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LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA ○ LOS ALAMOS NEW MEXICO

QUARTERLY STATUS REPORT ON
ADVANCED REACTOR TECHNOLOGY (ART)
FOR PERIOD ENDING JANUARY 31, 1965

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LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA LOS ALAMOS NEW MEXICO

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All LAMS reports are informal documents, usually prepared for a special purpose. This LAMS report has been prepared, as the title indicates, to present the status on LASL Advanced Reactor Technology. It has not been reviewed or verified for accuracy in the interest of prompt distribution. All LAMS reports express the views of the authors as of the time they were written and do not necessarily reflect the opinions of the Los Alamos Scientific Laboratory or the final opinion of the authors on the subject.

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INTRODUCTORY NOTE

This is the first of a new series of LASL quarterly reports on Advanced Reactor Technology (ART). It includes material previously contained in the Plutonium Reactor Fuel Development and Ultra-High Temperature Reactor Experiment (UHTREX) reports, and the unclassified portions of the Plasma Thermocouple report. These three reports will, therefore, not be issued in the future. New activities at LASL of interest to reactor technology development, such as possible magnetohydrodynamic applications and heat pipes, will be described in the ART quarterly report.

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I. PLUTONIUM REACTORS AND FUELS

LAMPRE

Flip-Flop Effect

During operations with the Mark II core in LAMPRE, it was observed that, under ostensibly steady-state conditions, the core always possessed two distinct equilibrium thermal modes of operation. Transitions between the two states occurred at what seemed to be random time intervals, and the transition times were relatively rapid, i.e., the elapsed time during a transition was too short to be resolved by conventional recording equipment. The most pronounced effects of the transitions were seen in the shifts in core outlet thermocouple readings where temperature changes as large as 30°C were observed. Small changes in the nuclear power level were also detected. These phenomena were designated the "flip-flop" effect.

Analysis of data on the time intervals between transitions indicates that, for fixed operating conditions, the probability per unit time of transition from a given state to the other state was a constant. Mean lifetimes of 14.8 and 19.0 min were found for the two states during Run 57 at 1 MW with a fixed Na flow rate.

In addition to providing a means for summarizing the data on state lifetimes, such analysis can be used for determining whether significant changes occurred in the lifetimes during the course of the Mark II core operation.

Capsule Worths vs Radius

During the search for a failed fuel element, the multiplication change $\Delta k/k$ caused by raising a capsule 10 in. was observed at various core lattice positions. The data have recently been reevaluated to give the capsule worth as a function of radial position in the core.

In general, each radial point represents the average of measurements for six capsules in corresponding core lattice positions. As an indication of consistency of measurement, the worth of each of the capsules was compared with the average for its group of six. The maximum such deviation was $2 \times 10^{-4} \Delta k/k$ (10%), and the average deviation was 0.7×10^{-4} (3.5%). The failed fuel capsule differed by 27×10^{-4} (133%) from its group's average. Although the relative worths are reliable to about 1% (0.7×10^{-4}), the absolute values are known only to the precision of the multiplication calibration of the counter used. This uncertainty may be about 5% (3×10^{-4}).

Melting Point Data

The melt-freeze data obtained in LAMPRE have been reviewed. It is concluded that melting points of the fuel extended over temperatures ranging from 414° to 435°C . The variation in melting points observed is attributed to a spread in the actual constituent fractions of Fe alloyed with the Pu. The results illustrate the desirability of performing in-place melting point observations on future molten Pu cores in order to establish definitive low-temperature operating limits.

FAST REACTOR CORE TEST FACILITY

Construction

On January 31, construction of the FRCTF was 58.2% complete compared with 84.9% scheduled. Efforts are being made to have the contractor accelerate his work but little improvement has been shown.

It is doubtful that the construction will be completed on schedule (May 18, 1965), July or August being more realistic.

In general, the quality of the construction work is very good, with the exception of welding and radiographing of the reactor piping systems. To date, 2004 radiographs have been submitted by the contractor, but only a few of these have met specifications. After considerable correspondence and discussion, the general contractor obtained a new subcontractor to perform this phase of the work. The new subcontractor began work in the middle of January and the first few submittals appear to be within the specifications, indicating that the problem may have been solved.

Concrete work for the basic structure is practically complete and the contractor is now concentrating on the installation of mechanical and electrical systems within the building.

Phase B engineering, which constitutes installation of equipment after the building is complete, has been delayed due to the lack of a firm Na system design. Progress is being made, however, in some of the electrical equipment and piping systems which are independent of the Na system.

Phase B

Intermediate Heat Exchangers (IHx)

The 5-MW IHX has been delivered. Fabrication has been completed on the 15-MW unit and the factory mass spectrometer leak tests were started on January 20. The complete shipment should be received early in February.

Piping Systems

Additional system studies have been made in order to maximize primary system Na flow and hence minimize reactor Na temperature rise. These calculational studies are based upon use of components which are

on hand or are now being fabricated, viz., pumps, IHX's, heat dumps, surge tanks, flowmeters, etc.

The addition of a 3-in. IHX bypass pipe is proposed in order to improve maximum Na flow capability. Calculations indicate that the bypass will reduce Na system ΔP at 2500 gpm from 42 to 32 psi.

New piping drawings have been started which will reflect modifications in equipment layout, increased piping sizes, and new piping arrangement. An analysis has been made of a mixing chamber to be used to mix the bypassed (1100°F) Na with that from the IHX outlet (840°F).

Thermal Expansion Tanks

The vendor reports that thermal expansion tank fabrication is progressing satisfactorily although somewhat behind schedule. It is estimated that all four tanks will be shipped in February 1965.

Emergency D.C. Pump

Details of performance requirements of the emergency d.c. pump were discussed with the vendor. This battery-operated pump will supply primary coolant circulation for decay heat removal during site electrical power outages of up to 12 h. The specified performance of 50 gpm at a head of 0.55 ft of Na is to be available at the end of 12 h of operation. Secondary coolant circulation will come from natural thermal convection.

Data System

The repeat at IASL of the factory tests of the Data System was completed satisfactorily on January 20, 1965. In general, the system accuracies, reliability, and performance seem to be as good or better than they were at the factory. However, the reed-relay low-level multiplexer and the typewriters still have questionable reliability. Because of this, it has been requested that the 6-month reliability

demonstration option be performed. The supplier is to be allowed up to 9 months to achieve a 6-month operating period during which the entire system shall be available 99% of the time and no more than three failures shall occur (excluding some electro-mechanical devices, light bulbs, etc.).

Criteria and Design Data Collection

Considerable effort has been devoted to assembling criteria and design data for both the facility and reactor. The collection, to be continuously updated, will furnish information needed for design and planning.

Reactor System - Core I

Core Heat Transfer

It is necessary to compute the heat transfer for each core design in order to determine if the design parameters are compatible with the limits set for maximum fuel-container wall interface temperatures and maximum thermal stress in the container wall. The core heat transfer code (HTHX) used in these calculations has been revised and now incorporates the following features:

1. The Na coolant Nusselt number is calculated using the correlation of Dwyer and Kalish (BNL-8116) for pitch/diameter (P/D) range of 1.3 to 3.0. $[Nu = 6.66 + 3.126 (P/D) + 1.184 (P/D)^2 + 0.0155 (\bar{\psi} P_e)^{0.86}]$ and is reduced by 10% as recommended by Dwyer for $P/D \geq 1.2$ and $P/D < 1.3$.

2. The circumferential tensile thermal stress at the outer surface of the fuel capsule wall is calculated using material properties for the temperature at the container surface.

Calculations have been made to aid in establishing design values for P/D, D, ρ_F (g Pu/cm³). In these calculations the following parameters were established:

Core power = 20 MW

Primary sodium flow rate = 2500 gpm

Number of fuel capsules per module = 7

Core inlet Na temperature = 480°C

Max. allowable fuel-container wall interface temp. = 700°C.

The independent variables were set at:

$P/D = 1.20$ and 1.25

$D = 0.4$ in. and 0.5 in. (D = outside diameter of fuel capsule)

Container wall thickness = 0.0225 in.

$\rho_F = 6.2$ g Pu/cm³ fuel and 8.0 g Pu/cm³ fuel

Corrugated and straight sided hexagonal modules

Module wall thickness 0.025 in. with 0.012 in. module separation

Core height = 12 in.

The critical mass was estimated by the relation $M_c = 270 (\rho_F V_F)^{-1.3}$, where V_F , the volume fraction of fuel, for a straight sided hexagonal module is given by

$$V_F = \frac{6.348 (D - 0.045)^2}{(2.732 P + 0.062)^2}$$

and for a corrugated hexagonal module by

$$V_F = \frac{0.5291 (D - 0.045)^2}{0.5834 P^2 + 0.031 P + 0.006}$$

The results of these calculations have been used to establish the following design parameters:

Reactor power

Core: 19 MW thermal

Thermal shield and reflector: 1 MW

Total: 20 MW

Coolant flow rate through core: 1.05×10^6 lb/h

Inlet coolant temperature: 480°C

Container-fuel interface temperature: 700°C

Fuel element modules: corrugated hexagonal

Fuel capsule

Diameter: 0.4 in. o.d.

Wall thickness: 0.020 in. minimum; 0.022 in. nominal

Number/module: 7

Spacing, P/D: 1.25

Pu density in fuel: 6.2 g/cm^3

Doubling Time of Breeders Using FRCTF-Type Cores

The factors in the doubling-time expressions given in the last quarterly report (LA-3208-MS) have been reevaluated, using the latest obtainable materials constants for Ta. The resulting expressions are

$$\text{DT}(\text{yr}) \approx \frac{65,000 \text{ b}(\text{cm})}{\text{HF} (\text{W/cm}^2)} \cdot \frac{1}{R(0.9 - 0.3R)}$$
$$\text{DT}(\text{yr}) \approx \frac{1.24 \times 10^7 \text{ b}^2}{S_{\text{Ta}}(\text{psi})} \cdot \frac{1}{R(0.9 - 0.3R)},$$

where DT is the time to double the core inventory only, without compounding. As before, b is the Ta wall thickness, HF the core average heat flux, S_{Ta} the average thermal stress in the Ta wall, and R the Ta/Pu atom ratio. These expressions are based on a very approximate empirical relation for the breeding ratio, namely,

$$\text{BR} = 1.9 - 0.3R.$$

Additional assumptions are that the plant factor is 80% and the Pu capture-to-fission ratio is 0.14.

In using these relations, it is assumed that the heat flux or stress limited conditions are achievable. For example, it is assumed that temperature limitations will not set lower limits.

Recent reexamination indicates that the stress limitation may be increased from 5000 psi to 10,000 psi. Based on this value and on the newer factors above, the case of 0.025-in. wall thickness cited in LA-3208-MS might achieve a doubling time of 7.4 yr.

Core Parameter Study

Calculations have previously been made of the thermal behavior, including the hottest fuel-Ta interface temperatures, for a few possible FRCTF cores. In order to extend these calculations to show the effect of systematically varying one or more parameters, some approximate general relations were developed. Among the parameters included were fuel capsule o.d. and wall thickness, pitch-to-diameter ratio, operating power, allowable temperature difference between Na inlet and hottest fuel-Ta interface, and module can wall thickness and clearance. Flow is assumed constant and equal to that of a chosen reference case which has been calculated in detail by the heat transfer code (HTHX) now in use. Critical mass is assumed to vary as $\rho^{-1.3}$, where ρ is the average Pu density in the core. The effect on heat transfer of any change in Na velocity is neglected.

Although the treatment is an approximate one, the following qualitative conclusions may be drawn. For any chosen capsule diameter, power density increases with fuel density, and the optimum capsule diameter decreases as the fuel density is increased.

Module Can Calculations

The flow effects of the clearance between the outer fuel capsules of a module and the surrounding can have been evaluated to determine the frictional drag of the can wall. Some related calculations are given in Table 1. These are all for 0.400-in. o.d. capsules with 0.022-in. walls on a 0.500-in. pitch; the module can wall thickness is 0.025 in. and a clearance of 0.012 in. is allowed between module cans.

Table 1. Comparison of Module Can Characteristics

Description	Recent Designs		Present Proposals	
	Clearance	v/o Fuel	Clearance	v/o Fuel
Hexagonal	0.050	39.5	0.072	37.1
Corrugated	0.050	41.5	0.085	37.3

The proposed designs incorporate clearances making the mass Na flow rate for the poorest cooled 60° arc of any fuel capsule equal to that for the central capsule. Variations in flow rate per 60° arc are probably greater for the hexagonal cans.

Power Flattening (Zoned Core) Calculation

A single 2-D calculation was made to see if a zoned core loading might be of interest. For reference, problem No. 2700 was used; it contained 5.7-g Pu/cm³ fuel and a 20.5-cm radius. The region beyond 13 cm was loaded with fuel having 12% higher Pu density and a new critical radius was calculated by DDK, Problem No. 2709. If operated at the same power as the reference core, the central power density was increased about 4% and the maximum power density in the denser fuel was about 7% below that in the reference fuel at the core center. The peak-to-average power density ratio was 1.33 for the 2-zone core, compared to 1.47 for the 1-zone case.

Extrapolating from these results, it was estimated that a 20% fuel density step would have brought the maximum power density in the outer zone up to that at the core center and that this power density would have been some 6% greater than that of the reference core operating at the same total power. The peak-to-average power density ratio should be reduced somewhat further.

Shielding

Nineteen CRAM problems, in spherical geometry, have been run to evaluate thermal shield configurations for the latest FRCTF reference core design. Table 2 gives the geometry used in the calculations. The radial shielding at the FRCTF core height was used in the spherical calculational model. Results for three shield configurations of interest are given in Table 3.

Table 2. Regional Specifications for Spherical
Model FRCTF Shielding Survey

<u>Region</u>	<u>Description</u>	<u>Thickness (in.)</u>	<u>Composition</u>
1	Core	21.16	Fuel-Ta-Na-Stainless Steel
2	Margin	3.07	Ni-Na-Stainless Steel
3	Flow Divider	5.81	Na-Stainless Steel
4	Reflector	20.32	65 v/o Ni, 35 v/o Na
5	Shield	Variable (20.32)*	Variable (80 v/o SS, 20 v/o Na)*
6	Shield	Variable	Variable
7	Vessel	5.08	Fe
8	Borated graphite	Variable	1.28 g/cm ³ C with 0.93 w/o B (corresponds to 80 v/o borated graphite and 20 v/o void)

* Values used to obtain results in Table 3.

Table 3. Shielding Calculations for FRCTF
Based on Model in Table 2

Problem	Region 6		Region 8		Neutron Leakage (%)	Percent Absorption in Region				
	t*	Material	t*			4	5	6	7	8
C-12	16	80 v/o SS (1 w/o B) 20 v/o Na	24		0.032	20.4	11.3	16.5	0.13	1.40 (11)**
C-14	16	80 v/o SS 20 v/o Na	24		0.052	20.6	12.9	12.9	0.45	2.78 (22)**
C-19	12	80 v/o SS (2 w/o B) 20 v/o Na	32		0.0069	20.3	10.9	16.6	0.16	1.79 (14)**

* t = radial thickness of region in inches

** kW deposited in region at 20 MW reactor power

One of the criteria in evaluating the adequacy of the shielding is the energy current (neutrons and gammas) incident on the walls of the reactor pit. For the neutrons, at a reactor power of 20 MW, the maximum energy current for the cases cited in Table 3 is 1.2 mW/cm². This figure applies at the outer boundary of the system, approximately 6-ft radius, and is well below the allowable 20 mW/cm². Calculations of the gamma-ray energy current have not as yet been made, but estimates from Monte Carlo problems for similar systems indicate that this contribution will be a few mW/cm².

Another item of importance is the heat deposition in the borated graphite region of the shield. Considering only the heat deposition resulting from the B¹⁰ (n, α)Li⁷ reaction, a 1% neutron capture rate at 20 MW is equivalent to approximately 8 kW. As can be seen from Table 3,

the maximum heat deposition in Region 8 occurs for the design with the non-borated stainless steel, Problem C-14.

Additional calculations of the types discussed above are planned. When a reasonably firm shield design is arrived at, Monte Carlo calculations (MGC code) in cylindrical geometry will be performed to obtain the heat deposition and energy escape for the gamma rays. The question of burnup of B in the borated steel and the effect of neutron irradiation on 1 and 2 w/o B steel will also be investigated.

Large Core Study

A preliminary analysis has been made of the implications of a substantial decrease in container parasitic capture for large molten Pu systems. The results indicate that characteristic module sizes increase rapidly from the present 50-100 MW (th) for 4 g Pu/cm³, with presently understood stress and flow velocity limits. For 2 g Pu/cm³, the indicated characteristic size could be in the thousands of MW. Since this is a surprising and long extrapolation, a few simple neutronic calculations will be made to refine these estimates.

Fuel Capsule Development

The design of the Pu fuel capsule for the FRCTF is not completely firm, but it appears to be a tube approximately 24 in. long having an o.d. somewhat greater than 0.40 in. diam and a wall thickness of 0.025 in. The material may be an alloy of Ta containing 5 to 10 w/o W and possibly a minute amount of Y. The Pu would occupy approximately half the volume of the container.

Alloys of Ta-W-Y have been ordered as rods and tubes from two vendors for evaluation. Unfortunately, deliveries from both vendors have slipped badly so that evaluation of the material cannot be started for at least 6 to 8 weeks. Pure Ta and Ta-W alloys have become available recently for development work.

One end of the tube can be sealed prior to Pu alloy addition but the other end of the tube must be sealed after the Pu has been added; the seals must be as resistant to Pu alloy attack as the body of the tube. In the LAMPRE I reactor the capsules were formed as closed-end tubes so that only one weld seal was necessary. The length and diameter of the FRCTF capsule makes this approach extremely difficult. However, a limited amount of work was performed on the Dynapak machine and on a draw bench in an attempt to prepare a closed-end tube.

Heavy-walled closed end tubes were pulled through dies on a draw bench in an attempt to thin the tube wall and lengthen the tube. The experiments were performed on pure Ta, the only suitable material available. Without describing the several variations employed in the tube drawing operation, the net result was that the pure Ta lacked sufficient tensile strength to withstand the drawing forces and the tube pulled apart during the operation.

A co-extrusion process was attempted on the Dynapak machine, in which a hollow, closed-end Ta billet was filled with steel or Al, the billet encased in steel and then extruded. A limited amount of success was obtained in this effort but it is apparent that a long development program would be involved to obtain the desired capsule. In view of the time scale, this program will be phased out.

A more practical approach to FRCTF capsule fabrication appeared to be the use of high quality tubing of the desired composition and the development of techniques for sealing the ends of the tube. Welding is a definite possibility and there was some hope that solid state diffusion bonding would be beneficial. In this latter case, the molten zone is absent and the joint area would be similar to that of the parent metal and probably less susceptible to corrosion by the fuel.

Plugs of various configuration were designed for insertion into Ta tubes. The tubes were then collapsed around the plug by the magnetic compaction technique which produces an excellent mechanical bond. It is

essential in diffusion bonding operations that metal-to-metal contact be provided to permit diffusion to occur. In spite of changes in plug design, variations in power of the magnetic pulse, additives to the surface to promote diffusion and other devices, the Ta tube exhibited just enough elastic springback after compaction so that a very slight gap was present at the interface of the tube and the plug. Consequently, spotty rather than continuous solid state diffusion occurred when the assembly was heated to high temperature in vacuum. Although work is continuing on the diffusion bonding technique, it does not appear to be too promising at this time.

The most logical approach, pending actual evaluation at temperature in the presence of Pu alloys, is that of welding the end plugs onto the tube. The plug material, in all cases, would be the same Ta alloy as the tube. A new plug design has been proposed for the bottom end of the tube which is in contact with the molten Pu alloy. It is much longer than anything used previously and is a modified taper with a much smaller diameter at the exit end of the tube. Magnetic compaction of the tube around the plug involves a relatively small diameter of tube-plug weld at the very end of the tube, and this should be accomplished without difficulty. The compaction of the tube around the plug provides an excellent fit of the two components and a relatively long path from the Pu interface to the weld. Consequently, it is the hope that the fuel would never reach the actual weld area. It is quite possible that this approach would be successful although the proof still lies in testing at temperature with Pu alloy in the tube.

It has been argued that the seal at the top end of the tube is not as important as at the bottom since it is not in direct contact with molten Pu alloy and is a considerable distance away from the fuel. Here, again, a fusion weld could be employed after the Pu fuel has been added to the capsule. The design of this joint is not yet as well developed as the bottom seal described above.

FUEL AND CONTAINER DEVELOPMENT

Omega West Reactor Experiment (OWREX)

The purpose of OWREX is to test possible reactor fuels and containers in the Omega West Reactor, as described in LA-3208-MS, page 10. Assembly of the first outer containment and heat transfer unit for OWREX has been completed and that of the second unit has begun. X-rays taken of the first unit show that all tolerances were maintained during assembly. In the second unit, several minor design modifications will be incorporated to simplify assembly procedures.

Fuel Alloy Preparation

Methods have been developed for the preparation of homogeneous Pu-Ce-Co alloys of the following compositions:

Pu Density, g/cm ³	Composition (w/o)		
	Pu	Ce	Co
5.0	50.6	39.4	10.0
6.2	57.7	32.9	9.4
8.0	68.2	23.5	8.3

The three elements are melted and stirred in a bottom-pour Ta crucible in an inductively heated vacuum furnace. The molten alloy is poured onto a graphite distributor sprue and runs into a multiple rod mold of anodized Al at ambient temperature. The chill-cast product has a small-grain structure. The analysis of a typical batch (25 rods 0.35-in. diam, 10-in. long) is as follows:

Sample	Pu(w/o)	Ce(w/o)	Co (w/o)
Top of First Rod to Fill	57.68 $\pm$ 0.03	32.68 $\pm$ 0.06	9.44 $\pm$ 0.02
Bottom of First Rod to Fill	57.69 $\pm$ 0.03	32.69 $\pm$ 0.06	9.43 $\pm$ 0.02
Top of Last Rod to Fill	57.68 $\pm$ 0.03	32.78 $\pm$ 0.06	9.43 $\pm$ 0.02
Bottom of Last Rod to Fill	57.68 $\pm$ 0.03	32.76 $\pm$ 0.06	9.38 $\pm$ 0.02

Fig. 1. Volume expansion of Pu-Co-Ce fuels on freezing

Fig. 2. Densities of stabilized Pu alloys

Density of Pu Fuels

The volume expansion on freezing of Pu-Co-Ce alloys containing 0-8 g Pu/cm³ is shown in Fig. 1. It is evident that increasing the Pu concentration increases the percent expansion in this density range. An attempt will be made to obtain lower volume expansions by replacing part of the Ce with Mn. A number of these quaternary alloys have been prepared with melting temperatures below 450°C, and volume expansion on freezing will be measured with the NaK volumeter.

Density curves for stabilized and unstabilized Pu and Pu-2.55 w/o Fe alloys are shown in Fig. 2. Since the δ -Pu phase is stabilized by Ga addition, the stabilized solid phases in the two systems are less dense than are the corresponding unstabilized materials below 320°C. A 1% Ga

addition does not completely stabilize pure Pu in the δ phase above 500°C. However, stabilized Pu is considerably less dense than the unstabilized material in this region.

The Ga additions give a wider melting range than exists in the unstabilized systems. There are also differences in volume expansion on freezing between stabilized and unstabilized materials. The unstabilized Pu-Fe alloy expands 1.4% on freezing, as compared to 0.4% for the alloy containing 0.2% Ga. The density of molten Pu at 665°C is 16.38 g/cm³, whereas that of the Pu-1% Ga alloy at this temperature is 16.04 g/cm³. Thus it appears small Ga additions exert a large effect on the densities of Pu and Pu-Fe alloy in the molten state by short-range ordering.

Containment Studies of Pu-Co-Ce Fuels

Sodium loop testing of Ta, Ta-5% W, and Ta-10% W alloys with Pu-Co-Ce alloys containing 5, 5.7, 6.5, and 8 g Pu/cm³ have been carried out at 600° to 750°C for times as long as 9200 h. Of 77 capsules tested, there have been 7 failures. All of the latter were after relatively short test periods (2800 h or less), and were associated with welds.

Fifty-nine corrosion experiments have been performed in inert-gas atmosphere furnaces. The materials investigated were similar to those tested in the Na loops; test temperatures were as high as 850°C and times up to 4500 h. Consideration of the results leads to the following conclusions:

1. All failures except one (Ta containing 6.5 g Pu/cm³ fuel) were associated with welds, predominantly the top seal weld.
2. Only two of 19 capsules tested at 600°C failed. These failures were Ta capsules containing 5 g Pu/cm³ fuel to which 500 ppm C (as Ce₂C₃) was added.
3. The addition of C to the fuel did not improve corrosion resistance even at 600°C.

4. The Ta capsules that were tested at 800°C and higher all failed. All but one of these failures were in the welds. (See 1 above.)

A carbide layer on Ta appears to improve its corrosion resistance to molten Pu-Fe and Pu-Co-Ce alloys. The carbide layers that have been applied to Ta are 5-15 μ in thickness. A number of capsules pack-carburized for 15 min in vacuum at 1800°C were tested with 5.0 and 6.5 g Pu/cm³ fuels. The results of these tests are summarized in Table 4.

Table 4. Corrosion Tests on Carburized and Uncarburized Capsules

Capsule Material	No. of Capsules	Surface	Pu Content (g/cm ³)	Temp. (°C)	Time (h)	Remarks
Ta	1	Uncarburized	6.5	750	3000	Leaked (gas space)
Ta	1	Carburized	6.5	750	3000	No leaks
Ta-10 % W	2	Carburized	5.0	750	4500	No leaks
Ta	1	Uncarburized	6.5	850	1000	Leaked (top seal weld)
Ta	1	Carburized	6.5	850	3000	No leaks
Ta-10 % W	2	Carburized	5.0	850	4500	One leaked (top seal weld)

In summary, it is observed from Pu-Co-Ce alloy corrosion results presently available that:

1. The 136 corrosion tests with Pu-Co-Ce alloys have resulted in 24 failures of which 23 have been in welds.

2. Carburization of capsules enhances their corrosion resistance, probably by protection of seal welds below the fuel phase.

3. Tungsten additions to Ta improve weld performance in that no Ta-5% W (of nine tested) and one Ta-10% W (of ten tested) have failed at temperatures up to 850°C. The one failure noted was the top seal weld of a carburized Ta-10% W capsule tested 4500 h at 850°C.

Containment Studies of Pu-Fe Fuel

A total of 132 capsules containing Pu-Fe fuel have been tested in the Na loops at 600°, 650°, 700°, and 750°C. There have been 84 failures in these tests. A summary of the temperature and types of failure is given in Table 5.

Table 5. Corrosion Tests with Pu-Fe Fuel

Temp. °C	Total Capsules	Capsules that Failed	Area of Principal Leakage		
			Weld and Gas Space Immediately Below Weld	Fuel Level and Gas Space Immediately Above Fuel Level	Fuel Phase Below Liquid Level
600	31	10	8	2	0
650	42	31	14	11	6
700	51	39	8	21	10
750	<u>8</u>	<u>4</u>	<u>4</u>	<u>0</u>	<u>0</u>
	132	84	34	34	16

It is evident that corrosion by Pu-Fe fuel causes failures in all parts of the capsules, rather than exclusively in the welds, as was observed with Pu-Co-Ce fuels. Of the materials that were tested at 700°C and higher, only Ta-0.1% W-9 ppm Y and carburized Ta did not exhibit any failures within 2800 h.

Accelerated Container Corrosion Tests

Accelerated corrosion tests of Pu, Pu-Fe, and Pu-Co-Ce alloys contained in Ta are being run in sufficient numbers to obtain statistically significant data. Tests are being conducted at 900°, 1000°, and 1100°C to establish the temperature dependence of failure time. If such a dependence can be established, failure times at lower temperatures can be predicted. This extrapolation to lower temperatures (600°-700°C) is necessary because of the long time that is required to obtain any corrosion

Fig. 3. Temperature dependence of capsule lifetime

The capsules that lasted for an unusually long time, however, all exhibited evidences of having been carburized by backstreaming diffusion pump oil. Only one capsule was tested with 8 g Pu/cm³ Pu-Co-Ce alloy; this experiment was terminated after 660 h at 1000°C without indications of failure.

Fuel-Sodium Interactions

A Na loop with a topless LAMPRE-size capsule containing 110 g of 5 g Pu/cm³ Pu-Co-Ce alloy was run at 650°C for 1540 h. The Na flow rate was 0.48 gpm. The fuel/Na interface was well defined by radiography throughout the run and was 1.7 in. below the top of the capsule. Sodium samples taken from the loop throughout the run contained < 2 ppm Co, < 4 ppm Ce, and < 0.1 ppm Pu.

data at these temperatures; for example, if welds are omitted from consideration, it appears that test times longer than 400 days at 700°C would be required to obtain a significant number of failures with Pu-Co-Ce alloys. Corrosion data with Pu and Pu-Fe alloy are being obtained over the temperature range of 700°C-1100°C to assure that extrapolation is valid.

The results obtained so far are summarized in Fig. 3. The failures of capsules containing α -Pu fit a straight line plot in a satisfactory manner, but there are some anomalies in the data obtained with Pu-Fe fuel.

At the conclusion of the run, a deposit of alpha-active material was found around the capsule mouth and down the outside walls for a distance of 1 in. Reexamination of radiographs indicated that it was present as early as 10 days after the start of the test. Apparently fuel climbed 1.7 in. up the inside wall from the fuel/Na interface and over the lip of the open capsule (surface tension pumping). The deposit had a composition of 51.5 w/o Pu, 40.5 w/o Ce, 8.0 w/o Co, compared to the original fuel composition of 49.3 w/o Pu, 40.8 w/o Ce, 9.9 w/o Co. Although the stainless steel portions of the capsule holder did not touch the capsule at any point, the entire assembly was highly alpha active.

Four more topless capsules containing 5 g Pu/cm³ Pu-Co-Ce alloy have been loaded into the Na loop. The fuel/Na interface is 4 in. below the top in two of these and 6 in. below the top in the other two. The experimental conditions are the same as those of the first experiment. At the conclusion of the test the creep height of the fuel in the capsules will be measured.

Mass Transfer of Tantalum

A Ta diffusion experiment (see LA-3208-MS, page 7) with 5 g Pu/cm³ Pu-Co-Ce fuel was run for a total of 3358 h. The first one-third of the test was conducted under isothermal conditions at 700°C, and during the final two-thirds of this time a differential of 92°C was maintained along the fuel column with the maximum temperature of 700°C at the bottom. During this run a total of 48 µg transferred from the radioactive tab at the bottom of the capsule to the fuel/gas phase interface. Shortly after the temperature gradient was imposed, the Ta transferred at a linear rate of 3.8 µg/week, which is equivalent to 6.3×10^{-3} mil/yr. However, in the last 6 weeks of the run the transfer rate slowed to approximately 1.4 µg/week. The rate change resulted from the total conversion of the radioactive Ta tab to Ta₂Co. The maximum dissolution rate from the tab occurred within the first 50 h, giving a rate equivalent to 105 µg/week or 0.175 mil/yr.

Irradiation Hardening of Tantalum

In an attempt to obtain the hardness of Ta irradiated in a fast-neutron and high-temperature environment, the post-irradiation hardnesses of four LAMPRE fuel element end caps were measured. These caps were fabricated from National Research Corp. commercial purity Ta. The preirradiation hardness of the material was 75 kg/mm² using a 500 g load and a Vickers indenter. The pertinent data for the samples are listed in Table 6.

Fig. 4. Hardness of neutron-irradiated tantalum

results of tests performed in the MTR at 50°C. The fast flux in these experiments was a factor of 5 to 10 less than the thermal flux, and so the dosage shown is essentially the thermal nvt. The BMI data show two

The present data are compared in Fig. 4 with information published in BMI-1476. The BMI values were the

Table 6. Hardnesses of Irradiated Tantalum Samples

<u>Capsule</u>	<u>V_H as Irradiated</u>	<u>Anneal* Temp. (°C)</u>	<u>V_H Post- Anneal</u>	<u>Calculated nvt (fast)</u>
1194	190	900	172	9.6 x 10 ²⁰
1294	273	800	261	10.2 x 10 ²⁰
1400	249	700	208	8.8 x 10 ²⁰
1597	195	600	176	6.4 x 10 ²⁰

* 1 h at temperature in vacuum.

effects of irradiation: (1) alloying by transmutation of Ta to W and (2) irradiation hardening due to damage. Calculations show that the transmutation effect in the LAMPRE fast flux was about a factor of 100 less than that which would exist in the MTR, since the fast neutron cross section of Ta is 100 times smaller than the thermal value. The hardness increases found in the present measurements are felt to be solely due to a damage mechanism.

The fact that the sample annealed at 600°C decreased in hardness indicates the irradiation temperature of the LAMPRE fuel element end caps was not above 600°C. Calculations confirm this result in that they indicate a 500° to 550°C irradiation temperature.

Tantalum-Yttrium Studies

Corrosion tests on Ta-0.1% W and Ta-0.1% W-9 ppm Y alloys with Pu-Fe fuel have revealed a considerable improvement in corrosion resistance as a result of the small Y addition. The four capsules of Ta-0.1% W that were tested at 700°C failed within 700 h, while three capsules with Y additions survived 4900 h without failure. The effects of Y addition on the properties of Ta and Ta-W alloys are being investigated for possible application to the containment of Pu-Co-Ce fuels.

Annealing studies on 0.020-in. sheet Ta specimens containing 14 to 74 ppm of Y revealed that 1 h at 1500°C was insufficient to cause complete recrystallization, while 1 h at 1750°C was more than adequate for this purpose. There was little grain growth after recrystallization even at a temperature of 2250°C. No loss of Y by vaporization was observed at any annealing treatment up to 1 h at 2250°C.

In many cases the recrystallized grains were highly elongated, parallel to the rolling direction, with irregular grain boundaries. This highly distorted boundary condition is due to Y in the boundaries which inhibits their motion. Electron microscopic examination indicated random precipitates present throughout the samples. There appeared to be

more precipitate in high-Y specimens than in those low in Y; however, there was no evidence of significant grain boundary precipitates in any of the samples. The observed effects are, therefore, probably due to Y adsorbed at the grain boundaries rather than to Y_2O_3 particles.

Both TIG (tungsten inert gas) and EB (electron beam) welds were made on samples of these Ta-Y alloys. There was little refinement of weld grain size over that encountered in pure Ta, indicating most of the Y was lost by vaporization during welding.

Spectrochemical Determination of Y in Ta and Ta-W Alloys

Y can now be determined in Ta or Ta-W alloys by a conventional d.c. arc technique. Using La as an internal standard and an Ar- O_2 atmosphere for arcing, which eliminates interfering CN bands, Y can be measured in the concentration range 6 to 40 ppm with a relative standard deviation of 8%.

II. SUPPORTING RESEARCH: PLUTONIUM STUDIES

Plutonium Nitrides

Plutonium nitride is now prepared routinely by reaction of N_2 gas with partially hydrided Pu metal. Special high-temperature treatments have been incorporated into the preparative method to produce stoichiometric PuN powder which can be ground in air without spontaneous ignition. Pellets of 0.40-in. diam can be pressed to 72% of theoretical density but sintering for 3 h at $1600^{\circ}C$ in 1 atm of N_2 results in little densification. The use of sintering aids is now being examined.

Plutonium Carbides

The phase relationships in the pseudobinary carbide systems PuC-ZrC and PuC-TaC are being investigated. Both systems should form a continuous series of solid solutions. However, the results obtained to date indicate that producing solid solutions of controlled composition in

these systems is very difficult. Preparative methods that have been satisfactory in the past for forming PuC-UC solid solutions are not satisfactory in these cases. Although partial solid solutions have been achieved, the powder metallurgy and arc melting techniques are still being developed.

The structure of PuC (relative to the C atoms) can be determined over the temperature range 80° - 673° K by neutron diffraction. At this time, monophasic PuC powder has been prepared, and the specimen dies and specimen holder have been designed. Preliminary studies with UC-Al cermet specimens indicate that a PuC-Al cermet having about 30 v/o Al powder will be satisfactory.

Plutonium Oxides

Sintering studies on high-purity PuO_2 have been completed. Pellets having a diameter of 0.40 in. can be sintered at 1650°C to at least 95% of theoretical density in slightly oxidizing atmospheres such as dry CO_2 . Sintering in dry Ar, however, causes decomposition to PuO_{2-x} , followed by conversion to $\alpha\text{-Pu}_2\text{O}_3$ and PuO_{2-y} on cooling. The decomposition to PuO_{2-x} can be eliminated by sintering the PuO_2 as a cermet with 5 v/o Mo.

Self Radiation Damage in Refractory Plutonium Compounds

The rate of change in unit cell dimensions of PuO_2 , PuN, and PuC is being measured as a function of time and rate of energy deposition. Data obtained so far indicate that the rate of change of the PuO_2 lattice constant is slightly dependent on the rate of energy deposition in the lattice.

Thermodynamic Properties of Plutonium Compounds

The reversibility of the Pu metal- Pu(III)Cl^- molten salt electrode is fundamental to all measurements of thermodynamic properties of Pu

compounds by emf methods at high temperature. A study of the cell $\text{Pu}|\text{PuCl}_3(\text{LiCl-KCl})||\text{AgCl}|\text{Ag}$ in the temperature range 690-720°K is essentially completed. The data indicate that the cell behaves reversibly, and involves a three-electron change. At 708.6°K the standard free energy of formation of PuCl_3 was found to be -192.4 kcal/mole, with a standard entropy change of formation of -50.5 eu.

Additional thermodynamic data are being obtained on the cell $\text{Pu}|\text{PuCl}_3(\text{NaCl-KCl})|\text{PuFe}_2 + \alpha \text{Fe}$. The results will yield the formation thermodynamics of PuFe_2 .

Analysis of Pu-U-Mn Alloys

A rapid x-ray fluorescence method was developed for determining Mn in Pu-U-Mn alloys using Fe as an internal standard and x-rays from a W-target tube. For the Pu measurement, the technique is similar except that Th is used as an internal standard. The relative standard deviation for Mn is 2% at the 5% level and 0.8% at the 40% level. For the Pu the relative standard deviation is 1.3% if its concentration is no greater than 50%.

Pu can be determined also by an absorption edge method in which the intensities of the secondary K β line for Nb and the K α line for Mo are transmitted through the absorption cell filled successively with H₂O, suitable standards, and the sample solution. Measured transmitted intensities are related to Pu concentration using accepted absorption principles. The relative standard deviation varies from 0.9% to 0.55% for Pu concentrations between 30 and 100%.

Plutonium-Indium Alloys

The results of a study of the Pu-In system are being prepared for publication and are summarized in the phase diagram shown in Fig. 5. The study is now considered to be complete except for determining the crystal structures of Pu_3In_5 and the high-temperature η phase, and reducing the uncertainties in the composition ranges of several of the intermediate phases.

Plutonium-Uranium-Titanium Alloys

Four Pu-U-Ti alloys, in both the arc-cast and heat treated (one week, at 750°C) conditions, were examined by x-ray diffraction and metallographic techniques. The purpose of the work was to determine the approximate extent of the solid solubility of Pu in the intermediate phase U_2Ti , and thus provide some basis for a decision as to whether or not such alloys might be suitable for a replacement fuel element. Although compositions were selected so that the alloys would consist entirely or largely of $(Pu,U)_2Ti$, the observed amounts of the second phase (Pu-U ζ in the heat treated alloys) indicated that the alloys either lost Ti during preparation or that the composition of the intermetallic phase does not correspond to the stoichiometric composition on which the hexagonal crystal structure (AlB_2 type) is based.

The results in Table 7 indicate that: (1) Pu can replace at least 50% of the U atoms in U_2Ti , and (2) essentially no change in the unit cell dimensions, therefore no change in composition, of the $(Pu,U)_2Ti$ took place as a result of the heat treatment.

Plutonium-Borides

Considerable effort has been expended during the past few years on studying refractory compounds containing Pu, but little information has been published about the borides, except for crystal structures and

Fig. 5. Plutonium-indium phase diagram

Table 7. Study of U₂Ti Solubility in Plutonium

Alloy composition (a/o)			Unit cell dimensions (Å)				Arc cast density (g/cm ³)	M ₂ Ti hardness DPHN 25-g load	Approximate volume fraction	
			arc cast		heat treated				{	M ₂ Ti
Pu	U	Ti	a	c	a	c				
33.3	33.4	33.3	4.835	2.871	-		14.42	252	0.25	0.75
22.7	44.0	33.3	4.829	2.859	4.829	2.860	14.76	260	0.1	0.9
10.0	56.7	33.3	4.830	2.855	4.830	2.854	14.95	265	0.05	0.95
17.5	52.5	30.0	4.830	2.855	4.830	2.855	15.33	256	0.1	0.9
U ₂ Ti (literature values)					4.828	2.847				

melting points of compositions corresponding to PuB, PuB₂, PuB₄, and PuB₆. Since no unequivocal evidence has yet been found that proves the existence of metal monoborides having the NaCl type of structure, and since the existence of PuB₁₂ was suspected, a modest program to investigate the plutonium borides was recently undertaken. Five intermediate compounds were prepared, as shown in Table 8.

Table 8. Properties of Plutonium Borides

Compound	Structure	Unit cell dimensions (Å)
PuB ₂	Hexagonal (AlB ₂ type)	a ₀ = 3.1857 ± 0.0002
		c ₀ = 3.9485 ± 0.0004
PuB ₄	Tetragonal (ThB ₄ type)	a ₀ = 7.1018 ± 0.0003
		c ₀ = 4.0028 ± 0.0001
PuB ₆	Simple cubic	a ₀ = 4.1134 ± 0.0003
PuB ₁₂	Face-centered cubic	a ₀ = 7.4843 ± 0.0003
"PuB ₁₀₀ "	Simple cubic	a ₀ = 23.43 ± 0.04

Attempts to produce PuB were unsuccessful. Although face-centered cubic monoborides have been reported for Ti, Zr, and Hf, as well as for Pu, the existence of such a boride phase is questionable because borides usually form only when the B can bond to itself in chains or networks. In the face-centered cubic NaCl-type of structure the bonding is metal-to-B.

The composition of "PuB₁₀₀" is uncertain. An arc cast sample containing 1 w/o Pu and 99 w/o B was examined metallographically and found to contain two phases, one of which was PuB₁₂. When the PuB₁₂ lines were subtracted from the x-ray powder pattern, the remaining lines were completely indexed on the basis of a simple cubic unit cell with $a_0 = 23.43 \text{ \AA}$. Ho, Tb, and Yb are known to form cubic borides near the MB₇₀ composition, for which the lattice parameter has been given on the basis of x-ray powder and single-crystal studies as 11.75 and 23.50 $\text{\AA}$, respectively. Furthermore, cubic MB₁₀₀ phases with a lattice parameter of 16.50 $\text{\AA}$ have been reported for Gd and Yb. The 11.7 or 16.5 $\text{\AA}$ (1.414×11.7) cubic cells accounted for some, but not all, of the lines in the x-ray pattern of the Pu compound. On this basis, the hectoboride (PuB₁₀₀) composition was tentatively assigned to the latter.

Metallographic evidence indicates that: (1) PuB₄ melts congruently; (2) all the other borides, PuB₂, PuB₃, PuB₁₂, and "PuB₁₀₀" melt peritectically; and (3) a eutectic reaction occurs between PuB₁₂ and "PuB₁₀₀".

III. ULTRA-HIGH TEMPERATURE REACTOR EXPERIMENT (UHTREX)

CONSTRUCTION

Leakage Rate

Construction of the UHTREX building was completed and beneficial occupancy obtained November 6, 1964. The only serious deficiency in the construction was the inability of the contractor to obtain a 1% per day leakage rate in the secondary containment as provided in the specifications.

A final leakage rate of 2.64% was reported and the building was accepted by the AEC on this basis. A change order is being negotiated to modify the specifications and relieve the contractor of the 1% leakage rate requirement.

Upon receipt of beneficial occupancy of the facility, IASL conducted additional leakage rate tests and by January it was determined to be between 3.0% and 3.4% per day. A considerable but unsuccessful effort was expended in an attempt to locate large isolated leaks. It appears, therefore, that the design of the facility and the construction methods used are probably not capable of obtaining the desired rate of 1%. No evidence of poor workmanship or negligence has been found which could account for significant leakage.

Rather than continue to lose time by further attempts at repairs, the leakage problem was reexamined in the light of recent analyses of the consequences of the maximum credible accident (MCA). As reported in some detail in LA-3190-MS, the analysis shows that the peak pressure in the secondary containment system after the MCA should be about 2.1 psig, instead of the 4.3 psig level calculated for the original UHTREX safety analysis (LA-2689). Consequently the leakage rate from the building after the MCA should be slightly more than 1.5% per day, according to latest tests conducted at this revised peak pressure. Furthermore, if the guidelines of TID-14844 are followed, the recent MCA analysis would indicate that a substantially higher leakage rate is tolerable (up to about 10% per day). From the combination of these considerations, it was decided that the currently measured leakage rate is tolerable.

Design

Work is continuing on the computer listings of the reactor cooling panel wiring and the gas cleanup transducer wiring. Electrical drawings for the installation of the high-pressure compressors, low-pressure compressors, and the vacuum pump are complete. Design has been started on installation drawings for the primary and secondary He loops. Stress analysis of the loops has been completed.

Design work is approximately 60% complete on the fuel conveyor system. This consists of the drybox, charging conveyor, discharge conveyor, and alpha transfer device.

Equipment Installation

Work on the first phase of the equipment installation was started in January. Over a 9-month period, crews of Los Alamos Constructors Inc., will install the gas cleanup system and its utility systems, the water-cooled wall panels in the reactor cell, supports for the reactor and recuperator, shielding slabs, control and power wire systems, the Minotaur remote maintenance machine, and electrical equipment for emergencies and for providing variable-frequency power to the He blowers.

During the second phase which will start in June 1965, the reactor vessel, the recuperator, heat exchanger, and the rest of the primary and secondary coolant loops will be installed. Concurrently, work on instrumentation for the gas cleanup system will start in the control room.

Completion of the entire UHTREX equipment installation is planned for January 1966.

CRITICAL EXPERIMENT

Plans are nearly complete and the assembly of equipment is well advanced for the UHTREX critical experiment (UCX), which will make use of the actual graphite core and reflector of the UHTREX reactor. The core will be loaded with the same fuel that is later to be used for initial operation. Fuel will be loaded manually, one element at a time, and only five of the 16 control rods will be equipped with the remotely operated drives which have been devised for the experiment.

During the experiment, the calculations of critical fuel loading will be verified, and the calculated worth of the system of shutdown rods will be confirmed experimentally. The threshold of detectability of neutron sources for startup will be investigated. Various geometrical locations of neutron sources will be considered, and the feasibility of restarting the reactor with the inherent source will be studied. As time permits, other measurements will be made to determine the power profiles, reactivity effects of voids, and the temperature coefficient of reactivity.

The core and reflector components have been assembled in the "Reactor Test Pit." Mechanical alignment of the core components has been improved slightly by careful adjustment of clearances between keys and keyways.

REACTOR COMPONENTS

Fuel Elements

Graphite fuel elements containing pyrolytic carbon (PyC) coated UC_2 particles will be used initially in the UHTREX reactor. It is anticipated that, with such particles, the release of fission products (particularly Kr and Xe isotopes) from the fuel during startup and early operations will be considerably decreased.

The diameter range of the coated particles is about 350 to 450 μ , resulting from 100 to 110 μ of PyC on 147-208 μ UC_2 cores. This represents a compromise between the requirements of good particle strength and fission product retention on the one hand, and the need to keep the particle size and volume fraction of particles as small as possible to avoid difficulties in fabrication by the extrusion process, on the other. The PyC on the UC_2 particles is a General Atomic "Triplex" coating. The inner coat is a buffer layer of low-density porous PyC 20 to 30 μ thick, to catch fission recoils, dissipate stresses due to differential dimensional changes, and

provide a void volume to minimize fission-gas pressure buildup. The middle layer consists of 35 to 45 μ of "isotropic" (poorly-oriented) PyC with low permeability to fission gases and good resistance to irradiation damage. The outer layer is fine-grained columnar PyC. The interface between the isotropic and columnar layers is intended to act as a barrier to propagation of cracks should they develop. Particles with this type of coating were selected because of good performance in irradiation tests elsewhere, especially under conditions of severe thermal cycling.

The availability of these coated particles and their suitability for use in LASL's fabrication process were established by procurement and evaluation of GA "Triplex" coated UC_2 particles containing 3 kg of depleted U. Based on this experience, a specification was written for procurement of particles containing 25 kg of 93% U^{235} comparable in quality to the depleted GA "Triplex" particles.

About 70 kg of production material was received as 44 individual coating batches. Each batch was examined microscopically before incorporation into a crossblend. The particles were found to be very uniform in appearance from batch to batch and had shapes which were at least as good as the depleted "Triplex" particles tested earlier. Testing of representative samples from the crossblend showed 0.06 w/o of the U exposed. The entire crossblend was leached in portions with 8 M HNO_3 at 90°C for 8 h, and as a result the exposed U was reduced to 0.01 w/o. The leached particles were crossblended again before portions were split out for use in the fabrication of UHTREX fuel elements. It is anticipated that extrusion of the fuel elements will begin early in February.

The enriched U content of UHTREX fuel elements can now be determined nondestructively by measuring the γ -ray flux by means of a NaI scintillator detector which completely surrounds the element. The integrated current produced in scintillator phototubes is directly

related to the O₂ content. Less than 2 min are required for the measurement with a relative standard deviation of 0.3%.

Reactor Vessel

The UHTREX reactor vessel has undergone its last major stress relief operation. All welds on the vessel proper have been made, inspected, and accepted. Nooter Corp.'s inspection department is now determining the vessel's "best vertical centerline" and "best horizontal centerline." The latter is the centerline through the recuperator and elevator openings. After these centerlines are established and fixed, they will be used as reference lines for all future machining.

A series of DTK problems, in spherical geometry, has been run to estimate the thermal flux at the surface of the UHTREX pressure vessel due to a centrally located Pu-Be source. The geometry used corresponded to a radial traverse of the reactor at the core midheight. In order to compare the effectiveness of a centrally-located source with the natural source in the core, one problem was run with a fission spectrum source uniformly distributed in the core. Results of the calculation are shown below; fluxes quoted are values at the surface of the pressure vessel normalized to one source neutron.

<u>Source</u>	<u>Location</u>	<u>Multiplication</u>	<u>Thermal Flux</u>
Pu-Be	Central	20	9.6×10^{-7}
Pu-Be	Central	10	4.7×10^{-7}
Pu-Be	Central	No U in core	2.0×10^{-8}
Fission Spectrum	Core	10	3.3×10^{-8}

Outgassing Tests of Graphite Felt

Calculated results from an analysis of outgassing tests of Union Carbide's graphite felt are as follows:

Moisture, < 0.01%

Volatile matter not detectable, but < 1%

Ash, < 0.02%.

These results and the thermal conductivity data reported below show that graphite felt is well suited to the UHTREX application.

Thermal Conductivity of Carbons

The thermal conductivity of a selected sample of porous C Grade B-303, supplied by Great Lakes Carbon Co., has been measured in the steady-state apparatus at temperatures to 1150°F. The data are plotted in the top curve of Fig. 6. The values are rather high when compared with other results from similar material. The same material was tested by the thermal flash technique, and the data are plotted in the lower curves of Fig. 6. As with other samples, the results are quite different from those obtained by the steady-state technique and are highly dependent on sample thickness. Analogous results were obtained from measurements on Union Carbide Corp.'s porous carbon, Grade 45.

The completed curve for the thermal conductivity of graphite felt as measured in a He atmosphere is shown in Fig. 7. The curve for a similar test on C felt is shown for comparison.

The flash technique of determining thermal conductivity was used to measure values for dense C. The data, shown in Fig. 8, are considerably below those determined by the steady-state method, which are indicated by a note on the figure. The values for the thinner specimens show a very pronounced break in the range of 900° to 1000°F. Obviously there is a change in heat transfer mechanism in this range leading to an indicated decrease in thermal conductivity with temperature. The

Fig. 6. Thermal conductivity of a selected sample of B-303 porous carbon

Fig. 7. Thermal conductivity of graphite felt in a helium atmosphere

Fig. 8. Thermal conductivity of dense carbon

Fig. 9. Thermal conductivity of insulating carbon, Grade B-204

effect, characteristic of radiation transfer within the sample, also appeared in measurements of the thermal conductivity of Great Lakes Carbon Corp.'s insulating C, Grade B-204, determined by the flash technique for samples of various thicknesses. The curves in Fig. 9 show a distinct effect of sample thickness on the results.

Fuel System

The prototype fuel lock ball valve was assembled for testing with the two-way main seal and after 900 cycles at 300°F and 500 psig He a ball bearing and pinion gear failed in the gear train of the actuator. While the entire gear train is being analyzed for weak points, new gears of the same design and a higher load capacity bearing are being placed in the actuator for resumption of testing.

Elastomer seals are being ordered for testing in the two-way seal configuration in the test fixture. Tests will be made on two different elastomers: Viton 8000-71 and Silastic S-2048; the latter has an additive to improve its radiation resistance.

Tests on Physical Sciences electrical feedthroughs for possible use in the fuel system are proceeding at a slower rate than anticipated. Test results to date have indicated leakage rates of less than 1×10^{-9} cm³/sec of He at 300°F and 500 psid, but only one of the seven feedthroughs has been tested through the complete test program of electrical, pressure, and temperature environments. The voltage breakdown test shows that in air the breakdown voltage from pin to case is 1.5 kV d.c., and under He pressures greater than 50 psig, the breakdown voltage is greater than 1.5 kV d.c. However, at 1 atm of He, the breakdown voltage is 500 V d.c., and once it has been initiated, the breakdown is sustained at 400 V d.c. This relatively low breakdown voltage may not give a high enough safety factor, since these electrical feedthroughs will normally carry 120 V a.c. in an ionizing radiation field for the fuel loader application.

Control Rod Drop Tests

Deceleration tests on a full-size central core plug control rod have been completed. A schematic of the test apparatus is shown in Fig. 10. Thirty-one drops were performed ranging from 33 g to 230 g, with the first sign of failure at 120 g. Beginning at 120 g the inner graphite sheath began to chip, both at top and bottom, until at 222 g, after 12 more drops, the sleeve was broken into several short segments. About 1-1/2 in. of the original length broke into pieces small enough to fall out when the rod was disassembled. The failure could have been a result of the rebound of the impact

plate, as the chipping began where the poison cylinders could hit and where the sleeve itself could impact on the Nb flanged end after a rebound. Because of allowance for differential thermal expansion, the cylinders and the graphite sheath are free to move 1/2 in. relative to the Nb rod. On the thirty-first drop, at 230 g, the upper end of the outer graphite tube broke and the tube fell free until it was caught by the flange on the end of the rod. At this point in the test, the control rod could still be withdrawn with only the loss of the graphite tip and a few pieces of the inner sheath.

The pulse shapes obtained in the test were triangular with a ramp increase of 5 msec duration and a fall-off back to zero of about 1 msec. Two lead pellets were used to shape the pulse. These pellets, 15/16 in.

Fig. 10. Control rod drop test apparatus.

long and $1\frac{3}{32}$ in. in diameter, had one end pointed to a 40° included angle. In the most severe tests, two pellets were smashed to $\frac{3}{16}$ -in. thickness. In drop number 31, when the outer graphite broke, the free fall was 55 in.

Control Rod Materials

Test results indicate that the main Nb support rod in the core plug control rods should be made by swaging and finish-machining of a $1\frac{1}{4}$ in. diam rod rather than by welding. Room temperature tensile tests of welded Nb rod show the as-welded and recrystallized specimens to be strong and ductile. However, after exposure to 2400°F for 4 weeks in an impure He atmosphere containing 3000 ppm CO, 3000 ppm N_2 , 2000 ppm H_2 , and 100 ppm O_2 the weld area became brittle. On test, the specimens exhibiting only $1\frac{1}{2}\%$ elongation over the 2.3-in. length, rather than the 10% elongation of the as-welded and recrystallized material. Yield strengths remained essentially the same.

For another series of tests, Nb specimens were exposed concurrently with those described above, but for longer periods of time. Results of the long-term tests lead to the conclusion that Nb rod, after any reasonable time in the reactor at temperatures up to 2400°F , will have mechanical properties equal to or better than the properties of the rod initially placed in service.

Figure 11 shows the changes that occur in room-temperature mechanical properties of unalloyed Nb after exposure under a He atmosphere in a 2400°F graphite tube furnace for the times indicated. The 0.2% offset yield strength increased greatly for short exposure times, probably due to O and C absorbed into the Nb. After a peak at about 54,000 psi, there was a gradual decline in strength to 44,000 psi, but the yield strength after 150 days at 2400°F remained 2.2 times that of the as-received material.

Fig. 11. Changes in mechanical properties of unalloyed niobium after high-temperature exposure

Proportional limit is the maximum stress at which strain remains directly proportional to stress; past this point the material takes on a permanent set. Percent elongation is the total increase in length of the 2.3-in. test section of the tensile test specimen, measured before pulling and after breaking. The sudden drop in ductility for short exposure times is due again to gas and C absorption.

The energy absorption curve represents the total energy absorbed by the 2.3-in. long, 0.190-in. diam test sections at failure. A slow change in this value indicates that the shock absorbing qualities of the rod are almost as good after exposure for 150 days at 2400°F as when the rod was installed.

Two pieces of Nb rod from Wah Chang were analyzed for impurity content. Both specimens were recrystallized at 1250°C for 1 h, but one piece was also exposed for 125 days in the furnace with the tensile test specimens. Impurity levels are listed on the following page, with the concentrations given in parts per million:

<u>Impurity</u>	<u>Wah Chang Ingot Analysis</u>	<u>Analysis of Fabricated Bar</u>	<u>Analysis After 125 days</u>
O	< 50	110	980
N	36	125	200
H	3.8	15	60
C	35	< 20	640

The changes indicate that gases were absorbed during forming of the 1/2-in. diam bar from the ingot material, or during the local recrystallization of the stock. A further absorption of oxygen and carbon occurred during the 125 days in the test furnace. According to published data, up to 0.315 w/o O in Nb acts as a solid-solution strengthener; but from this concentration to 0.565 w/o, the ductility decreases rapidly. Carbon also acts as a strengthener and hardener, but the concentration of NbC formed at the grain boundaries appears to rise rapidly at first, then to increase slowly with time at temperature.

Control Rod Drive Mechanisms

The basic design of the mechanism for the control rod drive has been completed except for the position transmitter and the method for decelerating the free-fall rod drop. One basic mechanism has been devised for both the plug rods and the articulated core rods. The drive consists of an a.c. hysteresis synchronous motor, with d.c. brake, driving a rack-and-pinion through a 940:1 ratio planetary gear train at a single drive speed of 18 in./min. Scram capability is provided by a stationary magnetic field clutch that disengages the gear train from the pinion when power is interrupted. The gear rack is machined on a ball spline that is used for the vertical guide.

HELIUM BLOWER TESTS

Prototype Primary Loop Blower

The 60-hp, gas bearing blower has been successfully operated for a total of 5000 h at design flow and head conditions. During a sequence of tests to obtain data points, the blower surged while operating at 10,000 rpm near the surge region. Steady-state operation had been achieved about 2 min before the event occurred, and there was no apparent perturbation of flow conditions to cause surge. The surge condition lasted about 5 sec before the blower was decoupled. The blower was successfully restarted after the main loop valve was opened to move the blower operating characteristics away from the surge region by reducing the loop pressure drop.

To check blower performance at different temperatures, tests were run at gas temperatures of 450^o, 300^o, and 200^oF and a pressure of 500 psig.

A series of tests was conducted to determine the temperature of the primary loop blower motor windings at various levels of motor horsepower and cooling water flow through the heat exchanger. The blower was operated at 600^oF inlet He temperature and 500 psi pressure. To vary the motor horsepower the flow control valve was set for a range of mass flow rates between 8800 to 14,400 lb/h. Results of the test show that the relationship between temperature and horsepower is linear over the range of test conditions. It can be concluded that the design of the cooling system for this blower is extremely conservative, and that the useful life expectancy of the windings is far in excess of 20,000 h.

Rateau Blower

A second set of performance tests on the Rateau 6-hp centrifugal blower have been completed. In measuring flow, speed, and power, better

data were collected from more accurate instrumentation than was used in previous tests. The new data show that the overall efficiency of the blower is higher than previously reported.

Recuperator

A solution has been found to the problem of joining the interior graphite structure of the recuperator to the nozzles on the external steel pressure vessel. The final design makes use of a stainless steel bellows, welded at the outer end to a liner inside the nozzle and fastened at the inner end to a flange that is enclosed within a graphite shoe. The inner flange can slip sideways inside the shoe, which is joined with threads to the internal assembly, while the bellows compensates for movement in the axial direction.

Fabrication of the steel vessel will be delayed after a decision was made to reject 1-in. steel plate material intended for the vessel shell. Ultrasonic inspection had revealed numerous inclusions throughout the material. None of these defects was singly injurious, but the large number of defects was intolerable. New plate material has been ordered, and delivery is expected at the end of March. Since the vessel will be delayed, Union Carbide Corp. may delay the machining of the carbon and graphite components.

Heat Exchanger

Baldwin-Lima-Hamilton has completed the fabrication of the primary-to-secondary loop heat exchanger and delivery will be made in March.

An extensive inspection program was carried out at the B-L-H plant by IASL staff members during the assembly of the heat exchanger. Particularly successful was the use of a small-diameter Resistron x-ray tube for the radiography of the internal welds that join the bayonet tubes to the tube sheets. The Resistron tube was placed inside the

1-in. tubes and a clear radiograph was obtained of each of the 96 tube-to-sheet welds.

Prior to assembly of the bayonet tubes, each end cap was He leak tested with a mass spectrometer detector. Then, after the tube-to-sheet welds were made, the individual tubes were leak tested before the sheet assembly was joined to the shell. The complete exchanger was leak checked with the tubes at 500 psig He pressure and the shell at 20 μ . No rise above background was found at a detector sensitivity of 3.7×10^{-11} std cm^3/sec . Welds in the shell were tested externally with the IASL encapsulator technique.

Secondary Loop Heat Dump

The secondary loop heat dump is scheduled for completion by B-L-H by March 1, 1965. Manifold to tube welds are now being made. When they are completed, the unit will be ready for acceptance testing. The rigorous inspection program set up for the heat exchanger has been extended to applicable portions of the heat dump.

Pressure Transducers

Cycling tests at 200°F with the Pace 0-50 psid transducer were halted in November 1964 for final calibration checks. This unit has been cycled approximately 200,000 times during 4280 h at this temperature for a grand total of 5280 h and about 250,000 cycles since the tests were started. To date, no seal failures have been observed nor has the unit exhibited any significant shift in its characteristics. The "as-received" calibration test had shown the transducer to be a "2%" unit, as was claimed by Pace, and the results of this final calibration are still within this value.

The demodulation circuit which was purchased with the transducer unit and used in the tests has begun to change its characteristics such that saturation now occurs at maximum output (5 V). As a result, there

is an apparent gross change in linearity for the transducer-circuit combination when used in this manner; however, it disappeared when the system was operated at reduced though adequate signal output levels.

GRAPHITE STUDIES

Graphite Oxidation

A small furnace, capable of producing temperatures to 2600°F, was constructed for experiments that supplement the test loop oxidation tests. Samples of graphite, similar in type to the UHTREX core graphite and machined to shapes that have various mass/surface-area ratios, are heated to 2600°F in He and then quickly exposed to ambient air. The temperature history of each sample is recorded until it cools to 1000°F or is consumed. The results of the experiment will indicate what might happen if the secondary containment of UHTREX were to be littered with small pieces of hot graphite after a rupture of the reactor pressure vessel.

In the first experiment, six samples of ATL graphite with weight-to-surface area ratios from 2 to 5 g/in.² were heated in He to 2600°F. The samples were moved quickly from the furnace into ambient air, cooled to 1000°F in 1.6 to 3.5 min; the weight losses were from 1 to 2%.

Corrosion Test Loop

Reaction rates at five temperatures have been measured in the graphite corrosion test loop with the technique that involves the measurement of inlet and outlet CO₂ concentrations. A nominal average temperature for each profile across the channel was calculated to characterize the reaction rate data. A plot of the rates as a function of concentration for each of the five characteristic temperatures

is shown in Fig. 12. The data obtained in the experiments are summarized in Table 9.

The individual specific reaction rates from which the averages were calculated varied no more than 25% from the mean, although the CO₂ concentrations ranged from 1 to 30 ppm. The fact that the specific rates determined were independent of concentration suggests rate control by a gas film diffusion mechanism. It follows therefore that the actual reaction rate (in units of mg/h-cm²) is dependent upon CO₂ concentration and is of the first order. The current data are excellent verification of results obtained by gravimetric measurements of graphite specimens and reported in LAMS-3063.

Fig. 12. Effect of CO₂ concentration on corrosion rate of graphite

Table 9. Graphite Corrosion Test Loop Results

Temperature °F	Average Specific Reaction Rate (mg/h-cm ² -ppm)	Number of Data Points	CO ₂ Concentration (ppm)
1610	7.32×10^{-4}	5	1 to 30
1770	2.11×10^{-3}	12	1 to 30
1892	3.09×10^{-3}	17	1 to 30
2117	4.40×10^{-3}	19	1 to 30
2334	5.73×10^{-3}	38	1 to 30

An Arrhenius plot (log specific reaction rate vs $1/T$) of the current data shows a curved line very similar to that calculated from Fick's law of diffusion. The calculated curve is higher than the experimental one, but the diffusion coefficient is not well known at this temperature level and the estimated value is probably too large.

The test loop experiment now underway is producing interesting data and will be continued to the upper temperature limit of the loop, i.e., metal surface temperatures up to 600°F . Reaction rate determination for CO_2 concentrations in the range of 30 to 100 ppm will then continue as the loop is cooled from day to day. When the characteristic temperature drops to 1600°F the effects of a concentration range up to 500 ppm will be examined.

IV. MHD APPLICATIONS

Introduction

An ad hoc committee composed of representatives of K, N, T, and J Divisions at LASL has been formed for the following purposes: (a) to determine whether or not a nuclear reactor-MHD closed Brayton cycle is feasible as an electrical power supply for space or ground-based applications, and (b) to decide if an experiment to demonstrate such a cycle is worthy of consideration as a LASL project. The design and performance of a relevant MHD generator are being established by consultation with the AVCO-Everett Research Laboratory. The initial responsibilities at LASL are the loop thermodynamics of proposed cycles and the availability of high-powered turbomachinery.

Possible Experimental Cycle

Preliminary calculations yielded the following conditions for a possible experimental cycle:

Reactor power (thermal)	9.1 MW
MHD power (electrical)	4.0 MW
Turbocompressor shaft power	5200 hp
Reactor outlet temperature	4000°F
Turbine inlet temperature	1800°F
Compressor inlet temperature	100°F
Compressor inlet pressure	15 psi
MHD duct inlet pressure	93 psi

A flow diagram of the cycle under study is shown in Fig. 13. Equations have been written for an analog simulation of this system. When gas molecular weight, gas flow rate, and heat exchanger geometries are used as design parameters, 24 equations with 31 unknowns are obtained; thus, seven of the variables must be chosen as parameters to

obtain a solution. The variables, shown in Fig. 13, are T_4 , T_9 , T_6 , T_{15} , T_{13} , T_{12} , and P_9 . For the present, the MHD generator performance is assumed to be equivalent to an adiabatic expansion (similar to a turbine) until parameters for the generator performance can be obtained from AVCO.

A second set of calculations was made with a digital code to determine possible core configurations for an approximately 2000 MW-reactor as the energy source in a ground-based, 1000 MW(e) MHD power plant. These calculations provide number of channels, hydraulic diameter, and

Fig. 13. Flow diagram of possible reactor-MHD system

length for an axial flow core composed of hexagonal fuel elements. Following are typical results:

Pressure Drop	10 psi
Core Diameter	30 ft
Active Core Length	12 ft
Maximum Fuel Temperature	4250°R
Outlet Gas Temperature	4050°R
Inlet Gas Temperature	2700°R
Helium Flow Rate	1040 lb/sec

The fuel elements have seven 1-in. diameter coolant passage holes, and a 0.125-in. web thickness between holes.

Analog simulation studies of MHD cycles are being continued. Computer programs are being debugged with results near at hand. A MHD topping-steam cycle will be compared with the regenerative turbocompressor cycle presently under study.

Cesium-Seeding Investigations

In the present concept, an MHD generator is to be operated in a closed loop with a graphite reactor and a graphite recuperator. The working fluid for the generator is He seeded with 0.5-1.0 a/o Cs added just upstream from the generator and removed before the He reenters the reactor. Methods for removing the Cs seem to be limited, for practical reasons, to condensation in a 100°F section of the loop near the compressor inlet, followed by electrostatic precipitation of the uncondensed Cs, if necessary. The degree of Cs removal that must be attained depends upon the reactivity of low concentrations of Cs with the graphite in the entrance end of the return side of the recuperator.

At 100°F, the vapor pressure of Cs is 5×10^{-6} mm Hg; this determines the concentration that would remain in the He stream if all of the condensed Cs were removed in the 100°F zone. Whether or not the graphite, at 300°F, in the recuperator would form an interstitial compound with Cs at such a low concentration is not known. The formation of CsC_{32} has been reported at a graphite temperature of 400°C under 6×10^{-3} mm Hg of Cs pressure. However, the analogous graphite-Br compounds decompose at elevated temperatures, and there seems to be general agreement in the literature that the graphite-metal compounds decompose spontaneously with a decrease in the vapor pressure of the free metal.

To provide information on the compatibility of Cs with graphite, several experiments are in preparation. The first group, in a glass system, is designed to check the effect of low concentrations of Cs in He on graphite up to 2000°F over a period of hours. The second group will be conducted in a stainless steel system and a high temperature (3500°F) furnace. It is designed to study the effect of 1 a/o Cs in He on graphite at high temperatures.

For the first Cs-graphite compatibility test, cylinder He was passed over a desiccant at a rate of approximately 80 cm³/min. It was then bubbled through liquid Cs at 300°F to saturate the He with 260 ppm of Cs. The saturated He was then passed over six blocks of graphite (three of type H4LM and three of Graphitite-G) held at about 1800°F. The gas was subsequently passed through a water trap to remove the Cs before the He was released to the atmosphere. The preliminary results of this test were inconclusive. The H4LM samples lost weight, but Graphitite-G samples gained about the same amount. Dimensional changes ranged from 0 to 0.0012 in. of growth.

V. PLASMA THERMOCOUPLE RESEARCH

Oscillator Strengths for the Cesium Resonance Lines

In studies of the spectrum of the Cs atom, the most reliable experimental results have generally disagreed with the accepted theoretical study (Stone) on the relative oscillator strengths of members of the resonance doublets. To resolve this disagreement, a simple optical absorption method of measuring both relative and absolute oscillator strengths was developed and tested experimentally.

The resonance lines of Cs are doublet members of the principal series, $6S_{1/2}-nP_{3/2}$ and $6S_{1/2}-nP_{1/2}$. The oscillator strength ratio ($R = f_{3/2}/f_{1/2}$) of the members of each doublet was measured from $n = 7$ (second member of the series) through $n = 14$. The findings agree with previous experimental results for the lower members of the series and with Stone's predictions for the higher members of the series.

When computed with the standard Cs vapor pressure data of Taylor and Langmuir, absolute oscillator strengths were about a factor of two lower than the expected values. After a series of careful checks on the experimental arrangement and method of analysis, it was concluded that the discrepancy was due to failure to achieve equilibrium Taylor-Langmuir vapor concentration for the species Cs in the presence of approximately 1 atm Ar.

VI. HEAT PIPES

Satellite Heat Pipe

A heat pipe is being constructed for the Applied Physics Laboratory, Johns Hopkins University, which will transfer heat from an externally mounted radioisotope (Pu^{238}) to the electronic component section of a satellite. Its purpose is to extend component life by reducing the amplitude of temperature cycling when in orbit.

The heat pipe is stainless steel with H_2O as the working fluid. Heat from the radioisotope capsule is transferred to the pipe through an Al block. The pipe is pressed into a hole in the block and the capsule is mounted in a recess parallel to the pipe. Three stainless steel clamps hold the capsule in position and supply the pressure for good thermal contact. This arrangement permits simple removal of the radioisotope for shipping.

Several models have been built and tested using electric heaters to mock up the radioisotope heating and a satisfactory design has been developed.

Ta and Ta Alloys as Containers for Boiling Metals

An evaluation is being conducted with various Ta alloys for use as heat pipe containers at temperatures above $1500^{\circ}C$. Possible fluids for use at these temperatures are Ag, In, Pb, and Bi. These metals are enveloped in small containers made from Ta and Ta alloys and heated isothermally in a vacuum furnace.

Five containers constructed from Ta bar stock and containing Ag, In, Pb, Bi, and Pb-Bi were run for 1000 h at $1800^{\circ}C$. No corrosion was detectable by metallographic examination.

Tests on seven containers constructed with Ta-5% W alloy and containing Ag were terminated after 1000 h at $1900^{\circ}C$. All samples showed grain growth. Two of the containers failed at 580 h due to extreme grain growth and slight bulging. Although they lost all their Ag, later He leak checks did not reveal any leaks.