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ORNL 768 Reactors Progress Report

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## THE AIRCRAFT NUCLEAR PROPULSION PROJECT

at the

#### Oak Ridge National Laboratory

A. M. Weinberg, Project Director C. B. Ellis, Project Coordinator

QUARTERLY PROGRESS REPORT for Period Ending May 31, 1950

Edited by M. J. Nielsen and C. B. Ellis

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ORNL 768 Reactors Progress Report

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#### SUMMARY

**Reactor Design.** Operational methods are being developed for treating proposed intermediate reactor designs by multi-group analysis.

A new survey has been made of the impurities to be allowed in a primary coolant stream emerging from a reactor shield. Among the impurities most difficult to shield would be Na, V, Mn, Rh, and In. Calculations on the Bremsstrahlung which would arise in piping containing radioactive Li indicate that a primary Li<sup>7</sup> stream could probably not be brought out of the reactor without some external shielding.

Some calculations have been made on radioactivity in the secondary coolant of a circulating fuel reactor system which might arise from the delayed neutron flux in the intermediate heat exchanger. It appears that this effect would be negligible.

A survey has been made of the pressure drops to be expected across representative reactor core geometries with various liquid metal coolants. The superiority of the light metals Li and Na over Pb and Bi from this standpoint is very great.

A number of preliminary designs for a 1000 kw ARE reactor have been sketched out in first approximation. Most of these emphasize liquid metal coolants with solid fuel elements; however, one proposal involves sodium hydroxide cooling, with the eventual aim of developing a homogeneous circulating fuel aircraft reactor. All of these designs involve a considerable number of materials unknowns. Research aimed at clarifying these materials problems is now underway.

**Critical Experiments.** A joint ORNL-NEPA group is being established to carry out critical experiments of interest to the ANP program. A new building to be occupied by this group is expected to reach completion in July, 1950. Most of the apparatus for beginning the critical experiments has already been built and tested by NEPA.

Shielding. Data are presented in Figs. 4, 5, 6 and 7 for the neutron and gamma attenuation of shield assemblies containing 27% and 18% Pb by volume in water. Measurements of 33% Pb in water have been started. Data on the Fe-H<sub>2</sub>O-Pb shield for the Naval reactor are presented in Figs. 8 and 9. As a result of a

recalibration and improvements in the instrumental setup, it is believed that the accuracy of the intensity measurements is now within 30%.

A theoretical method called the neutron accountability system has been developed for estimating fast neutron leakage through a sample from the purely thermal measurements in the lid tank. Such calculations, when added to the new lid tank data, indicate that a unit shield to surround a spherical four foot aircraft reactor, in the idealized case neglecting ducts and heat exchangers, would weigh in the neighborhood of 140,000 pounds.

Construction has begun on the building to house the new shielding reactor. A reactor assembly and measurement equipment are expected to be ready for installation when the building is completed in the fall of 1950. It is believed that the neutron and gamma intensity through shielding samples will be high enough with this new facility to permit making spectral intensity measurements. A neutron camera similar to a Los Alamos model is now being built, and several other instruments suitable for neutron or gamma ray spectroscopy are under design.

Theoretical calculations have been made on the spectrum to be expected of neutrons emerging from various thicknesses of  $H_2O$  and Fe-H<sub>2</sub>O shields.

A study has been made on the desirability of using W for a major part of the shielding. It does not appear that W is likely to be enough better than Pb to be worth the difficulty and expense of fabricating it in large quantities. However,  $UH_3$  still appears theoretically to offer worthwhile shield weight savings. The possibility of obtaining this material at high density and in a non-pyrophoric state is being investigated.

The NEPA fast neutron spectrometer has been tested on the electrostatic generator at Bartol and at MIT. Its resolution appears adequate for shielding measurements.

Theoretical analyses are being carried out on several air duct shielding problems of simple geometry.

Boral has now been produced in large sheets of 1/8 in. thickness. Methods of joining Boral sheets are being investigated.

Heat Transfer. A figure-of-eight loop for circulating liquid metals to measure heat transfer coefficients is now nearing completion. This circulating system will employ a hydraulic bearing pump which eliminates any seals.

**Experimental Engineering.** A new group is being established to carry out all varieties of large-scale experimental engineering, particularly in liquid metals problems preliminary to the construction of the ARE reactor.

**Metallurgy and Materials.** Four-hour static corrosion tests of numerous materials in liquid Li and Bi at 1800°F have now been completed. The detailed effects upon the sample surfaces are discussed in this report. Those materials which so far show the greatest promise in liquid Bi are Fe, Mo, W and Ta. For Li at 1800°F the same materials, together with Zr and a number of stainless steels, seem most promising.

Studies are underway on mass transfer effects in liquid metal systems whose containing walls involve several different materials.

Four-hour static corrosion tests at 1800°F have also been completed on a number of materials in liquid Bi containing dissolved uranium—a fluid which might be used in a circulating fuel reactor. Mo and W show the least attack so far.

Twelve convection loops (harps) are now being made of various materials for dynamic corrosion tests. A small forced circulation system for corrosion tests is also being planned.

Because of evidence that much of the observed corrosion in molten Li systems is caused by oxygen and nitrogen impurities, a special high temperature Li purifier is now being constructed.

Lithium Isotope Separation. A separation factor of 1.02 has been obtained by molecular distillation. A two-stage refluxing still is being built. A packed column refluxing still to test feasibility of higher temperature and higher pressure distillation has been constructed.

More than 400 organic-organic systems have been investigated in a search for a suitable liquid-liquid countercurrent arrangement for isotope separation. Of these, 12 show sufficient promise for further investigation. In addition, a number of aqueous-organic systems are being studied.

Pulse columns are being studied for possible use with a Li amalgam system.

Additional research on Li isotope separation is underway with ion-exchange resins.

**Radiation Damage.** Deuteron bombardment of Fe immersed in Li at 1800°F is being carried out by North American Aviation, Inc. at Berkeley to study effects

of radiation on corrosion rates. Similar cyclotron bombardment will be started shortly at Purdue to study the effect of radiation on creep rate. Silicon carbide or magnesium silicide will probably be used there, since these are unclassified materials analogous to the  $\text{Be}_2C$  which is of interest for the aircooled reactor.

The creep test equipment to go into the ORNL reactor is partially completed.

Samples of Ti, Mo, Ni and Fe for before-and-after mechanical property measurements are expected to go into the Hanford pile shortly.

An apparatus has been designed for exposing plastics to a  $10^5$  R/hr gamma source.

Hydrides of Ti, Zr, and Li have been exposed in the ORNL reactor and will be analyzed for radiation damage after cooling.

An experimental liquid metal loop at 1800°F is being constructed for insertion within the ORNL reactor. It will be used initially in exploring the random radioactivity arising in liquid metal coolants. Later it should also serve for studies on radiation damage.

Nuclear Measurements. The high voltage laboratory building to house the 2 Mev and 5 Mev Van de Graff machines has been designed.

A mechanical neutron velocity selector is now being designed which will incorporate a 12-inch steel rotor running at 10,000 rpm. An 80-channel recording system is planned.

## INTRODUCTION

The Aircraft Nuclear Propulsion Project (ANP) is a joint effort of three laboratories: The Oak Ridge National Laboratory, the NEPA Division of the Fairchild Engine and Airplane Corporation, and the Lewis Laboratory at Cleveland of the National Advisory Committee for Aeronautics (NACA). The work of these three groups is guided in broad outline by the ANP Policy Committee in Washington, whose members are:

Director of Reactor Development, Atomic Energy Commission, Chairman Director of Research and Development, United States Air Force Director of Research, National Advisory Committee for Aeronautics Deputy Chief, Bureau of Aeronautics, Navy Department

This Policy Committee acts upon recommendations submitted by the ANP Technical Committee, which is composed of the Technical Directors of the three laboratories. During the past quarter these committees have held several meetings, with the result that the division of effort among the laboratories for fiscal year 1951 has been fairly clearly delineated.\*

The Oak Ridge National Laboratory has general responsibility for reactor research. Within this general field the Laboratory is responsible for overall coordination throughout the Project of the work on:

> Shielding theory and tests, Nuclear measurements, Critical experiments, Radiation damage, and Fuel reprocessing.

In reactor materials research each laboratory will proceed somewhat independently, but with continuous coordination by a joint materials committee.

The Aircraft Reactor Experiment. In addition to these general efforts, the Oak Ridge National Laboratory has proposed the construction of a small reactor at Oak Ridge. This project is called the Aircraft Reactor Experiment (ARE). The purpose of this experimental reactor, to operate at a power level of 1000 kw, is to obtain data and experience necessary for the design of a full-scale aircraft reactor. It is intended that the ARE will incorporate all

<sup>•</sup> The most recent survey of the work of the NEPA Division is NEPA-1374-IPR-52, Quarterly Report for the Period January 1 to March 31, 1950.

of the features which appear most promising for a large aircraft reactor of the liquid-cooled type. A feasibility report will be submitted for approval during the next quarter. It is planned that ORNL will carry the full responsibility for design and construction of the ARE, but that assistance will be received from NEPA and NACA in many phases of the work.

The development of the ARE reactor is being approached by attempting to first visualize an approximate design for a full-scale aircraft reactor. This design will then be arranged to operate at 1000 kw with as little alteration as possible. Liquid metal cooling is favored at present for ARE, although alternate proposals are still receiving consideration. A very considerable fraction of the experimental ANP work of the Laboratory, as described in the present report, has already been pointed along lines to answer problems confronting the ARE Design Group.

**Personnel.** Oak Ridge National Laboratory personnel participating in the ANP program now total 125. Of these, 87 are in engineering and scientific categories. In addition there are 19 NEPA men working full or part-time at the Laboratory, together with five men from the United States Air Force, one from NACA, one on leave from the Georgia Institute of Technology, and one on leave from Reaction Motors, Inc. Figure 1 is an organization chart for the ANP Project at this laboratory.

There are now six outside contracting groups associated with the ANP effort of the Laboratory: Nuclear Development Associates, Inc., H. K. Ferguson Company, and North American Aviation, Inc., for general design and analysis; Purdue University for radiation damage work; and the University of California and Stanford University for fundamental heat transfer measurements. Summaries of the work of these contractors are included in this report.



FIGI PAGE 13

### **REACTOR DESIGN**

The ANP General Design Group and its contractors have been engaged in a number of miscellaneous studies in reactor physics pointing towards the solution of problems arising in the ARE design. In addition, a number of problems in reactor engineering have been analyzed and several possible designs for the ARE have been roughed out. A number of these efforts are described briefly below.

#### MULTI-GROUP CRITICALITY CALCULATIONS

M. C. Edlund, N. M. Smith, J. W. Webster\*

An operational method for the evaluation of the so-called constants employed in multi-group calculations has been formulated. It is pointed out that the constants are functions of the macroscopic characteristics of the reactorthat is, size, shape, etc.—and that they are also functions of position within the reactor. The numerical integration of the multi-group equations for specific reactor designs proposed for the ARE (with or without hydrogen present) is contemplated by the method of Masket and Goertzel (MonP-360).

## **REACTIVITY CALCULATION TECHNIQUES**

Nuclear Development Associates, Inc.

Nuclear Development Associates plans to undertake a study of the various methods for reactivity calculations applicable to reactors of aircraft type. This work will include a survey of the older literature, and it may culminate in the production of a Reactivity Handbook. Following the survey, numerous reactivity calculations of direct interest to the ANP design will be undertaken.

#### GAMMA ACTIVITY OF PRIMARY COOLANTS

The question of whether the nuclear aircraft can be run on a binary cycleone in which a working fluid heated by the chain reaction is brought outside

\* NEPA

and carried directly to the radiators to heat the turbojet air-depends to a large extent on whether this working fluid will become too radioactive. Too much gamma activity in the primary fluid would necessitate extensive shielding of all the pipe lines and radiators, which would probably add a prohibitive weight to the plane. There are three principal ways in which such activity might arise: (a) release of fission products into the coolant stream, either by diffusion through the fuel element cladding or by rupture of an element, (b) activity induced in the coolant (or in its impurities) by the neutron flux, or (c) activity induced in the container materials (or minute impurities in these materials) anywhere in the coolant system-which might be dissolved by the coolant, or ejected from the walls by neutron recoil, and so carried outside the shield.

It is generally assumed at the present time that in practice one or more of these effects will arise and that the nuclear aircraft will have to employ a ternary system, with an intermediate heat exchanger embedded in the shield to transfer the heat from the primary fluid to a secondary external fluid having no contact with the region of high neutron flux. However, this question is not yet completely settled. Some calculations are presented here which bear on two facets of the problem, the nuclear gamma activities to be expected from various contaminants in the coolant stream, and the Bremsstrahlung gamma rays which would arise in the external piping from the Li<sup>8</sup> beta particles, if lithium were used as the primary coolant.

Nuclear Gamma Emitters (N. M. Smith). The results of the calculations are displayed in Table 1. The elements giving rise to high  $\gamma$ -activity are: Na, V, Mn, Rh, In, most of the rare earths, W, Re, Ir and Au. To a lesser extent Al, Cl, Sc, Cs and Ta are bad. Normal contamination amounts of Mg and Fe may be allowed.

In this work the absolute density of each likely constituent in the reactor coolant was computed which gives rise to an exposure of one Roentgen per day at the crew's position 100 feet away, when the lines and radiators are unshielded. The following assumptions were made:

1.	Distance, emitter to crew	100 ft
2.	Average flux level in reactor	$10^{14}$ neut/cm <sup>2</sup> sec
3.	Dosage at crew's position	1 Roentgen/day
4.	Self-shielding factor of emitter	3

## TABLE 1

## Gamma-Emitting Contaminants-Binary System

(Permissible  $\gamma$ -ray emitting contaminants in primary reactor coolant assuming  $\phi$ =10<sup>14</sup> neutrons/cm<sup>2</sup> sec, S = 3, D  $\approx$  100 ft, V<sub>0</sub>  $\approx$  14  $\times$  10<sup>6</sup> cm<sup>3</sup>, V<sub>i</sub>  $\approx$ 1  $\times$  10<sup>6</sup> cm<sup>3</sup>.)

					MAXIMUM PERMISSIBLE VALUES FOR ONE $\gamma$ /DAY AT COCKPIT				
ELEMENT	RELATIVE ABUNDANCE PERCENT	σ (barns) a	Ε (γ) (Mev)	T%	ATOMIC CONCENTRATION Nos/cm <sup>3</sup>	DENS ITY (gm/cm <sup>3</sup> )	TOTAL MASS (gm)		
*N <sup>15</sup>	0.38	$2 \times 10^{-5}$	6 (75%)	7.35 s	$2.5 \times 10^{23}$	6.2	8.7 × 10 <sup>8</sup>		
<sup>+</sup> Na <sup>23</sup>	100	0.5	2.75, 1.39	14.8 h	3.6 × 10 <sup>16</sup>	1.4 × 10-6	20		
Mg <sup>26</sup>	11.3	$4.8 \times 10^{-2}$	0.84, 1.01(20%)	10.2 m	$1.3 \times 10^{19}$	$5.6 \times 10^{-4}$	7900		
*A1 <sup>27</sup>	100	0.18	1.8	2.4 m	$2.3 \times 10^{15}$	$1.0 \times 10^{-5}$	140		
*Si <sup>30</sup>	3.05	0.12	Νο γ	2.8 h	R	-	<b>63</b>		
*C1 <sup>37</sup>	25	0.6	1.6 (36%) 2.1 (47%)	37.5 m	$2.6 \times 10^{17}$	$1.5 \times 10^{-5}$	212		
+K <b>4</b> 1	6.7	1.0	3.6 (75%) 2.1 (25%)	12.44 h	$3.5 \times 10^{17}$	$2.4 \times 10^{-5}$	340		
Sc <sup>45</sup>	100	22	1.12 (98%) 0.88	85 d	$1.7 \times 10^{15}$	$1.3 \times 10^{-7}$	1.8		
*Ca <sup>4 8</sup>	. 185	. 02	0.8	2.5h 30m	$2.0 \times 10^{21}$	. 15	$2.1 \times 10^{6}$		
V <sup>5 1</sup>	100	5	1.45	3.74 m	$1.0 \times 10^{16}$	$8.5 \times 10^{-7}$	11.9		
* Mn <sup>5 5</sup>	100	12.8	0.84	2.6 h	$7.0 \times 10^{15}$	$6.6 \times 10^{-7}$	9.2		
<sup>+</sup> Fe <sup>58</sup>	0.32	0.31	1.10 (50%) 1.30 (50%)	46 d	$5.8 \times 10^{18}$	$5.4 \times 10^{-4}$	76009		
Rh <sup>103</sup>	100	149	04, .18 0.95	42 s	$4.3 \times 10^{14}$	$7.3 \times 10^{-8}$	1.02		
In <sup>115</sup>	95.8	56	.4, 1.12 1.31, 2.32	53.9 m	$2.6 \times 10^{14}$	$4.9 \times 10^{-8}$	0.69		

			1	MAXIMUM PERMISSIBLE VALUES FOR ONE Y/DAY AT COCKPIT				
E LE ME NT	RELATIVE ABUNDANCE PERCENT	$\sigma_{a}$ (barns)	Ε (γ) (Mev)	T%	ATOMIC CONCENTRATION No./cm <sup>3</sup>	DENSITY (gm/cm <sup>3</sup> )	TOTAL MASS (gm)	
Cs <sup>133</sup>	-100	25.6	.60, .79 (95%)	2.3 y	2.1 × 10 <sup>15</sup>	4.6 × $10^{-7}$	6.4	
La <sup>139</sup>	99.9	9	1.63 (75%)	40 h	$5.2 \times 10^{15}$	$1.2 \times 10^{-6}$	17.0	
Pr <sup>141</sup>	100	11	$\Sigma E = 2.65$	19.3 h	$3.3 \times 10^{15}$	$7.7 \times 10^{-7}$	10.8	
5 Sm <sup>1 5 2</sup>	26.6	280	1.03 .07	47 h	$1 \times 10^{15}$	$2.5 \times 10^{-7}$	3.5	
Eu <sup>151</sup>	47.7	1530	0.12, 0.16 0.72	9.2 h	$1 \times 10^{14}$	$2.5 \times 10^{-8}$	0.35	
Dy <sup>164</sup>	27.3	2700	0.37 0.83	2.4. h	$8.5 \times 10^{13}$	$2.3 \times 10^{-8}$	0.32	
Ho <sup>165</sup>	100	67	1.2	27.2 h	$9.3 \times 10^{14}$	$2.5 \times 10^{-7}$	3.5	
Yb <sup>174</sup>	97.5	30	0.35	4.1 d	$7.5 \times 10^{15}$	$2.2 \times 10^{-6}$	31	
Lu <sup>176</sup>	2.5	3200	·112, ·206 ·317	6.6 d	$1.5 \times 10^{15}$	$4.4 \times 10^{-7}$	6.2	
Ta <sup>181</sup>	100	20.6	1.13 (37%) 1.22 (55%)	113 d	$3.6 \times 10^{15}$	1.1 × 10 <sup>-6</sup>	15	
W186	28.4	37.2	∑E = 3.07	24.1 h	$2.3 \times 10^{15}$	$7.1 \times 10^{-7}$	10	
Re <sup>187</sup>	62.9	75	∑E = 3.65	18 h	$4.4 \times 10^{14}$	$1.4 \times 10^{-8}$	2.0	
Ir <sup>191</sup>	38.5	260	$0.05 \Sigma = 2.20$	1.5m 70d	$3.4 \times 10^{14}$	$1.1 \times 10^{-7}$	1.5	
Au <sup>197</sup>	100	96	. 41	2.69 d	$1.9 \times 10^{15}$	$6.2 \times 10^{-7}$	8.7	

TABLE 1 (Cont'd)

+ Normal Impurities in Li

\* Normal Impurities in Fe

Feld, in Lex P 157, estimates total mass of  $3 \ge 10^4$  gm of Fe permitted in (Bi) coolant. Converting this assumption as to flux (2.4 x  $10^{14}$ ), volume (1.8 x  $10^5$ ), self-shielding (zero), and dosage ( $10^9$  2 Mev  $\gamma/r$ ) to those used here one gets satisfactory agreement. This is equivalent to 2.8 x  $10^{-4}$ % in Li. Compare this with typical analysis.

NOTE: The limitation on Li<sup>7</sup> (and other  $\beta$ -emitters) shall be treated separately. The above table is limited entirely to  $\gamma$ -ray emitters.

5.	Volume	of	coolant	external to shield	14	×	10 <sup>8</sup>	cm <sup>3</sup>
6.	Volume	of	coolant	inside shield	1 >	<. ]	.0 <sup>e</sup>	cm <sup>s</sup>

It may be noted that none of the pure elements listed in Table 1, with the possible exception of Ca, could be permitted to penetrate the core in massive amounts and then become exposed to the unprotected crew. Pure sodium, for instance, would, after penetrating the core, give an exposure at the crew's position of  $7 \times 10^5$  Roentgens per day in the case of the assumptions stated. A secondary heat-exchanger on the off-crew side of the shield proper, assuming a factor of ten for shadow shielding and another factor of ten for self-shielding, would still require a thickness of lead of 22 cm or an additional weight of 17 metric tons for an exchanger a cubic meter on a side.

Bremsstrahlung from Lithium Beta Rays (N. M. Smith, D. K. Holmes). The gamma-activity produced by the 12 Mev  $\beta$ -ray from the disintegration of Li<sup>8</sup> was estimated. The method employed assumes that the Li is intimately dispersed in an element of atomic number of 26-30 (the piping and radiators). The Bethe-Heitler theory for the emission of  $\gamma$ -rays was assumed correct. This theory essentially assumes that a  $\beta$ -particle makes not more than one Bremsstrahlung collision in slowing down. Furthermore, the extreme relativistic formula was assumed for the Bremsstrahlung cross section. The first assumption should result in an under estimation of the  $\gamma$ -activity; the second an over estimation. The resulting estimate may therefore be in error bymore than an order of magnitude.

There resulted a dosage of 30 R/day at the crew position as above, making an allowance of a factor of ten for self-shielding and shadow shielding. While this number should not be taken too seriously, it does indicate that  $\text{Li}^7$ , if used for a coolant, may have to be confined to a heat-exchanger and the latter covered by light shielding.

Further work on this problem is contemplated either making use of a more exact theory by M. E. Rose (ORNL 697) and the more exact Bremsstrahlung cross sections, or by means of a Monte Carlo calculation. The latter is expected to give approximate results with less calculation time.

# RADIOACTIVITY IN THE EXTERNAL COOLANT OF THE CIRCULATING FUEL REACTOR N. M. Smith

In the case of the circulating fuel reactor another effect might make the external coolant radioactive even though it came from an intermediate heat ex-

changer in the shield and never passed through the core. The delayed neutrons from the fission products in the inner circuit of the heat exchanger might react on the dissolved fuel in such a way as to build up a high neutron flux within the heat-exchanger. Thus even though the exchanger was far enough away from the chain reacting core so that the ambient neutron flux in the shield was not great enough to make the coolant in the secondary circuit (or its impurities) active, still the exchanger itself might be a strong enough neutron source to activate the external coolant. Some examples of this effect are calculated below.

The primary fluid in the case of a circulating fuel reactor is assumed to be a bismuth-lead-uranium alloy; the secondary (external) coolant being normal lithium. The total power is taken to be 300 MW.\* The heat-exchanger is assumed to be constructed of Fe, with one-third of its volume each of primary and secondary coolants.

The theory of the neutron multiplication in the heat-exchanger resulting from the presence of fuel and the emission of delayed neutrons has been published as a separate report.<sup>(1)</sup> It is shown that the slowing down density,  $q(r,\tau)$  in the bare intermediate reactor is given by:

$$q(\underline{r}, \tau) = \sum_{n} F(n) \overline{P}(B_{n}, \tau) Z_{n}(\underline{r}), \qquad (1)$$

where

$$F(n) = \frac{\varphi_n \, Q_n}{1 - \frac{1}{1 - k_{th \ eff}(B_n)} \int_0^{\tau th} \alpha(\tau') \overline{P}(B_n, \tau') \, d\tau'} \,. \tag{2}$$

11 0

r = position vector.

 $\tau$  = Fermi age.

 $P(B_n, \tau)$  = the three-dimensional Fourier-transform of the pointslowing down function from age 0 to  $\tau$  (including the resonanceescape probability).

 $B_n^2$ , the buckling, is an eigenvalue of the Helmholtz Equation:  $\nabla^2 Z_n(\underline{r}) + B_n^2 Z_n(\underline{r}) = 0$  obtained by the boundary condition of  $Z_n(\underline{r})$  finite everywhere and  $Z_n(\underline{r}_b) = 0$ , where  $\underline{r}_b$  is the vector position of the boundary.

(1) Smith, N. M., The Bare Intermediate Reactor: the Approach to Critical, ORNL 665 (Apr. 18, 1950).
\* Megawatts.

 $\psi_n = e^{-\tau_0 B_n^2}$ , where  $\tau_0$  is the age at which the delayed neutrons are born.

 $Q_n$  is the weight of the  $n^{th}$  mode of the source distribution, *i.e.*,  $Q(r) = \sum_n Q_n Z_n(\underline{r})$ , and since the  $Z_n(\underline{r})$  are orthonormal,  $Q_n = \int_{reactor} Z_n(\underline{r})Q(\underline{r}) d\underline{r}$ .

 $k_{\text{th eff}}(B_n^2) = \frac{k_{\text{th}}\overline{P}(B_n^2, \tau_{\text{th}})}{p_{\text{th}}(1 + L^2B_n^2)}$  thermal effective multiplication constant corresponding to the  $n^{\text{th}}$  mode.

L = the diffusion length.

 $k_{th} = \eta_{th} \frac{\sigma_{fth}}{\sigma_{ath}} p_{th}$  = thermal infinite multiplication constants.

 $\eta_{\rm th}$  = fission neutrons produced per thermal neutron absorbed

 $\sigma_{fth}$  = thermal fission cross section per cm<sup>3</sup>.

 $\sigma_{ath}$  = thermal absorption cross section per cm<sup>3</sup>.

$$\alpha(\tau) = \frac{3 \eta(\tau) \sigma_{f}(\tau)}{\lambda(\tau)} .$$

 $n(\tau)$   $d\tau$  = fission neutrons produced per neutron absorbed of age between  $\tau$  and  $\tau$   $d\tau$ .

 $\lambda( au)$  = transport mean-free-path as a function of age.

 $\sigma_{f}(\tau)$  = fission cross section per cm<sup>3</sup> as a function of age.

 $p_{th}$  = thermal resonance-escape-probability.

The denominator of (2) is the negative of the excess multiplication factor of the entire reactor. A very approximate evaluation of the excess multiplication factor of the fundamental mode for the heat-exchanger indicated to a first approximation that the neutron multiplication in the heat-exchanger just compensates for the leakage. The strong  $1/\nu$  absorption of the lithium has the property of cutting off the spectrum of neutrons before thermal energy is reached, making the heat-exchanger a sub-critical intermediate reactor.

The ratio, R, of the neutron absorption in one component of the materials making up the heat exchanger and its contents to the total absorption, was computed by approximate means, using the neutron spectrum of equation (1). There results that

$$R = \frac{\sigma_{ai}}{\Sigma \sigma_{ai}(0)} g(y), \qquad (3)$$

where  $\sigma_{ai}(0)$  is the fast macroscopic absorption cross section of the  $i^{th}$  component, and g(y) is a very slowly varying function of y, where

$$y = \frac{\sigma_{ai}(0)}{\sigma_{ai}(\tau_{th})} \frac{\Sigma \sigma_{ai}(\tau_{th})}{\Sigma \sigma_{ai}(0)}$$

g(y) is very close to unity. Thus a second simplifying result is obtained that the various components of the assumed heat-exchanger absorb neutrons in proportion to their fast absorption cross sections.

Table 2 has been constructed assuming a power level of 300 MW, a chainreacting core volume of  $.6 \times 10^6$  cm<sup>3</sup>, volume of circulating fuel in heat-exchanger =  $.3 \times 10^6$  cm<sup>3</sup>, volume of lithium in heat exchanger =  $.3 \times 10^6$  cm<sup>3</sup>, volume of lithium outside shield and exposed to crew's position 100 feet away is taken to be  $14 \times 10^6$  cm<sup>3</sup>.

The concentration of various contaminants producing one Roentgen per day at the crew's position is tabulated. Note that normal contamination amounts of all substances listed is permitted. At the same time, pure substances (such as sodium) would produce radiation in excess at one Roentgen per day.

One need not be much concerned with the  $(\beta, \gamma)$  emission from Li<sup>8</sup> in the case of the circulating reactor.

## EVOLUTION OF FISSION GASES FROM A FUEL ELEMENT D. K. Holmes, N. M. Smith

In order to examine the possibility that significant amounts of fission products may diffuse out of bare fuel elements at  $1000^{\circ}$ C, the general expression for the current of fission product ions from the surface of a slab inside which P fission product ions are produced per cc per sec has been obtained.<sup>(2)</sup> The accurate result may be approximated for small Dt by

$$j = \frac{2P}{\sqrt{\pi}} \sqrt{Dt}, \qquad (1)$$

where t is the elapsed time from the start of fissioning, and D is the diffusion coefficient  $(cm^2/sec)$  which must be obtained from experimental data. A series of reports of the Ames Chemical Group<sup>(3-10)</sup> give diffusion coefficients for many fission products from uranium slugs, all ranging down from  $10^{-10}$  cm<sup>2</sup>/sec. (2-21) A complete list of references is given on pages 29-30.

## TABLE 2

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## Gamma Emitting Contaminants --- Circulating Fuel System

I TEM	АТОМ	ABUNDANCE (percent)	REACTION	γ's EMITTED (Mev)	β's EMITTED (Mev)	HALF LIFE	FISSION ENERGY CROSS-~SECTION (millibarns)	THERMAL CROSS SECTION (barns)	ATOMIC CONCENTRATION (No./cm <sup>3</sup> )	DENSITY (grams/cm <sup>3</sup> )	TOTAL MASS (IF LESS THAN PURE SUB- STANCE) (grams)	REMARKS
1	N <sup>14</sup>	99.62	$N^{14}(n, p)C^{14}$	Νογ		5700 y	40					Fast reactions in nitro- gen are negligible
2	N <sup>14</sup>	99.62	$N^{14}(n, \alpha)B^{11}$	Stable		Stable						
3	Na <sup>23</sup>	100	$Na^{23}(n, \gamma)Na^{24}$	1.4, 2.76	1.39	14.8 h	0.29	0.6	$2.4 \times 10^{20}$	. 0092	$1.29 \times 10^5$	
4	Na <sup>23</sup>	100	Na <sup>23</sup> (n, p)Na <sup>23</sup>	Νο γ	4.1	4,0,7 s	0.7*	-2				Neglect
5	Na <sup>23</sup>	100	$Na^{23}(n, \alpha)F^{20}$	2.2	5	12 s	0.4	n.	$> 3.3 \times 10^{20}$	> .013	$> 1.8 \times 10^5$	
6	Na <sup>23</sup>	100	Na <sup>23</sup> (n, 2n)Na <sup>22</sup>	1.3	B <sup>+</sup> 0.6	2.6 y		-				May be neglected
7	Mg <sup>24</sup>	78.6	Mg <sup>24</sup> (n, p)Na <sup>24</sup>	1.4, 2.76	1.39	14.8 h	1.0	5	$> 5.1 \times 10^{19}$	> .0022	$> 3.0 \times 10^4$	
8	Mg <sup>25</sup>	10.1	$Mg^{25}(n, p)Na^{25}$	1.?	3.4	61 s	2.5	-	→ 1.2 x 10 <sup>21</sup>	> .046	$> 6.4 \times 10^5$	
9	Mg <sup>26</sup>	11.4	Mg <sup>26</sup> (n, γ)Mg <sup>27</sup>	0.84 (100%) 1.05 ( 20%)	1.8 (80%)	9.6 m	0.60	.048	$4.2 \times 10^{21}$	. 170	$2.4 \times 10^{6}$	
10	A1 <sup>27</sup>	100	$\mathrm{Al}^{27}(n, \gamma)\mathrm{Al}^{28}$	1.8	2.75	2.3* m	0.4	0.18	$4 \times 10^{20}$	.012	$1.7 \times 10^{5}$	
11	A1 <sup>27</sup>	100	$Al^{27}(n, p)Mg^{27}$	0.84 (100%) 1.05 (20%)	1.8 (80%)	9.6 m	2.8		$> 1 \times 10^{20}$	> .003	$> 4 \times 10^4$	
12	A1 <sup>27</sup>	100	Al <sup>27</sup> (n, a)Na <sup>24</sup>	1.4, 2.76	1.39	14.8 h	0.6	-	$> 1.2 \times 10^{20}$	> .0035	$> 4.9 \times 10^4$	
13	Si <sup>28</sup>	92.27	$Si^{28}(n, p)Al^{28}$	1.8	2.75	2.3 m	3.0	13	$> 5.8 \times 10^{14}$	$> 2.7 \times 10^{-4}$	$> 3.8 \times 10^3$	
14	Si <sup>29</sup>	4.68	Si <sup>29</sup> (n, p)Al <sup>29</sup>	?	2.5	6.7 m	2.7	9				May neglect
15	Si <sup>30</sup>	3.05	$\mathrm{Si}^{30}(n, \gamma)\mathrm{Si}^{31}$	Νο γ	1.8	2.83 h	1.1	0.12				May neglect
16	C1 <sup>35</sup>	75.4	$Cl^{35}(n, \alpha)P^{32}$	Νο γ	1.71	14.3 d	6.0					
17	C1 <sup>37</sup>	24.6	$\mathrm{Cl}^{37}(n, \gamma)\mathrm{Cl}^{38}$	1.60 (36%) 2.15 (47%)	1 19 (36%) 2.79 (11%) 4.94 (54%)	37.5 m	0.80	0.60	$9.3 \times 10^{20}$	.056	7.8 x 10 <sup>5</sup>	
18	K <sup>41</sup>	6.7	$K^{41}(n, \gamma)K^{42}$	1.51 (25%)	2.07 (25%) 3.50 (75%)	12.44 h	2.9	1.0	3.9 x 10 <sup>21</sup>	. 27	3.8 x 10 <sup>6</sup>	
19	Ca		No bad gammas combine	d with high cross secti	ons or high abundance					· · · · · · · · · · · · · · · · · · ·		
20	Mn <sup>55</sup>	100	$Mn^{55}(n, \gamma)Mn^{56}$	0.84 (100%) 1.81 (25%) 2.13 (15%)	0.75 (100%) 1.05 ( 25%) 2.88 ( 60%)	2.59 h	3.5	12.8	$5.9 \times 10^{19}$	.0054	$7.6 \times 10^4$	
21	Fe <sup>56</sup>	91.7	Fe <sup>56</sup> (n, p)Mn <sup>56</sup>	0.84 (100%) 1.81 (25%) 2.13 (15%)	0.75 (100%) 1.05 (25%) 2.88 (60%)	2.59 h	4.7		> 4.8 x 10 <sup>19</sup>	> .0045	$> 6.3 \times 10^4$	

(Heat exchanger separating circulating fuel from external coolant. Concentration leading to one R/day at crews quarters of  $\gamma$ -emitter of some common or possible contaminants.)

\* The application of this theory to fast threshold reaction is expected to give an upper limit to the radioactivity.

This value for D makes Eq. (1) valid for times as long as  $10^8$  seconds, and a simple estimate from Eq. (1) using  $P = 6.10^{15}/\text{sec}$  (corresponding to about 100,000 watts per cc) shows that even after  $10^8$  seconds only 1.5 cc of fission products considered as gases at STP will have crossed a square centimeter of surface. Thus, the fission products will be effectively trapped inside the fuel even at 1000°C unless (a) the fuel is in the form of a powder or a porous compact, or (b) if the uranium itself suffers sufficient radiation damage that it essentially breaks down into a powder. The Ames reports<sup>(3-10)</sup> which concern UO<sub>2</sub> are in agreement with (a), while recent GE experiments (unpublished results) indicate that for one percent burn-up (b) is indeed the case.

For the ANP, then, it must be concluded that essentially 100 percent of the fission products will escape from the fuel itself during operation.

The possibility remains that the fuel may be "canned." By enclosing the fuel element in a container of, say, aluminum, the fission fragments which come from the uranium on first flight are readily stopped in container walls of a few mils thickness.<sup>(18)</sup> Further, experimentally measured values for the diffusion coefficients of fission products in aluminum and stainless steel<sup>(15,16)</sup> are as low as those in uranium. It must be concluded that even at  $1000^{\circ}$ C, diffusion through undamaged container walls will not be appreciable,<sup>(17)</sup> so that the fission products which do escape from the fuel itself may be canned in, unless the effect of radiation damage, which may be toward increasing the effective diffusion coefficients,<sup>(19,20)</sup> becomes important at the proposed high flux. It is this latter possibility which stands in greatest need of experimental work, since available theory is entirely insufficient for even an estimate of effects of high fluxes on diffusion coefficients.

## ADVANCED SEMINAR IN REACTOR PHYSICS

N. M. Smith

An advanced seminar in reactor physics has been conducted for any interested members from the Oak Ridge National Laboratory or NEPA; the meetings are held once each week. The theory of the bare reactor has been covered in detail, including:

a. Slowing down and diffusion of neutrons in the finite medium;

b. The approach to critical of the thermal reactor: the critical conditions;

c. Elementary kinetics of the bare reactor,

1. One velocity group, no delayed neutrons,

2. One velocity group, one delayed group of neutrons,

3. Bare thermal reactor with general slowing down kernel and arbitrary number of delayed neutron groups and no external source;

d. Same as c (3) but with an arbitrary internal source present;

e. Kinetics of the bare reactor with uniformly increasing reactivity; \*

f. The approach to critical of the bare intermediate reactor: the critical conditions (ORNL 665);

g. The theory of the oscillating absorber in a chain-reactor;

h. Perturbation calculations of kinetic response of a reactor as influenced by various physical characteristics;

i. Kinetics of the Homogeneous Reactor; \*\*

j. First-order perturbation theory.

Copies of these seminar lectures are available on request.

#### COOLANT DYNAMICS

#### J. F. Haines

Studies have been made of the influence of the physical properties of potential coolants on the mechanical design of the nuclear aircraft power plant. Li, Na, Pb and Bi have been considered as the primary choices, but results can be interpolated for other materials by comparison of their physical properties with those of Li, Na, Pb and Bi.

Table 3 gives the properties of the four coolants having the major effect on the engineering design of heat exchangers, pumps and other coolant system elements. It will be noted that the boiling point is comfortably high for all but Na. Pressurization at approximately eight atmospheres might permit operation with Na at 1000°C; the vapor pressure of Na at 1000°C being about 2.5 atmospheres, and reaching only eight atmospheres at about 1200°C.

The product of specific heat and density is the heat capacity per unit volume. This value is of primary importance since it determines the effectiveness of the coolant in removing heat on a velocity basis. Table 3 indicates

Lecture by W. L. Carlson

\*\* Lecture by J. M. Stein

TABLE 3	5
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Properties	of Some	Potential	Reactor	Coolants	
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<u></u>	MELTING POINT (°F)	BOILING POINT (°F)	HEAT CAPACITY AT 1500°F (Btu/ft <sup>3</sup> -°F)	VALUES RE FOR A GIV	LATIVE TO LI AT EN GEOMETRY AND	SURFACE HEAT TRANSFER COEFF h* AT 1500°F	PRICE	
COOLANT				VELOCITY	PRESSURE	PUMP Hp	(Btu/hroft - F)	PER FT
Li	366.8	2928	29.8	1.00	1.00	1.00	30,900	\$ 400
Na	207.5	1616	14.8	2.01	8.52	17.1	32,000	<b>\$</b> 12
Pb	620.6	2935	22.6	1.32	42.7	56.3	7,300	\$ 84
Bi	519.8	2678	23.3	1.28	34.9	44.6	7,500	<b>\$122</b> 0

\* Values calculated for flow through .1 in. tubes at a velocity of 10 ft/sec.

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the relative velocities, dynamic pressures and pump horsepowers for the four coolants considered for a given geometry and power.

From a practical standpoint, the limiting factor of the coolant system and reactor structural design is probably reactor pressure drop, which will account for the major stresses. In order to maintain equivalent stresses in systems designed around the different coolants, it is apparent that coolant velocities must be much lower for the heavy coolants, resulting in a large increase in size of ducts, heat exchangers, etc. Design studies indicate further that in order to avoid excessive size of the reactor, with a corresponding major increase in shield and structural weight, the choice of core construction is much more limited for Pb and Bi than for Li and Na. The indicated superiority of Li as a coolant is in agreement with previous surveys.<sup>(22,23)</sup>

## LIQUID METAL COOLED ARE DESIGNS A. Fraas, R. Schroeder, J. Wyld\*

The ARE Design Group is attempting to achieve a 1000 kw reactor design which will simulate as far as possible an eventual large liquid metal cooled aircraft reactor. The first step in the process must of course be to develop at least a rough picture of the large reactor itself. During the past quarter a number of reactor arrangements have been sketched out which satisfy the following set of general engineering specifications. These specifications are believed to apply, in first approximation, to a reactor capable of powering an aircraft at Mach 1.5:

- 1. Coolant  $Li^7$ ,
- 2. Core pressure drop 60 pounds per square inch,
- 3. Core size and shape right circular cylinder 2-1/2 feet height and 2-1/2 feet diameter,
- 4. Maximum temperature of fuel element in contact with coolant 1800°F,
- 5. Power 500,000 kw,
- 6. Coolant temperature rise 400°F maximum,
- 7. Coolant outlet temperature 1700°F minimum,
- 8. Structural working stress 6,000 pounds per square inch,
- \* On leave from Reactor Motors, Inc.
- (22) Lyon, R. N.; Heat Transfer Experience on the Project, M-3974 (Aug. 5, 1949).
- (23) Kitzes, A. S., A Discussion of Liquid Metals as Pile Coolants, ORNL 360 (Aug. 10, 1949).

- 9. Acceleration load 12 g vertical,
- 10. Maximum attainable moderator volume and minimum structural poisoning,
- 11. Thermal stresses assumed to be self annealing.

All of the designs studied during the quarter contemplate BeO as moderator and  $UO_2$  as the fuel. Possibilities considered for fuel element designs include:

(a) 1/4 in. stainless steel coolant tubes coated on the outside with  $UO_2$  grains embedded in an iron matrix,

(b) Rectangular sandwich-type plates .025 in. thick, consisting of a UO<sub>2</sub>-stainless steel ceramel layer coated on both sides with stainless steel,

(c) Small cylindrical stainless steel capsules containing possibly a sintered powder of BeO plus  $UO_2$ , or perhaps  $UO_2$  plus Cr.

In each case molybdenum would be substituted for stainless steel if it turns out to have a low enough intermediate neutron cross section. Figure 2 illustrates some examples of the types of fuel-moderator arrangements being studied.

All of these designs are faced at present with a large number of experimental unknowns. In every case the feasibility of the reactor design will be decided by materials questions for which the relevant data are yet inadequate. During the next quarter experiments will be carried out in an attempt to settle as many of the materials unknowns as possible.

## NaOH--COOLED ARE DESIGN fl. K. Ferguson Co.

The Atomic Energy Division of H. K. Ferguson Co. is under contract with the Oak Ridge National Laboratory to make an exploratory study of an aircraft reactor operating with circulating liquid fuel. The contract covers a ninemonth period which started in March, 1950. The first two months of this period were devoted mainly to a critical review of the available information relating to the problem. For this purpose members of the Ferguson Atomic Energy Division have spent considerable time at Oak Ridge, and information







visits were made to Battelle Memorial Institute and to Argonne National Laboratory. In addition to informing themselves by reading reports, visiting the experimental facilities, and holding discussions with various people at X-10, Y-12, and NEPA, these personnel have participated in the ARE design conferences held twice a week.

As a first step in development of a liquid fuel reactor, Ferguson will submit an ARE proposal. This proposal is directed toward the ultimate use of an aircraft reactor in which NaOH serves both as the fuel solvent and the moderator for a homogeneous circulating-fuel reactor.

Calculations were made which indicated that NaOH was satisfactory from the standpoint of heat transfer. A preliminary survey has indicated that the possible fuel-solvent combinations for a circulating fuel for high-temperature use are limited in number, but this is at least partly due to lack of information. In particular, experimental data on the solubility of  $UO_3$ , or other uranium compounds, in NaOH are scanty and inconclusive. Such a system would have important and obvious advantages. Consequently, Ferguson has requested ORNL to make an experimental study of the pertinent properties of this system.

The Ferguson ARE proposal comprises three phases. In Phase I, a heterogeneous low power ARE reactor is used, comprised of fixed fuel elements (which may or may not contain the fuel in liquid form) and circulating NaOH moderator. The objective is to achieve as quickly as possible a reactor operating at high temperatures, to gain operating experience and to test certain container materials against molten NaOH under radiation. The heterogeneous low power reactor is scaled up to a full-size reactor not by increasing the heat transfer surface between fuel and moderator, but by eliminating it. This is done in successive steps (Phases II and III) in one of which the surface is removed making a homogeneous reactor, and in the other the fuel is circulated outside at a rate sufficient to get losses in delayed neutrons. Depending on the available uranium-bearing liquids, either of these steps may be taken first.

Design work on Phase I is proceeding in New York.

#### **GENERAL COOLANT CYCLE STUDIES**

North American Aviation, Inc.

North American Aviation, Inc. is under contract with the AEC to perform general analytical and materials studies pointed toward an eventual aircraft reactor. During the past quarter the work consisted of a survey of various possible power cycles for the aircraft reactor, in an effort to assess the relative merits of the flowing liquid metal system as compared to a number of vapor-cycle possibilities. A report on this work will be issued during the next quarter.

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## **CRITICAL EXPERIMENTS**

#### A. D. Callihan, K-25, J. F. Coneybear, NEPA

Organization of the newly formed joint ORNL-NEPA group to study aircraft reactor design by essentially zero power critical assemblies is just being effected. The construction of the Oak Ridge Area Critical Mass Laboratory, Building 9213, is about 60% completed. The building is scheduled for occupancy late in July. Provision has been made in the laboratory for two sets of experiments to be carried on simultaneously. A part of the personnel in the building will continue the joint K-25—Y-12 program of experiments designed to yield data of value to the production groups. The other personnel will participate in the ANP program. Early experiments in the latter will be directed to procurement of design data for the Aircraft Reactor Experiment. Requests have been made for the allocation of beryllium and uranium to the program. However, in the event of delayed delivery of the beryllium, it is expected that preliminary experiments will be done with graphite moderator to test instruments and controls.

The efforts of the NEPA experimental physics section over the past few years, directed towards criticality experiments, form a substantial background for the inauguration of this ANP program. Assembly equipment for critical experiments has been built and tested at NEPA and will be installed in the new laboratory. The following paragraphs descriptive of this NEPA work are reprinted from NEPA documents.

"The general arrangement of the assembly equipment is shown in Fig. 3. The operation of the assembly table, which is now set up at NEPA, has been tested with a 20,000 lb static load on the movable half which is driven by a speed-controlled electric motor. Limit switches are located at intervals to reduce the speed in increments as the movable part of the table approaches the contact position. No difficulty was experienced in the mechanical or electrical operation."

"Tests were started on prototype, safety, and control rods mounted on the assembly table. Deflection tests were performed on the three-inch square aluminum tube to be used in the honeycomb. This was done by loading a bank of tubes with lead bricks considered to be equal in weight to the maximum load for the honeycomb. The observed deflection in the bottom row of tubes was



0.005 inch which is considered satisfactory. There was no apparent buckling of the tube walls. An order has been placed for fabrication of a sample section of the aluminum honeycomb."

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"The overall system is electrically controlled. Control and safety rods have been worked out and tested in part. Control room equipment is being assembled in a mock-up of the control panel."
#### SHIELDING

#### BULK SHIELDING EXPERIMENTS

C. E. Clifford J. D. Flynn H. E. Hungerford H. W. Newson E. P. Blizard T. V. Blosser R. Lewis\* Beactor Technology Division

Results for Experiments 5 and 6, 27% and 18% Pb by volume in water, have been re-evaluated and the data are shown in Figs. 4, 5, 6, and 7. These are now believed to be accurate to within about 30% in intensity.

The data on the  $\text{Fe-H}_2^0$ -Pb shield for the Naval Reactor are also complete and here included as Figs. 8 and 9.

Experiment 8, measurement of 33% Pb, 67% H<sub>2</sub>O has been started. No data are yet available, but extensive foil measurements have been made for the first 10 inches of shield.

Much of the last period has been devoted to calibration of instruments with emphasis on the gamma ionization chambers. The latter have been standardized in free air by means of a radium source, and this measurement agrees with a calculated value within 20%. The absolute gamma fluxes as reported in NEPA-1374-IPR-52, Fig. 39, should be reduced by a factor of  $1.9 \pm 20\%$ . Uncertainties arise from the effect of the orientation of the cylindrical chamber with respect to the source. Further work will be required for a final calibration, but in the meantime data can be taken which will be internally consistent.

All neutron detectors are now fitted with new type waterproof cases and individual preamplifiers. The latter, it is hoped, will considerably improve the internal consistency of measurements.

The monitor, which records the incoming flux from the pile, has been replaced with a much larger  $B^{10}$  ionization chamber which is powerful enough to run a Brown recorder directly, thus eliminating the necessity of an intermediate amplifier and its corresponding uncertainties and servicing requirements.

The positioning apparatus for the long (25 in.)  $BF_3$  counters has been completely revamped so that uncertainties in position are now kept to about 1/16 in.

\* NEPA













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A new air-conditioned instrument room is being installed, and it is hoped that this will considerably improve the reliability of the instruments. Heretofore almost all delays in the program have been directly attributed to instrument failure.

As a service to the MTR, gamma measurements around the MTR Mock-up have been made by the group during the shutdown of operations incident to installation of the air-conditioned room.

The new fast neutron spectrometer, reported on elsewhere in this section, has been installed in the tank and a few spectra close to the source will be measured. The instrument is not sufficiently sensitive to measure at very great attenuations in the lid tank.

In an effort to assist Brookhaven National Laboratory in setting up their shielding program, the radioactivity induced in their tap water after one hour's exposure in the lid tank thermal flux of 10<sup>8</sup> was measured. The preliminary results were:

Distilled H <sub>2</sub> 0:	.010 microcuries/cc
Brookhaven water:	.016 microcuries/cc

#### INTERPRETATION OF LID TANK DATA

E. P. Blizard

#### Reactor Technology Division

Thus far only thermal neutron fluxes have been measured in the lid tank. Since intensity limitations make difficult the measurement of fast neutrons directly through the required attenuation, it is necessary to develop a reliable method of estimating fast neutron leakage from thermal flux. Since there is no resonance capture in water, this medium is ideal for correlation of fast and thermal neutrons, by what is referred to as neutron accountability.

The power of the source plate has been measured by means of thermocouples to be about six watts. From this value the fission rate in the source plate is known and with a guess about the reflection coefficient, one can estimate the total number of neutrons entering the tank. This number agrees well with the integral over the tank of thermal flux times thermal absorption cross section. A simple derivation yields a relation between location of removal of a fast neutron from the incident beam and its appearance as a thermal neutron. This displacement,  $\delta$ , is found at large distances from the source, to be:

$$\delta = \frac{\tau}{\lambda}$$
,

where  $\tau$  is the Fermi Age, and  $\lambda$  is the relaxation length of the fast current. Since  $\delta$  is a slowly varying function,  $\lambda$ 's for slow and fast neutrons will be about the same.

The relaxation length is crudely found to be just the reciprocal of the total fast cross section, and from this one can find, again crudely, a dominant energy of neutron for any relaxation length in water. The pure water data so analyzed show a reasonable approximation to the fission spectrum, as would be expected.

From the energy associated with the relaxation length one can find calculated values of the Fermi Age,  $\tau$ , and from this obtain the displacement  $\delta$ .  $\tau$  is chosen for the energy before collision, which gives an overestimate, but on the other hand the scattered neutrons are distributed in the forward direction, which probably compensates adequately.

Neutron accountability demands that for plane geometry,

$$\frac{dI(z)}{dz} = \frac{1}{\lambda} I(z) = \Sigma_a \phi_{th}(z') ,$$

where

$$z' = z + \delta ,$$

I(z) =fast neutron current at z,

 $\Sigma_{a}$  = thermal macroscopic absorption cross section,  $H_{2}0$ ,

 $\phi_{th}$  (z') = thermal neutron flux at z'.

It follows directly that

$$I(z) = \lambda \Sigma_a \phi_{th}(z').$$

Thus, for a given measured  $\phi_{th}$  and  $\lambda$  at z' one can estimate the fast neutron current passing the plane at z. Since back-scattering in water is not great, this then gives a measure of the fast neutron current which would be observed with a shield ending at z.

This method has been applied to some of the shield mock-ups that were tested in the lid tank and a weight estimate for an ideal unit shield (no ducts, no shield cooling, no structural members) has been made.

For purposes of calculation the following values were used:

	REACTOR	LID TANK SOURCE PLATE			
Power	$4 \times 10^8$ watts	6 watts			
Leakage of fast neutrons	20%	25%			
Size	2 ft radius (sphere)	14 in. radius disc = $4000 \text{ cm}^2$			

Reactor-crew separation = 100 ft

Tolerance: It is assumed that the military dose is eighty times that allowed at the Laboratory.

	LABORATORY (8 hr day)	MILITARY (25 hr mission)
Gamma dose rate	1/80 R/hr	1 R/hr
Fast neutron dose rate	$50 n/cm^2/sec$	$400 n/cm^2/sec$

Note that the dose can be taken either in gammas or neutrons, and if both types of radiation are incurred that the sum of fractional doses should not exceed unity.

The following symbols will be used:

r = inner radius of shield  $r_0 =$  outer radius of shield  $H(r_0 - r) =$  attenuation of spherical shell shield F(z) = attenuation of plane shield, thickness z h = factor to apply to lid tank centerline data to convert to plane geometry a = lid tank source plate radius = 14 in. S = source strength, neutrons/cm<sup>2</sup>/sec

We use the following geometrical factors:

$$H(r_{0} - r) = \frac{r}{r_{0}} F(r_{0} - r)$$

$$h = \frac{2\lambda^2}{a^2} \left( \frac{z}{\lambda} + 1 \right) + \frac{1}{2}$$
 (See ORNL 629, p. 10)

Ratio of surface strengths of lid tank source to reactor:

$$\frac{S_{LT}}{S_r} = \frac{\left(\frac{6 \times 0.25}{4000}\right)}{\left(\frac{4 \times 10^8 \times 0.20}{4 \pi (60)^2}\right)} = 2.1 \times 10^{-7}$$

In order to minimize the heavy gamma shielding required, it is decided to accept 75% dosage in gammas and 25% in fast neutrons. Slow neutrons are assumed negligible.

A distance z' in the lid tank is then chosen so that the thermal flux is:

$$\phi_{th}(z') = \frac{1}{h} \times \frac{r_0}{r} \times \left(\frac{100 \times 30}{r_0}\right)^2 \times 2.1 \times 10^{-7} \times \left(\frac{1}{\lambda \Sigma_a}\right) \times 1000 \text{ n/cm}^2/\text{sec}$$

where the factors in order are:

- 1) correction of lid tank centerline measurements to plane geometry
- 2) correction, plane geometry to spherical geometry
- 3) allowance for reactor-crew separation
- 4) allowance for lower specific source strength
- 5) conversion factor, thermal flux to fast current
- 6) 25% of fast neutron dose rate.

$$\phi_{th} = \frac{1630}{hR_0\lambda}$$
$$r_0 = r + z' - \delta$$

Choose  $\delta$  conservatively at 10 cms, and from Fig. 4 for 27% Pb and 73%  $H_20$ , it is seen that z' = 128 cms satisfies the above on the curve labelled 11 slabs. The shield thickness determined by neutrons is then 118 cms.

The next assumption is that the emergent gamma rays all result from neutron scattering or capture within the shield. It is then required that the gamma flux at z = (128 - 10) cms must be measured in the lid tank to be:

$$\Gamma_{LT} = \frac{1}{h} \times \frac{r_0}{r} \times \frac{(100 \times 30)^2}{r_0^2} \times 2.1 \times 10^{-7} \times .75 \ R/hr$$

$$= \frac{.018}{hr_0} \text{ Roentgens/hr} = 4.5 \times 10^{-5} \text{ R/hr}$$

These requirements are satisfied at z = 118 cms by the estimated value for about 10½ slabs (Fig. 5). 95 cms of matched shield, plus 23 cms of H<sub>2</sub>0 are adopted.

Note that the second lead slab does little or no good for gammas. It is neglected in the weight estimate.

Weight estimate:

$$W = \frac{4}{3}\pi (178^3 - 60^3) + \frac{4}{3}\pi \times .27 \times 10.3 (155^3 - 60^3)$$
 grams

= 64 long tons, or 70 short tons.

It is to be noted that in the measurements no advantage is taken of boron to reduce capture gammas. This would probably introduce a saving of 5 to 10 tons.

#### NEW BULK SHIELD TESTING FACILITY

W. M. Breazeale J. L. Meem\*

Reactor Technology Division

Formal approval for construction of the new bulk shield testing facility has been received, and the period since the last Quarterly Report has been used in completing mechanical design work. Since the construction is to be done by an outside contractor on a lump sum bid, it was necessary for the ORNL Engineering Department to prepare complete plans and specifications for the building and pool. The low bidder was John A. Johnson and Company, and work began on May 25. The contract calls for completion within 150 days. The reactor, supporting bridge, and the bridge for the measuring equipment have been designed and will be built at Oak Ridge or under subcontract. Schedules are such that this equipment will be ready for installation when the building and pool are completed.

Figure 10 gives the fundamental dimensions of the facility and indicates the possible reactor locations. Details of the reactor design, which essentially duplicates the MTR design, are shown in Fig. 11. The fuel elements (viewed end-on in section A-A of Fig. 11) consist of curved plates contained in aluminum boxes and running the length of the core. The curved plates are U-Al alloy clad with aluminum.

A three-dimensional sketch of the pool, reactor, and reactor bridge is shown in Fig. 12. A similar bridge (not shown) will support the measuring equipment.

It is still too early to predict accurately when the facility will go into operation, but allowing a reasonable time for installation of equipment, calibration of instruments, etc., and adding this to the contractor's time for construction of the pool and building, it appears that investigation of the initial sample will begin during the first quarter of the next year.

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With the relatively high intensity from the reactor, it is hoped that measurements can be made of neutron and gamma ray spectra through shielding samples in addition to the thermal neutron and total gamma ray ionization measurements, such as are being made in the lid tank. A neutron camera similar to that used at Los Alamos is under construction. Neutrons are collimated through a two foot pipe, one inch in diameter, and strike a paraffin radiator, ejecting protons. Experimentally, the Los Alamos group has found little change in the neutron spectrum above 1 Mev upon collimation through such a pipe. The protons are collected on a plate coated with a proton-sensitive photographic emulsion, the plate being placed at a given angle with respect to the neutrons incident on the radiator. The plates are examined under a microscope after development, and from the number of proton tracks and the length of the tracks, the intensity and energy of the neutrons may be calculated.

Several other instruments suitable for either neutron or gamma ray spectroscopy are being considered for use in the new facility. It is hoped that the feasibility of such devices will be determined by the time the first shielding sample goes in the pool.

#### SHIELDING ANALYSIS

W. K. Ergen\* F. Murray K. Keyes S. Podgor\*

Reactor Technology Division

Attempts are being made to summarize shielding theory, and to express every step in as simple a form as possible. This is a long term project, but some items are made available in the form of memos as soon as they are derived.

In CF 50-4-3, identical with NEPA STRM-57, the neutron spectrum emerging from a water shield was computed, assuming an incident fission neutron spectrum of the form  $e^{-0.72E}$ , and a total molecular water cross section which can be fitted by  $\sigma(E) = 11.6 \ E^{-1/2}$  barns, in accordance with the H cross-section data of B. E. Watt, LA-718, and 0 cross-section data of H. Feshback, ORNL 417.

The emerging neutron spectrum has a maximum, since neutrons below the maximum are attenuated strongly by the large cross sections, and neutrons with

\* NEPA

energies above the maximum are not very abundant in the incident spectrum. The maximum moves to higher energies with increasing shield thickness because the attenuation, which is less for higher energies, becomes more important than the abundance in the incident spectrum.

Similarly, the neutron spectrum emerging from an iron water shield was computed in CF 50-3-108 (NEPA STRM-55). It was found that the emerging spectrum for 2/3 iron by volume, as used in lid tank Experiment 7, has a peak at about 1 Mev because of the cut-off in inelastic scattering in iron at low energies.

Since the fabrication troubles with metallic wolfram sheets proved to be greater than anticipated, the advantages of W for shielding were reexamined in CF 50-3-103, NEPA STRM-53. The high density of W gives savings in shield weight which amount to 10%, as compared to lead, in representative cases. This is partially offset by the lower Z, which makes W a less efficient gamma shielding material than lead. Both the density and the Z effect are easily and accurately computable and would not justify separate W experiments. The unknown shielding properties of W are connected with the numerous and low lying levels of W, which give hope of cascading of capture and inelastic scattering gammas, and hope of a low inelastic scattering threshold. Information on some of these points is, however, appearing in the open literature. All-in-all, experiments with metallic W appeared not worth the difficulty in fabrication, and it was recommended that fabrication of metallic W sheets be abandoned for the time being. (As reported elsewhere, W experiments are expected to be carried out with a more easily obtainable material, such as W powder bonded by tygon plastic,)

Among hydrides,  $UH_3$  seems still worth investigation as to obtainable density, reduction of pyrophoric behavior, and filling of interstices with useful shielding material (CF 50-3-107, NEPA STRM-54).

Another interesting hydride is  $\text{LiBH}_4$ , which probably can be cast and hence obtained in theoretical density. Its main advantage would be the low weight of compound per gram atom of hydrogen, 5.4 gm as compared to 9 g of water.  $\text{LiBH}_4$  of theoretical density also contains somewhat more hydrogen atoms per cc than water, and Li and B are better neutron attenuators than oxygen.

Titanium hydride is not too good as shielding material but holds the hydrogen at high temperature. Some attempt should be made to obtain  $TiH_2$  at

theoretical density, or to fill the interstices.

These comments on hydrides were set forth in somewhat more detail in CF 50-5-24 (NEPA STRM-63).

Liaison was maintained with the Westinghouse-Argonne and the KAPL Naval Reactor Shielding groups and one of the conferences was summarized in CF 50-3-6 (NEPA STRM-52).

As a service to the MTR group, the expected relaxation length in Barytes concrete was computed (CF 50-4-54) and found to be about 8 cm.

The analysis of the penetration of fast neutrons through thick shields with the Boltzmann equation was further developed to include an approximate calculation of the contribution of the continuous spectrum, for the special reduced form of the Boltzmann equation in which the only scattering assumed is elastic scattering from heavy elements, and possibly others considered as pure scatterers.

#### NEUTRON ENERGY SPECTROMETER

B R Gossick\* K. Henry\* Reactor Technology Division

During the past quarter the fast neutron energy spectrometer was tested at Bartol Research Foundation and at Massachusetts Institute of Technology. The electrostatic generator at Bartol was used to obtain 4.4 Mev neutrons from the  $d(d,n)He^3$  reaction. Points at 1.9 Mev and 14 Mev using the  $Li^7(p,n)Be^7$ , and  $H^3(d,n)He^4$  reactions, respectively, were obtained on the Rockefeller generator at MIT. The resolution checked with design calculations, giving on the average a 23% half level width, which is considered sufficient for shielding measurements. Results of the tests at Bartol and MIT are given in the joint reports, ORNL 711 and NEPA-1407.

#### DUCT THEORY

D. W. Whitcombe\*

#### Mathematics Panel

The effect of shield ducts upon flux distribution is very important when a shield must be designed for minimum weight. Theoretical results have been

\* NEPA

obtained for some simplified arrangements.

A diffusion solution has been obtained for a cylindrical duct with walls perfectly absorbing thermal neutrons, the duct being filled with a pure scatterer. Laplace's equation is solved for a plane source of thermal neutrons. The solution is found in terms of the extrapolated boundaries  $r_0$  and  $z_0$ . These are then related to the actual boundaries  $r_1$  and  $z_1$ . A condition on the diffusion analysis is obtained which requires  $r_1$  to be greater than 1.6  $\lambda$ where  $\lambda$  is the mean-free-path. When  $r < 1.6 \lambda$  the first collision density should be used. A detailed report of this problem is being prepared.

Another problem, diffusion for a cylindrical air-duct for infinite geometry, has been solved by two methods. The first solution in an iteration solution; the second uses the steepest descent approximation. Both methods solve the same diffusion equation and boundary conditions. The results have been published as ORNL 668.

Some transport equations have been set up and the matrix elements are being computed so that the solution can be found on the NEPA computing machine. Some work has been done on the three-dimensional cylindrical duct and it is hoped to reduce this transport integral equation to linear equations so that it may be solved on the NEPA machine.

#### SHIELDING SURVEY

Nuclear Development Associates, Inc.

Although the NDA contract for shield analysis is supervised by the AEC Washington Office, The Oak Ridge National Laboratory maintains a strong interest in those phases of the work which are applicable to ANP reactors. The program involves study and application of analytical and numerical methods for predicting shield performance, study of results obtained from transmission and bulk-shielding experiments, and work on problems arising in moving toward actual shield designs, with liaison on engineering and materials problems. In general the work is along lines indicated in ORNL 437, Summary Report on Summer Shield Work, by Gale Young, with increased attention to practical design problems encountered in mobile reactors.

#### SHIELDING MATERIALS

A. S. Kitzes V. L. McKinney

#### Reactor Technology Division

Two additional attempts were made to roll Boral into large sheets at Lukens Steel Company. One sample was heated to 1050°F and the other to 1150°F. Both attempts were unsuccessful, probably due to rough handling since the mill used is designed for steel ingots and the rolls could not be slowed to the desired speed.

At X-10, twelve ingots 4 in.  $\times$  4 in.  $\times$  1 in. with  $B_4C$  content varying from 20% to 50% by volume were successfully rolled into 1/8 in. sheets. The ingots were heated after each reduction. Initial temperature was 1100°F and the rolls were lubricated with kerosene. Two ingots containing 50%  $B_4C$  were successfully rolled to 1/8 in. without heating between rolls by completing the rolling within 3-1/2 minutes.

Two ingots are now being cast for rolling into sheets 24 in.  $\times$  84 in.  $\times$  1/8 in. Methods of joining Boral sheets are also being investigated.

### HEAT TRANSFER

C. P. Coughlen H. C. Claiborne A. R. Frithsen\* R. N. Lyon

Reactor Technology Division

Experimental heat transfer equipment is now nearing completion. The system, Fig. 13, is a figure-of-eight apparatus with liquid passing from a pump through a preliminary heat exchanger, to the test exchanger, and from there to a heater. The heated liquid is sent back through the test exchanger, countercurrent to the cool liquid, then through the preliminary exchanger, and finally through a cooler, before being returned to a sump attached to the pump inlet. By-passes are built around both streams in the test exchanger, so that flow rate on one side can be varied without changing flow on the other side. Calrod tracing of all components permits preheating of the system to prevent premature freezing of the lithium charge. Piping work is essentially complete; Globar mounts have been constructed in the heater. The main heater control and control board have been constructed and installed. Clean out runs and pressure testing are scheduled for the near future.

Theoretical investigations of shape effects on local heat transfer coefficients have progressed to the point of locating velocity distribution data on rectangular and triangular tubes. Attempts will be made to interpret these data in terms of local heat transfer coefficients.

A level indicator for flow rate determination using a catch tank has been developed. A small amount of additional work is required, however, to make this indicator operate a timer.

To date, pump development for experimental systems has been restricted to the development of hydraulic bearings as a means of eliminating seals in pumping systems.

The hydraulic bearing presently undergoing tests is a modified type of journal bearing which derives its load carrying capacity from high pressure fluid which is introduced through small orifices. After passing through the orifices, the fluid pressure is distributed longitudinally along the pump shaft by means of eight slots; this produces a constant pressure around the shaft periphery as long as no transverse loads are experienced. When transient



FIG 13 FIGTHE "8" LILUID DECALC OVEREN

transverse loads are introduced, the shaft tends to move to one side of the hydraulic bearing, and the flow from that side is reduced which, in turn, increases the fluid pressure on this side but reduces the pressure on the opposite side. Due to the pressure differential then opposing the transverse load, the shaft tends to move back to its central position.

Figure 14 shows a complete pumping system, excluding pump impeller, which has been set up for the purpose of testing hydraulic bearings. The pump impeller is omitted in all tests in order to eliminate pump parameters from influencing data obtained for evaluating the bearings. It should be noted that an enclosed ("canned") rotor motor is used for the pump drive, which, when combined with hydraulic bearings, not only gives a pumping system void of seals and ball bearing, but also one that can be completely welded together thus becoming leakproof.

During the next quarter, approximately 10 different variations of this type of hydraulic bearing will be tested. In addition, one bearing of this type has been made entirely out of transparent plastic so as to observe, with the aid of dyes, the hydrodynamics involved in its operation.

Editing of the Liquid Metals Handbook, written jointly with representatives from other AEC and Navy Laboratories and contractors, is now essentially complete, thanks to invaluable assistance by the publications staff at the Naval Research Laboratory. The final proof will be submitted to the Government Printing Office by June 17.



# EXPERIMENTAL ENGINEERING

#### H. W. Savage

The objective of this group, newly formed, will be to provide data from engineering experiments to determine the validity of design conclusions and to test the components of the Aircraft Reactor Experiment. It will also apply the findings of other groups performing fundamental studies to full scale engineering tests in components or mock-ups of the reactor.

Among the components anticipated to be developed, tested and evaluated are the fuel, moderator, container and reflector designs of the ARE assembly; the primary and secondary cooling systems including the heat exchangers and the required instrumentation; the internal and external power control systems; the components of the shielding system; and equipment for handling components remotely.

This activity will be initially centralized in Building 9201-3, Y-12. The group will work collaboratively with the ANP groups engaged in fundamental studies, and will be closely coordinated with related activities carried on by NEPA.

# METALLURGY AND MATERIALS

# E. C. Miller

#### Metallurgy Division

The OPNL Metallurgy Division is engaged in a materials program whose objectives are: (1) the selection and study of materials suitable for the construction of a high temperature, liquid metal cooled, airborne reactor---and its prototype, the aircraft Reactor Experiment; and (2) the development of methods for fabricating these materials into reactor components.

This program includes:

- (1) Static corrosion sorting tests in which a wide variety of possible structural materials are being exposed to several potential coolants for different times and at different temperatures to select the more promising combinations of materials for further investigation.
- (2) Dynamic corrosion tests, using
  - (a) thermal convection loops to investigate the effects of thermal coefficients of solubility on corrosion in a circulating system subjected to a temperature differential,
  - (b) forced circulation corrosion loops intended as simulated service tests, and
  - (c) spinner tests to investigate erosion in small scale tests at constant temperature.
- (3) Investigation and control of observed static corrosion phenomena and the factors affecting corrosion. Some of these include surface finish, crystal structure, presence of precipitated phases, form and concentration of alloy components, stress-corrosion, addition agents and gettering agents, mass transfer effects.
- (4) Fabrication of reactor components and associated equipment. This includes composite fuel elements, moderator matrix, and heat transfer equipment.

#### STATIC SORTING TESTS

The tests now available for reporting consist of exposing the metals listed in Tables 4 and 5 to the action of liquid lithium, and to bismuth, in

# TABLE 4

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# Corrosion of Materials by Liquid Lithium in Static Tests - 4 hrs., at 1000°C

	· ·		WEIGHT CHANGE (mg/cm <sup>2</sup> ) RATIO OF SPECIMEN AREA TO COOLANT		RATIO OF SPECIMEN AREA TO COOLANT	
MATERIAL	SOURCE AND ANALYSIS	RUNS	RÅNGE	AVERAGE	VOLUME . em²/em3	REMARKS
Titanium	Remington Arms	3	+ 3.5 to + 4.4	÷ 3∘5	0 - 4	Discontinuous adherent film. Identified by X-ray as TiN.
Zirconium	Bureau of Mines	3	+ 0.3 to + 0.5	+ 0∘4	0 <sub>°</sub> 6	Thin continuous adherent film of ZrN.
Molybdenum	Fansteel	3	+ 0∘1 to + 0∘4	++ 0₀3	0 . 6	MoC and Mo <sub>2</sub> C detected
Zirconium	Bureau of Mines	2	- 0.1 to + 0.1	÷ 0∘1	0 . 4	$Z_{\mathcal{E}}N_{e}$ same as $Z_{\mathcal{E}}$ above
Niobium	Fansteel	3	- 1.9 to + 0.1	- 0. <u>1</u>	0 - 2	
Iron (Armeo)	Corry Steel Co.	3	· 0.2 io · 0.1	- 0∘1	0.6 (?)	Uniform attack
Tungsten	Fansteel	3	= 5∘6 ¢∾ = 0∘ <u>1</u>	- 0·2	0 - 6	Uniform attack - no film detected
Tanîalum	Fansteel	3	- 1.5 to - 1.0	÷ 1∍5	0∘б	Uniform attack with thin carbide film formed
1040 Steel	SAE Steel Co.	3	- 2.4 to ≈ 1.9	- 2∘2	0.6 (?)	Selective type corrosion, probable carbide solution
Cobalt	Kulite Tungsten Co.	2	- 5.2 to - 4.8	<b>⊸ 4</b> ∍8	0 . 6	Penetration attack of sintered metal
Chromium	Univ. of Cincinnati	1		<del>-</del> 5.7	0 . 7	
L - 605	Haynes Stellite Co.	3	~ 7.4 to - 6.0	<del>∘</del> 6∘8	1.3	Selective type corrosion
Vanad ium	KAPL	1		- 7.7	0 . 4	Continuous non-uniform film of VN and $V_2N$ or $V_2C$
Beryllium		3	- 18 to - 16.4	- 17	0 • 6	Unidentifåed discontinuous surface film. Fairly uniform attack
Thorium	Iowa State College	1	- 67°7 to - 45°4	- 56.8	0.6	
Nickel	International Nickel	3	- 100°4 to - 67°9	- 86	0 . 6	Severe intergranular attack
Manganese	Electro Manganese Corp.	3	Thin strip tested -	some solu	bility noted.	
Silicon		3	Dissolved and depos	ited on ca	psule wall.	

#### TABLE 5

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# Corrosion of Materials by Bismuth in Static Tests - 4 hrs. at 1000°C

MATER IAL	SOURCE	RUNS	WEIGHT CHANGE(m) RANGE	g/cm <sup>2</sup> ) AVERAGE	RATIO-SPECIMEN AREA TO COOLANT VOLUME. cm <sup>2</sup> /cm <sup>3</sup>	REMARKS
Niobium		2	+ 14.1 to + 24.5	+ 19.3	0 . 2	Continuous film
Molybdenum		2	+ 9.9 to + 11.0	+ 10∘4	0 . 6	Continuous uniform film plus fairly continuous intermetallic
Vanadium	KAPL	1		+ 9,9	0.4	Heavy continuous film formed some intergranular attack
Tungsten		2	- 39,6 <sup>*</sup> tő + 3,7	+ 3.7	0.6	Uniform attack
1040 Steel	SAE Steel Co.	3	- 0∘3 to + 4∘6	÷ 2.₀6	0 - 6	Uniform attack with selected corrosion of carbide
Tantalum		2	÷ 2 ⋭õ÷ 2₊9	÷ 2÷4	0 . 6	Uniform film over original surface plus penetration layer
Fe (Armco)	Corry Steel Co.	2	0 to + 0.2	+ 0.1	0.6	Uniform attack
I, ~ 605	Haynes Stellite Co.	3	-4 to+0.1	÷ 3.5	1 - 3	Continuous film intergranular type attack
Beryllium		3	- 25°4 to - 14°8	- 21	0.6	Fairly uniform attack
Cobalt	Kulite Tungsten Co.	2	∾ 30 to = 18∘4	- 24	0.6	Severe penetration type of attack
Chromium	Univ. of Cincinnati	1		- 126	0.7	Severe uniform attack
Silicon		3	Some weight loss no	Some weight loss noted		
Manganese	Electro Manganese Corp.	3	Thin strip tested	- partial	ly dissolved	
Nicke l	International Nickel	3	Essentially dissolved			
Zirconium	Bureau of Mines	3	Essentially dissolved		<i></i>	
Titanium	Remington Arms	3	Dissolved			
Thor ium	ISC	. 3	Dissolved			
Uranium	<u> </u>	3	Dissolved			

\* This value is probably not due to corrosion.

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evacuated Armco iron capsules for four hours at 1000°C. Samples were run in triplicate. The lithium was melted in the protective atmosphere of a heliumfilled dry box, drossed to remove floating impurities, and cast into inverted hollow capsule plugs, one of which is shown in Fig. 15. After cooling, the plugs were placed in a desiccator before removing from the dry box. In runs using bismuth, the dry box was not used but loading was carried out by charging a small ingot of bismuth, previously cast to fit into the capsules. To prepare for a run, the sample was placed in a capsule along with the coolant metal and plug, the plug was cold pressed into the capsule, the capsule evacuated, and the seam between plug and capsule heli-arc welded. The weld was helium leak tested and the capsule and contents heated to the melting point of the coolant with a vacuum still applied. The portion of the plug extending above the capsule was then crimped twice, sheared off by a third crimp, and spot welded. Heat for the test was furnished by four tilting Burrell Globar-tube furnaces, each capable of holding two samples. The tilting feature allows inversion of the capsules after each run, to drain the coolant metal from the sample. The furnaces were continuously flushed with helium to prevent oxidation of the iron capsule and to help prevent fire in case of container failure. After completion of a heating cycle the capsules were placed in a lathe, grooves cut through the capsule walls, and the plugs removed. This method is preferred to sawing since it results in less contamination of coolant due to chips from the capsule. Lithium or bismuth remaining was removed and the samples were weighed. The metal samples were sectioned and mounted for microscopic study, and surface films, if present, were identified by X-ray diffraction. The lithium and bismuth were analyzed spectrographically for sample components.

The bismuth used was specified as 99.95% pure, supplied by Belmont. Lithium was obtained from the Maywood Chemical Works of Maywood, New Jersey. The manufacturer's specifications indicated a product of 99.5% purity, the remainder being 0.20 calcium, 0.07 heavy metals, 0.03 iron and aluminum, 0.02 sodium and 0.009 silicon. Subsequently, lithium with 0.005% sodium, as supplied by the Metalloy Corporation of Minneapolis, Minnesota, was substituted.

The data for the various four hour runs at 1000°C are presented in Tables 4 and 5. They represent, with a few exceptions, weighted values for three specimens. The results are given in terms of weight change per unit of surface area rather than as a time rate of weight change, because the corrosion rate is



not likely to be linear, and at present data is available for one time interval only. Samples for test were made up of flats 3/4 in.  $\times 3/4$  in.  $\times 1$  in. when ever possible. Since the volume of coolant material placed in each capsule was uniform, variation in sample surface is reflected in the column headed "Ratio-Specimen Area to Coolant Volume." The value of this ratio for a standard specimen is 0.6.

Corrosion samples from each of the various elements and alloys tested are being studied to determine the nature and depth of attack, thickness of film formation and selective leaching of impurities.

Transverse sections were prepared and mounted adjacent to steel backing plates used to prevent rounding of the specimen edge during polishing. All specimens were carefully polished and examined microscopically in the unetched condition. Etching, it was felt, would perhaps destroy films and other corrosion effects at the liquid-metal interface.

Pure Metals in 1000°C (1832°F) Lithium. Zirconium (graphite melted) (Mag. 1000X) shows good resistance to attack. A thin continuous adherent film, identified by X-ray diffraction as ZrN, was found. Film thicknesses were approximately 0.1 mil and 0.4 mil respectively for two lots of zirconium metal tested.

Cobalt (sintered) (Mag. 500X) shows poor resistance to attack. Average corrosion depth is one mil. Several deep stringers were noted running to a depth of 6 mils. The intergranular attack is typical of that due to impurities at the grain boundaries.

Tantalum (Mag. 1000X) shows good resistance to 1000°C lithium, with formation of continuous thin TaC film.

Armco Iron (Mag. 1000X) shows extremely good resistance to attack. Edge roughness is presumably due to machining during preparation. All specimens are now being finished on 600 grade metallographic paper.

Columbium (Niobium) (Mag. 1000X) shows good resistance to attack. The gain in weight is attributed to a uniform film formation, approximately 0.1 mil deep. The coating has not yet been identified.

Molybdenum (Mag. 1000X) shows good resistance to short time exposure. The weight gain is attributed to irregular build-up of  $Mo_2C$  and MoC.

Tungsten (Wolfram) (Mag. 1000X) shows good resistance to lithium attack.

Nickel (electrolytic) (Mag. 100X) shows severe intergranular attack and deep penetration.

Alloy L605 (Mag. 1000X) shows poor resistance to attack. Selective leaching of one or more impurities is apparent from the zone of voids immediately below the surface layer. Depth of the selective type of corrosion was approximately 0.7 mil.

Titanium (Mag. 500X) shows fair resistance. The thin film formed on the outer surface, 0.1 mil thick, was identified as TiN. The heavier film underneath ( $\sim$  1 mil thick) and penetrating into the base metal, is believed to be TiC. This was Remington Arms graphite-melted grade Ti.

Beryllium (Extruded) (Mag. 500X) shows a scattered non-continuous film formation and large voids. Attack was severe.

Vanadium (Mag. 500X) shows a weight loss and a continuous non-uniform, unidentified film. Intercrystalline attack, commencing at the specimen surface and running to a depth of approximately 6 mils, was noted.

1040 Steel (Mag. 100X) showed decarburization throughout the entire sample. Apparently all the carbon has been removed from the sample.

Figures 16, 17, 18, and 19 show micrographs of the various metals tested in 1000°C (1832°F) lithium in the unetched condition.

Pure Metals in  $1000^{\circ}$ C (1832°F) Bismuth. Tantalum (Mag. 1000X) shows continuous but unidentified uniform corrosion product build-up on the surface. Depth of the product was approximately 0.4 mil. The product is composed of a heavier film on the outside specimen surface, approximately 0.3 mil thick, and a diffusion layer penetrating approximately 0.1 mil into the specimen.

Beryllium (Mag. 500X) shows a moderate to heavy attack, of the straight solution type.

Chromium (Mag. 500X) shows a severe attack, with pure bismuth adhering to the corroded surface.

Molybdenum (Mag. 1000X) shows a continuous heavy film formation which has been identified as alpha iron. This presumably results from mass transfer of iron from the container wall. A thin unidentified layer exists between the molybdenum specimen and the iron coating.

# CORROSION SAMPLES TESTED IN MOLTEN LITHIUM

1000°C FOR FOUR HOURS

EXPOSED SURFACE AT TOP OF PHOTOMICROGRAPH



i.







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Fig. 16

# CORROSION SAMPLES TESTED IN MOLTEN LITHIUM

UNCLASSIFIED Fig. 17

EXPOSED SURFACE AT TOP OF PHOTOMICROGRAPH



UNETCHED

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R.J. GRAY

# CORROSION SAMPLES TESTED IN MOLTEN LITHIUM

EXPOSED SURFACE AT TOP OF PHOTOMICROGRAPH

4


# CORROSION SAMPLES TESTED IN MOLTEN LITHIUM

UNCLASSIFIED Fig. 19

4

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### EXPOSED SURFACE AT TOP OF PHOTOMICROGRAPH







UNETCHED VANADIUM -7.68 mg./cm<sup>2</sup>

R.J. GRAY

Armco Iron (Mag. 1000X) shows a good resistance to attack with no apparent weight change or damage to metal specimens for short-time exposure, but this may be due to the fact that Armco iron was the container.

Vanadium (Mag. 500X) shows formation of a continuous heavy film (1.8 mils) identified by X-ray diffraction as alpha iron. It also appears that some selective corrosion occurred at the grain boundary.

Columbium (Niobium) (Mag. 1000X) shows a continuous unidentified film formation (approximately 0.1 mil thick) with pure bismuth metal adhering to the specimen surface.

1040 Steel (Mag. 1000X) shows selective corrosion (decarburization) occurring near the surface of the sample and extending to a depth of approximately one mil.

Sintered Cobalt (Mag. 500X) shows drastic attack, penetrating deeply into the specimen. Apparently the attack proceeded down through the sintered powdered particles and left a matrix of unattacked metal behind. Pure bismuth is present in the network.

Tungsten (Wolfram) (Mag. 1000X) shows a heavy uniform straight solution type of attack.

Alloy L605 (Mag. 500X) shows severe intercrystalline attack to a depth of approximately 6 mils. A continuous film was formed on the specimen surface, approximately 1 mil in thickness, identified as gamma iron.

Figures 20, 21 and 22 show micrographs of the various metals tested in 1000°C (1832°F) bismuth in the unetched condition.

The static tests are as yet quite incomplete, but even from this fragmentary information some materials can be eliminated from consideration. Others can be selected as having sufficient promise to justify further study and consideration, but the available information does not by any means warrant their ungualified recommendation as reactor materials.

In the case of bismuth the materials which show promise are jron, molybdenum, tungsten, and tantalum. The same materials, together with zirconium and a number of iron-base stainless-type alloys, warrant further study in lithium.

These and other tests not yet reported indicate the importance of giving consideration to the presence and nature of any third component which may be present in the system. Mass transfer of relatively insoluble materials-so-

# CORROSION SAMPLES TESTED IN MOLTEN BISMUTH 1000°C FOR FOUR HOURS

EXPOSED SURFACE AT TOP OF PHOTOMICROGRAPH



UNETCHED

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UNCLASSIFIED

Fig. 20



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# CORROSION SAMPLES TESTED IN MOLTEN BISMUTH

EXPOSED SURFACE AT TOP OF PHOTOMICROGRAPH

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R.J. GRAY

UNCLASSIFIED

Fig. 21

# CORROSION SAMPLES TESTED IN MOLTEN BISMUTH Fig. 22

EXPOSED SURFACE AT TOP OF PHOTOMICROGRAPH



76







UNETCHED

lution from one location and precipitation at another location, probably on the third component, without at any time being present in high concentrations in the liquid--appears to be a serious factor.

**Dissolved Fuel Tests.** Because of the recent interest in the liquid metal fuel type reactor, some exploratory corrosion tests were conducted to find potential materials for containing uranium bearing coolants. The lead-uranium and bismuth-uranium binary systems look somewhat promising from a uranium solubility viewpoint and for this reason were selected for these tests. The literature shows lead will dissolve up to 0.5 percent uranium at 900°C (1652°F) while upwards of 25 atomic percent uranium is soluble in liquid bismuth at 1000°C (1832°F). The minimum solubility of uranium in the coolant thought necessary for successful reactor operation is of the order of one atomic percent.

The materials tested were confined to some of the high melting metallic elements:

ſungsten	Titanium
Cantalum	Iron
Molybdenum	Nickel
Columbium	Beryllium
Zirconium	

Corrosion specimens 1 in.  $\times$  1/4 in.  $\times$  3 in. were prepared from strip stock and were immersed in the uranium bearing coolant by means of a vertical positioning rod. (See sectional view of test apparatus in Fig. 23.) The BeO crucible containing the coolant was housed in a cylindrical quartz tube heated externally by an electric resistance-type furnace. Tube ends were sealed with metal and rubber gasket fittings to permit conventional evacuation and purging of the test chamber prior to operation under an inert atmosphere.

The bismuth and lead bath metals contained approximately two atomic percent uranium for all tests. This amount may be in excess of the solubility of uranium in lead, however. Test temperature was 1000°C and test duration was four hours at temperature.

Preliminary results showed substantial attack on most materials with formation of films in most cases. Beryllium in the lead-uranium bath formed a UBe, coating, and also formed a coating (X-ray identification not completed) in bismuth-uranium. Molybdenum and tungsten showed the least attack of materials tested, but the results should be confirmed in longer tests which are being made.



SECTIONAL VIEW VACUUM ARGON CORROSION TESTING APPARATUS

#### DYNAMIC CORROSION TESTS

Thermal Convection Loops (Harps). Twelve convection loops are being made by the Philadelphia Pipe Bending Company. Six of the loops will be of 310 stainless steel, three of V-36 alloy, and three of L-605 alloy. Three of the 310 loops and one each of the L-605 and V-36 loops will be constructed of material shrouded over a tube of 1010 steel. The fabricator has experienced considerable trouble in trying to weld the sections of shrouded material together and still maintain an unbroken inner surface of iron. At present the base metal is 0.080 in. thick and the inner shell is 0.020 in. thick. One of the 310 stainless steel loops fabricated at Philadelphia will be sent to Battelle Memorial Institute for internal cladding with pure iron by vapor deposition.

The ORNL machine shop has fabricated one loop of a low carbon, deep-drawing, fully-killed steel and is fabricating two more of this low carbon steel and four each of 304 and 347 stainless steels. Material has been ordered for 446 stainless steel and for nickel loops, and efforts are being made to obtain loops of zirconium and molybdenum.

The engineering design group has drawn up a specification for a cylindrical pressure vessel to serve as a dry box for the thermal convection loops. The specification has been sent out for bids for three of the pressure vessels. The auxiliary equipment for the pressure vessels, such as heater and thermocouple cap assemblies, mounting and terminal plate assemblies, and air cooling jackets, are either on order or have been designed and are ready to be turned over to the shop for fabrication.

A lithium purifier constructed of 316 stainless steel with a capacity of ten pounds of lithium has been designed. The 316 stainless steel plate tubing and piping have been ordered and delivery of the material is expected within two weeks.

A thermal convection loop of the very low carbon steel has been filled with a lead bismuth alloy containing 47% bismuth. This loop will be operated at temperatures up to 800°F. As soon as a loop of high temperature oxidation resistant material is obtained, a similar setup will be made to operate at temperatures up to 1800°F. The heating characteristics and power requirements of the various parts of the loop will be determined, and the control and electrical equipment needed to operate the harps will then be determined.

Forced Circulation System. A system is being planned to serve as a simulated service corrosion test for reactor and heat transfer components. It is intended that this system will duplicate, as far as possible, the anticipated materials, component construction, velocities, and temperatures of the proposed Aircraft Reactor Experiment, although it will not duplicate the radiation effects or the total number of components planned for the reactor.

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#### LITHIUM ISOTOPE SEPARATION

G. H. Clewett

Chemical Research Division

The very superior heat transfer properties of lithium make it look very attractive as an aircraft reactor coolant. However, the normal 7.35 percent Li<sup>6</sup> content must be largely eliminated if the liquid is not to have too great a neutron absorption cross section; therefore, all of the likely techniques for producing tonnage quantities of separated Li<sup>7</sup> isotope, in a purity about 99.93% at a reasonable price, are now being explored. The various physicochemical methods are discussed in this section.

The small weight of the lithium isotopes with resultant large percentage difference in mass would seem to make the problem of separation of these isotopes a relatively easy matter if purely physical methods are used. Because of this a considerable portion of the total effort on this problem has been placed on development of the molecular distillation method which now appears to be entirely feasible, but must undergo considerable engineering development. It is further apparent that the very simplicity of the chemistry of lithium with its single valence state in compounds, the tendency of the ion toward extreme solvation in solution, and the dearth of complex forms of the element would appear to make the search for chemical methods somewhat difficult. This is true; however, the ease of handling a chemical process, possibly a two phase liquid liquid system, in conventional equipment at ordinary temperatures and the fact that it operates at, or close to, thermodynamic equilibrium at each stage appears to be sufficient justification for putting a sizable portion of the effort into the search for such a chemical system. Two phase liquid-solid systems must also be studied with perhaps less vigor but with some care and thought since sizable improvements continue to be made in methods of engineering liquid-solid contacting processes.

#### MOLECULAR DISTILLATION

Definite progress has been made toward the separation of lithium isotopes by distillation methods. A positive separation factor has been established and a tentative production rate determined. Several additional experiments in a single stage still (Fig. 24) have established a separation factor  $\alpha = 1.02$ or slightly greater. As a result of distillation over a fairly narrow temperature range (at around 450°C) some indication of evaporation rate, and therefore production rate, has been determined.

The experiments with the one stage still pointed to several changes and improvements in design that would be essential to smooth operation. As a result of this experience a two stage still (Figs. 25 and 26) was designed and fabricated which incorporated these modifications. This still has provision for reflux of liquid and the condenser-roof is so designed that the vapor progresses from stage to stage. Considerable time was spent in testing this apparatus and it has been only recently that sufficiently satisfactory operation was achieved to warrant sampling the product for assay.

A further step in the evaluation of the molecular distillation method has been the design and construction of a simple evaporation apparatus. This unit consists of a single evaporating surface and condenser and is to be used to determine the rate of evaporation of liquid lithium at various temperatures and pressures. The equipment has been test run only. Data which will be secured from this equipment should prove very useful in engineering calculations on multi-stage still design.

A packed column refluxing still has been designed and fabricated. This will be used to test the feasibility of higher temperature and higher pressure distillations of lithium. It should be placed in operation in the near future.

Considerable thought and effort have been devoted to the design of a multi-stage vertical molecular still. It is being held in abeyance until more information is available from the smaller equipment now being operated.

#### CHEMICAL EXCHANGE IN A LIQUID-LIQUID SYSTEM

Because of the many advantages of a liquid-liquid countercurrent process over a liquid-solid countercurrent process the search for such a system has been rather painstaking and thorough during this quarter. Furthermore, since one of the methods reported in the published literature, using lithium amalgamlithium salt solution, was of this category, it appeared that some of the development work should be in this direction.



### KEY

() ROOF

2 BOX

- 3 STILLPOT
- (4) INSULATION
- 5 SPARK PLUGS
- 6 HEATER

## 7 ROOF SUPPORT

- 8 RECEIVER
- 9 BOTTOM PLATE
- () GAUGE CONNECT.
- () VACUUM LINE

## BONNET

(3) WATER LINES FOR BOTTOM PLATE





FIGURE 24 SINGLE STAGE LITHIUM STILL

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- 1. Vacuum gage connection
- 2. Vacuum bonnet

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COLUMN.

- 3. Thermocouple wells
- 4. Radiation baffles
- 5. Coolant ducts
- 6. Still pot assembly
- 7. Condenser assembly
- 8. Heating elements
- 9. Flexible connection for coolant
- 10. Radiation baffle
- 11. Reflux ducts
- 12. Vacuum gasket
- 13. Spark plugs for electrical leads
- 14. Still support
- 15. Wilson vacuum seal
- 16. External coolant connection





Lithium Amalgam-Lithium Salt Solution. Considerable effort has been devoted to a study of the lithium amalgam-organic solution exchange as first reported by Lewis and McDonald. (24) Initial experiments using ethanol (95%) made it very apparent that the rapid reaction of the water and alcohol with the lithium in the amalgam would make this system unsuited for sustained operation unless this reaction could be either eliminated or materially reduced. Experiments with absolute alcohol (H,0 about 700 ppm) indicated that although the moisture content was a very important contributor to the reaction, the alcohol itself reacted too rapidly to be practicable. This discovery led to an investigation of the higher alcohols up to isoamyl. It was found that the rate of reaction decreased substantially as the carbon chain was lengthened and it appeared from static tests on hydrogen evolution that isoamyl alcohol had definite possibilities. It soon became apparent that a new difficulty must be overcome, namely emulsions. Isoamyl alcohol, with or without dissolved lithium chloride, forms very stable emulsions with lithium amalgam. More than 150 organic solvents of suitable structure were examined for their ability to stabilize the system against the formation of emulsion. No success was achieved with this program. It was discovered, however, that those materials which were known to react chemically with the lithium in the amalgam were very effective in breaking and preventing emulsions. In this group are water, methanol, ethanol, etc.

Following this initial disappointment with the amalgam system both from reaction and emulsion standpoint, a survey was made to find some organic solvent which would neither react chemically nor cause emulsions. Of a rather wide group of organic liquids representing nearly all the major classes of compounds not a single example has been found which measures up to the desired characteristics. As a matter of fact it appears that the following generalization can be made: those organic liquids which do not have a replaceable hydrogen (or hydroxyl group) atom when contacted with lithium amalgam form stable emulsion with the amalgam. All indications point to the conclusion that the lithium is selectively located at the emulsion interface as the stabilizing agent for the dispersion.

The magnitude of the separation factor of the amalgam exchange system has been determined by batch shake-out methods. The procedure was to contact

(24) Lewis, G. N. and MacDonald, R. I. "The Separation of Lithium Isotopes," Am. Chem. J. 58, 2519-2524 (1936). lithium amalgam (about 0.5 molar) with an absolute ethanol (700 ppm moisture) solution of lithium chloride for two minutes. The amalgam was then divided in two equal parts and a new alcohol solution made from the lithium in one of the portions. These operations were carried through five stages.

Reaction of the lithium in the amalgam with the alcohol solutions caused a 33% depletion during the five operations. The over-all separation effect observed from this experiment shows  $\alpha$  to be 1.0495 as based on a single set of assay results. This high separation factor makes it almost mandatory to continue the search for a suitable method of utilizing the amalgam system for separation of the lithium isotopes.

Aqueous-Organic Systems. Although an aqueous phase in contact with an immiscible organic phase is one of the first possibilities which suggested itself for a two phase system, it now appears that only selected examples of this type or greatly modified solutions such as lightly salted aqueous solutions may approach the arbitrary criteria set up for suitable exchange systems. Since the distribution of lithium chloride between water and isoamyl alcohol is not completely unfavorable, two separate batch extraction experiments were carried out on this system. It now appears, after a recheck on the assay of the first experiment that no significant enrichment was obtained in these two attempts. A somewhat more favorable distribution ratio of lithium salt between phases can be obtained if the aqueous phase of any two phase system is highly salted with some inert salt. This phenomenon is being studied at some length and multiple batch separation experiments will probably be conducted on selected systems of this type.

Organic-Organic Systems. The predominantly hydrophillic properties of lithium ions prompted an investigation of the possibility of discovering a suitable pair of immiscible organic solvents as the liquid-liquid media for a separation system. More than 400 organic-organic systems were examined. Of these, approximately 60 were found to be immiscible pairs. Many of these pairs were rejected from qualitative observations of viscosity and phase separation. Of the remaining systems 12 pairs show sufficient promise to warrant more thorough investigation. These pairs are:

- 1. Butyl acetate vs. ethylene glycol
- 2. Hexone vs. ethylene glycol
- 3. Benzaldehyde vs. ethylene glycol

- 4. Dimethyl aniline vs. ethylene glycol
- 5. Ethyl benzoate vs. ethylene glycol
- 6. Nitromethane vs. octyl alcohol
- 7. Formamide vs. butyl acetate
- 8. Formamide vs. hexone
- 9. Formamide vs. octyl alcohol
- 10. Formamide vs. ethyl ether
- 11. Formamide vs. dimethyl aniline
- 12. Formamide vs. ethyl benzoate

These systems are now in process of investigation to determine the distribution of lithium in equal volumes of the liquids and the saturation solubilities of various lithium salts.

An added phase of this study has been the preparation of special lithium compounds which would appear to possess the desired solubility properties for for these organic solvents. Among those already prepared in this laboratory have been the following:

- 1. Lithium perfluorobutyrate  $(LiC_4F_70_2)$
- 2. Lithium thiocyanate (in hexone)
- 3. Lithium salicylate

(The work on these varied projects is being intensified.)

**Pulse Column Application**. Pulse columns<sup>(25)</sup> have been investigated for their possible application to the lithium amalgam system. These columns have several advantages over ordinary countercurrent gravity columns, the principle one being short stage length.

The pulse column work was carried out concurrently with the laboratory examination of the amalgam system and, if anything, highlighted the difficulties inherent in the reaction and emulsion problems.

A series of experiments were made to test the effective height of a theoretical stage. These were made by contacting a lithium amalgam with a solution of sodium ions. The apparatus used is shown in Figs. 27 and 28. The results were inconclusive because of the fact that reaction with the solvent medium in all cases nearly depleted the alkali metal content of the amalgam.

(25) A description of the pulse column is given in U. S. Patent 2,011,186 (1935); HW 14728, The Design and Operation of the Pulse Column, Burns, Groot and Slansky (Oct. 12, 1949).





A series of tests were made to determine the relationship between screen size and flooding rate in the one-inch glass pulse column. This data is summarized in Table 6 which shows flooding rates observed in the system mercury vs. 95% ethanol which is believed to be a close approximation to any amalgam system which might be used.

SIZE OF SCREENS (1 in. Separations)		RATE OF FLOODING
WIRE (in.)	HOLE (în.)	(cc/min of Mercury)
.010	.015	>240
.009	.011	>218
.0075	₀ 0092	>227
.0065	。0078	>242
.0055	<b>∝0070</b>	>247
.00525		26.7
.,0035	0055	19.9
	EENS (1 in. Se WIRE (in.) .010 .009 .0075 .0065 .0055 .00525 .0035	EENS (1 in. Separations)   WIRE (in.) HoLe (in.)   .010 .015   .009 .011   .0075 .0092   .0065 .0078   .0055 .0070   .0035 .0055

#### TABLE 6

Several runs were made to test the action of amalgam against lithium chloride solutions in the pulse column. With the lower alcohol solvents the reaction rate proved excessive. In one experiment using absolute alcohol (700 ppm moisture) the initial rate of loss of lithium from the amalgam (0.5 molar in lithium) was 2.8% loss per minute and an over-all loss of 96% was observed in 47 minutes in this experiment. Special efforts were made prior to this test to insure that the equipment was absolutely dry and the entire test was made under inert atmosphere to reduce moisture pickup.

The emulsifying tendency of such solvents as n-butyl alcohol, isoamyl alcohol, and pyridine was demonstrated to be a major problem by operation in the pulse column, and the selective removal of the lithium from the bulk of the mercury to the dispersed phase was clearly demonstrated in several of these tests.

The pulse column is now being used to make preliminary investigations on some of the more promising organic organic systems that are under investigation in this laboratory.

#### CHEMICAL EXCHANGE IN A LIQUID-SOLID SYSTEM

The practical application of a countercurrent liquid-solid system is considerably more difficult than either the liquid-liquid or liquid-gas systems. In spite of this fact, some of the more promising liquid-solid systems have been investigated.

A cellulose column was set up using filter aid pads as the packing material. Five separate runs were made using various eluents to remove the lithium ions previously deposited at the head of the column. The results of these tests were sufficiently discouraging to cause abandonment of this project.

Another liquid-solid system tried comprised a column filled with finely ground lithium carbonate and eluted slowly with water. As was the case in the cellulose experiment, no enrichment of the isotopes was observed and this project was also discontinued.

Isotope separation by the use of synthetic zeolites, such as Dowex 50, is being investigated in the X-10 area of ORNL. Some success has been achieved by use of this technique. This work is reported in detail in the Chemistry Division quarterly reports.<sup>(26)</sup>

#### AUXILIARY STUDIES

All the lithium assays made to date have been determined by use of a modified Nier mass spectrometer. Another method of assay is being developed using fission counting technique in which the  $\alpha$  particles from the <sup>6</sup>Li neutron reaction are counted. Although still in the development state, this method appears promising.

Several attempts have been made to determine the vibration frequency of the lithium ion-solvent bond in the Raman range. No distinct band which could

(26) Swartout, J. A., Chemistry Division Quarterly Progress Report for Period Ending March 31, 1950, Part I (June 16, 1950).

be attributed to the lithium-solvent bond was observed. This work has been abandoned, at least temporarily.

#### **RADIATION DAMAGE**

#### **REACTOR MATERIALS**

#### D. S. Billington, Metallurgy Division

The behavior of materials when exposed to the high neutron flux anticipated in an aircraft reactor is of extreme importance in the design of such a reactor since it must be assumed that material properties of importance may change significantly. There is no adequate theory which can give useful predictions and experimental data are at present meager. Experiments in this field are difficult and often very lengthy, so it is important that experiments which will answer the unknowns involved in the construction of the reactor be initiated in the near future. Planning such experiments is complicated by the present uncertainties about the reactor itself, as neither materials to be used nor exact design have been determined. A further complication is that neutron fluxes of the proper magnitude do not exist. However, experiments are in progress and more are planned making use of both accelerators and piles, which will yield information of particular interest for reactors as envisaged at present as well as information of a general nature. The equipment and experience resulting from these experiments can be applied to different materials and design problems as the requirements become known. Fuel element design has not yet progressed to the point where testing of samples can be started profitably.

#### Accelerator Experiments.

(a) An experiment has been set up by North American Aviation, Inc. that makes it possible to study the effect of deuteron bombardment on the corrosion rate of Armco iron by lithium at  $1000^{\circ}$ C. It is thought the deuteron bombardment will simulate to a high degree the behavior of this system under neutron bombardment, and thus will give answers on this important problem in a relatively short time. The apparatus has been assembled and is being bench-tested at the present time. Irradiation data are expected very shortly. It is planned to extend the experiment to include molybdenum in contact with both lithium and sodium, also at 1000°C. An ORNL man has joined the NAA group for the purpose of assisting in the experiments.

(b) The Purdue Group is expected to begin work by July 1. The following experiments are tentatively being planned:

(1) Study of SiC as an analog material for  $Be_2C$ .\* Measurement of electrical property change induced by deuteron bombardment.

(2) Effect of bombardment on creep rate.

(2a) Repetition of Andrade experiment (27) using alpha particles and deuterons.

(2b) Effect of bombardment on creep rate of stainless steel and other high temperature materials

(2c) Effect of bombardment on creep rate of single crystals above and below recrystallization temperature.

In-Pile Creep Tests. A creep test apparatus (illustrated schematically in Fig. 29) capable of operation under irradiation in a stringer in the ORNL pile is being developed. In the preliminary model now being bench-tested, a specimen nine inches long and one-eighth inch in diameter is enclosed in a constant-temperature furnace throughout about two-thirds of its length. The specimen is to be stressed by means of cables extending outside the pile where a dead weight furnishes a constant load. The central four-inch section of the specimen is the gauge length over which the strains are measured. Longitudinal strain is transmitted to a microformer by extensometer elements welded to the ends of the gauge length. A duplicate microformer whose core may be moved a known distance by an electromagnet controlled from outside the pile will be included in the apparatus alongside the strain measuring microformer so that any effect of temperature and radiation may be compensated for. An entire duplicate creep apparatus may be placed in series with the load supply cables outside the pile so that the difference in creep rates, in-pile and out-of-pile, may be observed.

In bench tests the temperature control system now in use has proved capable of holding the test bar temperature to within  $\pm$  1° at 1200°F. The variation of temperature over the gauge length is now  $\pm$  4°F. Work is now directed toward minimizing this value to  $\pm$  1°F, at which time the bar will be loaded to test the loading system and strain measuring instruments.

<sup>\*</sup> Accelerator experiments on Be, BeO, Be<sub>2</sub>C, and Zr using particles above 1 Kev are now classified. The possibility of using Mg<sub>2</sub>Si as an isoelectric analog material for Be<sub>2</sub>C is being studied.

<sup>(27)</sup> Andrade, E. N. da C., "Effect of Alpha~Ray Bombardment on Glide in Metal Single Crystals," *Nature* 156, 113 (1945).

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Graphite Stringer	Quartz Iron Furnace Tube Mica Insulation Vichrome Heater Windings Quartz Tube Quartz Tube Magnesium Oxide Powder
	Test Bar 4 Extensometer Elements Sont Microformer
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Dead Weight At Pile Face

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IN PILE CREEP APPARATUS For Insertion In Stringer In ORNL Pile

Dwg. No. 9146 Fig. 29

The above apparatus is designed to supplement the creep experiments of the NEPA Group It is felt that between the two groups all ranges of temperatures and types of materials of interest can be studied in the existing pile facilities.

The NEPA and ORNL Groups are investigating jointly with Argonne the use of special facility at Hanford that will permit dead-weight loading of creep units. Use of dead-weight loading permits considerable simplification in the design of creep units.

At this time it appears desirable to study the creep properties of pure metals such as molybdenum and iron and alloys of the stainless type in addition to the ceramics being studied by NEPA.

Diffusion of Fission Products. It appears necessary to obtain more data on the diffusion of fission gases and fission products in various metals than is presently available in the literature. For example, it seems evident that the fission gases do diffuse out of undamaged uranium, though at a rate so small as to be insignificant in times of interest in an aircraft reactor. It also seems evident that the inert fission gases will not diffuse through metals such as stainless steel even at 1000°C in the absence of radiation. This seems to sum up our immediately useful knowledge. Thus it is necessary to measure diffusion rates of uranium in molybdenum and other high temperature materials with and without irradiation.

There seems to be a lack of information also on the diffusion of the halogens at high temperatures. Nor is it known whether a thin cladding of inert metal on uranium would prevent accumulation of fission gases. In an attempt to gain an answer to these and other problems diffusion experiments in and out of the reactor are being planned.

Hot laboratory space is being made available, and it is expected that some measurements such as hardness, electrical resistivity, elastic modulus, magnetic susceptibility, and tensile strength on some samples of Inconel, Hastelloy, stainless steel, and zirconium that were irradiated in the Argonne channel experiment in the X-10 reactor will be obtained. Measurements will also be started on some stainless steel samples irradiated at Hanford. Samples of Ti, Mo, Ni, and Armco Fe suitable for mechanical property measurements will go into the "H" pile via the Argonne channel experiment, according to the latest revised Hanford schedule, on June 29, 1950.

#### AUXILIARY MATERIALS

#### O. Sisman, Reactor Technology Division

**Plastics**. This work has been temporarily delayed because of the construction work which has been going on in the pile building. It is expected, however, that most of the normal pile irradiations will be completed in the next three months. Work is also planned to determine the effect on radiation stability due to irradiation rate, oxygen and antioxygens, and to determine the damage due to thermal neutrons, fast neutrons and gamma radiation. Design of an apparatus for subjecting specimens to a  $10^5$  r/hr gamma source is nearly complete.

Metal Hydrides. The samples of titanium, zirconium, and lithium hydrides, on which data were presented in the last quarterly report, have been removed from the pile and are being held in storage until they have decayed sufficiently to permit a chemical analysis on the materials and the containers. It is hoped that a better interpretation of the data will be possible after the chemical analysis.

Preparations are being made for further studies on titanium hydride, lithium hydride and uranium hydride. The apparatus for determining the dissociation pressures of these materials under pile radiation up to temperatures of about 500°C will be installed in the pile during the coming period.

Liquid Metals In-Pile Experiment. To fully evaluate the feasibility of using liquid metals as a primary reactor coolant it is desirable to study these materials in a flowing system under radiation. Work was started in April on the design of a loop (Fig. 30) in which liquid metals may be circulated through the X-10 reactor at high temperatures. The primary data to be gained from such an experiment will be (1) to determine the activity pickup of the liquid metal from the pipe through which it is circulated, (2) to determine the radiation effect on corrosion (and erosion) rate, and (3) to develop techniques for handling liquid metals under radiation at high temperatures.



## LIQUID METALS "IN-PILE" EXPERIMENT

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The first experiment will use 316 stainless steel as the container and the liquid metal will be lithium, but the equipment will be designed so that the loop containing the liquid metal may be readily interchanged and any combination of container and liquid metal may be used. It would be most desirable to use Li<sup>7</sup>, instead of natural lithium, and a request has been placed with Y-12 for an estimate of the time required and the cost to produce two pounds of Li<sup>7</sup> by electromagnetic separation. It is thought that a purity of 99.5% Li<sup>7</sup> will be sufficient for this experiment although 99.93% is the estimated requirement for the final reactor. In the event that Li<sup>7</sup> is not available, a highly purified natural lithium will be used (the sodium, in particular, must be removed).

Design of all internal portions (parts that will fit into the pile proper) has been completed and turned over to the shop for fabrication. This portion of the apparatus consists of an electrically heated loop of 5/16 in. OD tubing approximately 20 feet long to contain the lithium. A 2 in. evacuated tube jackets the loop and contains foil radiation shields. The jacket also serves as a vacuum chamber which will insulate against heat loss by convection and at the same time aid in detecting lithium leaks since the lithium vapor will cause a decided pressure rise in the vacuum system. Insulation will be packed around the jacket and will be retained by an aluminum can of 3-3/4 in.  $\times 3-3/4$  in.

Since the maximum allowable temperature of the outside of the aluminum can is 250°C in the pile and 75°C in the concrete of the pile shield and since it is desired to maintain the lithium ultimately at 1000°C, the internal parts and vacuum system will be assembled first and heat transfer data obtained to determine whether a cooling coil is needed in the insulation.

The external portion of the system will consist of an electrically heated reservoir over which an inert atmosphere will be maintained, an electromagnetic pump, an electromagnetic flow meter, and activity detecting equipment. An a-c electromagnetic pump is on order from General Electric, and delivery is expected in June The flow meter has not yet been designed.

Calculations are being made on the activity to be expected in the loop based on the pessimistic assumption that half of the 316 tubing will be removed to the lithium From the results of these calculations decisions will be made about the type of activity counting equipment required and the necessary shielding for the portions of the system which will be outside of the pile.

#### NUCLEAR MEASUREMENTS

#### A. H. Snell, Physics Division

It seems quite likely that the eventual aircraft reactor as well as the ARE will be of the intermediate type. This will create a demand for greatly improved intermediate cross-section data for several elements. Both experimental and theoretical efforts are underway to accumulate this information.

#### HIGH VOLTAGE PROGRAM

A High Voltage Laboratory building has been conceptually designed. It will house the 5 Mev NEPA Van de Graff machine along with the present 2 Mev machine and the Cockcroft-Walton set. It is hoped that detailed design and construction can be rushed so that the 5 Mev machine will not wait in idleness for more than a few months. Two men have been obtained for cross-section measurements. They are experienced and interested in spectrometry of this nature.

#### MECHANICAL VELOCITY SELECTOR

AEC approval of the change recommendation covering construction of the mechanical velocity selector is expected momentarily. After considerable study, a tool steel rotor about 12 inches in diameter has been decided upon. This will contain hydrogenous scattering material with slits, and will rotate at about 10,000 rpm. An 80-channel recording system is envisaged. A second man will join the project this month

### PUBLICATIONS

(Li:	st of External Reports Issued During the Last Quarter)
ORNL 535	Determination of the Fast Flux in Hole 19 O. Sisman and C. D. Bopp
ORNL 649	Uranium Hydride, A Survey A. S. Kitzes
ORNL 665	The Approach to Critical With the Bare Intermediate Reactor N. M. Smith
ORNL 668	A Diffusion Solution for the Cylindrical Ducting Problem of Infinite Geometry D. Whitcombe
ORNL 684	The Atomic-Powered Aircraft, January 1950 C. B. Ellis
ORNL 710	Theoretical and Practical Aspect of Shielding A. S. Kitzes and T. Rockwell