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

QUARTERLY STATUS REPORT ON LAMPRE PROGRAM
FOR PERIOD ENDING AUGUST 20, 1962

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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 of the LASL LAMPRE program. 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.

LAMPRE I

Operations

The core loading of LAMPRE I was increased to 135 capsules plus four test specimens (23.02 kg Pu total, less burnup). Approximately 25 MWD of operation have been accumulated with Core II at powers up to 1 MW. The design power of 1 MW was achieved on July 15 and held for about 13 h before excessive heating of the reactor cell ceiling dictated reduction of power. Modifications in the cell cooling have been made to permit extended operations at the design power. The reactivity loss history of this core is illustrated in Fig. 1 and a summary of the operating history is given in Table I.

Fission Product Leakage from Fuel Capsules

Activity in the cover gas and in Na samples gives continuing evidence of capsule leakage; however, none of the capsules removed from the core showed detectable signs of having leaked. The levels of the activities observed suggest that the leaks are probably from the upper portions of the capsules, i.e., above the fuel level.

Fig. 1. Reactivity history of LAMPRE I

TABLE I

Resume of LAMPRE I Core II Operations

Run No.	Dates		Power MW	Cumulative MWH
	Start	Stop		
42	4/23/62	4/24/62	0.05	0.8
43	4/24/62	4/25/62	0.1	2.6
44	4/25/62	4/26/62	0.2	6.8
45	4/26/62	4/27/62	0.4	14.5
46	4/30/62	5/1/62	0.4	21.9
47	5/1/62	5/2/62	0.5	29.7
48	5/2/62	5/3/62	0.6	40.0
49	5/3/62	5/3/62	0.7	45.1
50	5/4/62	5/5/62	0.6	55.9
51	5/7/62	5/11/62	0.6	112.7
52	5/29/62	6/13/62	0.6	322.9
53	6/26/62	7/3/62	0.7	437.3
54	7/5/62	7/12/62	0.8	546.2
55	7/13/62	7/16/62	0.9 } 1.0 }	608.3
56	8/13/62	8/13/62	10^{-3}	608.3
57	8/20/62	8/20/62	10^{-3}	608.3
	8/20/62	(Running)	1.0	

Between the two 1 kW points on Runs 56 and 57, ten fuel elements were replaced.

Capsule Fabrication

An order is being processed for 220 standard LAMPRE I capsules to be used as needed for replacement in the core. These capsules are being made from ingot K596, high-purity, electron beam melted Ta.

Fabrication of 150 half-length capsules has been started to provide material for studies of corrosion film effects. High-purity Ta and Ta - 0.75 w/o W alloy are being used as the container materials.

RESEARCH AND DEVELOPMENT

Core Calculational Program

Work has begun on a series of calculations which will compare results from the current transport theory neutron codes with data obtained from the fast reactor experiments LCX I, LCX II, and LAMPRE I.

In the study of a particular case, one-dimensional spherical calculations are initially performed utilizing the DTK code, which is the latest adaptation of the S_n technique to one-dimensional transport theory. Top, bottom, and side reflectors of the actual cylindrical reactor geometries are mocked-up in the spherical calculations in detail. Then a parameter study is made to ascertain to what extent specifications may be relaxed without sacrificing computing accuracy. Typical parameters for study include space interval widths, material homogenization, neutron group collapsing, definition of outer boundaries, temperatures of outlying regions, etc. After consolidating the data, a two-dimensional calculation of each case is performed using the TDC (Two-dimensional S_n) code. Results from the study indicate what allowances should be made in future engineering designs for calculational uncertainties.

In addition to the reference study outlined above, survey calculations have been started of simple geometries containing dilute fuels, such as the Pu-Co-Ce alloy. In the initial computations, the effectiveness of reflector-shim control versus fuel dilution will be studied. Other questions for study in the future will include the effects of container materials, plenums, fixed reflectors, etc., on the neutron economy and control problem in dilute systems.

Estimates have been made of some operating characteristics for several cores which might be tested in the Core Test Facility. Results are summarized in columns 3, 5, and 6 of Table II along with three other comparison cores. The first four cores assume the same capsule diameter, wall thickness, and spacing as used in LAMPRE I with adjustment of cooling capacity, fuel density, capsule length, and number of capsules to give the indicated operating conditions. The Common-Header Tubular core assumes an annular array of tubes of approximately LAMPRE I size but joined at their tops to a tube sheet above which a thin layer of fuel forms a common header; coolant would flow down inside the annulus, then radially outward across the tubes, and finally back up the outside. The fuel of the direct contact core is assumed to be pumped by an upflowing annular Na jet. This jet entrains the fuel which flows out through orifices near the bottom of the cylindrical core vessel.

The estimates are not precise and are intended only to serve as a guide to preliminary thinking. The first four reactors are scaled from LAMPRE, while the last two are based on preliminary six-group calculations using the TDC in S_4 approximation. The first two values of dk/dT are the LAMPRE measured values, while the others are reasonable variations on a value calculated for the Common-Header Tubular core. Better estimates will be made when the expansion coefficients of the dilute fuels (here taken as Pu-Co-Ce ternary alloys) are better known. The value used for volumetric expansion coefficient in the dk/dT

TABLE II
ESTIMATED CHARACTERISTICS OF SEVERAL CORE TYPES

	Fuel Capsule Cores				Common-Header Tubular (CTF)	Direct Contact (CTF)
	LAMPRE I	LAMPRE Added Cooling	15 MW Size (CTF)	Power Station Size		
Power (MW thermal)	1.0	2.3	15.0	76.0	15.0	15.0
Pu in Fuel (g Pu/cm ³)	15.6	15.6	6.3	3.1	6.8	3.0
Critical Mass (kg Pu)	23.0	23.0	61.0	151.0	70.0	84.0
Average Heat Flux (10 ⁶ BTU/hr-ft ²)	0.43	1.0	1.0	1.0	1.0	--
Specific Power (watts/g Pu)	43.0	100.0	240.0	500.0	210.0	180.0
Core Volume (liters)	3.0	3.0	19.0	99.0	18.4	20.0
Core Power Density (MW/liter)	0.33	0.77	0.77	0.77	0.82	0.75
Δ T Na (°C)	113.0	150.0	150.0	150.0	150.0	150.0
Δ T Fuel (°C)	180.0	420.0	420.0	420.0	400.0	250.0
Na Volume Flow (gal/min)	133.0	230.0	1500.0	7600.0	1500.0	1500.0
Fuel Volume Flow (gal/min)	--	--	--	--	--	460.0
Core* Na Velocity (ft/sec)	4.0	7.0	13.0	20.0	30.0	30.0
Breeding Ratio (if blanketed)	--	1.7	1.6	1.3	1.6	1.7
dk/dT, Prompt Fuel (10 ⁻⁶ /°C)	-34.0	-34.0	-8.0	-6.0	-8.0	-8.0
Core Height (cm)	15.0	15.0	29.0	51.0	20.7	24.2
Core Diameter (cm)	15.0	15.0	29.0	51.0	18.0	32.6
Annular Core I.D. (cm)	--	--	--	--	6.4	--
Fuel Tube O.D. (in.)	0.430	0.430	0.430	0.430	0.400	--
Tube Wall (in.)	0.030	0.030	0.030	0.030	0.020	--
Tube Spacing (in. on centers)	0.500	0.500	0.500	0.500	0.460	--
Velocity of Mixed Fuel-Na(ft/sec)	--	--	--	--	--	7.5
Velocity of Fuel in Core (ft/sec)	--	--	--	--	--	1.1

* Except: Maximum velocity of direct contact core, excluding jet.

calculation was $-25 \times 10^{-6}/^{\circ}\text{C}$. Breeding ratios are rough estimates based on one-dimensional S_4 calculations made on cores of similar compositions.

The designs reported here are not optimized. The engineering limitations assumed were rather conservative in regard to heat fluxes, temperature limits, etc. Even so, the performance potential indicated is quite promising. Giving more attention to optimization and relaxing the conservative engineering limitations as experience permits should lead to cores giving even more attractive performance.

FUEL AND CONTAINER DEVELOPMENT

LAMPRE Fuel Containers

The present LAMPRE core loading utilizes four different Ta alloys as container material for the 90 a/o Pu - 10 a/o high-purity Fe liquid fuel. These materials are:

1. Electron-beam melted pure Ta
2. Double arc-melted pure Ta
3. Ta - 0.1 w/o W
4. Ta - 0.75 w/o W - 0.2 w/o Y

Corrosion studies have recently shown that pure Ta containers will withstand a higher temperature corrosion condition than will the W-containing alloys.

The corrosion test conditions consisted of varying time-temperature histories at temperatures of 625, 675, 700, and 725°C for periods of up to 2100 h at each temperature. Although the results indicate that, at temperatures of 675°C, pure Ta is superior to the W-containing alloys in withstanding intergranular penetration, it is significant that no containers failed (approximately 36 tested) at 625°C.

Until further testing can be accomplished in an effort to understand the effect of W additions, it has been recommended that the hottest LAMPRE capsule-fuel interfacial temperature be maintained at 625°C. In addition, future LAMPRE capsules used for routine replacements will be fabricated from unalloyed Ta with only selected irradiation test capsules being made from Ta alloys.

Hardness tests on fusion welded Ta - 10 w/o W sheet gave values for the weld area considerably higher than in either the heat affected zone or parent metal. Average for the parent metal was DPH 204; heat affected zone, DPH 254 welded in air, DPH 210 welded in dry box; weld metal DPH 352 welded in air, DPH 236 welded in a dry box. The results indicated that contamination occurred in the dry box, probably from oxygen dissolved in the molten weld metal, to form solid solution hardening. Weld microstructures were about the same. There was very little porosity or inclusions present. Specimens heli-arc welded in air appeared to have more pronounced grain boundaries--possibly from segregation of contaminants.

Plutonium-Cobalt-Cerium Alloys

Studies of the Pu-Co-Ce fuel alloy contained in Ta have shown that a very definite reaction layer is formed on the container wall. The layer also forms when the Pu is omitted from the alloys, i.e., with Co-Ce binary alloys. Preliminary x-ray analysis has established the composition of the reaction phase as TaCo. Because of potential difficulties arising from this reaction zone in obtaining a satisfactory fuel element, effort is being directed at establishing the kinetics of formation, spalling tendencies, and ways of minimizing the reaction layer.

A program has been initiated using Co-Ce alloys of 6 w/o, 12 w/o, and 18 w/o Co. Preliminary results indicate a strong effect of Co concentration, e.g., the layer does not form in a 6 w/o Co-Ce alloy

at up to 200 h at 750°C. The layer does not appear uniform, but varies in thickness by a factor of two over the surface of the Ta specimen. There is strong evidence that the layer is under high compressive stress and spalls readily, and that there is some solubility of the phase in the fuel alloy. Long lamellae of this phase are observed in the solidified fuel which is well separated from the Ta walls of the container.

Since Pu-Co-Ce alloys are being considered for use in a direct-contact core, studies have been initiated to determine the compatibility between these alloys and Na. In one such test, Na and Pu-Co-Ce fuel (5 g Pu/cm³) were placed in a Ta capsule and rotated through 360° in 10 min cycles for 96 h at 600°C. Subsequent sectioning after allowing a settling time of 1 h showed the following:

1. Na had wet the entire Ta surface and a layer of Na was visible at all points between fuel and Ta.
2. The fuel had a considerable amount of Na entrained throughout.
3. The Na appeared clean without evidence of fuel entrapment.

Although this experiment was of a preliminary nature, it does point up some of the areas that must be given careful study with regard to Na directly contacting Pu-Co-Ce fuel. Analytical results for Na in fuel and for fuel components in Na are pending.

In regard to the problem of fuel-Na compatibility, an initial Pu-Co-Ce irradiation test has been inserted into LAMPRE to obtain information concerning the effect of a stagnant layer of Na in contact with the fuel. Three LAMPRE capsules were loaded with Pu-Co-Ce fuel (3 g Pu/cm³) and a 1-in. layer of Na; three other capsules contained fuel without Na. One of each of the two types was tested out-of-pile at 625°C for 500 hr and showed no leakage. The other four capsules were inserted in the outer row of LAMPRE and will remain in the reactor as long as convenient.

U-Base Alloys

Scoping tests of the U-Mn-Pu system have been continued. Crucibles of ZrC, NbC, and yttrium were prepared for corrosion testing.

It is generally accepted that the use of a molten U-Pu-X alloy system will require extremely high temperatures with reference to present Na technology, since the lowest melting points observed are above 700°C. Because of these high ternary melting points, consideration is being given to the employment of fuels of this type as solids with periodic melting, either in place or external to the reactor, for the release of fission gases. The fuel would then be frozen within the original container and be returned to service. This process, while not as "clean" as a continuously molten fuel, should permit a combination of the absence of corrosion of solid fuel, fuel temperatures of less than 700°C minimum, and a marked increase in fissions per unit volume. Experiments along these lines have not yet been initiated.

Uranium Oxide - Plutonium Oxide Paste Fuel

Previous efforts directed toward determining whether a settled UO_2 -Na paste can be moved, or pushed mechanically, have ended in failure. Initial work using $<10\text{-}\mu$ UO_2 particles was reported in LAMS-2720.

A further experiment using 105- to 150- μ UO_2 particles was attempted. The paste was 37 v/o UO_2 , 1.5 v/o U metal, balance Na. A 1/2-in. internal diameter cylinder fitted with a 13/32-in. diameter piston fabricated from Type 440 stainless steel was used for this experiment at approximately 500°C. A pressure of approximately 59,000 psi proved insufficient to cause UO_2 movement, although the piston rod fractured.

No further experiments on paste movement will be undertaken for the present. A report will be prepared describing the work performed to date.

PLUTONIUM ALLOY STUDIES

Plutonium-Carbon Alloys

The vapor pressure over PuC_2 in equilibrium with graphite was measured by the Knudsen effusion method in the temperature range $2000^\circ\text{--}2400^\circ\text{K}$. Assuming that the gaseous species over this system is monatomic Pu, as is indicated by experimental evidence, the temperature-pressure relationship is: $\log_{10} p(\text{atm}) = -17920/T + 2.779$. This gives $\Delta H_{298}^\circ = 95.1 \text{ kcal/mole}$ as the heat of vaporization.

Plutonium-Indium Alloys

The congruent melting point of the intermediate phase PuIn_3 was tentatively placed at $1225^\circ \pm 10^\circ\text{C}$, and the reaction temperature and composition of the θ (PuIn)- κ (Pu_3In) eutectic were found to be $1068^\circ \pm 10^\circ\text{C}$ and 59 a/o In. The solubility of In in liquid Pu at 803°C was determined as $2.7 \pm 0.2 \text{ a/o}$.

Plutonium-Rare Earth Alloys

A series of preliminary experiments with arc-melted specimens has indicated that there are no intermediate phases in the binary systems of Pu with Sm, Eu, Gd, and Tb. It was also found that Pu and Eu form immiscible liquids.

Plutonium-Silicon Alloys

Three filtration experiments were performed to investigate the extent of solubility of Si in liquid Pu. The results are given in the accompanying table.

<u>Temperature</u> ($^\circ\text{C}$)	<u>Silicon Solubility</u> (a/o)
614	3.75
719	4.9
832	8.1

The maximum solubility of Si in δ -Pu was estimated to be 1.2 ± 0.1 a/o at 495°C , the peritectoid temperature for the reaction $\epsilon\text{-Pu} + \text{Pu}_5\text{Si}_3 \rightleftharpoons \delta\text{-Pu}$. This estimate is based on experimental results obtained at 485°C , which indicated that 1.2 a/o Si is soluble in δ -Pu at that temperature, and an extrapolation of the δ solvus to the peritectoid temperature at 495°C . In this 10°C temperature interval the change in solubility appears to be less than 0.1 a/o, which is less than the ± 0.1 a/o uncertainty in the solubility value.

Plutonium-Cerium-Cobalt Alloys

The approximate densities at room temperature of alloys in the Pu-Ce-Co ternary system are shown in Fig. 2, based on data obtained from the measured densities of a large number of arc-melted alloy specimens. Although there were large gaps in the data, trials have shown that densities taken from the figure have a maximum uncertainty of ± 0.2 g/cm³.

FUEL REPROCESSING

Solvent Extraction

Solvent extraction run 14-1 was completed by precipitation of $\text{Pu}_2(\text{C}_2\text{O}_4)_3$. This solid was fired to convert it to PuO_2 and dissolved, and $\text{Pu}(\text{C}_2\text{O}_4)_2$ was then precipitated. The resulting overall decontamination factor was 514 for β and 767 for γ .

A glass column was set up and mock solutions were run through to study operating procedures. It was found that emulsions formed to a greater extent than was expected. For this reason, solvent extraction run 15-1 was made with an in-line bed of silica gel to absorb the aqueous phase dispersed in the organic phase before it contaminated the product. The silica gel picked up 5 r/h activity which is about

Fig. 2. Iso-density lines for the Pu-Ce-Co system
at room temperature

10% of the total. However, the decontamination factors remained low at 21 for β and 21 for γ . An oxalate treatment is planned for this solution also.

Electrorefining

The analytical results of the third electrorefining experiment in the Hot Cells are complete. This was the second run in which irradiated Pu was used as feed material. An 83.5-g Pu foil irradiated to 0.12% burnup was combined with 212 g of unirradiated Pu and 7.7 g of Fe and the mixture was melted and filtered through a Ta frit. The filtrate which was collected in a graphite crucible served as feed to the electrorefining cell. The filtration yield was 87%. The metal was electrorefined at ~ 3 A until back EMF measurement indicated the conclusion of the electrorefining. The overall recovery, i.e., (Pu in product ring)/(Pu in anode feed), in the electrorefining process was 85%.

The purification achieved in both the filtration and electrorefining steps is summarized in Table III.

TABLE III

REMOVAL OF FISSION PRODUCTS FROM PLUTONIUM
HOT CELL ELECTROREFINING RUN IP-3

<u>Fission Product</u>	<u>Filtration, C_o/C_f *</u>	<u>Electrorefining, C_f/C **</u>
Sr	1.6×10^3	1×10^6
Mo	1.2	4×10^2
Ru	0.8	3×10^2
Cs	52	3×10^3
Ba	1.5×10^3	5×10^2
Ce	1.5	7×10^3
Nd	8.8	5×10^3
Pr	7.2	5×10^3
Zr	1.5×10^2	1×10^3
Gross	4.3	5×10^2
Gross	2.7	5×10^2

* C_o = concentration of fission product in filtration feed

C_f = concentration of fission product in filtrate

** C_f = concentration of fission product in electrorefining feed

C = concentration of fission product in electrorefining product

CORE TEST FACILITY

Architect-Engineer

Title II drawings and specifications for the Core Test Facility reached LASL on August 6, 1962. It is expected that evaluation, review, revision, and reproduction will be completed by the first week in November.

A preliminary examination of the drawings and specifications has been disappointing. The electrical drawings in particular are not as well done as had been expected. In several instances specifications were either absent or inadequate.

Phase B

Safety Analysis Report

The Safety Analysis Report for the CTF (LA-2735) has been completed and transmittal to AEC, Washington, is expected to be made August 31. Based on previous experience, permission to ask for CTF construction bids may be received about December 1. Construction could then be started about February 1, 1963, at the earliest.

Sodium Systems

Layouts of the Na systems are being altered to reflect recent drawings showing nozzle locations of the intermediate heat exchangers and heat dump. An effort is being made to finish the layout prior to the completion of Title II review so that no penetration changes will be necessary after contract negotiations.

The two non-radioactive Na loops establish adequate natural thermal convection upon loss of site electrical power, but the two radioactive Na loops are marginal in this respect. The actual core configuration must be known before convection calculations can be made with confidence. The use of battery-powered, low-performance,

dc electromagnetic pumps in series with the main pump in each radioactive Na loop is being considered as a means of maintaining decay power removal flow in the event of electrical power outage or main pump failure.

An examination of the pressure rise in the radioactive Na space, following a vessel rupture and release of fuel and all radioactive Na to the pit, indicates that ventilation system No. 2 will limit the period of positive space pressure to about 7 min.

Heat Dumps

A purchase order has been issued to the Struthers Wells Corporation for providing the two sodium-to-air heat dumps. Equipment delivery is scheduled for July 1, 1963. Preliminary general arrangement drawings have already been received from Struthers Wells and a representative of the Corporation visited LASL on August 14 to discuss design details. The conference was quite satisfactory and no problems are anticipated in procurement of heat dump equipment.