because of the rush to fabricate these elements, it was impossible to conduct a systematic investigation to isolate the variables involved, and establish the fundamental reasons for the cause of the reaction between aluminum and UO_2. The unit cost of the Geneva Conference fuel element is estimated at $3,000 and speaks for itself. On the basis of our experience in manufacturing this fuel element, it is not likely that the unit cost can be decreased to less than $1,500 until the yield of acceptable material can be appreciably increased.

Further investigation of manufacturing fuel plates containing high percentages of uranium in the uranium-aluminum alloy is deemed necessary. An alloy containing 25 weight-percent uranium was utilized successfully in the past. The segregation was a bit more severe than that encountered with the 18.5 weight-percent alloy, but it is felt that it will be acceptable in pool-type reactors. With a modification in design of the Geneva Conference fuel element from 18 plates to 14 plates, the uranium-concentration requirements can be decreased to approximately 27 weight percent. The fact that the plates would be approximately 0.100 inches in thickness, and the metal-to-water ratio would be 1:1 is not too desirable; but, for minimum metallurgical development, this type of fuel element appears very promising. It is proposed to generally confine development of this type of fuel element to the physics aspects as related to pool-type reactors. Metallurgical investigations will be restricted to improving the segregation in the alloy.

Because of complete lack of knowledge of manufacturing methods for aluminum alloys containing high weight percentages of uranium, it is proposed that an investigation be conducted to determine the limiting concentration of uranium in the alloy from both segregation and fabrication considerations. In the event an alloy containing 45 weight-percent uranium proves to be sufficiently ductile to be roll-clad successfully, considerable effort will be concentrated on improving the segregation to an acceptable level.

In order to avoid confining the success of this project to development of uranium-aluminum alloys, it is recommended that powder-metallurgical processing of fuel plates be further investigated. In this connection, Dr. M. L. Picklesimer conducted a preliminary survey of the field, and selected the following compounds as possible sources for fueling aluminum plates with uranium:

TABLE I

Compound Number	Compound Formula	gm/cm^3 Density	Percent Uranium in Compound
1	UC	13.6	95.2
2	UC_2	11.7	90.8
3	U_3Si	15.6	96.2
4	U_3Si_2	12.2	92.7
5	$U\text{-}Al_2$	8.2	81.5
6	$U\text{-}Al_3$	6.7	74.6
7	$U\text{-}Al_4$	-	68.8
8	UO_2	10.6	88.1

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The desirable features which are considered in the selection of a compound for powder-metallurgy processing into fuel plates are listed in the order of importance:

1. Stability in the presence of aluminum
2. Pyrophoricity
3. Density
4. Percent of uranium in the compound

The stability of the compound is essential in order to eliminate dimensional changes during fabrication and subsequent irradiation.

A high degree of pyrophoricity would be disadvantageous and lead to uneconomical methods for handling the material.

High density and percent uranium in the compound are desirable to minimize the volume of compound in the aluminum matrix, and in turn ease fabrication problems.

A review of Table I reveals that all compounds except Nos. 5, 6 and 7, have desirable requirements in density and percent of uranium in the compound.

The preliminary work of Dr. M. L. Picklesimer has revealed that UC_2 appears to be very promising. Aluminum compacts, prepared with UC_2 and containing approximately 50 weight-percent uranium, have shown no instability after a 24-hour heat treatment at 630°C. Fuel plates, prepared from similar compacts, have been satisfactorily fabricated and heat treated for 24 hours at 630°C with no dimensional changes. Although the compound is somewhat pyrophoric, simple procedures for handling in an inert atmosphere can be utilized with no difficulties. This compound appears to be the solution to the problem; however, it is to be emphasized that the investigation has been of a preliminary nature, and extensive development work and testing will be required before a reliable fuel element can be marketed.

The compound, UC, potentially may be more suitable than the compound, UC_2, since it has a higher density and a higher percentage of uranium in the compound. A preliminary study of this compound is in process.

The compound, U_3Si, appears to be unfavorable, since it exhibited a reaction with aluminum. A similar effect may also be observed with the compound, U_3Si_2.

The uranium-aluminum-type compounds do not appear promising because of lower density and percentage of uranium in the compounds. However, a preliminary investigation should be conducted to determine the compatibility of these compounds with aluminum.

UO_2 has satisfactory density and sufficient percentage of uranium, but has exhibited instability during the manufacturing processing of aluminum fuel elements. The degree of instability appears to be related to the physical characteristics of the UO_2 particle. At the present time, the fundamental cause or causes of the dimensional changes which occur during the fabrication of UO_2-aluminum fuel elements is not completely known. It has already been demonstrated that fuel elements of this type can be manufactured. However, until the mechanism of UO_2-aluminum reaction can be fully understood and the yield of acceptable material increased, considerable expense and some risk will be attached to this fuel element. It is felt that further efforts should be extended to improve the cost and reliability of this element.

During the manufacturing of fuel plates, the present practice is to remove the hydrogen trapped in the plate by fluxing and annealing. This procedure generally results in pitting of the plate surface, and thus introduces possible sources for galvanic-corrosion attack. It is felt that the integrity of existing or future aluminum fuel plates can be improved by development of methods to either eliminate flux annealing or minimize the effect of the flux on the surface quality during this treatment.

The status of the fabrication of foreign-reactor fuel elements is not well defined at the present. There exists a real need for development work. The Fabrication Laboratories have been conducting a portion of this work on extremely limited funds; and, unless further appropriations are available in the very near future, progress in this direction will soon be at a standstill. In a recent visit, Mr. Howard Schwartz, of the Washington AEC, expressed a great desire for ORNL to develop a reliable manufacturing method as soon as possible. Within the next month, it will be possible to place three technical men and two technicians on this type of work, compared to one technical man and one technician at the present. It is proposed that, for the balance of fiscal year 1956, $100,000 be appropriated for investigation of aluminum-fuel-element manufacturing methods for foreign-reactor application. Under this program, the following investigations would be conducted:

1. Development of manufacturing methods for aluminum fuel elements containing 27.5 weight-percent uranium in the uranium-aluminum alloy.

2. Investigation to determine the limit of uranium additions to the uranium-aluminum alloy, based on segregation and fabrication difficulties.

3. The application of UC and UC_2 compounds in powder-metallurgy processing of aluminum fuel elements.

4. Development of methods for improving the yield of acceptable UO_2 for aluminum-fuel element processing.

5. The mechanism of the reaction between UO_2 and aluminum at elevated temperatures.

6. The mechanism of the reaction between U_3Si and U_3Si_2 and aluminum at elevated temperatures.

7. The application of U-Al_2, U-Al_3, or U-Al_4 compounds in powder-metallurgy processing of aluminum fuel elements.

8. Elimination of pitting in aluminum fuel plates as a result of flux annealing.

As soon as funds are made available, efforts will be concentrated on projects 1, 2, 3, 4 and 8, with limited efforts on projects 5, 6 and 7.

The reactor-power and other design limitations on foreign pool-type reactors is not clearly defined at the present time. The Geneva Conference Reactor has performed satisfactorily at 100 kilowatts, and the possibility exists that it could be operated

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successfully at 500 kilowatts. However, before any reliable information could be provided, it would be necessary to conduct further experiments at power levels between 100 kilowatts and one megawatt.

On the basis of present experience, the design of pool reactors is limited to the BSR-type fuel unit. Elements with less plates than the BSR element would probably cause a decrease in the peak-power level of a foreign-pool reactor. The possibility does exist that improvement of circulation in the fuel elements will result in an increase in attainable power.

In connection with the inquiries received from the Danish Commission, we submit the following information;

1. Ratio of Aluminum to Uranium in the Fuel Alloy

   In the event a 14-plate element is acceptable, plates can probably be fabricated from an aluminum-uranium alloy containing 27½ weight-percent uranium.

   Experience also has demonstrated that fuel elements containing 48 weight-percent uranium in the form of UO_2 can be manufactured, although further development appears essential.

2. Thickness of Aluminum Canning

   Examination of a dummy and two active fuel elements, which were exposed in the BSR for two and three years, respectively, disclosed pits deep enough to penetrate the 20-mil cladding. However, water-purity control during this period was quite haphazard. Recent examination of some Geneva Conference fuel elements, exposed in the BSR (demineralizer has been installed which controls the resistivity of the water to approximately 100,000 ohms), has disclosed a pit penetrating 12 mils into the cladding, as well as shallow pits, after 6 weeks' exposure. Another Geneva Conference element was exposed in the LITR for 15 weeks, and showed only superficial pitting attack approximately 2 mils deep. The LITR elements are cooled with circulating water with a resistivity greater than 500,000 ohms.

   Experience with aluminum fuel elements has been quite varied; however, there appears to be no good reason why fuel plates with 20-mils aluminum cladding should not be satisfactory in pool-type reactors for a number of years, providing water purity and material control are exercised.

3. Shapes in Which Fuel Elements can be Manufactured

   Experience with pool reactors has been limited to plate-type fuel elements.

4. Range of Widths in Which Plate-Type Fuel Elements can be Manufactured

   The width of fuel plates in all pool-type reactors has been approximately three inches. Flat plates, approximately one-inch wide, were

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fabricated for Phillips Petroleum Company in their SPERT element. ORNL is confined to manufacturing plates with a three-inch maximum width because of equipment limitations.

With proper equipment, it is very conceivable that plates six inches in width could be manufactured.

5. Radiation Damage to Fuel Elements

Investigations conducted on LITR and MTR fuel plates containing 18.5 weight-percent uranium have revealed that both neutron and fission-fragment damage occurs. However, the damage is not so severe to cause any noticeable distortion or warpage of the fuel plates.

The only available information on elements containing uranium enriched in U-235 to the 20% level are the recent results obtained from examination of the Geneva Conference Reactor test element in the LITR. Limited information is also available on a similar element tested in the MTR.

These elements contained 48 weight-percent uranium in the form of UO_2. The LITR element was in the reactor for 83 days and received approximately 5% burn-up of U-235 atoms. The MTR element was in the reactor for 42 days and received approximately 30% burn-up of U-235 atoms. The LITR element appears to be in excellent condition after its irradiation period. At this writing, the MTR element has completed its testing after an uneventful period in the reactor. A visual examination will be made shortly to determine whether or not any gross damage occurred during irradiation.

6. Cladding Failures

There has been no leakage of fission products which has been shown to be a result of defective cladding.

Very truly yours,

OAK RIDGE NATIONAL LABORATORY

M. E. RAMSEY

for C. E. Larson
Director

CEL:RJB:mw

cc: H. M. Roth (2)
C. E. Center
A. M. Weinberg
R. A. Charpie
Beas Stringfield, Jr.
J. H. Frye, Jr. (2)
R. J. Beaver
C. E. Larson (2)
Laboratory Records (2)

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