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QUARTERLY TECHNICAL PROGRESS REPORT
AEC UNCLASSIFIED PROGRAMS
JULY-SEPTEMBER 1965

AEC Research and Development Report

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS



ATOMICS INTERNATIONAL

A DIVISION OF NORTH AMERICAN AVIATION, INC.

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The preceding Technical Progress Report
was NAA-SR-11450

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- I -

FAST-REACTOR DEVELOPMENT

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Program: Fast Reactor Development

Project: Parameter Surveys

Project Engineer: A. S. Gibson

Reporting Period: July-September 1965

AEC Category: 04-01-04-19

General Order: 7661

Subaccount: 1311

I. PROJECT OBJECTIVES

The general objectives of this project are: to review certain parameters which sensitively affect the economic, safety, or mechanical design characteristics of fast-reactors and determine the nature of these effects; to determine the safety-economic tradeoffs which may be made by emphasizing one feature over another in fast-reactor design; and to evaluate those components and operating characteristics which have an effect on operating reliability. A specific objective is to determine the safety-economic tradeoffs of various types of fuel. These studies will be designed to determine the relative economics and development problems associated with sodium bonded and unbonded carbide fuel, unbonded oxide fuel, nitride fuel, and problems associated with venting or containing fission products released from these fuels. Similar tasks are associated with identification of safety-economic interrelationships and development problems for reactor mechanical components and systems.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

The project was opened in the current quarter. Major accomplishments to date are: (1) the demonstration that system pressurization does not improve the safety characteristics of sodium cooled fast reactors by materially increasing the time required to reach boiling for a given transient, and (2) the demonstration that safety-economic interrelationships can be used to assist in establishing development goals and design criteria, particularly with regard to fuel development.

III. PROGRESS DURING REPORT PERIOD

A. SODIUM PRESSURIZATION STUDIES

To determine the effect of sodium pressurization with the resulting boiling point increase on the progression of potential fast-reactor accidents, a study was conducted of various accident situations with and without sodium pressurization. A 1000-Mwe fast-reactor with characteristics described in NAA-SR-11378, Vol I (for the core design designated as SB Mod 3) was used as the basis for the study, and the kinetic code AIREK 3C (described in NAA-SR-11378, Vol III) was used to make the transient analysis. Initial effort involved an evaluation of the effects of a flow reduction accident and a reactivity insertion accident on different fuel types without significant sodium pressurization ($p = 33$ psia). The boiling phenomenon was handled identically in all cases. At the conclusion of the vaporization of 5% of the coolant within a node, it was assumed that film boiling coefficients would apply and heat transfer coefficients were switched to take this into account. In addition, in the bonded vented core the

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bond was assumed to vaporize when the cladding reached the saturation temperature of the coolant and heat transfer coefficients were switched accordingly.

Table 1 presents the response of five different types of cores to a reduction in coolant flow. Coolant flow was reduced step-wise to 10% of the removal value at $t = 0$. The numbers in the table represent the elapsed time in seconds before fuel melting, clad melting, and coolant boiling. The time for coolant boiling and clad melting is representative of the average channel; the time for fuel melting is representative of the maximum channel. Fuel melting did not occur in any of the cores in the average channel before termination of the transient. The power densities are per fuel pin in the fast region of the core.

TABLE 1
FLOW REDUCTION ACCIDENT
(Reduction of flow to 10%)

Run No.	Core Type	Average Core Power Density (kw/ft)	Fuel Melt Time (sec) at 4900° F (Oxide) and 4300° F (Carbide)	Clad Melt Time (sec) at 2500° F	Coolant Boil Time (sec)
5	Oxide Unbonded	11	0.75	1.60	0.65
1	Carbide Bonded Unvented	19.5	4.00	2.55	0.65
17	Carbide Bonded Vented	19.5	2.00	6.00*	0.65
12	Carbide Unbonded	16.6	2.80	1.60	0.45
20	Carbide Unbonded	20.2	1.40	1.37	0.80

*Clad melts when fuel melts in average channel.

In Run 20 the mass of the cladding was lumped with that of the coolant and in Run 12 it was ignored. This accounts in part for the longer time required to reach the saturation temperature. Fuel and cladding melting occurred sooner in Run 20. This is undoubtedly due in part to the approximately 20% greater power density and may also be affected by changes which were made in the blanket region time constants. The blankets contribute approximately one-half of the Doppler feedback.

Runs 1 and 17 were identical except for the assumption that the bond would vaporize in the vented core when the cladding reached the coolant saturation temperature. This virtually stopped all heat transfer from the fuel causing it to melt in half the time that it did in the unvented core where no vaporization of the bond was assumed to occur. When the bond vaporizes, the cladding temperature drops to very close to that of the coolant temperature and the model shows no cladding melting. The cladding would melt, however, upon contact with molten fuel and it is this time which is indicated in Table 1.

Figure 1 shows the effect of the loss-of-flow (to 10%) accident on cladding temperatures (outlet node) for the oxide fueled core at 33, 100, and 240 psia. These pressures correspond to sodium boiling temperatures of 1800, 2100, and 2400°F respectively. The cladding temperature follows coolant temperature

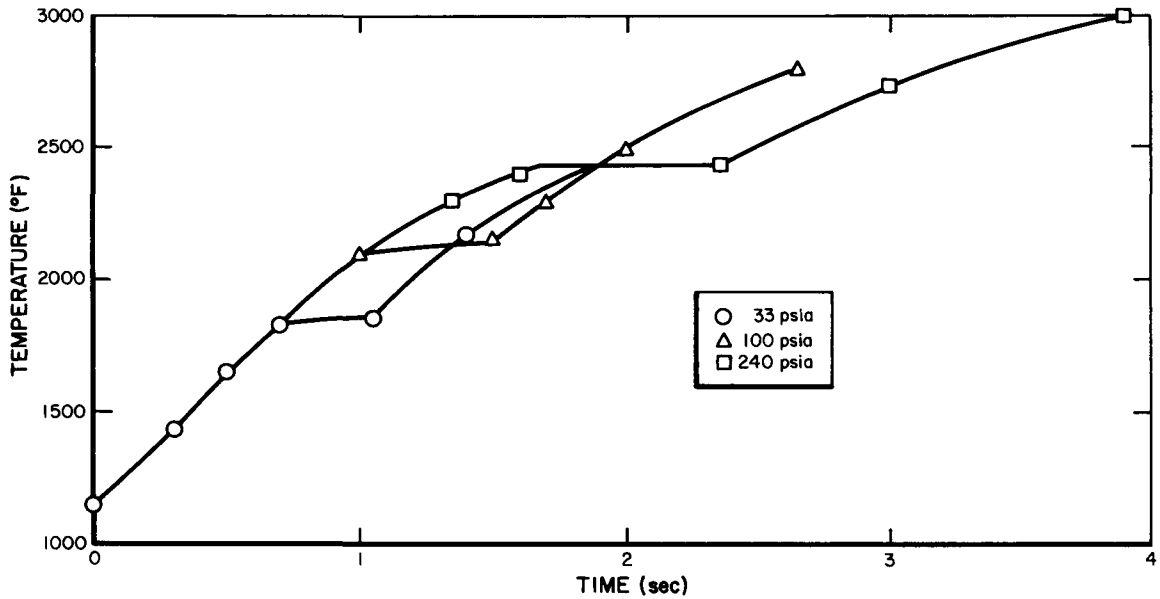


Figure 1. Cladding Temperature of Average Oxide-Core Channel

closely as coolant temperature increases to the boiling point and through the early stages of boiling. When film boiling occurs with its resultant poor heat transfer, the cladding temperature seeks a new level much closer to that of the fuel. Figure 1 shows that there is no advantage to pressurizing the system as it has little effect upon the time required for the cladding to reach an unsafe temperature (approximately 2500°F). At the time cladding reaches 2500°F in these transients, only the centerline fuel in the center node of the hot channel has reached its melting point while the average fuel temperature is still at 3000°F or below.

Table 2 presents the response of three of the cores to a reactivity insertion followed by a scram. \$1.50 was inserted at \$100/sec at $t = 0$; beginning at 0.2 second, -\$4 was inserted at -\$10/sec. No coolant boiling or clad melting occurred during Runs 13 and 23. No reactivity insertion transients were considered in the oxide core; since for any appreciable insertion, the fuel vaporizes before the sodium reaches its boiling point at even the lowest system pressure. The system pressure is therefore of no concern in the reactivity insertion transient.

Loss of flow studies (to 10%) were made for the sodium bonded, unvented carbide core shown on Figure 2, leading to the same conclusion as that reached for the oxide core. Actually the cladding reached melting temperature (2500°F) slightly quicker at the higher pressures than at 33 psia; however, considering the approximations which must be made in this type of study, slight differences are inconsequential.

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TABLE 2
 REACTIVITY INSERTION ACCIDENT
 (\$1.50 reactivity insertion followed by \$4. scram)

Run No.	Core Type	Average Core Power Density (kw/ft)	Fuel Melt Time (sec) at 4300° F	Clad Melt Time (sec) at 2500° F	Coolant Boil Time (sec) at 1800° F
6	Carbide Bonded Unvented	19.5	0.0346	0.35	0.20
13	Carbide Unbonded	16.6	0.0223	-	-
23	Carbide Unbonded	20.2	0.0202	-	-

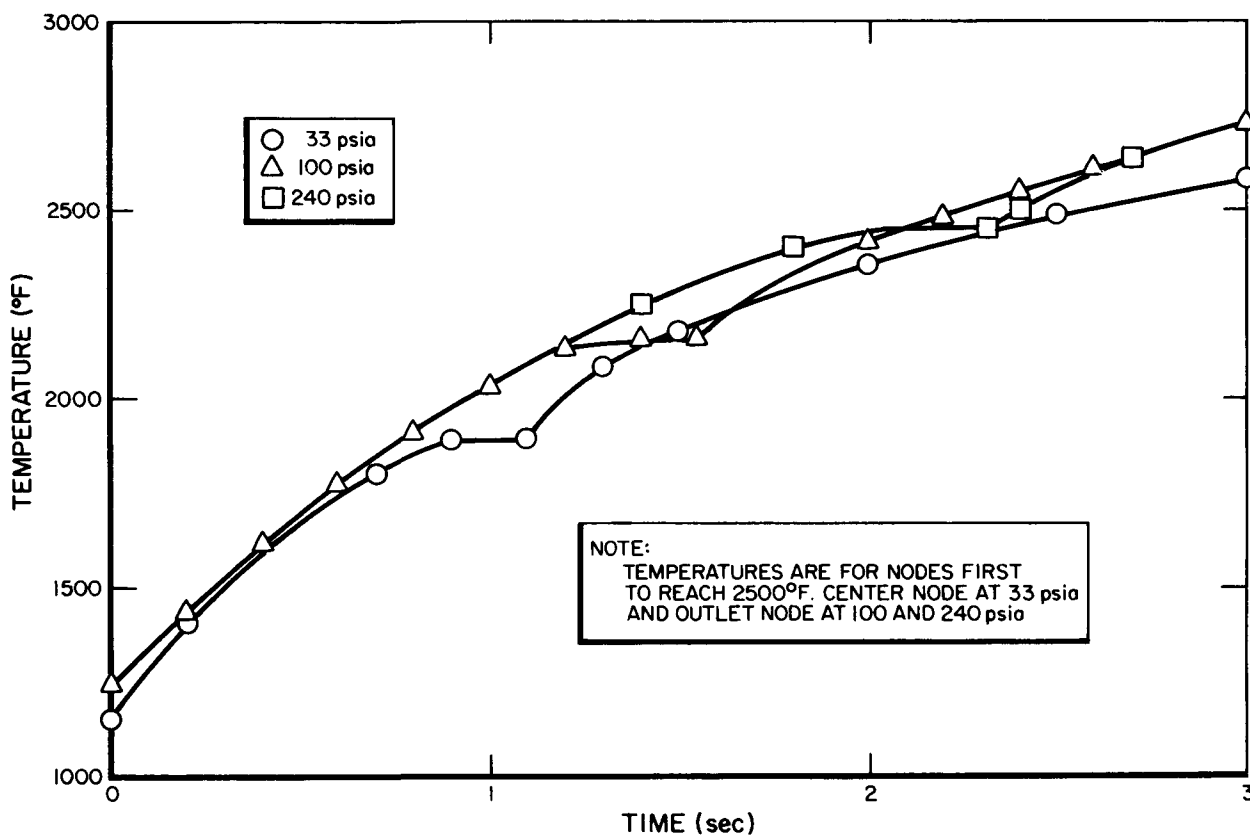


Figure 2. Cladding Temperature of Average Carbide-Core Channel

The higher conductivity and lower temperature of the carbide fuel makes the system pressure a matter of some concern during reactivity insertion transients. Consequently, studies of reactivity insertions of \$1.50, \$1.75, \$2.00, and \$2.25 have been made. Preliminary results show that while \$1.50 causes cladding melting at 33 psia, \$1.75 may be tolerated at 240 psia without cladding damage. There is no sodium boiling with the \$2.00 insertion at 240 psia; however, cladding

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temperatures are too high. In all of these reactivity insertion transients, a scram was assumed to occur at 0.2 sec which inserted $-\beta$ in 0.4 sec.

B. FUEL CYCLE STUDIES

To determine the economic potential and the relative safety-economic trade-offs between various fuel concepts, a study of these effects was initiated. Parameters being investigated are shown in Table 3.

TABLE 3
FUEL CYCLE PARAMETERS

Item	Parameters
Type of Fuel	Mixed-Oxide Mixed-Carbide Mixed-Nitride
Fission Gas Containment	Vented Nonvented
Fuel Properties	Density Thermal Conductivity
Thermal Bond	Gas Sodium
Geometry	Diameter
Temperature	Fuel Center Cladding
Irradiation	Burnup

Initial work has involved scoping studies on mixed-carbide to compare the economic and safety interrelationships between nonbonded carbide, bonded carbide, and nonbonded oxide. The bonded carbide and nonbonded oxide fuels are compared in NAA-SR-11378, Vol I, on an economic basis. Table 4 shows the thermal-hydrodynamic data for a mixed-carbide core with gas bonded fuel pins in a 1000-Mwe reactor operating at an average linear power in the fast region of 11 kw/ft. For a gas bonded fuel pin with a contact resistance of 1500 Btu/hr-ft²-°F, an average linear power in the fast region of 11 kw/ft results in a peak center temperature in the hottest fuel element of 1300°F.

Previous studies in NAA-SR-11378, Vol I showed that sodium bonded carbide fuel operating at an average linear power in the fast region of 13 kw/ft and at a peak center temperature in the hottest element of 2100°F enjoyed a slight economic advantage compared with oxide fuel operating at a peak temperature in the hottest element of 4700°F (and an average linear power of 9.6 kw/ft). Based upon extrapolation from this data, it appears that the nonbonded carbide core described in Table 4 will show approximately equivalent economics to the oxide fuel. Based upon limited kinetics studies, it also appears that the nonbonded

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TABLE 4
1000-Mwe REACTOR WITH GAS BONDED FUEL
(Preliminary Design Data)

Parameter	Fast Region	Inner Blanket	Outer Blanket
Type of Fuel	PuC - UC	PuC-UC	PuC-UC
Number of Modules	16	16	16
Average Enrichment (%)	23.5	3.0	3.0
Active Core Height (ft)	4.3	4.3	4.3
Axial Blanket Height (ft)	2.0	2.0	2.0
Fuel Diameter (in.)*	0.220	0.350	0.350
Clad Thickness (in.)	0.0150	0.010	0.010
Fuel Rod OD (in.)	0.250	0.370	0.370
Fuel Rods/Element	127	91	91
Total No. of Elements	256	288	224
Volume Fractions of Module (%)			
Fuel and Gas Gap	23.3	42.3	42.3
Sodium	61.5	44.9	44.9
Steel	15.2	12.8	12.8
Overall Power Split (%)	63.6	21.5	14.9
Total Power/Region (Mw)	1590	540	370
Axial Peak/Average Power	1.28	1.28	1.28
Rod Radial Peak/Average Power	1.29	1.76	1.83
Power Hot-Channel Factor	1.41	1.41	1.41
Average Linear Power (kw/ft)	11.4	4.8	4.2
Nominal Maximum Linear Power (kw/ft)	18.8	10.8	9.9
Peak Linear Power (kw/ft)	26.5	15.2	14.0
Coolant Inlet Temperature (°F)	750	750	750
Nominal Coolant Temperature Rise (°F)	400	400	400
Peak Fuel Temperature (°F) [†]	3015	1869	1823
Peak Clad Temperature (°F)	1304	1187	1186

*Including a gas gap of ~2 mil

†Based on a fuel conductivity of 10 Btu/hr-ft-°F, a gap heat transfer coefficient of 1500 Btu/hr-ft-°F, and 100% mixing of coolant.

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carbide core described in Table 4 will behave approximately the same as the oxide core for the two accident situations described under III-A above. Due to the higher thermal conductivity of the sodium bonded carbide fuel, it is expected that its response to transient inputs will be different from that of the gas bonded fuel.

C. PLANT SYSTEM TEMPERATURE AND PRESSURE STUDY

An evaluation was conducted of the economic effects of varying plant sodium and steam temperatures and pressure in a 1000-Mwe reactor. This effort was confined to cost variations in the plant system and equipment excluding the reactor. The resulting data are therefore not sufficient to justify final selection of plant temperatures since, for example, cost variations in the fuel cycle caused by different temperature selections are not included.

A summary of the system parameters studied is shown in Table 5. Major equipment costs included in the study were as follows.

- 1) Steam generator (surface computed at \$68, \$102, and \$136 per ft²)
- 2) Intermediate heat exchanger (surface computed at \$57/ft²)
- 3) Primary and secondary sodium pumps
- 4) Primary and secondary sodium piping
- 5) Turbine-generator and main steam condenser
- 6) Steam piping
- 7) Feedwater heaters

Economic cost factors were as follows.

- 1) \$50,000 per 1% change in reactor thermal rating
- 2) 80% load factor
- 3) Use demand charge of \$75 per kilowatt
- 4) 12% capital charge rate
- 5) Fuel cycle costs of 6¢ and 12¢ per 10⁶ Btu

TABLE 5
1000-Mwe REACTOR PARAMETER STUDY
(System Temperatures °F)

Reactor Mixed Mean Sodium Outlet Temperature	1200	1150	1100	1050
Secondary Sodium Hot-Leg Temperature	1100	1050	1025	1000
Reactor (Primary) Sodium Delta T	250-350	250-350	250-350	250-350
Secondary Sodium Delta T	325-400	325-400	325-400	325-400
Main/Reheat Steam Temperature	1000/1000	950/950	950/950	900/900
	950/950	900/900	900/900	850/850
	900/900	850/850	850/850	850/850

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The turbine heat rate was computed for each steam condition selected to determine capitalized fuel cycle costs, and pumping power requirements were computed to determine demand charges. All of the studies completed to date were based upon a 2400-psig steam pressure. A typical schematic of the reactor flow arrangement is shown in Figure 3 and a typical cycle displayed on a temperature-enthalpy diagram is shown in Figure 4.

A summary of the resulting calculated optimum temperatures under the different assumed parametric conditions is shown in Table 6. It is noted from this table that optimum primary sodium ΔT is between 275 and 325°F, optimum secondary sodium ΔT is approximately 50°F higher than primary sodium ΔT . For 6¢ fuel cycle costs, optimum main/reheat steam temperatures are 900°F/900°F; and for 12¢ fuel cycle costs, main and reheat temperatures are 950°F/950°F.

TABLE 6
OPTIMUM SYSTEM TEMPERATURES

Reactor Outlet Temperature (°F)	1200	1150	1100	1050
Secondary Hot-Leg Temperature (°F)	1100	1050	1025	1000
Steam Generator Cost (\$/ft ²)	68 102 136	68 102 136	68 102 136	68 102 136
IHX Cost (\$/ft ²)	57.25	57.25	57.25	57.25
<u>Fuel Cycle Cost 6/10⁶ Btu</u>				
Best Primary ΔT (°F)	325 325 300	300 300 300	285 275 275	210 220 230
Best Secondary ΔT (°F)	400 375 350	350 350 325	350 325 325	350 310 325
Best Steam Temperature (°F)	950 950 900	950 900 900	900 900 900	900 900 850
<u>Fuel Cycle Cost 12¢/10⁶ Btu</u>				
Best Primary ΔT (°F)	350 325 300	300 300 300	275 275 275	200 210 220
Best Secondary ΔT (°F)	400 375 350	350 325 325	325 325 325	350 350 325
Best Steam Temperature (°F)	950 950 950	950 950 950	950 900 900	900 900 850

Table 7 shows the comparative cost differences for reactor outlet temperatures varying from 1050 to 1200°F at the parametric conditions noted. Although the greatest savings is shown for the 1200°F case for all parametric conditions shown, as noted previously, care should be exercised in using this table since reactor costs are not included. For example, the core structural costs for increasing temperature from 1150 to 1200°F will result in a debit to the 1200°F system. Additionally, the higher sodium temperature will result in higher fuel cladding temperature which could restrict burnup, thus increasing fuel cycle costs. The capitalized value of this increased fuel cost would also be a debit against the higher temperature system.

The steam generator costs shown in Table 7 vary from \$68/ft² to \$136/ft². Based upon a modular steam generator design, these values correspond to the estimated fabricated and installed costs respectively. Steam generator cost differences in going from 1100 to 1200°F were estimated to be negligible. It is possible that a materials change can be incorporated in the steam generator at 1050°F thus lowering the cost somewhat.

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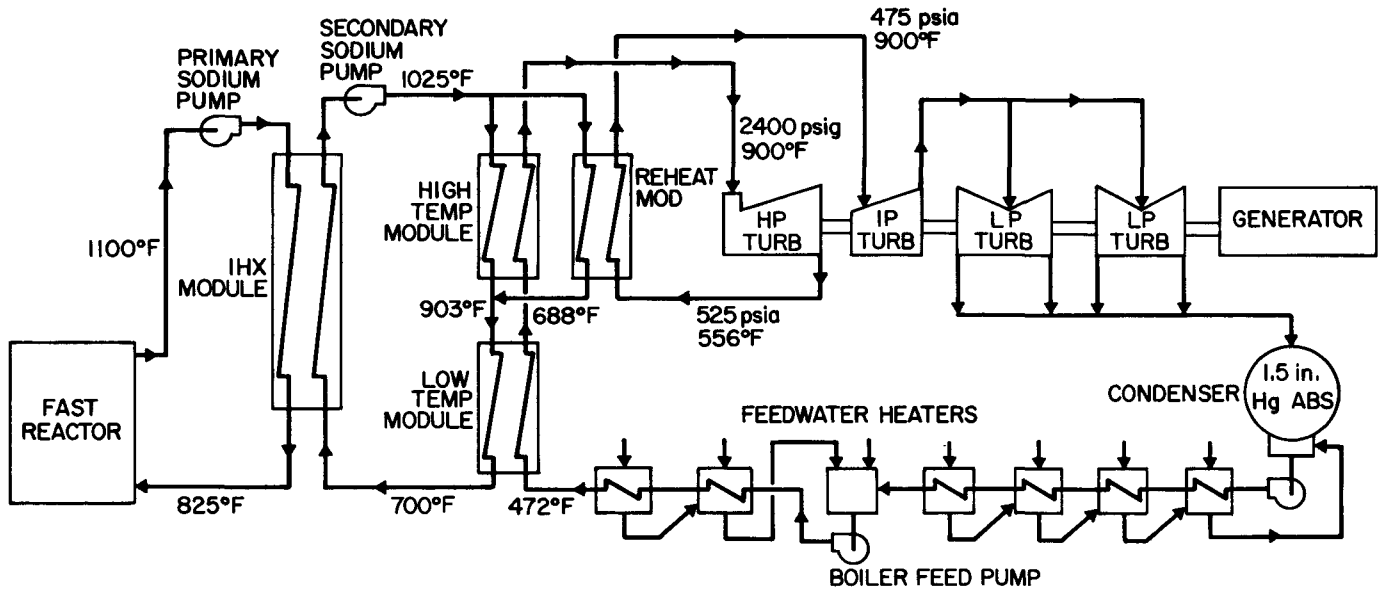


Figure 3. Typical Sodium-Cooled Fast-Reactor

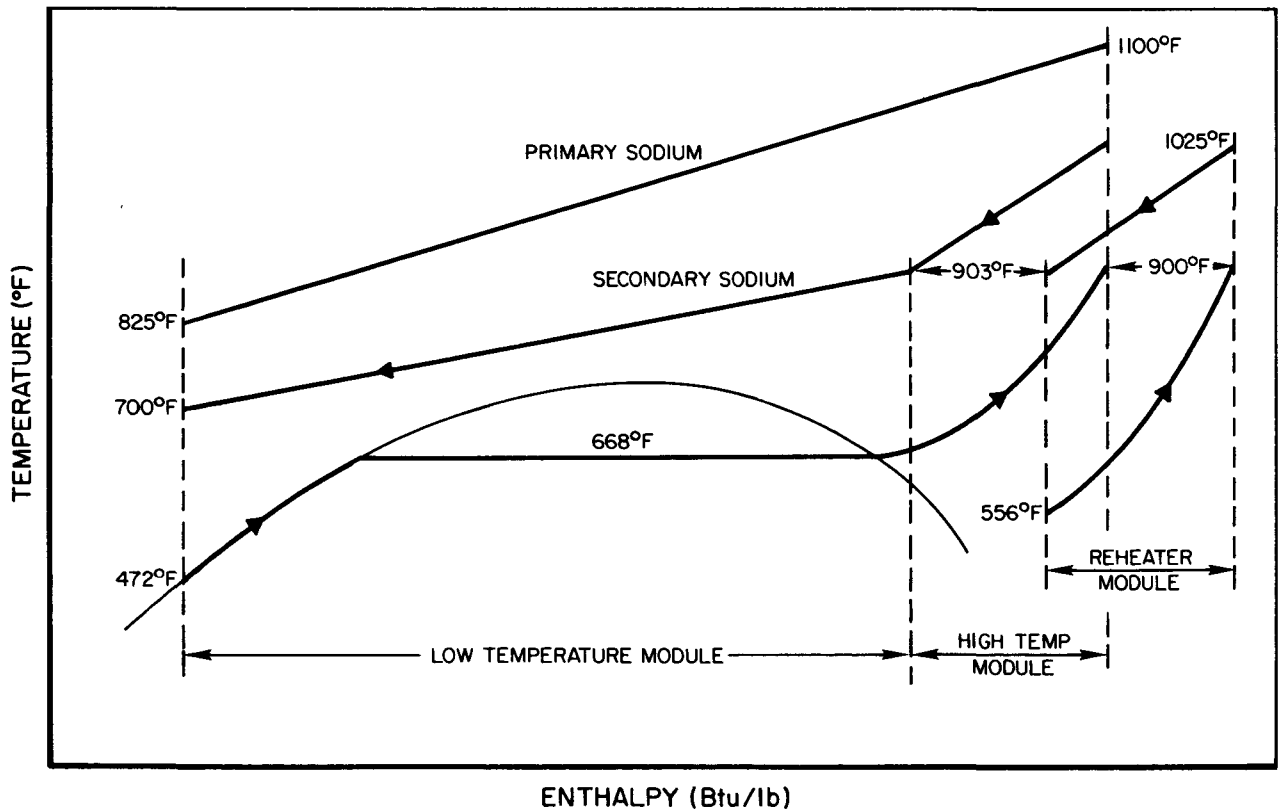


Figure 4. Sodium/Steam Parameters on a Temperature/Enthalpy Diagram

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TABLE 7
COST EFFECT OF REACTOR OUTLET TEMPERATURES

Fuel Cycle Cost	Reactor Mixed Mean Sodium Outlet Temperature (°F)	Potential Savings Over Base Case With Steam Generator Cost		Additional Costs Over Base Case With Steam Generator Cost	
		at \$68/ft ²	at \$136/ft ²	at \$68/ft ²	at \$136 ft ²
6¢ per 10 ⁶ Btu	1050	-	-	\$2.2 x 10 ⁶	\$3.4 x 10 ⁶
	1100	Base	Case	Base	Case
	1150	\$2.3 x 10 ⁶	\$3.2 x 10 ⁶	-	-
	1200	\$3.5 x 10 ⁶	\$4.9 x 10 ⁶	-	-
12¢ per 10 ⁶ Btu	1050	-	-	\$2.3 x 10 ⁶	\$4.0 x 10 ⁶
	1100	Base	Case	Base	Case
	1150	\$2.7 x 10 ⁶	\$3.3 x 10 ⁶	-	-
	1200	\$3.8 x 10 ⁶	\$5.2 x 10 ⁶	-	-

D. CONTAINMENT-ENERGY RELEASE EVALUATION

The magnitude of energy release of the maximum hypothetical meltdown accident is often computed by methods developed by Bethe-Tait;⁽²⁾ and modified by Jankus⁽³⁾, Brout⁽⁴⁾, Nicholson⁽⁵⁾, and Meyer-and-Wolfe.⁽⁶⁾ The percentage of the total energy produced in the excursion, which is available for mechanical work, is dependent on the properties of the core materials. The equations of state of the reference core materials; i. e., heat of fusion, heat of vaporization, and heat of sublimation, are derived from Hougen, Watson, and Ragatz.⁽⁷⁾ After the fuel is brought to the point of vaporization, the remaining energy is converted to an equivalent high-explosive yield using the relationship that 2 Mw-sec = 1 lb TNT. This quantity of free energy is used as the containment design loading.

A TNT explosion is a chemical reaction which has its effect by three distinct and separable phenomena, namely: the shock wave, the blast effect, and heating effect. The three phenomena are separable mainly because they occur at, and persist for, different times. The shock wave, a microsecond affair, carries and dissipates a fixed amount of the total available energy (~25%). It is essentially a step function in pressure which decays with a microsecond time constant and proceeds spherically through the media surrounding the source of the explosion at several times the speed of sound. Its destructiveness is large because it dissipates its energy by giving an almost instantaneous acceleration to everything in its path.

The blast effect, a quasi-static gas pressure, results from the kinetic energy of the gaseous explosion products. It also consumes a finite amount of the initial energy, the magnitude of which, however, has as yet not been determined accurately. The blast effect does mechanical work on the surrounding medium and would in a reactor vessel; and, since it is a quasi-static effect, the degree of physical damage resulting from this source is usually much larger than that from the preceding shock wave, in spite of the fact that the blast peak-pressures are several decades smaller than shock pressures. The third factor is the heat-

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ing effect, or heat losses to all media in contact with, and surrounding the explosion products. This effect dissipates the remaining portion of the released energy by radiation and convection; and, although this is the largest share of the energy (between 50 and 60%), the overall effect is only a small increase in temperatures because of the large inherent heat capacities.

Figure 5 shows a schematic representation of the progression of an explosion in a reactor. This explosion flow-diagram also schematically represents a digital computer code developed as a part of a company-sponsored effort to study explosion effects on containment. The code calculates: the shock wave energies at various distances from the explosion center; the reactor vessel deformation at various points, computing the total increase in vessel volume; and, if the shock energies are larger than required to rupture the wall at any point, it calculates the kinetic energies of the resulting missile fragments. In the upper region of the vessel, the code calculates the kinetic energy of the sodium spray which is propelled by the shock wave towards the top shield; after comparing this with the energy requirements to shear off the shield support, it determines the distance the shield would be lifted by this effect alone.

The blast effect is formulated in a similar manner. First the maximum pressure is calculated by using the original vessel volume plus the deformation volume increase from the shock wave. Then, by an iterative process which is based on the clausius inequality principle, the masses of fuel, steel, and sodium which are vaporized in the explosion are calculated and the temperature of the mixture is determined.

The mechanical work done by the explosion gases is computed as a function of time. The time rate of expansion of the product gases into the reactor cavity is calculated (if the primary containment is breached by the shock or blast effect), as is the gas flow past the moving top shield (again only if the explosion is large enough to lift the shield). Finally, pressures, temperatures, and maximum lift of the shield are determined.

To determine the effect of energy release in hypothetical accidents on containment requirements, this code is being applied to typical reactor vessel arrangements. Energy release is an independent parameter, and its effect on different geometric arrangements is being evaluated. The adequacy of alternative containment schemes to meet varying levels of energy release is also being evaluated.

IV. EVALUATION OF EFFORT TO DATE

Effort completed to date demonstrates the usefulness of parametric studies in defining reactor- and plant-design criteria, and in setting development program objectives and direction.

The results of fuel cycle studies demonstrate that there are definite safety-economic interrelationships which can be determined quantitatively for the different fuel types being considered. Furthermore, the relative economic potential resulting from the selection of different fuel design, fabrication, and operating parametric conditions can be determined. The sodium-pressurization study demonstrates that sodium pressurization does not appear to result in an improved

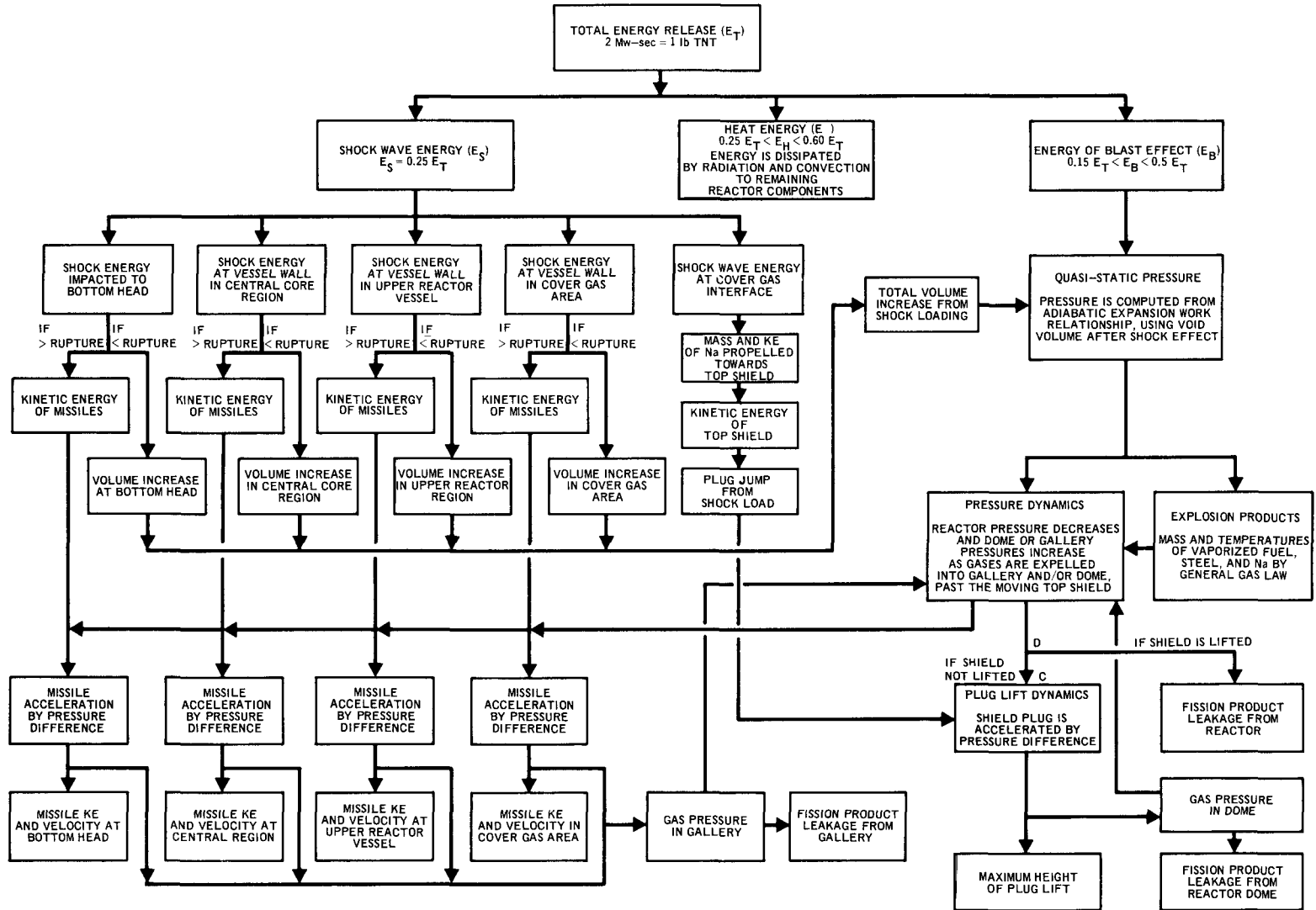


Figure 5. Flow Path of the TNT Computer Code

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safety situation. The system temperature-pressure study indicates an economic trend towards higher system temperatures and pressures if the reactor effects are ignored. These effects should be evaluated.

V. NEXT REPORT PERIOD ACTIVITIES

This project was terminated.

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Program: Fast Reactor Development

Project: Calculation Methods Development

Project Engineer: H. A. Morewitz

Reporting Period: July-September 1965

AEC Category: 04-01-04-19

General Order: 7661

Subaccount: 1331

I. PROJECT OBJECTIVES

The general objective of this project is to develop reactor physics calculational techniques and codes for reactivity, neutron economy, and critical mass for sodium-cooled fast reactors that are efficient, reliable, and accurate. Specific objectives include improvement in the methods for calculating multi-group cross-sections for use in multigroup codes, and the improvement of various features of the codes themselves. Special emphasis is placed on the treatment of resonance capture, inelastic and anisotropic scattering, and neutron leakage, emphasizing the neutron spectrum and the cross-sections in the kilovolt energy region and above. The influence of various reactor characteristics on the energy release from core meltdown accidents will be evaluated. Improvement will be sought in the calculation of reactivity coefficients, and in the determination of their dependence on core burnup. Machine codes and neutron cross-section libraries for making fast-reactor neutronic calculations will be improved. Comparison will be made between calculated results and experimental measurements.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

Checkout was finished on most of the features of CAESAR-IV which were contained in the FORTRAN-II version. Coding and checkout continues on improvements in the edits and code options. A preliminary version of the code was placed in production status.

Work is proceeding on the GRAVE-II (GRISM) code, and successful runs were made with the GRAVE-TRIXIE combination.

The SIZDEK-II code was finished.

Analysis of reactivities and central fission ratios have been made for 4 ZPR-III fast critical assemblies. Results indicate that our fission cross-sections for U^{235} and Pu^{239} may still be about 2% too high at energies above a few kilovolts. The values for U^{233} appear to be about 10% high.

An evaluation of dilute resonance integrals and effective cross-sections for fissionable isotopes calculated using single level resonance theory was started. A literature survey on the effects of resonance overlap on group cross-sections and the Doppler coefficient was finished.

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III. PROGRESS DURING REPORT PERIOD

A. CAESAR-IV

All the features which were contained in the FORTRAN-II version of the multigroup diffusion theory CAESAR code were debugged except for the punched card output options which cannot yet be included in CAESAR-IV because of a current shortcoming in the computing system. A number of improvements were made in the CAESAR-IV output edits, and a new 5-element concentration search was added. Work was started on an option to interpolate resonance cross-sections from a table on each iteration of concentration searches. Additional improvements in the code options was planned. A preliminary version of the code was placed in production status so that wider experience in its use may be obtained. This version will be updated as additional improvements are made in the code options and edits.

B. GRAVE-II (GRISM)

In cooperation with the cross-section analysis and application project (7604-1310), work proceeded on combining the GRAVE, PRISM, TRIXIE, and CONTRACT codes into a single code for the production of multigroup cross-section libraries. Coding was finished on the combination of GRAVE and TRIXIE, and successful test runs were made to produce group-averaged cross-sections for resonance isotopes. Debugging of the GRAVE and PRISM combination is in progress. Certain problems with the neutron spectrum generation techniques in these codes remain to be resolved. The inclusion of CONTRACT, which properly formats the various output libraries, remains to be done.

C. SIZDEK-II

SIZDEK-II is a code for constructing punched card cross-section libraries for the SIZZLE, AIM-6, BIP, ELMOE, and DTF codes from a FAIM or CAESAR-II library. Originally devised to automate the construction of special purpose SIZZLE libraries, the code has now been extended to include the other libraries as well. This makes it relatively easy to extend or refine calculations made with FAIM or CAESAR-II (the codes used at AI for making most of the survey calculations) by running any of the other codes with an identical set of microscopic cross-sections. SIZDEK-II is the only code currently available for linking the libraries of these codes.

D. ZPR-III CALCULATIONS

Revised 35-group cross-section libraries were constructed for ZPR-III critical assemblies 29, 31, 34, and 36 which had previously been selected as a part of the cross-section testing program. Libraries for all 4 assemblies were prepared by using the revised U^{235} cross-sections based on data recently obtained from Aldermaston and resonance cross-sections generated with the TRIX code. The neutron spectra for cross-section weighting for each assembly was calculated with ELMOE. The transport cross-sections were weighted directly with the current spectrum and other cross-sections with the flux spectrum except that the TRIX resonance cross-sections were not weighted with the ELMOE spectrum. However, the correction to the group lethargy width to account for

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flux depression by the resonances of the absorbing isotope was included for the resolved resonances. The highest resolved resonance for U²³⁸ was at 3,904 ev; that for U²³⁵ was 61 ev.

Criticality calculations were made for the above assemblies with the CAESAR-IV code. The calculated value of k_{eff} for each assembly should be unity in order to agree with experiment. Results are presented in Table 1, along with results reported by other laboratories, for comparison.

TABLE 1
ZPR-III REACTIVITY CALCULATIONS

Assembly No.	CAESAR	Davey ^(a)	Meneghetti ^(b)	Yiftah ^(c) (1964 set)	Yiftah ^(c) (ANL 26-group)
29	1.0310	1.003	0.979	0.9951	0.971
31	1.026	1.006	-	0.9947	-
34	1.027	0.997	0.982	1.001	0.977
36	1.006	0.994	0.996	-	-

(a) W. G. Davey, NSE 19, 259 (1964).

(b) D. Meneghetti and J. R. White, ANL-7010, page 110, (1964).

(c) S. Yiftah and M. Sieger, IA-980, page 16 (1964).

The CAESAR calculations are based on diffusion theory; the others S-4 on transport theory. An S-4 calculation for assembly 29 was made with the DTF code using the same spectrum for weighting the DTF cross-sections as with CAESAR. The S-4 result was $k_{eff} = 1.0286$, a reduction of 0.0024 in k_{eff} , showing that diffusion theory introduces very little error for these assemblies.

The transport correction to the diffusion coefficient, given by

$$\frac{1}{D} = 3 \Sigma_{tr} \left[1 - \frac{2 \Sigma_a}{5 \Sigma_{tr}} \right]^2$$

was also tried for assembly 29. The result showed a reduction in k_{eff} by the amount 0.0071 as compared with the value obtained with the option $D = 1/3 \Sigma_{tr}$.

The neutron spectra obtained with CAESAR were somewhat softer than those reported by Davey and also showed some structure that he did not find. Davey observes that his spectra are too hard. The reason for the CAESAR spectra showing structure that Davey's calculations did not obtain can be attributed in part to the fact that (1) Davey used a single ELMOE fine group spectrum to weight the coarse group cross-sections for all assemblies while we calculated an ELMOE spectrum for each assembly, and (2) the AI light element cross-sections show a few minor differences from the corresponding ELMOE cross-sections. A group-by-group comparison of coarse group cross-sections for these assemblies generated by ELMOE and by GRAVE-PRISM show a few

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significant differences. These differences appear to be explainable in terms of small displacements in the locations and magnitudes of resonances in light elements.

The neutron spectra for these ZPR-III assemblies show a peak near 400 kev (group 7 in the 35-group scheme). The two softest spectra (Assemblies 29 and 34) have dropped by a factor of 1000 in group 21 which has a lower energy of 275 ev. Contributions below this energy are negligible.

Measurement of spectral indices with various fission counters was reported in Davey's article. The reported measurements were all relative to the Pu-239 fission rate. The experimental and CAESAR-calculated results for these fission ratios are compared in Table 2. It is evident that agreement with experiment is good in most cases although a few instances appear where a difference of about 10% is found.

TABLE 2
CENTRAL FISSION RATIOS RELATIVE TO Pu²³⁹ FISSION

Isotope	Assembly No.							
	29		31		34		36	
	Calc	Exp	Calc	Exp	Calc	Exp	Calc	Exp
U ²³³	1.438	1.39	1.335	1.29	1.430	1.36	1.320	1.23
U ²³⁴	0.230	0.254	0.288	0.294	0.234	0.240	0.277	0.273
U ²³⁵	0.907	0.943	0.843	0.847	0.902	0.935	0.835	0.840
U ²³⁶	0.070	0.082	0.086	0.095	0.072	0.075	0.079	0.084
U ²³⁸	0.035	0.036	0.043	0.040	0.035	0.034	0.039	0.037
Pu ²⁴⁰	0.255	0.283	0.318	0.276	0.261	0.264	0.300	0.294

The central fission ratios shown in Table 2 can be used to form corresponding fission ratios relative to each of the seven isotopes involved in the measurements. Examination of the data expressed in this manner indicates that the U²³⁵ fission cross-section is still about 2% too high, which is supported by the k_{eff} values shown in Table 1. It is also found that the fission cross-sections for U²³³ appear to be about 10% high, that for U²³⁶ about 5% low, and that for Pu²³⁹ possibly about 3% high. Values for U²³⁸ and U²³⁴ fission cross-sections appear to be fairly good although the evidence regarding Pu²⁴⁰ is inconclusive.

E. RESONANCE CROSS-SECTIONS

In cooperation with the cross-section analysis and application project, the procedures for adding new isotopes to the ARES resonance parameter library in the GRAVE-I code and the TRIXIE resonance parameter library in the GRAVE-II (GRISM) code were reviewed in preparation for the addition of new data to these libraries.

The problem of calculating realistic dilute resonance integrals and effective group cross-sections for fissionable isotopes using single level resonance theory

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is being studied. It is proposed to apply an empirical correction to the single level results by adding a suitable smooth background capture and fission cross-section which produces agreement with measured values of dilute resonance integrals and cross-sections. The current analysis involves the splitting of the resonance region into several energy groups for which resonance integrals calculated with the TRIX code and obtained from studies at Karlsruhe* are compared with integrals over the measured fission and capture cross-sections. An extensive analysis for U²³⁵ is nearing completion and some of the results obtained to date are presented in Tables 3 and 4.

TABLE 3
 CALCULATED FISSION AND CAPTURE RESONANCE
 INTEGRALS FOR U²³⁵

Energy Range	Fission		Capture	
	TRIX (barns)	Karlsruhe (barns)	TRIX (barns)	Karlsruhe (barns)
0.5 to 10 ⁵	238.7	265.7	185.3	169.4
1.0 to 10 ⁵	199.0	223.2	178.4	163.0
60.0 to 10 ⁵	66.5	67.6	38.6	32.6
0.5 to 10 ⁵	241.9	268.7	185.9	171.4

TABLE 4
 MEASURED FISSION AND CAPTURE RESONANCE INTEGRALS FOR U²³⁵
 (Cutoff Energy is 0.5 ev)

Experimenter	Reference	RI _{fis} (barns)	RI _{cap} (barns)
E. Hellstrand	AE-181 (1965)	278 ± 9	-
J. Hardy et al.	NSE <u>9</u> , 341 (1961)	274 ± 11	-
Baumann	DP-817 (1963)	276 ± 9	-
E. D. Clayton	AECD-4167 (1955)	269 ± 25	-
C. B. Bigham	EANDC-211 (1964)	263 ± 8	-
F. Feiner et al.	Trans. ANS <u>7</u> , 78 (1964)	292 ± 18	140 ± 10
H. D. Conway et al.	Trans. ANS <u>7</u> , 78 (1964)	288 ± 18	158 ± 10

The sensitivity of the calculated dilute resonance integral of Pu²³⁹ to the cadmium cutoff energy (about 0.5 ev) was investigated. The large Pu²³⁹ resonance at 0.3 ev makes a large contribution to the resonance integral, and the value calculated is sensitive to the choice of the cutoff energy. Since the

*J. J. Schmidt, AEC-ENEA Seminar on the Evaluation of Neutron Cross-Section Data at Brookhaven, N. Y. (May 1965)

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Doppler width for this resonance is small compared with its natural width, the use of the natural line shape to calculate the dependence of the resonance integral on the cutoff energy introduces only a few percent error in the result. By using the natural line shape, the contribution of the 0.3 ev resonance to the fission resonance integral over the energy interval from 0.45 ev to 0.50 ev was found to be 44 barns. (The corresponding quantity for U^{235} is given by J. J. Schmidt of Karlsruhe as 9.4 barns.) The experimental value for the Pu^{239} fission resonance integral above a cutoff energy of 0.5 ev is quoted as about 327 ± 22 barns. It is evident that comparison between the experimental and calculated resonance integral values for Pu^{239} is strongly dependent on careful evaluation of the cutoff energy of the experiment. In particular; use of the experimental resonance integral value to assess the accuracy of the calculated contribution of resonances located above the cutoff energy appears to be difficult. The use of high resolution measurements of point values of the cross-section may be more reliable for this purpose unless a very accurate determination of the effective cutoff energy of the resonance experiment can be made.

The investigation of resonance overlap and its effect on the formation of flux weighted multigroup cross-sections and on the Doppler coefficient was continued. A review of the literature was finished and a memorandum prepared summarizing the results and conclusions.

Of the documents examined, the one by Graves* is the only one which gives material in a form immediately usable to calculate cross-sections with overlap effects included. Various graphs giving data for calculating correction factors for overlap and statistical distributions are presented. The results are based on analytic approximations to the effective resonance integral rather than on numerical methods for solving the neutron slowing down equation.

Sample calculations for a typical $U^{238} - Pu^{239}$ mixture were made by using Graves' method. The results indicate that the effective cross-sections and the Doppler reactivity coefficients are somewhat dependent on corrections for level width distributions, level spacing distribution, correcting the potential scattering cross-section to the average total cross-section, and dividing the effective resonance integral by the actual group integrated flux rather than a $1/E$ flux. The significant corrections usually range from about 5 to 15%. It also appears that the assumption of isolated resonances is a reasonably good approximation for calculating these correction factors for a wide range of fast reactor cores. However, obtaining highly accurate results will require further development in the methods for taking resonance overlap effects into account.

F. ARGONNE CROSS-SECTION COMPARISON STUDY

A report, NAA-SR-TDR-11517, describing the methods used and giving the results of the analysis of the fast-reactor assemblies for the Argonne comparison study was written. The study analyzed most of the fast-reactor configurations suggested by ANL. The analysis was based on 15-group diffusion theory calculations having an energy structure the same as that used in calculations for the split-blanket modular concept.†

*C. C. Graves, "General Methods for the Evaluation of Effective Cross-Sections Used in Doppler Effect Calculations for Fast Reactors," UNC-5064 (May 1964)

†D. T. Eggen et al, "Feasibility Study of a Sodium-Cooled Coupled Reactor," NAA-SR-11378

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In order to compare the results of 15-group and 35-group calculations, assembly A-3 was selected for further analysis. The ELMOE spectrum was used in GRAVE and PRISM to produce 15-group and 35-group libraries for this assembly. Self-shielded U²³⁸, and Pu²³⁹, and Pu²⁴⁰ cross-sections were generated for each library by the TRIX code. Work on this study has been suspended for the present.

G. FAST REACTOR SAFETY

Safety analyses completed for the sodium-cooled fast-reactor feasibility study* are being reviewed to determine those areas in which improved analytical methods are wanted. The reactor kinetics and nuclear transients are complicated by the interaction between modules of the core. When methods of evaluating these interactions are developed, the reactor behavior will be calculated for transients caused by disturbances in one or several modules. The previous treatment of delayed neutrons can be improved by considering more delayed groups.

Details of the maximum hypothetical accident may be modified as information on fuel meltdown in sodium is obtained from the transient heating studies being conducted in TREAT by AI. Specific mechanisms which lead to the explosive energy release of the maximum hypothetical accident will be evaluated to assure that they provide realistic estimates of the consequences of this accident.

IV. EVALUATION OF EFFORT TO DATE

The need for improved neutron cross-section libraries for fast-reactor calculations is being met. Improvement is being made in the tools for calculating fast-neutron spectra and Doppler coefficients. These improvements will contribute toward the reliability of calculated reactivity coefficients. The mechanics of producing multigroup, spectrum-weighted, cross-section libraries is being streamlined. When completed, this will result in a considerable reduction in the time and labor involved in making neutronic calculations.

The analysis of critical assemblies, fission ratios, and resonance integral data is uncovering certain areas where improved basic cross-section data is needed. Much remains to be done in this area, along with acquiring and evaluating new measurements of cross-sections and resonance parameters.

V. NEXT REPORT PERIOD ACTIVITIES

The CAESAR-IV code will be finished. Analysis of the ZPR-III critical assemblies will be continued, including the calculation of neutron generation time and central reactivity coefficients. Evaluation of cross-section data for fissionable isotopes will be continued.

This project was terminated. The activities may be continued under the Reactor Physics Program.

*loc. cit.

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Program: Fast Reactor Development	
Project: Physics Experiments	
Project Engineer: H. A. Morewitz	
Reporting Period: July-September 1965	AEC Category: 04-01-04-19
General Order: 7661	Subaccount: 1332

I. PROJECT OBJECTIVES

The general objectives of this project are to supply information pertinent to the safety and economic studies of fast-reactor systems and to confirm calculational methods and parameters used in preliminary design. The specific objectives of this project include the study of Doppler effects for specific fuel geometries, sodium void effects, and the interaction between the two in reactor spectra resulting from core compositions of interest. Because limited information is available on Doppler coefficients for specific fuel geometries, considerable effort must be spent in experimental studies. Information on sodium void effects will be obtained including the interaction in fast spectra of the Doppler and sodium void effects. Physics measurements will be made for a variety of materials, composition, and spectra utilized in fast-reactors, particularly sodium because of its large volumetric fraction in fast-reactor cores. The spectral effects and energy and spatial distributions resulting from the use of these materials will be determined and compared with theoretical predictions. Liaison with other laboratories will continue so that a rapid and useful exchange of information can be maintained.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

Calculations have been made for loadings of the ECEL core required to perform measurements of the sodium void coefficient. The test region of the core contained 50 vol % sodium, 15 vol % stainless steel, and 25 vol % uranium with a U²³⁸ to U²³⁵ atom ratio of 8:1, and 10% nickel and aluminum cladding and/or structure. Calculations were made for full sodium voiding and for 50% sodium voiding. These indicated that the reactivity changes expected could be compensated for by rod positioning alone.

III. PROGRESS DURING REPORT PERIOD

The initial calculations for the sodium void measurements were made, using CAESAR II, with a cross-section set equivalent to that used in calculating the present ECEL core so that a direct comparison could be made. For a test region of 30.18 cm radius and a driver increased over the present core by 1 cm, the following values of k_{eff} were obtained:

	<u>k_{eff}</u>
Sodium present (50 vol %)	0.9915
Sodium 1/2 voided	0.9938
Sodium fully voided	0.9960

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A second round of calculations used an improved cross-section set with the absorption cross sections of all the fertile and fissile elements (U^{238} , Th^{232} , and U^{235}) and the fission cross-section of U^{235} shielded in the resonance regions (groups 6-14 for 18 group calculations) using the TRIX code. The cross sections were determined by using the narrow resonance approximation. The interference between resonance and potential scattering was excluded in U^{235} and included in U^{238} . The unresolved resonance contributions were found using the narrow resonance approximation for the $l = 0$ and $l = 1$ neutrons excluding interference.

Values of k_{eff} using this latter cross-section set are given below for the following cases: (A) identical-core to previous-calculation, and (B) driver region is increased only 0.6 cm over presently loaded reactor.

	k_{eff}	
	(A)	(B)
Sodium present (50 vol %)	1.0278	1.0036
Sodium 1/2 voided	1.0286	1.0055
Sodium fully voided	1.0303	1.0075

Stainless steel boxes for sodium containment are now being produced. The boxes are 2 in. by 2 in. by 1/2 in. with 15-mil walls and are being deep drawn to eliminate welding of bottoms on the 5000 boxes. The initial shipment of the boxes was found to be out of tolerance and are now being reworked.

IV. EVALUATION OF EFFORT TO DATE

Calculations indicate that the sodium void measurements in the present test region can be made with a minimum modification to the present driver and buffer region of the ECEL critical assembly.

Some delay has been experienced in preparation of the canned sodium due to rework required on the stainless steel boxes. It is apparent that a deep draw box of the required wall thickness is a more complex problem than the vendor had anticipated.

V. NEXT REPORT PERIOD ACTIVITIES

An improved set of calculations will be made by using the 35 groups allowed in CAESAR IV. The 35-group cross-section set will be generated by use of the TRIX, ELMOE, and GRAVE-PRISM series of codes. The stainless steel cans will be procured and filled with sodium.

This project was terminated. The activities may be continued under the Reactor Physics Program.

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Program: Fast Reactor Development	
Project: Boiling Studies for Sodium Reactor Safety	
Project Engineer: A. A. Jarrett	
Reporting Period: July-September 1965	AEC Category: 04-01-04-19
General Order: 7661	Subaccount: 1341

I. PROJECT OBJECTIVES

The general objective of this project is to develop information on two-phase flow required in the safety evaluation of sodium-cooled reactors. This information is important because of the key role that sodium boiling plays in reactor dynamics, in fuel meltdown accidents, and in the ultimate shutdown mechanism of the reactor. Specific experimental objectives include the measurement of boiling heat transfer characteristics, critical heat flux, two-phase pressure drop, void fractions, transient voids and pressures, and the study of hydrodynamic instabilities. Specific objectives of the theoretical study, in addition to any analyses required to support the experimental work, are the development of a digital computer code which will predict transient void fractions, flow rate, and heat transfer for single and multichannel sodium flow, and the incorporation of this code into a general reactor kinetics code.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

A topical report "Steady State Sodium Boiling and Hydrodynamics," NAA-SR-11586, has been completed.

III. PROGRESS DURING REPORT PERIOD

A new high-temperature heater for the 6-ft test section was designed, built, and installed in the two-phase sodium loop. Repeated difficulties with the previously installed graphite heater necessitated this change. The loop had been redesigned and modified in preparation for the study of hydrodynamic instabilities and transient flow due to boiling. The new loop contains the 6-ft annular (1/4-in. by 1/2-in.) test section uniformly heated along its length. Valves at the inlet and outlet of the test section enable simulation of variable orifices, and a surge tank in parallel with the test section permits boiling instabilities and transients to be induced.

The original design called for heat to be generated along 4-1/2 ft of the test section (maximum heat flux of 200,000 Btu/hr-ft²) by using graphite cloth as a resistance heater insulated from the test section by a thin sprayed-on layer of aluminum oxide. Repeated difficulty with this type heater, however, necessitated the above mentioned change. The new heater is a clam-shell type with nichrome heating elements to be used in the low-power, low-temperature range. To obtain the maximum heat fluxes required, a high-temperature heater is needed and tantalum heating elements will be used. To prevent oxidation of the heating elements at these high temperatures, especially with tantalum, a pure-argon purge will be maintained on the furnace at all times.

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The heat loss calibrations have been repeated and tests for two-phase pressure drop with uniform heating along the test section have been initiated.

In addition to the experimental work covered above, the second phase of the digital computer code TRANS-FUGUE has been initiated. In the first phase of the code, TRANS-FUGUE-I,* the classical equations of continuity, momentum, and energy were solved for two-phase flow by using the momentum integral technique and solving the equations sequentially; i. e., the equations were solved in sequence for each time interval.

In the new method of solution, TRANS-FUGUE-II, the momentum integral model is replaced by a sectionalized compressible model. The basic equations are first converted to explicit relations for the time derivatives of the enthalpy, pressure, and mass flow rate, and these equations are then solved simultaneously. This solution is expected to give greater numerical stability than TRANS-FUGUE-I in the type of transients studied in fast-reactor safety analyses. To enable larger time increments to be used and maintain the numerical stability, higher order integrations will be incorporated into the code to replace the simpler numerical techniques of the previous code.

The topical report "Steady State Sodium Boiling and Hydrodynamics," NAA-SR-11586 was completed and is being prepared for publication. This report is a final comprehensive summary of the steady state heat transfer and hydrodynamics studies of sodium under forced convection flow.

A paper, "The Development and Growth of In-Core Voids Due to Boiling During Fast Reactor Transients" was prepared for the International Conference on Safety, Fuels, and Core Design in Large Fast Power Reactors. A second paper, "Sodium Boiling Heat Transfer and Hydrodynamics" was presented at the Conference on Application of High Temperature Instrumentation to Liquid-Metal Experiments held at Argonne National Laboratory. An abstract of a third paper "Sodium Boiling Heat Transfer," was accepted by the Conference Committee of the Third International Heat Transfer Conference, Chicago, August 1966.

IV. EVALUATION OF EFFORT TO DATE

Progress on the experimental phase of this project has not been as rapid as anticipated due to the heater problems outlined above. The difficulties are being overcome, however, and operation of the loop and the experiments is imminent.

On the other hand, considerable progress has been made on the digital code TRANS-FUGUE. It is anticipated that the new method of solution of the equations of motion will greatly improve the stability of the calculation and the accuracy required for fast-reactor safety analyses.

V. NEXT REPORT PERIOD ACTIVITIES

The study of the two-phase steady-state pressure drop with uniform heating along the test section will be completed. These results are required for the analysis of the transient flow experiments. Following these tests, the "check curves," or the inception of instability, will be determined for the annular test section under study. Development of TRANS-FUGUE-II will continue.

*R. C. Noyes, J. G. Morgan, and H. H. Cappel, "TRANS-FUGUE-I" A Digital Code for Transient Two-Phase Flow and Heat Transfer, NAA-SR-11008, July 1965

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Fast Reactor Development	
Project: Fuel Element Transient Heating and Accident Dynamics	
Project Engineer: A. A. Jarrett	
Reporting Period: July-September	AEC Category: 04-01-04-19
General Order: 7661	Subaccount: 1343

I. PROJECT OBJECTIVES

The objective of this project is to experimentally examine the events which occur under various conditions of nuclear transient heating of typical sodium-cooled, fast reactor fuel element configurations, and from such studies to develop the foundation upon which to base the understanding of fast-core behavior during power transients. Emphasis will be placed on the characterization and generalization of the phenomenon of fuel dispersion noted during a prior TREAT meltdown experiment.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

Planning for Phase-III experiments and capsule design have been completed. Modifications to the analog computer program for analysis of the transient thermal behavior of the TREAT capsules have been extended to include phase change effects which played an important role in the Phase-II tests. These improvements assist considerably in understanding the course of events during the severe transients leading to fuel dispersion observed in Phase II.*

III. PROGRESS DURING REPORT PERIOD

A. PHASE-III EXPERIMENT PLANNING

Planning of the Phase-III experiments was initiated and the design of the Phase-III capsules was completed. Fabrication of components for mocking-up certain complex phases of the capsule assembly was initiated and purchase orders for long-lead time components were placed.

A drawing of the Phase-III capsule is shown in Figure 1. It consists of a 5/16-inch diameter fuel pin which is 12 inches long. The pin is surrounded by stagnant coolant and a graphite heat sink and crucible. Because of the considerable importance to the fast-reactor safety program in the dispersion of fuel noted in the Phase-II meltdown of a sodium bonded UC fuel rod, the Phase-III capsules will be used to examine the behavior of unbonded (gas bonded), sintered uranium carbide and uranium oxide under similar conditions. These capsules will be even more highly instrumented than the Phase-II capsules in spite of the smaller diameter pin which is being examined. The additional effort for extensive instrumentation is considered worthwhile on the basis of experience with preceding capsules as noted in the following analysis.

*See FY 1965 Annual Technical Progress Report, NAA-SR-11450.

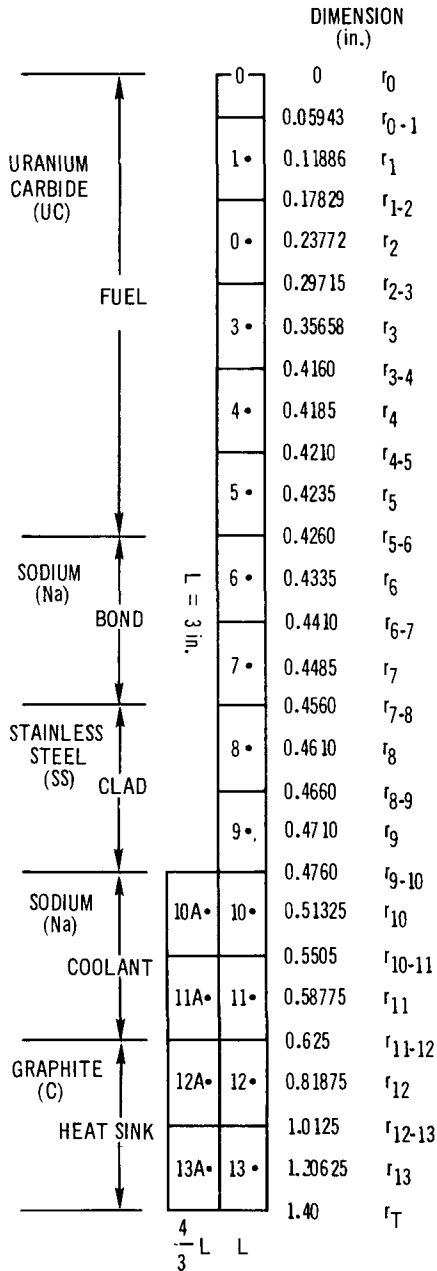
ATOMICS INTERNATIONAL

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Energy release calculations (capsule energy relative to TREAT) for several fuel enrichments have been made for the Phase-III capsule design. A final choice of the required enrichment must await completion of the thermal analysis currently in progress.

B. ANALYTICAL DEVELOPMENT

The analog computer model which was being used for analysis of the thermal

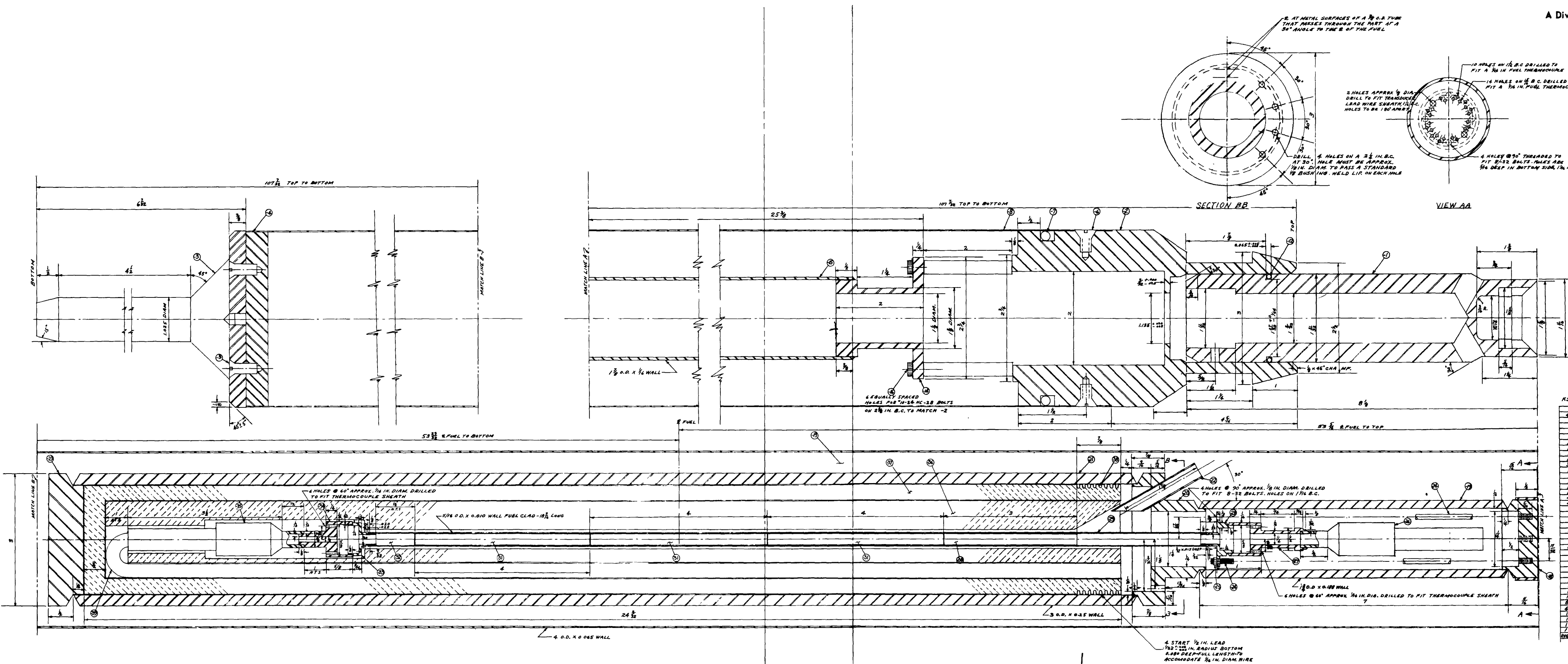


behavior of the TREAT capsules included provisions for latent heat of fuel melting and adjustments in bond conductivity on boiling. However, it was noted in Transient 4 (Phase II) that bond boiling was accompanied by very constant bond temperature which was not simulated by the thermal model as originally devised. Consequently, adjustments were made to the conductivity and heat capacity of the bond to permit the boiling simulation. Figure 2 indicates the node positions used in Figures 3 through 6. Figure 3 shows the results with the improved model; the bond property changes were made primarily within the region between Nodes 6 and 7, so that these temperatures show the temperature difference across the entire bond region rather than between the midpoints of the region. Comparison of these results with those of the old model (Figure 4) shows that in the new model the central and midradius temperatures are brought more closely together; the bond temperature (Node 7) levels off; the coolant temperature (Node 9) reaches about 1500°F; and the fuel temperatures drops more slowly and reaching 2500°F in about 35 seconds. In all of these cases, the new model agrees more closely with the experimental data.

The application of the new model to Transient 2 (Phase II) showed (Figure 5) that melting of the edge of the fuel was likely to occur at about the time the TREAT burst terminated (t = 1 to 1.5 seconds). Since this still left some uncertainty regarding the cause of the delay before cladding failure, provisions were made to revert back to the original thermal properties at one second in order to simulate the collapse of the fuel into the cladding, thus overcoming the large resistance of the vapor gap.

7661-2527

Figure 2. Node Positions For Figures 3 Through 6

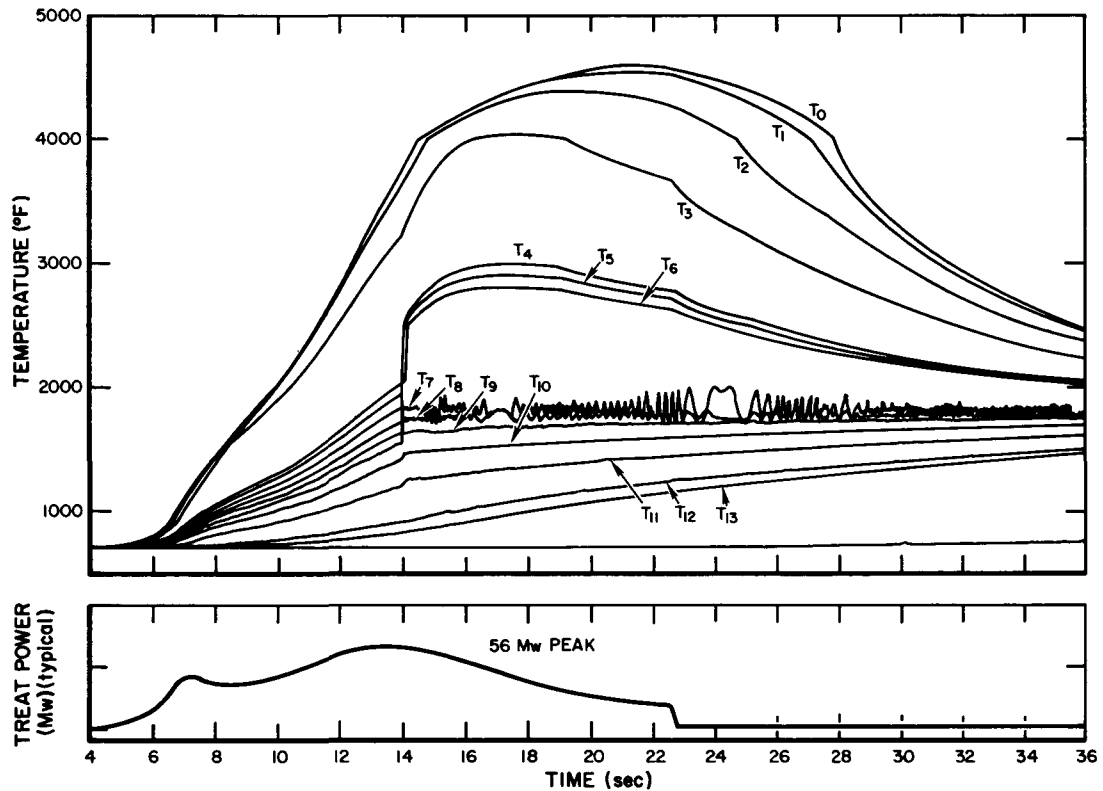


RS. - FUEL SECTION IN CAR - INNER CAPSULE GRAPH - GRAPHITE

ITEM NO.	DESCRIPTION	MATERIAL	SIZE
1	40 GUSSETS		
2	38 ENCL. LEAD TUBE	S.S. TYPE 304	1/4 O.D. X .001 WALL
3	38 HEATERS		
4	37 GRAPHITIC CARBIDE GRAPHITE AGENT		
5	36 GRAPHITIC CARBIDE GRAPHITE AGENT		
6	35 PREL. TRANS. LUMP		
7	34 FUEL		
8	33 FUEL		
9	32 TOP GRAPH. ROD	GRAPHITE AGENT	
10	31 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
11	30 TOP CAP	S.S. TYPE 304	
12	29 TOP CAP	S.S. TYPE 304	
13	28 PREL. TRANS. LUMP		
14	27 PREL. TRANS. LUMP		
15	26 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
16	25 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
17	24 LOWER CAP	S.S. TYPE 304	
18	23 LOWER CAP	S.S. TYPE 304	
19	22 TOP CAP	S.S. TYPE 304	1/4 O.D. X .001 WALL
20	21 TOP CAP	S.S. TYPE 304	1/4 O.D. X .001 WALL
21	20 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
22	19 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
23	18 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
24	17 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
25	16 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
26	15 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
27	14 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
28	13 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
29	12 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
30	11 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
31	10 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
32	9 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
33	8 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
34	7 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
35	6 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
36	5 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
37	4 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
38	3 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
39	2 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
40	1 FUEL CLAD	S.S. TYPE 304	1/4 O.D. X .001 WALL
41	SCREENS	S.S. TYPE 304	1/4 O.D. X .001 WALL
42	SUPPORT TUBE	S.S. TYPE 304	1/4 O.D. X .001 WALL
43	TOP SURFLANG	S.S. TYPE 304	
44	THROTTLE		
45	METAL LOCK RING		
46	SET SCREW	S.S. TYPE 304	
47	RETAIN RING		
48	OUTER CAP TUBE	S.S. TYPE 304	
49	SPRING	S.S. TYPE 304	
50	SCREENS	S.S. TYPE 304	1/4 O.D. X .001 WALL
51	SCREENS	S.S. TYPE 304	1/4 O.D. X .001 WALL
52	BASE SUPPORT	S.S. TYPE 304	
53	BASE SUPPORT	ALUMINUM	
54	BASE TUBE	ALUMINUM	
55	ALU. HOLDER	ALUMINUM	
56	DESCRIPTION		

LIST OF MATERIAL

7661-2526
Figure 1. Phase-III TREAT Capsule
NAA-SR-11650
I-27
OSTI ID: 000019411



7661-2528

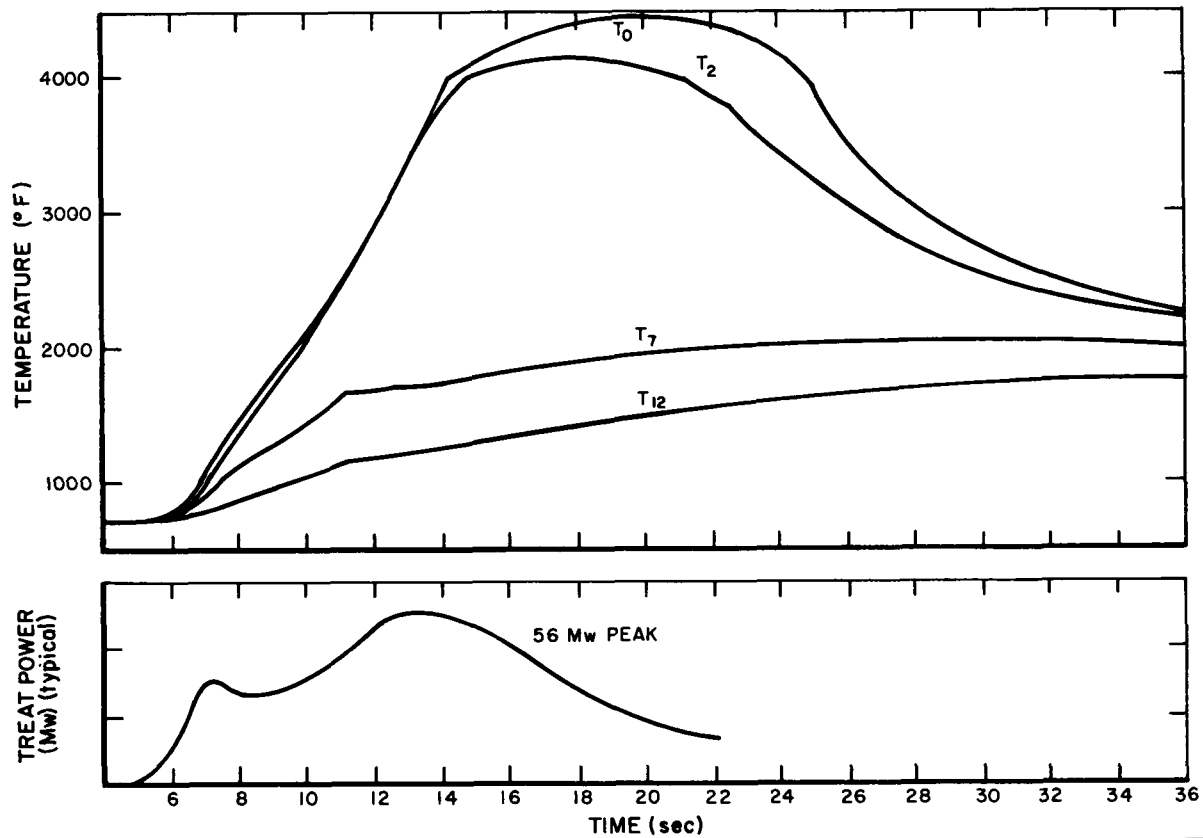
Figure 3. Predicted Thermal Behavior (Improved Nodes)
Slow Transient

Figure 6 presents the results of this analysis and shows that a few tenths of a second might be required to bring the relatively cold cladding to its melting point (2500°F). An additional few tenths of a second would be required to completely melt the cladding.

The coolant thermal data show a substantial temperature rise between 1.5 and 2 seconds, resembling the rise in the predicted behavior between 1 and 1.5 seconds (due to simulated fuel clad contact). This implies that fuel-clad contact occurred at approximately 1.5 seconds and that edge melting and sagging of the fuel was completed at that time.

C. PUBLICATIONS

A paper was completed for the International Fast-Reactor Conference at Argonne National Laboratory entitled, "The Application of Experimental Data from TREAT Meltdown Studies to Reactor Accident Analysis." A summary paper of the TREAT program on uranium carbide transient heating experiments is in preparation.



7661-2529

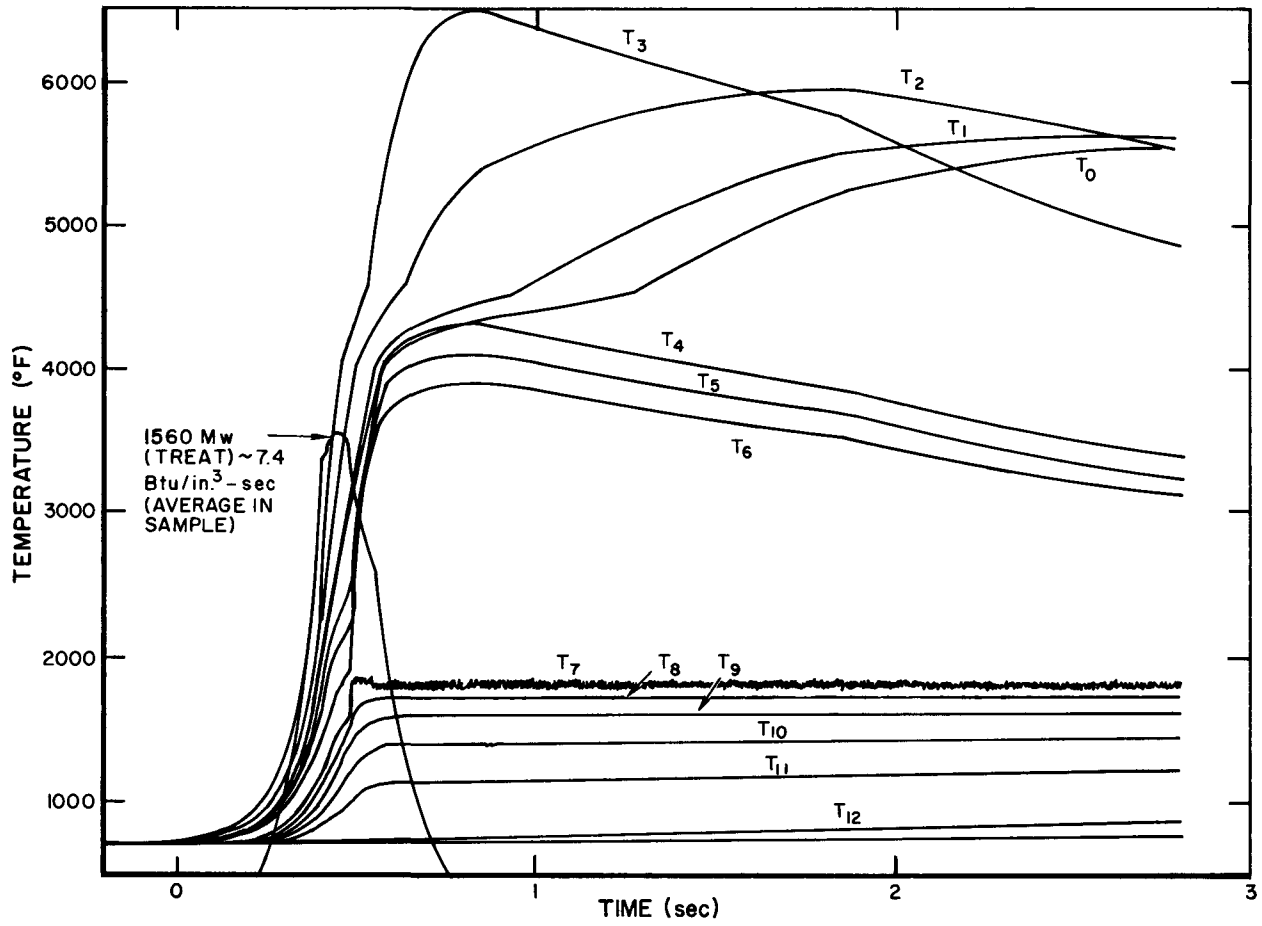
Figure 4. Comparison of Thermal Behavior With or Without Bond Boiling

IV. EVALUATION OF EFFORT TO DATE

All of the necessary groundwork, in terms of planning and design for the Phase-III experiments, has been completed. Fabrication of the capsules is in the initial stages. Improvements to the analog program for predicting the thermal behavior of the capsules have provided additional understanding of the Phase-II results regarding the mechanisms of fuel dispersion.

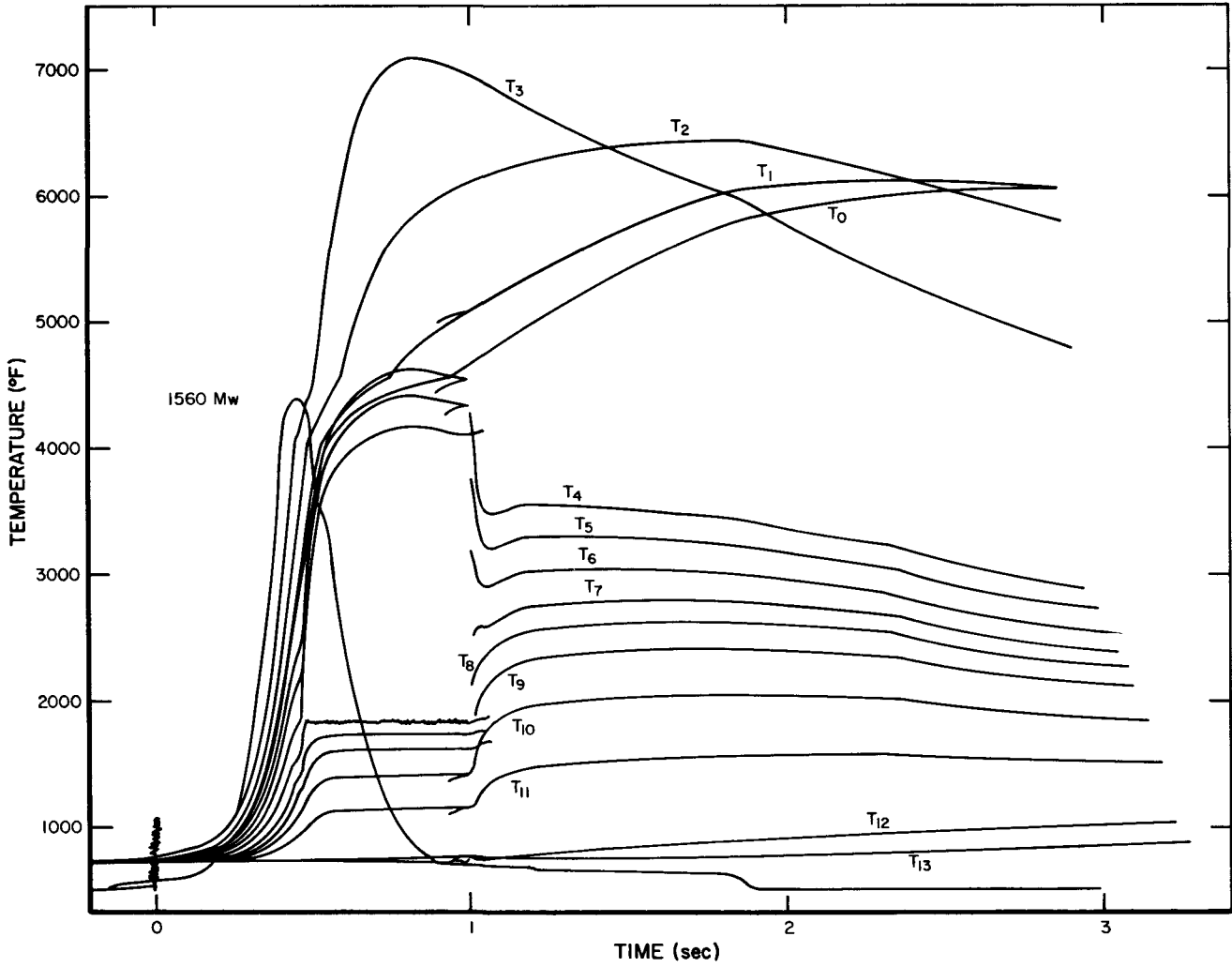
V. NEXT REPORT PERIOD ACTIVITIES

This project was terminated.



7661-2530

Figure 5. Application of Improved Model to Rapid Transient



7661-2531

Figure 6. Thermal Behavior Incorporating Simulation of Fuel Collapse

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A Division of North American Aviation, Inc.

Program: Fast Reactor Development

Project: Carbide Fuel Material Development

Project Engineer: B. R. Hayward

Reporting Period: July-September 1965

AEC Category: 04-01-04-19

General Order: 7661

Subaccount: 1921

I. PROJECT OBJECTIVES

The objectives of this project are: (a) to perform an evaluation of published data on the fast-reactor irradiation behavior of uranium-plutonium carbide and uranium-plutonium oxide fuel materials; (b) to perform an evaluation of thermo-physical and mechanical properties of U-Pu-C system materials and to determine the feasibility of performing property measurements on samples in this material system; (c) to identify the materials properties which require further investigation and suggest methods for determining these properties; (d) to define procedures to be used in the chemical analysis of plutonium-containing materials for carbon, oxygen, nitrogen, and/or plutonium; and (e) to define the required procedures and physical constants for determining burnup in U-Pu fuels.

II. PROGRESS DURING REPORT PERIOD

A. FUEL MATERIALS EVALUATION

An evaluation of the available irradiation data on uranium and uranium-plutonium carbide fuels was completed. Areas of uncertainty exist in the experimental thermal histories and in the burnup analyses for plutonium-containing fuels. In summary, it was concluded that there are essentially no published data on mixed carbides irradiated in a fast flux. These data are being generated at present in the USSR BR-5 reactor and in the UKAEA Dounreay Fast Reactor. The only operating fast-flux test facility in the U. S. is the EBR-II at NRTS, Idaho, and it has minimal test volume (~3 ft³) with no provision for any experiment instrumentation. With major problem areas existing in the definition of flux energy spectra in the U. S. test reactors and in capture-to-fission cross-section ratios as a function of neutron-energy for Pu and U isotopes, the need for instrumented experiments to develop reliable fast-reactor materials irradiation data is critical. There are no facilities in the U. S., existing or planned in the next three years, which can satisfy this need.

In addition to the search of the literature on mixed carbide fuel materials behavior, the results of a literature survey on mixed oxide fuel materials were published as NAA-SR-Memo-11465.*

B. REVIEW OF FUEL PROPERTIES

A review of the thermophysical properties of the uranium-plutonium-carbon system was completed during the quarter. Data available on the U-Pu-C system

*R. D. Hahn, P. S. Olson, and S. Strausberg, "U-Pu Oxide Fast Reactor Fuel Technology," NAA-SR-Memo-11465

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were on thermal conductivity, electrical resistivity, thermal expansion, and volatility. No data are available on thermal diffusivity and heat capacity. The available data are sporadic and a need is indicated for substantial experimentation. Specifically, the available data are limited to low temperatures, narrow composition, high density, and narrow impurity range. All of these variables can cause large variations in thermal conductivity. The highly successful laser pulse technique used on UC would be used for the mixed carbides.

The possibility of operating a relatively simple and inexpensive facility for obtaining thermal properties (thermal diffusivity, heat capacity, and electrical resistivity) of the U-Pu-C system is being evaluated. The problems associated with the toxicity of plutonium do not appear to be overwhelming. A safe experimental setup appears to be easily obtainable due to the very small quantities of Pu needed for the measurement and the methods of handling contained material.

The general feasibility of property determinations in a glovebox facility at Atomics International's headquarters facility was considered and evaluated by Radiation Engineering (AI) and detailed experimental procedures were prescribed.

Two reports were completed during this report period: "Thermophysical Properties of the (U, Pu)C System," NAA-SR-TDR-11659, by M. Nakata, and "Proposed Research of the Thermophysical Properties of (U, Pu)C Systems," NAA-SR-TDR-11660, by M. Nakata.

Data on the mechanical properties for (U-Pu)C fuels were found to be practically nonexistent. There was a range of room temperature compressive strength and hot hardness values for 25, 500, and 900°C.* These data were for fuel materials of essentially the same composition. Data on the variation in properties, as a function of composition, were not found.

The measurements for hot hardness and elastic properties (Young's modulus, shear modulus, and Poisson's ratio) seem feasible for testing at AI in FY 1966. A sample 1/2-in. -diameter and 1/4-in. -thick (11 grams) could be used for both measurements. The elastic properties could be measured to 800°C with the pulse-echo (sonic) technique. This measurement would depend on the grain size of the samples which is reported to be in the appropriate range, and on the density which must be 98% or higher. The hot hardness could be measured to 1000°C independent of the sample properties, but is most meaningful in samples of low porosity. The measurements are possible on samples with no metallic second phase. Because of the appreciable vapor pressure of Pu in the metallic phase, the temperature limitation would be considerably lower for substoichiometric samples. If the microscope used to measure the hardness indents is equipped with a hot stage, phase transformations, recrystallization and precipitation could also be observed to about 800°C.

C. IDENTIFICATION OF AREAS REQUIRING STUDY

A review and evaluation of physical-chemical properties of PuC and mixed carbides was completed and a progress report issued as TDR-11506.† The

*Paper 154 - 1964 Geneva Conference

†G. Ervin, Jr., and A. K. Smalley, "Materials Problems Relative to the Use of Plutonium Compounds as Fuels in Fast Breeder Reactors - A Literature Review and Evaluation," NAA-SR-TDR-11506 (July 21, 1965)

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general conclusion reached is that much of the important information needed to make effective use of uranium-plutonium mixed carbides as fuels is still unknown, and that research is needed in most areas. Particular areas of critical need are: (1) high temperature phase equilibria in the U-Pu-C system, (2) effects of oxygen and nitrogen on the phase equilibria together with kinetics of high temperature reaction of (U, Pu)C with low partial pressures of oxygen and nitrogen, and (3) volatility of (U, Pu)C. The effects of oxygen and nitrogen need to be better understood to remove uncertainties about both phase equilibria and volatility. Experimental research is in progress on the U-C system and on oxygen-UC interactions; this work is a necessary preliminary to work with U-Pu-C. In order to perform experiments in which effects of oxygen and nitrogen are known and controlled, ion-pumped systems are being installed for vacuum atmospheres, and inert gas atmospheres are being purified to the 1 ppm impurity (oxygen, nitrogen, and water vapor) range. Monitoring of the impurity content by gas chromatography and other methods to measure less than 1 ppm of these gases is planned.

The effects of low partial pressures of oxygen on UC are under study. A report of preliminary results has been accepted for publication in *Acta Metallurgica*. This report shows that at 10^{-4} torr oxygen pressure, oxidation of UC is rapid at 900 to 1000°C, forming epitaxial layers of UC₂ and UO₂ on the surface; but at 1800°C at the same pressure, rapid loss of uranium and carbon occur and there is no surface coating produced. Work in progress is expected to ascertain the pressure and temperature limits for these radically different types of behavior, as well as the conditions under which oxygen solution and other changes in the UC lattice occur.

D. ANALYSIS OF ANALYTICAL CHEMISTRY PROCEDURES

Based on the information available, most of which is derived from metal or mixed oxide work (only AERE reports deal specifically with PuC), the following procedures are considered most applicable to the requirements of the Fast-Reactor Program.

1) Carbon

A combustion procedure utilizing a thermal conductivity detector as a readout (now in use for uranium carbide) reported and evaluated by Larsen and Glass,* is deemed to be adequate. Such a technique can be made compatible with glove-box operation and is applicable to both irradiated and unirradiated material.

2) Oxygen

Either vacuum fusion or inert gas fusion is applicable to glove-box operation. The latter would be preferable from a maintenance-of-equipment standpoint and should be investigated to remove the inconsistencies in operation observed in its application to UC analysis.

3) Hydrogen

Vacuum extraction, as currently used, is applicable to this determination. Simultaneous determination with oxygen may be possible and should be investigated using gas chromatographic readout with inert gas fusion.

*C. M. Larsen and D. N. Glass, "Evaluation of the Combustion-Thermal Conductivity Technique for the Determination of Carbon in Uranium Carbide," NAA-SR-TDR-11495

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4) Nitrogen

Kjeldahl distillation into boric acid and titration of the collector with strong acid is applicable to glove box operation and, if required, can be adapted for irradiated material.

5) Plutonium

Several different analytical methods for the determination of plutonium should be verified and evaluated for glove-box or shielded remote operation.

a) Spectrophotometric

Plutonium is separated by solvent extraction with tetrapropylammonium nitrate and methyl isobutyl ketone. Its absorption spectrum is observed spectrophotometrically. This technique is useful for unirradiated plutonium. However, irradiated fuel would necessitate setting up a remote extraction operation. It can be used on mixed uranium-plutonium matrices.

b) Coulometric

Controlled potential coulometry should be ideally suited to glove-box operation and/or remote shielded analysis. According to the literature, analysis for both U and Pu can be accomplished by this technique. This technique should be further investigated and evaluated as the first choice of method.

c) Volumetric

A conventional redox titration of separated plutonium is applicable to unirradiated fuel and with suitable modification could apply to irradiated fuel. This technique requires a separation of plutonium from uranium, iron, molybdenum, and similar interferants.

d) Polarographic

Utilization of characteristic half-wave potentials may enable the analysis for plutonium in the presence of uranium. This technique, reported in 1957, from LASL, has not received wide acceptance and therefore may not be as tempting as the literature indicates.

E. ANALYSIS OF BURNUP MEASUREMENT TECHNIQUES

Plutonium burnup requirements fall into two categories: (1) facility requirements, and (2) technical requirements. This discussion will deal only with the latter category, although the former must be dealt with early in the time scale.

There is, as yet, no well agreed upon technique for the determination of burnup in mixed fissile-element fuels. Those under active consideration which require careful evaluation before making a choice (1) to determine the yield of one or several fission product nuclides, either stable or radioactive, or (2) measure the isotopic distribution of the heavy elements.

1. Fission Product Determination

Several fission products, e.g., Te^{99} , La^{139} , Nd^{148} , etc, have nearly the same yield for the fission of U^{235} , U^{238} , and Pu^{239} . If these yields are independent of the energy range over which the fast reactor operates (requires verification); then, measurement of the fission products provides an indication of the number of fissions which have occurred. The three fission product nuclides mentioned above are long-lived or stable and require nonradiochemical measurement techniques. Argonne proposes spectrophotometric determination of technetium. G. E. Vallecitos proposes isotope dilution mass spectrometric analysis for neodymium. The latter is also an approach for the lanthanum determination requiring lanthanum spike-enriched in La^{138} .

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The basic requirements of this technique are: (1) an accurate knowledge of the fission yields from U and Pu; (2) independence of fission yield with neutron spectrum or the known function with respect to energy; (3) knowledge of the capture cross-section of the fission product species for high flux, long-time exposures for estimating the "disappearance fraction;" and (4) as in-pile times become longer, the contributions from heavier fissile species must be considered; hence, the knowledge of these fission yields is required. A compilation of fission yields and energy dependence, where documented, is in process.

2. Heavy Element Mass Spectrometry

This technique involves a determination of the isotopic distribution of the heavy elements and a ratio of one of the isotopes to an isotope of the original material. This technique requires a knowledge of α , the capture-to-fission ratio, and its energy dependence for the fissile elements. The neutron spectral distribution likewise must be known.

Both preirradiation and postirradiation heavy element isotopic distribution are required. Present techniques for the separation of U-Pu are adequately documented in the literature. Computational details have not as yet been formulated nor has an error analysis been undertaken.

III. EVALUATION OF EFFORT TO DATE

The reviews and evaluations of the out-of-pile and irradiated properties of the mixed U-Pu carbides indicate a general lack of well-defined data. Although some specimens of U-Pu-C have been irradiated to over 100,000 Mwd/MT, the resultant data is limited to scoping value. It is apparent that an extensive materials development program is required for both out-of-pile and irradiated material properties. Due to the large volume increases resulting from the high burnup, this development program is affected by basic materials design criteria such as the retention or release of fission gases, volatile fission products, phase equilibria, and the original density of the fuel. It is important to note that much of the data currently being developed within the US and in the UKAEA are at relatively low specific power, ~100 to 125 w/g, whereas the economic evaluations indicate specific powers of 200 to 400 w/g are required. Thus, data obtained at low specific power must be carefully analyzed and evaluated for extrapolation to high specific power. One of the major properties requiring immediate attention is thermal conductivity. A substantial continued effort is also required on irradiation tests with the primary effort to evaluate the relationship of composition, density, and impurities with dimensioned changes, temperature, burnup, and fission gas release. This fuel materials data must coordinate with the fuel rod system requirements and the fuel fabrication methods development. The supporting analytical techniques that are required to prepare specimens and evaluate results should continue to receive attention.

IV. NEXT REPORT PERIOD ACTIVITIES

This project was terminated.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program: Fast Reactor Development

Project: Carbide Fuel Process Development and Fabrication

Project Engineer: B. R. Hayward

Reporting Period: July - September 1965

AEC Category: 04-01-04-19

General Order: 7661

Subaccount: 1931

I. PROJECT OBJECTIVES

The purpose of this project is to develop economical fuel processing methods for the reliable synthesis and fabrication of carbide fuels, within specifications required for high-performance, fast-reactor systems. The primary objective of this project is to evaluate the capability of existing and potential processes, and to demonstrate the reliability of those processes which prove optimum for the fabrication of mixed (U, Pu)C fuels.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

A report has been prepared summarizing the status of existing technology for the preparation and fabrication of solid-solution mixed carbides (U and Pu) to fast-reactor fuel sizes.

A preliminary experimental work has been carried out to evaluate potential problems of plutonium evaporation during fuel processing operations.

The feasibility of casting carbides to fast-reactor fuel pin size, 0.25 and 0.19-in. dia., in approximately 6-in. lengths has been demonstrated.

III. PROGRESS DURING REPORT PERIOD

A detailed report was completed that evaluates potential problem areas in the fabrication of mixed carbides for fast-reactor use, current practices and potential methods were analyzed. Experimental activity was initiated in fabrication techniques considered most promising for (U, Pu)C fuels; namely; (1) synthesis from the mixed oxides, (2) consolidation by cold-press and sinter techniques, (3) arc-melting and casting and (4) vibratory compaction. The chief problem areas appear to be potential vaporization losses of plutonium during processing and the feasibility of casting carbides to fast reactor fuel pin sizes.

Considerable information on losses of Pu and phase structure of sintered pellets was obtained under subcontract by UNC (at Pawling, N. Y.), to an experimental program initiated at AI. A terminal report (UNC-5126) was recently issued. The data are summarized in Tables 1 and 2. Solid solution (U, Pu)C with an intended Pu/U ratio of 0.25 was synthesized by the reaction of UO_2 , PuO_2 , and carbon in vacuum at 1550 and 1650°C, and in helium (1/2 atm) at 1650°C. Carbon contents for the carbide "clinkers" were near the stoichiometric value of 4.80 wt % C. Losses of Pu during synthesis were essentially zero for the 1550°C run, ~2.4% for the 1650°C helium atmosphere runs, and ~5.0% for the 1650°C vacuum (3×10^{-4} Torr) runs. Oxygen contents were

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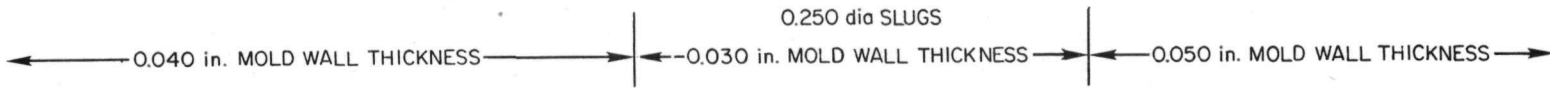
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~2300 and 4400 ppm for the carbide products synthesized at 1650°C under vacuum or helium, respectively. Oxygen contents for the 1550°C run were not determined.

The (U, Pu)C "clinkers" were comminuted by ball milling and the powders were cold pressed to 0.220-in. dia. compacts and sintered with and without nickel sintering aid (0.2 wt % Ni). Sintering conditions were 1590°C in helium or 1950°C under vacuum. Plutonium losses at 1590°C ranged from zero to 3.1% and at 1950°C they were 1.6 to 5.2%. The pellets sintered at 1590°C with nickel were of high density, 12.7 to 13.0 gm/cm³; the pellet sintered at 1590°C without nickel was of low density. The pellets sintered at 1950°C without sintering aid were of high density (12.5 to 12.7 gm/cm³). Sintering at 1590°C in commercial high purity helium resulted in negligible changes of carbon content with oxygen values increasing from ~2300 to 6800 ppm. Vacuum sintering at 1950°C reduced carbon contents to ~4.50% and oxygen levels from ~4400 to ~2300 ppm. The microstructure of all pellets showed the typical monocarbide matrix. Pellets sintered at 1590°C with nickel contained a second phase of sesquicarbide; pellets sintered at 1950°C contained minor amounts of free metal. The pellet sintered at 1590°C without sintering aid was essentially single phase, though of low density.

Similar experiments (at AI) using cerium as a "stand-in" for the Pu in the synthesis and fabrication of (U_{0.8}Ce_{0.2})C pellets indicated slightly greater losses of cerium. Synthesis of the mixed carbides under vacuum (2×10^{-4} Torr) at 1650°C indicated cerium losses of 0, 9.2 and 27.3% in attempts to prepare hyperstoichiometric, stoichiometric, or hypostoichiometric mixed carbides, respectively. Similar losses for vacuum synthesis at 1750°C were 8.5, 31.9, and 41.8%, respectively. Most of the cerium collected on the wall of the crucible as golden, crystalline deposits, suggesting that Pu recovery may not present too great a problem. It was observed that the U-Ce carbides are much more susceptible to hydrolysis during normal laboratory handling than similarly prepared uranium carbides. Under precautions minimizing laboratory exposure, oxygen levels of ~1100 ppm were obtained for these synthesized U-Ce carbides. When U-Ce pellets were sintered at 1590°C with 0.2 to 0.4 wt % Ni in 1/3 atm of helium, densities >94% of theoretical were obtained. The structure of these pellets appeared similar to those of the (U, Pu)C pellets prepared by UNC. Photomicrographs indicated a two-phase structure, one phase probably being a sesquicarbide; there were no dicarbide platelets (Widmanstätten lines) evident, even in hyperstoichiometric carbide preparations. A report was completed on this U-Ce-C fabrication work, NAA-SR-TDR-11570.

Scoping tests were carried out to evaluate losses of cerium resulting from arc-melting operations for comparison with reported Pu losses. Compositions of approximately (U_{0.8}Ce_{0.2})C were used, from the above mentioned synthesis or prepared from the elements with graphite. Oxide-derived carbides, when arc-melted under 1.0 atm argon and kept molten for 2 minutes, lost from 5.5 to 12.8% cerium, with losses varying inversely with the carbon contents. The carbides obtained from the elements lost from 2.5 to 5.3% Ce when arc-melted in 1.0 atm argon for from 2 to 8 minutes, with little difference observed between stoichiometric or hyperstoichiometric carbon compositions. Losses increased with increased melt times. Arc-melting similar buttons under a lower pressure, 0.1 atm argon, for 8 minutes increased the cerium losses 7.0 to 8.5%. These



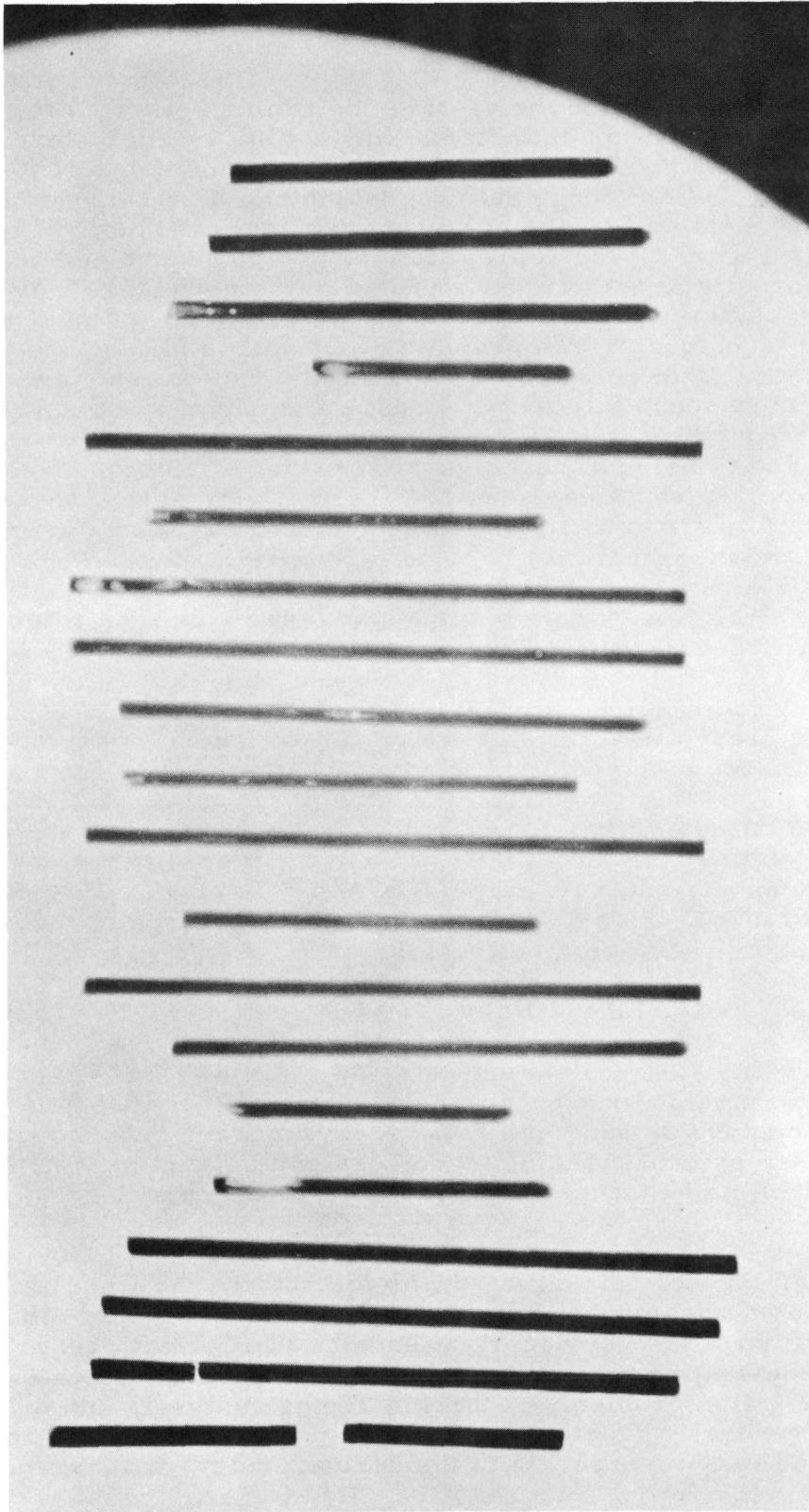
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7661-2532

Figure 1. Typical As-Cast 1/4-in. Diameter UC Pins

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Figure 2. Typical Gammagraphs Of As-Cast UC Slugs

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cerium vaporization losses and photomicrographs of the U-Ce buttons agree closely with fabrication results obtained with $(U_{0.85}Pu_{0.15})C$ as reported at DERE.

Experiments were initiated on the casting of UC pins (~4.8% C) to fast-reactor sizes; i. e., 0.250- to 0.187-in. diameter. Single-pin casting techniques developed to date successfully produced cast pieces >3-in. long, 90 to 100% of the time. Full length, 6-1/4-in. long castings, which can be obtained ~50% of the time, are still dependent on furnace operator technique. Typical as-cast UC slugs are shown in Figure 1, and representative gammagraphs can be seen in Figure 2; slight porosity is sometimes obtained in the upper 1 to 2-in. of a 6-in. long casting. The casting operation was carried out by conventional skull-melting, tilt-pour techniques. The graphite mold wall thickness was varied 0.020 to 0.060-in. To date, molds of ~0.030-in. wall thickness appear near optimum for obtaining sound full-length slugs. The use of previously outgassed molds (at 2350°C) heated to ~1000°C prior to pouring improves the surface of the casting compared to using a nonoutgassed, nonheated mold. The principal surface defects are cold-shuts, as seen in Figure 1. Cursory tests were carried out with these small diameter castings to determine the feasibility of obtaining a good slug surface finish, which may become a requirement if fast-reactor fuel designs incorporate a gas-bonded fuel rod. No difficulties were encountered in surface grinding. Preliminary data obtained on 1/4-in. diameter castings of 2 to 5-1/4-in. length having initial TIR* values of 0.002 to 0.007-in. and diametric "out-of-roundness" of 0.001 to 0.002-in., indicated diametric stock removals of 0.017 to 0.032-in. were required for complete surface "clean-up". These data indicate that at the present state-of-the-art, ~20% of the cast carbide product may have to be removed by grinding if "contact-bond" finishes are required for fast-reactor fuels. The fuel pin surface finish after grinding was ~120 to 180 rms. Tests were also initiated on multiple casting techniques (7 slugs per pour). Results show that on the basis of a limited number of tests, there was partial success with 3/16-in. diameter molds; only 2 to 4-in. lengths of sound pins can usually be obtained in a 6-1/4-in. length mold. Similar tests are in progress for the multiple casting of 1/4-in. diameter pins.

IV. EVALUATION OF EFFORT TO DATE

Process engineering analysis showed that one of the key mixed carbide fabrication problems is Pu volatilization. Scoping tests indicate that Pu vaporization losses during mixed carbide synthesis from the oxides and subsequent fabrication to pellets by cold-press and sintering techniques can be held below 3%. Fabrication by arc-melting and casting techniques may result in somewhat higher losses of Pu.

In order to study Pu fuel fabrication problems without the limitations of Pu, Ce has been used as a cheap substitute for many years in both the US and UK. Comparison of cerium losses by vaporization with similar data reported for plutonium fuels show that cerium is an economical stand-in for evaluation of potential Pu vaporization problems occurring during synthesis and processing. Losses of Ce are generally greater than those expected for similar operations with plutonium, and vary inversely with the carbon content of the mixed carbide.

*TIR - Total Indicator Reading is a measure of the bow or nonstraightness in the total slug length.

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TABLE 1
 SYNTHESIS OF URANIUM-PLUTONIUM CARBIDE FROM THE OXIDES

Synthesis Run	A	B	C	D	E
Atmosphere	Vacuum*	Vacuum*	Vacuum*	0.5 atm He ⁺	0.5 atm He ⁺
Temperature (°C)	1650	1650	1550	1650	1650
Hold Time (hr)	2	2	2	2.5	2.5
Initial Pu Comp'n (%)	15.5	15.5	15.6	15.6	15.5
Clinker Content:					
Carbon (wt %)	4.76	4.86	4.73	4.76	4.83
Oxygen (wt %)	0.23	-	-	-	0.44
Pu (wt %)	18.2	18.1	19.2	18.6	18.7
Plutonium Loss (wt %)	4.7	5.2	0.0	2.6	2.1

* 3×10^{-4} Torr

+The He atm runs were static with five evacuations and refills of the furnace during the run.

It is recommended that tests with Ce be performed prior to the actual Pu tests in all applicable phases of fuel fabrication. Preliminary arc casting tests with stoichiometric UC demonstrate that high quality carbide pins as small as 0.187-in. diameter and up to 6-in. long can be obtained by modified arc-melting and casting techniques. Expansion of the multiple mold casting is required to put the promising small diameter casting work on a practical basis. Additional analytical effort is required to evaluate microstructure segregation and composition variations. Further tests are required to define the capability as well as the need for surface finish specifications. Grinding appears to be a required finishing method until process improvements are developed. It should be noted that the results obtained apply to the specific compositions studied. A more thorough analysis should be made of the effects that other potential fast-reactor compositions may have on the choice of fabrication methods.

V. NEXT REPORT PERIOD ACTIVITIES

This project was terminated.

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TABLE 2
 SINTERING OF (U, Pu)C PELLETS

Sinter Run	F	F	G	G	H	H
Charge from Synthesis Run	A	C	B	D	C	E
Temp (°C)	1590	1590	1590	1590	1950	1950
Atmosphere	He**	He**	He**	He**	Vacuum	Vacuum
Hold Time (Hr)	2	2	2	2	1	1
Ni @ start (wt %)	0.2	0.2	none	0.2	none	none
Pellet Content:						
Carbon (wt %)	4.92	4.73	4.72	4.77	4.50	4.52
Oxygen (wt %)	0.68	-	-	-	-	0.23
Pu (wt %)	18.1	18.6	17.9	18.6	18.2	18.4
Plutonium Loss (wt %)	0.6	3.1	1.1	0.0	5.2	1.6
Pellet Density (gm/cm ³)	12.94	12.68	12.47	13.00	12.70	12.52
Metallography	Major (U, Pu)C, moderate sesqui-carbide	Major (U, Pu)C, moderate sesqui-carbide	Single-phase (U, Pu)C, porous	Major (U, Pu)C, minor sesqui-carbide	Major (U, Pu)C, minor metal	Major (U, Pu)C, minor metal

**1/2 atm of static helium. Furnace evacuated and backfilled three times with high-purity He.

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Program: Fast Reactor Development	
Project: Carbide Fuel Element Development	
Project Engineer: B. R. Hayward	
Reporting Period: July-September 1965	AEC Category: 04-01-04-19
General Order: 7661	Subaccount: 1941

I. PROJECT OBJECTIVES

The objectives of this project are to develop reference designs and prepare preliminary fuel specifications for fast reactor fuel elements; establish design criteria for irradiating mixed uranium-plutonium carbide fuel element test assemblies; and perform a fast reactor fuel cycle cost survey based on proposed plant requirements.

II. PROGRESS DURING REPORT PERIOD

Liaison was established between the Laboratory Director's Office of Argonne National Laboratory and Atomics International to evaluate irradiation space in the fast-flux Experimental Breeder Reactor-II (EBR-II). Irradiation experiment requirements were received from the EBR-II Irradiation Testing Liaison Engineer at NRTS, Idaho, and evaluated with regard to what conditions could be attained in the EBR-II and what the tentative experiment design would require in fuel fissile contents, containment, heat transfer media, etc, to satisfactorily perform under those conditions.

A review and evaluation of the characteristics and criteria for the fuel rod systems were prepared. A total plan for the fast reactor fuel systems development activities has been written in draft form.

III. EVALUATION OF EFFORT TO DATE

The key factors involved with fuel rod system development are (1) allowable strain on the clad, (2) dimensional stability and fission gas release of the fuel, (3) gas bond versus sodium bond, (4) vented versus nonvented rod design, and (5) original density versus performance requirements. These factors in turn require analysis and evaluation based on fuel cycle economics. The results of this evaluation indicated (a) the need for expanded materials effort on both fuel and clad, (b) expanded fuel processing development to meet the needs for long burnup, and (c) the need for system tests of a scoping nature in parallel with the main materials development. Many of the initial fuel rod system design criteria are based on very limited material data.

There appear to be some advantages of the sodium bond design over the gas-bonded design. The sodium bond concept provides (1) an annulus for the volume increase caused by solid fission product accumulation, (2) lower temperature operation, which has advantages of avoiding high-temperature phase changes at certain compositions but has the disadvantage of lower diffusion rates of fission gases, which could result in fuel swelling, (3) higher specific power operation, (4) less apparent safety characteristics for sodium boiling as compared

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with the gas-bonded design. This latter evaluation depends on assumptions used for the analysis, and requires additional effort supported by experimental results.

There are three problem areas in the fuel rod system that require plant engineering support: (1) vented fuel rod design, (2) fuel rod safety characteristics during steady-state and transient conditions, and (3) heat transfer analyses involving fission gas mobility with the bond and coolant.

The change in properties of the clad materials under irradiation and the effect of coolant on cladding are both being evaluated through coordination with the clad development program 7661-4553.

These analyses of existing engineering data compared with the required performance criteria have provided excellent guidance for the technical scope, and emphasis on the materials development program and the process development activities.

IV. NEXT REPORT PERIOD ACTIVITIES

This project was terminated.

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Program: Fast Reactor Development	
Project: Cladding Evaluation	
Project Engineer: B. R. Hayward	
Reporting Period: July-September 1965	AEC Category: 04-01-04-19
General Order: 7661	Subaccount: 1951

I. PROJECT OBJECTIVES

The major objective of this project is to determine and demonstrate the validity of specific clad-design criteria for fast-reactor fuel clad-materials. The effect of chemical composition and grain size on the biaxial low strain rate behavior of austenitic stainless steels will be evaluated.

II. PROGRESS DURING REPORT PERIOD

The cladding stress-rupture tests continued. When these "tests" ruptured, measurements were taken but no attempt was made to thoroughly analyze the data.

Specimens tested were from six different heats of type 304 stainless steel and two heats of type 316 stainless steel. These heat compositions are identified in Table 1. The tests were biaxially loaded at 1250 and 1350° F, and were performed with seamless tubing.

TABLE I
CHEMICAL COMPOSITION OF TASK 2B SPECIMENS

Heat	Code	C	N	Mn	P	S	Si	Ni	Cr	Mo
27898X	A	0.06	0.060	1.67	0.021	0.011	0.50	10.41	18.93	-
06737	B	0.06	0.056	1.64	0.022	0.025	0.39	9.82	18.54	-
06820	C	0.06	0.047	1.84	0.023	0.009	0.44	9.79	18.54	-
00042	D	0.05	0.052	1.77	0.023	0.021	0.55	9.70	18.55	-
90213	E	0.05	0.037	1.68	0.018	0.021	0.45	9.67	18.47	-
26980X	F	0.06	0.059	1.68	0.027	0.013	0.66	10.27	18.71	-
96180	G	0.03	0.027	1.54	0.003	0.021	0.30	13.39	17.01	2.28
90106	H	0.04	0.043	1.68	0.022	0.017	0.51	13.63	17.33	2.32

Sixty-four specimens ruptured during this quarter. The results of these tests are summarized in Table 2. Previous tests, under 20,000 psi hoop stress at 1350° F, exhibited axial tears of one to three inches in length. The current tests, at lower hoop stresses, were all characterized by pinhole failures. These pinhole failures are similar to those experienced in previous tasks where welded-

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TABLE 2
SUMMARY OF LOW STRAIN-RATE, BIAXIAL, CREEP-RUPTURE TESTS
FOR
JULY-SEPTEMBER, 1965

Heat and Specimen No.	Temperature (°F)	Hoop Stress (psi x 10 ³)	Time to Rupture (hr)	Maximum Strain at Failure (%)
A-9	1250	13.5	1802	11.0
A-10	1250	13.5	2336	15.5
A-11	1250	13.5	1992	14.2
A-16	1250	15	1217	-
A-8	1350	10	612	*
A-25	1350	10	520	10.0
A-26	1350	10	612	12.5
A-27	1350	10	564	12.0
B-9	1250	13.5	1992	20.5
B-10	1250	13.5	1808	-
B-16	1250	15	1049	-
B-8	1350	10	608	-
B-25	1350	10	612	17.1
B-27	1350	10	288	13.8
B-28	1350	10	488	10.4
C-9	1250	13.5	3772	-
C-10	1250	13.5	3027	-
C-13	1250	15	2440	17.7
C-14	1250	15	2456	18.8
C-15	1250	15	2040	14.6
C-8	1350	10	908	-
C-24	1350	10	142	-
C-25	1350	10	756	7.3
C-26	1350	10	600	8.1
C-1	1350	13.5	186	19.0
C-2	1350	13.5	184	37.8
C-22	1350	15.0	72	25.5

(continued)

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TABLE 2 (Continued)

Heat and Specimen No.	Temperature (°F)	Hoop Stress (psi × 10 ³)	Time to Rupture (hr)	Maximum Strain at Failure (%)
D-11	1250	13.5	2907	-
D-16	1250	15	1536	-
D-8	1350	10	632	-
D-21	1350	13.5	141	30.4
D-22	1350	13.5	153	16.8
D-23	1350	13.5	160	21.6
D-24	1350	13.5	160	35.4
D-1	1350	15	72	29.8
D-2	1350	15	144	14.5
E-9	1250	13.5	1600	-
E-10	1250	13.5	1490	-
E-51	1250	13.5	1730	-
E-24	1350	13.5	72	28.4
E-2	1350	15	49	33.2
E-4	1350	15	49	34.2
F-13	1250	15	2711	17.5
F-14	1250	15	3708	-
F-30	1350	13.5	381	22.7
F-21	1350	15	130	28.3
F-22	1350	15	260	24.0
G-9	1250	13.5	2704	-
G-11	1250	13.5	2696	-
G-13	1250	15	1487	-
G-14	1250	15	1332	-
G-15	1250	15	1308	-
G-25	1350	10	541	9.0
G-26	1350	10	584	10.5
G-27	1350	10	584	10.7
G-28	1350	10	536	9.5

(continued)

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TABLE 2 (Continued)

Heat and Specimen No.	Temperature (°F)	Hoop Stress (psi x 10 ³)	Time to Rupture (hr)	Maximum Strain at Failure (%)
G-29	1350	13.5	129	15.0
G-30	1350	13.5	178	14.6
G-33	1350	15	129	17.8
G-34	1350	15	69	21.4
H-15	1250	15	3680	-
H-29	1350	13.5	464	16.1
H-33	1350	15	216	29.6
H-34	1350	15	260	27.4

*Dimensional measurements have not been made of all specimens.

and-drawn specimens were evaluated in a sodium environment containing a carbon source and a helium environment. Failures in the current task have occurred at 7 to 35% plastic strain. These failures are indicative of a creep type failure. Previous metallographic analysis has shown large amounts of deformation in the grain boundaries.

III. EVALUATION OF EFFORT TO DATE

Average rupture times for a given stress level are plotted versus stress for 1250°F in Figure 1 and for 1350°F in Figure 2. Of the various heats of 304 stainless steel, heats E and F are the weakest and strongest heats, respectively. Heat G, which is 316 stainless steel, does not appear to be stronger than the majority of the 304 stainless steel heats.

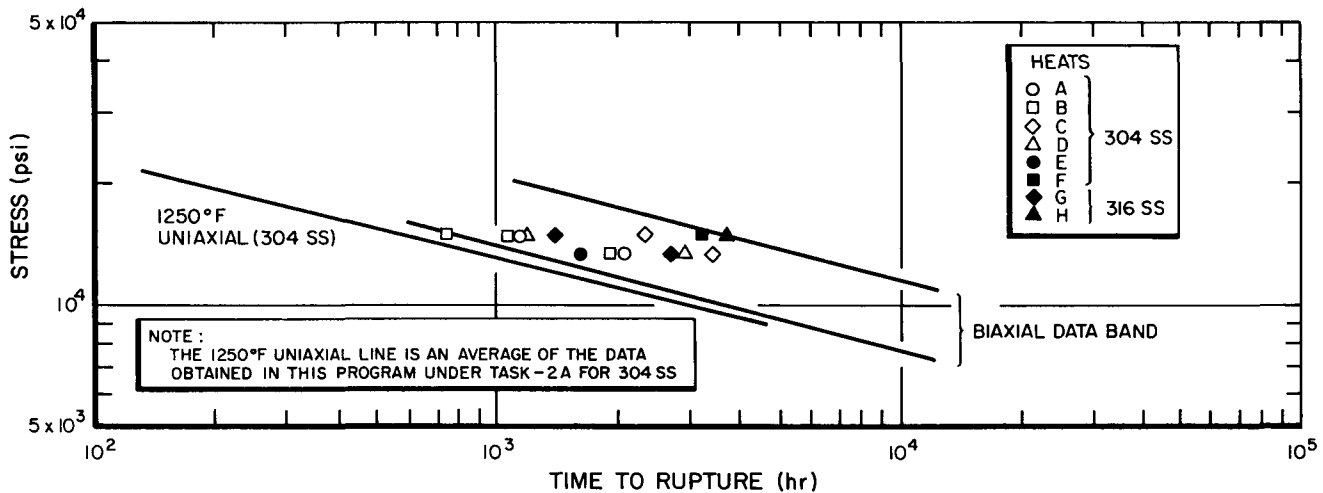
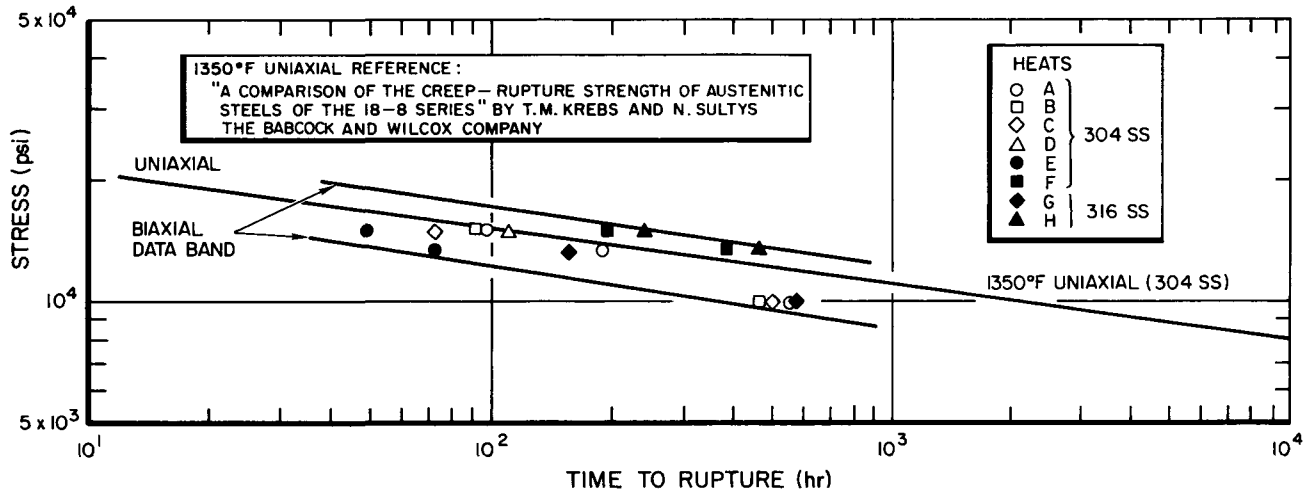


Figure 1. Rupture Data for 304- and 316 SS at 1250°F

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Figure 2. Rupture Data for 304- and 316 SS at 1350°F

The biaxial data, along with some previously reported data, are plotted as a function of hoop stress, and are compared to uniaxial data in Figures 1 and 2. Analysis of the data, by using the maximum shear or von Mises theories, would lower the two data bands and perhaps provide a better correlation with the uniaxial data. However, in order to evaluate the proper choice of stress level, tests other than biaxial creep tests would have to be conducted (i. e., biaxial pressure plus a superimposed compressive or tensile stress).

IV. ACTIVITIES FOR NEXT REPORT PERIOD

The ruptured specimens will be analyzed and evaluated.

This project was terminated. The activities will be continued under the High Temperature Materials Program.

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A Division of North American Aviation, Inc.

Program: Fast Reactor Development

Project: Irradiation Testing

Project Engineer: B. R. Hayward

Reporting Period: July-September 1965

AEC Category: 04-01-04-19

General Order: 7661

Subaccount: 1961

I. PROJECT OBJECTIVES

The objective of this project is to obtain irradiation data on the performance of fast-reactor carbide fuels, fuel element systems, and cladding materials which will be used as the basis for design of fast-reactor carbide fuel systems. Specific objectives are: materials scoping irradiations to evaluate fuels and cladding materials; materials systems irradiations to provide data on the compatibility, swelling, fission gas release, microstructure changes, and operational characteristics of the fuel-cladding system; prototype fuel rod and fuel element irradiations to provide a reference fuel rod design; and special test irradiations to provide information on fission gas pressure buildups due to step increases in fuel operating temperatures, effects of short time temperature excursions on fuel and cladding materials, and performance of vented fuel systems.

II. TECHNICAL PROGRESS DURING REPORT PERIOD

Progress on the NAA-123, a multicompartmented in-pile stainless steel clad stress-rupture experiment, is proceeding according to plan. All clad specimens were prepared and preirradiation measurements taken. Approximately 95% of the parts have been fabricated and approximately 50% of the sub-assemblies have been assembled. The temperature-measurement and control consoles have been received. Testing has disclosed that the valves in the argon flow lines, used for temperature control, are of excessive size. Their size is being reduced to restrict the flow rate and to increase the control sensitivity. This additional effort is not expected to delay the assembly schedule.

A decision has been made to determine the flux spectrum as well as the flux distribution in the test facility. In addition to monitor wires on the test assembly, a separate flux monitoring capsule will be built for installation at the downstream end of the test assembly. The data obtained from this portion of the NAA-123 experiment will characterize the neutron environment and will permit a more qualitative evaluation at the times of the postirradiation examination.

A survey has been made of test reactor facilities to determine which would be technically suitable and available for fast-reactor fuel and materials irradiation tests. The EBR-II and Dounreay Fast Reactor appear to be the only fast-spectrum facilities that can schedule experiments in the next 1 or 2 years. The availability of the Fermi reactor is questionable at significant power levels. Many thermal spectrum facilities, such as the MTR, SRE, ETR, GETR, Hanford, etc, are available which could provide the desired power densities and temperatures for irradiation of mixed carbides.

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The least expensive and still the most informative single experiment that could be conducted in the near future would be one using a 19-rod element in the EBR-II core. The experiment would be relatively inexpensive because EBR-II standard reactor element designs would be used, only a minimum amount of engineering analysis would be necessary, and currently available parts and techniques would be used for fabrication. The experiment could utilize several different fuel forms and claddings, thereby providing the valuable scoping information needed for the fast-reactor fuel element design. The preliminary nuclear, heat transfer, and hydraulic calculations have been made for an EBR-II test design. The results show that a position in one of the inner three reactor rows would be needed if the desired specific power (200 to 400 watts/gram, U & Pu) is to be attained. Limitations of the EBR-II are the lack of instrumentation of test capsules and the low operating level of the reactor. These limitations are however more important to long burnup tests than to short burnup scoping tests.

An analysis has been performed to determine the variations in radial power density of a nominal fast-reactor carbide fuel rod design (0.25-in. diameter, 20 wt % Pu) operating in a typical thermal test-reactor spectrum. The results indicate a power density depression in the thermal spectrum of about three, from the rod surface to centerline, yielding centerline temperatures which are 100 to 200°F lower than those which would exist in an identical rod operating in a fast-spectrum reactor. The fuel surface thermal stresses are correspondingly greater in the thermal spectrum. Calculations indicate that the characteristics of the SRE thermal flux spectrum and the high coolant temperatures have advantages over other thermal reactors for testing fast-reactor fuels. For example, the power density depression is approximately two, for the SRE.

A study of the feasibility of bringing mixed-carbide fuel rods into the AI Headquarters assembly laboratories for testing and experiment-assembly has been completed and a preliminary draft (RSA-1) has been written. The conclusion of this study is that the risks of contamination do not preclude the operation as long as certain special precautions are taken.

III. EVALUATION OF EFFORT TO DATE

Assembly of the NAA-123 clad experiment has proceeded satisfactorily and there appears to be no technical reason why the experiment objectives cannot be met. By using a thermocouple for the control instrument in an out-of-pile mockup test, rather than the gas bulb thermometer planned for the in-pile test, temperature control of $\pm 8^\circ\text{F}$ was achieved. The sensitivity of the gas bulb thermometer is about four times that of a thermocouple and is expected to provide the $\pm 5^\circ\text{F}$ temperature control which is desired.

The initial resolution of handling small quantities of Pu at AI allows the possibility of purchasing small samples of fast-reactor fuels for both irradiation tests and for physical property measurements.

IV. NEXT REPORT PERIOD ACTIVITIES

Modification of the argon flow system on NAA-123 will be completed and temperature control within $\pm 5^\circ\text{F}$ should be demonstrated. Assembly of the experiment and the flux monitoring capsule will be completed.

This project was terminated. The activities will be continued under the High Temperature Materials Program.

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A Division of North American Aviation, Inc.

Program: Fast Reactor Development

Project: Fuel Irradiation

Project Engineer: B. R. Hayward

Reporting Period: July-September 1965

AEC Category: 04-01-04-19

General Order: 7661

Subaccount: 1981

I. PROJECT OBJECTIVES

The primary objective of this project is to evaluate the in-pile performance of the AI 24-1, AI 25-1, and AI 25-2 capsule irradiations, initiated under the Sodium Cooled Reactor Program. This objective will be accomplished by: in-pile surveillance of the AI 25-1 and AI 25-2 capsules; and postirradiation examination of AI 24-1, AI 25-1, and AI 25-2.

II. TECHNICAL PROGRESS DURING REPORT PERIOD

The irradiations of the AI 25-1 and -2 experiments at GETR were terminated on September 11, 1965. These hypostoichiometric UC experiments achieved 14,000 Mwd/MTU burnup at clad and fuel centerline temperatures of 1250 and 1850°F, respectively, and at a specific power of about 120 watt/gm U.

The NRX-86-1 and -2 long rod hypostoichiometric UC irradiations were terminated after a burnup of 13,000 Mwd/MTU and operation at design temperatures of 1800°F fuel centerline.

III. EVALUATION OF EFFORT TO DATE

The AI 25-1 and -2 experiments were discharged due to program redirection, and will be buried. The AI 24-1 experiment currently in storage will be buried. The two NRX-86 irradiations were transferred by AEC direction from the HNPF Fuel Surveillance program to the fast reactor program. These capsules will also be buried during the next quarter. No examinations are planned on these capsules.

IV. NEXT REPORT PERIOD ACTIVITIES

Reports will be completed on UC irradiations from the FY 1965 program. The irradiations terminated in this quarter will be buried.

SECTION

- II -

SODIUM-REACTOR EXPERIMENT
ADVANCED SODIUM-COOLED REACTORS
&
SODIUM-COMPONENT DEVELOPMENT

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program:	Sodium Reactor Experiment
Project Manager:	L. E. Glasgow
Reporting Period:	July-September 1965

I. PROGRAM OBJECTIVES

The primary objectives are to: (a) maintain the plant in a safe shutdown condition, (b) operate the systems necessary for testing of systems and components, (c) maintain accountability of Core III fuel, and (d) improve reliability of new sodium systems.

These objectives will be in effect until the reactor is restarted. At that time the objectives will be to: (a) operate the reactor at PEP design conditions, (b) evaluate long-term system characteristics, and (c) utilize the plant as a fuel irradiation facility.

II. PROJECT MANAGER'S SUMMARY AND EVALUATION

The preoperational testing, beyond that required for startup that could be done without loading fuel, has been completed. Safeguards evaluation work, operator recertification, and some system refurbishing work, disallowed under the PEP program, was completed. The SRE hot-cell was reworked and put on standby.

The SRE staff has been reduced to a minimum size consistent with the latest budgetary level and program objective requirements.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program: Advanced Sodium Cooled Reactors	
Project: Sodium Reactor Experiment Operations	
Project Engineer: J. E. Owens	
Reporting Period: July-September 1965	AEC Category: 04-01-04-22
General Order: 7602	Subaccount: 1310

I. PROJECT OBJECTIVES

The primary objectives are to: (a) safely maintain the plant in a shutdown condition, (b) operate the systems necessary for testing of systems and components, (c) maintain accountability of Core-III fuel, and (d) improve reliability of new sodium systems.

These objectives will be in effect until the reactor is restarted. At that time, the objectives will be to: (a) operate the reactor at PEP design conditions, (b) evaluate long-term system characteristics, and (c) utilize the plant as a fuel irradiation facility.

The specific objectives which support the above are to: (a) operate the plant with adherence to approved methods and procedures, (b) assist with plant modifications and maintenance, (c) generate, collect, and organize operational data, and (d) maximize efficiency and economy in the use of manpower and material through a well-organized operator training program.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

Operational support was provided for the SRE-PEP preoperational testing program. All sodium systems were filled and operated for determination of system hydraulic characteristics and function testing of components.

During operation of the sodium systems for testing purposes, the primary and secondary service systems were activated and the oxide content of all systems was reduced to less than 10 ppm.

III. PROGRESS DURING REPORT PERIOD

Upon completion of moderator element replacement, the equipment used for this program was decontaminated, painted, and properly stored.

The primary gallery shield blocks were installed and sealed after system testing and modifications were completed. The openings between the blocks were sealed utilizing a two-part styrofoam compound which expands to fill and seal the space.

A complete central print-file system is being established for the SRE and associated components. All necessary prints will be in a central location and clearly indexed as to number and function.

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Rewiring of the sodium service building heater panels has been initiated. The wiring was stripped from the panels and all multicolored extension wire removed and replaced.

Certification testing of all operations personnel was accomplished this report period. Examinations were administered encompassing plant changes made during PEP modifications.

IV. EVALUATION OF EFFORT TO DATE

Repair, replacement of broken or worn parts, and proper storage of moderator replacement equipment will reduce the time required for activation. As a result of decontamination and painting, the equipment may be stored in a building without need of radiation and contamination restrictions.

The use of styrofoam for sealing of vaults and galleries results in a reduction of nitrogen used to maintain the gallery atmospheres to less than the required 4% necessary to prevent fire in the event of sodium leak.

Establishment of a central print-file will provide easy access to needed material. Previously, needed drawings had to be ordered from a central file at another location. This caused unnecessary delays in work schedules.

Rewiring of sodium service heater panels reduces the time required for pre-heating of systems and recircuiting of heaters. Standard arrangement of wiring will reduce possibility of electrical shock to personnel working in the panels.

Certification of operations personnel assures safe, uniform operating methods and thorough understanding of equipment. Continued training will be given as new equipment or procedures are introduced.

V. NEXT REPORT PERIOD ACTIVITIES

All sodium systems will be operated for testing purposes.

Final gallery and vault closures will be made and the cooling and dehumidification systems operated for determination of cooling capacity and heat loads.

Work will be completed establishing a central print-file. Operator training will be conducted as required to familiarize operations personnel with new equipment and operating methods.

The reactor will be prepared for startup with an expected fuel loading date of January 1, 1966.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program: Advanced Sodium Cooled Reactors	
Project: SRE Operational Support	
Project Engineer: J. E. Owens	
Reporting Period: July-September 1965	AEC Category: 04-01-04-22
General Order: 7602	Subaccount: 1320

I. PROJECT OBJECTIVES

The primary objectives of this project are to: (a) provide a preventive maintenance program, (b) provide for breakdown maintenance as required, (c) update standard operating procedures, (d) provide radiological health and safety support for SRE personnel, (e) provide for hot-cell support as required, and (f) provide an updated operator training manual.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

Modification of the primary sodium service system was completed. Significant changes were made to separate piping for the inlet to the hot-traps and cold-traps, and installation of a sample tab exposure facility.

The main primary pump case vent line was modified to provide adequate drainage and ensure gas communication between the reactor and primary pump cases.

III. PROGRESS DURING THE REPORT PERIOD

Modification of the primary sodium service system was completed. Changes include the addition of separate inlet lines for the cold-trap and hot-traps, a facility for exposure of metal samples to primary coolant, conversion of all valves to the freeze-stem bellows-backup type, and enclosure of all wiring in terminal boxes or conduit.

Cleanup of the SRE hot-cell was completed. All waste accumulated during Core II operation and all operating equipment except the remote manipulators were removed. The cell and associated work areas were then decontaminated to acceptable levels and painted.

Radiological health and safety support was provided for SRE personnel during: modification of the primary sodium service system, cleanup of the SRE hot-cell, and transfer of core components to and from the reactor.

Modification to the main primary pump case and associated piping was completed to permit operation of the pump at flow rates necessary to achieve design conditions of the SRE-PEP reactor.

The plant instrument air system modifications were initiated. Two small compressors were replaced with a temporary compressor of larger capacity providing a working pressure for valves and instruments of 80 to 100 psig.

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Preventive maintenance was performed on a minimum amount of equipment. Because components had been operated only for testing purposes, minimum maintenance was required.

IV. EVALUATION OF EFFORT TO DATE

Separation of the inlet piping to the cold-trap, and hot-traps, and installation of a tab exposure facility allows increased flexibility of operation. An inlet temperature limitation of 700° F prevented operation of the cold-trap when the reactor was at full power. A line was installed from the reactor return line (which never exceeds 700° F) to the cold-trap permitting operation of the service system, irrespective of reactor power. The tab exposure facility precludes the necessity for shutdown of the hot-traps for removal of samples which were previously exposed in the inlet and outlet lines of the hot-traps.

Cleanup of the SRE hot-cell removes a contamination and radiation hazard from the SRE facility. The cell decontamination and painting will permit the tooling for future work to be installed quickly and with a minimum radiation exposure to personnel.

Radiological health and safety support provided for operations and maintenance activities permitted completion of scheduled activities involving radiation and contamination without unnecessary delay. The insignificant exposures to personnel reflects the safety aspects of the procedures used and the close cooperation between Health and Safety and performing personnel.

The main primary pump case overflow line was modified to include a throttle valve for control of the pump case sodium level. Removal of a low section of piping ensures gas communication between the pump cases and reactor.

The instrument air system installed during PEP modifications was inadequate to supply the requirements to the facility. Existing equipment located at other facilities was used to increase the working pressure to the required level.

V. NEXT REPORT PERIOD ACTIVITIES

The main primary pump case overflow line valve installation will be completed and placed in operation.

Modification of the instrument air system will be completed upon arrival of the replacement air compressor in December. Additional in-core heaters will be fabricated to replace the present elements which are not usable with the SRE-PEP core.

Spare control and safety elements will be assembled, checked out, and placed in storage cells for use if required.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Sodium Cooled Reactors	
Project: SRE Engineering	
Project Engineer: J. E. Owens	
Reporting Period: July-September 1965	AEC Category: 04-01-04-22
General Order: 7602	Subaccount: 1330

I. PROJECT OBJECTIVES

The objective of this project is to provide engineering and technical support necessary to operate the SRE-PEP. This specifically includes: (a) performance and evaluation of all tests, (b) determination and analysis of plant performance, (c) design engineering studies for plant improvements, and (d) development of advanced testing and analysis techniques.

The purpose of the project is to provide engineering information required to determine and improve SRE safety, reliability, and performance.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

Preoperational testing beyond that needed for startup of the SRE was completed. Tests of the control room instrumentation, systems, and components in all cases met or exceeded the established acceptance criteria.

Analysis and modifications necessary to eliminate gas entrainment in the main primary pump were completed.

A detailed test-procedure was prepared for operation of the present SRE core with vented fuel elements. The experiment would establish the practicality and economics of operating vented fuel in sodium-cooled fast reactors, and provide engineering data based on good statistics for design of any sodium-cooled vented fuel system.

III. PROGRESS DURING REPORT PERIOD

All of the preoperational testing that can be done without loading fuel required for startup of the SRE was completed. These tests include the fill and function check of the SRE-PEP sodium systems, sodium component vibration checks, and the control room instrumentation checkout. Evaluation of the test results was completed and documented. In summary, all system performance with the exception of main primary system gas entrainment, met the established acceptance criteria set forth in the test procedure summaries.

Detailed testing established that the main primary gas entrainment occurred through the main free surface pump and its associated overflow dip-leg piping. The specific cause was a low pump sodium level coupled with high sodium velocity in the overflow dip-leg. The low sodium level caused surface turbulence and vortexing in the pump barrel to carry gas into the pump overflow. The high velocity in the overflow dip-leg did not allow the entrained gas to separate and the high velocity sodium falling in the dip-leg entrained more gas by aspiration.

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Three minor system modifications have been made which correct the entrainment and allow gas free operation at SRE-PEP conditions. These modifications are: (a) installation of a cap in the dip-leg vent pipe to eliminate the source of gas causing the aspiration effect, (b) installation of a dip-leg control valve to raise the pump sodium level above the overflow nozzle, and (c) modification to the pump to minimize turbulence in the sodium pool.

An experiment was proposed of operating the existing UC core loading for SRE-PEP as a "vented fuel" system under controlled conditions. The experiment consists of three separate tests. The first, using eight fuel rods vented through the loading face shield by means of tubing, would give an accurate measurement and identification of the fission gas released inside the fuel cladding. The second test, comprising two rods located in a sodium filled thimble, would give a measurement of the gas release rates to the sodium and the cover gas. The final test would consist of a controlled, increasing number of fuel rods venting directly to the sodium; this would provide the system effects which in turn would be related back to the release rates. This inter-test correlation would yield the necessary information for extrapolation to other reactors.

A test was performed to determine the consequences of overheating the O-ring seal at the loading face during fuel handling operations. An irradiated fuel element was simulated by using electrically heated cladding. The test showed that if a fuel element were stuck at the O-ring, the O-ring temperature would exceed 500°F and probably cause failure unless mitigative steps are taken. The fuel cladding temperature would not exceed 1300°F and therefore would not be damaged.

Modification of the Mark II fuel handling machine was completed. Changes made include a revised interlock system, pneumatic hoists for the biological shield, and a revised gas system.

Fission product monitor modifications were completed. Existing components were replaced with solid state electronics, sample piping was changed, and the readout equipment was installed nearer the control room.

As-built drawings were completed for all systems affected by PEP modifications. Old prints of existing systems were also reviewed and revised.

IV. EVALUATION OF EFFORT TO DATE

The preoperational tests were successfully completed on schedule for the original reactor startup date. The performance of these tests by use of the new test procedure summary together with the detail test procedures was effective in determining the system performance in an efficient and safe manner. The overall preoperational testing effort has uncovered a number of minor problem areas in sufficient time to permit correction, thus minimizing delays during operation to full power.

The modifications to the main primary system have eliminated gas entrainment and the system presently meets the acceptance criteria.

The vented fuel experiment would establish the practicality and economics of operating venting fuel in sodium-cooled fast reactors, and provide engineering

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data based on good statistics for design of any sodium-cooled vented fuel system. The detailed experiment has been designed so that no major safeguards studies or revisions to the existing safeguards report would be required. Likewise, no system additions or modifications would be required except for inexpensive fuel element changes. Equipment and procedures for identifying, measuring, storing, and disposing of the vented gases exist and are ready for service.

The test results from the simulation of a stuck fuel element in the fuel handling machine indicate that a large release of fission products from damaged fuel cladding will not occur. It is anticipated that the failure of the O-ring from overheating will not increase the core cover gas leakage above the 45% per day operating limit. Therefore, the consequences of such a postulated accident do not represent a hazard.

The completed modifications to the fuel handling machine and the fission product monitor were the last major modifications required for steady full power SRE-PEP operation.

V. NEXT REPORT PERIOD ACTIVITIES

Instrumentation will be prepared for testing during startup and operation of the reactor.

Testing of the gallery cooling systems will be done after final vault and gallery sealing is completed.

Because of recent AEC direction, no further work will be done on vented fuel elements. Engineering support will be limited to that necessary to 1200° F operation.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Sodium Component Development	
Project: Operation of Sodium Component Test Installation	
Project Engineer: J. A. Leppard	
Reporting Period: July-September 1965	AEC Category: 04-01-04-31
General Order: 7593	Subaccount: 1322, 1323

I. PROJECT OBJECTIVES

The project objective is to operate the Sodium Component Test Installation (SCTI) in order to test, obtain data for, and evaluate the performance of prototype intermediate heat exchangers and steam generators.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

The entire SCTI system was operated under isothermal conditions at a temperature of 350°F. Several failures of major equipment occurred; solutions to these problems have been obtained.

III. PROGRESS DURING REPORT PERIOD

Late in the last quarter, the drive motor for the secondary pump burned out. The mode of failure was a short between two coils and the stator. The motor was rewound, satisfactorily tested at the factory, and reinstalled in its operating position. High potential (d-c) tests performed in this location proved that the insulation was inadequate for the application; namely, operating a 4160-v machine in an argon atmosphere. The motor was returned to the repair shop where additional insulation was placed on the coil ends, and certain metallic hardware was replaced with electric materials. The motor was again returned to its operating position and successfully run for approximately 5 days. During a time when it was running at about 25% of full load, it burned out again. On this occasion the failure was a phase-to-phase short between a lead and adjacent coil connections. This incident is the fifth failure of this motor in a total operating life of less than 3000 hr. No further effort will be made to use this particular motor.

In order to apply an air-cooled motor to the secondary pump, it is necessary to change the configuration of the pump case. A contract has been let to Byron Jackson Company to construct a new pump case closure, jack shaft, bearing housing, and shaft seal assembly. Integral with this pump case closure will be a new mount for an air-cooled motor of standard design. A used motor, available from Borax V in Idaho, was loaned to the SCTI for this application. This motor is currently being examined and serviced. Additionally, bids are being requested for a new standard design motor specified for the requirements of this application.

Nearly coincident with the secondary pump motor failure, a bellows pipe expansion joint in the primary sodium piping leaked. Examination of the joint revealed that the stainless steel material was sensitized and had subsequently undergone massive intergranular corrosion. Photomicrographs of sections of

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both the inner and outer plies showed that both the interior and exterior surfaces had corroded about the same amount. Because sodium does not cause this type of effect, it seems that the corrosion occurred prior to sodium filling. The specification for these bellows has been revised to eliminate a high temperature bakeout which may have given rise to the sensitization to the bellows.

A second bellows expansion joint was cut out of the piping, cleaned, and examined. It was determined by a dye-penetrant technique that no cracks or corrosion pitting existed on the interior surface of the bellows. Appearance of this second bellows was excellent, and no corrosion was evident. This joint has now been reinstalled in the primary piping. An order has been placed for six spare bellows expansion joint assemblies. This will yield a 50% replacement capability. Leak detectors are being installed on all of the bellows expansion joints.

Because of the failure of several bellows seal valves, these valves are being modified to have freeze-stem seals. All but three of the total of 25 have now been modified.

The body of a plastic valve (polyvinyl chloride), located immediately adjacent to the demineralized water storage tank, split open. This valve and two other similar valves in lines to this tank have been replaced with stainless steel valves. The plastic pipe lines associated with the demineralized water system are supported on the same structure which supports the main circulating system piping. Vibration transmitted from the main circulating pump and the instrument air compressor induced considerable vibration in these lines. Action was taken to both dampen and isolate the vibrations from the plastic piping.

A 2-in. bypass line was installed around the primary plugging meter assembly to decrease the pressure differential across the plugging valve. Also the entire assembly was relocated to provide better channeling of the oxide to the plugging valve.

A leak developed in the main instrument air compressor lower head gasket, which necessitated shutting down the compressor for repair. In addition to repairing the defective gasket, the entire unit was disassembled, inspected, and worn parts replaced. An order has been placed for a new instrument air compressor of larger capacity than the existing unit. The present air compressor will be used as a standby unit in preference to the present standby unit, which is inadequate.

The bubbler-type level controller for the primary sodium pump was modified in order to assure its proper operation. After making several seemingly minor changes, this unit functioned properly.

A meeting was held with a representative of Baldwin, Lima, Hamilton to review the ALCO steam generator and IHX test program. A letter was prepared summarizing the results of this review and defining the mutual recommendations of AI and BLH for revisions to the program described in APAE 122. These recommendations included reduction of the number of test points in the steady state series (without losing the engineering value of the program), and the addition of several test conditions not specified in that report.

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A draft was completed of the step-by-step test procedure to be used by the operating crew in performing the first series of ALCO steam generator and IHX tests. The first test-completion report of the series to be prepared on the pre-operational test program was written.

Preventive maintenance activities during this period included replacement of bearings in one of the preheat air compressors and in the circulating water pump. In the case of the circulating water pump, a grease seal on the original bearing was missing; this fact probably led to the early failure of the bearing.

All corrective action procedures were completed and the rest of the plant performance test procedures were finished and reviewed by the Steam Generator Coordinating Committee. The plant description section of the Operating Manual was revised and issued to each person on the staff.

IV. EVALUATION OF EFFORT TO DATE

Completion of the preoperational test program and the start of the test performance work has been seriously delayed because of equipment failures experienced during this period. However, valuable experience and information have been gained in obtaining solutions to these problems. Completion of all of the required plant performance procedures and most of the ALCO test procedures will enable the program to proceed immediately after mechanical equipment is fully operative.

V. NEXT REPORT PERIOD ACTIVITIES

Revisions to the secondary pump will be completed. The failed bellows expansion joint will be replaced. Replacement of bellows seal valves with freeze-stem seals will be completed. The rest of the preoperational tests will be performed and the plant performance tests initiated.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program: Sodium Component Development	
Project: High-Temperature Strain Measurements	
Project Engineer: J. E. Owens	
Reporting Period: July-September 1965	AEC Category: 04-01-04-31
General Order: 7593	Subaccount: 1328

I. PROJECT OBJECTIVES

The objective of this project is to develop suitable, reliable, high-temperature strain gages for experimental stress or strain measurements, or for application in process instrumentation components. Commercially available gages will be tested to determine their limits, applicability, and reliability as analytic instruments for use on component test loops and systems, and as sensing elements in pressure transducer and force-arm applications. Suitable high-temperature strain gages will be developed, as well as application techniques for high-temperature use. Methods of accomplishing temperature compensation and sealing of the sensitive element will be investigated to permit use in contact with sodium or sodium vapor.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

Pilot evaluation of the 1200° F Armour D-Platinum, 900° F Microdot weldable half-bridge gage and the B-L-H, 1200° F platinum-tungsten wire gage was completed. Fixtures and techniques for winding special high-temperature gages were further perfected. New approaches for improving gage reliability were also studied.

III. PROGRESS DURING REPORT PERIOD

A. STRAIN GAGE EVALUATION

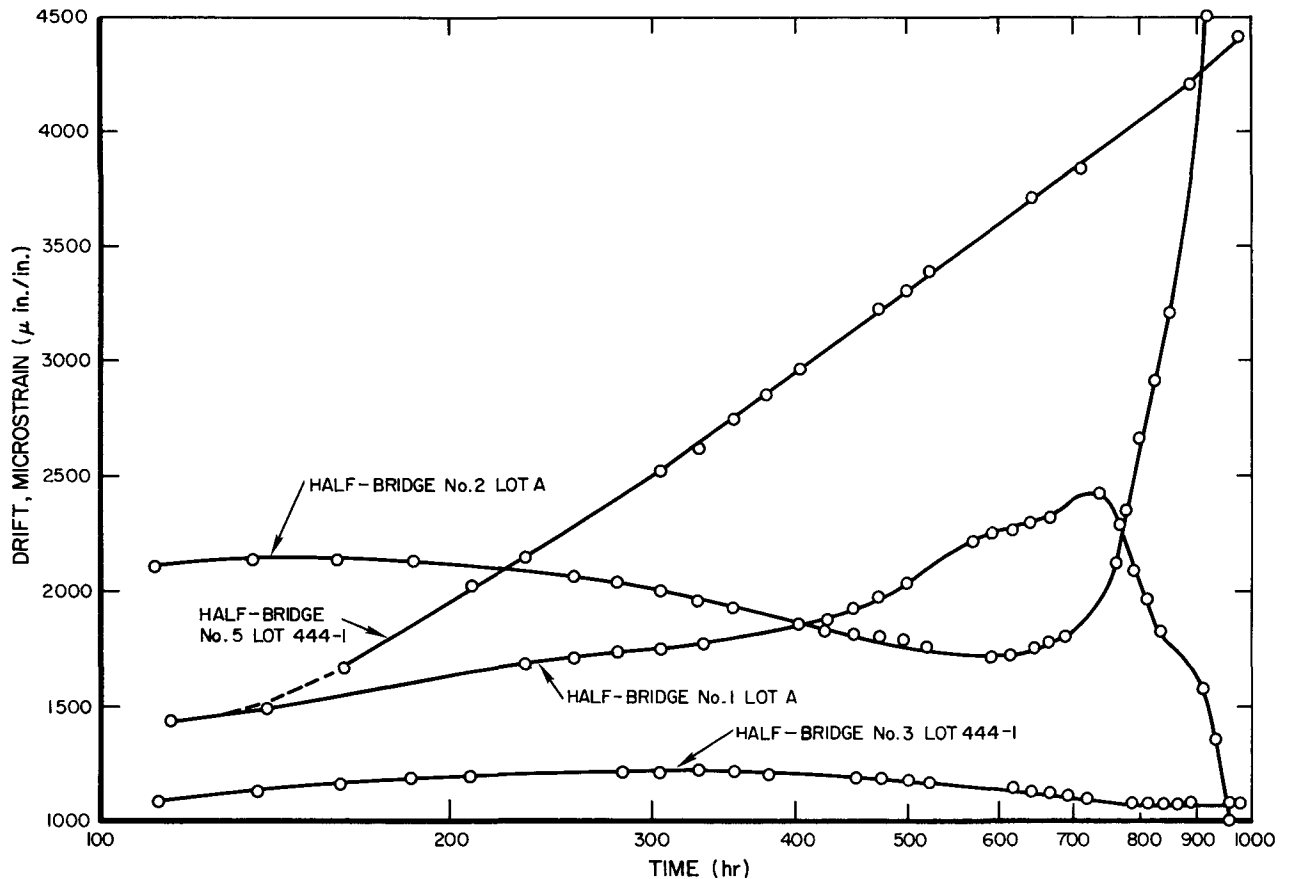
1. B-L-H Type HT-1212-5A Strain Gage

Pilot studies on this gage have been completed. Earlier studies of the performance of this gage indicated that the gage had excellent performance with respect to drift characteristics, under steady-state conditions at temperature up to 1200° F in an air environment. After 1675 hr of exposure at 1200° F, the drift rate was still constant, with only 0.5 microstrain per hr. However, upon observing its performance under transient heating conditions, at heating rates as low as 5° F/sec, it was found that the gage would open (fail) electrically. The steady-state performance of the gage was sufficiently attractive to justify further evaluation of the gage. A careful examination of the cyclic strain versus temperature oscillographic records and photomicrographs of the joint between the gage filament and the nickel gage tabs indicated that the spotweld was responsible for the erratic gage behavior under transient conditions. Welding techniques used in making the spotwelds for these gages were subsequently changed and gages were procured with Nichrome V, Evenohm, and Alloy 479 ribbon tabs, all tabs spotwelded to the gage filament using the new welding procedures. It was hoped that the change in welding technique and/or tab material

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would result in a satisfactory gage for transient work. The table summarizes results of tests on four Type 304 stainless steel specimens, each instrumented with 4 flame-spray-bonded (Al_2O_3) HT 1212-5A gages. Each specimen was stabilized in an air furnace at 1225°F for 16 hr prior to transient testing in a special transient heating test facility. Each specimen was subjected to 10 cycles at 5°F/sec, followed by 10 cycles at 10°F/sec, followed by 10 cycles at 25°F/sec, all cycling being between 325 and 1150°F. These heating rates bracket most of the heating rates anticipated during sodium component loop testing.



7593-2505

Figure 1. Drift Characteristics of B-L-H Strain Gages

Results from the steady-state pilot drift tests conducted at 1200°F for periods up to 1000 hr for four gages are shown in Figure 1. The gages were flame-spray-bonded to a Type 304 stainless steel specimen and stabilized for 16 hr in an air furnace, before commencement of steady-state (drift) testing. Gages were selected from two different lots. These gages, which are not self-temperature compensating, were hooked up in a half-bridge arrangement, which is the manner in which these gages would normally be used in evaluation of sodium components, where both steady-state and transient behavior are investigated.

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SUMMARY OF TRANSIENT RESULTS

Specimen No.	Gage No.	Number of Shocks Withstood, Without Erratic Behavior, at			Lead Tab* Type	Remarks
		5°F/sec	10°F/sec	25°F/sec		
B1	1	10	10	10	P	No erratic behavior throughout test
	2	4	1	0	P	Started opening intermittently on 5th cycle at 850°F
	3	10	10	1	N	Started opening intermittently on 21st cycle at 900°F
	4	10	5	0	N	Started opening intermittently on 15th cycle at 900°F
B2	1	10	10	10	P	No erratic behavior throughout test
	2	9	0	0	P	Started opening intermittently on 9th cycle at 1100°F
	3	9	0	0	N	Started opening intermittently on 9th cycle at 1100°F
	4	1	0	0	N	Started opening intermittently on 1st cycle at 1100°F
B3	1	10	10	4	P	Started opening intermittently on 24th cycle at 800°F
	2	10	9	0	P	Started opening intermittently on 19th cycle at 800°F
	3	8	0	0	N	Started opening intermittently on 8th cycle at 1000°F
	4	-	-	-	N	Damaged during installation
B4	1	2	0	0	E	Started opening intermittently on 2nd cycle at 800°F
	2	3	0	0	E	Started opening intermittently on 3rd cycle at 750°F
	3	10	1	0	E	Started opening intermittently on 11th cycle at 900°F
	4	1	0	0	E	Started opening intermittently on 1st cycle at 600°F

*Legend

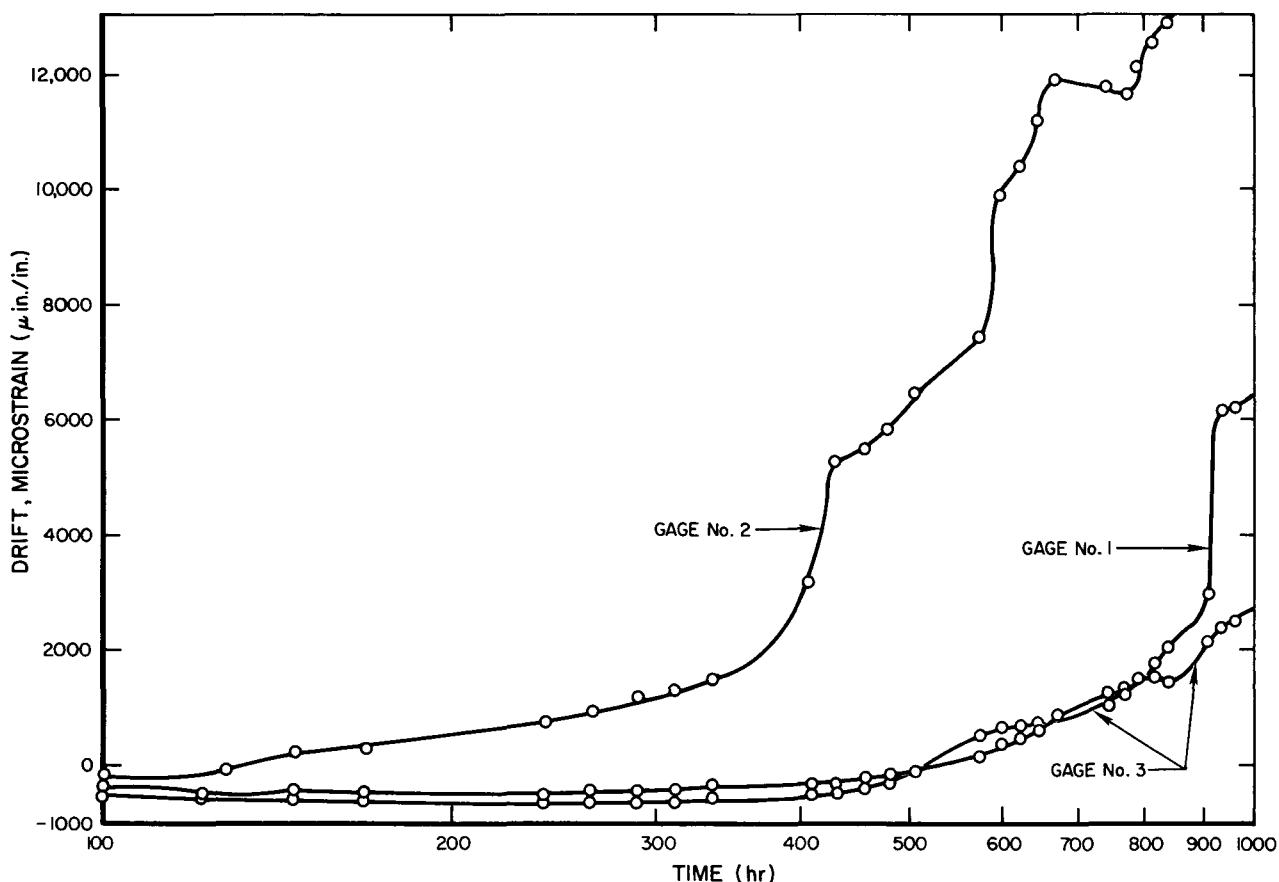
N = Nichrome V
E = Evenohm
P = Alloy 479

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2. Armour D-Platinum Strain Gage

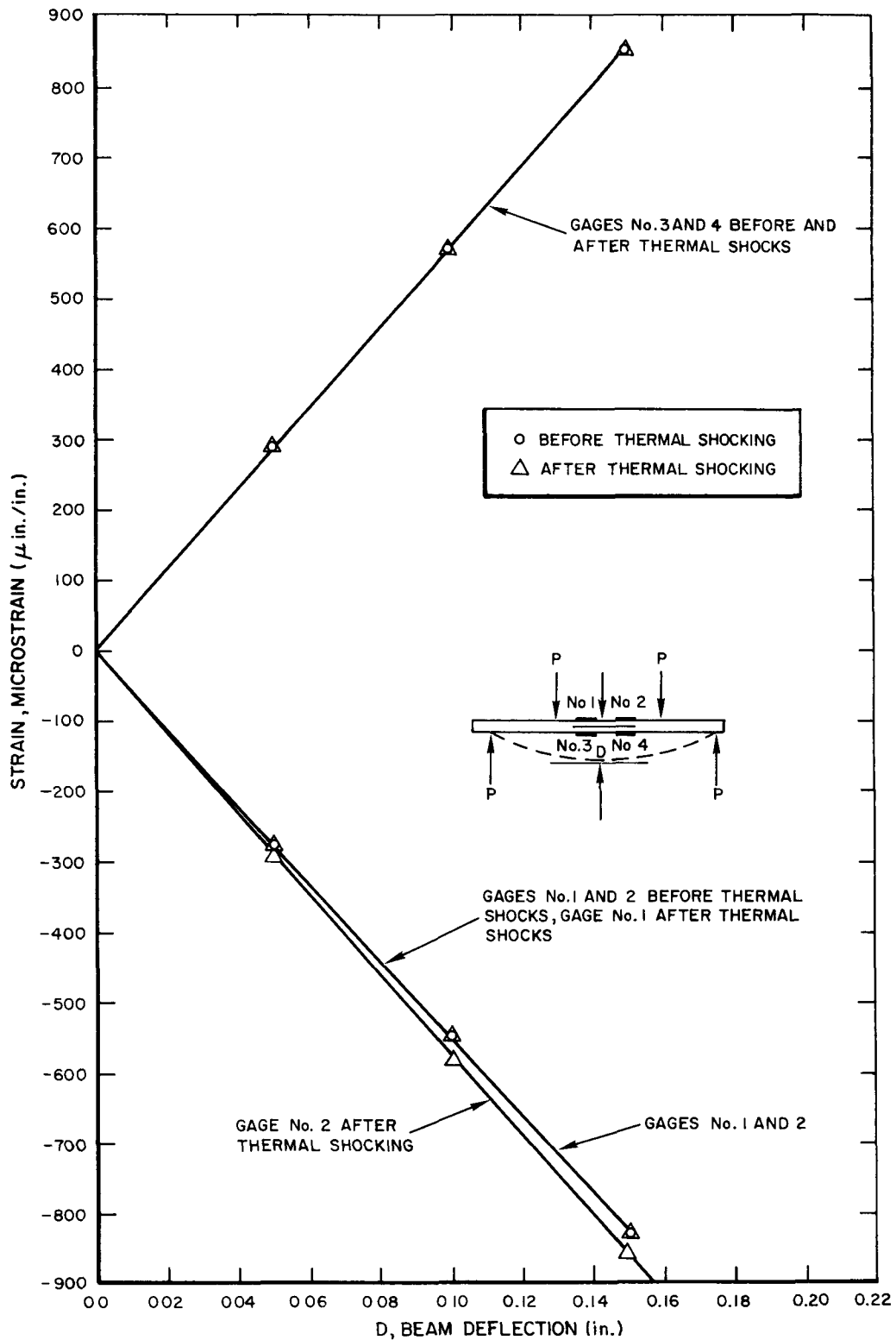
The characteristics of this gage, under steady state and transient conditions, were investigated. The gages were evaluated, individually, since they are self-temperature compensating and need not be used in a half-bridge arrangement. Figure 2 shows the drift versus time curves for three Armour D-Platinum gages, flame-spray bonded to Type 304 stainless steel bars. To determine the effect of prior transient history, Gage 1 was subjected to a total of 60 shocks from 800 to 1200°F, consisting of 10 shocks at 6°F/sec, 10 shocks at 14°F, and 10 shocks at 20°F/sec, 10 shocks at 27°F/sec, 10 shocks at 37°F/sec, and 10 shocks at 53°F/sec. Gages 2 and 3 were not shocked.



7593-2506

Figure 2. Drift Characteristics of A D-P Strain Gages

Additional shock-testing was performed on a similar test specimen with four gages. The bar was subjected to a total of 30 shocks from 325 to 1150°F, consisting of 10 shocks at 5°F/sec, 10 shocks at 10°F/sec, and 10 shocks at 25°F/sec. Three out of four of the gages satisfactorily withstood all thermal shocks, without any erratic behavior. The fourth gage withstood a total of 22 shocks satisfactorily. However, during the third cycle at 25°F/sec, it began to exhibit the same erratic behavior as the B-L-H type HT 1212-5A gage.



7593-2507

Figure 3. Thermal Shock Characteristics

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The three gages which withstood the thermal shocks were then evaluated with respect to the effects of thermal shock on their strain response (linearity, etc). The evaluation was performed in a special "constant-moment" beam test apparatus, which is contained within an air furnace. The gages were mounted back-to-back on opposite faces of the bar. All gages were within the constant moment portion of the beam. Theoretically, Gages 1, 2, 3, and 4 should be of the same magnitude, with Gages 1 and 2 of opposite sign to that of Gages 3 and 4. Figure 3 shows the strain versus beam deflection curves at room temperature, before and after thermal shocking. It is noted that Gage 4, which showed erratic behavior during the thermal shock tests after a total of 22 cycles, responded satisfactorily - at room temperature - as did the other gages with respect to strain response. Similar characteristics were observed at 1200°F for Gages 1, 2, and 3. Gage response was quite linear and similar to that shown in Figure 3, except for a small difference in gage factor. Gage 4 was found inoperative at 1200°F, because of an electrical opening which developed at elevated temperature, after thermal shocking.

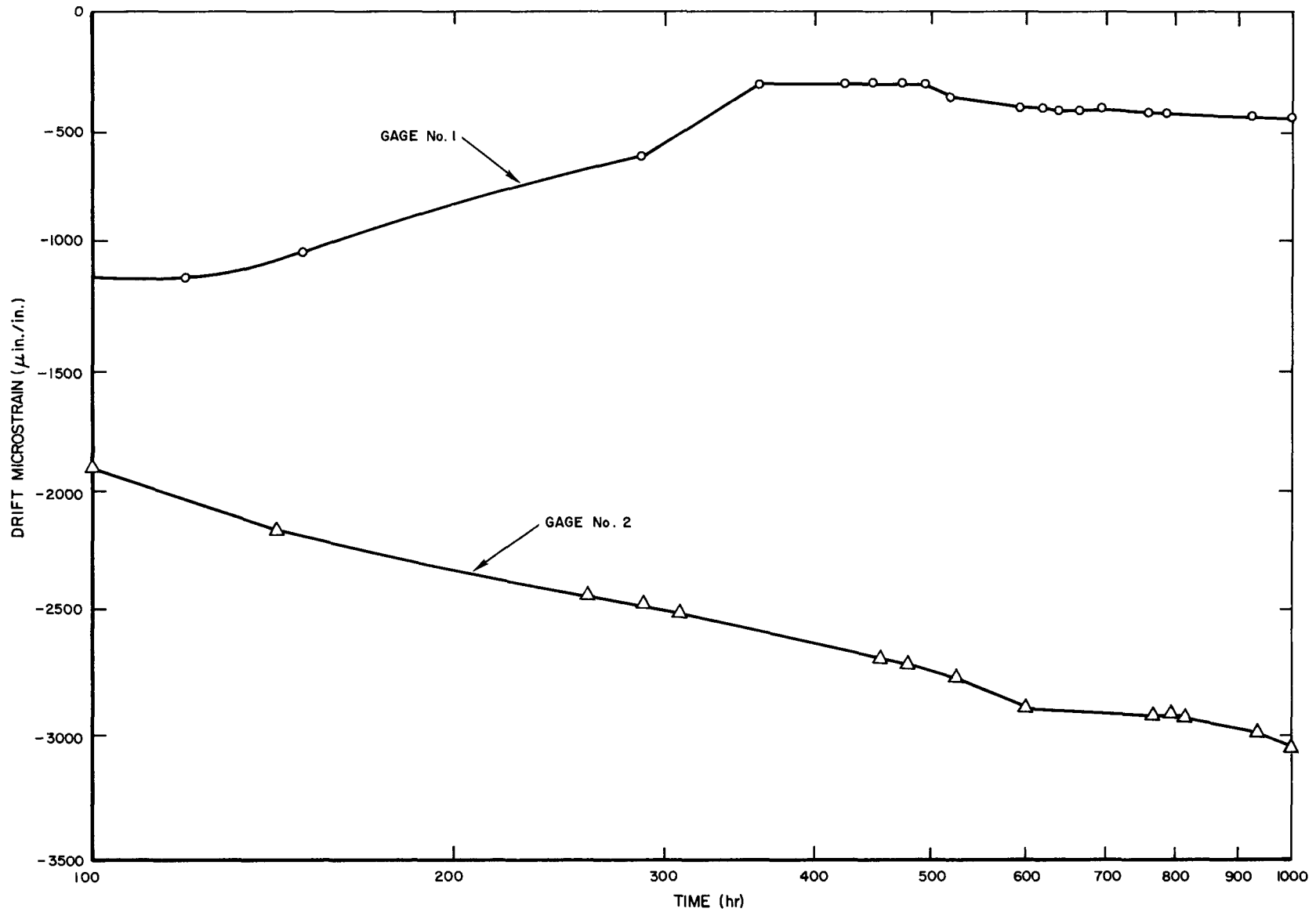
3. Microdot Type SG-420 Strain Gage

Steady state and transient tests were conducted on this gage which is a completely encapsulated weldable, half-bridge gage, with a maximum steady state operating temperature of approximately 950°F. This gage was selected for evaluation because of its potential for use in relatively high-temperature steam environments or under field conditions when moisture or corrective action over extended periods of time might prohibit the use of nonencapsulated gages. Figure 4 shows the drift characteristics of two of these gages in an air furnace at 900°F. These gages were spotwelded to Type 304 stainless steel specimens. Drift testing on these gages was discontinued after they had been subjected to slightly more than 1000 hr.

After conducting steady state drift test on Microdot Gage 1, the gage was subjected to 107 thermal shocks in the temperature range of 500 to 900°F, consisting of 11 shocks at 22°F/sec, 11 shocks at 33°F/sec, 22 shocks at 40°F/sec, 40 shocks at 50°F/sec, and 23 shocks at 60°F/sec, with no erratic behavior noted. Prior to conducting the steady state drift test on Microdot Gage 2, the gage was subjected to 58 thermal shocks in the same temperature range as for Gage 1, consisting of 11 shocks at 9°F/sec, 11 shocks at 30°F/sec, 12 shocks at 40°F/sec, and 12 shocks at 50°F/sec. The soak-shock sequence for these two gages was reversed to determine the effect, if any, of prior shocks on drift characteristics.

B. STRAIN GAGE DEVELOPMENT

Existing fixtures and techniques for strain gage fabrication were modified to facilitate fabrication of gages of various sizes and shapes, including the Armour D-Platinum strain gage. Elements for approximately 20 Armour D-Platinum gages have already been fabricated in this fixture. Gages from these elements have been fabricated, and four sample gages are in the process of being evaluated with respect to steady state and transient performance. These gages were fabricated by use of spotwelding. Figure 5 is a schematic representation of the Armour D-Platinum gage. It is noted that four spotwelded joints exist in the present design. The symmetrical configuration with two platinum elements is used to assure temperature compensation in locations where transverse temperature gradients exist across the gage width.

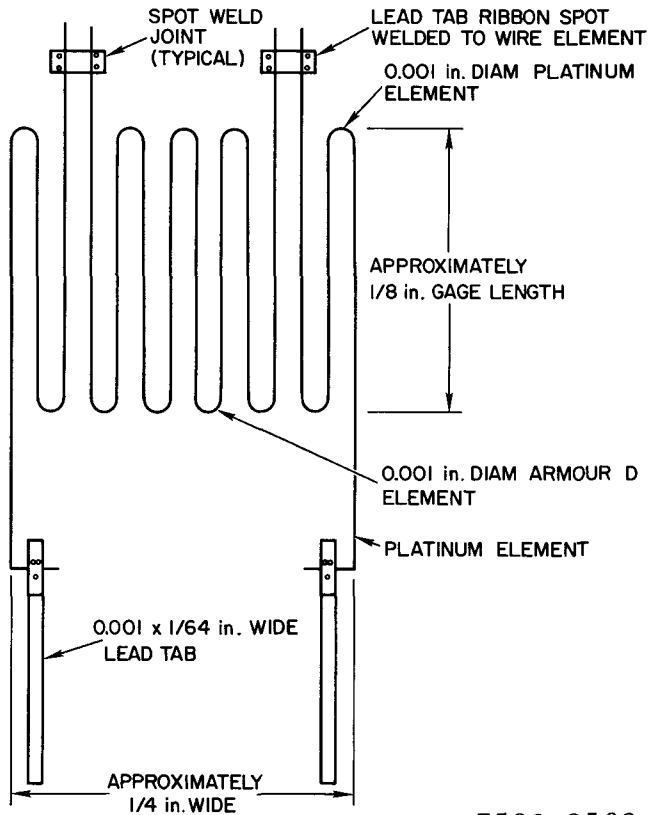


7593-2508

Figure 4. Characteristics of M. Gage

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7593-2509

Figure 5. Installation of A. D-P Strain Gages

Successfully flame-spray-bonded without damage to the filament, which in conventional foil gages is very thin (approximately 0.0002 in. thick) and has a large projected area. Specifically, gages are being fabricated from thicker foil, having a reduced projected area. Because of the thinness of the foil and the large projected area, conventional metal film gages are quite susceptible to damage during the flame-spray process. By using foil, rather than wire, more reliable and uniformly consistent weld joints should result, since it is obviously easier to spotweld two relatively large flat surfaces together than it is to "spot" a weld or welds on a 0.001-in. -diameter wire. Further, making the lead tab and one element of the gage from one piece of foil eliminates the need for separate lead tabs and the weld joint required when a separate lead tab is used.

IV. EVALUATION OF EFFORT TO DATE

Based upon results of the effort to date, there are no reliable off-the-shelf, commercially available electric resistance strain gages that can be flame-spray-bonded for use in evaluating structural behavior of sodium components, where the temperatures imposed are in excess of 900°F and the heating rates in excess of 5°F/sec. If the joints of the HT 1212-5A gage can be improved, it may then be a satisfactory gage for both transient and steady state testing for periods of time up to at least 500 hr, provided that it is used in conjunction with a dummy gage. However, until then, it is recommended that it not be used at elevated temperatures for heating rates in excess of 5°F/sec.

In conjunction with the preceding effort, several ways of improving gage reliability and uniformity from gage to gage are being investigated, with particular emphasis on improvement of the reliability and uniformity of the joints which are presently spotwelded. Because of the small 0.001-in. diameter of the wire, extreme precision is required in making gages which are consistently uniform, from an electrical standpoint, and which are capable of functioning reliably, under transient heating conditions, in the 500 to 1200°F temperature range and heating rates greater than 5°F/sec.

As a consequence, the use of other joining methods including brazing and welding techniques used in making microminiature electronic components is being evaluated. Full use will be made of the extensive technical "know-how" and experience of the Autonetics Division of North American Aviation.

In addition to the development of ways of improving wire element gages, consideration is being given to ways of modifying the geometry of foil (metal-film) gages so that they may be suc-

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Below 900°F, the Microdot gage has exhibited very good performance both under steady state and transient heating conditions. The main drawbacks to this gage are (a) its relatively large gage length – approximately 9/16 to 3/4 in.; (b) its reinforcing effect on thin-shell structures; and (c) its relatively high cost – approximately 10 times that of other commercially available gages. However, where strain gradients are not steep and thin structures are not involved, these drawbacks are not considered serious.

The Armour D-Platinum gage, which must be custom-made, appears to be the only gage that can withstand both the steady state and transient time-temperature histories associated with sodium component loop testing, with a present restriction on service life at 1200°F of approximately 500 hr.

Before a reliable and uniformly consistent 1200°F self-temperature compensating gage can be developed and produced, it will be necessary to continue pilot fabrication of the Armour D-Platinum gage, incorporating changes in joint design, joining techniques, fabrication techniques, or perhaps going over to the foil type Armour D-Platinum gage. Unless this is done, the variation in gage to gage behavior may be too large for many stress measurement applications.

An examination of Figures 1 and 4, for example, indicates a substantial difference in drift characteristics in "production-run" commercial gages. Once the reason(s) for this variation is clearly established, it should then be possible to produce gages that are uniformly consistent, within specified engineering tolerances and accuracy.

In Figure 2, one out of three of the Armour D-Platinum gages exhibits a drift rate that differed substantially from that of the other two, which essentially had a truly zero drift rate up to approximately 400 hr at 1200°F. The fact that it was possible to fabricate two of three gages with essentially zero drift rate, using the same materials and fabrication techniques, strongly suggests that by changing or improving the joint design and paying meticulous attention to each step of the fabrication, it should be possible to eliminate the variable(s) responsible for this variation, and to produce gages having both a high degree of uniformity and reliability.

Interestingly enough, the fabrication factor(s) responsible for the difference in drift rate, did not introduce material differences in the room temperature strain response or linearity of any of four gage samples that were fabricated from wire from the same spools and using the same fabrication technique as evidenced from the curves in Figure 3. This suggests that time and temperature have a very pronounced effect on the variable(s) responsible for gage-to-gage variations.

V. NEXT REPORT ACTIVITIES

During this period, no further evaluation of the B-L-H 1212-5A will be carried out, unless the manufacturer makes substantial improvements in the performance of this gage under transient heating conditions. Additional evaluation of the Microdot SG 420 gage will be carried out primarily to establish statistical information from which life probability and confidence levels can be derived. Particular emphasis will be devoted to further improving the gage-to-gage uniformity of the Armour D-Platinum gage. The performance of this gage is believed better than any commercial gage for use at temperatures up to 1200°F,

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where both steady state and transient conditions exist. Specifically, new joining techniques and joint designs will be developed and pilot gages fabricated and evaluated, in order to determine the best type of joint for the wire filament Armour D-Platinum gage. It is also anticipated that some results on the feasibility of fabricating Armour D-Platinum gages from foil will be forthcoming by the end of this period.

Concurrently with the above effort, a systematic investigation will be undertaken to determine the optimum time-temperature history for prestabilizing the Armour D-Platinum gage, with respect to apparent strain, drift rate, strain response, gage factor, leakage to ground, linearity, repeatability, and resistance to thermal shock. Compilation of statistical data for this gage will not get under way until the optimum joint design and configuration of this gage are established.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program: Sodium Component Development

Project: Mechanisms in Sodium

Project Engineer: J. E. Owens

Reporting Period: July-September 1965

AEC Category: 04-01-04-31

General Order: 7593

Subaccount: 1329

I. PROJECT OBJECTIVES

The objective of this project is to evaluate materials, configurations, and surface treatments for use in high-temperature liquid sodium or sodium vapor. Information generated on this project will be applicable to the designs of mechanism such as control elements, handling equipment, and variable orifices which must operate in sodium-cooled reactors.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

Tests have been designed and equipment partially fabricated to provide information on candidate materials for use as bearings and gears in 1200° F sodium.

The testing of a third Bell-Metrics type 316 stainless steel bellows has been completed.

III. PROGRESS DURING REPORT PERIOD

A material evaluation task has been initiated to provide general performance data on candidate materials for use in bearings and gears operating in sodium at 1200° F. Two identical test assemblies which had been built under the LSGR program (7519-4450) in FY 1963, on which a limited number of quantitative tests have been performed, are in the process of reconditioning. These assemblies have been designed to test a set of meshed spur gears in conjunction with two journal bearings under controlled load conditions. The equipment has been modified to permit sodium sampling to determine the sodium oxide concentration. This information is required because qualitative tests performed in FY 1963 showed that for mechanisms in sodium, friction factors for some of the materials tested are substantially influenced by the sodium oxide concentration.

A linear bearing test has been initiated with a fixture which had been used in the evaluation of linear seals for liquid sodium service. The tests had been unsuccessful because the seals passed a film of sodium which vaporized when the shaft moved outboard of the seal. The seal materials of tungsten carbide on Inconel X base and tungsten over 304 stainless steel were found to be compatible at 450° and 800 to 1000° F when rubbed together at slow surface speeds (3 in./min). The linear bearing test will determine the self-welding and frictional characteristics of these materials in sodium at 1200° F.

To provide basic information for materials for use in journal bearings designed to operate in sodium at 1200° F, a test assembly has been designed and fabrication initiated. Materials found to be acceptable in the combination gear and bearing test fixture will be evaluated for wear and frictional characteristics.

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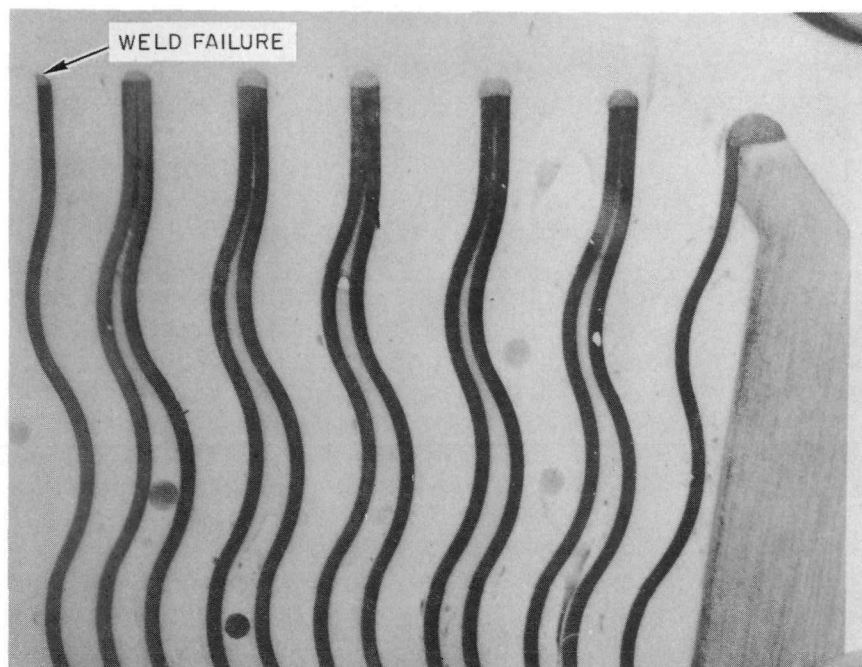
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A motion-multiplying (4x) mechanism which consists of a 2-stage differential rack and pinion gear system currently is being tested. The test mechanism has been placed in continuous operation for cycling a 100-lb load at a rate of 1 ft/min over 76-in. distance with a 20-in. input stroke. Cycling in this mode has been in progress for approximately 14,000 cycles, with no sign of failure. Cycling will continue until failure occurs, to provide basic information regarding problem areas associated with rack and pinion mechanisms.

A motion-multiplying mechanism (using racks and pinions) for operation in sodium to 1200°F has been designed, and fabrication initiated. With this mechanism, the testing of gears without the complication of bearings is possible. The mechanism will have an input stroke of 3 in. with a 12-in. output travel. Tests conducted with the mechanism will determine wear as a function of tooth loading and allowable gear tooth loads for operation in high-temperature sodium. Hastelloy C has been selected as the initial candidate material for the racks and pinions.

Testing of bellows in liquid sodium at a stroke to compressed-length ratio of 4:1 has been concluded. A third Bell-Metrics, type 316 stainless steel bellows has been tested in 1200°F sodium, and failed after 1895 shim cycles (approximately 20% of design life). Disassembly revealed that the seventh outer weld (from the top) failed, leaving the bellows in two sections (Figure 1). Metallographic examination of the bellows revealed that a weld bead thickness of only 66% of the 0.010-in. parent metal was obtained during manufacture of the bellows. The inner welds were approximately 20% heavier than the parent metal. The premature failure of the bellows was caused by insufficient weld penetration on an outer weld. Two previously tested Bell-Metrics bellows of the same configuration were cycled 40,400 and 43,000 shim cycles, respectively (over 400% of design life), at the design stroke to compression ratio of 4 to 1 in sodium at 1200°F before developing a leak.

It is believed that sticking of orifice devices in sodium can be prevented by providing a position seal to exclude sodium vapor and cover gas from the orifice actuator shaft annulus. A test is in progress to evaluate bellows designed for this application. The effect of environmental and cycling rate on the life, and sodium fouling characteristics of the bellows will be determined. Four Bell-Metric type 347 stainless steel bellows assemblies have been installed in a test vessel. Reactor conditions are approximated by using reflective insulation and cooling coils at the upper end of the vessel. Provision has been made for controlled cover gas outleakage. Two bellows assemblies are immersed in liquid sodium, and two are in the cover gas region. The testing of one pair of bellows (one at 1200°F in sodium and the other at 450°F in sodium vapor) has been completed. This pair of bellows has been cycled continuously 21,200 times without failure at a stroke to compressed-length ratio of 3:1 (4-in. stroke) at 8 in./min. The other pair has been half-cycled 12 times (once every half-hour) by manual means. Automatic cycling of this second pair of bellows is continuing at a half-cycle rate every 7 hr with 140 half-cycles completed. The temperature of the bellows located in the sodium vapor has been increased from 450 to 800°F at the completion of 52 half-cycles. Cycling will continue until the bellows have accumulated 500 half-cycles.



7593-2511

Figure 1. Fatigue Failure of Outer Convolution
Weld Bead

Difficulties associated with the rotation of the HNPF loading face shield (LFS) have been attributed in part to the condensation and freezing of sodium vapor within the annulus between the LFS and the cavity liner. In order to provide information regarding this phenomenon, a test utilizing a rotatable plug above a pool of molten sodium had been set up, and placed into service in FY 1965. The sodium pool is maintained at 1200°F as a source of sodium vapor, and agitated continuously by a mechanical device. This facilitates transformation of liquid sodium to the vapor state. By cooling the upper end of the plug, the necessary temperature gradient through the length of the plug (170°F at the top) is provided. Thermocouples in the plug and in the sleeve measure the actual temperature profile. The weight of the plug is supported by bearings so that it can be rotated to measure resistance caused by the deposition of sodium. Torque required to rotate the fixture has increased from 2 in. -lb (initial torque) to 3 to 4 in. -lb after 73 days of exposure. Exposure of the plug to high-temperature sodium will be continued to determine the rate of increase in resistance to rotation.

Tests are in progress to determine the type and effect of sodium vapor deposits in various size annuli and nonannular gaps and surface finishes on the operability of orifice actuator rod and sleeve configurations. The annular and nonannular gaps exist inside 9 small sleeves (0.640-in. ID), and one large sleeve (1.04-in. ID). Eight of the 9 small sleeves contain circular rods. Four of these have normal finishes (125 microns), and diametrical clearances of 0.015, 0.055, 0.105, and 0.205 in. The other 4 have fine surface finishes (16 microns) and clearances which are the same as described for the first 4. The remaining small sleeve and the large sleeve contain cruciform-shaped rods. None of the rods extends into the molten sodium. The sleeves are in a simulated shield

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plug region with reflective insulation to establish temperatures of 200°F at the top and 650°F at the bottom, similar to those in the lower 120 in. of a loading face shield. After 86 days exposure, no increase in the breakaway force was required to move the cruciform-shaped rods or the 0.015- and 0.205-in. diametrical clearance rods; 3 lb additional force was applied to move the 0.055- and the 0.105-in. diametrical clearance rods. After 120 days exposure, the rods will be examined for sodium deposition in radial gaps.

In the same test vessel are four additional sleeves (1.049-in. ID) with (Wiggins Bar-X 17-4 PH stainless steel) metal seal rings at the lower end. These seals act as vapor seals and scrapers for removal of sodium deposits from the operating shafts or rods. Two seal rings (for Shafts 11 and 14) are located near the lower reflector plate region in a 600 to 625°F sodium vapor environment, and two seal rings (for Shafts 12 and 13) are located 10-1/2 in. below the lower reflector plate in a 970 to 995°F sodium vapor environment. Because of differential expansion, none of the 1-in. -diameter, type 304 stainless steel shafts with hard chrome plating (4-micron finish) in the seal area could be operated above 400°F. New shafts (of 17-4 PH bar stock with hard chrome plating in the seal area) have been installed in three of the sleeves.

IV. EVALUATION OF EFFORT TO DATE

The assembly of various testing fixtures for evaluation of candidate materials for use as bearings and gears in high temperature sodium has been progressing as scheduled.

The premature failure of the third Bell-Metrics Type 316 stainless steel bellows during test was a manufacturing problem which the vendor has rectified by a change in welding specifications. Additional testing of this type of bellows will be continued, to verify bellows manufacturing procedures.

V. NEXT REPORT PERIOD ACTIVITIES

Reconditioning of the combustion gear and bearing test assemblies will be completed, and material testing in sodium at 1200°F will be initiated. The linear bearing test to determine the compatibility of tungsten carbide versus tungsten in sodium (1200°F) will be in operation. Fabrication and installation of test equipment to provide basic information for journal bearing materials will be completed. Bench-testing of the motion-multiplying mechanism will be continued. Fabrication of the motion-multiplying mechanism for operation in 1200°F sodium will be completed, and preliminary operational tests will be in progress. The orifice seal bellows cycling tests will be completed. The loading face shield rotational and orifice actuator rod sodium vapor deposition tests will be continued.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Sodium Component Development

Project: Installation of B & W Steam Generator

Project Engineer: J. A. Leppard

Reporting Period: July-September 1965

AEC Category: 04-01-04-31

General Order: 7593

Subaccount: 4127

I. PROJECT OBJECTIVES

The objective of this project is to install the B and W steam generator in the SCTI in preparation for testing.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

Liaison was established between Atomics International and Babcock & Wilcox. The first review meeting was held.

III. PROGRESS DURING REPORT PERIOD

Representatives of Babcock & Wilcox and Atomics International met for a full day's exchange of data about the installation of the B and W boiler at SCTI. A detailed review of the B and W unit revealed several areas where additional investigations are required to assure adequate provision for testing the unit. Lines of communication were established between the two organizations and some transmittal of data occurred.

IV. EVALUATION OF EFFORT TO DATE

Establishing liaison between the two organizations is the initial step required to successfully complete the project.

V. NEXT REPORT PERIOD ACTIVITIES

No meetings are anticipated during the next quarter; however, additional correspondence will pass between the two companies.

SECTION

- III -

HALLAM NUCLEAR POWER FACILITY

&

PIQUA NUCLEAR POWER FACILITY

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program: Hallam Fuel Surveillance
Program Manager: F. C. Gronemeyer
Reporting Period: July-September 1965

I. PROGRAM OBJECTIVES

The purpose of this program is to evaluate the performance of fuel elements in the Hallam reactor. Equipment, methods, and procedures will be provided to perform pre- and postirradiation examinations of fuel elements and fuel element components. The surveillance of the fuel operating parameters is to be incorporated into the performance evaluation.

II. PROJECT MANAGER'S SUMMARY AND EVALUATION

There was no progress made on the major technical objectives of the program. Observations by operations of the fuel storage area revealed the presence of water in some fuel storage thimbles. None of the thimbles containing fuel contained water. The source of the water is not directly known, but is believed to be due to condensation from the water-filled storage pit. A further check of 11 thimbles containing fuel showed the fuel storage atmospheres to be dry. A minor change in the storage atmosphere was recommended during the orifice rework effort. Argon will be used rather than helium.

The limited effort this fiscal year has consisted mainly of surveillance of fuel handling operations as related to the potential reuse of these elements. It is important that all observations should be evaluated. Since the majority of elements are unwashed, the contamination of the storage atmosphere or contamination during handling by water vapor must be avoided. This surveillance directly contributes to determining the criteria for reuse. Continued surveillance of all fuel handling operations is recommended. The report describing progress to date should be completed. If the HNPF Core II uranium carbide fuel is planned for near term use, the technical specifications do not allow washing of these elements. This situation should be resolved prior to use of these elements such that procedures exist for washing if it is necessary.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: HNPf Fuel Surveillance	
Project: Hallam Alloy Fuel Element Surveillance and Examination	
Project Engineer: B. R. Hayward	
Reporting Period: July-September 1965	AEC Category: 04-01-04-21
General Order: 7617	Subaccount: 1910

I. PROJECT OBJECTIVES

The primary objective of this project is to evaluate the in-pile performance of the HNPf Core I U - 10 wt % Mo fuel elements. This objective is to be accomplished by in-pile surveillance of the fuel elements, and performing nondestructive and destructive examinations of selected fuel elements at specific burnup levels.

Comparisons of these results will provide the basis for prediction of the irradiation behavior and burnup capability of the U - 10 Mo fuel elements in the HNPf.

II. TECHNICAL PROGRESS DURING REPORT PERIOD

Due to the extended reactor shutdown no in-pile surveillance or postirradiation examinations were performed during this report period.

The discovery of water in four unoccupied thimbles of Storage Cell 2 at the HNPf initiated a study to determine the possible source and also the effect on the fuel elements. CPPD checked the atmosphere of eleven storage thimbles, containing unwashed fuel elements, for moisture and reported moisture contents of 1 to 3 gm/m³. This reported atmosphere is equivalent in dryness to the minimum humidity in the Sahara Desert at 100°F. No change was recommended for the storage condition of the fuel elements, i. e., unwashed in a dry helium atmosphere. The source of water in the four empty thimbles was not definitely determined.

A change in storage conditions for the fuel elements was later recommended in connection with an orifice rework proposal. Washing the fuel elements was recommended to free sticking orifice drives and to eliminate the hazard of handling fuel element shield plugs coated with sodium. This proposal required removal of the shield plug for modification to the orifice drive, and during this operation the cell atmosphere is not sealed. Therefore, it was recommended that prior to removal of the shield plug the helium cover gas be replaced with argon.

III. EVALUATION OF EFFORT TO DATE

No effort has been expended to date on the primary objectives of this project. All fuel handling operations are being surveyed with respect to observations and incidents that could influence reuse of the stored elements.

IV. NEXT REPORT PERIOD ACTIVITIES

An interim topical report on the accomplishments to date on this program is scheduled for completion. Liaison with orifice cleaning operations will be maintained.

NAA-SR-11650

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: HNPF Fuel Surveillance Program	
Project: HNPF-UC Fuel Element Surveillance and Evaluation	
Project Engineer: B. R. Hayward	
Reporting Period: July-September 1965	AEC Category: 04-01-04-21
General Order: 7617	Subaccount: 1920

I. PROJECT OBJECTIVES

The primary objective of this project is to evaluate the in-pile performance of the ten UC fuel elements inserted in HNPF Core I and all Core II UC fuel elements. This objective will be accomplished by: (a) surveillance of the in-pile performance of the elements, (b) performing nondestructive and destructive post-irradiation examinations at predetermined burnup levels, and (c) irradiation in the NRX and postirradiation examination of two NRX 86 long rod capsules.

Comparison of these results will provide the basis for prediction of the irradiation behavior and burnup capabilities of the UC fuel elements.

II. PROGRESS DURING REPORT PERIOD

Due to the extended reactor shutdown, no operating data were accumulated.

The NRX 86, UC-fueled, long-rod experiments adopted by this program from the LASCOR program were discharged and will be buried. The calculated peak operating temperature of NRX 86-1 has ranged between 1600 and 1800°F, and the peak burnup is approximately 13,000 Mwd/MTU. The peak operating temperature of NRX-86-2 ranged between 1350 and 1600°F (calculated), and the peak burnup achieved is approximately 11,000 Mwd/MTU.

III. EVALUATION OF EFFORT TO DATE

The NRX-86-1 and -2 experiments have been removed from the NRX and arrangements are underway to dispose of the experiments.

No effort has been expended to date on objectives A and B.

IV. NEXT REPORT PERIOD ACTIVITIES

No effort is anticipated. Liaison with orifice cleaning operations will be maintained.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Piqua Technical Assistance Program	
Project: PNPFF Fuel Element Examination	
Project Engineer: S. O. Arneson	
Reporting Period: July-September 1965	AEC Category: 04-02-02-01
General Order: 7615	Subaccount: 1970

I. PROJECT OBJECTIVES

The objectives of this project are to examine and evaluate the performance of irradiated Piqua fuel elements. Cylinders in ten fuel elements have been given extensive preirradiation measurements and inspection. Systematic data will be obtained from the postirradiation examination of these cylinders with regard to film formation on the heat transfer surfaces, fuel swelling due to irradiation, and interdiffusion of the aluminum, nickel, and uranium alloy. These data will be analyzed and evaluated as a primary basis for establishing the performance capabilities of the PNPFF Core I fuel elements.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

No major activities were undertaken during the first quarter of FY 1966.

III. ACCOMPLISHMENTS DURING REPORT PERIOD

The data obtained from the postirradiation examination of Piqua fuel elements P-1071 and P-1091 are being consolidated in a single topical report. In preparing this report, further evaluation was given dimensional change versus burnup data for the two fuel elements. Using an extrapolation of a least squares fit of the data, an increase of 0.84% in the outer fuel ring diameter is predicted at a peak burnup of 1.96×10^{20} fiss/cc (4000 Mwd/MTU). However, based on the upper envelope of dimensional change data, an increase of 1.2% in outer fuel ring diameter could occur at this burnup. Firm contact between the outer fuel cylinder and outer process tube would result if 1.2% increase in the fuel diameter actually occurs. Based on the latter possibility, it is planned that a fuel evaluation element be removed from Piqua Core I when a peak burnup of about 1.96×10^{20} fiss/cc (4000 Mwd/MTU) is attained. An evaluation element should reach this burnup late in the next report period.

IV. EVALUATION OF EFFORT TO DATE

No major activities were undertaken during the report period.

V. NEXT REPORT PERIOD ACTIVITIES

It is planned to remove a fuel evaluation element which has attained about 1.96×10^{20} fiss/cc peak burnup from Piqua Core I. This should take place during the latter part of the next report period. Postirradiation examination will begin in the third quarter of this fiscal year.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program: PNP Development

Project: Core II SAP Clad UC Fuel Element Development

Project Engineer: S. O. Arneson

Reporting Period: July-September 1965

AEC Category: 04-02-02-01

General Order: 7629

Subaccount: 1920

I. PROJECT OBJECTIVES

The objectives of this project are the development of processes, specifications, and procedures for fabricating SAP-clad UC fuel elements for PNP Core II; the fabrication of a mockup fuel element for hydraulic testing; the fabrication of a Core II prototype element for irradiation in Piqua Core I; and initial surveillance and evaluation of the prototype element during irradiation in Core I.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

A decision was made to use the collet technique for making eutectic-bonded end-closures in SAP-clad fuel tubes for the Core II prototype fuel element. Development of the magneform-eutectic bonding technique was discontinued.

Continuing investigations of nondestructive testing processes have developed equipment and testing techniques that are capable of detecting inclusions or other defects in finned SAP cladding tubes and unbonded areas in eutectic-bonded end-closures in SAP fuel tubes.

The design of the Core II prototype fuel element was established, and procurement of SAP tubing was initiated. Preparations were made for dilution of 4.9% enriched U-metal to 3.8% enriched U-metal for fabricating the prototype UC fuel slugs.

III. PROGRESS DURING REPORT PERIOD

A comparison of current cost and reliability data for the collet-eutectic and magneform-eutectic SAP end-closure bonding techniques indicated an advantage in favor of the collet technique. As a result, the magneform development effort was discontinued. The reasons for this decision include the following.

- 1) Cost comparisons on these two processes show no apparent cost savings for the magneform process for PNP Core II.
- 2) Bond quality and reproducibility appear no better for the magneform process.
- 3) Significant development and inspection problems remain to be solved for the magneform process.
- 4) With the selection of SAP-UC for PNP Core II, the number of end-closures required for core fabrication is reduced by approximately a factor of two as compared with the UO₂ design. As a result, the slower collet bonding process could be used for Core II production within the schedule, on single-shift operation.

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Hydraulic tests were performed on the Core II SAP-UO₂ fuel element mockup which was fabricated last fiscal year. Although the mockup was significantly different from the present Core II SAP-UC fuel element design, the test was made to obtain data on the pressure drop across finned tubes in a close-pack array. All previous data had been obtained for finned tubes in a spaced-rod array. The actual pressure drop was determined to be considerably higher than had been predicted using the data for a spaced-rod array.

Several samples of eutectic-bonded end-closures in SAP cladding tubes were submitted to two nondestructive testing firms for ultrasonic evaluation of the bond quality. Both firms were able to detect unbonded areas indicating that this technique is feasible. Ultrasonic C-scans submitted by the two firms were evaluated, and both were found to give comparable bond quality information. One of the techniques, however, gave higher sensitivity and resolution and, therefore, was easier to read and interpret.

The investigations of radiography and eddy-current techniques for the evaluation of SAP tubing, and ultrasonic techniques for the evaluation of SAP billets, were continued. Inclusions and other defects were detected by all three techniques. Radiography was found to be capable of detecting iron inclusions as small as 0.002 by 0.002 by 0.002 in. in the 0.030-in. -thick tube walls. However, the high cost of radiography will probably prohibit its use for large-scale production. Evaluation of the ultrasonic and eddy-current inspection techniques is continuing. SAP billets scheduled for extrusion to finned cladding tubes are being inspected by conventional ultrasonic techniques. Internal and external eddy-current coils are being procured for the inspection of SAP tubes to be used for the prototype fuel element.

A 42-fuel rod, concentric ring array was selected as the Core II SAP-UC prototype design. Procurement action has been initiated for the SAP tubing for delivery in December. AEC permission was obtained and effort was initiated to dilute approximately 150 kg of excess HNPF 4.9% enriched U-metal to 3.8% enriched U-metal for fabrication of the prototype fuel slugs.

IV. EVALUATION OF EFFORT TO DATE

The decision to use the collet technique for end-closure bonding will assure high quality end-closure bonds on the Core II prototype fuel rods. This technique has been used successfully in the fabrication of irradiation test elements for the AOMR and HWOCR programs. It is believed that the magneform bonding technique could also be used successfully; however, additional development effort would be required with only marginal potential improvements in process capability. Magnetic force welding, being developed under the HWOCR program, will be considered as the end-closure process for Core II, if developed in time.

The continuing development of equipment and techniques shows significant promise for utilizing conventional nondestructive testing processes in the inspection and quality control of SAP-clad UC fuel rods. Additional evaluations are required to determine the applications and limitations of these techniques in the production of Core II.

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Completion of the mockup and prototype fuel elements is now scheduled for February 1966, instead of as originally planned in November and December 1965, respectively. This delay was due to the additional design and analysis effort required, and the longer than expected lead-time for SAP tubing procurement.

No delay is anticipated for completing the materials specifications, process specifications, and assembly procedures prior to their being required for fabrication of the Core II fuel elements.

V. NEXT REPORT PERIOD ACTIVITIES

Fabrication of the prototype and mockup fuel elements will begin and be nearly completed by the end of the report period. Material specifications for SAP billets and tubing will be issued, and procurement of billets for Core II will be begun. Preparation of the nuclear safety feasibility report and fabrication process specifications will be initiated.

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Program: PNPf Development

Project: PNPf Core II Design

Project Engineer: H. J. Rubinstein

Reporting Period: July-September 1965

AEC Category: 04-02-02-01

General Order: 7629

Subaccount: 1940

I. PROJECT OBJECTIVES

The objectives of this project are to design a SAP-clad UC fuel element for Core II at PNPf, and to prepare a safeguards summary report for operation of the PNPf with this new core.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

This project was initiated in FY 1966.

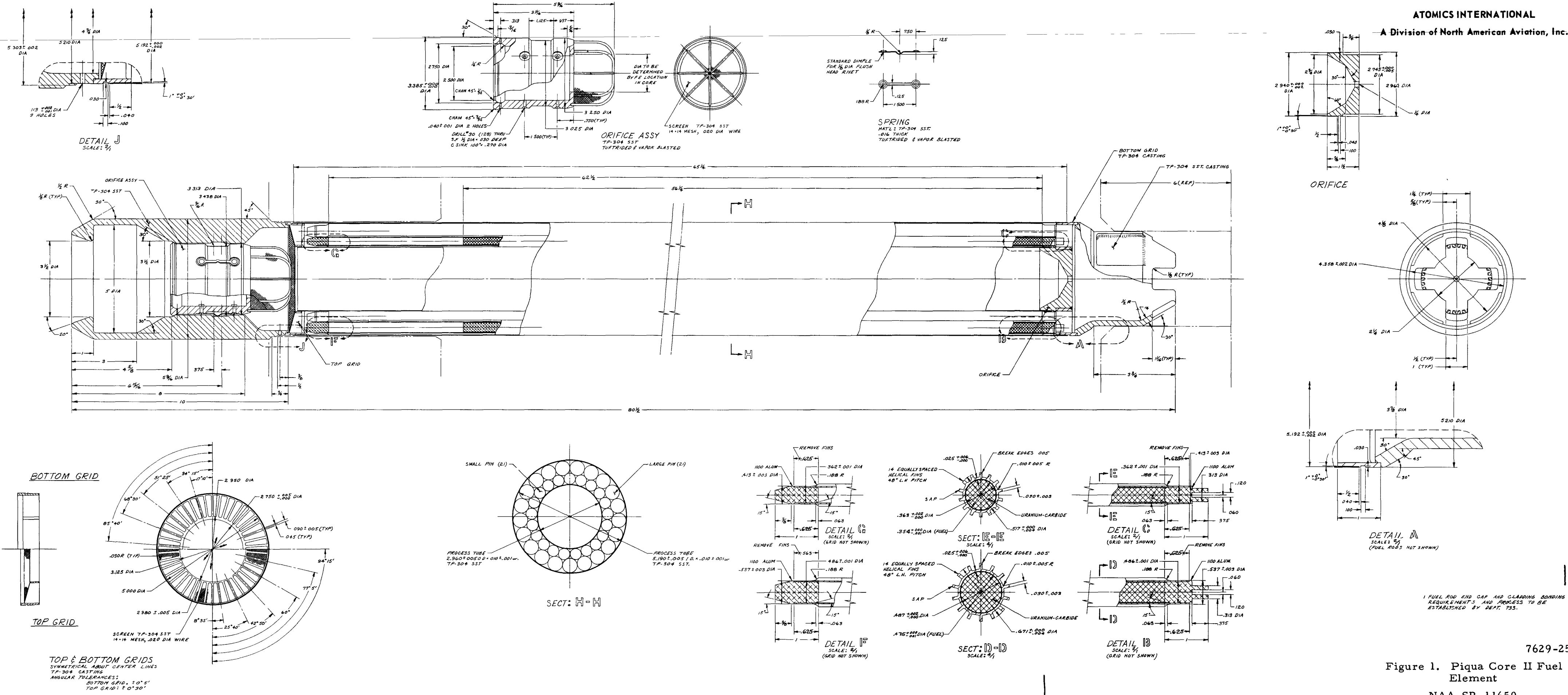
III. PROGRESS DURING REPORT PERIOD

A fuel element optimization study was carried out, resulting in selection of a reference fuel element having 42 SAP-clad UC fuel rods arranged in two tightly packed concentric annular rings. This element has a stretch capability, for a 61-element core, well within fuel and cladding temperature limitations of 2000 and 850°F respectively. It is the smallest number of fuel rods, arranged in this type of configuration and sized within existing dimensional limitations, that will allow a single-fuel element cross-section to fit rodded as well as un-rodded core (control rod position) positions. In comparison with the 48-rod element presented in the UC feasibility study, the 42-rod reference fuel element offers a significant reduction in fuel fabrication costs and, because of fewer feet of fuel rod cladding and fewer end plug bonds, gives greater reliability.

At the end of this report period the following major work has been accomplished.

- 1) Layout drawings of the 42-, 44-, 46-, and 48-pin elements were made to provide necessary nuclear and heat transfer parameters for calculations.
- 2) Heat transfer analysis was made for the prototype to be placed in Core I, and complete analysis of the hot-channel element in Core II for the 42- and 48-pin design.
- 3) Determination of the nuclear control requirements for Core II, and an evaluation of the available control rod worth for the reference (48-pin) design.
- 4) Calculation of the enrichment required for the prototype element to give a maximum power of 1.6 Mwt.
- 5) Design layout and detailed manufacturing drawings of the prototype element are under way.
- 6) The Safety Analysis Report for the Prototype in Core I is 50% complete.
- 7) A hydraulic test of a mockup (originally made for the UO₂ design) was underway for pressure drop measurements.

Figure 1 presents the design of the 42-rod element for Core II.



7629-2501

Figure 1. Piqua Core II Fuel Element

NAA-SR-11650

III-9

OSTI ID: 060019411

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A. PROTOTYPE

In order to meet the schedule for completion of the prototype and insertion in Core I by February 1966, it was necessary to proceed with a design which was most probable for Core II. The prototype must also provide the following objectives.

- 1) Provide manufacturing and assembly experience for an element similar to the Core II loading.
- 2) Provide about 14 months of operating experience at power levels up to 1.6 Mwt before insertion of Core II in PNPf.
- 3) Provide in-core surface temperature measurements which simulate the 45-Mwt as well as the 60-Mwt stretch condition.
- 4) Provide shipping and handling experience for UC fuel with particular interest in the storage pool and the fuel handling machine.
- 5) Provide an approved safeguards report for the use of UC fuel at PNPf.

The prototype design was selected on September 16, 1965 and is to be identical with that shown in Figure 1 except for cladding surface thermocouples, which are not shown, and the upper extension piece to bring out the thermocouple leads. Design parameters and dimensions for the prototype are given in the following discussions.

B. HEAT TRANSFER AND HYDRAULIC ANALYSIS

A 42-rod element in two concentric rings of equal number of rods has been selected for the prototype UC fuel element. The geometric data for this design together with that of the previous reference design (48-rods) is shown in Table 1. The 42-rod design has the minimum number of rods that can be nested within the constraints of the fixed outer process tube ID and the minimum OD inner process tube set by control rod size.

The thermal and hydraulic basis for selection of the 42-rod prototype is presented in the following. Basic thermal and hydraulic data applicable to the analysis used in making the comparison of the 42- and 48-rod element are given in Table 2. The results of this analysis are shown in Table 1, which also includes the expected performance of each element as a prototype in Core I.

The results at this point are preliminary and do not represent the final thermal performance evaluation, but they are believed to be sufficiently accurate for the prototype selection. Power peaking factors, hot-channel factors, and pressure drop values are under study and may change the final results. Should the higher temperatures of the 42-rod design be increased more as a result of further study, the 48-rod element could be selected for Core II without too much loss of simulation with the 42-rod prototype in Core I.

Ground rules for the designs include: (1) 61 fuel elements, (2) fuel length of 56.25 in., (3) coolant inlet temperature of 525°F, (4) 60-Mwt reactor power capability with a maximum clad steady-state surface temperature of less than 850°F, and (5) core pressure loss limited by the capacity of the existing pump with either the small or large impellers.

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TABLE 1

CORE GEOMETRIES WITH 42- OR 48-ROD FUEL ELEMENTS

No. of fuel elements	61			
No. of control rods	13			
Length of fuel (in.)	56.25			
Finned length of rods (in.)	62.5			
ID of outer process tube (in.)	5.190			
Thickness of process tubes (in.)	0.010			
Fuel rod cladding thickness (in.)	0.025			
No. of rectangular fins per fuel rod	14			
Cladding fin thickness (in.)	0.030			
Fin twist (degrees per ft)	90			
No. of fuel rods per element	42-rod		48-rod	
No. of small and large rods per element	21		24	
OD of inner process tube (in.)	2.960		3.180	
Element coolant flow area (in. ²)	5.578		4.589	
Total heat transfer surface (ft ²)	3160		2994	
	Large Rod	Small Rod	Large Rod	Small Rod
Fuel diameter (in.)	0.480	0.358	0.446	0.342
Cladding ID (in.)	0.487	0.363	0.453	0.347
Cladding OD (in.)	0.537	0.413	0.503	0.397
Gas bond thickness (in.)	0.0035	0.0025	0.0035	0.0025
Fin OD (in.)	0.671	0.517	0.597	0.475
Fin height (in.)	0.067	0.052	0.047	0.039
Heat transfer surface area per unit length of rod (ft ² /ft)	0.2969	0.2294	0.2414	0.1949

It was assumed that the flow distribution in the core would be the same as that in Core I. In this core, 41 elements of the inner region have the same flow, and 20 elements of the outer region are orificed for equal flow. The distribution of total reactor flow assumed is, then:

$$\begin{aligned}
 \text{Inner flow region} &= 70.4\% \\
 \text{Outer flow region} &= 23.6\% \\
 \text{Bypass flow} &= \underline{6.0\%} \\
 &100 \% .
 \end{aligned}$$

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TABLE 2
BASIC THERMAL AND HYDRAULIC DATA

Reactor coolant inlet temperature (°F)	525
Reactor coolant inlet pressure (psi)	120
Power peaking factors	
Radial	1.5
Axial	1.5
Elemental	1.2
Symmetric axial cosine power distribution, L/L_0	0.953
Hot-channel factors	
Coolant ($F_{\Delta T}$)	1.20
Film (F_{Θ})	1.40
Heat flux (F_{ϕ})	1.15
Fraction of heat generated in moderator	0.06
Fraction of heat generated in coolant	0.02
Fin heat transfer efficiency (%)	95
Coolant properties at 560°F	
Density (lb/ft ³)	56.4
Viscosity (lb/ft-hr)	1.40
Specific heat (Btu/lb-°F)	0.517
Thermal conductivity (Btu/hr-ft-°F)	0.069
Prandtl number	10.49
Surface roughness (in.)	0.6×10^{-4}
Element flow orificing arrangement	2 velocity zones
Ratio flow maximum power element to average element	1.115

The ratio of flow in the maximum powered central element-to-core average element flow is 1.115.

The fin lengths for the two sizes of rods in each element was determined so that maximum temperature of the surface of both small and large rod are equal. A balance between heat flux from the rods and size of coolant subchannel removing the heat was achieved by proper sizing of the fins on each rod.

The pressure drop and element velocity calculations were based on the equivalent diameter concept for flow through straight noncircular passages. A cladding surface roughness of 0.6×10^{-4} in. was divided by the equivalent diameter to give a relative roughness for the channel. This parameter and the Reynolds number determined the Moody friction factor. An entrance-plus-exit loss of 2.5 velocity heads inside the element was assumed.

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Future thermal and hydraulic analysis of the UC fuel element will consist of completing an optimization study of a range of rods/element including 44 and 46 rods in addition to the 42- and 48-rod element presented here. Factored into this study and future work will be an analysis of hot-channel factors, and any new and better data on power peaking factors and hydraulic friction losses. Tables 1, 2, and 3 give the results of hydraulic and heat transfer calculations to date.

C. NUCLEAR ANALYSIS

Preliminary analysis of the nuclear performance characteristics of the UC 61-element core loading was initiated. Group constants as a function of enrichment, over the enrichment range from 2.0 atom % U^{235} to 4.0 atom % U^{235} , were calculated for the 48-pin "reference design" fuel element. Burnup calculations covering the same enrichment range were completed.

A fuel enrichment of 3.8 wt % U^{235} was selected for the prototype fuel element. The relative power of the prototype element in the Piqua core was calculated as a function of enrichment. The calculations were based on determining the fraction of core power produced by a fuel element located in the center of the core. FOG one-dimensional multigroup diffusion theory calculations were performed with a Piqua Core I element in the center of the core, and also with the prototype element in the center. The fuel enrichment was selected such that the prototype element could operate at up to a power level of 1.6 Mwt. The estimated peak fuel element power for Piqua Core II is 1.48 Mwt.

The equilibrium cycle fuel management scheme for the UC 61-element core loading of Piqua Core II is an 8-batch refueling program. With a 0.80 capacity factor between shutdowns, this results in a 170-day operating period at 60 Mwt. Fuel is discharged from the central part of the core at an exposure of 15,000 Mwd/MTU, and fresh fuel is loaded near the core periphery. The fuel enrichment for the fresh elements necessary to maintain this cycle is 2.38 wt % U^{235} .

The initial core loading will be selected to simulate the equilibrium core by having two enrichment zones. The outer zone will contain fuel of the feed enrichment and the inner zone will contain fuel of a lower enrichment. Up to four dummy fuel elements will be used to enhance the exposure of the first core fuel loading.

Control requirements for the equilibrium core have been estimated for both 45.5-Mwt and 60-Mwt operation. These requirements are as follows.

Requirement	% $\Delta K/K$	
	45.5 Mwt	60 Mwt
Equilibrium Xe	2.25	2.45
ΔT organic	2.01	2.01
ΔT fuel	1.25	1.54
Burnup	2.81	2.81
Center control rod stuck out	1.40	1.40
Shutdown margin	<u>1.00</u>	<u>1.00</u>
Total	10.72	11.21

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TABLE 3

CALCULATED THERMAL AND HYDRAULIC PERFORMANCE*

Parameter	Prototype in Core I	Core II			
		Small		Large	
Reactor power (Mwt)	45.5	60		45.5	
Maximum element power (Mwt)	1.60	1.475		1.119	
Pump impeller size	Small	Small	Large	Small	Large
42-Rod Element					
Core pressure loss (psi)	27.5	18.4	20.4	18.4	20.4
Reactor total flow (gpm)	-	14,880	15,750	14,880	15,750
Element flow (gpm)	320	256	271	256	271
Core outlet temperature (°F)	-	583	580	569	567
Element outlet temperature (°F)	594	604	600	585	582
Core average heat flux (Btu/hr-ft ²)	-	59,700	59,700	45,290	45,290
Maximum linear rod power (kw/ft)	14.62	13.49	13.49	22.6	22.6
Maximum specific power (kw/kg U)	32.3	29.8	29.8	22.6	22.6
Peak heat flux (Btu/hr-ft ²)	154,900	142,840	142,840	108,360	108,360
Burnout heat flux (Btu/hr-ft ²)	778,220	649,300	678,450	680,045	710,625
Average velocity (ft/sec)	18.39	14.71	15.56	14.71	15.56
Maximum velocity (ft/sec)	20.27	16.27	17.19	16.27	17.19
Heat transfer coefficient (Btu/hr-ft ²)	1545	1295	1354	1295	1354
Maximum clad surface temperature (°F)	792	820	807	748	738
Maximum fuel surface temperature (°F)	1531	1503	1490	1264	1254
Maximum fuel temperature (°F)	1847	1795	1782	1485	1475
48-Rod Element					
Core pressure loss (psi)	27.5	27.0	30.6	27.0	30.6
Reactor total flow (gpm)	-	13,830	14,780	13,830	14,780
Element flow (gpm)	240	238	254	238	254
Core outlet temperature (°F)	-	588	585	573	571
Element outlet temperature (°F)	616	610	605	590	585
Core average heat flux (Btu/hr-ft ²)	-	63,010	63,010	47,800	47,800
Maximum linear rod power (kw/ft)	12.80	11.80	11.80	8.95	8.95
Maximum specific power (kw/kg U)	32.7	30.2	30.2	22.9	22.9
Peak heat flux (Btu/hr-ft ²)	166,600	153,680	153,680	116,600	116,600
Burnout heat flux (Btu/hr-ft ²)	686,040	691,870	733,660	728,520	772,310
Average velocity (ft/sec)	16.80	16.61	17.76	16.61	17.76
Maximum velocity (ft/sec)	18.62	18.43	19.65	18.43	19.65
Heat transfer coefficient (Btu/hr-ft ²)	1491	1478	1556	1478	1556
Maximum clad surface temperature (°F)	811	792	778	727	716
Maximum fuel surface temperature (°F)	1509	1437	1423	1216	1205
Maximum fuel temperature (°F)	1786	1693	1679	1410	1399

*Data not specifically referred to core or core average apply to maximum powered element.

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This indicates that a total control rod worth of 11.21% $\Delta k/k$ would be sufficient to provide a 1% shutdown margin in all cases even in the event that the center rod remains withdrawn. These results are only approximate; the temperature reactivity defects are extrapolations of the feedbacks calculated for the UO_2 core loading. It is anticipated that the temperature coefficients for the UC core will be slightly less negative than those of the UO_2 core. Specifically excluded from the control requirements is any requirement for transient Xe override. The criteria for a transient Xe override requirement have not as yet been established.

Initial calculations of control rod worth in the UC core indicate that the total worth of 13 control rods is 9.6% $\Delta k/k$. This compares with a measured worth in Piqua Core I of 12% $\Delta k/k$ and a previously calculated worth for the UO_2 core of 12% $\Delta k/k$. The reason for the discrepancy between the calculated and measured rod worths has not yet been established; indications are that the surrounding UC fuel shields the control rod more effectively than does the U-Mo or the UO_2 fuel. Calculations are under way to evaluate this hypothesis.

D. SAFEGUARDS REPORT

Two Safety Analysis Reports will be prepared, one for the insertion of a UC-SAP clad prototype in Core I and one for the operation up to 45.5 Mwt of PNPf with a complete loading of a new UC-SAP clad core designated as Core II.

Initial safety analysis work has been directed toward preparing the prototype fuel element report. A rough draft of more than 50% of this report has been completed and work is in progress on the remainder of the report. Completion of the prototype SAR is scheduled for November 1, 1965.

Criticality analyses of the pool and turret storage facilities show it is safe to store the 3.8% U^{235} prototype fuel element in the specified array of Core I elements.

Several postulated fuel handling incidents were examined. Even for the hypothetical case of dropping a fuel element on the reactor room floor, analysis shows that the 2-hour dose at the controlled-area-boundary is more than two orders of magnitude below the reference emergency limits specified in 10 CFR 100.

The fission gas concentration at the controlled area boundary was calculated for a fuel rod cladding failure at steady-state full-power operating conditions. Based on pessimistic assumptions, this concentration is below MPC for an unrestricted area.

IV. EVALUATION OF EFFORT TO DATE

The design of the Core II element is progressing satisfactorily. As design progresses, two important unresolved questions may require reevaluation of the design. These are the results of the hydraulic testing and the results of the control rod worth calculations.

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V. NEXT REPORT PERIOD ACTIVITIES

Fabrication drawings for the instrumented prototype element will be released early next report period. The design layout of the Core II element will be completed and approved. The Core II fabrication drawings and product specification will be nearing completion in preparation for release early in the following report period.

Optimization of the design will be completed. The effect of the hydraulic testing and control rod worth calculations will be factored into the design.

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Program: PNPf Development	
Project: PNPf Catalytic Hydrocracking Coolant Processing System	
Project Engineer: H. J. Rubinstein	
Reporting Period: July-September 1965	AEC Category: 04-02-02-01
General Order: 7629	Subaccount: 4310

I. PROJECT OBJECTIVES

The objective of the Piqua hydrocracker program is to establish the technical feasibility and economic aspects of hydrocracking as a means of reducing coolant makeup costs on an organic cooled reactor. In particular, this program will provide a basis on which to establish the merit of polymerized organic coolant reclamation for the HWOCR concept. To carry out this objective, a hydrocracker will be installed at PNPf where recycling conditions and irradiation of hydrocracked products can be achieved.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

The following major accomplishments have been achieved during FY 1966.

- 1) Preparation of a program plan
- 2) Review of the Phillips Petroleum Co. Title I design* of the hydrocracker system
- 3) Points of piping connections established between PNPf and the hydrocracker system

III. PROGRESS DURING REPORT PERIOD

A program plan was prepared. At the request of the AEC, the program plan delineated the extent of the efforts of Phillips Petroleum Co., City of Piqua, and Atomics International. The responsibilities of the three contractors included the following. (1) Title II design; performance tests of the system, and consultation involving technical aspects of hydrocracker operation and performance; (2) Review of all aspects of the design, making recommendations to the Commission, operation of the system, and conducting of chemical analyses; and (3) Consultant to AEC-COO on all phases of the work. Responsibility for all aspects of the program as they affect PNPf, including preparation of a Safety Analysis Report. Analysis of the hydrocracker operation and performance in conjunction with the HWOCR program.

The Phillips preliminary design was reviewed and a number of comments and suggestions were presented. Principal among the comments was the suggestion that smaller catalyst beds be used for the initial stages of operation. It was further suggested that no attempt be made to convert HB at the rate of its formation in PNPf during the early stages of hydrocracker operation. Thus the

*Phillips Petroleum Co., "Revised Design for Piqua Hydrocracker AEC Contract No. AT(10-1)-1215," June 1965

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hydrocracker and PNPf would be operated independently during the early test period, when performance of the new system may be uncertain. The use of smaller catalyst beds during this period makes possible the use of smaller and more easily handled, quantities of coolant.

A proposed schedule of milestones over the next six months was established as follows.

<u>Date</u>	<u>Milestone</u>
October 22, 1965	P&I drawings, preliminary to Title II drawings, to be completed.
November 1, 1965	First of Title II drawings delivered to COO. The first drawing will be the hydrocracker mechanical design.
February 1, 1966	Safety Analysis Report (SAR) completed.
March 1, 1966	Complete review of SAR
May 1, 1966	Review by DR&L completed and construction permit issued.

Points of tie-in between PNPf and the hydrocracker system were selected. Coolant to be treated will be taken from the PNPf system immediately upstream of the column feed heater. Provision will also be made to feed high boilers from the HB decay tank to the hydrocracker system, so that highly concentrated HB streams can be treated on the catalyst bed. Hydrocracked product coolant will return to the PNPf coolant system through the degasifier. During the early stages of operation, when hydrocracked coolant will not be returned directly to the PNPf system, the hydrocracker product will be stored in one of the PNPf new coolant storage tanks. If analyses indicate that the coolant is of reactor quality, it will be pumped to the auxiliary building drain tank, and from there metered into the PNPf system. If the hydrocracker product is initially not suitable for use on the reactor, it will be transferred to the HB decay tank for disposal in the PNPf waste-fired boiler.

IV. EVALUATION OF EFFORT TO DATE

The Piqua Hydrocracker Program was initiated during this report period. Thus the effort during the quarter has been mainly one of organization and planning. The Program Plan recommended the program as a 4-yr effort covering all phases of design, construction, testing, analysis, and licensing. The Title I design of the Phillips Petroleum Co. was reviewed, and suggestions were made for the continuation of design under Title II.

V. NEXT REPORT PERIOD ACTIVITIES

During the next report period, the Title II design, as completed by Phillips, will be reviewed. The proposed method of connecting the hydrocracker to PNPf will be submitted. Work on the SAR will be started and specifications for additional chemistry laboratory space at PNPf will be prepared.

SECTION

- IV -

REACTOR PHYSICS
&
FUELS AND MATERIALS

ATOMICS INTERNATIONAL
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Program: General Research, Reactor Physics Studies	
Project: Age Measurements	
Project Engineer: H. A. Morewitz	
Reporting Period: July-September 1965	AEC Category: 04-40-01-01
General Order: 7604	Subaccount: 1311

I. PROJECT OBJECTIVES

The objective of this project is to investigate the slowing-down properties of fission neutrons in various metal and moderator mixtures. These measurements are compared with Monte Carlo calculations, to determine the accuracy of the microscopic, fast, neutron cross-section data used in obtaining group-averaged cross-sections for reactor design codes. In many cases, a direct measurement of these cross-sections is not feasible over the energy range needed. In such cases, the cross-section must be calculated; in other cases the measured cross-section must be evaluated. Age measurements represent an indirect method of checking out these cross-sections.

II. PROGRESS DURING REPORT PERIOD

A. AGE OF ALUMINUM-WATER MIXTURES

The age measurements in several aluminum-water moderator mixtures were completed. The theoretical analysis has been compared in considerable detail with corresponding experiments.¹

Comparison of the Monte Carlo calculations with experiments for the neutron age and higher moments is presented in Table 1.

TABLE 1
NEUTRON AGE AND HIGHER MOMENTS IN ALUMINUM-WATER MIXTURES

Al/H ₂ O	Age (cm ²)	M ₄ (10 ⁴ cm ⁴)	M ₆ (10 ⁸ cm ⁶)	
0.000	calculation	26.5 ± 0.1	9.99 ± 0.15	1.86 ± 0.06
	experiment	26.6 ± 0.3	9.45 ± 0.50	1.39 ± 0.20
0.250	calculation	33.8 ± 0.2	14.36 ± 0.27	2.78 ± 0.11
	experiment	33.9 ± 0.6	14.50 ± 0.25	2.68 ± 0.04
0.500	calculation	41.3 ± 0.3	19.77 ± 0.35	4.07 ± 0.18
	experiment	43.2 ± 0.8	21.60 ± 0.70	4.40 ± 0.30
1.000	calculation	57.2 ± 0.3	33.41 ± 0.50	7.62 ± 0.27
	experiment	59.6 ± 0.9	37.10 ± 0.65	8.83 ± 0.23
2.000	calculation	90.1 ± 0.4	70.56 ± 0.77	18.65 ± 0.44

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The effect of the inelastic scattering and elastic scattering anisotropy in aluminum on the neutron age and higher moments is given in Tables 2 and 3, respectively.

TABLE 2
EFFECT ON NEUTRON AGE AND HIGHER MOMENTS OF
INELASTIC SCATTERING IN ALUMINUM

Al/H ₂ O	Age (cm ²)	M ₄ (10 ⁴ cm ⁴)	M ₆ (10 ⁸ cm ⁶)
0.25 *	33.8 ± 0.2	14.36 ± 0.27	2.78 ± 0.11
†	35.2 ± 0.2	16.42 ± 0.34	3.60 ± 0.17
0.50 *	41.3 ± 0.3	19.77 ± 0.35	4.07 ± 0.18
†	44.3 ± 0.3	24.70 ± 0.50	6.25 ± 0.29
1.00 *	57.2 ± 0.3	33.41 ± 0.50	7.62 ± 0.27
†	63.6 ± 0.4	46.96 ± 0.97	15.88 ± 0.89
2.00 *	90.1 ± 0.4	70.56 ± 0.77	18.65 ± 0.44
†	103.2 ± 0.6	106.96 ± 1.72	42.95 ± 1.56

*Inelastic scattering in aluminum included.

†No elastic scattering in aluminum included

TABLE 3
EFFECT ON NEUTRON AGE AND HIGHER MOMENTS OF
ELASTIC SCATTERING ANISOTROPY IN ALUMINUM

Al/H ₂ O	Age (cm ²)	M ₄ (10 ⁴ cm ⁴)	M ₆ (10 ⁸ cm ⁶)
0.25 A	33.8 + 0.2	14.36 + 0.27	2.78 + 0.11
I	32.2 + 0.2	12.63 + 0.21	2.22 + 0.08
0.50 A	41.3 + 0.3	19.77 + 0.35	4.07 + 0.18
I	38.2 + 0.2	16.03 + 0.24	2.80 + 0.09
1.00 A	57.2 + 0.3	33.41 + 0.50	7.62 + 0.27
I	51.4 + 0.2	24.96 + 0.27	4.44 + 0.09
2.00 A	90.1 + 0.4	70.56 + 0.77	18.65 + 0.44
I	80.1 + 0.3	52.39 + 0.44	10.90 + 0.19

A = Anisotropic elastic scattering in aluminum.

I = Isotropic elastic scattering in aluminum.

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The theoretical analysis of the age measurements was extended by using two other sets of aluminum cross-section data. The results of this analysis is presented in Table 4.

TABLE 4
CALCULATED NEUTRON AGE AND HIGHER MOMENTS IN
ALUMINUM-WATER MIXTURES

Al/H ₂ O	Cross-Section Set	Age (cm ²)	M ₄ (10 ⁴ cm ⁴)	M ₆ (10 ⁸ cm ⁶)
0.25	Ref. 2	33.8 ± 0.2	14.36 ± 0.27	2.78 ± 0.11
	3	33.8 ± 0.2	14.77 ± 0.30	3.04 ± 0.14
	4	33.9 ± 0.2	14.61 ± 0.28	2.89 ± 0.12
0.50	Ref. 2	41.3 ± 0.3	19.77 ± 0.35	4.07 ± 0.18
	3	41.1 ± 0.3	19.71 ± 0.35	4.08 ± 0.18
	4	41.3 ± 0.3	19.75 ± 0.35	4.04 ± 0.18
1.00	Ref. 2	57.2 ± 0.3	33.41 ± 0.50	7.62 ± 0.27
	3	56.7 ± 0.3	32.98 ± 0.49	7.49 ± 0.27
	4	57.5 ± 0.3	33.82 ± 0.50	7.73 ± 0.27
2.00	Ref. 2	90.1 ± 0.4	70.56 ± 0.77	18.65 ± 0.44
	3	89.3 ± 0.4	70.96 ± 0.85	19.55 ± 0.58
	4	90.5 ± 0.4	71.71 ± 0.88	19.51 ± 0.85

Since the metal plates used in the measurements were 1100-H 14 aluminum alloy (aluminum and approximately 0.5% iron and 0.2% copper), a final calculation was performed to determine the effect on the neutron age and higher moments of the impurities. The copper was mocked up by iron and the results of the calculation is given in Table 5.

The results presented in Table 1 are illustrated by Figures 1, 2, 3, while Figures 4, 5, 6 illustrate the results of Tables 2 and 3.

B. NEUTRON AGE IN WATER

A new set of oxygen cross-section data, constructed at KAPL,⁵ was obtained recently. The data was based on both new experimental results and sophisticated theoretical models. This information was readily transformed into the AIENDF format and several Monte Carlo calculations for the neutron age and higher moments in water were performed. The results of these calculations, compared with

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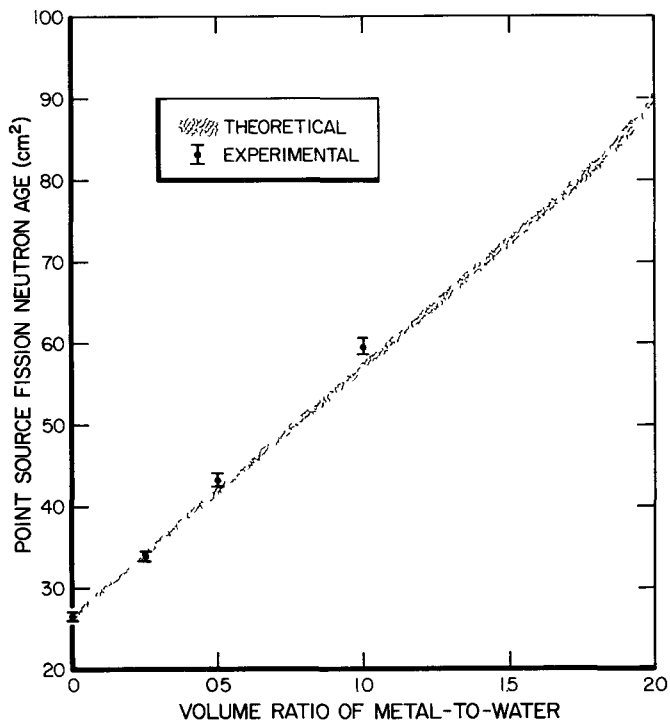
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TABLE 5
EFFECT ON THE NEUTRON AGE AND HIGHER MOMENTS OF THE
IMPURITIES IN 1100-H 14 ALUMINUM ALLOY

Al/H ₂ O		Age (cm ²)	M ₄ (10 ⁴ cm ⁴)	M ₆ (10 ⁸ cm ⁶)
0.25	*	33.8 + 0.2	14.36 + 0.27	2.78 + 0.11
	†	33.8 + 0.2	14.37 + 0.26	2.77 + 0.11
0.50	*	41.3 + 0.3	19.77 + 0.35	4.07 + 0.18
	†	41.5 + 0.3	20.01 + 0.36	4.17 + 0.19
1.00	*	57.2 + 0.3	33.41 + 0.50	7.62 + 0.27
	†	57.5 + 0.3	33.59 + 0.47	7.53 + 0.22
2.00	*	90.1 + 0.4	70.56 + 0.77	18.65 + 0.44
	†	91.0 + 0.4	72.25 + 0.79	19.38 + 0.45

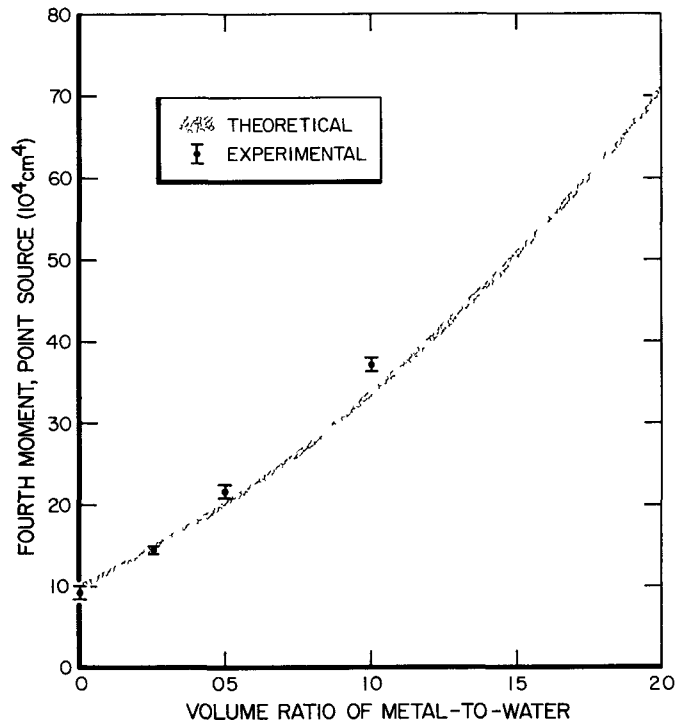
*Calculation does not include impurities.

†Calculation does include impurities.



7604-1560

Figure 1.



7604-1561

Figure 2.

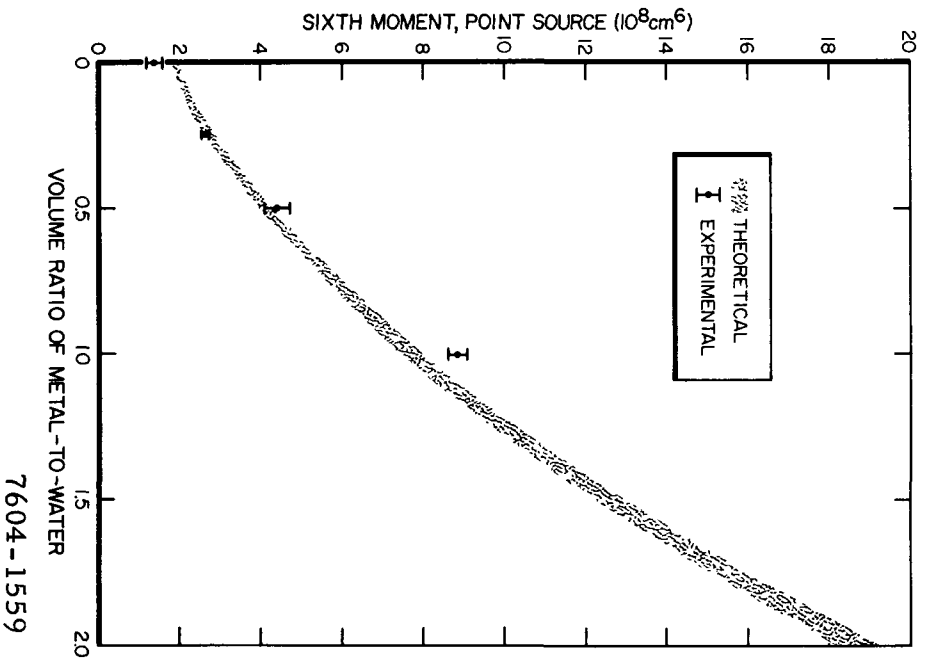


Figure 3.

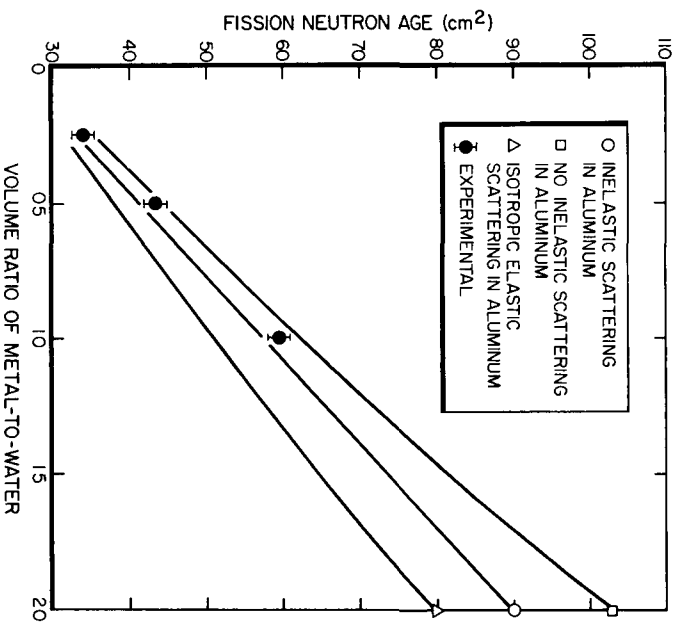


Figure 4.

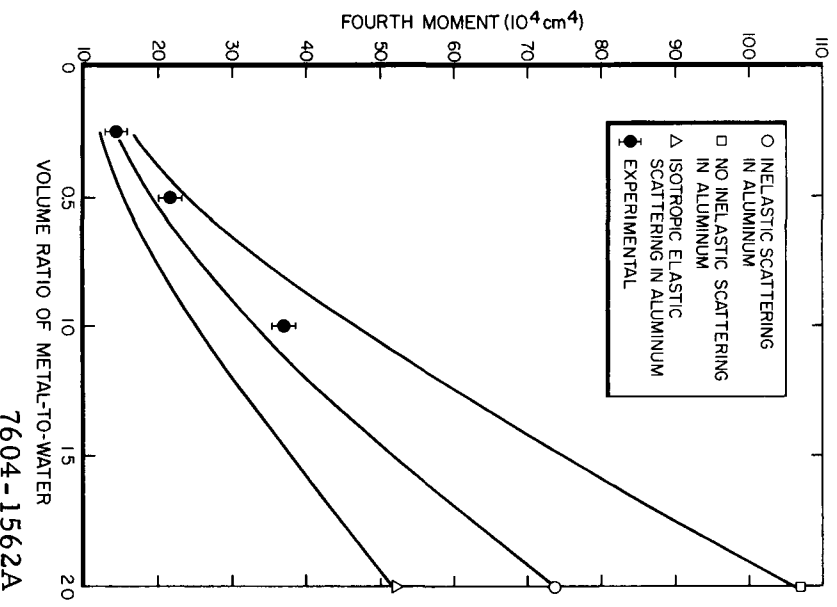
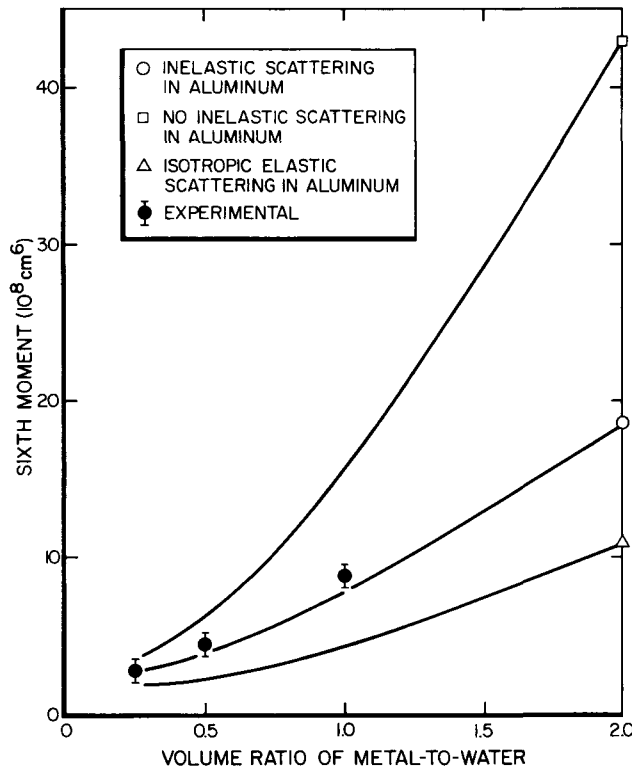


Figure 5.



7604-1563A

Figure 6.

both previous calculations⁶ and measured values,⁷ are presented in Table 6. An analysis and comparison of the several sets of oxygen data is given in NAA-SR-MEMO-11239.

TABLE 6
 NEUTRON AGE AND HIGHER MOMENTS IN WATER

	Age (cm ²)	$M_4(10^4 \text{ cm}^6)$	$M_6(10^8 \text{ cm}^6)$
Experiment	26.46 + 0.32	9.34 + 0.50	1.37 + 0.20
AIENDF	26.14 + 0.19	9.67 + 0.22	1.76 + 0.10
KAPL	26.46 + 0.18	9.90 + 0.20	1.80 + 0.10

III. EVALUATION OF EFFORT TO DATE

Theoretical calculations of the neutron age and higher moments of the slowing-down distribution for several aluminum-water mixtures were compared with results of the experimental program of age measurements. The reasonable agreement between measured and calculated values for the neutron age and fourth moments indicates that the cross-section and associated angular data used in the calculations are satisfactory. The discrepancy between the measured and calculated sixth moments is believed due to uncertainty in the extrapolation of the

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measured distribution to infinity. While the contribution of the extrapolated portion of the curve is negligible for the case of the second and fourth moments, it represents a large fraction of the measured sixth moment. The value of the measured sixth moment is highly sensitive to the method of extrapolation.

The technique of measurement and the theoretical analysis have been developed to a high degree, permitting routine measurements of the age and higher moments with high precision. The total, the elastic scattering, and the differential elastic scattering cross-sections for oxygen, hydrogen, carbon, zirconium, iron and aluminum have been validated to a satisfactory degree from 1 to 10 Mev.

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Program: General Research, Reactor Physics Studies	
Project: Extrapolation Length Measurements	
Project Engineer: H. A. Morewitz	
Reporting Period: July-September 1965	AEC Category: 04-40-01-01
General Order: 7604	Subaccount: 1312

I. PROJECT OBJECTIVES

The objective of this project is to investigate the diffusion properties of neutrons in different materials such as water, graphite, and beryllium, using both steady state and pulse techniques. As the neutron temperature from a steady state source is different from that of a pulsed-source, a comparison between the two results provides a check on the accuracy of the scattering kernel. The technique is a fairly sensitive test, as small changes in the geometric size of the medium cause correspondingly small changes of the asymptotic neutron spectrum. A good scattering theory should be able to predict the diffusion parameters which may be calculated from these experiments.

II. PROGRESS DURING REPORT PERIOD

A. PULSED MEASUREMENTS IN DIPHENYL

Pulsed neutron measurements in diphenyl at 85, 105, and 24°C have been completed and the data analyzed, using both the conventional 3-parameter fit of λ versus β^2 and the heat transfer approach. Table I summarizes the results. Average density of the solid was 1.066 gm/cm³. Density of the liquid was about 0.99 gm/cm³ at 85°C, and 0.98 gm/cm³ at 105°C.

TABLE I
SUMMARY OF PULSED MEASUREMENTS IN DIPHENYL

Power Series Fit				
Temperature	$\lambda_0 (10^3 \text{ sec}^{-1})$	$D_0 (10^4 \text{ cm}^2 \text{ sec}^{-1})$	$C (10^4 \text{ cm}^4 \text{ sec}^{-1})$	$Y (10^4 \text{ sec}^{-1})$
24	3.114 ± 0.019	4.84 ± 0.05	2.19 ± 0.21	—
85	2.901 ± 0.033	6.26 ± 0.08	4.29 ± 0.43	—
105	2.854 ± 0.029	6.59 ± 0.08	3.72 ± 0.43	—
Heat Transfer Fit				
24	3.108 ± 0.029	4.90 ± 0.08	—	4.09 ± 0.70
85	2.895 ± 0.022	6.37 ± 0.07	—	3.15 ± 0.27
105	2.829 ± 0.026	6.73 ± 0.08	—	4.16 ± 0.50

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A final article to be submitted for publication is in preparation. A talk on the diphenyl results will be presented at the Washington, D. C., meeting of the ANS.

B. PULSED MEASUREMENTS IN GRAPHITE

The variation of the diffusion length of thermal neutrons in graphite with temperatures was measured initially at Hanford⁽¹⁾ and later by Zherzerun and Korolev.⁽²⁾ The Hanford measurements covered a temperature range from 22 to 600°C. After corrections were made for changes in graphite density and nitrogen content, the measurements were fitted to the expression

$$\frac{L^2}{L_o^2} = \left(\frac{T}{T_o}\right)^w \quad \dots(1)$$

with $w = 0.48 \pm 0.005$ (where the standard error rather than the quoted 95% confidence limit of 0.01 is used). If we assume $v\Sigma a$ is a constant,

$$\frac{D}{D_o} = \left(\frac{T}{T_o}\right)^w \text{ or } \frac{\lambda_{tr}}{(\lambda_{tr})_o} = \left(\frac{T}{T_o}\right)^w - 1/2 \quad \dots(2)$$

where T_o is the reference (room) temperature.

For this type of measurement, the transport mean free path has a slight temperature variation equal to

$$\left(\frac{T}{T_o}\right)^{-0.02 \pm 0.005}$$

The Russian measurements confirmed this dependency up to 350°C and, in addition, an auxiliary experiment was performed. To separate the variation caused by the change in the temperature of the spectrum from the effect caused by the thermal vibrations of the scattering atoms, a beam transmission measurement of the scattering cross section was performed. The temperature of a 2-cm-thick disk of graphite and the exit temperature of a neutron beam from a thermal column were varied independently. When the temperature of the spectrum was varied from 10 to 362°C with the temperature of the scattering disk constant, no change was observed in the scattering cross-section. With the spectrum at room temperature, a 3.5% increase in the scattering cross-section was observed for a change in disk temperature from 18 to 340°C. Within the experimental error, the variation of the transport mean free path of graphite may be ascribed entirely to the temperature change of the moderating atoms and is independent of the spectrum temperature. Baumann⁽³⁾ has introduced a convention to indicate both the moderator and spectrum dependence. If the diffusion coefficient is written as $D(T_o, T_n)$, the first temperature term is that of the moderator and the second that of the spectrum.

In both static and pulsed diffusion length experiments, the difference between the spectrum temperature and the moderator temperature was not taken into account. For an infinite medium, diffusion heating occurs and the spectrum is

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warmer than the Maxwellian at moderator temperature. In the Hanford measurements, the finite size of the graphite stack was such that the leakage caused the spectrum to be slightly cooler than the moderator temperature at the higher temperatures where L is large and warmer near room temperature; however, $w = 0.48 \pm 0.01$ is probably valid for spectrum and moderator at the same temperature, where the standard error has been doubled.

In experiments at room temperature where the spectrum is hardened by poisoning, the transport mean free path must be regarded as constant. In pulsed measurements where diffusion cooling occurs, the transport mean free path may vary if the spectrum is cooled enough so Bragg scattering occurs; however, valid decay constants may not be obtained when crystalline effects become important.⁽⁴⁾ For most of the valid range of pulsed decay data in graphite, the best assumption is that the transport mean free path is constant or $w = 1/2$.

In the previous spectral correction analyses^(5,6) of pulsed decay data in graphite, a value of $w = 0.48 \pm 0.01$ was assumed. In this approach a least squares fit is made to $v\Sigma a$, $D_0(T_0, T_0)$, and γ , the heat transfer coefficient from moderator to the neutron spectrum (in place of C the diffusion cooling coefficient). To the first approximation w^2/γ is constant, so γ is sensitive to the input value of w , while $v\Sigma a$ and $D_0(T_0, T_0)$ are practically unaffected. For a buckling range of $-3.685 \times 10^{-4} \text{ cm}^{-2}$ (steady state point) to $1.057 \times 10^{-2} \text{ cm}^{-2}$ in graphite of density 1.689 g/cm^3 , the values obtained using the latest technique⁽⁶⁾ are listed in Table 2.

TABLE 2
PULSED DIFFUSION PARAMETERS AT 23.5°C
GRAPHITE ($\rho = 1.689 \text{ g/cm}^3$)

w	0.50 ± 0.01	0.48 ± 0.01
$v\Sigma a(\text{sec}^{-1})$	77.76 ± 0.47	77.79 ± 0.47
$D_0(10^5 \text{ cm}^2 \text{ sec}^{-1})$	2.0942 ± 0.0056	2.0940 ± 0.0055
$\gamma(\text{sec}^{-1})$	1772 ± 99	1620 ± 97

The preferred value of γ is $1772 \pm 99 \text{ sec}^{-1}$ and the inverse of γ is the thermalization time constant, $\tau = 564 \pm 32 \mu\text{sec}$. Corrected to a density of 1.6 g/cm^3 , the value is $\tau_{1.6} = 595 \pm 34 \mu\text{sec}$.

C. STEADY STATE EXTRAPOLATION LENGTH MEASUREMENTS

A detailed analysis is being made of the data from the horizontal measurements in the 36- by 36-in. graphite stack and the 28- by 28-in. stack in an attempt to resolve the discrepancies in the resulting extrapolation lengths. Several possibilities are being investigated: (1) 5th harmonic effects, (2) heterogeneous stack density, (3) effect of micarta under the stack, (4) foil orientation, (5) foil-holder effects, and (6) foil calibrations.

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III. EVALUATION OF EFFORT TO DATE

The pulsed measurements in diphenyl have been completed, and a final report is being prepared for publication. The report on the pulsed measurements in graphite have been published previously. This completes the pulsed neutron studies in support of the project objectives. The program is progressing satisfactorily.

IV. NEXT REPORT PERIOD ACTIVITIES

The static measurements in graphite are continuing in order to resolve some apparent discrepancies in the measured extrapolation lengths.

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Program: General Research, Reactor Physics Studies	
Project: Cross-Section Analysis and Applications Project	
Project Engineer: H. A. Morewitz	
Reporting Period: July-September 1965	AEC Category: 04-40-01-01
General Order: 7604	Subaccount: 1313

I. PROJECT OBJECTIVES

The objectives of this project are related to the production of a good, up-to-date, basic set of nuclear data. To accomplish this we intend to: (a) survey, analyze, and compile existing experimental and theoretical information on neutron cross-sections and angular data; (b) provide automated accessibility to users of cross-section data (microscopic data, group libraries); (c) provide automated reference to and retrieval of cross-section information; and (d) use nuclear model calculations to help fill gaps in cross-section data.

II. PROGRESS DURING REPORT PERIOD

A. LIBRARY DEVELOPMENT

A multigroup library in 35-group structure was produced for use with the CAESAR¹ code. This library is designated as the "standard library," having a 1/E spectrum weighing of up to 3.29 mev and a fission spectrum weighing from 3.29 to 10 mev. Further work on multigroup library development will continue following the updating and expansion of the Atomics International Evaluated Nuclear Data File (AIENDF). The latter project will be completed by January, 1966.

B. THEORETICAL MODEL ANALYSIS AND DATA DEVELOPMENT

A revised set of calculated nuclear data has been produced for Pu²³⁸ and Cm²⁴⁴. Negative energy resonance parameters in the single-level Breit-Wigner approximation were used to calculate thermal neutron data. Measured resonance parameters were used in the resolved resonance region, while in the unresolved resonance region average resonance parameters derived from the resolved data, were used. A nonspherical optical model was used to generate fast neutron cross-sections; capture, fission and inelastic scattering were assumed to proceed by means of the compound nucleus mechanism yielding to a Hauser-Feshbach statistical treatment. Group constants in 16- and 35-group structures, for Pu²³⁸ and Cm²⁴⁴, are presently being prepared for the multigroup diffusion and transport theory neutronic codes at Atomics International.

C. PROCESSING PROGRAM DEVELOPMENT

1) A number of IBM 7094 computer programs which further enhance the automation of the AI cross section project, were written or modified and checked out. These include ADT, a program which maintains, updates, and corrects the AI angular data tape; AROCS, a code to average rapidly oscillating cross-sections (i. e., the sodium data of Hibdon) and thus allows the use of an average cross-

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section on the coarser AIENDF energy grid; the usefulness of the MALINDA code was extended by simplification of input and the addition of log-log and log-linear interpolation subroutines.

The work of combining GRAVE,² TRIXIE,³ PRISM,⁴ and CONTRACT⁵ into the comprehensive program GRISM is nearing completion. Problems of intercode compatibility have been resolved and some initial runs of the GRAVE-TRIXIE combination have been rewarding. The addition of PRISM and CONTRACT will permit this package of programs to generate multigroup libraries from start to finish in minimum time with maximum performance.

The tape conversion program, TCP, which permits intercomparison between the Aldermaston and Atomics International Evaluated Nuclear Data Files has been extended to permit comparisons of the AIENDF with the Los Alamos evaluated nuclear data files. Further extension of this program to permit comparison of the AI data files with those of the Livermore Radiation Laboratory and Brookhaven National Laboratory is currently underway.

A computer code to generate Doppler broadened point cross section data in the resonance region was written and checked out. This program has been used to generate room temperature data for U²³⁵ and U²³⁸ based on the zero degree Kelvin Karlsruhe⁶ data. The theory and related programming of this code is currently being documented.

D. AI NUCLEAR DATA FILE

Evaluated data from the Aldermaston Data File has been retrieved, punched on data cards and plotted. This data will be used in the forthcoming updating and expansion of the AIENDF. The elements taken from the Aldermaston data tape are as follows.

Tritium	Fluorine	Calcium	Th-232
Helium 3, 4	Aluminum	Copper	U-233
Lithium 6, 7	Silicon	Cadmium	U-234
Beryllium	Sodium	Titanium	U-236
Boron 10	Chlorine	Zirconium	Pu-239
Carbon	Chromium	Gallium	Pu-240
Nitrogen	Iron	Xenon	Pu-241
Oxygen	Nickel	Lead	

A comparison of measured and calculated infinitely dilute resonance integrals for the fissionable isotopes U²³³, U²³⁵ and Pu²³⁹ has been undertaken so that criteria and procedures for determining the amount of smooth background capture and fission cross-sections to be added to the single-level resonance calculation may be assessed. Calculations for the fissionable isotopes, based on the single-level Breit-Wigner formalism underestimate the resonance integral. Evidence indicates that resonance integral calculations based on multilevel formalism tends to minimize this discrepancy. Efforts, as yet unsuccessful, are being made to obtain the series of multilevel Fortran programs developed by the Adler's.^{7,8}

A considerable amount of elastic scattering angular data has been compiled preliminary to the updating and expansion of the AIENDF. In all cases, the differential scattering data is converted into a Legendre expansion of order nine or higher. The Legendre coefficients are then used to reconstitute the cross section. The following nuclides have been successfully analyzed by the foregoing process.

Tritium	Fluorine	Copper	Tantalum
Helium 3, 4	Magnesium	Zinc	Tungsten
Lithium 6, 7	Silicon	Niobium	Lead
Nitrogen	Calcium	Cadmium	Titanium

The initial data was taken primarily from BNL-400⁹; however, more recent measured information as well as theoretical calculations are being used to fill gaps. Documentation of data sources and methods of evaluation and compilation is proceeding.

III. EVALUATION OF EFFORT TO DATE

Satisfactory progress is being made in the efforts associated with the updating and expansion of the AIENDF. The extension of the tape conversion program to treat data files of other installations has greatly increased the utilization by the cross-section project of available nuclear data files. The integration of the various individual processing programs for the production of multigroup libraries, has proceeded to the point where optimal production of multigroup libraries, in terms of computer time and personnel effort, will soon be achieved.

IV. NEXT REPORT PERIOD ACTIVITIES

Programming, checkout and documentation of GRISM will have been completed. Efforts associated with the updating and expansion of the AIENDF will continue, as will the programming work related to converting the Livermore and Brookhaven data files. Documentation of theoretically determined Pu²³⁸ and Cm²⁴⁴ cross-sections will be finished. Methods for the production of higher-order Pn anisotropic scattering matrices for use in transport theory codes will be developed.

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Program: General Research, Reactor Physics Studies	
Project: Fast-Spectrum Doppler Measurements	
Project Engineer: H. A. Morewitz	
Reporting Period: July-September 1965	AEC Category: 04-40-01-01
General Order: 7604	Subaccount: 1330

I. PROJECT OBJECTIVES

The objectives of this project are to: (a) refine further the techniques originally developed at this laboratory for measuring Doppler reactivity coefficients; (b) investigate the Doppler coefficients of various fuel and fertile materials in a variety of fast spectra; (c) provide better understanding of temperature-induced reactivity changes by analytically and experimentally investigating these changes at cryogenic as well as very high temperatures; and (d) investigate resonance-overlap and interference effects, and the effect of varying the surface-to-mass ratio.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

Measurements of reactivity and temperature coefficients have continued in Core 13, with a 30-vol % carbon test region. The temperature coefficients of tungsten, tantalum, thorium (metal and oxide), U^{235} (metal, oxide, and carbide), 22% enriched uranium, and U^{238} (metal and oxide) have been measured in the standard core and in some polyethylene blankets. Improved blankets have been constructed. Apparatus has been assembled for measurements down to 20°K.

III. PROGRESS DURING REPORT PERIOD

The temperature coefficients of several nonfissile materials were measured in the standard core and in some polyethylene blankets. These materials are tungsten, tantalum, U^{238} (metal and oxide), thorium (oxide and 0.25-in.-diameter metal). The expansion coefficient for these materials is generally a small fraction of the total temperature coefficient and so these measurements closely approximate the Doppler coefficients.

The measurements of the tungsten reactivity have been corrected for expansion by a calculated value for the expansion reactivity. The Doppler effect of a 187-gm sample is $-2.9 \times 10^{-5} \text{ } \beta / ^\circ\text{C}$ in the standard core and $-1.1 \times 10^{-4} \text{ } \beta / ^\circ\text{C}$ in the 1/2-in. polyethylene blanket, between 20 and 800°C.

A sample composed of $U^{235}O_2$ powder in an Invar capsule has been used in an attempt to reduce or eliminate the expansion effect. Invar has a very low thermal expansion in the temperature range involved and so constrains the fissile material. A negative temperature coefficient was measured in the 1/8-in. polyethylene blanket and also in the 3/8-in. polyethylene blanket, greater in magnitude than that of the standard size metal sample. However, the sample is much less dense than planned (20% TD), resulting in a very large mass correction; and the powder is sufficiently loose that it shifts during oscillation.

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Measurements of the reactivity of a 1/4-in.-diameter U^{235} sample in the 1/2-in. polyethylene blanket show very clearly the competing effects of expansion and Doppler broadening. In this core the expansion effect is positive and linear with temperature, while the Doppler effect is negative and varies approximately as the square root of the temperature above 20°C. Thus, the reactivity change is negative initially, reaches a minimum, passes through zero at about 500°C, and becomes positive.

A thorough study of the polyethylene blankets with thermal and threshold fission counters has shown that the flux and spectrum inside the blanket are severely affected by a gap in the polyethylene caused by the faces of the fuel drawers. In addition, these blankets are poorly represented by the spherical approximation used in the reactor calculations.

New 1/8-in. and 1/2-in. blankets have been constructed to improve the uniformity of the neutron flux. These blankets are longer and extend continuously across the parting plane of the reactor. Also, some parts of the blanket not directly surrounding the sample have been eliminated. Fission counter traverses through the new blankets indicate that the flux and spectrum are much more uniform across the sample space than in the old blankets. A repetition in these new blankets of the experiment to determine the reactivity coefficient of expansion of U^{235} indicates, however, that this coefficient does not change significantly between the old and new blankets; i. e., the change is less than 10%.

The cryogenic apparatus using the Cryotip refrigerator has been operated successfully, cooling a stainless steel sample to 60°K with an axial temperature difference of 8°K.

Twelve spherical photoneutron sources have been fabricated for source-importance measurements. The sources are Sb-Be, La-Be, Na-Be, La-D, Na-D.

A prototype high-temperature oven has been constructed. This oven has operated satisfactorily at above 1250°C. This appears to be the upper operating limit of some of the materials used, and so more stable replacements have been obtained.

IV. EVALUATION OF EFFORT TO DATE

Additional methods for determining or reducing the expansion effect in temperature coefficient measurements have been developed. The temperature range in which measurements may be made has been greatly increased by development of the high-temperature oven and the cryogenic refrigerator.

V. NEXT REPORT PERIOD ACTIVITIES

Comprehensive investigation and improvement of the moderating blanket technique will be conducted. Measurements with the cryogenic apparatus will be made in the temperature range where crystal binding effects may be observable. The neutron importance shape in all core configurations will be measured with small photoneutron sources. The next core loading will be planned. Apparatus may be borrowed for spectrum measurements between 10 kev and 2 Mev by use of hydrogen-filled proportional counters.

ATOMICS INTERNATIONAL

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Program:	Reactor Fuels and Materials; High Temperature Materials Development		
Project:	Structure, Properties, and Fission Effects in UC and Other Refractory, Interstitial Fuels		
Project Engineer:	C. E. Weber		
Reporting Period:	July-September 1965	AEC Category:	04-40-02-09
General Order:	7551	Subaccount:	4539

I. PROJECT OBJECTIVES

The objectives of this project are to: (a) use and develop, where necessary, the most advanced methods to prepare pure, single and polycrystalline material of controlled structure and stoichiometry; (b) precisely determine selected microstructural attributes and mechanical and physical properties required both to establish the potential for these materials as nuclear fuels and to provide a basis for interpreting and understanding irradiation behavior in terms of atomistic transport phenomena; (c) apply current solid state theory developed for metallic fuels to obtain an understanding of the processes and mechanisms involved in fission damage in order, and (d) use knowledge gained therefrom, and also apply to these materials the general theory and experimental techniques developed to reduce and control swelling in metallic fuels and demonstrate such control in refractory fuels.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

A study of the effects of electron-beam traverse-rate and rotation on the quality of single crystals grown by zone-refining has resulted in the production of a single-phase single crystal of UC directly by zone-refining, without a decarburization step. The key was the use of a fast traverse rate to minimize uranium vapor loss.

Analysis of data on induction zone-melting has permitted calculation of the shape of the phase diagram near the UC composition at its melting point. The shape agrees with the currently accepted diagram, which has been estimated from other types of data.

Further work on quenching UC compositions containing tungsten has shown that tungsten is soluble to the extent of several per cent at 2100°C, and that most of the tungsten can be retained in solution on quenching.

Low-pressure oxidation of UC has disclosed some interesting effects and some important differences between the behavior at 900°C and that at 1800°C. A paper giving the preliminary results has been accepted for publication in Acta Metallurgica, and the work is continuing.

A suitable experimental procedure for simultaneous measure of self-diffusion of U and C in UC has been evaluated. The method involves deposition of U²³³ and C¹⁴ on the UC surface and proportional counting to determine the concentrations of these isotopes in sections removed after diffusion annealing.

III. PROGRESS DURING REPORT PERIOD

A. PREPARATIVE RESEARCH

A series of UC single-crystal, electron-beam, zone-refining studies in which the rotation rate and zone traverse rate were varied has been completed. The most straightforward result is that rotation interfered with the perfection of the single crystals grown; and, at 250 rpm, the product was definitely polycrystalline. With no rotation, the product was a single crystal in which the subgrains showed less than a 2° mismatch. Crystals grown at 60 rpm were single but with subgrains mismatched by 3 or 4° and with correspondingly more twist and strain. The smoothness of cleavage in these crystals also reflected these differences.

Attempts to make a perfect crystal by second-pass zone-refining and by zone-annealing at a temperature just below the melting point were unsuccessful.

The purpose of these experiments is to prepare UC single crystals which are sufficiently flawless to permit the use of x-ray microscopy methods for the study of radiation damage. Presently available crystals have so many imperfections that any structural changes due to irradiation are obscured. The new crystals with about 2° of tilt are not yet perfect, and cannot be used for such studies.

Chemical analyses of the series mentioned above are shown in Table 1. There are no clear trends indicating any effect of rotation rate on composition. There is an indication that slower beam traverse rate causes more loss of uranium (as shown by higher carbon analysis). Changes in nitrogen content are erratic, and changes in oxygen are substantial but puzzling. The large oxygen content found for samples 4-9 could not have been picked up during melting because it can be calculated from kinetic theory that the maximum pickup at the measured pressure of 5×10^{-6} torr can be only 10 ppm. The metal analyses results are also puzzling in that there was essentially no purification with respect to Fe, Mg, and Ti at the slowest traverse rate, but substantial purification at the higher rates. Perhaps this is related to carbon content in which case the UC₂ platelets could conceivably serve as a trap for impurities.

One sample prepared at a 6-in./hr traverse rate lost no uranium and ended up as a stoichiometric or slightly hypostoichiometric single crystal in the electron-beam zone melter. If this result proves to be reproducible, it might be possible to eliminate the hydrogen decarburization step in preparing single-phase zone-refined UC, by using a fast traverse rate and low initial carbon. The carbon content shown in Table 1 was determined chemically, but the single-phase characteristic was shown by metallography, Figure 1, which also shows that some uranium was lost during zone-refining, causing the top end of the crystal to contain a few UC₂ platelets.

Data reported earlier on induction zone-refining in inert gas atmosphere (such that essentially no uranium is lost by vaporization) now have been evaluated in terms of conventional zone-melting parameters in order to get a better understanding of the zone-melting process and at the same time to learn what the data signify with regard to the U-C system near the melting point of UC.

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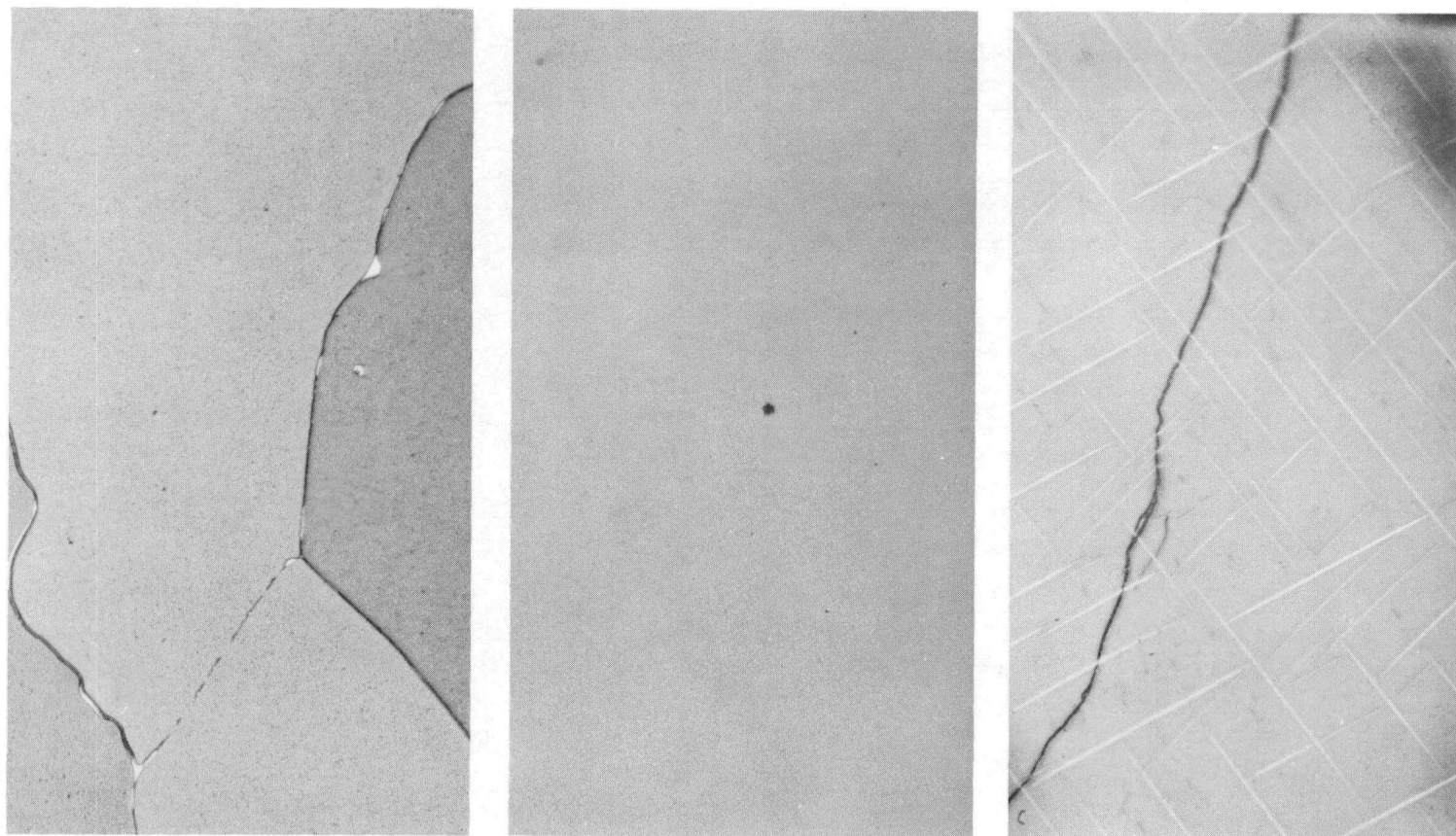
TABLE 1
 ZONE REFINING OF UC IN AN ELECTRON BEAM ZONE MELTER

Sample Number	Nominal Starting Carbon Content (wt %)	Zone Refining Traverse Rate (in./hr)	Base Rod Rotation Rate (rpm)	Carbon (wt %)	Analyses Results (ppm)					Crystal- linity
					Nitro- gen	Oxy- gen	Metals*			
							Fe	Mg	Ti	
260192-control	4.70-4.80	-	-	4.76	245	60	50	5	15	Poly
260192-1	4.75	1	0	5.09	87	20	50	3	10	Single
260192-2	4.70		60	5.16	40	25	50	5	10	Single (?)
260192-3	4.79		250	5.16	72	50	50	2	10	Poly
260192-4	4.70	3	0	5.05	155	700	<10	<2	<10	Single
260192-5	4.75		60	5.05	98	930	35	5	15	Single
260192-6	4.75		250	5.02	80	1015	10	2	10	Poly
260192-7	4.80	6	0	5.00	42	1380	10	<2	<10	Single
260192-8	4.75		60	4.74	168	685	<10	<2	<10	Single
260192-9	4.80		250	4.98	100	1120	15	<2	<10	Poly

*All other metals below detection limit

Single-pass zone-refining will reduce the concentration of an impurity only over the first part of the crystal, extending only a few zone lengths, depending on the value of the distribution coefficient, k , for the ratio of the concentration of the impurity in the solid to its concentration in the liquid. The data agree with this picture, showing essentially no change in carbon concentration for the top or last portion of the zone-refined rod, compared with the original, as-cast carbon concentration, but a slight shift for the bottom or first portion. The shift is in the direction of the stoichiometric value, 4.80 wt % C, as expected for congruent melting of UC.

The data shown in Tables 2a and 2b are for five of the six zone-refining runs reported earlier (Annual Report FY 65, NAA-SR-11450). The sixth was eliminated because of an unusually high oxygen analysis, 960 ppm, compared with less than 75 ppm for the others. The carbon analyses shown are "effective carbons" taking into account oxygen and nitrogen which are assumed to substitute atom-for-atom in the carbon sublattice of UC. The changes shown by carbon analysis are small but are significant with respect to the reported precision of 0.002%. The quantities X_u and X_c are mole fractions of excess uranium and carbon respectively, when treating excess uranium as the impurity in UC for hypostoichiometric compositions and excess carbon as the impurity in hyperstoichiometric composition. $(X_u)_0$ is the initial mole fraction of excess uranium, $(X_u)_{4\ell}$ is the same value at an average distance up the zone-refined rod of four zone lengths, and $(C/C_0)_{4\ell}$ is the ratio of the two.



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Figure 1. Electron-Beam Zone-Melting at Fast (6-in./hr) Traverse Rate
(left, as-cast; center, Zone-refined - bottom;
and right, Zone-refined - top)

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TABLE 2a
 REMOVAL OF EXCESS URANIUM DURING ZONE-REFINING UF UC

Traverse Rate (cm/sec)	Carbon (wt %)		$(X_u)_0$	$(X_u)_{4\ell}$	$(C/C_0)_{4\ell}$	k
	As-cast	Zone-refined bottom				
3.5×10^{-4}	4.505	4.598	0.062	0.043	0.69	0.2
2.8×10^{-3}	4.545	4.564	0.055	0.051	0.093	0.5
3.5×10^{-4}	4.77	4.78	0.007	0.005	0.72	0.2

TABLE 2b
 REMOVAL OF EXCESS CARBON DURING ZONE-REFINING OF UC

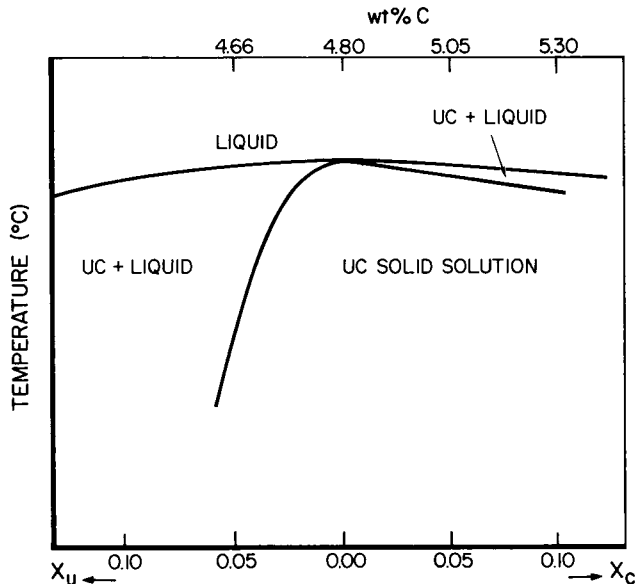
Traverse Rate (cm/sec)	Carbon (wt %)		$(X_c)_0$	$(X_c)_{4\ell}$	$(C/C_0)_{4\ell}$	k
	As-cast	Zone-refined bottom				
3.5×10^{-4}	5.22	5.16	0.084	0.073	0.87	0.4
2.8×10^{-3}	5.17	5.107	0.075	0.075	0.87	0.4

The values shown in the last column are the effective distribution coefficients calculated from the C/C_0 values at a distance of four zone lengths, using the relation,

$$C/C_0 = 1 - (1-k)e^{-kx/l} \quad (x = \text{length})$$

or plots of this equation given by Pfann (W. G. Pfann, Zone Melting, John Wiley & Sons, New York 1958). The effective distribution coefficient is always larger than the equilibrium distribution coefficient for finite growth rates and the relation depends on the parameter, $f\delta/D$, in which f is the growth rate, δ is the thickness of the laminar flow region at the solid/liquid interface, and D is the diffusion coefficient in the liquid. About all that can be concluded from the very few data points now available is that the equilibrium distribution coefficient (the coefficient which applies to the phase equilibrium diagram) is only slightly less than 0.2, perhaps 0.15, on the uranium-rich side, and very close to 0.4 on the carbon-rich side. These figures are in qualitative agreement with published data on the U-C phase diagram but indicate a much better agreement with the higher UC-U solubility values given by Magnier and Accary (1963 Harwell Conference on Carbides in Nuclear Energy) than with the values given by Buckley (AERE-R-3872, November 1961). Buckley's distribution coefficient values vary from about 0.03 to 0.05, while those calculated from Magnier and Accary's data are about four times as great. The general shape of the phase diagram in the

neighborhood of the melting point of UC is shown in Figure 2 based on the distribution coefficients given in Tables 2a and 2b. The shape agrees with that reported in the literature, for example, that in Storm's recent review (E. K. Storms, LA-2942, 8/13/64). Temperatures have been omitted in Figure 2 because the zone-melting data did not include temperature measurement and because a range of values for the melting point of UC have been reported.



The above is given not with the idea that the results represent a more certain measure of phase equilibria than that in published data, but as an indication of how zone-melting data can be used to estimate phase relations for reactive high melting systems. Zone-melting experiments over a wider composition range, coupled with temperature measurements of the molten zone, would indeed constitute an independent determination of the phase diagram.

Figure 2. Phase Diagram Determined with Zone-Melting Data 7551-2528

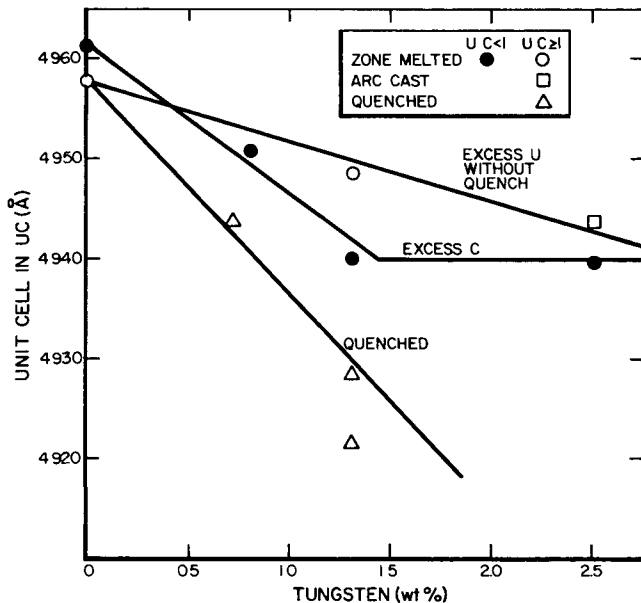
B. TUNGSTEN SOLUTIONS AND DISPERSIONS

A rough draft of a paper, "Preparation of UC with Fine Dispersions of Refractory Metals" has been completed. Three methods of preparing UC containing precipitates are described: (1) induction zone melting, (2) arc-melting, and (3) arc casting. In all three processes, precipitates are formed when tungsten is added to UC having a U:C ratio of one or less, but no precipitates are observed in material with a U:C ratio of less than one; that is, no precipitates are observed in material in which UC₂ platelets are present. The microstructure of the monocarbide phase is markedly affected in material having a U:C ratio of one or greater; the grain boundaries become irregular and the grain size is smaller by a factor near ten.

Figure 3 shows the effect on the uranium carbide lattice of adding tungsten to materials varying in U:C ratio and of using different cooling rates. The lattice constant decreases with the addition of tungsten in all cases, but there is more decrease in materials with a U:C ratio of less than one. When samples with a U:C ratio greater than one are rapidly cooled by quenching in a low temperature liquid metal, the lattice parameter is much further contracted. The implication is that tungsten dissolves in UC and causes a lattice contraction, but that more tungsten dissolves at higher carbon content.

On the basis of recent reactor irradiation tests on metal fuels, control of fission gases may depend on the presence of a dissolved material capable of forming precipitates during irradiation. This postulation gives incentive to our efforts to obtain phase equilibrium data on the system U-C-W and similar systems at low additive concentrations. In order to prepare solid solutions of known supersaturation, the phase equilibrium diagram must be known. Our progress on determining the U-C-W phase diagram has included testing of annealing and quenching procedures and the preparation of two series of samples, one on the

UC-W join, and the other on the UC-WC join. The work remaining is the actual annealing and quenching of these, followed by metallography and x-ray to characterize phases and structures.



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Figure 3. Change in Lattice Parameter with Tungsten Content

The first group of samples was prepared using UC feed stock containing 5.2 to 5.3% carbon and tungsten additions of 0, 1, 2, 3 and 4% tungsten. The second group was prepared using UC containing 4.75 to 4.80% carbon and tungsten additions of 0, 1/2, 1, 2, and 3% tungsten. The tungsten was incorporated into the UC in the induction zone refiner by wrapping the feed rods with tungsten wire with a density such as to give the desire addition, except that the first 1/4 in. was wound with an additional amount of wire such as to provide twice the nominal addition. This figure was based on the effective distribution coefficients of 2.0 to 2.4 observed for tungsten in UC during previous work. This procedure is called zone leveling.

Chemically determined compositions of these samples are shown in Table 3, for 1/4-in. sections cut at various positions along the rods. In some cases the sections were cut beyond the tungsten zone, but otherwise there are substantial lengths showing rather uniform tungsten and carbon composition such that a number of 1/4-in. sections of each composition are available for annealing and quenching experiments.

As a part of testing the procedure, and to obtain a preliminary indication of the phase diagram, three quench tests were run with six samples in each test; lattice parameter results are shown in Table 4. The results do not follow a regular order in all cases, and there is some possibility of a mixup in samples in one or two cases. It may be that the data will prove to be consistent when variations in carbon content and temperature are considered and observations are made of phases. The results show that tungsten is soluble in UC at 2100°C, probably to the extent of at least 4 wt % W; these results are consistent with the similar data of Figure 3.

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TABLE 3
 COMPOSITION OF ZONE-MELTED RODS PREPARED FOR PHASE
 EQUILIBRIA IN THE U-C-W SYSTEM (wt %)

Rods	C or W	Initial End	Middle		Final End
UC _{1.1} ^{-W} Series	C	4.97	5.10		-
	W	-	-		-
	C	5.01	5.09		5.02
	W	1.10	1.02		1.01
	C	4.82	4.98		4.97
	W	1.60	2.18		1.67
	C	5.15	4.98		5.01
	W	2.65	-		3.01
	C	4.79	4.84		4.80
	W	3.52	-		3.60
UC _{1.1} ^{-W} Series	C	4.83	4.80	4.86	4.85
	W	0.08	0.05	0.05	0.05
	C	4.74	4.64	4.61	4.60
	W	0.43	0.42	0.45	0.03
	C	4.70	4.68	4.64	-
	W	0.63	0.01	1.01	-
	C	4.70	4.58	4.56	4.62
	W	1.66	1.74	1.53	0.20
	C	4.73	4.67	4.57	-
	W	1.94	2.43	2.47	-

TABLE 4
LATTICE PARAMETER VARIATIONS IN THE UC-W SYSTEM

Tungsten (wt %)	Carbon (wt %)	UC Lattice Parameters After Annealing (Å)		
		1 hr at 2100°C	2 hr at 2100-2175°C	4 hr at 2100°C
0.11	4.61	4.957	4.955	4.956
0.64	4.56	4.953	4.940	4.953
1.11	4.47	4.914	4.946	4.948
2.31	4.54	4.939	4.952	4.943
3.98	4.43	4.924	4.915	4.918
4.34	4.56	4.941	4.919	4.925

C. REACTION OF CARBIDES WITH GASES

A paper on the subject of UC-O₂ interactions has been written and approved for publication in *Acta Metallurgica*. The paper covers observations on kinetics and structural changes at UC surfaces exposed to low oxygen pressures.

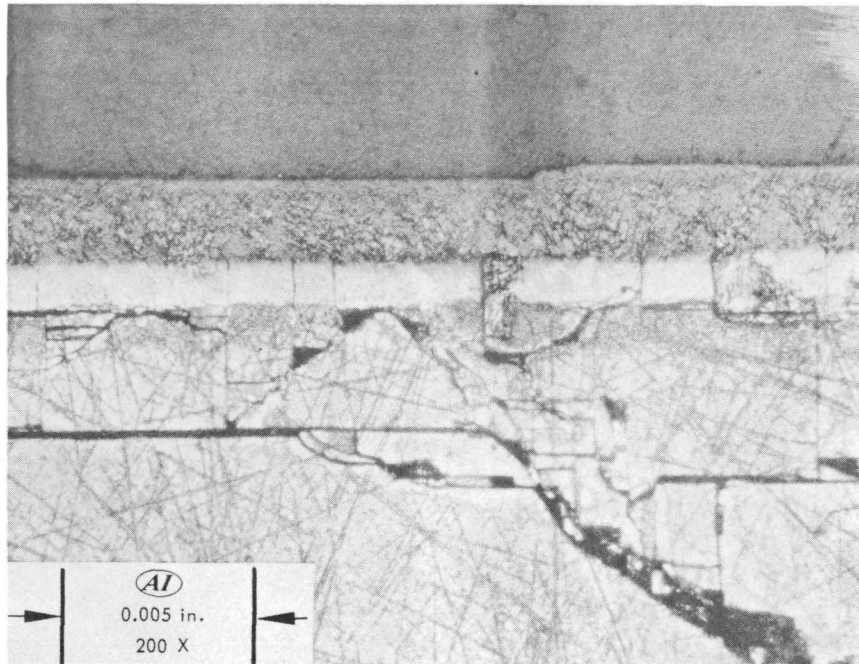
Recent results include more detailed observations of the epitaxial oxidation products which form at low temperatures (~900°C) and further data which permit a description of the probable reaction occurring at high temperatures (~1800°C).

The low-temperature structures are shown in Figure 4 for oxidation of a single crystal of UC_{1.01} in oxygen at 10⁻⁴ torr at 950°C to a total weight gain of 10.6 mg/cm², and in Figure 5 for oxidation of a single crystal of UC_{0.98} in oxygen at 2 x 10⁻⁵ torr at 900°C to a total weight gain of 1.0 mg/cm². The polished cross section in Figure 4 shows two layers on the surface, an inner layer about 30μ thick and an outer layer about 100μ thick. X-ray diffraction patterns of the untouched outer layer indicate UO₂ with no preferred orientation. Another portion of the oxidized crystal was mounted face up in order to grind off the outer layer and leave the inner layer. X-ray diffraction then showed that the inner layer was UC₂ highly oriented on the 100 face of the UC substrate. The relative amounts of UC₂ and UO₂ as determined by measuring the thickness of each layer, and by weight gain, indicate that the predominant reaction was:



but that a part of the UO₂ was formed by a reaction not producing UC₂, probably





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Figure 4. Cross-Section of Oxidized Surface of UC Single Crystal Showing UO_2 and UC_2 Layers

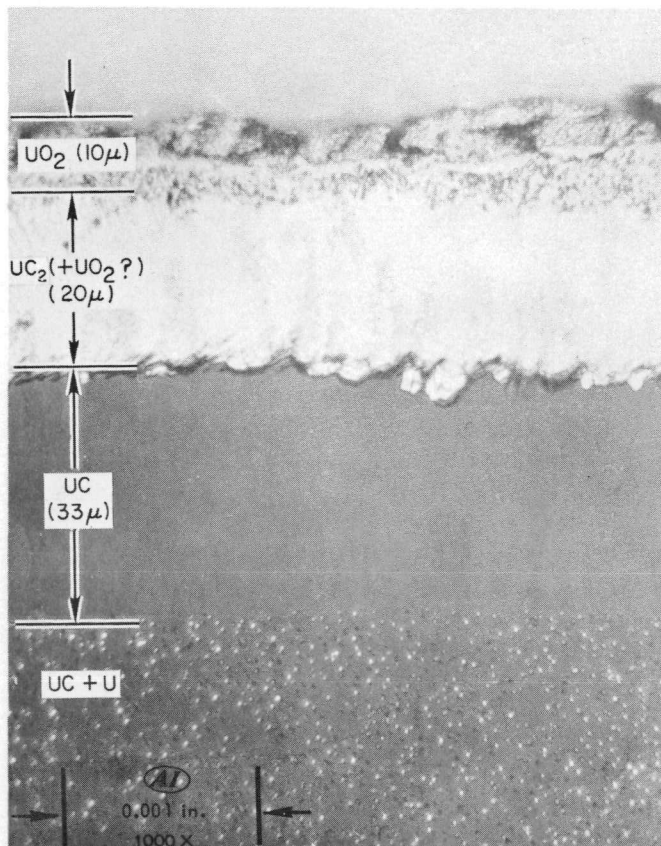


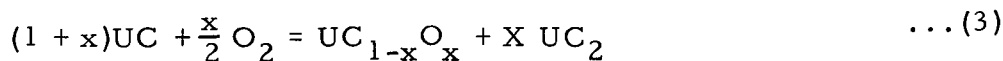
Figure 5. Surface Layers on Oxidized Hypostoichiometric UC Single Crystal

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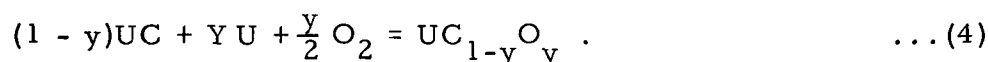
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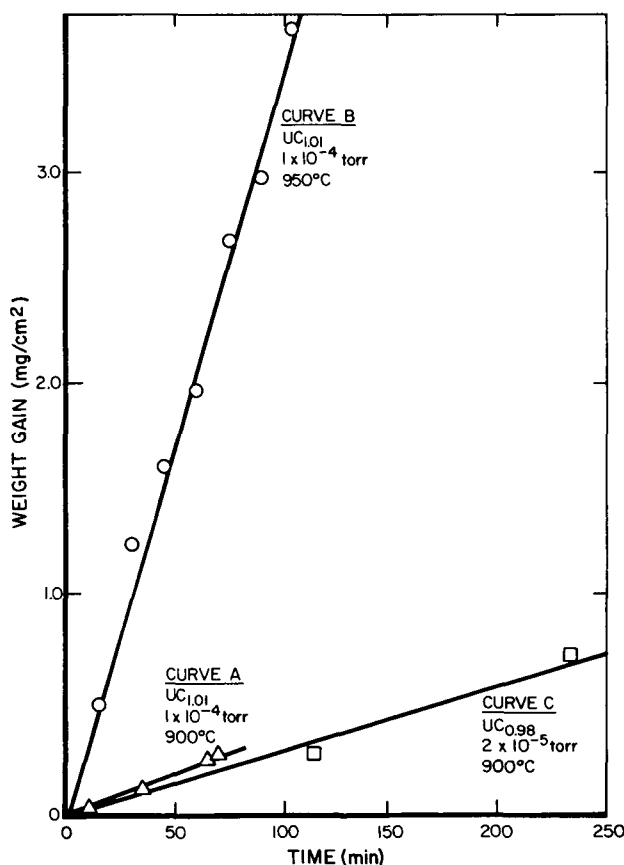
In the case of the hypostoichiometric sample, the cross-sections seem to show much less UO_2 unless there is some intergrown in the UC_2 layer. Another feature that may be observed on Figure 5 is the sharply defined zone in the UC crystal underneath the UC_2 layer which has been cleared of the free uranium particles present in the original crystal. To explain the small amount of UO_2 and the reaction with uranium, it may be assumed that oxygen has dissolved in the UC phase according to the equations:



and



According to the maximum solubility of oxygen in UC reported by various workers, x may be as high as 0.35, based on a volumetric estimation of the free uranium present, y is about 0.02. Hence, reaction (4) would account for most of the cleared zone seen and its thickness (33μ) agrees well with that calculated from the weight gain calculated by using reaction (3).



The kinetic data for the runs at 900 to 950°C are summarized in Figure 6. As reported previously, UC oxidized at 10^{-4} torr oxygen pressure and 1800°C showed no oxidation layer and lost weight instead of gaining. Analysis of the condensate evolved during this experiment now shows that all of the weight loss can be accounted for by uranium and carbon evolution, that is, without identifying the species vaporized. Presumably, the carbon was in the form of CO and the uranium was either free uranium released by the combination of the carbon with oxygen, or gaseous UO. At the same time, the UC lattice parameter was reduced to 4.9557 Å, which corresponds to an oxygen solid solution of $UC_{0.75}O_{0.25}$; however, only 500 ppm of oxygen was found by analyses. Our current thinking is that the contracted lattice parameter is due to a carbon deficiency in the UC resulting from the oxidation.

Figure 6. Low-Pressure Oxidation of UC Single Crystals

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In order to determine and control the atmosphere conditions under which all of the various testing of carbides is done on this program, two types of equipment changes are in process. One is to install ion-pumping systems for high vacuum and the other is to instrument and analyse inert gases for traces of oxygen, nitrogen, and water vapor, and to use purification trains adequate to reduce the concentration of each of these gases to below 1 ppm whenever the exposure of the sample warrants this precaution. The gas analysis equipment is now in operation. Thus far, a gas chromatograph has been calibrated and it has been found that oxygen and nitrogen in the helium delivered by the purification train on the induction zone melter are about 2 and 7 ppm, respectively, compared with about 3 times as much in tank helium. An improved purification train seems desirable.

D. SELF-DIFFUSION STUDIES

The technique for depositing radioactive U^{233} and C^{144} on the surface of UC, followed by high temperature annealing, sectioning by grinding off surface layers, and counting of both tracers in a gas flow proportional counter has been explored; the only problem still remaining is that the precision of counting for the C^{14} is low due to self-shielding from the grindings. Other methods are being checked for accuracy. A preliminary run at $1600^{\circ}C$ for 45 min gave a diffusion coefficient figure of $8 \times 10^{-8} \text{ cm}^2/\text{sec}$ for carbon, about twice that reported by Chubb et al. (BMI-1551).

E. MECHANICAL PROPERTIES

Hot hardness of near-stoichiometric single crystals of UC have been measured in the 100 and 111 directions and the data are shown in Table 5. The differences found indicate that there is appreciable slip even at room temperature, since the differences are consistent with the resolved shear stress on slip planes.

F. GAS MOBILITY IN UC AND W-DOPED UC

Activity in this area has not actually started as yet, but it is planned to carry out three types of experiments: (1) low level irradiation and annealing, (2) helium injection by accelerator and annealing, and (3) high-temperature high burnup irradiations in a reactor. Some progress in this phase of the work is expected during the next quarter.

TABLE 5
HARDNESS OF SINGLE CRYSTAL UC
(kg/mm²)

Temperature (°C)	100 Orientation	111 Orientation
200	650	425
350	610	425
500	415	280
600	220	160
700	110	80
800	75	55
900	75	60
1000	75	58
1100	72	48
1200	51	37

IV. EVALUATION OF EFFORT TO DATE

Zone-melting continues to be a useful procedure, not only for preparing both doped and undoped material for use in studies of self-diffusion, gas mobility, reaction with gases, and phase equilibria, but also as a source of data which are useful in themselves; for example, distribution coefficients for U and C between liquid and solid phases indicate the shape of the phase diagram. Preparation of perfect single crystals would be of value for radiation damage studies, but efforts to prepare such crystals by zone-refining techniques have not been successful.

Our interest in W-doped and Mo-doped UC data for use in determining a means of fission gas control is still strong and we are making efforts to start various testing programs for study of fission gas mobility in these materials; the tests would include high burnup irradiations. Solid solutions which are supersaturated at reactor fuel temperatures now appear promising, as do preparations containing fine particle dispersions, in the light of results of a recent test on metal fuels. We have apparently prepared solid solutions of this kind in the UC-W system, and have made considerable progress towards obtaining phase equilibrium data which are needed for work on supersaturated solutions.

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A better understanding of the UC-U system (and the related PuC-Pu and mixed carbide systems) and in particular the influence of oxygen on these phase relations is needed in carbide fuel research. The work on interaction of carbides with low pressure oxygen is one step towards supplying that need. Another step is our recent procurement of gas analysis equipment to determine impurities of less than 1 ppm in inert gases used for furnace atmospheres, and the further procurement of ion-pumped furnace chambers to permit long-time vacuum heating of carbides without risk of significant contamination by oxygen or nitrogen.

V. NEXT REPORT PERIOD ACTIVITIES

Rough drafts have been written of two papers which are expected to be submitted for publication in journals during the next quarter. One deals with the preparation and properties of UC containing fine dispersions of tungsten, the other with stoichiometry changes in UC during zone-melting and decarburization.

Experimental work is expected to be completed on the low tungsten portion of the U-C-W phase diagram. Further progress is expected in the controlled preparation of W-doped and Mo-doped UC, including a start on the preparation of enriched samples for a reactor irradiation test.

Newly acquired high-purity-atmosphere controls will be used for studies of a portion of the UC-U phase diagram to include high-temperature x-ray studies.

The self-diffusion work is now at the data-taking stage, and sufficient data to provide a preliminary view of results by the chosen technique are expected during the next quarter.

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Program: Reactor Fuels and Materials: High Temperature Material Development	
Project: Refractory Moderator Materials	
Project Engineer: C. E. Weber	
Reporting Period: July-September 1965	AEC Category: 04-40-02-09
General Order: 7551	Subaccount: 4549

I. PROJECT OBJECTIVES

The broad objectives for this program are to investigate defect structures and radiation damage effects of selected materials in a broad category of "refractory moderator materials," and to relate these properties to material behavior. More specifically, they are: (a) to develop, understand, and to utilize means of growing single crystals of graphite, and, as necessary, of other materials consistent with other objectives of the program; (b) to characterize crystal perfection and crystal defect structures of selected materials of interest to this program; and (c) to study radiation damage mechanisms and effects, with the ultimate objective of understanding and possibly controlling radiation damage effects, and of assessing relative radiation damage tolerance for selected materials of interest.

The materials of interest to this program are those refractory, nonfissile materials that can be considered to have potential use as moderator, as diluent, or in structural nuclear application. This may include oxides, carbides, nitrides, beryllium-rich compounds, and other structurally related compounds.

II. MAJOR ACCOMPLISHMENTS IN FISCAL 1966

Procedures for growing graphite single crystals have been reviewed, and those showing the most promise of success have been selected. Suitable furnaces were designed and built, and recently have been delivered to Atomics International.

A recently acquired Lang-type camera for making x-ray diffraction topographs has been put into operation and extensive investigation of defect structure in BeO crystals initiated. This is coupled with etching studies of surface-emergent defects.

Some other materials of interest to this program have been acquired in the form of presumably highly perfect single crystals.

III. PROGRESS DURING REPORT PERIOD

The objectives under the present program represent significant redirection, in that some new tasks have been included, notably the effort to grow graphite crystals of significant size and high crystalline perfection. The remainder of the program is broadened from that of last year primarily by embracing refractory moderator and related materials in addition to BeO. Because of this redirection, many of the tasks are in preliminary status.

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A. GRAPHITE CRYSTAL GROWTH

The adopted procedures for graphite crystal growth are based on experiments described in the literature. They are, in order of priority, precipitation of carbon from saturated molten metal (Fe, for example), and deposition on a substrate from carbonaceous vapor; initial laboratory effort will be directed toward the first of these. A recent attempt to grow graphite crystals was made by the research group at Union Carbide Corporation by the two methods noted above. A visit was paid to this research group to review their experiences in growing graphite crystals. They were somewhat successful in producing small crystals and in gaining some insight into growth processes and crystal quality, that will be helpful in the present program.

Perusal of the literature discloses that two important factors generally have been given inadequate attention. These are (a) crystal nucleation control, and (b) control of growth rates which are commonly far too great to obtain crystals of good quality. The present experiments are designed to at least minimize the adverse effects of these two factors.

Two furnaces have been designed, built, and delivered for the graphite crystal growth task. Utilities hookup, inspection, and operational check-out are presently under way. The furnaces are dissimilar only in the heater elements; one will be heated with superkanthal-type rods and the other with Pt-Rh alloy wire winding. Each furnace has a main heater element and an auxiliary heater element. With these, it is planned to establish a controlled temperature gradient along the furnace core. The heater elements surround a muffle tube of dense Al_2O_3 oriented in a vertical direction. The crucible containing c-saturated molten metal will be supported on an elevated hearth. Provisions are under study for removal without damage of the graphite crystals following their growth.

In the furnaces just described, it is planned to grow crystals by two techniques. One is to very slowly cool a saturated metal solvent to create supersaturation and hence promote crystal growth. The other is to establish a steady-state thermal gradient across which dissolution-precipitation processes can slowly operate over fairly long periods of time.

B. RADIATION DAMAGE IN BeO

In theories of radiation damage in BeO at nominally room temperature ($<100^\circ\text{C}$), the possible role of the β -BeO structure or related lattice disorder continues to be without satisfactory assessment. An unusual experimental technique that may yield information useful to assessing this matter involves the so-called Borrmann anomalous x-ray transmission that is observed with highly perfect crystals. The degree of normal photoelectric x-ray absorption when the crystal is oriented at the Bragg angle depends on the extent and character of disturbances in the crystal structure; hence, it is expected to be strongly modified by radiation-induced structure defects. Selected BeO crystals are being examined for their suitability for this experiment.

The topic of low-temperature radiation damage in BeO was recently discussed with B. S. Hickman, Australian AEC. There is now concurrence on the following items that are central to interpreting the effects.

- 1) The existence of β -BeO has not been directly observed in irradiated material, in spite of attempts in many laboratories to find evidence for it.

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2) The large amount of expansion in the c-axis cannot be attributed to the presence of the radiation-induced basal-plane clusters.

3) Some other type of dilating centers is present, whose character is still unknown; it may be an interstitial-type defect or some type of disorder with a tendency toward the β -BeO structure, or some other type of defect not yet proposed.

Of particular interest to this same topic, it has been reported that β -BeO has been found in the exhaust products of rocket solid fuels containing Be metal. This further indicates that under some circumstances, β -BeO can be retained at room temperature, in agreement with electron microscope observations. Subsequent laboratory experiments (not at AI) indicate that it may be possible to produce quenched β -BeO in research quantities. This may open some promising avenues of research in radiation damage effects and resistance in BeO.

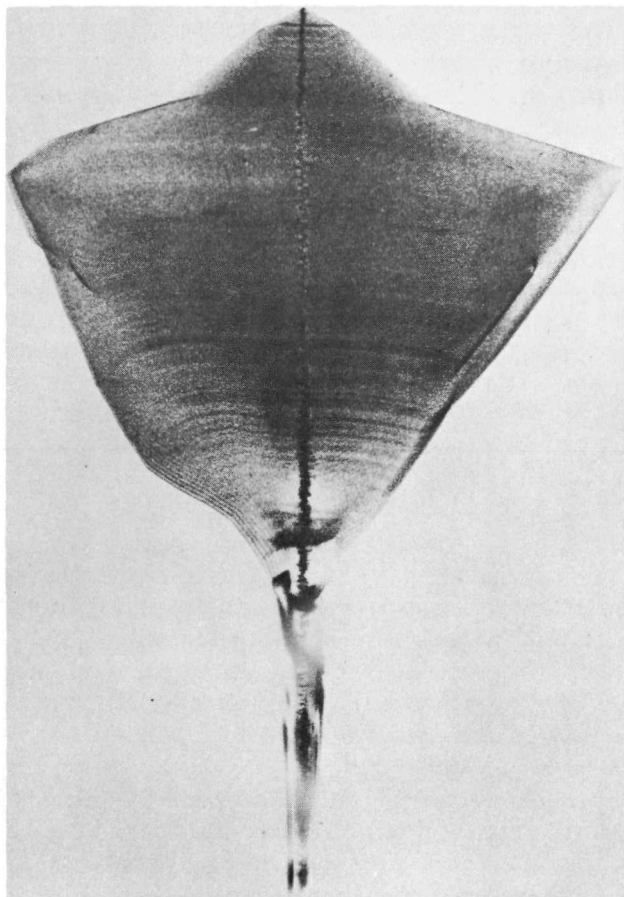
C. DEFECT STRUCTURE STUDIES

The recently acquired Lang-type camera for making x-ray diffraction topographs of single crystals and the associated microfocus x-ray generator have been put into more or less continuous operation. Since the microfocus unit requires considerable maintenance attention and has limited x-ray intensity output, a more satisfactory unit has been ordered. This will permit far more rapid examination of the many crystals that will be used in this program.

Figures 1 and 2 show a topograph of a crystal in which the axial screw dislocation appears to have undergone configuration alterations. (The unusual shape of the crystal is due to severe etching in phosphoric acid.) Near the growth end of the crystal, the dislocation is straight and appears as a narrow line. In older parts of the crystal, away from the growth end, the trace of the dislocation is broader and irregular. This is interpreted in terms of precipitation of vacancies or of interstitial atoms on the screw dislocation, causing it to climb into helical configuration. This effect has been observed only in a few crystals, and may be related to the specific types of impurities incorporated into those crystals.

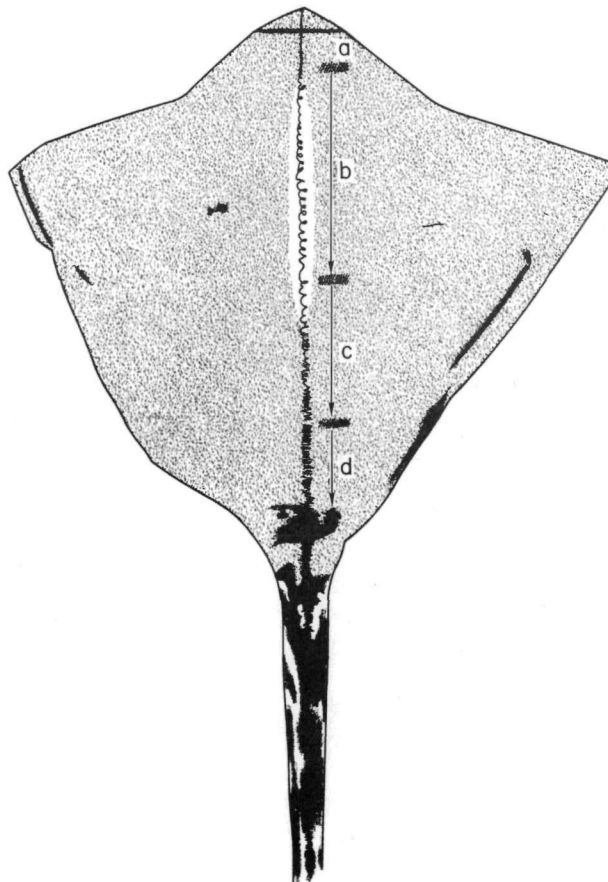
Although topographs have been made of enough crystals to draw some general conclusions, many more will be necessary before acceptable conclusions can be made. It is apparent, however, that the BeO crystals grown from flux are almost universally defective in some manner that can be detected by x-ray topography, and that the distribution of defects, whether line or point type, varies considerably between crystals and even within a given crystal. Even crystals that appear to be perfect by visual examination contain considerable defect concentrations as shown by x-ray means.

Associated with the x-ray techniques, means of studying the defects in BeO by chemical etching are being developed. Molten salts in which BeO are soluble were determined to be useful as etchants. A particular class of etchant examined was lithium molybdate (Li_2MoO_4) with small percentages of MoO_3 , V_2O_5 , or LiBO_2 . It was found that these etchants are specific in their action, in that screw dislocations emergent on (0001) and inversion twin boundaries emergent on basal plane surfaces are preferentially attacked. This as well as other specific manners of etching are under study and are being correlated with defect structures observed by x-ray topography.



7551-51191

Figure 1. Lang Topograph of Heavily Etched AgK Crystal (002) Reflection (Notice the lateral extensions of axial dislocation)



7551-1544

Figure 2. Drawing Emphasizing Character of Dislocation and Depleted Region Around Portion of the Dislocation Length

D. SINGLE CRYSTAL PROCUREMENT

For defect structure and radiation damage studies, a variety of materials in the form of relatively perfect single crystals are required. Those that have been procured are α -SiC (five sources), Al_2O_3 (both vapor and flux-grown), Fe_2O_3 and Cr_2O_3 (flux-grown), and CdS (vapor-grown). AgI (wurtzite structure) in relatively poor crystal form was also obtained.

A newly developed material which is intriguing for potential application as a reactor material is glass-carbon, samples of which have been acquired. It has the same moderator properties as the commonly used graphites but may have considerably more attractive physical properties. It is reputed to have a glassy, therefore isotropic structure, to be stronger than bulk graphite, and to resist graphitization at temperatures as high as 3000°C . If these representations are valid, carbon-glass may become important in reactor technology. In this sense, we intend to assess the properties of this material.

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E. PRESENTATIONS AND PUBLICATIONS

The following papers have been accepted for publication:

1. G. G. Bente, "The Elastic Constants of BeO at Room Temperature," J. Am. Ceram. Soc.
2. S. B. Austerman, J. B. Newkirk, and D. K. Smith, "Study of Defect Structures in BeO Single Crystals by X-ray Diffraction Topography," J. Appl. Phys.
3. S. B. Austerman and J. W. Wagner, "Cation Diffusion in Single Crystal and Polycrystal BeO," J. Am. Ceram. Soc.
4. S. B. Austerman, "Degradation of Growth Perfection in BeO Crystals by Silica," J. Am. Ceram. Soc.
5. S. B. Austerman and K. T. Miller, "Dimensional and X-ray Diffraction Changes in Irradiated Single Crystal BeO," Physica Status Solidi.
6. A. C. Hott and V. C. Davis, "Multiple Thin Section Grinding," J. Am. Ceram. Soc.

The status of each of the following items is as noted:

7. "The Application of Radionuclides to the Determination of Phosphorus in As-grown Beryllium Oxide Crystals," Int. J. Appl. Rad. and Isotopes 16, 499-500 (1965); published
8. "Thermal Conductivity of BeO Over a Range of Density," (approximate title) C. J. Ambrose and R. S. Carpenter. To be presented at the Fifth Annual Thermal Conductivity Conference, Denver, October 13, 1965.

IV. EVALUATION OF EFFORT TO DATE

The various tasks described in this report are generally well underway. The techniques for growing graphite single crystals have been thoroughly reviewed, promising techniques selected, and suitable furnaces have been acquired. In the study of BeO crystals the growth of crystals has been discontinued except for a small effort, and has concentrated more heavily on investigation of the defect structures related to crystal growth, impurities, mechanical treatment, radiation damage, etc. Some additional materials, such as single crystals, that are appropriate to this program, have been acquired for incorporation into the defect-structure and radiation-damage studies.

V. NEXT REPORT PERIOD ACTIVITIES

During the next quarter, the graphite crystal-growth furnaces will be put into operation and temperature distribution characteristics as a function of power settings will be evaluated. Hopefully, a few preliminary runs to produce graphite crystals will be made. Examination of defect structures, radiation damage, and related properties in single crystal materials appropriate to this program will continue.

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Program: Reactor Fuels and Materials Development— Other

Project: Fission Effects in Metal Fuels

Project Engineer: C. E. Weber

Reporting Period: July-September 1965

AEC Category: 04-40-02-09

General Order: 7652

Subaccount: 4537

I. PROJECT OBJECTIVES

The objectives of this project are: (a) to obtain a detailed understanding of the processes and mechanisms involved in fission damage, including those responsible for swelling or reactor fuel materials during irradiation and those arising from dynamic fission processes; and (b) as a result of such understanding, to propose and test procedures by which swelling can be minimized.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

The NAA-118 experiment, which explored further the concept of fuel swelling control by additives and heat treatment, has demonstrated that solution-quenched alloys are superior to thermally produced fine dispersion alloys. This observation has led to new concepts of the influences of fission spike and fission product production on the stabilization of fuel swelling. In essence, the kinetics of precipitation or the factors that influence nucleation are altered by the fission events to the point where more numerous and more finely dispersed particles can be formed during fission than can be formed during thermal treatments. Accordingly, maximum volume stability can be obtained by the economical, solution-quenching procedure.

Rare gas-atom mobility studies of fission product-bombarded copper foils have indicated either that there is no unique activation energy for the release of rare gas atoms from this material, or that there are sufficient traps of various strengths so that the extent of gas release is temperature-dependent. This very pure material does not conform to ideal diffusion theories for rare gas atom release from solids.

The in-cyclotron creep device for measuring the properties of fuel materials during fission has been proof-tested. Its response is well within the specified sensitivity of load and extension measurements. Published data on the creep of alpha uranium as a function of strain rate and temperature have been reevaluated, and very consistent correlations have been obtained which subdivide the creep process into at least four different stages or substages.

III. PROGRESS DURING REPORT PERIOD

A. NAA-118 CAPSULE

The swelling resistance of specifically designed fine-dispersion metal fuels that contain trace amounts of additives is being investigated at high temperatures, 550 and 645°C (990 and 1152°F) and at high burnups, 0.8 and 1.9 atom % burnup.

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The following conclusions, which may be subject to minor modifications, have been reached.

1) The unrestrained weakly alloyed uranium fuel materials have swelled to the extent that they are unsuitable reactor alloys in their prescribed metallurgical state. (U - 0.1 Si, U - 0.1 Al, U - 0.1 Si and 0.01 V, U - 0.1 Al-0.01 V, U - 0.01 Ce, U - 0.01 Cr, U - 0.01 V, irrespective of heat treatment.) The stability of all of these alloys was disappointing.

2) U - 4 Mo and U - 10 Mo alloys without additive were grossly unstable with respect to volume changes during test.

3) The silicon containing U - 10 Mo alloys showed definite improvement in volumetric stability. These alloys can be rated in terms of relative stability in the following manner.

a) Solution-quenched alloys are superior to both coarse and fine dispersion alloys which are in turn quite superior to unalloyed base materials.

b) No firm decision can be made concerning coarse (0.1 Si) and fine (0.2 Si) dispersion alloys, due to existing differences in silicon content. The volume changes are generally larger for U - 4 MoSi fine dispersions but this may not be a consequence of initial microstructure. Recent theorems (Brinkman), which are substantiated by the NAA-118 test results, suggest that the absolute level of alloying content may be important. Savannah River test data also tend to support this viewpoint.

4) Volume changes for the more stable molybdenum alloys are reported in the table. Measured density changes have been converted to volume changes by the relationship

$$\frac{\Delta V}{V_0} = \frac{\Delta \rho / \rho_0}{1 + \Delta \rho / \rho_0} \cdot$$

This conversion is desirable because $\Delta V/V_0$ is linearly related to burnup, while $\Delta \rho / \rho_0$ is not. The comparison of density changes and area change as measured on macrophotographs is presented in Figure 1. These data indicate that the volume changes are nearly isotropic.

5) The observed volume changes are rather high in the case of the U - 4 Mo and U - 10 Mo alloys when compared to other published results. Burnup rate may be a factor in this higher volume increase, following Barnes et al. (Geneva 1964). No real operational control factors appear to be responsible for this larger volume change.

6) The replication data for samples of fine dispersion U - 10 Mo - 0.1 Sn that were incorporated into Capsule 4 suggest consistent behavior in these materials. (Dry weights pre- and postirradiation agree within 0.001 gm. All reported samples were monitored before irradiation by spectrometry to ensure that sample markings and composition were consistent.)

7) Only the U - 10 Mo - 0.1 Sn fine dispersion and solution-treated samples in Capsule 3 provide inconsistent material response data. The solution-treated material has transformation products at grain boundaries following irradiation, although the fission rate was sufficiently high to promote a single-phase alloy during test at this temperature (as shown by the behavior of the companion sample in Capsule 2, which operated at the same temperature). Further analysis requires electron microscopy studies, which are in progress.

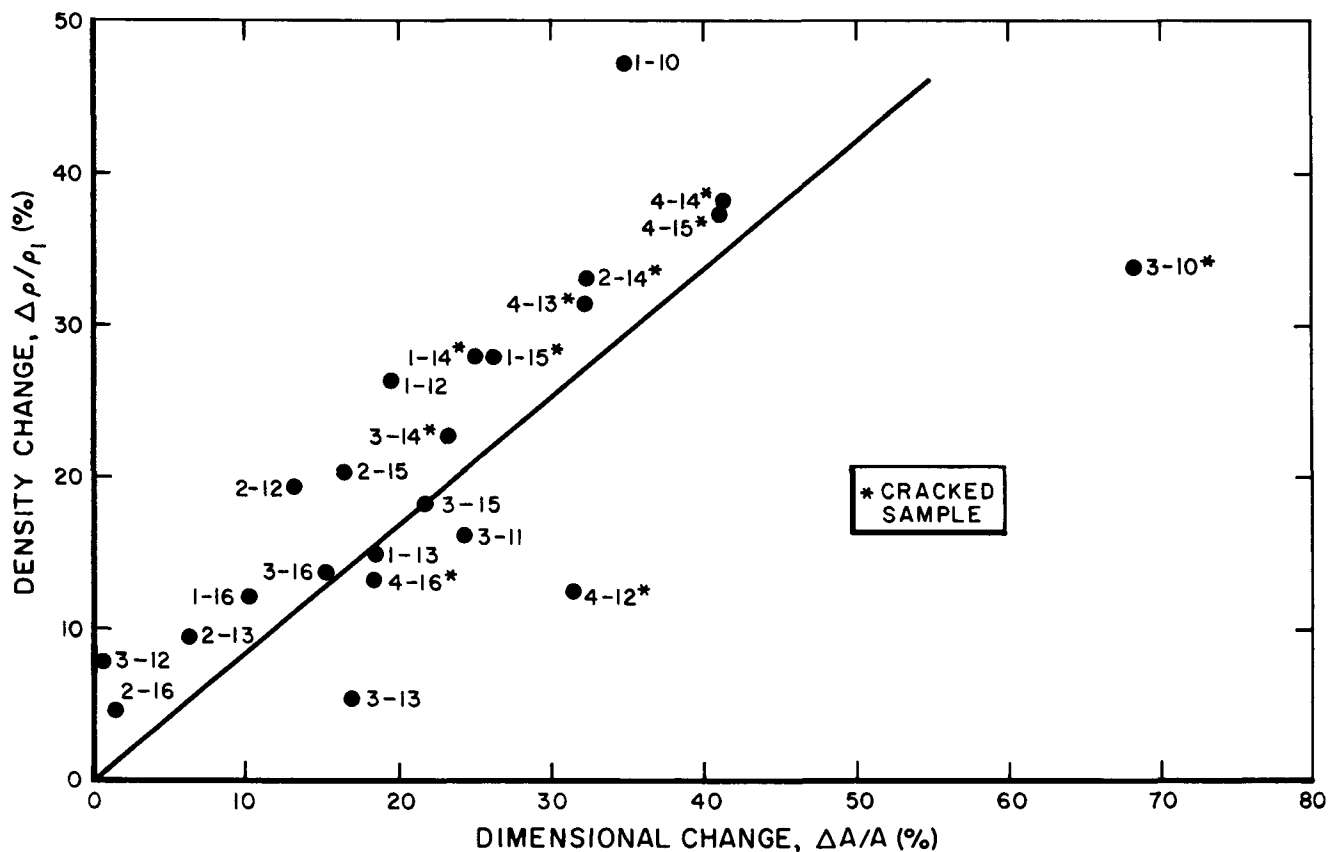
8) These results can be interpreted in terms of an irradiation-induced (more specifically a displacement spike damage-induced) re-resolution of particles. Presentations by Brinkman and Johnston of the NAA Science Center,

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VOLUME CHANGES IN NAA-118 SAMPLES

Capsule 0.8 atom % Burnup 645°C		Capsule 2 1.4 atom % Burnup 550°C		Capsule 3 0.8 atom % Burnup 550°C		Capsule 4 1.4 atom % Burnup 645°C	
Alloy	$\frac{V}{V_0}$	Alloy	$\frac{V}{V_0}$	Alloy	$\frac{V}{V_0}$	Alloy	$\frac{V}{V_0}$
U - 4 Mo Control	91.1	U - 4 Mo Control	-	U - 4 Mo Control	51.5	U - 4 Mo Control	-
U - 4 Mo - 0.2 Si f. d.	68.9	U - 4 Mo - 0.1 Si	-	U - 4 Mo - 0.2 Si f. d.	19.3	U - 4 Mo - 0.1 Si s. t.	-
U - 4 Mo - 0.1 Si c. d.	36	U - 4 Mo - 0.1 Si c. d.	27.5	U - 4 Mo - 0.1 Si	8.7	U - 4 Mo - 0.1 Si s. t.	34.8
U - 4 Mo - 0.1 Si s. t.	17.6	U - 4 Mo - 0.1 Si s. t.	10.7	U - 4 Mo - 0.1 Si s. t.	5.8	U - 10 Mo Control	46.3
U - 10 Mo Control	38.9	U - 10 Mo Control	50	U - 10 Mo Control	29.4	U - 10 Mo - 0.1 Sn f. d.	62
U - 10 Mo - 0.1 Sn f. d.	39	U - 10 Mo - 0.1 Sn f. d.	25.1	U - 10 Mo - 0.1 Sn f. d.	27.9	U - 10 Mo - 0.1 Sn f. d.	59.7
U - 10 Mo - 0.1 Sn	13.9	U - 10 Mo - 0.1 Sn	4.9	U - 10 Mo - 0.1 Sn	17.9	U - 10 Mo - 0.1 Sn	15.2

f. d. = fine dispersion
c. d. = coarse dispersion
s. t. = solution-treated



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Figure 1. Comparison of Changes in Density and Dimensions

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McDonnell and Angerman of Savannah River, and Bierlein and Leggett of HAPO at the coming Asheville meeting will reinforce the technical and engineering significance of these results.

9) A most significant contribution arises from a reinterpretation of the NAA-60 test results. These similar alloys were irradiated at low temperature (300 to 400°C) and annealed to 850°C following irradiation, with remarkable volume stability in doped alloys. Current concepts (complements of Brinkman) suggest that the stability arises from the solution-treatment of these doped alloys during irradiation as a consequence of fission-induced mixing. Kramer has deduced from quenching tests, prior to the NAA-60 experiment, that vacancy aggregates were nucleation sites for particle precipitation. Barnes (currently at NAA Science Center) has speculated that fission gas clusters may play a similar role in fuel materials. The observed stability may then be a difference in the response of irradiated materials, rather than a preirradiation treatment control. A supersaturation caused by irradiation may have been responsible for bubble stabilization during postirradiation heating. Heating could cause fine-scale precipitation of the excess silicon on the bubbles.

10) The arguments for bubble stabilization on the basis of (a) xenon migration to particles or (b) particle precipitation on xenon bubbles is immaterial at this point. The significant point is that dynamic fission processes appear to influence fuel stability and that substitute, short-range tests have to be evaluated very carefully.

11) Details of the table show the following.

a) Solution-treated alloys (or solution-treated and quenched alloys) are always superior to any of the thermally produced dispersions.

b) Unalloyed materials are unacceptable under these conditions for reactor application.

c) The precipitate stabilization concept cannot be confirmed until electron microscopy of irradiated samples demonstrates particle size distributions and bubble-particle association.

d) A point to be considered concerns the time scale required to supersaturate a fine-dispersion alloy to the extent that it is equivalent to a solution-treated (solution-quenched) alloy. Alternatively, one might be interested in the rate of thermal unmixing (the growth rate of existing precipitates which are of stable size) relative to the rate of fission-product mixing. These phenomena have not been measured or even considered in an engineering sense.

B. FISSION GAS RELEASE MEASUREMENTS

"Isochronal" annealing experiments in which samples were heated 200°C in 8 to 16 hr time-periods at a uniform heating rate have shown that there is no unique activation energy for the release of rare gas atoms from recoil-bombarded OFHC copper foils. The amount of gas release is temperature-dependent, with the bulk of the gas release occurring in the temperature range of 290 to 350°C.

The temperature programming has been such that following the temperature rise, the maximum temperature is maintained for several hours. The minimum setpoint was then increased and the heating cycle was repeated. A saturation in gas release was obtained at the highest annealing temperature, but additional gas was released during subsequent heating to higher temperatures. All of the contained gas was not released during the annealing treatment.

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This response suggests that there is a spectrum of activation energies for the release of the rare gas atoms from internal traps. Under such conditions the standard diffusion theories for the release of these gas atoms for solids are inapplicable.

C. COMPUTER STUDIES

Analogue studies have been deferred, due to NAA-118 commitments. The digital studies, which try to identify the most plausible mode of xenon diffusion in a host lattice, have continued. The computer code evaluates the energy of an oversize atom in a host lattice.

Exponential inverse sixth power potentials for both copper and xenon have been explored. This potential has three independent parameters:

- a) r_0 , the value of the radius at the energy minimum;
- b) ϵ , the depth of the potential well at the energy minimum;
- c) α , the steepness of the exponential repulsion.

In combining separate potentials for copper and xenon to form a composite copper-xenon potential, the parameters have been evaluated by the following combination rules.

- a) The r_0 parameters have been calculated in terms of an arithmetic mean, i. e.,

$$r_0(\text{Cu} - \text{Xe}) = 0.5 [r_0(\text{Cu}) + r_0(\text{Xe})] \quad ;$$

- b) ϵ has been evaluated in terms of a geometric mean, i. e.,

$$\epsilon(\text{Cu} - \text{Xe}) = [\epsilon(\text{Cu}) \epsilon(\text{Xe})]^{1/2} \quad ; \text{ and}$$

- c) The α parameter has been defined in terms of both types of mean values.

Successive interactions which allow the relaxation of the surrounding copper lattice atoms about an oversized xenon atom have resulted in a reduction in total energy; but an equilibrium configuration, with zero forces between atoms has yet to be reached.

A second type of potential is also being explored and a computer program has been written, using similar relaxation techniques. Rimmer and Cottrell [Phil. Mag. 2, 1345 (1957)] proposed an arithmetic mean for the composite Cu-xe potentials. In distinction to the approach outlined above, this approach permits the use of dissimilar individual potentials for the gas and metal. The results are not necessarily more reliable, but the computation provides a reference point for existing theory, present codes, and a measure of the dependence of calculated results to different potential functions.

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D. IN-PILE STRESS RELAXATION

The cyclotron creep device has been proof-tested at room temperature, using aluminum test samples. Excellent creep curves were obtained in all tests. The dual sensitivity extensometer system permitted the required 10^{-5} creep rate measurements over a total strain range of 3×10^{-4} . Irradiation testing is tentatively scheduled for November, with contractual arrangements being the only schedule-limiting problem.

The device has two modes of loading four samples during a single irradiation. Two samples have variable load imposed by pressurized bellows attachments. The other two samples are loaded by "dead weights" to provide a uniform load level over theoretically frictionless bearings.

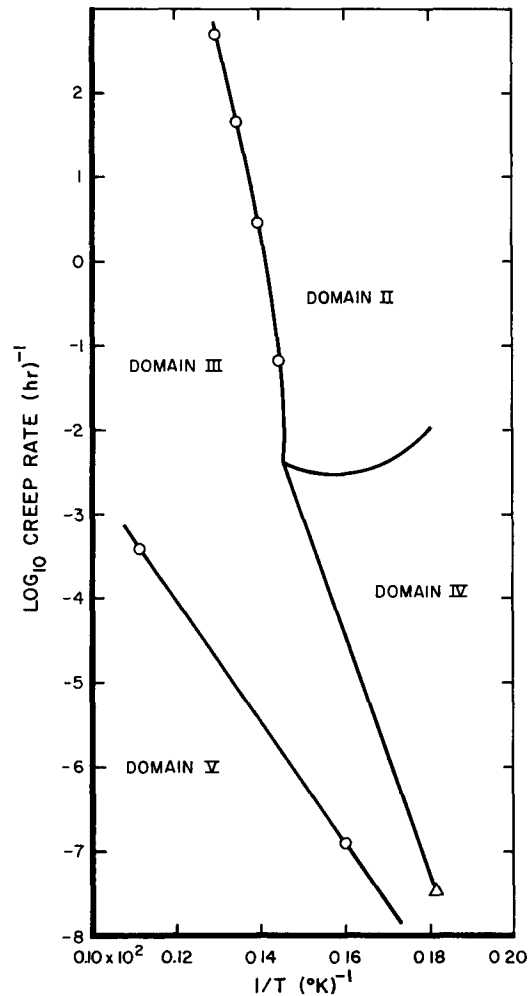
Computer analysis of creep data on alpha uranium obtained from the literature, which was reported in preliminary form in last month's report, has been completed. As previously reported, the existence of at least two different creep-rate controlling processes has been definitely established. Final expressions for creep rates in four of these domains, as evaluated from a best statistical fit using codes defined by Rocketdyne, are as follows.

	<u>MCC</u>
Domain II $\epsilon = \exp \left(11.5 + 3.42 \times 10^{-4} \sigma - \frac{31,400}{RT} \right)$	0.901
Domain III $\epsilon = \exp (-10.01) \sigma^{6.17} \exp \left(\frac{-76,600}{RT} \right)$	0.988
Domain IV $\epsilon = \exp (-213.9) \sigma^{30.4} \exp \left(\frac{-121,000}{RT} \right)$	0.993
Domain V $\epsilon = \exp (2.67) \sigma^{2.33} \exp \left(\frac{-49,000}{RT} \right)$	0.976 ,

where MCC is the multiple correlation coefficient, an estimate of the "goodness" of the proposed correlation.

Domain I is not listed because creep rates in this domain are very sensitive to history; and, because of this, data from various investigators cannot be correlated as a single group.

The boundaries of operation of each of the domains, II through V, are shown in Figure 2. The defined boundaries have several technologically important features. First, the temperature limits of cavitation swelling correspond well with the limits of temperature and creep rates for Domain III indicating that a quantitative relationship between cavitation swelling and Domain III creep should exist. Additional theoretical analyses and laboratory work must be conducted to fully explain this possibility. Secondly, all predictions of "Cottrell" irradiation creep have been based on Domain V behavior. Actually, Domain III creep is the dominant mechanism for almost all reactor applications. "Cottrell" creep should be re-examined as the basis of Domain III behavior. Finally, Figure 2 provides an explanation for the mechanical behavior of alpha uranium in terms of such parameters as ultimate tensile strength and microstructure.



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Figure 2. Boundaries of Controlling Creep Mechanisms in Alpha Uranium

IV. EVALUATION OF EFFORT TO DATE

The NAA-118 experiment has demonstrated that the initial microstructure in thermally treated, precipitated alloys is not a major factor in the control of fuel swelling in materials that are irradiated at elevated temperatures. Solution-treated alloys are more stable than fine dispersions under such conditions. The concept of re-solution and in-pile precipitation of particles during irradiation has been proposed. The dynamic aspects of irradiation damage again require closer scrutiny.

The isochronal annealing tests on fission product damaged copper have indicated that the release mechanism is more complicated than that described by

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simple diffusion theory. There appears to be a distribution of traps throughout the material such that the fractional gas release is temperature-dependent. Therefore, the evaluation of a unique activation energy by isothermal annealing processes is misleading.

The computer calculation codes have been written, and calculations of migration energies for different diffusional paths are in progress.

The cyclotron device is operative and has been proof-tested. Supplementary evaluation of out-of-pile creep data has provided excellent base line correlations for the interpretation of in-cyclotron tests.

V. NEXT REPORT PERIOD ACTIVITIES

The possibility of further experiments on solution-treated alloys at high temperatures and high burnups will be explored. Emphasis on tracer level annealing studies will concentrate on burnup dependence and the influence of microstructure on gas release. The in-cyclotron stress relaxation tests will be initiated.

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Program: Reactor Fuels and Materials Development — Other

Project: Synthesis, Structure, and Behavior of Metal-Hydrogen Systems

Project Engineer: C. E. Weber

Reporting Period: July-September 1965

AEC Category: 04-40-02-09

General Order: 7652

Subaccount: 4552

I. PROJECT OBJECTIVES

The objectives of this project are: (a) preparative research on new binary and ternary hydrides; (b) selected correlated studies of physical, thermodynamic, and mechanical properties; (c) the study of the structure, solid-state physics, and nature of bonding in hydrides as relates, primarily, to thermal stability and movement to its defect structure; and (d) application of advanced experimental and theoretical techniques to study fission gas mobility and radiation damage in various hydride structures to optimize the high-temperature swelling resistance of this class of materials.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

The self-diffusion of Zr in ZrH_x made from both reactor grade sponge and iodide zirconium has been measured over a temperature range of about 750-1000°C.

III. PROGRESS DURING REPORT PERIOD

A. SOLID-STATE EFFECTS

The self-diffusion of Zr in zirconium hydride is being studied by a tracer method. Zr^{95} is deposited as the oxalate on the surface of a specimen and allowed to diffuse through the lattice. After a suitable time at temperature, the γ activity is measured as a function of depth from the surface.

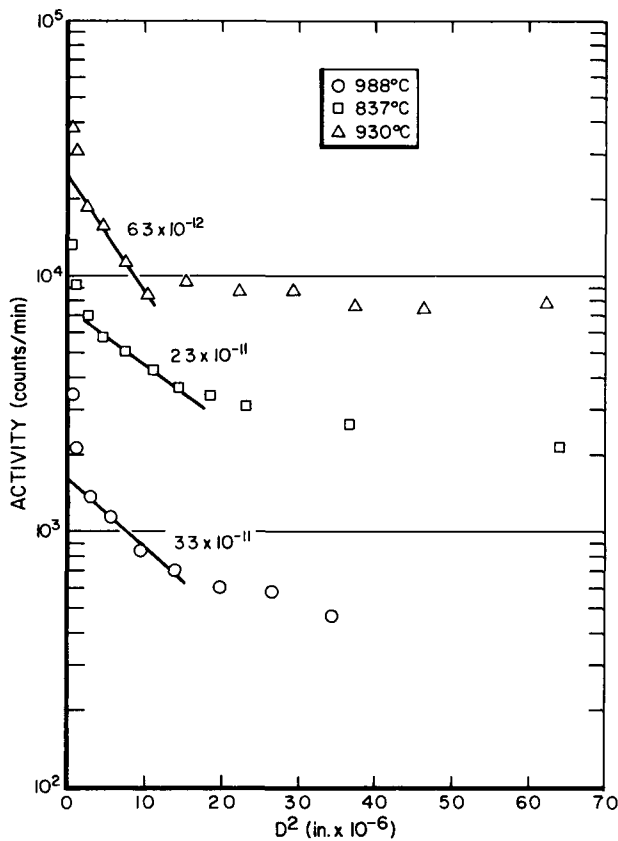
In the Annual Report for GFY 1965, the data for self-diffusion of Zr in $ZrH_{1.71}$ made from reactor grade sponge Zr were reported. At diffusion depths of 0.001- to 0.003-in. into the specimen, a bulk volume diffusion coefficient was determined. At greater depths, grain boundary diffusion predominates and a different diffusion coefficient for this process was found.

During this quarter, the self-diffusion of Zr in zirconium hydride made from crystal bar zirconium was determined. The composition of the hydride was $ZrH_{1.63-1.69}$. Two distinct diffusion processes were again noted, with volume diffusion predominating near the surface of the specimens and grain boundary diffusion predominating at greater penetration depths where the concentration gradient is low. The volume diffusion coefficients measured are reported in Table 1. The experimental data from which these values were calculated are shown in Figures 1 and 2. An approximate value for the activation

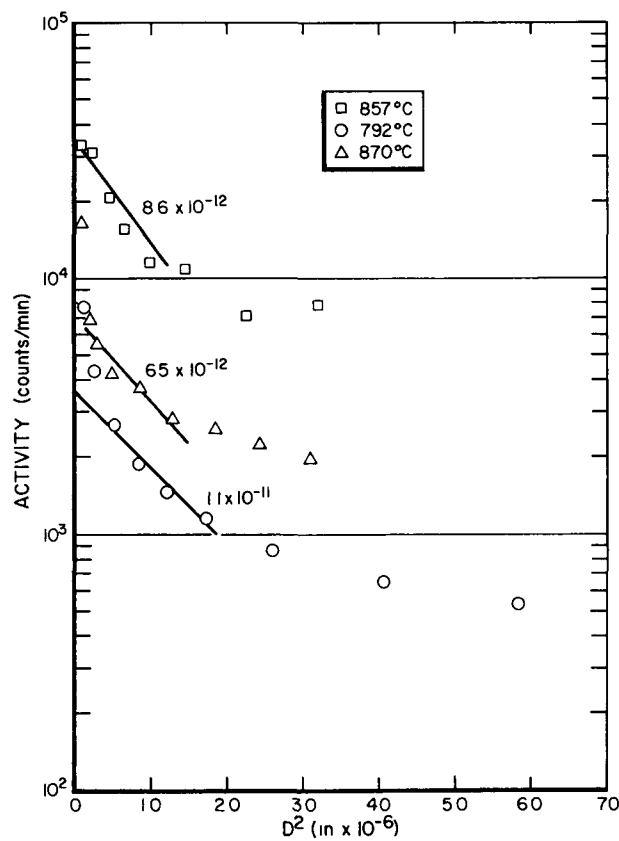
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**VOLUME DIFFUSION OF Zr in H_{1.63-1.69}
(CRYSTAL BAR)**

Temperature (°C)	Diffusion Constant (cm ² /sec)
770	3.1×10^{-12}
792	6.5×10^{-12}
837	6.3×10^{-12}
857	1.1×10^{-11}
870	8.6×10^{-12}
900	1.2×10^{-11}
930	2.3×10^{-11}
988	3.3×10^{-11}



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Figures 1 and 2. Activity Penetration Curves for Crystal Bar Zr in ZrH

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energy for volume diffusion was calculated, and is about 28,000 calories per mole (compared to about 35,000 calories per mole for the arc-melted reactor-grade sponge material). The frequency factor for the crystal bar material is about an order of magnitude lower than that for the sponge material.

During the coming month, a complete mathematical analysis of the data will be made. Least squares calculations of the activation energies will be carried out and standard deviations computed.

B. TERNARY HYDRIDES

A very accurate powder x-ray diffraction analysis was made of the ternary compound $ZrC \cdot ZrH_2$. The various lines observed, and their relative intensities were determined. The hexagonal lattice constants determined from these data are:

$$a_o = 3.3467 \pm 0.0001\text{\AA}, C_o = 5.4898 \pm 0.0001\text{\AA}, c/a = 1.640.$$

A literature survey on the interactions of mercury with metal hydrides was conducted; some of the observations made in these systems suggests the formation of ternary compounds of metal + mercury + hydrogen on the surface of rare earth hydrides when exposed to mercury.

IV. EVALUATION OF EFFORT TO DATE

The efforts on fission gas mobility in the zirconium hydride lattice have reached a logical concluding point; therefore, this work has been terminated.

The largest single effort during this report period was on self-diffusion, and significant progress was made on that task. After a few more experiments, and a thorough analysis of the data, a topical report can be written.

The ternary hydride task is being reoriented from structural determination to preparative research on large monolithic bodies. The task so far has been concerned only with planning, and setting up of equipment.

V. NEXT REPORT PERIOD ACTIVITIES

A complete mathematical analysis will be made of the self-diffusion data.

Experiments to prepare $ZrC \cdot ZrH_2$ bodies by a powder metallurgy process will be conducted.

SECTION

- V -

NUCLEAR SAFETY
&
PHYSICAL RESEARCH

ATOMICS INTERNATIONAL
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Program: Nuclear Safety - Reactor Kinetics	
Project: Measurement of Doppler Coefficients	
Project Engineer: A. A. Jarrett	
Reporting Period: July-September 1965	AEC Category: 04-60-01-09
General Order: 7549	Subaccount: 1351

I. PROJECT OBJECTIVES

The fundamental objective of the project is to develop rapid and inexpensive calculational methods for the Doppler coefficient of reactivity of proven validity that will be attractive to reactor designers. The secondary objective is to demonstrate the validity of the calculational method by comparing calculations with experiments having well-understood accuracy and limitations so that the experimental measurements can be extended with confidence to materials of importance to reactors, but which have resonance parameters which are not well understood. Specific objectives are: (1) to establish gold (with a simple resonance structure at low energies and well-established resonance parameters) and U^{238} metal (with well-established parameters and an extended resonance structure) as resonance integral and Doppler effect standards to which other materials can be compared both in experiment and in theory; (2) to find and evaluate the magnitudes of systematic errors present in measurement techniques; (3) to test the range of applicability of present theory by making measurements over an extended range of surface/mass ratios and temperatures under neutron spectral conditions that can be evaluated most accurately by calculation (i. e., in classical 1/E spectra); (4) to test both theory and experiment by auxiliary measurements designed to emphasize differential effects and test specific approximations and predictions of theory; (5) to measure the Doppler coefficients of uranium metal and oxides and compare to previous work; (6) to extend the resulting experimental and theoretical techniques to other fertile materials such as thorium metal and oxide, uranium alloys and uranium carbide; and (7) to extend the studies to structural and control materials and to those materials of interest to reactor design where insufficient basic resonance data are now available.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

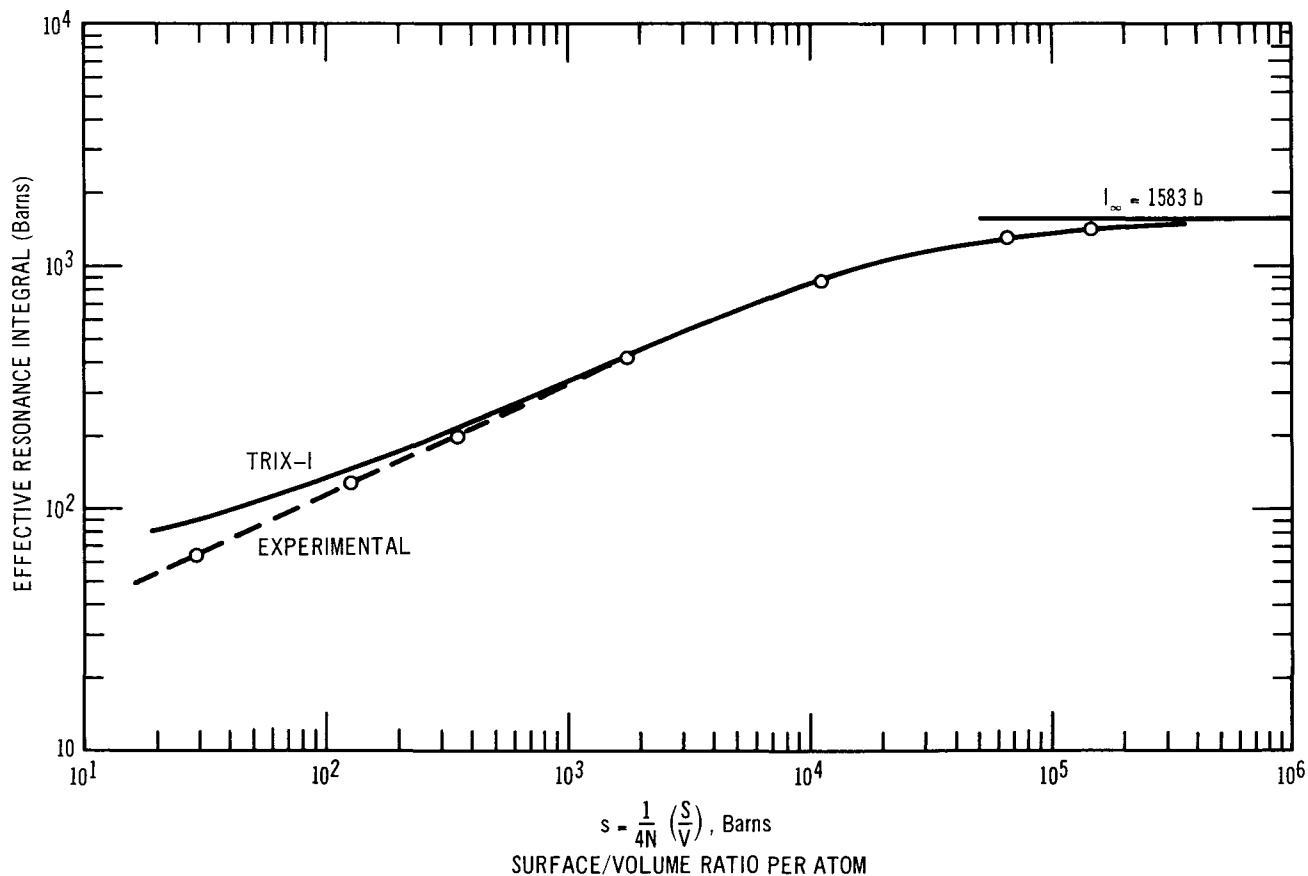
Further analysis and comparison of the experimental results in measurements of the effective resonance integral and Doppler effect in gold with the predictions of TRIX-I have revealed a potentially serious discrepancy between the two. Measurements and calculations to define further the area of discrepancy have been started. Equipment and methods which will greatly improve the accuracy and speed of measurements have been built and checked out. Improved data reduction and analysis codes have been written and are in final stages of checkout. The TRIX-I code has been converted to FORTRAN-IV and documentation has been completed.

III. PROGRESS DURING REPORT PERIOD

A. ANALYSIS OF RESULTS

NAA-SR-Memo-11511, describing present results in measurement of the effective resonance integral and Doppler effect in gold and comparing them briefly with predictions of TRIX-I, was published during this report period. In general, theory and experiment were in reasonable agreement. Further analysis has revealed, however, that there appears to be a disagreement of potentially serious magnitude in some details.

In particular, the theory predicts a much higher effective resonance integral at low surface/mass ratios (S/M) than was in fact observed. The theory also predicts a large peak in the Doppler effect at high S/M. The measured peak appears to lie at an S/M higher by a factor of 5 to 10 than was predicted (See Figures 1 and 2). Analysis shows that the differences cannot be explained by uncertainties in resonance parameters or by experimental uncertainties as they are presently understood. The analysis also shows that the discrepancy is clearly associated only with the giant resonance at 4.9 ev. (This assignment was made possible because of the fact that measurements were made over a wide range in S/M. Information from both extremes in S/M were used in the determination. Resonances of varying width and energy contribute strongly to the

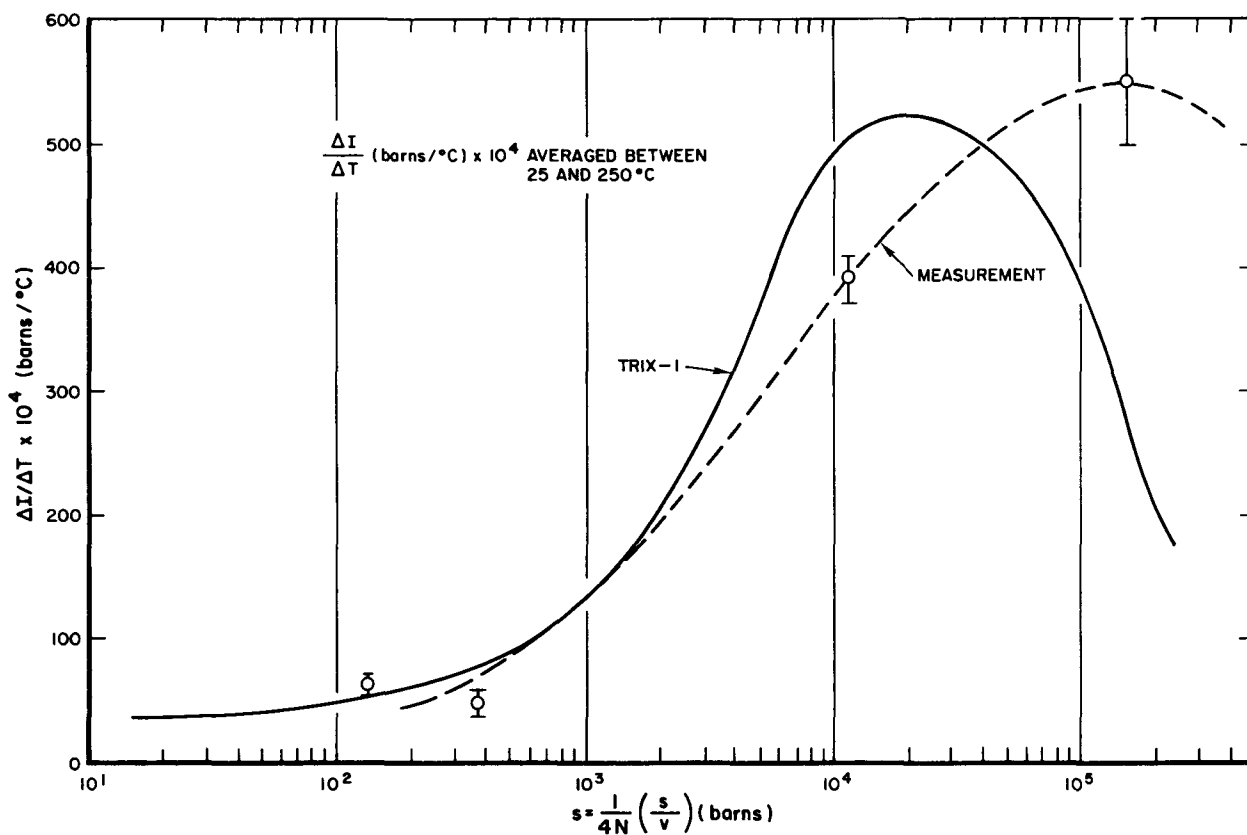


7549-2505

Figure 1. Effective Resonance Integral for Au¹⁹⁷ at 25°C

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7549-2506

Figure 2. Temperature Derivative of Au¹⁹⁷ Resonance Integral

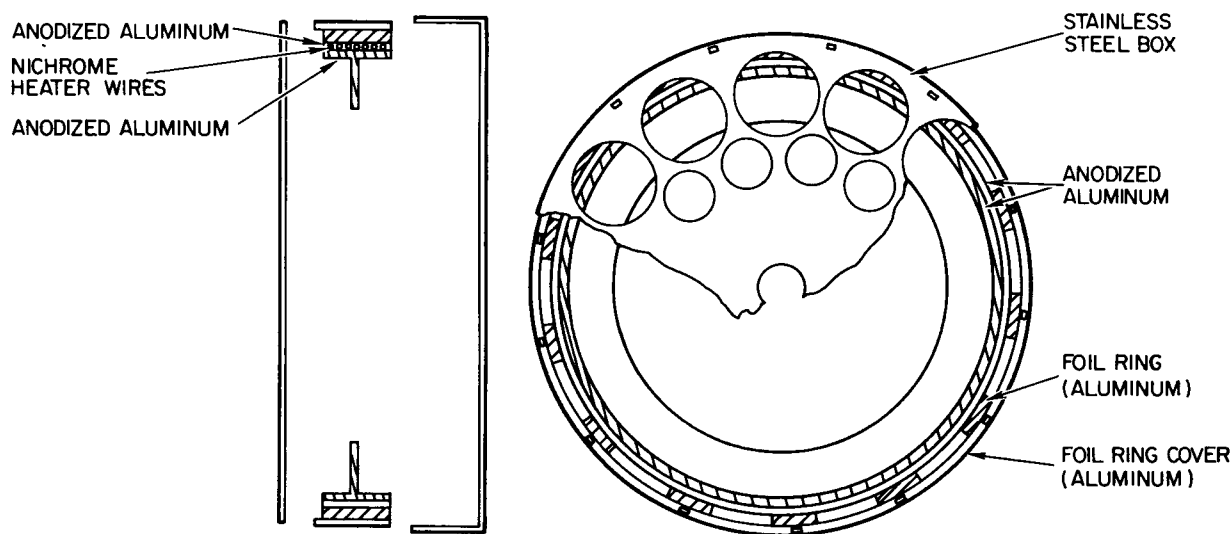
Doppler effect or resonance integral only in a portion of the complete curve. Many resonances contribute strongly to the resonance integral at different S/M from their strongest contribution to the Doppler effect, thus making it possible to isolate their separate contributions.) The exact amount of disagreement depends somewhat on the experimental normalization, and on better definition of where the Doppler peak does lie. Experiments which will provide the required information are now being conducted.

Three possible causes of the disagreement have been proposed. Part of the disagreement is due to improper truncation of the calculation at cadmium cutoff for any temperature other than zero degrees Kelvin. The fact that the giant resonance is so broad may place the temperature broadening in an inaccurate region of the Doppler tables used in the calculation. This effect appears to be unlikely to explain more than a small part of the discrepancy, however. Finally, the validity of the narrow resonance approximation for moderator collisions is in question when applied to broad, low-lying resonances like those in gold, thorium, uranium, and the higher plutonium isotopes.

The difficulty in presence of a discontinuity in the spectrum (such as would occur near control elements) and the questionable validity of the narrow-resonance approximation for broad, low-lying resonances are potentially serious problems to any thermal reactor and to the present generation fast reactor designs.

B. EXPERIMENT

The present measurements have been designed to provide much better definition of the structure in the Doppler effect as a function of S/M in gold and to provide an experimental normalization to the relatively well known 2000 m/sec cross section and thus be completely independent of calculation. To aid in these measurements, a new Doppler capsule has been constructed. It is in the form of a rotating cadmium-covered calibrating wheel that can be heated to high temperature under vacuum. The wheel can hold up to fourteen separate foils in an arrangement designed to minimize any possible flux-depression interactions. (The exact extent of interactions was evaluated in a separate experiment which is now being analyzed.) In principle such an arrangement can evaluate the entire effective resonance integral curve in gold at a given temperature in a single exposure (See Figures 3 and 4). This, in turn, more than doubles the possible experimental precision because it eliminates most of the uncertainties in inter-exposure normalization, varying thickness or placement of cadmium, and foil calibration factors.

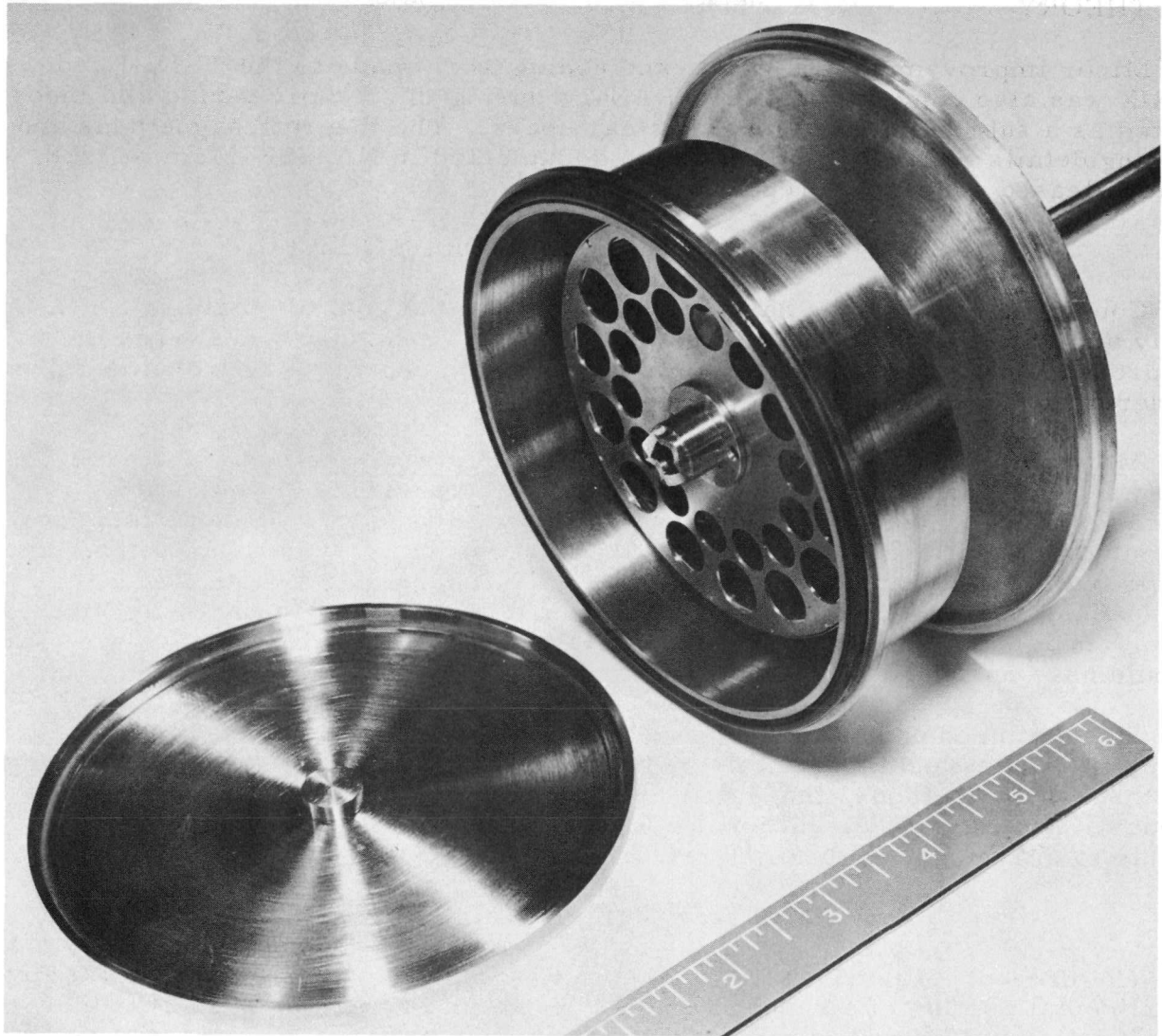


7529-2507

Figure 3. Sample Wheel with Heater Assembly

The apparatus described above was constructed, tested, and placed in service during this report period.

Other measurements using foils of many resonance absorbing isotopes were performed in an effort to establish the validity of neutron spectrum calculations for this core. These consist of measurements of the spatial dependence of the flux in the test region with dysprosium (absorbing mainly in Group 14, of the standard group split) indium (Group 13), gold (Group 12), tungsten (Group 11), and cobalt (Group 10). The measurements will be compared to the predicted radial dependence in each group and to the predicted flux gradient at the center of the test region in each group. (The gradient changes sign in this energy region.) These measurements are continuing, but analysis is not yet complete.



7549-1841

Figure 4. Capsule Assembly with Bottom Plate Removed

An improved data reduction and analysis code package has been completed and is undergoing checkout. The statistical analysis provisions for foil activation measurements have been greatly extended in scope and validity. Changes in input formats should greatly speed up data reduction.

NAA-SR-Memo-11321, describing improved methods for determination of paired-pulse resolution in counter systems, was released during this report period. Accurate knowledge of the paired-pulse resolution, τ , is very important to the success of these measurements, since uncertainties in τ are the largest single source of systematic error.

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C. THEORY

Minor improvements in format and coding were made to the TRIX-I code. TRIX was also converted to FORTRAN-IV during this report period and incorporated as a subroutine in several nuclear codes. The theoretical methods and coding details embodied in TRIX were documented in NAA-SR-Memo-11538, which was released during this report period.

IV. EVALUATION OF EFFORT TO DATE

Considerable progress has been made toward the goal of obtaining a reasonably accurate, reliable, and inexpensive calculational tool. TRIX-I is the first approximation to this goal, and represents a high degree of improvement over older analytical methods.

Although TRIX-I is an improvement in calculational methods, the experimental portion of the program has shown that there are some potentially serious areas of inadequacy. It must be noted that the present experimental approach, combining Doppler effect and resonance integral measurements in "clean" materials over a wide range in S/M, is primarily responsible for uncovering the discrepancies. It is very unlikely that standard measurements involving a narrow range in S/M in materials with comparatively complicated resonance structures would have uncovered the discrepancies.

The continued program, using additional experiments designed to test specific approximations and theoretical predictions, is expected to lead a second iteration in theoretical methods, that is, to further improvements in the TRIX series of codes. As verified calculational methods, these codes will be applicable and valuable to either fast or thermal reactors.

V. NEXT REPORT PERIOD ACTIVITIES

The present measurements in gold will be completed, analyzed, and the experimental portion of a report will be started. Comparison of TRIX with experiment and with the numerical calculations of ZUT or similar codes will be started. Data reduction will be continued on the present U²³⁸ measurements and, hopefully, completed. Apparatus to allow U²³⁸ measurements at selected S/M at 1000 to 1500°C will be constructed and preliminary measurements made.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program:	Nuclear Safety — Chemical Reactions		
Project:	Fission Product Retention by Reactor Coolants		
Project Engineer:	A. A. Jarrett		
Reporting Period:	July-September	AEC Category:	04-60-10-01
General Order:	7608	Subaccount:	1331

I. PROJECT OBJECTIVES

The general objective of the project is to determine the ability of reactor coolants to retain specific fission products released to the coolant stream as a result of fuel cladding failure or fuel meltdown. Specific objectives of the project are: (a) the determination of the ability of sodium to retain radioactive iodine as a function of such parameters as sodium temperature, elapsed time from incident, sodium to fuel and sodium to gas volume ratios and geometric relationships, condition of released iodine; and (b) the determination of the ability of boiling sodium and sodium in the vapor phase to retain radioactive fission products.

II. PROGRESS DURING REPORT PERIOD

Maypack and diffusion tube components were characterized in a sodium vapor environment. The retention characteristics of sodium were verified in a capsule rupture experiment under conditions of inert gas-fission product bubble transfer. This completed the experimental activities on this project.

Three experiments were performed to study the effectiveness of Maypack and diffusion tubes for the selective retention of various iodine forms in a sodium vapor atmosphere. In the first of these experiments, a preirradiated uranium foil was melted/vaporized by the condenser discharge and the resulting effluent transferred by a helium flow of 200 cc/min directly to a Maypack and diffusion tube network as described in previous reports.

In the second experiment, the Maypack and diffusion tube were subjected to sodium vapor and subsequent condensation and deposition for a time equivalent to the total helium flow from the condenser discharge reaction chamber. The foil was then melted/vaporized similarly to the first experiment, and the effluent was conducted through the pretreated Maypack and diffusion tube network.

The third experiment conducted the effluent gas from the vaporized fuel ribbon directly to a sodium plenum where it was allowed to bubble through 4 in. of sodium at 900°F for the same time period as in the preceding two experiments.

The second experiment showed a considerable deposition of sodium (approximately 3 gm) on the first three copper screens and as far downstream as the charcoal filter paper of the Maypack. The activity, however, was not homogeneously distributed throughout the condensed sodium, as was determined by comparing several portions of the sodium. Considerable iodine and other fission products were observed on the Maypack components. These results, shown

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in Figure 1, demonstrate the relative retention of the Maypack components for the various isotopes measured under the pretreated sodium vapor condition.

The third condenser discharge experiment, in which a sodium plenum was used, showed little deposition of fission products on the Maypack components. A small quantity of sodium (1/2 gm) was observed to have condensed on the first copper screen filter, as in the preceding experiment. Analysis of this sodium indicated no appreciable radioactivity. Diffusion tube data were obtained.

The capsule rupture experimental equipment was assembled and tested, and a capsule discharge experiment performed. Figure 2 shows the overall arrangement of the components, while Figure 3 is a closeup of the gas chamber and pressure pulse initiating device to rupture the capsule.

The capsule rupture experiment was conducted by preirradiating 129 mg of enriched uranium foil in a capsule 1 in. in diameter by 3.25 in. long. This capsule was affixed to a long tube and inserted in the simulated sodium plenum. The quantity of sodium was 4500 gm, and the height of sodium above the discharge rupture disc of the smaller capsule was approximately 5 in. The sodium was contained in a cylinder 7-in. in diameter by 19-in. high and the temperature of the sodium was elevated to 1000° F for two hours, after which a pulse of 100 psi of argon gas was transferred to the small cylinder. This ruptured the discharge disc (calibrated for rupture at 75 psi and 500° F) and transferred the capsule contents through a 1-in. orifice nozzle, 3/16 in. in diameter, to the sodium column in the large cylinder.

The plenum above the sodium was charged with helium flowing at 200 cc/min, and the resulting cover gas transferred through a Maypack diffusion tube arrangement. A photograph of the disassembled Maypack components is shown in Figure 4. Analysis of the Maypack and diffusion tube components indicated no measurable radioactivity on the diffusion tube; however, I^{131} and Xe^{135} were observed throughout the Maypack. Subsequent analysis of the sodium was effected by lowering the temperature of the sodium to 400° F, and sampling at this temperature.

Ten sodium samples were obtained at different sodium levels as the sodium in the plenum was drained into the lower tank. Analysis of the sodium samples indicated a rather complete spectrum of fission product activity, including iodine, Zr-Nb, Ba-La, and Cs^{137} . Results indicate an iodine release approximating 1×10^{-4} , as obtained by relating the total iodine measured in the gas sampling devices to the extrapolated iodine activity in the sodium.

A paper* was prepared for presentation to the International Fast Reactors Conference, October 9-11, at Argonne National Laboratory. An outline has been prepared for the summary report.

*S. Berger and W. P. Kunkel, "Fission Product Retention in Sodium and Applications to Vented Fuel Element Design."

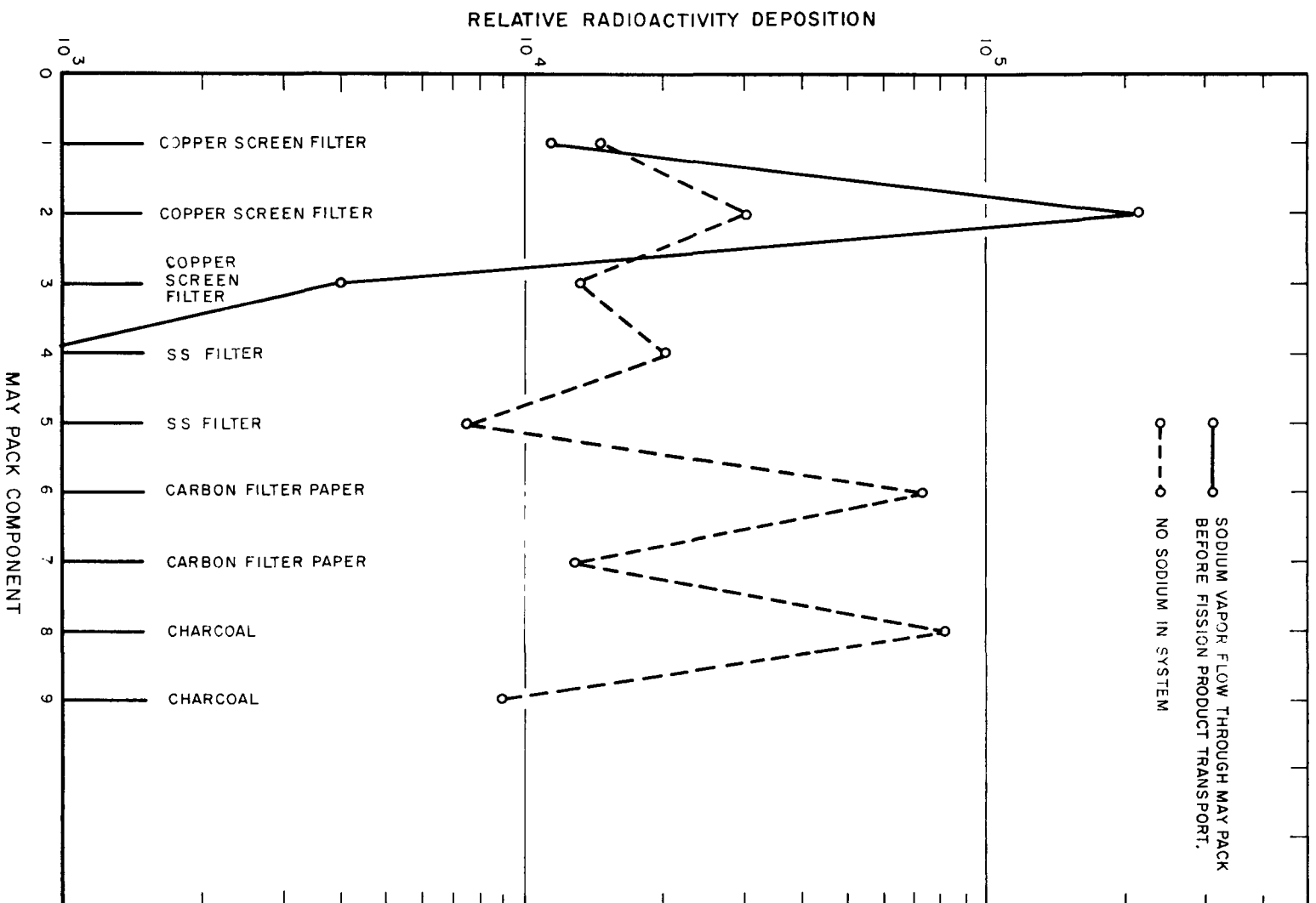
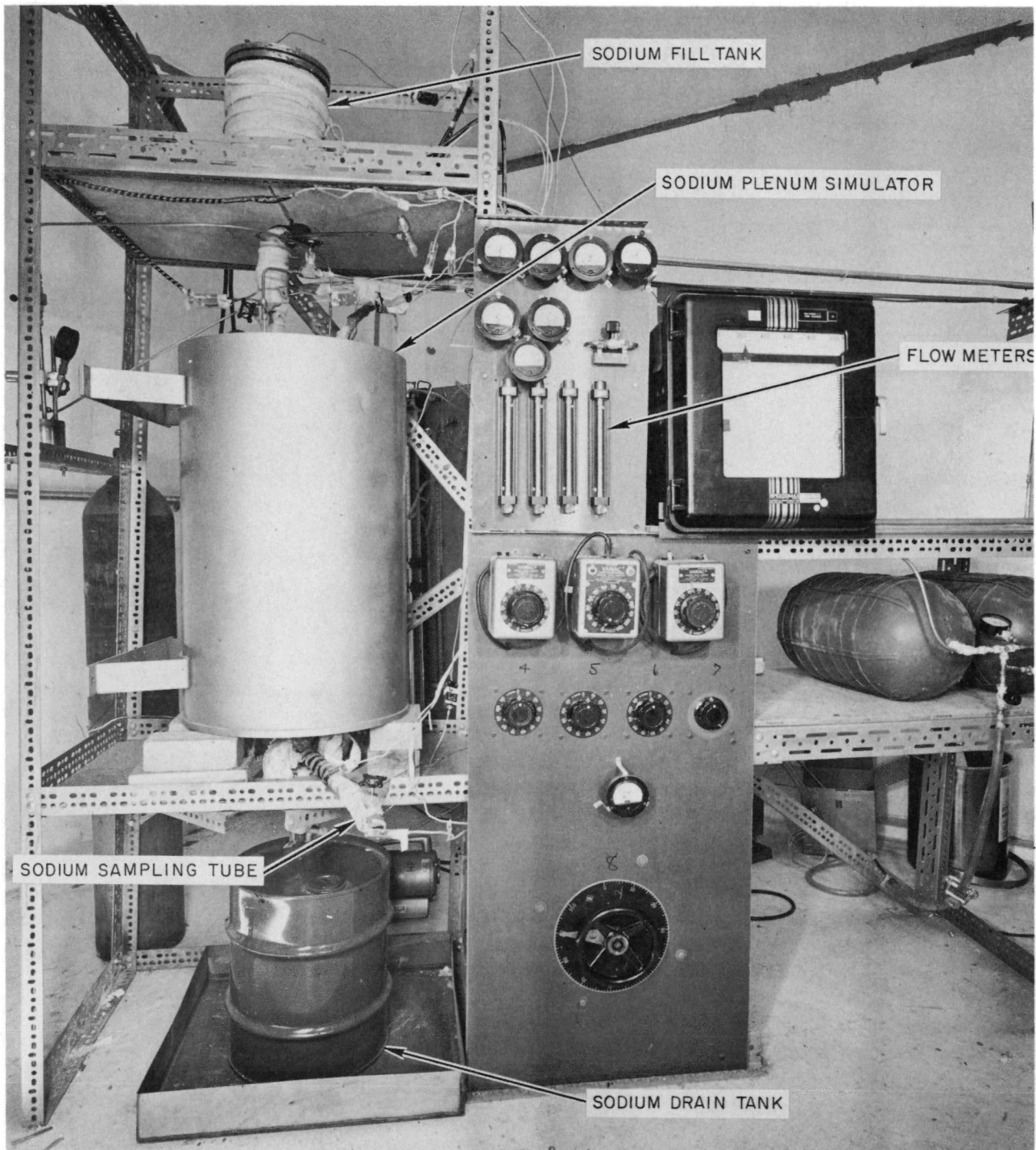


Figure 1. Comparative May Pack Retention With and Without Sodium Vapor in Effluent (I¹³¹ Results)

7608-13310

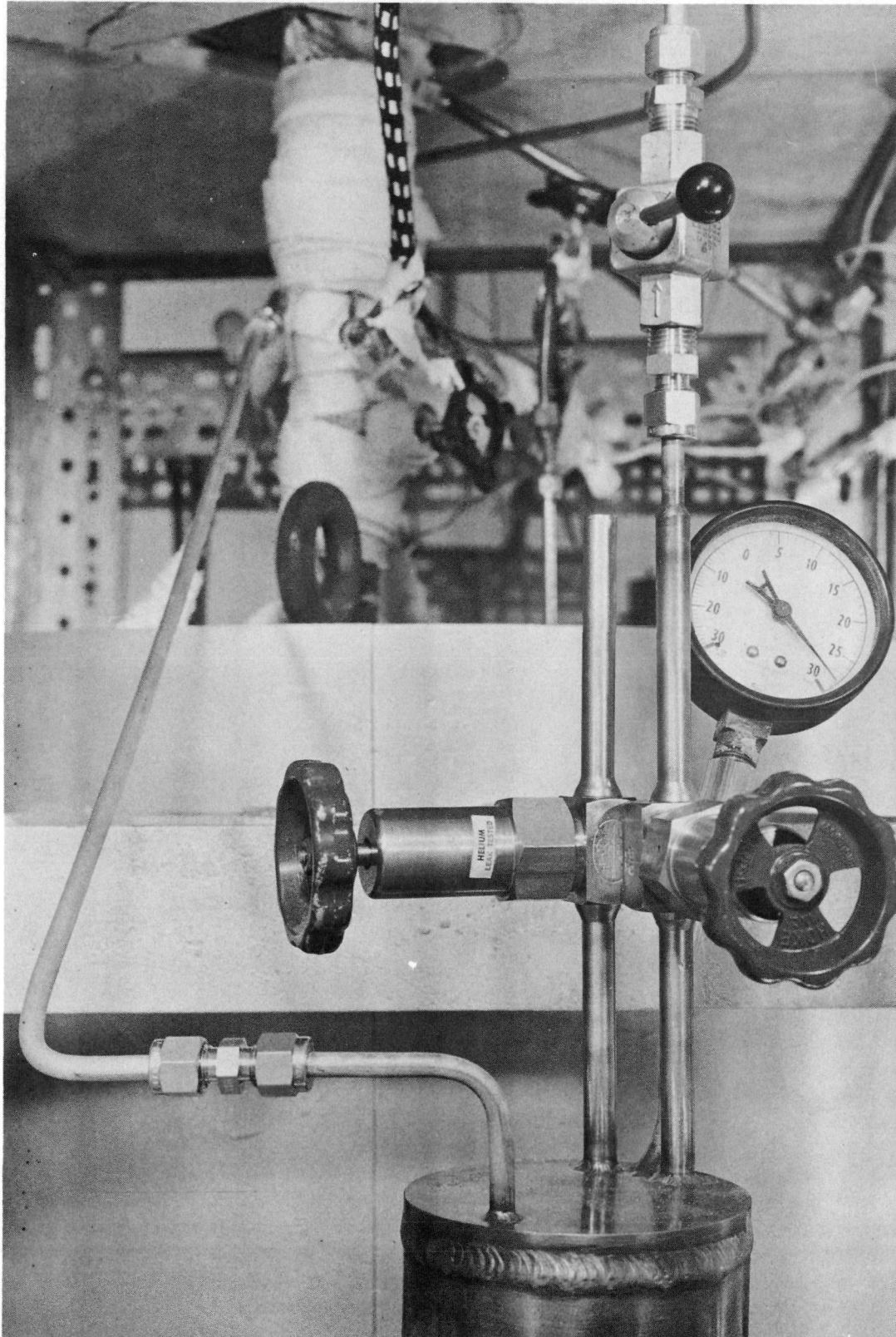
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Figure 2. Component Arrangement in Capsule Rupture Experiment



7608-4016

Figure 3. Gas Chamber and Pressure Pulse Initiator

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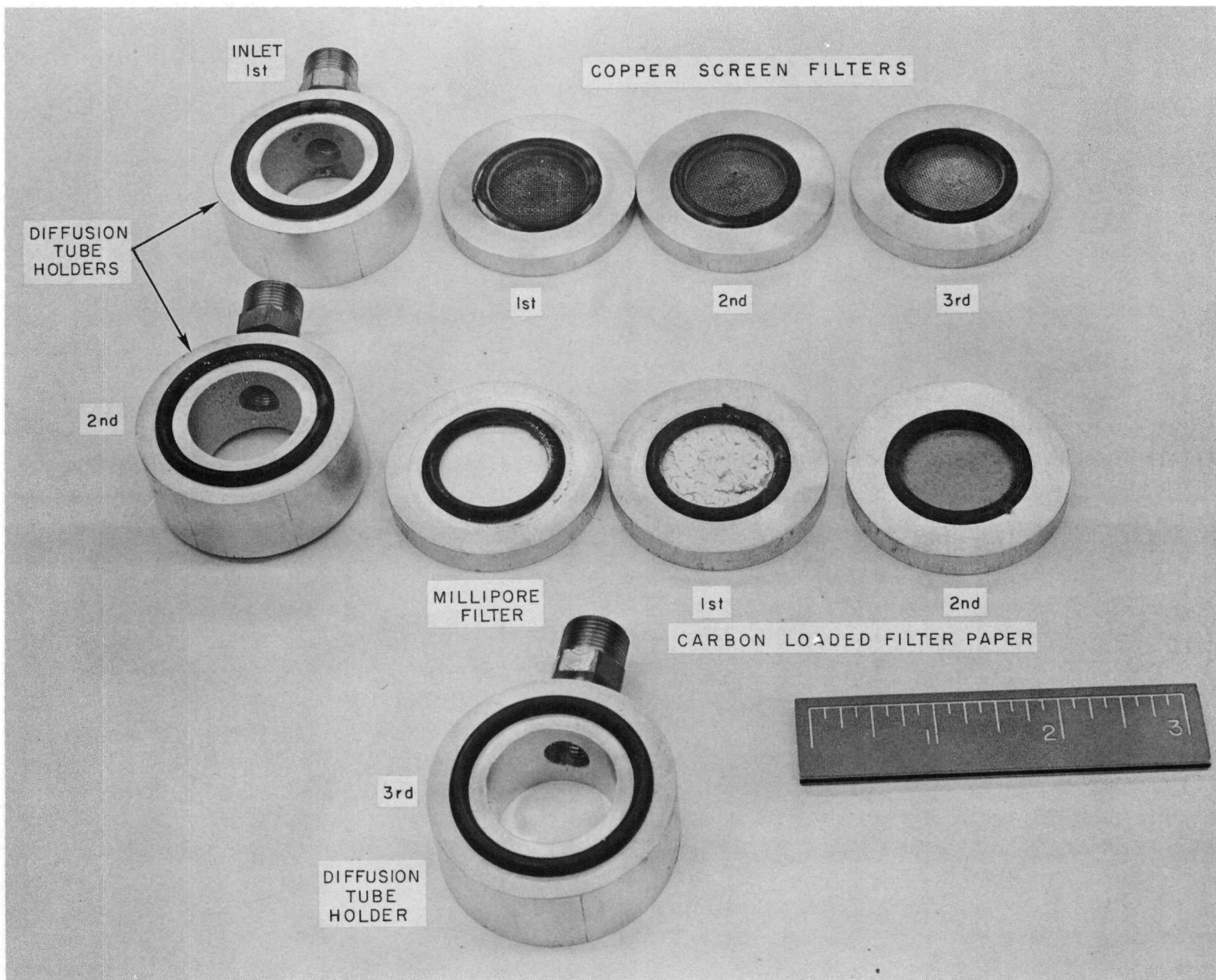


Figure 4. May Pack Components

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III. EVALUATION OF EFFORT TO DATE

The three condenser discharge experiments associated with the Maypack characterization have yielded significant information concerning the effect of sodium vapor on the Maypack components. The data show that even after condensation, the sodium may represent a sink for radioiodine activity and thus obscure the validity of the Maypack device for characterizing the forms of iodine.

The successful operation of the capsule rupture experiment has resulted in some data which indicate that, under the specific conditions of this experiment, the iodine retention properties of sodium (as measured in previous capsule experiments) may be subject to modification.

However, it is probably appropriate to employ the earlier figure of 0.02% as the release of the iodine in the sodium with more caution, since this figure had included a safety factor of approximately 10 over the capsule results.

IV. NEXT REPORT PERIOD ACTIVITIES

No further experimental effort is planned in accordance with plans for project termination. The summary report of the project will be completed and published.

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A Division of North American Aviation, Inc.

Program: Nuclear Safety – Gaseous Effluent Studies

Project: Characterization of Sodium Fires and Fission Product Release

Project Engineer: A. A. Jarrett

Reporting Period: July-September 1965

AEC Category: 04-60-40-04

General Order: 7640

Subaccount: 1374

I. PROJECT OBJECTIVES

The general objective of this project is to develop experimental information and analytical methods which characterize the release and transport of effluents and energy generated during a primary-coolant-sodium accident. The source of energy generation may be from the escaping coolant (by rapid thermal energy transfer) and/or subsequent combustion of the sodium coolant. The effluents are sodium or its oxide in particulate form and selected fission products, available as potentially serious dispersions of radioactivity. An analytical model will be developed for the design and safeguards analysis of sodium cooled fast reactors.

II. MAJOR ACCOMPLISHMENTS DURING REPORT PERIOD

Planning and evaluation were completed to define the detailed requirements of a 5-yr program for characterizing sodium fires and fission product release following primary coolant accidents in sodium cooled fast reactors. The program planning effort was summarized and presented to the AEC. As part of the planning effort, the test installation requirements for the project were established and a Construction Authorization Request was submitted.

III. PROGRESS DURING REPORT PERIOD

A. PROGRAM PLANNING

A major effort involved completion of a detailed planning and evaluation study to define requirements of a 5-yr program for accomplishing objectives of the project. The planning study consisted of a state-of-the art review of sodium fires research, and of fundamental concepts of combustion, mass and energy transport, and aerosol behavior, all of which serve as the base for future work. The problems requiring solution were evaluated on the basis of postulated sodium-coolant accidents which are pertinent to the program. A technical approach to the solution of these problems was outlined, and includes experimental and analytical studies of fundamental mechanisms and modeling principles regarding the release and transport of matter and energy. Proposed experiments and the apparatus (existing and proposed) and facilities for conducting them were delineated, including: fundamental energy, matter, and fission product release studies for pool and spray fires; large fundamental and modeling fires; and test chambers for studying pressure generation from coolant sprays. A conceptual description of a test installation for conducting large fundamental and modeling experiments was incorporated in the study, based upon utilization of an available, existing facility. The scheduling aspects of the 5-yr program, including milestones, were also evaluated under the planning study.

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B. SODIUM FIRES TEST INSTALLATION PLANNING

The test installation requirements for conducting large fundamental and modeling sodium fires experiments were established during the program planning studies described above. These requirements were summarized in a Construction Authorization Request for the installation and transmitted to the AEC Canoga Park Area office. The test installation will be located in an available portion of North American Aviation Critical Facility, Building 009, Santa Susana. This facility was evaluated and found desirable for the test installations. Two critical assembly rooms are located in this building. The assembly room and control rooms previously occupied by the OMR Critical Assembly will be utilized for the project. The test room contains 1520 ft² of floor area, 33 ft high, with a 4-ft-thick concrete wall separating the room from the rest of the facility. A 5-ton bridge crane covers the test assembly area. A 19- by 12- by 10-ft deep pit is located within the test room. A schematic flow diagram of the process system and equipment for the burning experiments is shown in Figures 1 and 2.

The design and construction of the test installation is divided into two phases:

Phase I – Modified large fires apparatus for performing large fundamental release experiments, and equipment and instrumentation common to Phase I and II apparatus.

Phase II – Modeling fires apparatus.

C. FUNDAMENTAL EXPERIMENTS

1. Bench-Scale Fires Experiment

An inlet manifold was assembled which permitted variations in nitrogen flow from 5 to 36,000 cc/min, and from 0 to 1200 cc/min for the oxygen. Appropriate pressure-relief devices were added to limit the flow meter pressure to 4 psi. The manifold was designed to allow calibration of all flow meters in the circuit without disassembly from the small fires apparatus.

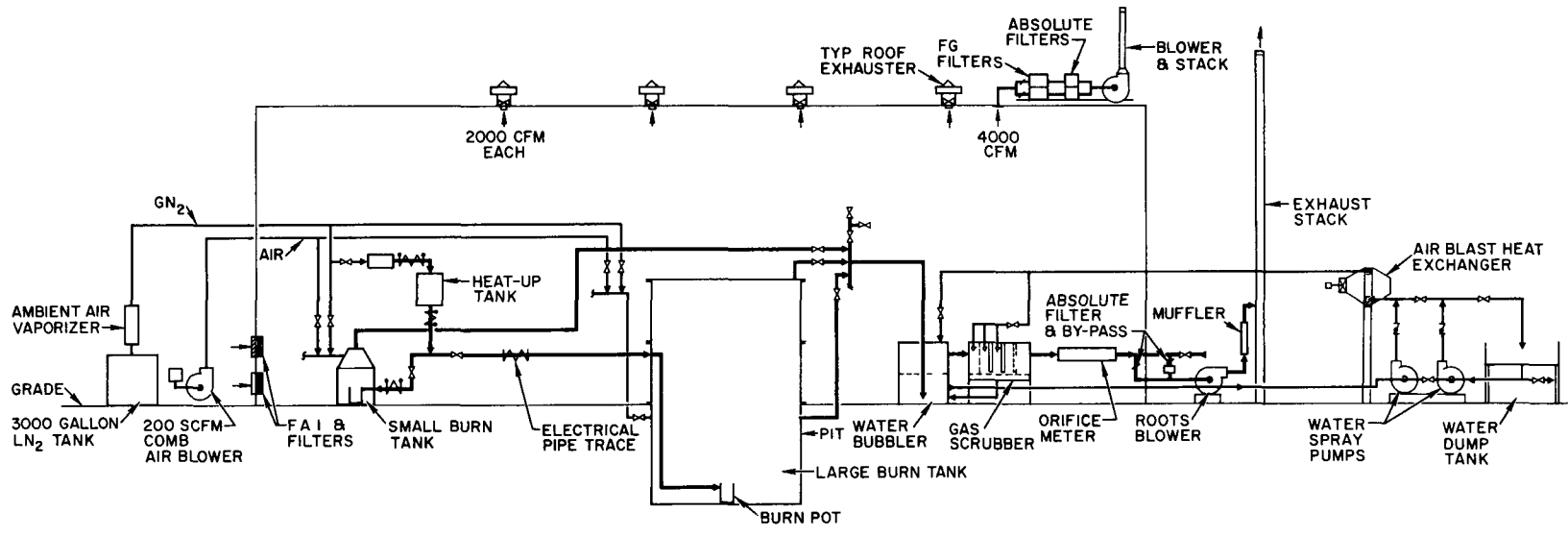
A second manifold was built which allowed the O₂ and H₂O analysers to sample the N₂ line, air inlet to the burn pot, burnpot exhaust, and to be calibrated during the burning experiment.

The cleanup system of the bench-scale apparatus was designed for greater flow and more efficient oxide removal. Checkout of the modified apparatus was initiated at the close of the report period.

2. Large Fires Experiment

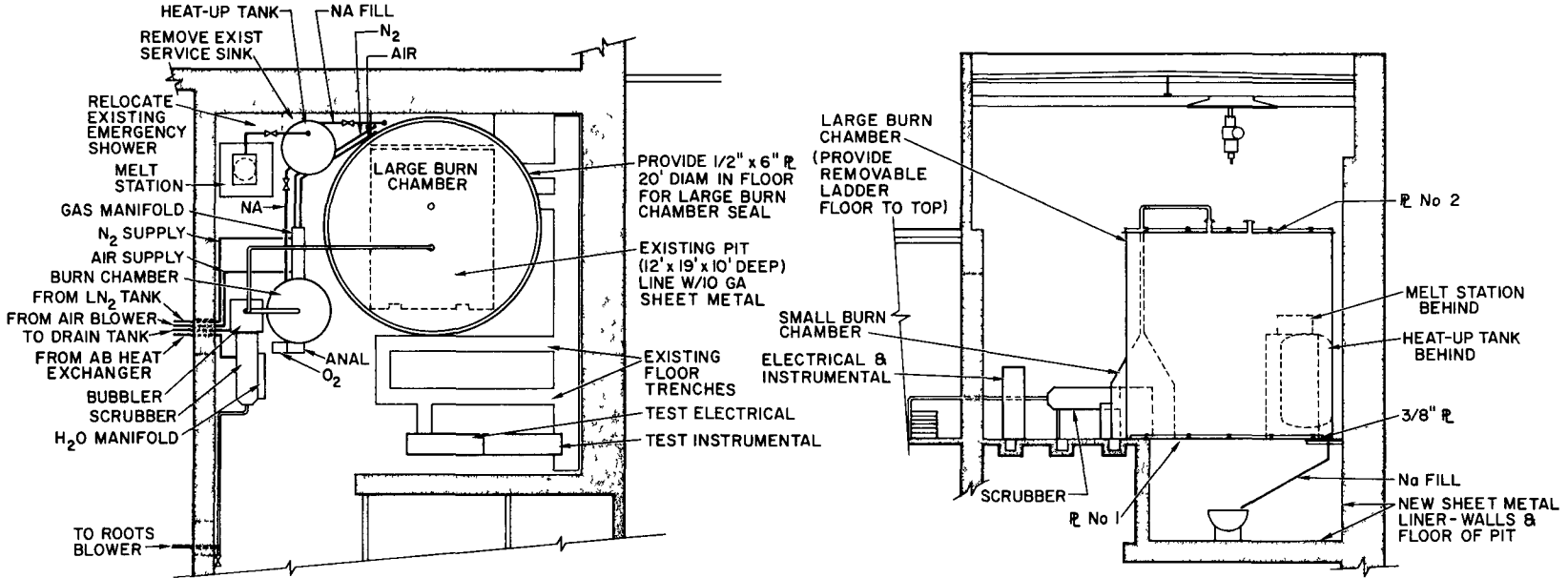
The equipment for the large fires experiment which was operated at the old outdoor facility was moved to the Sodium Fires Test Installation in Bldg 009. The experimental equipment (burn chamber and bubbler-scrubber) will be located inside of the building, and the process and motorized equipment will be located outside.

The following changes for the modified large fires apparatus were proposed. The burn pot-burn chamber annulus will be fully insulated and the burn chamber cover will be insulated. At least 30 thermocouples will be placed in the gas space above the fire. More gas samples will be taken in the gas



7640-2510

Figure 1. Conceptual Process and Facility Flow Diagram



7640-2511

Figure 2. Conceptual Building Layout

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space. A new heatup tank and melt station will be used for both the large fires and modeling experiments. A low-pressure N_2 gas feed system will include a large liquid nitrogen storage tank and an ambient air vaporizer. An airblower will provide air for mixing with N_2 for obtaining desired gas mixtures. An electric heater will be provided for adjusting gas temperatures. Considerable attention is being given to the design of gas sampling and temperature measurement near the combustion surface as well as some distance from it.

3. Initial Sizing Apparatus

The purpose of the initial sizing apparatus is to obtain measurements of the sodium particle-size distribution immediately after its production. This apparatus has been designed, and the components required for its fabrication have been ordered. The technique by which this is done is similar to tranquil settling, except that laminar flow is used to separate the particles as they fall. One advantage of this technique is that it allows one to obtain different groups of particle sizes for other studies. Another advantage may be that one can vary the rate at which the oxide is produced and thus observe any difference in the distribution which flows downstream. The relation between the length required for 100% deposition and size for circular pipes is:

$$L = \frac{8 Q l}{3 R \pi \nu} , \quad \dots(1)$$

where

Q = flow rate, $\frac{\text{cm}^3}{\text{sec}}$

L = pipe length (in.)

R = pipe radius (cm)

ν = settling velocity $\left(\frac{\text{cm}}{\text{sec}}\right)$.

For $Q/R = \frac{\text{cm}^2}{\text{sec}}$, the relation between L and diameter D in microns for sodium oxide is $\sim L \frac{(\text{cm})}{100} = \frac{100}{D^2}$; i. e., a micron particle will travel 100 cm and a 10-micron particle will travel 1 cm in a pipe of 1-cm radius for a flow of $1 \text{ cm}^3/\text{sec}$, or a 5-cm radius and a $5\text{-cm}^3/\text{sec}$ flow rate. The designs of sampling ports and plating distances are all based on this simple relationship.

4. Particulate Sampling and Sizing Development

a. Midget Impinger

The midget impinger is an air sampler normally used to collect aerosols which are ultimately sized by optical microscopes. An airstream is pulled through a specially designed nozzle which forces the particulates to impinge on the bottom of the container. A solution is provided as a collection medium, and is also used for dispersion of the collected material when microscopic studies are made. When the particulate material is insoluble in the solution, it is frequently found

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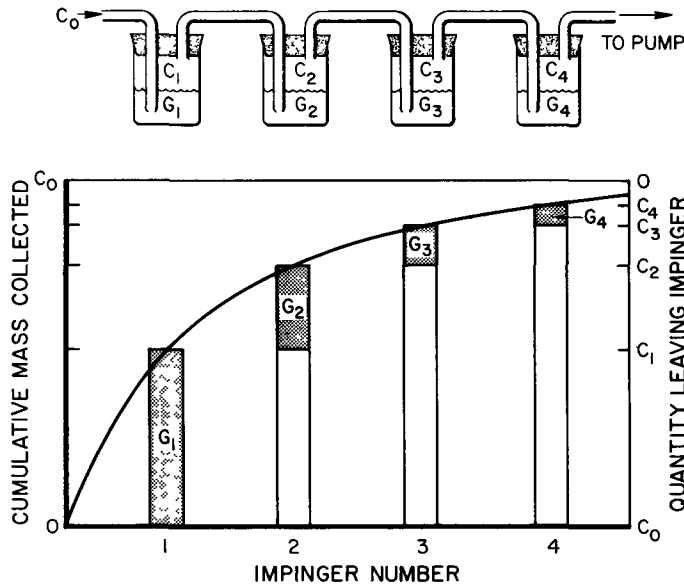
that the efficiency of collection decreases with decreasing particle size, since impingement, like impaction, is a function of inertia and the size of jet.

In the present program, the impinger is used exclusively as a sodium concentration measuring device; i. e. it is desired that the sodium oxide aerosol be collected and dissolved in the collection media. In this way, a simple measurement of the sodium in solution will yield the desired information.

Several experiments have been performed to determine the efficiency of the midjet impinger for sodium vapor and/or its oxide when water is used as the collecting agent and solvent. These experiments were performed so as to gain some knowledge of efficiency, since the data generated from impinger samples and from the scrubber-bubbler collection system of LF-2* were not compatible; i. e., all impinger samples contained less sodium per ft³ of sampled air than that collected in the scrubber from the same air.

Sodium smoke was allowed to partially fill an 18-in. -high, 0.15-ft³ container which was covered on both ends after filling. An inlet to the impinger was placed at the bottom of the container. A 1-min sample at 0.1 cfm was obtained from the partially closed container. Approximately 10 minutes later, another sample was obtained over a 2-min interval at the same sampling rate. In order to evaluate the efficiency, 4 impingers were placed in series and the concentration of sodium was measured in each container. The measurements of concentration were performed with a flame photometer whose sensitivity is about 0.5 ppm. The first samples in the series had to be diluted in order to be measured with the photometer, and thus were measured with no more accuracy than the latter collectors.

Figure 3 gives a schematic representation of the technique used to evaluate the efficiency.



7640-2512

Figure 3. Impinger Efficiency Test

*Annual Technical Progress Report, Fiscal Year 1965, NAA-SR-11450, p 2 VII-110

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The quantity of sodium entering the impinger is

$$C_o = S_o Q t \quad \dots (2)$$

where

C_o = inlet airborne content (μgm)

S_o = concentration being sampled ($\mu\text{gm}/\text{cm}^3$)

Q = sampling rate (cm^3/sec)

t = time (sec).

The quantity of sodium leaving an impinger is equal to the quantity entering less the quantity collected,

$$C_i = C_{i-1} - G_i \quad \dots (3)$$

where

C = quantity leaving (μgm)

G = quantity collected (μgm)

i = impinger number.

The efficiency of an impinger is

$$\epsilon_i = \frac{G_i}{C_{i-1}} \quad \dots (4)$$

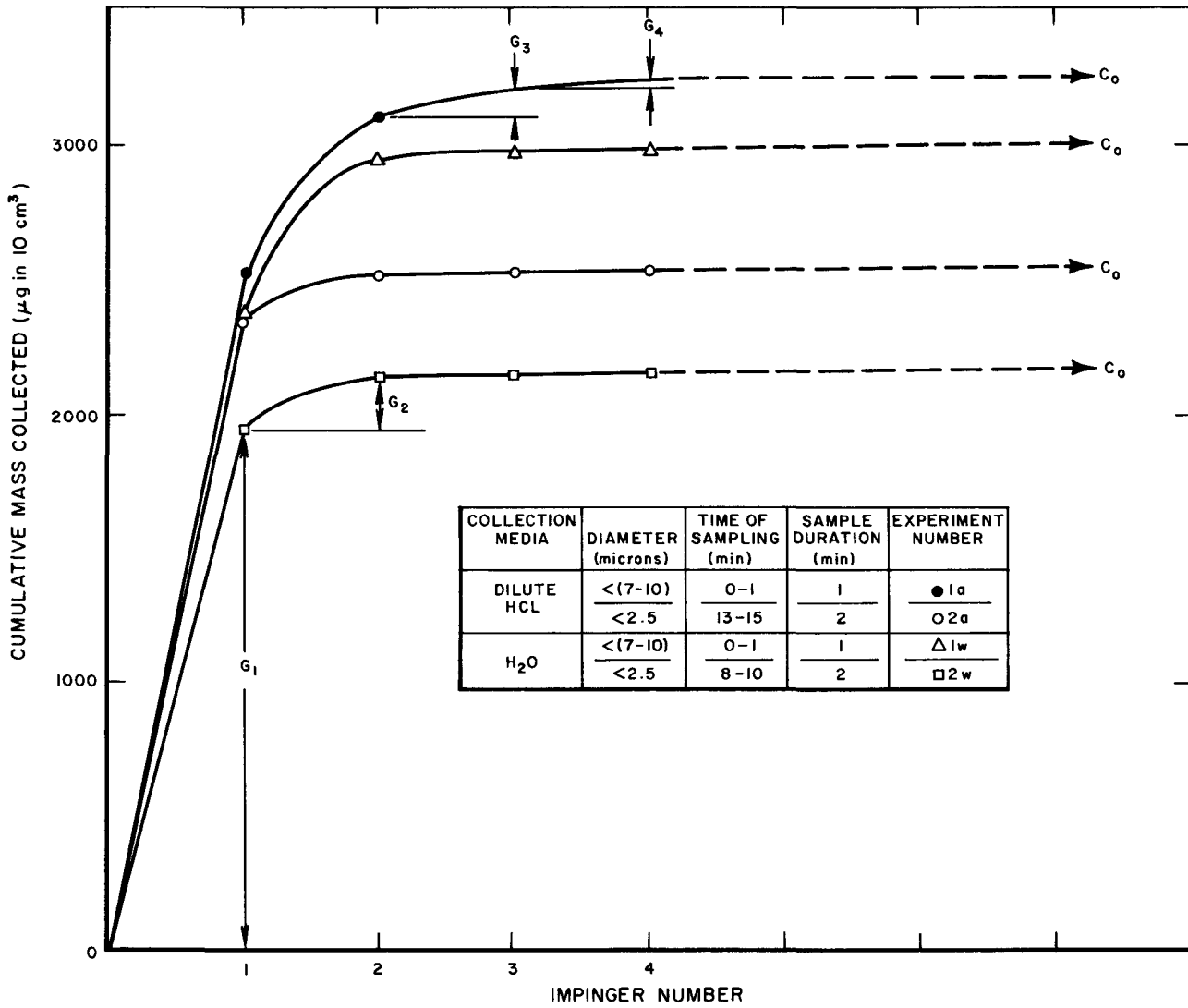
The relative efficiency of an impinger is

$$E_i = \frac{G_i}{C_o} \quad \dots (5)$$

The cumulative efficiency of a series of impingers is

$$\Sigma E_i = \frac{\Sigma G_i}{C_o} \quad \dots (6)$$

If all impingers have the same fairly large efficiency, one should approach 100% collection with a small number of collectors, and the efficiency would be approximately equal. If, however, the efficiency decreases as the collectors are added in series (due to particle-size decreases), then there may not be equal collector efficiency efficiency (i. e., if $\epsilon_4 < \epsilon_3 < \epsilon_2 < \epsilon_1$). The table presents the data and gives



7640-2513

Figure 4. Impinger Efficiency as Indicated by Total Mass Collected

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Experiment No.	Impinger No.	Quantity Collected G_i (μgm)	Cumulative Quantity Collected ΣG_i (μgm)	Relative Efficiency E_i	Cumulative Efficiency ΣE_i	Efficiency E_i
1a	1	2530	2530	0.77	0.77	0.77
	2	585	3115	0.18	0.95	0.80
	3	103	3218	0.03	0.98	0.73
	4	28	3245 3255*	0.008	0.988	0.73
2a	1	2340	2340	0.915	0.915	0.915
	2	177.5	2517.5	0.07	0.985	0.865
	3	12.5	2530	0.005	0.990	0.44
	4	6.3	2536 2546*	0.0025	0.992	0.39
1	1	2370	2370	0.790	0.79	0.79
	2	582	2952	0.194	0.984	0.92
	3	22.5	2974	0.007	0.991	0.47
	4	16.0	2990 3000*	0.005	0.996	0.64
2	1	1940	1940	0.895	0.90	0.90
	2	182	2122	0.084	0.98	0.805
	3	22.5	2145	0.010	0.99	0.50
	4	12.5	2157	0.005	0.995	0.60

*Inlet airborne content – extrapolated value

the computation of efficiency based on C_o , the extrapolated total mass collected. The data are plotted in Figure 4. The preliminary conclusions from these experiments are as follows.

1) Impingement of particulates of sodium in solutions in which they are soluble is sufficiently efficient in one impinger for concentration studies where only mass is of interest.

2) If the data taken by impingers are to be used to establish particle sizes (as in a tranquil settling chamber when the concentration at a particular height is measured), then two samplers in series should be used to assure a total efficiency which is greater than 90%.

b. Cascade Impactors

One measurement made during the LF-2 experiment was of considerable interest to the overall project goal. This measurement was concerned with the relative distribution of the I^{131} activity as a function of the particle size of the sodium aerosol which was released from the sodium surface. Since the Lovelace impactor samples were small (~10 micrograms of sodium per stage) and the specific activity of the sodium was only $5 \mu\text{c}/\text{lb}$, it was most difficult to measure the I^{131} activity ($10^{-7} \mu\text{c}/\text{stage}$). Measurements were finally obtained

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by grouping 10 stages together and counting in a low-level iron shield for several hours using a multichannel analyzer.

A new cascade impactor called an Anderson sampler has been purchased. The design of this sampler represents an improvement over the Lovelace cascade impactor. It is a round-jet impactor with sizes similar to the Lovelace-Mercer design. However, this sampler has 400 holes per stage, each of which can sample at rates similar to the Lovelace. Thus, for equivalent sampling times one will obtain $400 \times 10^{-7} \mu\text{c}/\text{stage}$ for the same low specific activity. Several measurements of the oxide distribution of a hood fire have been made with the Anderson and Lovelace impactors. The 50% sizes and standard deviation as measured with both instruments agreed to within 10%. The manufacturer of the Anderson impactor provides extensive information on the distributions per stage, mass median diameter per stage, and the 50% cutoff value per stage in terms of the aerodynamic diameter.

A modification of the design of the Lovelace impactor has been made. This design reduces the number of parts necessary, thereby lowering the cost of construction. The modified design will also save time in loading and unloading the impactor.

c. Turbimetric Particle Size Analysis

An Eagle-Picker turbidimeter has been obtained which is a settling device used to determine size distribution of materials which are suspended in liquids. A beam of light is passed through the suspension, and the light transmitted through the suspension is measured by a photcell. As the particles fall in the liquid, the change in light transmission is measured. From these measurements, the particle size and distribution of the material are calculated. Liquid media make excellent tranquil settling devices since the turbulence involved in loading lasts only a few seconds. The main concern is in the mechanics of getting an airborne sample collected in the suspension liquid. Preliminary calibration of this device indicates that it will be difficult to measure size distribution of Na_2O ($\rho = 2.3$) in suspensions of organic liquids ($\rho = 0.8$ to 1). The time of fall is directly related to density differences of fluid and particle, viscosity of the media, the square of radius, and the height of chamber. Since the density of the particles may be less than the true density, the typical use of the device may not be practical. The instrument has been modified into an air sedimentation tube by adding an additional settling height (~ 2 ft) onto the existing cell housing which contains the sedimentation cell used for liquids. This technique improves the density difference ($\rho_{\text{air}} = 1.2 \text{ mg/cc}$), and increases the height which compensates for the viscosity decrease of air. The first attempt at comparing the distribution as a function of time for transmission in the air suspension with that obtained from the same smoke which was allowed to settle in an organic liquid, differed by a factor of ~ 1.5 for the mmd (mass median diameter). The standard deviation was the same for both methods. This difference is probably due to the uncertainty in the actual height from which the particles are falling in air. Additional information on the technique will be obtained with standard particle size distributions.

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d. Royco - Light Scattering Instrumentation

During the last fiscal year, the Royco output pulse was modified to work with a multichannel analyzer. The system was calibrated, indicating a linear relation between the square root of the pulse height and the particle diameter over the range from 0.5 to 1.2 μ . Others have reported sensitivities down to 0.3 μ , and with the exception of the high noise level (due to preamplifier and high voltage power supply modifications), the modified instrument at AI would have achieved the same sensitivity.

There are several necessary variations in the physical design of the instrument, which need to be incorporated into the system along with the pulse shaper that has been designed. The improvements are necessary in order to count both higher concentration and smaller-sized particles. These improvements may be obtained in the following ways.

- 1) Increasing the linear flow rate in order to increase concentration.
- 2) Increasing the slit width in order to improve the amount of light received from the scattering particle (signal to noise).
- 3) Decreasing the pulse duration by decreasing the slit height or using lasers to define a smaller sensitive volume.
- 4) Decreasing the effects of signal to noise due to the scattered light resulting from the carrier gas. (He scatters 0.014 times that of air).
- 5) Varying the light source (ordinary to ultraviolet) lenses (to quartz) and incorporating suitable filters and detectors, one may change the entire response characteristic and lower the limit of detection.

D. ANALYTICAL DEVELOPMENT

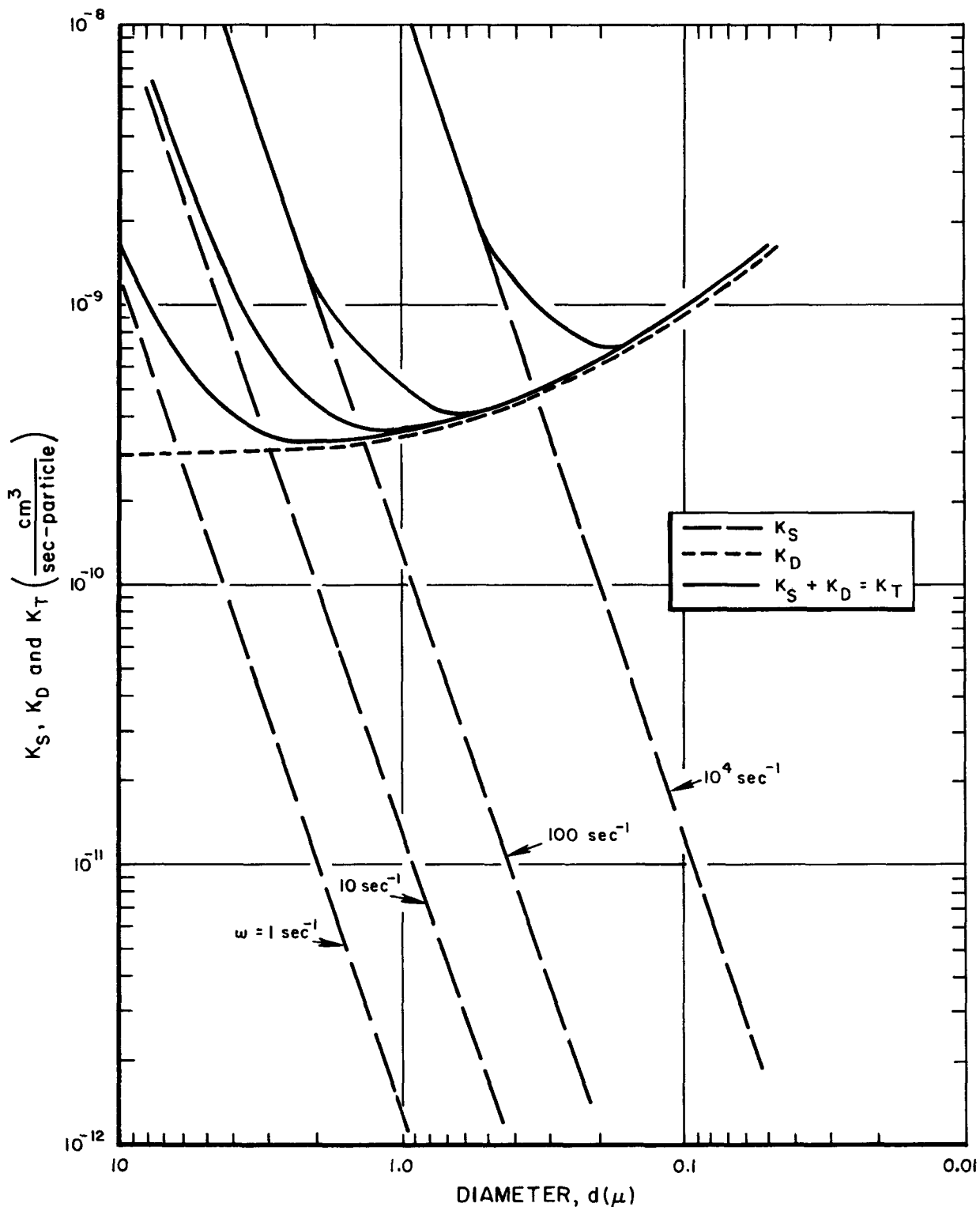
1. Particle Studies - Modeling

The theory underlying application of the phenomenon of agglomeration to particle-size distributions is necessary in order to predict or model the transport of material from sodium fires. The general equation describing agglomeration and settling for a uniform population has been derived, and is a nonlinear integro-differential equation of the convolution type. A solution for this equation is available for two limiting cases, i. e. where either agglomeration or settling was important. Solutions for nonuniform initial populations have not been reported.

The following discussion is based on a review of the literature on the subject of agglomeration, with particular emphasis on applications to the role of agglomeration in modeling large sodium fires.

a. Diffusion

One of the important phenomena that will influence the transport of the small particles formed in a sodium fire is agglomeration. Smoluchowski¹ appears to have been the first to state that Brownian movement or diffusion of the small particles causes coagulation at a rate proportional to n^2 , with n the concentration.



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Figure 5. Agglomeration Constants (K_D Diffusion, K_C Turbulent, and $K_T = K_D + K_C$) as a function of $d(\mu)$ diameter for various values of $w(\text{sec}^{-1})$ the velocity gradient

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Thus

$$\frac{dn}{dt} = Kn^2$$

where K is the Smoluchowski constant for agglomeration due to diffusion and is $= \frac{4kT}{3\eta} = 3 \times 10^{-10} \frac{\text{cm}^3}{\text{part-sec}}$ in air at T - 20°C; k is Boltzman's constant and η is the viscosity of air = $1.82 \times 10^{-4} \frac{\text{dyne-sec}}{\text{cm}^2}$. The equation holds exactly for large spherical particles of the same size; it holds satisfactorily for moderately nonuniform sized aerosols of nonspherical particles.^(2,3) For small particles with diameter comparable to the mean free path in air, one applies the Cunningham correction factor:

$$-\frac{dn}{dt} = Kn^2 \left(1 + A \frac{\lambda}{d}\right) = K_D n^2 \quad \dots (7)$$

$$K_D = \frac{4kT}{3n} \left(1 + A \frac{\lambda}{d}\right) \quad \dots (7d)$$

In air at 20°C, the mean free path $\lambda = 10^{-5}$ cm. A is a constant which corrects for slip; d is the particle size. The correction factor essentially takes account of the fact that very small particles have a better chance of colliding with each other by "slipping" the air molecules. The agglomeration constant, K_D , as given above related to the expression for the diffusion constant D for small particles:

$$D = \frac{kT}{3Tnd} \left[1 + A \frac{\lambda}{d}\right] \left(\frac{\text{cm}^2}{\text{sec}}\right) \quad \dots (8)$$

$$K_D = 4\pi d D \frac{\text{cm}^3}{\text{part-sec}} \quad \dots (9)$$

The applicability of Equation 8 for particles as small as gas molecules has been tested by J. W. Thomas.⁴ He showed that using literature values of D in Equation 8 produced values of the molecular diameter, d, that agreed with values in the literature with errors that do not exceed 50%. He concluded that the expression for D is accurate for all particles in air down to 0.001-micron radius, and that "it is accurate to better than 30% for other materials in this size range." In the same report⁴ Thomas showed the validity of using a "diffusion battery" technique for determining particle sizes. He checked his results by comparisons based on a standard light scattering method, with results agreeing to within 30%. Thomas gives a convenient graph of D vs d for a range of values extending from d = 0.004 to 2.4 microns. Equation 9 above was used to obtain K_D based on Thomas' curves, and the result is given graphically as K_D in Figure 5. The graph shows that K_D is virtually constant for particle sizes greater than 0.5μ , but increases rapidly with diminishing sizes less than 0.1μ . Clearly diffusion is an important mechanism for agglomeration for particles with diameters less than 0.1μ .

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b. Turbulence

The presence of velocity gradients in turbulent flow results in an increase in coagulation rates, since particles moving in adjacent streamlines tend to collide more frequently. An agglomeration coefficient K_S can be defined by an equation similar to 7:

$$-\frac{dn}{dt} = K_C n^2 \quad \dots(10)$$

Smoluchowski⁽¹⁾ gave a theoretical derivation for an expression for K_S based on the idea of a dispersion subjected to simple shearing stress:

$$K_S = \frac{4}{3} d^3 \omega, \quad \dots(11)$$

with

ω = velocity gradient.

Teverovskii⁽⁵⁾ reports values for K_S that are 10 to 4,000 times that due to diffusion in atmospheric smokes. If $d = 2\mu = 2 \times 10^{-4}$ cm, then $K_S = 10^{-11} \omega$; thus some of the large values reported by Teverovskii would be possible only with very large values of ω of the order of 10^2 to 10^4 sec⁻¹. However, Fuks⁶ criticizes these computations of the coefficient of coagulation made by Teverovskii. S. M. Greenfield⁷ has calculated the effect of coagulation of particles in a mist or cloud due to turbulence, using values of ω ranging from 4 to 30 sec⁻¹. E. G. Richardson⁽⁸⁾ discusses the difficulties in theoretical calculations of turbulent agglomeration.

It is clear that experiments are needed to check the adequacy of Equation 11 for expressing the effect of turbulence, and also that the parameter will have to be related to observable parameters (e. g., Reynolds number) in turbulent flow.

For the purposes of the present discussion, Equation 11 is accepted as correct, and it is represented graphically in Figure 5. The coefficient K_S increases as the third power of the particle size. The differing lines were drawn for assumed values of ranging from 1 sec⁻¹ to 10^4 sec⁻¹. The solid lines are the sum ($K_S + K_D$). Thus, the total coagulation coefficient will have minimum values ranging from 3×10^{-10} to $7 \times 10^{-10} \frac{\text{cm}^3}{\text{sec-part}}$, at values of particle diameters ranging from 2μ down to 0.2μ , depending on the assumed value of the velocity gradient, ω . The rapid increase of the combined coagulation coefficient with either increasing or diminishing values of particle diameter suggests that whatever the original distribution of particle sizes may be, there will be a tendency to "compress" the distribution about some central value, determined by the degree of turbulence present.

c. Mean Life-Time of Particles

The effect of coagulation on the distribution of sizes may be approached from another point of view, namely, the mean life of a given sized particle. For this purpose, let us consider Equation 1 with $K_T = K_D + K_S$.

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$$-\frac{dn}{dt} = K_T n^2 \quad \dots (12)$$

Equation 12 has been tested for uniform-sized particles, and is known to hold at least during initial stages(9). If it is written as

$$-\frac{1}{n} \frac{dn}{dt} = K_T \quad , \quad \dots (13)$$

one may define a mean lifetime,

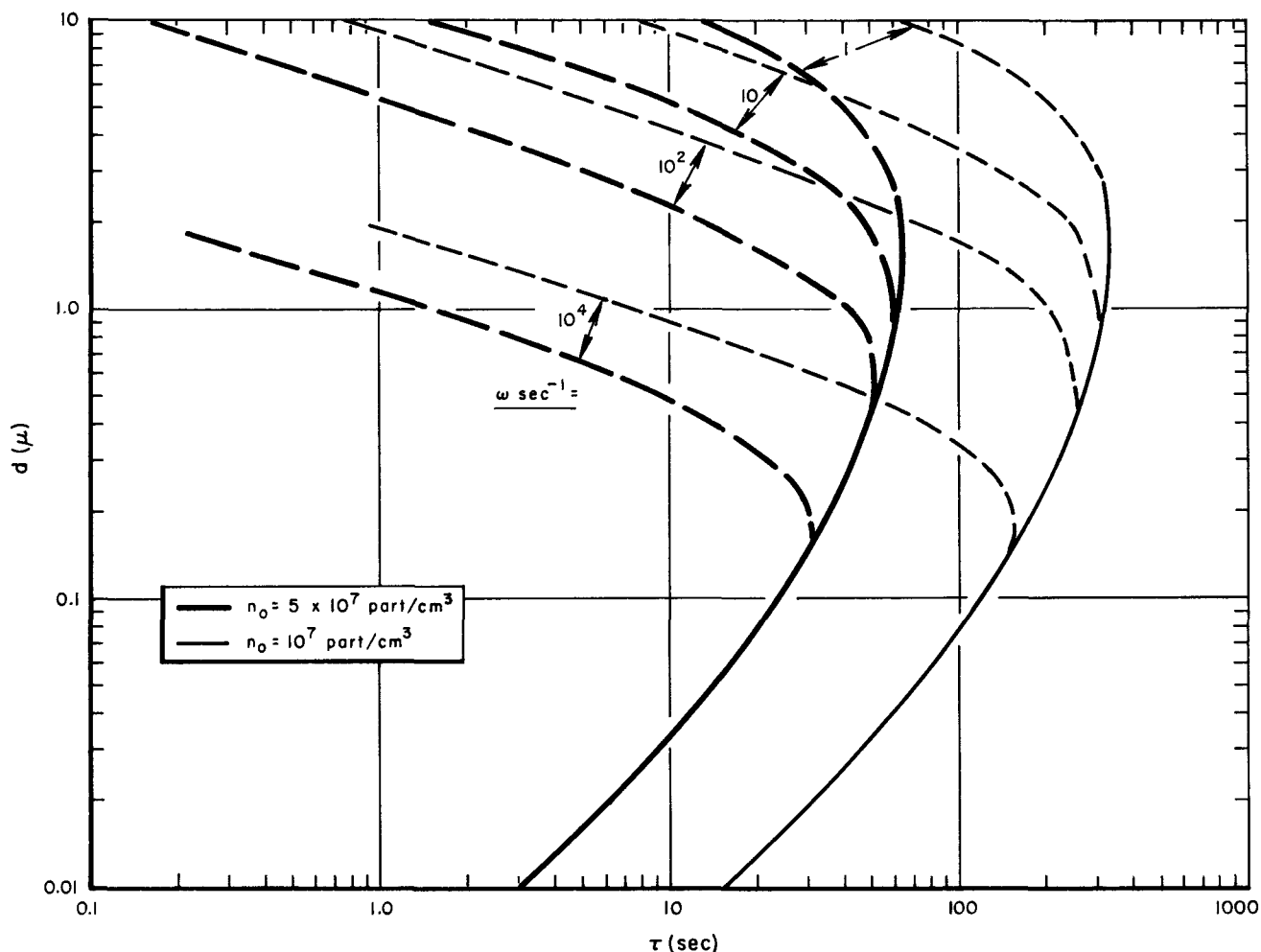
$$\tau = \frac{1}{K_T n_0} \quad , \quad \dots (14)$$

where n_0 is the initial concentration. τ will give some idea of the stability of a given size of particle in terms of the magnitude of its expected lifetime. Figure 6 is a representation of values of τ as defined by Equation 14, based on the K_T values of Figure 5. Two sets of curves are given for two assumed values of n_0 : 1×10^7 and 5×10^7 $\frac{\text{part}}{\text{cm}^3}$. These values for n_0 are those observed in ex-

perimental sodium fires for LF-2 in a region just above the burning sodium surface. Figure 6 indicates that for such concentrations, particles formed in the burning process with diameters less than a few hundredths of a micron should not be observed; i. e., they have a lifetime of the order of a few seconds only. These particles will have a continued airborne existence as parts of larger particles. On the other hand, particles which are larger than say, 10μ will also have a short life-span, the value depending on the degree of turbulence; but in this instance, agglomeration means probable removal of a particle from the airborne state due to settling. Thus, it is likely that in a gallery with a high density of airborne particles, the consequence of agglomeration would be two-fold: (a) a narrowing of the size distribution and (b) an accelerated fallout of larger particles. These processes would occur with time constants whose values would be less than 100 sec. The effect of the fallout of large particles on the burning fire itself may be quite important as a quenching mechanism.

d. Experiment Design

As the above discussions have indicated, the agglomeration and distribution of particles are strongly related to the parameter ω , the velocity gradient. The experimental program must therefore be oriented toward ways of measuring or controlling the velocity gradient. The degree of turbulence is frequently related to Reynolds number, and this has been used previously to correlate deposition rates in tubes. Since correlations exist which relate velocity gradient and Reynolds number in turbulent flow, fundamental experiments with measurement of agglomeration rates as a function of concentration and degree of turbulence (Reynolds number) will be valuable in particle behavior modeling.



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Figure 6. Mean Life of Particles which Agglomerate τ (as a function of particle diameter $d(\mu)$ for various values of the turbulence parameter $\omega(\text{sec}^{-1})$ for two values of particle density N_0)

2. Gas and Energy Transport Models

a. Sodium Fire Thermal Analysis

The detailed mechanisms of heat transfer between regions of sodium fire LF-1 were reviewed¹⁰ to evaluate means of predicting temperatures and energy release for developing state-of-the-art accident models involving sodium fires. From the observations of a temperature maximum slightly above the liquid surface, it has been assumed that the Na-O₂ reaction takes place in the gas phase about 0.02 in. above the surface. Vaporization and diffusion of sodium from the surface and diffusion of oxygen from the cloud of air and smoke to the flame front determine the burning rate.

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The heat transferred from the flame to the sodium is about 2/3 conduction and 1/3 radiation, with only a small contribution from convection. Most of this heat is required to vaporize more sodium (about 32% of combustion energy is required for vaporization). Most of the heat transfer from the flame to the cloud is by radiation (particle-to-particle), with convection and conduction together accounting for about 17% of the heat.

The cloud loses most of its heat to the air flowing through the chamber, with some loss (~18%) to the hood enclosing the burn chamber. Future fires will have reduced heat losses through the sodium and the hood because of more effective insulation in these regions.

The method of analysis used for LF-1, particularly with regard to flame conditions and burning rate, will be used to check the measured conditions in the low oxygen fires of LF-2. Future analytical effort will attempt to describe the effect of a large sodium fire in an open cell on the building temperature and pressure. The ability of air to flow or diffuse into the hot, open cell containing sodium is expected to establish the likelihood that a fire will burn after initial depletion of oxygen by the sodium.

b. Experimental Design-Release and Transport

The fundamental experiments for studying mass and energy release parameters will utilize the modified large fires apparatus and the bench-scale fires apparatus. The experiments will examine the dependence of matter and energy release rates from a sodium surface on surface temperature, gas temperature, oxygen concentration and geometry. The small apparatus is sufficiently smaller than the large apparatus to be of value in determining the existence of geometric effects. In particular, it is felt that matter and energy release rates per unit surface area become constant at a sufficiently large pot diameter. The particular diameter or size would vary with temperatures and oxygen concentration, however. Results in the large apparatus will be compared with results in the small apparatus. If the results are different, indicating that the so-called constant or "critical" size is greater than the small diameter, several intermediate diameters will be tried, probably in the large apparatus.

In the accident situation, the gas may become quite hot. Gas temperature can influence turbulent and diffusive transport coefficients and reaction kinetics. For this reason, gas temperature has been added as an important experimental variable. Consideration will be given to the experimental difficulties encountered in the use of forced fresh gas convection. At high forced flows the natural convection patterns may be altered, thus changing the release characteristics. At low flow such a large fraction of oxygen may be consumed that the inlet concentration bears no relationship to the concentration found in an enclosure in the accident situation. In any event, the need is clear for measurements of oxygen concentration in the gas space above the fire. A significant portion of the inlet gas may be by-passed right out the chamber while "old" gas is being recirculated several times. At least the concentration should be known as it enters the flame region and related to the concentration measured some distance above the pot, as well as the inlet and outlet concentrations. If substantive oxygen concentration

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gradients exist over large distances above the burn pot, then more careful attention will have to be paid to measuring this in many locations in all types of fires. The same comments apply to temperatures and temperatures gradients in the gas space.

The transport processes involved in moving quantities of matter and energy through large distances are complex, sensitive to geometry changes and not amenable to direct analytical solution. Thus, in order to characterize completely the sequence of events in the accident case, it is necessary to utilize large gas volumes and substantial heights to accurately simulate these events. For this reason, the modeling type of experiments have been conceived utilizing large tank volumes in the new test installation.

IV. EVALUATION OF EFFORT

The extensive planning and evaluation effort which was conducted to define a 5-yr program for characterizing sodium fires has provided excellent guidance for the future course of the project. The various phases of this program have been planned so that the experimental information and accident consequence prediction techniques will be provided in a form which will be useful for safeguards analyses and evaluation, and engineering safeguards design.

The planning and evaluation have emphasized the need for adding gas temperature as a primary independent variable to the experimental program in order to fully understand the fundamental matter and energy release phenomena. Careful attention must be paid to O₂ concentration and temperatures throughout the entire gas space in order to subtract the effects of forced convection when extrapolating results to modeling or prototype cases. Experimental activities formerly conducted outdoors will now not be weather-dependent nor will heat balances be comprised by solar radiation. The new design allows faster approach to equilibrium and more accurate heat balances.

Use of the bench-scale fires apparatus in conjunction with the large apparatus will allow rapid study of geometrical effects and indicate a range of respective results for comparison with the large apparatus.

A multiplicity of particle sizing instruments which collect the sample by several different techniques are being fabricated, purchased, and calibrated. This will lead to an overall better estimation of the mean size of any distribution.

The study of the agglomeration coefficient in the presence of a turbulent environment has shown that the coefficient is turbulence-dependent for particles greater than 0.2 microns, that it is nearly constant with a minimum in the region of 0.2 to 2 microns depending on the velocity gradient, and highly diffusion dependent for particles less than 0.2 microns. The implications of the study are that the observed distribution should lie within the size range in which the minimum for the agglomeration constant occurs. For the LF-2 experiments, 80% of the particles were distributed between 0.2 μ and 1.0 μ , and 50% were larger or smaller than 0.36 μ (all by number). This could imply that velocity gradients ω between 100 and 10,000 per second existed. The mean life of the agglomerate for this size and for high concentration (5 x 10⁷ particles/cm³) would be approximately 60 sec (see Figure 4), which is consistent with the flow rates over the fire.

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V. NEXT REPORT PERIOD ACTIVITIES

a. Assuming approval of the Construction Authorization Request for the Sodium Fires Test Installation early in the report period, the design of the installation will be completed; Phase I construction bids will be requested, Phase I construction will be initiated, and Phase II design will commence.

b. Parametric small fires experiments will be conducted with the bench-scale fires apparatus.

c. The initial particle size apparatus will be constructed and calibrated.

d. Testing and evaluation of particle size measurement and sampling techniques will continue.

e. An experiment will be designed for determining agglomeration constants for particles in turbulent flow.

f. A state-of-the-art analytical model for a sodium fire in an opened cell will be described. Analytical studies of particle behavior will continue.

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A Division of North American Aviation, Inc.

Program: Physical Research Program

Project: High Temperature Chemistry

Project Engineer: S. J. Yosim

Reporting Period: July-September 1965

AEC Category: 05-02-02-01

General Order: 9305

Subaccount: 5302

I. PROJECT OBJECTIVES

The project is divided into two general areas, namely, the study of fused salts and the study of metal-metal salt solutions. The objective of the study of fused salts is to achieve an understanding of ionic melts (halides, oxides, oxy-salts, and glass systems) by (a) establishing the nature of the species existing in ionic melts, (b) determining the physico-chemical interactions between species, and (c) formulating systematic relationships between the physico-chemical properties of fused salt systems and the molecular parameters of the species.

The objectives of the metal-metal salt study are to determine phase diagrams for, and the nature of interactions between metals and their salts at high temperatures, to determine the species in these solutions; and, finally, to predict the solubilities in metal-salt systems.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

A. PUBLICATIONS

The following papers were published.

1. "The Excess Entropy of Mixing of Liquids," by S. J. Yosim, J. Chem. Phys., 43, 286 (1965)
2. "Electrochemical Studies of NO^+ and NO_2^+ in Concentrated Sulfuric Acid," by L. E. Topol, R. A. Osteryoung, and J. H. Christie, J. Electrochem. Soc., 112, 861 (1965)
3. "The Electrical Conductivities of Molten Bi-BiCl₃ and Bi-BiBr₃ Solutions," by L. F. Grantham, J. Chem. Phys., 43, 1415 (1965)

The following papers were submitted for publication.

1. "The Electrical Conductivities of Molten Cadmium-Cadmium Halide Solutions," by L. F. Grantham, J. Chem. Phys.
2. "Corresponding States Melting Curve for the Alkali Halides at High Pressure," by B. B. Owens, J. Chem. Phys.

B. TALKS

1. The paper "Calculation of Thermodynamic Properties of Liquids from a Rigid Sphere Equation of State," by S. J. Yosim, was presented at the XXth International Congress of Pure and Applied Chemistry in Moscow, U.S.S.R.
2. The paper "Phase Equilibria in Solid Solutions of the Alkali Halides at High Pressure," by A. J. Darnell, was presented at the American Ceramic Society in Pittsburgh, Pa., Sept. 1965.

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The following talks were presented at the American Chemical Society Meeting in Atlantic City, September 1965.

3. "The Electrical Conductivities of Metal-Metal Salt Solutions," by L. F. Grantham.
4. "The Soret Effect in the Molten Bismuth-Bismuth Tri-iodide Redox Thermocell," by J. D. Kellner.
5. "The Compression of Solid Bismuth Trihalides to 40,000 Bars and a New Solid-Solid Phase Transition in BiI_3 ," by A. J. Darnell and B. B. Owens.

C. TECHNICAL PROGRESS

The study of the Soret effect in Bi-BiI_3 solutions has been completed. A new high pressure metastable phase of BiI_3 has been found.

III. PROGRESS DURING REPORT PERIOD

A. THE SORET EFFECT IN Bi-BiI_3 SOLUTIONS

The steady state thermoelectric powers of BiI_3 solutions containing 1 and 90 mole % bismuth were measured during the report period. In addition, thermocells containing 35, 50, and 80% Bi were repeated and more precise data recorded. The thermoelectric potentials range from $-4600 \mu\text{v}/\text{deg}$ at 1% metal to $18 \mu\text{v}/\text{deg}$ at 90% metal. This completes the experimental portion of this study.

B. STATISTICAL MECHANICS OF FUSED SALTS

Previously in our work on fused salts, the melt was compared to a fluid of identical rigid spheres, the diameters of which equalled the sum of the anion and cation radii. We have now re-derived new expressions for thermodynamic properties of fused salts by using Lebowitz' equation of state for a mixture of rigid spheres. Thus we are now taking into account the anion-cation ratio. Preliminary calculations show that the results do depend on this ratio.

C. THE SOLUBILITY OF INERT GASES IN MOLTEN SALTS AT HIGH PRESSURES

Several laboratories have recently been investigating the effect of pressure upon the properties of molten salts. In our investigation of melting properties and compressibilities the salts were pressurized by a mechanical piston. However, in most of the other laboratories the melts were compressed by inert gas pressure. In the latter studies one must be concerned with the degree of solution of the gas in the fused salt. Extrapolation of one value reported for the solubility of Ar and He in molten NaNO_3 indicates that at 1000 atm as much as 10 mole % inert gas may dissolve into the melt.

We propose to systematically investigate this phenomenon and to interpret the results in terms of the structural models for molten salts. The equipment for this study has been assembled; the procedure that will be followed is to measure the freezing point of a molten salt saturated with gas. From the freezing point depression we will calculate the concentration of solute species. The initial measurements will be carried out on KNO_3 under a 200 to 500 atm pressure of Ar. The study will then be extended to other systems at pressures up to 2000 atm.

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D. HIGH PRESSURE FORM OF BiI_3

The solid phase of BiI_3 which is stable above ~24 kilobars and 100°C has been recovered in a metastable form at atmospheric pressure. This metastable form, $\text{BiI}_3(\text{II})$, reverts to the stable form $\text{BiI}_3(\text{I})$ when heated to -14°C . The volume increases by $8 \pm 2\%$ during this $(\text{II}) \rightarrow (\text{I})$ transition. This corresponds to about 4 kcal/mole for the $\text{BiI}_3(\text{I}) \rightarrow \text{BiI}_3(\text{II})$ transition.

E. PHASE EQUILIBRIA IN SOLID SOLUTIONS OF ALKALI HALIDES

Studies on the solid-solid phase transitions in solid solutions of the alkali halides were initiated. The systems KCl-KBr , KCl-RbCl and KCl-RbBr have been studied. The above salts which form continuous solutions in the NaCl type structure apparently also form continuous solid solutions in the CsCl type structure. The pressure composition phase diagrams (at 100°C) indicate that "near ideal" solid solutions are formed in both solid phases. The regular manner in which the transition pressure varies in these substitution type solid solutions indicates that the cation plays the predominant role in determining the pressure at which the NaCl-CsCl transition occurs.

IV. EVALUATION OF EFFORT TO DATE

The statistical mechanical approach for developing liquid theory so that the physico-chemical properties of pure liquids and solutions can be calculated from equations with no adjustable parameters continues to be very fruitful. The emphasis in the experimental approach continues on high pressure and transport experiments.

V. NEXT REPORT PERIOD ACTIVITIES

The study of the Soret effect in Bi-BiBr_3 solutions will be initiated. Attempts to measure the electrical conductivities of metal-salt solutions above their critical temperatures will be resumed. Measurements of the electrical conductivity of the Hg-HgX_2 system will also be resumed. The new calculations on the thermodynamic properties of fused salts from the Lebowitz equation of state will be completed. The high-pressure study of the solubility of gases in fused salts will be under way. The high-pressure studies of transitions in solid solutions of alkali halides will be completed.

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Program: Physical Research Program	
Project: Radiation Chemistry	
Project Engineer: R. B. Ingalls	
Reporting Period: July-September 1965	AEC Category: 05-02-02-01
General Order: 9305	Subaccount: 5305

Principal Scientists: R. B. Ingalls, R. A. Holroyd, G. T. Tiedeman

I. PROJECT OBJECTIVES

The objectives of this project are to delineate the fundamental physicochemical processes occurring in the radiolysis of organic systems and to study these processes in separate experiments designed to increase our understanding of their nature and their role in radiolysis.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

A. PAPERS AND PRESENTATIONS

The following papers have been published.

1. "Radical Intermediates in the Mercury (ep) Photosensitized Decomposition of Cyclopropane" by R. A. Holroyd and G. W. Klein, *J. Phys. Chem.*, 69, 2129 (1965)
2. "ESR Study of Excited Rare-Gas Initiated Reactions of Polystyrene and Polymethylmethacrylate" by R. B. Ingalls and W. A. Young, *J. Chem. Phys.*, 43, 1759 (1965)
3. "Determination of Radiation Lifetime of Vibrationally Excited sec-Butyl Radicals using Methylethylamine as a Model" by G. T. Tiedeman, R. B. Ingalls, and Jack S. Margolis, *J. Chem. Phys.*, 42, 2627 (1965)

The following papers have been accepted for publication.

1. "Radical Yields in the Radiolysis of Hydrocarbons II, Neopentane, Isopentane, and Cyclopentane" by R. A. Holroyd and G. W. Klein, *J. Am. Chem. Soc.* (1965)
2. "Radical Yields in the Radiolysis of Cyclohexene with Different Kinds of Radiation" by W. G. Burns, R. A. Holroyd, and G. W. Klein, *J. Phys. Chem.* (1965)
3. "Radical Yields in the Radiolysis of Hydrocarbons" by R. A. Holroyd in "Advances in Chemical Research" published by Springer Publishing Co., Germany, early 1966
4. "On the Thermal Spike Model of Low L. E. T. Radiolysis" by R. B. Ingalls, *J. Chem. Phys.*
5. "ESR Spectra of Six Selectively Deuterated Biphenyl Negative Ions," by T. W. Lapp, J. G. Burr, and R. B. Ingalls, *J. Chem. Phys.*

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The following paper has been submitted for publication.

1. "A New ESR Technique for Investigating Reactions of Free Radicals with Compounds of Biological Interest," by R. B. Ingalls, D. E., Holmes, and L. S. Myers, Jr., Nature

The following papers were presented.

1. R. A. Holroyd served as chairman of the session on "Reaction of Hydrogen Atoms in Organic Systems" at the Gordon Conference on August 5.

2. "ESR Studies on the Course of Addition of Deuterium Atoms to Polystyrene Fluff" by G. T. Tiedeman and R. B. Ingalls was presented by Dr. Tiedeman at the ACS Atlantic City meeting, September (1965)

3. "Thermal Spike Model of Low LET Radiolysis," by R. B. Ingalls, was presented at the XXth International Congress of Pure and Applied Chemistry in Moscow, U.S.S.R.

III. PROGRESS DURING REPORT PERIOD

A. RADICAL YIELDS IN THE RADIOLYSIS OF CYCLOHEXENE WITH DIFFERENT KINDS OF RADIATION

Dilute solutions (1-20 mM) of $^{14}\text{CH}_3\text{I}$ in cyclohexene were irradiated, and yields of radioactive methyl-3-cyclohexene, methylcyclohexene, methyl-1-cyclohexene, ethane, and methane were determined as functions of $^{14}\text{CH}_3\text{I}$ concentration, energy input, dose-rate, and L.E.T. of the radiation. The results are interpreted on the basis that thermal $^{14}\text{CH}_3$ radicals are formed and react with the surrounding molecules or radicals. The effect of dose-rate indicated that as the dose-rate is decreased, all radicals react in greater proportion with the cyclohexene rather than in inter-radical reactions. The effects of concentration and of L.E.T. at high dose-rates are consistent with the concept that methyl radicals are generated by electron capture. At high dose-rates of low L.E.T. radiation only inter-radical reactions occur, and the G-values of 3-cyclohexenyl, 1-cyclohexenyl and cyclohexyl radicals determined from the above radioactive product yields agreed with those deduced from the C_{12} product yields from the pure liquid. With increasing L.E.T. the apparent radical yields from the activity measurements fall more rapidly than those deduced from the C_{12} product yields. This may be due to the inhomogeneity of track reactions, and can be accounted for if the CH_3 radicals, formed possibly by electron capture, have a larger diffusion constant and a larger initial track radius than the cyclohexenyl and cyclohexyl radicals. Diffusion theory calculations indicate that the ratio of the track radii is ~ 5 .

B. ALKYL RADICAL YIELDS FROM BRANCHED HYDROCARBONS

Continued investigations of the yields of radicals from various hydrocarbons have led to a general semi-empirical equation for calculating all of the fragment alkyl radical yields from any alkane. It was found that the probability that a particular bond will break increases with the number of adjacent carbon-carbon bonds. Further a theoretical basis for this approach has been found in that it can be shown that the strength of a carbon-carbon bond in the molecular ion or molecule is reduced with the addition of adjacent carbon-carbon bonds. Calculated values of radical yields are compared with experimental values obtained in this study in the following table.

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RADICAL YIELDS FROM HYDROCARBONS

Hydrocarbon	Radical	G(radicals/100 ev)	
		Calculated	Experimental
n-Pentane	CH ₃	0.12	0.14
n-Pentane	C ₂ H ₅	0.44	0.48
n-Pentane	C ₃ H ₇	0.44	0.38
2,2-Dimethylbutane	CH ₃	1.12	0.99
2,2-Dimethylbutane	C ₂ H ₅	1.20	1.14
2-Methylpentane	C ₂ H ₅	0.14	0.29
3-Methylpentane	C ₂ H ₅	0.80	0.82
2,4-Dimethylpentane	CH ₃	0.39	0.28
2,4-Dimethylpentane	i-C ₃ H ₇	0.56	0.53
2,2,4-Trimethylpentane	CH ₃	0.75	0.69
2,2,4-Trimethylpentane	i-C ₃ H ₇	0.20	0.12
2,2,4-Trimethylpentane	i-C ₄ H ₉	0.61	0.74

C. EXTENSION OF FLUFF TECHNIQUE TO LOW MOLECULAR WEIGHT MATERIALS

The investigation of free radical reactions by electron spin resonance techniques using high surface substrates has been extended to materials of moderate molecular weight. In previous reports we described the use of high polymers in a finely divided fluff state to stabilize the reaction products of hydrogen atoms, other gaseous free radicals and excited states. However, a limitation of the technique was that only high molecular weight materials can be prepared as fluffs. During this report period, a new and more general method of radical stabilization which permits the investigation of low molecular weight materials as well as high polymers has been developed.

The essential feature of this method is the use of quartz wool as a supporting medium. A solution of the material to be investigated is added to a tube containing quartz wool, quickly frozen, and freeze-dried. During the drying the solute is trapped between the quartz strands to produce a highly dispersed sample. Such a sample has several square meters of surface area per gram, so that even though the penetration of the reacting free radicals is probably much less than 10⁻⁵ cm, sufficient filling factor in the ESR cavity is obtained. Thus, the radicals produced in the substrate are easily observed. Obviously, this technique is restricted to materials with low vapor pressure and so far only solids have been investigated.

D. KINETICS OF RADICAL FORMATION IN POLYMER FLUFFS

The kinetics of radical formation in fluff systems is not simple to analyze and is currently under investigation. It appears that a steady state radical concentration is set up on the surface of the substrate in a very short time, and that in

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order for additional radicals to accumulate, the initiating species must diffuse through this steady state region and react with the relatively inaccessible material further from the surface. Such a model has been described mathematically and it predicts a sharp break in the curve of total substrate radical concentration with time, in agreement with experimental results. Details of this investigation will be reported in the next progress report.

IV. EVALUATION OF EFFORT TO DATE

The effects of dose rate and L. E. T. on inter-radical reactions and radical yields have been investigated using the $^{14}\text{CH}_3\text{I}$ radical sampling technique. These results throw light on the inhomogeneous nature of tracks and reactions therein which are more important at high L. E. T. The investigation of the yields of radicals from various hydrocarbons has led to the development of a semi-empirical equation for calculating all of the fragment alkyl radical yields from any alkane. These developments contribute to our fundamental understanding of the interaction of ionizing radiation with organic materials.

V. NEXT REPORT PERIOD ACTIVITIES

A general kinetic model for the reaction of atoms with polymer fluffs will be developed. The properties of this model will be compared with experimental results which have already been obtained.

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Program:	Physical Research Program		
Project:	Electronic Structure of Metals and Alloys		
Project Engineer:	R. G. Breckenridge		
Reporting Period:	July-September 1965	AEC Category:	05-03-02-01
General Order:	9305	Subaccount:	5401

Principal Scientists: R. G. Breckenridge, L. J. Barnes, H. J. Fink,
W. J. Tomasch

I. PROJECT OBJECTIVES

This effort is devoted to the acquisition of knowledge regarding the electronic structure of metals and alloys (configuration of electronic energy states in momentum space) and the role of this structure in determining electrical, thermal, magnetic, vibrational, and alloying characteristics. Detailed information on the shape of the Fermi surface is provided by de Haas-van Alphen studies in magnetic fields up to 200 kilogauss, and the density of electronic states at the Fermi level is deduced from low-temperature specific heat measurements. The latter also yield information on lattice vibrational modes and on the interactions involved in superconductivity and magnet ordering. Further characterization of magnetic interactions is accomplished by means of conventional magnetic susceptibility techniques. Considerable effort is devoted to the exploration of high-field superconductivity with emphasis on thermodynamic and transport characteristics, as well as on the electron energy spectrum as deduced from thin-film electron-tunneling measurements. These experimental investigations are correlated with current theory, and attempts are made to characterize quantitatively the relationship between superconductivity and the normal-state electronic structure.

II. PROGRESS DURING REPORT PERIOD

A. PUBLICATIONS

1. "The Exact Solutions of the Ginzburg-Landau Equations for the Superconducting Surface Sheath," by H. J. Fink and R. D. Kessinger, American Physical Society Meeting, Honolulu (September 1965)
2. "Influence of a Magnetic Field on Tunneling from a Type-II Alloy Superconductor," by W. J. Tomasch, Phys. Rev. 139, 3A (1965)
3. "Critical State of Superconducting Surface Sheath," by H. J. Fink and L. J. Barnes, submitted to Phys. Rev. Letters
4. "Exact Solutions of the Superconducting Surface Sheath," by H. J. Fink and R. D. Kessinger, submitted to Phys. Rev.
5. "Internal Currents and Magnetic Fields Close to the Surface for a Type II Superconductor for Applied Magnetic Fields Near the Upper Critical Field H_{C2} ," by H. J. Fink, Phys. Rev. Letters 14, 21 (1965)

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6. "Geometrical Resonance in the Tunneling Characteristics of Superconducting Pb," by W. J. Tomasch, to be published in Phys. Rev. Letters

7. "Intrinsic Hysteresis in Type II Superconductors," by H. J. Fink, to be published in Phys. Letters

B. THEORETICAL STUDIES ON SUPERCONDUCTIVITY (H. J. Fink and L. J. Barnes)

1. Exact Solutions of the Superconducting Surface Sheath

The exact solutions of the superconducting surface sheath of a semi-infinite superconducting half-space for various values of the Ginzburg-Landau parameter κ were obtained as a function of the applied magnetic field between the critical fields H_C and H_{C3} (for $0.417 < \kappa < 0.707$; Type I), and between H_{C2} and H_{C3} (for $0.707 < \kappa < \infty$; Type II). For Type II superconductors, the solutions were also extended into the mixed state well below H_{C2} . The order parameter, the supercurrents, the internal magnetic field, the vector potential, the thickness of the surface sheath and the magnetization per unit volume for a "thick" film were determined. These quantities vary strongly with applied magnetic field for $\kappa < 3$. In order to obtain these solutions a boundary condition was derived which relates the absolute value of the order parameter at the surface to the phase of the order parameter and the value of the vector potential at the surface. Two supercurrents of equal magnitude, but opposite direction, flow near the surface for magnetic fields above H_C (Type I) or H_{C2} (Type II) respectively. Below H_{C2} these currents are slightly different due to the bulk solution and give rise to a small net surface current.

2. Critical State of the Superconducting Surface Sheath

When a long superconducting cylinder is placed into an axial magnetic field whose value is between the upper critical field H_{C2} and the surface nucleation field H_{C3} , then the surface is in the superconducting state and the core of the cylinder is in the normal state provided the thickness of the surface sheath Δ is small compared to the radius of the cylinder R . Although the metal is singly connected, the superconducting surface is multiply connected. By analogy with a superconducting ring one could imagine that a persistent current could be induced in the surface sheath provided the surface sheath is able to carry a total current. Such a current could easily change the magnetic properties of a superconducting cylinder.

We have shown that such a current can indeed be induced in the surface sheath and that its magnetic moment can be either diamagnetic or paramagnetic which depends on whether the magnetic field is increased or decreased respectively. It was found that these currents are size dependent and have an appreciable effect on the magnetization for Ginzburg-Landau κ -values of order unity and for applied magnetic fields close to H_{C2} . It is also shown for a cylinder that the induced magnetization per unit volume is always considerably larger than the inherent diamagnetic magnetization per unit volume of the surface sheath (in its lowest energy state) although the induced surface current is always considerably smaller than the inherent surface currents provided R is very large compared to the penetration depth λ .

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C. ELECTRON TUNNELING STUDIES (W. J. Tomasch)

1. Resonance Effects in Superconducting Tunnel Diodes

A new type of structure in the current-voltage characteristics of Al/AlO_x/Pb superconducting tunnel diodes was recently discovered at this laboratory. This phenomenon has been studied as a function of temperature, alloying, annealing, junction geometry, barrier (AlO_x) properties, Al film thickness and Pb film thickness. Although temperature and alloying affects the strength and sharpness of the periodic structure, only Pb film thickness produces a systematic change in the voltage period. (The other factors investigated apparently exert little or no influence so long as both Al and Pb films remain superconducting.) To be more specific, the voltage period has been found to be proportional to the reciprocal of the Pb film thickness. This result strongly suggests that the Pb films exhibit geometrical resonances (standing wave modes) which serve as monochromatic reservoirs to which tunneling electrons may lose energy in quantum assisted transitions. The propagation velocity associated with the phenomenon has been determined and has a value comparable to the Fermi-velocity, indicating that the standing wave is electronic in nature. In view of the large Pb film thicknesses involved (2-26 μ), collective electronic excitations appear to be involved. The present data probably constitute the most direct and detailed evidence for the existence of such modes. Very fundamental theoretical considerations require the existence of such modes, and the lack of reasonably direct experimental confirmation has been a source of sustained concern.

Preliminary results indicate that the effect is also present in thick In films. This demonstrates that the effect is not restricted to strong coupling superconductors (Pb and Hg) but probably occurs quite generally. Effort is underway to determine the velocity associated with the effect in In, and to discover whether or not the phenomenon can be observed in still other soft superconductors.

III. EVALUATION OF EFFORT TO DATE

The concentrated activity in the field of superconductivity continues to produce a variety of interesting theoretical and experimental results. In spite of the rather formidable mathematical difficulties encountered in the application of the Ginzburg-Landau-Abrikosov-Gor'kov theory to the physical situation now being investigated the theory continues to yield quantitative descriptions of observed phenomena as well as an improved understanding of the situation. The two papers discussing aspects of the sheath phase are excellent examples of this type of activity. The tunneling studies have apparently discovered a completely novel effect in this system and are continuing the exploration of this new phenomenon. The observations have some similarity to oscillatory effects recently reported from Bell Labs and elsewhere but the detailed studies reported in this quarter show that they are not the same. While at present the observations are only of theoretical interest their practical implications are being followed up.

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Program: Physical Research Program

Project: Radiation Damage in Crystalline Solids

Project Engineer: S. C. Carniglia

Reporting Period: July-September 1965

AEC Category: 05-03-03-01

General Order: 9305

Subaccount: 5404

Principal Scientists: A. Anderman, W. Bauer, D. Keefer, A. Sosin,
K. Thommen, and K. Garr

I. PROJECT OBJECTIVES

The materials requirements of modern technology demand an increased understanding of the effects of lattice defects on the physical properties of crystalline solids, and on their behavior when subjected to various kinds of radiation. Studies of radiation damage in solids not only directly yield information on the effects of radiation, but also provide a means of investigating defects in solids. It is the objective of this project to study the production of defects in crystalline solids by radiation, and to attain an understanding of their structure, their interactions with each other and with impurities and dislocations, their effect on the physical properties of solids, and their recovery kinetics. An understanding of these points should assist in the understanding of lattice defects produced in solids by other means and help make possible the development of materials of superior properties as the ability to better control their defect structure is developed. The materials chosen for study are metals, metal alloys, and semiconductors.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1966

A. PUBLICATIONS

W. Bauer, "Influence of Impurities on the Recovery of Electron Irradiated Gold," accepted for publication in Physics Letters.

A. Sosin, "Interaction of Interstitials with Solute Additions in Face-Centered Cubic Metals," invited paper presented at the American and Japanese Physical Society Meeting in Hawaii. Bull. Am. Phys. Soc. 10, 697 (1965). An expanded paper on this subject is being prepared for a book edited by R. R. Hasiguti on the lattice defects section of the meeting.

W. Bauer, "Damage Production in Pt near 7°K by Electron Irradiation," Bull. Am. Phys. Soc. 10, 711 (1965). An expanded paper is being prepared for the book on the lattice defects section of the Physical Society Meeting.

A. Sosin and D. W. Keefer, "Trapping of Point Defects by Dislocations," AIME Proceedings of Radiation Effects Symposium in Asheville, North Carolina (September 8, 1965)

W. Bauer and A. Sosin, "Recent Threshold Energy and Displacement Cross Section Measurements in Metals," AIME Proceedings of Radiation Effects Symposium in Asheville, North Carolina (September 8, 1965)

A. Sosin and D. W. Keefer, "Trapping of Point Defects by Dislocations," invited paper presented by A. Sosin at the Gordon Conference on Physical Metallurgy in Meridan, N. H., July 19-23, at a seminar at RPI, July 14, and at a seminar at the University of North Carolina (July 2, 1965)

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D. W. Keefer and A. Sosin, "Modulus and Internal Friction Effects in Irradiated Metals," invited paper presented by D. Keefer at the Conference on Internal Friction in Solids sponsored by the Institute of Physics and the Physical Society. Conference held in Manchester, England, September 7-11, 1965. Invited paper also presented at a seminar at SCK-CEN, Mol, Belgium (September 15, 1965)

III. PROGRESS DURING REPORT PERIOD

A. SEMICONDUCTOR IRRADIATIONS (K. Thommen)

Efforts towards a systematic study of radiation defects in GaSb have been started (April 1965). Experimental equipment has been built and assembled for irradiation and electrical measurements on samples at temperatures between 4°K and 460°K.

Resistivity and Hall coefficient have been measured for an undoped GaSb-sample between 20°K and room temperature. A simple impurity level scheme has been constructed which gives good agreement with the experimentally observed carrier-concentration between 25°K and room temperature. Below 25°K an anomalous behavior of the carrier concentration is observed, in agreement with the findings of other authors, which is most probably due to impurity band conduction.

A p-type GaSb single crystal has been irradiated with 1.0-Mev electrons at 77°K. Measurements of resistance and Hall voltage revealed a decrease of mobility and an increase of carrier concentration as a result of the irradiation. The fractional change in mobility brought about by 1.75×10^{16} e/cm² was -30% whereas the corresponding change in carrier-concentration was only +4%. It was observed that the reciprocal Hall-mobility varied linearly with the irradiation dose.

Isochronal pulse annealing after the irradiation revealed distinct annealing stages centered at 170, 210, and 375°K. The fractional recovery in these 3 stages was +0.33, +0.13, and +0.46 for the resistivity and +1.85 (change from increased carrier concentration to decreased carrier concentration with respect to the pre-irradiation value), -0.2 (further decrease of carrier concentration) and +1.2 for the carrier concentrations. These numbers should be regarded as preliminary results. It appeared that a small amount of damage remained in the sample even after annealing at 460°K, which was the highest annealing temperature in this experiment.

B. RECOVERY OF ELECTRON IRRADIATED ALUMINUM (K. Garr and A. Sosin)

The following abstracts have been submitted to the American Physical Society for inclusion in the Los Angeles meeting to be held on December 20 to 22, 1965.

The Stage I recovery of high purity aluminum (99.9999%) has been investigated following electron irradiation near 4.2°K. A movable slit arrangement was employed so that two samples could be irradiated separately, to different doses or at different energies, and then annealed simultaneously, thus ensuring the same thermal history during the recovery process. Isochronal annealing pulses were taken every 1/2° between 10 and 50°K with the temperature being controlled to

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$\pm 0.03^\circ\text{K}$. A multiplicity of substages in the Stage I region has been observed, in agreement with Dawson, et al. The ratio of slopes, $R = (d\rho/dT)_H / (d\rho/dT)_L$, of the isochronal plots for samples irradiated to different doses and annealed simultaneously has been plotted versus T . Let r represent the ratio of the total damage, high to low dose, ($r = \Delta\rho_H / \Delta\rho_L$) for two samples in the above mentioned plot. The following has been observed: between 25 and 38°K R increases slowly from slightly below r to slightly above r . At about 38°K there is a sharp increase in R lasting about 5°. R then decreases to well below r and after about 2° again sharply increases to well above r , after which R once more decreases to below r .

Let $r > 1$ be the ratio of increases in defect concentrations in two irradiated samples. If the postirradiation decreases in concentrations are due to spectra of first order processes, whose amplitudes are proportional to dose, a plot of the ratio, R , of decreases versus time or temperature should be a straight line: $R = r$. Deviations from this line are indicative of amplitudes not proportional to dose and/or higher order processes. For processes of order $\nu > 1$, $R = r^\nu$ at the beginning of annealing, decreases rapidly to unity. Stage I recovery in metals has been frequently described as a spectrum of first order processes terminating in random migration leading to higher order characteristics. In the absence of nonlinear amplitude-dose effects, recovery in Stage I would be expected to be characterized by a ratio R constant at $R = r$, then increasing toward $R = r^\nu$, then decreasing toward unity. The behavior of R in Al (see previous paragraph) between 25 and 45°K is in qualitative accord with such a model. The remaining deviations from the $R - r$ line must be due to other effects to be discussed.

C. THRESHOLD ENERGY OF ALUMINUM (H. H. Neely)

The increase in the residual electrical resistivity per unit electron flux $d\rho/d\phi$ was measured near 4.2°K as a function of electron energy. The electron energy ranged from 0.19 to 1.6 Mev. The specimens were of 99.9999% purity and <0.8 mils thick. Potential leads were spot-welded to the specimens well within the irradiated portion to reduce the uncertainty in the geometrical factor. The displacement threshold energy was extrapolated from the $d\rho/d\phi$ versus T_m curve to yield a value of $T_d \approx 17$ ev; this is to be compared with the value of Mehner et al., $T_d = 16.5 \pm 3$ ev. The usual tail in the damage rate curves observed in Cu, Au, and Pt near threshold is absent in Al. This is consistent with the model of Bauer and Sosin where the tail is attributed to the ejection of light substitutional impurity atoms by focussons and thus may not be as important in Al. The data do not fit the simple step-function displacement theory with a reasonable value of ρ_f .

D. MODULUS CHANGES AT LOW TEMPERATURES - DISLOCATION PINNING IN PURE Cu, Au, AND Al (D. Keefer, A. Sosin, and J. Robinson)

The following is an abstract of an invited paper entitled "Modulus and Internal Friction Effects in Irradiated Metals," by D. Keefer and A. Sosin. Modulus and internal friction measurements have been made on Cu, Au, and Al following low-temperature electron irradiation or quenching and subsequent thermal treatment. Upon isochronal annealing, changes in modulus and internal friction resulting from dislocation pinning by interstitials and vacancies have been observed in all three metals, although the temperature ranges in which the effects occur are distinctly different. Results will be discussed in terms of point defect types in the

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three metals. Aluminum results will be contrasted with those obtained by yield stress measurements. Final release of point defects from dislocations occurs at higher temperatures in Cu than in Au or Al.

Dislocation pinning during extended irradiation at low temperatures has been studied in Cu and Au. Analysis of data for both metals requires cognizance of two dislocation components, these being different in their average loop length, density, and cross section for pinning. Analysis shows the pinning cross-sections for Au to be about 20 times those for Cu dislocations.

The rate of change of modulus with integrated electron flux has been studied for Cu and Au at 20°K. Characteristics of these damage rates yield information on the modes of defect production in the two metals.

The following is an abstract of an invited paper entitled "The Trapping of Point Defects by Dislocations," by D. Keefer and A. Sosin. Symposium sponsored by the AIME. The Symposium was held in Asheville, North Carolina, September 8 to 10, 1965.

A discussion is presented concerning the major aspects of point defect-dislocation interactions, comparing the extensive observations in copper with more recent data in gold and aluminum. Dislocation pinning effects are emphasized and compared with related experiments.

The study of damage rates in Au at 20°K as studied by modulus measurements is now almost complete. We have observed dislocation pinning during irradiation at electron energies below that at which direct lattice displacements are believed to occur. This substantiates our previous suggestion that dislocation pinning in Au can occur by focusing processes. The threshold energy for production of a pinning point at a dislocation by a focussed electron (about 29 ev) is below that for lattice displacements (33 to 36 ev). Comparison of calculated with experimental displacement cross-sections shows that focusing processes predominate in the creation of pinning points in the energy range studied, up to 2 Mev. This is in contrast to Cu, where direct displacements at dislocations appear to account for all pinning points created during irradiation at energies somewhat below 1 Mev. In the case of 1-Mev irradiation, some focusing process (possibly the dynamic crowdion) may be operative.

A paper on the modulus work in Au is nearing completion.

E. POINT DEFECT RELAXATIONS IN IRRADIATED METALS (D. Keefer)

An investigation of the internal friction peak observed near 230°K in irradiated Cu is continuing. The height of the peak has been found to be a function of the integrated electron flux up to 9×10^{17} electrons/cm² (defect concentration about 3×10^{-7}). In an attempt to clarify further the origin of this peak, we will presently investigate the dependence of peak height on the dislocation density.

F. ELECTRON IRRADIATION HARDENING OF METALS (O. Buck, D. Keefer, and J. C. Robinson)

Two samples from a single crystal of pure Al have now been irradiated and tested in the tensile apparatus. In general, the apparatus operated extremely

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well in all phases of the operation. Unfortunately, at least one of the samples tested was cold-worked during the procedure. We are presently trying to find the source of this cold-work.

G. DAMAGE PRODUCTION IN PLATINUM NEAR 7°K (W. Bauer)

A paper was presented at the APS meeting in Hawaii on the damage rate work in platinum. The abstract is as follows.

"The increase in electrical resistivity per unit flux $d\rho/d\phi$ of 99.999% pure 0.0002-in. -thick platinum foils was measured as a function of incident-electron energy from 1.3 to 2.1 Mev. Reasonable agreement with the theoretical displacement cross section, using a unit probability of displacement, is achieved with an effective threshold energy of 26 ev and a value of $\rho f = 6.0 \times 10^{-4} \Omega \text{ cm/unit}$ fractional concentration of Frenkel pairs. In gold, the best agreement between the theoretical and experimental cross-section was only achieved for an unusually low value of ρf . The ratio of $d\rho/d\phi$ for platinum to $d\rho/d\phi$ for gold is a constant (≈ 6) for energies above approximately 1.7 Mev, as expected for elements of similar atomic number, but decreases to zero near the threshold. These results are interpreted to indicate that (1) directional effects in the displacement processes are more important in gold than in platinum, and (2) that indirect displacement processes due to focussons are more important in gold near threshold than in platinum."

H. THRESHOLD ENERGY IN METALS (W. Bauer and A. Sosin)

A paper entitled "Recent Threshold Energy and Displacement Cross Section Measurements in Metals," was presented at the Radiation Effects Symposium of the AIME at Asheville, September 8. The abstract is as follows.

"We present a general discussion of threshold energy measurements and the inherent limitations imposed by theoretical and experimental considerations. A discussion of displacement cross-section calculations and measurements, including their relationship to the effective threshold energy, is given. We review the experimental results of measurements on copper and gold and discuss the latest results on platinum and aluminum. Finally, an attempt is made to present a coherent picture of displacement processes near threshold in these metals."

I. INFLUENCE OF IMPURITIES ON THE RECOVERY OF ELECTRON IRRADIATED GOLD (W. Bauer)

This work has been accepted for publication in Physics Letters. A summary of the salient features of the results follows.

1. Between 50° and 200°K the recovery of the 5-9's and 6-9's samples is practically identical both in the amount of recovery and in the position of the stages as evidenced in the slope plot.

2. Between 200° and 340°K the amount of recovery depends on the purity in a systematic manner, being smaller the purer the sample. Since the recovery in Stage III was not observed to completion, no statement can be made as to whether the center of the stage is shifted in temperature as a function of purity or whether the total amount of recovery in Stage III is a function of purity.

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3. The recovery of the 4-9's sample is significantly different from the 5-9's and 6-9's samples over the complete temperature range. There is suppression of the amount of recovery ($\approx 5\%$) near 20°K and subsequent enhancement of recovery in the Stage III region.

4. The results of the measurements made on the 4-9's + 0.1 at. % Cu sample basically confirm the trend indicated in (3), that is suppression of recovery at low temperatures and enhancement in the Stage III region with decreasing purity.

5. We have found reproducible resistivity increases near 20°K and from 70° to 80°K in the Cu-doped sample. The resistivity reading at 4.2°K after the anneal at 20°K is larger than the resistivity after irradiation.

6. The damage rate (resistivity increase per unit electron flux) near 10°K by 2 Mev electrons is a systematic function of the purity of the samples. The rate increase approximately 10% with a decrease of a factor of 10 in purity.

J. EVALUATION AND INTERPRETATION OF THRESHOLD ENERGY MEASUREMENTS (W. Bauer and A. Sosin)

This work has been considerably revised and expanded to include a section on multiple low angle electron scattering and its implications on single crystal irradiations. The work has been resubmitted to J. of Appl. Phys.

IV. EVALUATION OF EFFORT DURING REPORT PERIOD

This period has been characterized largely by consolidation of views in several areas of our research and presentation of these views at a number of meetings. The large number of invited as well as contributed papers given during the period is particularly gratifying to the group. A part of the period has been occupied by construction of new equipment and revision of some of the existing apparatus. This is a reflection of the expanding interests of the group as well as our intention to do more detailed work in several areas.

V. NEXT REPORT PERIOD ACTIVITIES

Work will be continued in all areas, in the directions indicated by the status summaries of Section III. Relatively little further reporting activity is planned for the next period, while experimental work is expected to be emphasized.