MASTER

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AEC RESEARCH AND
DEVELOPMENT REPORT

GA-7981

HTGR BASE PROGRAM
QUARTERLY PROGRESS REPORT
FOR THE PERIOD ENDING
MAY 31, 1967

Prepared under
Contract AT(04-3)-167
Project Agreement No. 17
for the
San Francisco Operations Office
U.S. Atomic Energy Commission

September 29, 1967

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INTRODUCTION

This report covers the work performed by General Atomic Division of General Dynamics Corporation under U.S. Atomic Energy Commission Contract AT(04-3)-167, Project Agreement No. 17. This Project Agreement calls for preliminary design and related exploratory development of a 1000-Mw(e), gas-cooled, nuclear power reactor incorporating advanced technology, and a 150-300 Mw(e) prototype. This program is based on the concept of the High-temperature Gas-cooled Reactor (HTGR) system developed by General Atomic.

The related exploratory development effort is aimed at a 1000-Mw(e) gas-cooled reactor plant that could be placed in operation in the 1970's, with operation of the prototype at an earlier date. The plant would demonstrate such new design features as:

1. Breeding or near breeding of fuel in a high-temperature, gas-cooled, thermal reactor using a Th-U²³³ fuel cycle.
2. Advanced reactor fuel elements, which may include new and simpler configurations and new methods of processing and fabricating to extend fuel lifetime and reduce fuel costs.
3. Alternative plant arrangements to reduce plant capital costs, including such possibilities as alternative materials (e.g., concrete pressure vessels) and the assembly of the reactor and steam-generating equipment in a single containment.
4. A supercritical-pressure steam cycle that is particularly adaptable to a 1000-Mw(e) plant and to the inherent high-temperature capabilities of the HTGR system.

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Task I
UPDATED 1000-MW(E) HTGR CONCEPTUAL DESIGN

ADVANCED CONVERTER TASK FORCE EVALUATION

Reactor plant designs, fueling requirements for the principal mode of operation (U^{235} fissile makeup), and research and development requirements have been submitted to ORNL for both the backup and reference design plants. The basic arrangements for these plants were established for consideration by the task force early in the evaluation procedure and no changes have been made in the designs as the studies have progressed. The basic design parameters for the plants are given in Table 1.1.

Table 1.1
COMPARISON OF BACKUP AND REFERENCE 1000-MW(E) DESIGNS

Parameter	Backup Design	Reference Design
Vessel arrangement	Steam generators under core	Steam generators under core
Steam cycle	1000°/1000°F/2400 psig	1050°/1050°F/3500 psig
PCRV	PSC type with linear prestressing	PSC type using wire-wound prestressing and without head tendons
Steam generators	6 PSC-type	6 radial-flow steam generators
Refueling method	Off-line region refueling	On-line column refueling

The supercritical steam cycle was selected as one of the design features for the reference plant. A further comparison of 1000°/1000°F and 1050°/1050°F steam cycles at 3500 psig has shown that the electrical output would be 1.6% higher for the 1050°/1050°F steam cycle at a given reactor thermal output. The capital cost of the plant excluding the steam generators would be approximately \$2.3 million higher for the 1050°/1050°F cycle. For a fuel cycle cost of 1.4 mill/kw-hr, a capital charge on initial investment of 12%, and a capacity factor of 80%, the 1050°/1050°F plant would yield lower power costs if steam generators cost \$200,000 less than for the 1000°/1000°F plant. However, it appears certain that steam generators for the higher temperature cycle would be more, rather than less, expensive, and the 1050°/1050°F steam cycle therefore appears to offer no possibility of economic gain. Since it has no other obvious advantages, it was decided not to consider the higher temperature steam cycle further at this time.

A comparison between 1000°/1000 F cycles at 2400 and 3500 psig led to the conclusion that if the cost of the steam generators for the supercritical cycle did not exceed the cost for the 2400-psig cycle by more than about \$2 million, choice of the supercritical cycle would lead to lower power costs. In addition, for a 1000-Mw(e) plant the 3500-psig cycle makes possible a choice of turbine blade sizes, whereas 33-1/2-in blades would be mandatory with the 2400-psig cycle.

The 3500-psig steam generators would certainly be more expensive than the 2400-psig ones, but no accurate cost comparison or supporting design study has been made. However, it is believed that the cost differential could be as high as \$2 million. The preferred design is therefore 2400 psig at 1000°/1000 F, but a supercritical steam generator design and cost comparison would be desirable at some future time to assist in optimizing the steam cycle.

There are several other potentially interesting steam cycles. The most important of these is the steam reheat cycle. It has been determined that this cycle could be designed to suit standard turbine generator equipment, although the steam conditions would be somewhat unconventional. The attractiveness of this cycle is tied to the amount of the cost saving on the steam generators from eliminating the reheater. It is not now possible to make any valid economic comparison for the steam reheat cycle.

The steam reheat cycle might make desirable a different circulator drive scheme from the reheat steam turbine in the reference design. A high-pressure steam turbine drive may be an attractive possibility, partly because it should make possible a simplification in the steam piping, valving, and control arrangements. A variant that might be considered is the use of high-pressure, low-temperature steam from the steam generator economizer-evaporator tube bank in the circulator turbine and then returning this steam to the superheater bank of the steam generator.

1000-MW(E) HTGR DESIGN STUDY

Basic Considerations

Work has proceeded towards an updated conceptual design for a 1000-Mw(e) HTGR based on the technology developed for the Peach Bottom and Fort St Vrain reactors and the reactor designs developed for the Advanced Converter Task Force evaluations, thereby exploiting the improvements in the state of the art since the TARGET reactor design was completed. The ground rules for the current studies may be summarized as follows:

- 1 The updated design should consist of a 1000-Mw(e) reactor with a single turbine generator
- 2 All mechanical components, such as circulator units and control rod drive units, should be removable and replaceable as a routine maintenance operation. The steam generator units should be removable,

but removal and replacement need not necessarily be possible within the scheduled shutdown time for maintenance and refueling, or within the equipment and manpower resources of the reactor operator. The steam generator units should be designed to last for 30 years without need for removal.

3. The reference design should be based as far as possible on concepts and component designs formulated for the Fort St. Vrain reactor developed for Public Service Company of Colorado (PSC). However, it could be assumed that several large HTGR reactors would be built, and changes to the PSC design concept should be considered if they improve the performance or the economy of the 1000-Mw(e) reactor.
4. The design concept should permit the incorporation of future engineering developments such as on-line refueling without the need for drastic rearrangement or complete redesign of the reactor.
5. The reference design should be for a reactor that could be on line in July 1973.

Based on the design study work performed for the task force evaluation, it has been recognized that the backup and reference design parameters established are not necessarily optimum and that the 1000-Mw(e) conceptual design should be updated. To enable the work to proceed, the following tentative design decisions have been made to establish a basis for a comparison of some attractive variations. These design decisions will be reviewed as the work progresses. Alternatives to the design features used in the task force work are described.

1. Reactor Core. Fuel elements, core height, and maximum power density in the core are the same as those in the PSC 330-Mw(e) reactor. This yields a rather flat core, avoiding xenon instabilities, but probably does not optimize either capital or fuel costs. The average power density is slightly higher, because the peaking factors are lower than for PSC. A four-year fuel cycle is assumed.
2. Reactor Layout. Two designs are being considered, a PSC-type layout with steam generators under the core (the basic configuration used in task force evaluations) and a layout with the steam generator modules located in separate cavities in the walls of the prestressed concrete reactor vessel. These two layouts will be under consideration until a clear technical or economic choice can be made between the two alternatives.
3. PCRV Design. The PSC-type and multicavity layouts involve different vessel designs, which are to be compared. Also, circumferential prestressing by linear tendons and by wire winding are to be compared, and a choice made between the two systems as early as possible. The head tendons have been eliminated for both of these designs.

4. Coolant Circuit. Primary and secondary coolant temperatures and pressures are the same as for the PSC design, but steam reheat may have advantages in conjunction with alternative circulator drives and should be evaluated before the studies are completed.
5. Steam Generators. The PSC-type design is to be scaled up to the required size, with minimum changes, and will be compared with schemes using multiples of the PSC-size modules. Other steam generator types, such as radial flow, will be considered only as possible variants to the reference design.
6. Coolant Circulators. Circulators are the PSC-type axial flow, steam turbine driven using cold reheat steam with the same pressure rise as PSC. Designs utilizing other types of drives such as superheated steam or low-temperature steam will be considered after the preliminary work leading to a choice of reactor layout has been made. Variations in the choice of circulator drives affect the steam cycle.

Circumferential Prestressing

Wire winding the circumferential prestress on the vessel is a possible alternative to the linear tendon scheme being used for the PSC reactor. It offers the possibility of greatly reduced capital costs, primarily because the expensive tendon anchorages needed with linear tendons would be eliminated.

There are two reasons for considering wire winding at this stage, the cost saving compared with linear prestressing, and the possibility of additional flexibility in vessel design due to the elimination of prestressing ducts in the walls. The prestressing forces that can be applied per unit height of vessel can also be greatly increased, although there seems to be no immediate need to exploit this potential.

The possible cost savings were first evaluated by comparing the current estimated costs for a 1000-Mw(e) linear prestressed PSC-type vessel with the estimated costs for the same basic vessel without cap tendons and with the wire-wound circumferential prestress. The relative costs are summarized in Table 1.2. Circumferential prestressing by wire winding rather than by linear tendon results in cost savings of more than 60%. The \$1 million saving projected on the prestressing cost represents the main source of the over-all cost saving. However, there are small additional contributions due to the reductions in concrete, form work, and reinforcement costs. For the multicavity design, the savings with wire-winding would be even greater. It was therefore recommended that the reference design should have wire-wound circumferential prestressing.

To date, a wire-winding system has not been fully developed for this requirement

Table 1.2
COST COMPARISON FOR LINEAR PRESTRESSING AND WIRE WINDING^a

	Linear Prestress (%)	Wire Winding (%)
Prestressing System:		
Vertical	10	9
Circumferential	52	18
Concrete	19	17
Reinforcement	15	14
Form work	<u>4</u>	<u>4</u>
TOTAL DIRECT COST	100	62

^a1000-Mw(e) PSC-type PCRV. Total direct cost of the linear prestressing system taken as 100%.

Separate studies have been made to evaluate head designs with and without head tendons. It was concluded that the vessel for the PSC plant could be constructed without head tendons by applying the head prestress circumferentially, and that this was economically attractive with linear prestressing systems. The difficulty of demonstrating the performance of the scheme in time for the PSC vessel precluded its use. The 1000-Mw(e) vessels, both of the multicavity- and PSC-type, have been conceptually designed without crosshead tendons, and appear to be fully satisfactory. With wire-wound circumferential prestress, the possible savings from eliminating head tendons are, of course, significantly greater than with linear tendons. It is unnecessary to make a detailed cost estimate to show that the potential savings justify the decision to eliminate head tendons from the 1000-Mw(e) reference design. A further advantage results from the additional flexibility in design available when accommodation of crosshead tendons need not be considered. In Britain, the Dungeness B reactor vessel now under construction, and probably all future designs, will have no crosshead tendons. Similar developments are taking place in France.

The development costs required to permit utilization of wire-winding or the designs without head tendons must be carefully assessed and evaluated against the potential advantages.

Plant Arrangement

Work has proceeded in parallel on the conceptual design for two reactor layouts, a scaled-up PSC-type reactor design, and a multicavity design. Both appear technically feasible. A list of the design parameters is provided in Table 1.3, and the reactor layouts are illustrated in Figs. 1.1 and 1.2.

Table 1.3
MAJOR DESIGN PARAMETERS

	PSC	1000-Mw(e) Plant	
		PSC Type	Multicavity
Reactor:			
Reactor thermal power, Mw	837	2469	2469
Active core diameter, ft	19.6	31.1	31.1
Active core height, ft	15.6	15.6	15.6
Core power density, kw/l	6.3	7.3	7.3
Core-reflector helium flow, 10 ⁶ lb/hr	3.33	9.85	9.85
Core helium pressure drop, psi	8.3	7.5	7.5
Number of control rod drives	37	91	91
PCRVR:			
Circumferential prestressing	linear	wire-wrap	wire-wrap
Inside diameter, ft	31.0	44.0	37.7
Inside height, ft	75.0	97.5	39.4
Outside diameter or across flats, ft	49.0	70.5	99.0
Outside height, ft	106.0	141.5	77.4
Steam generator cavity diameter, ft	31.0	44.0	13.2
Primary System:			
Number of loops	2	3	6
Helium pressure, psia	700	700	700
Total helium pressure drop, psi	14	12.5	12.5
Helium temp at circulator inlet, °F	742	745	745
Helium temp at core inlet, °F	758	755	755
Helium temp at core-reflector outlet, °F	1449	1443	1443
Helium temp at steam generator inlet, °F	1427	1427	1427
Primary system efficiency, %	99.65	99.65	99.65
Helium Circulators:			
Number of circulators	4	6	6
Compressor type	axial	axial	axial
Drive type	steam turb	steam turb	steam turb
Auxiliary drive type	water turb	none	none
Helium flow rate, 10 ⁶ lb/hr	3.49	10.32	10.32
Steam inlet pressure, psia	866	836	836
Steam inlet temperature, °F	738	727	727
Steam outlet pressure, psia	645	645	645
Turbine power/unit, hp	5750	9820	9820
Turbine power margin, %	5	5	5

Table 1.3 (continued)

	PSC	1000-Mw(e) Plant	
		PSC Type	Multicavity
Steam Generators:			
Number of modules	12	6	6
Shroud outside diameter, ft	5.4	12.0	12.0
Helium flow rate, 10 ⁶ lb/hr	3.41	10.09	10.09
Helium pressure drop, psi	3.0	4.0	4.0
Feedwater flow rate, 10 ⁶ lb/hr	2.31	6.86	6.86
Feedwater temperature, °F	404	420	420
Outlet steam temperature, °F	1005	1005	1005
Outlet steam pressure, psia	2512	2515	2515
Reheat steam flow rate, 10 ⁶ lb/hr	2.24	6.78	6.78
Reheat steam inlet temp, °F	673	670	670
Reheat steam outlet temp, °F	1002	1002	1002
Reheat steam outlet pressure, psia	600	600	600
Reheat steam pressure drop, psi	36	36	36
Steam Cycle:			
Main generator output, Mw	341	1010	1010
House generator output, Mw	0	9.4	9.4
Net station output, Mw	330	1000	1000
Net station heat rate, Btu/kw-hr	8660	8425	8425
Net station efficiency, %	39.4	40.5	40.5
Turbine arrangement	TC2F-33.5	TC6F-33.5	TC6F-33.5
Throttle flow, 10 ⁶ lb/hr	2.31	6.83	6.83
Throttle pressure, psia	2412	2415	2415
Throttle temperature, °F	1000	1000	1000
High pressure turbine exhaust pressure, psia	866	854	854
Reheat steam flow, 10 ⁶ lb/hr	2.24	6.78	6.78
Intermediate pressure turbine inlet pressure, psia	567	567	567
Intermediate pressure turbine inlet temp, °F	1000	1000	1000
Turbine backpressure, in. Hg abs	2.5	1.75	1.75
Number of feedwater heaters	6	6	6

Fig. 1.1--1000-Mw(e) reactor, P&C-type design

Fig. 1.2--1000-Mw(e) reactor, multicavity design

Scaled-up PSC Design. Layout work on the backup design evaluated by the task force has produced the following results:

1. For compact reactor layout, the optimum number of steam generator modules is six.
2. In order to make the best use of the PCRV internal space for the reference core design, which is 15.6 ft high and 31.1 ft in diameter, the frontal area of the steam generator modules should be smaller than for directly scaled PSC modules. A diameter of 12 ft was chosen for the reference design, whereas the directly scaled diameter of the modules would have been 13.75 ft. This results in an increase in tube bank height 2.2 ft greater than that for the PSC module. In scaling up the size of the steam generator modules from the PSC size to the reference design size for the 1000-Mw(e) reactor (factor of 6), there are additional increases in module height involved, because of the need to accommodate a much larger number of tube ends and subheaders. The height of the design module is 35.8 ft from the mounting flange face to the top of the module; the corresponding height of the PSC steam generator module is 25.5 ft.
3. The weight of a steam generator module for the 1000-Mw(e) reactor is estimated to be about 200 tons. This can be handled by lifting devices that run on rails attached to the lower side of the core support floor, very similar to the devices used for the PSC reactor. Steam generator removal is therefore also similar in nature. In order to satisfy these requirements, the connecting ducts between the core support floor and the steam generator modules are 11 ft long, compared with 6.8 ft on the PSC reactor.
4. The core support floor is a cooled structure supported on columns, as for the PSC reactor. The passage of the steam generator inlet ducts through the floor presents a problem that is solved in the reference design by making the duct cross section elliptical at the top end and circular at the bottom. Apart from this, the internal structure design is straightforward and appears to present few new problems. All the graphite core support components are removable through a 4.5-ft-diameter access penetration in the top head of the vessel.
5. The PCRV pressure was fixed at 845 psig (the same as for the PSC reactor) for preliminary design purposes. The pressure rise in the vessel resulting from a steam generator tube rupture is probably not as high as for the PSC reactor, if the same size steam generator subheaders are used. No optimization involving definition of credible accidents, sizing of subheaders, and choice of vessel reference pressure has been attempted yet.

Multicavity Design. An alternative multicavity design has been based as far as possible on the same ground rules and similar components for easy

comparison with the scaled-up PSC design. Layouts with PSC-type steam generators beside, and above the reactor core showed that with these generators in separate cavities, the steam generators should be around the core. With the down-flow core, gas flow in the steam generators is upward. The circulators are in the top head of the vessel, with the steam generators directly under the circulators. A steam generator module is removed by lifting it directly through the top head penetration after removing the circulator. There are some alternatives to this layout, involving return gas flow around the steam generator shrouds, which enables the gas flow in the steam generators to be downward. These alternatives appear to offer no real advantage over the more straightforward one described above.

The following conclusions have been made from the layout studies and calculations:

1. A preliminary vessel design was calculated using the same ground rules as for the PSC-type vessel. It is 99 ft in diameter by 77.4 ft high (the scaled PSC design is 70.5 ft in diameter and 141.5 ft high). The steam generator holes are 13.2 ft in diameter to accommodate 12-ft-diameter modules. The core cavity is 37.7 ft in diameter, smaller than the scaled PSC design cavity, whose diameter of 44 ft is fixed by the dimensions of the core support floor and the steam generator modules, rather than by the core and reflector.
2. Six steam generator and circulator modules can easily be fitted in the walls around the reference design core with space available for several additional holes. It might be economical to use these extra holes for helium storage; this depends largely on whether the liners in the storage holes would need insulation and cooling. A layout showing two such holes is shown in Fig. 1.2. However, it is assumed that for the present there will be no large holes in the walls other than the six for steam generators and circulators.
3. There is no clear need seen for a steel core barrel in the multi-cavity design. It may be possible to locate the core and reflector directly from the concrete vessel walls. Thermal insulation is needed between the vessel liner and the reflector graphite. The PCRV liner would receive a higher neutron irradiation than that in the PSC reactor, and this might require that part of the liner be made from an alloy steel, such as 9% nickel, to meet NDT criteria. This would still be less expensive than providing a steel core barrel.
4. As in the PSC-type design, the core support structure consists of graphite and carbon posts and blocks resting directly on the steel liner of the PCRV bottom head. There is no equivalent of the PSC core support floor. The insulation requirements for the vessel bottom head liner are the same as for the PSC core support floor.
5. The steam generators are of the PSC type, but the tube banks are inverted so that gas flow is upward, and the reheater is at the

bottom. There are no obvious technical difficulties with the downhill boiling in the evaporator-economizer section. The steam and water headers all pass through the hot-gas stream at the inlet to the module and need insulation.

6. It is not apparent that the PSC axial flow circulator design is necessarily optimum for this reactor layout. However, this circulator design is quite possible and is used in the reference design. Draining of the Pelton wheel cavity is an unresolved problem with the inverted circulator. Steam lines for the circulator turbine drives come up the outside of the PCRV. Some alternative schemes with internal steam connections between steam generators and circulators seem to justify further design effort.
7. The reactor building is wider, but not as high as that of the scaled-up PSC design. Sufficient layout work has been done to obtain over-all building dimensions for comparison.

Cost Comparison and Evaluation

To determine whether there is a significant cost difference between these two layouts, a capital cost comparison was made. Only those items obviously different in design or cost for the two designs were included in the comparison.

The cost estimates, which are summarized in Table 1.4, favor the multicavity design by about \$1.7 million, which represents about 12% of the costs included in the cost comparison, or about 1% of the total plant cost. The cost difference between the two reactor designs is not large, and it is unlikely that any other cost comparison that could be done without extremely detailed design as its basis would yield a different conclusion. The choice between the two designs must therefore be influenced heavily by considerations other than plant capital cost.

Research and development needs are much the same for both designs, except for development of the multicavity vessel itself. The multicavity vessel is a novel design and would almost certainly require a model to demonstrate the soundness of the design approach, whereas the scaled-up PSC vessel might well be built without a model. In both cases a scaled-up steam generator and circulator are needed, and these items would need some development, about the same in total for either reactor design, provided scaled-up PSC components were used.

Design and engineering work, apart from that included in the PCRV model program, is considered about the same for either design, but some additional cost must be debited to the multicavity design to allow for the additional preliminary design work needed for a new layout, as opposed to an existing concept. This would be an insignificant contribution to the cost of the whole job, however.

Table 1.4
1000-MW(E) REACTOR COST COMPARISON
(Costs in \$1000)

<u>Item</u>	<u>Scaled PSC Design</u>	<u>Multicavity Design</u>
PCRV Structure		
Concrete	533	614
Form work	114	132
Rebar	440	507
Vertical prestress	319	296
Circumferential prestress	575	706
Indirects and fee	<u>1206</u>	<u>1372</u>
Subtotal	3187	3627
PCRV Liner, Penetrations, and Closures		
Liner	886	947
Steam generator access hole	240	---
Core support access hole	52	32
Steam generator (6)	312	312
Circulator (6)	312	1118
Refueling (91)	931	931
Filter absorber	38	38
Purification wells	<u>159</u>	<u>159</u>
Subtotal	2930	3537
Insulation		
Class A ^a	568	553
Class B ^b	345	624
Class C ^c	<u>169</u>	<u>157</u>
Subtotal	1082	1334
Reactor Internal Structures		
Core support floor	1035	---
Core barrel	400	---
Circumferential gas seal ^d	---	100
Lower floor	200	---
Steam generator ducts and clamps ^e	250	---
Bellows	224	128
Steam generator seals (Δ cost)	---	<u>90</u>
Subtotal	2109	318
Reactor Building		
Excavation and substructure	910	1070
Superstructure	3063	2484
Indirects and fee	<u>667</u>	<u>568</u>
Subtotal	4640	4122
Miscellaneous Items		
Helium	230	86
Helium storage	575	215
Circulator removal (Δ cost)	290	---
Steam piping (Δ cost)	---	<u>100</u>
Subtotal	1095	401
TOTAL	<u>15043</u>	<u>13339</u>
COST DIFFERENCE		<u>1704</u>

^a Between cool gas and concrete liner.

^b Between hot gas and carbon steel.

^c Between hot gas and concrete liner.

^d Between liner and reflector.

^e Excluding bellows.

The selection of the design should therefore be made on the basis of which reactor is safer and cheaper, easier to operate and maintain, more adaptable to larger and smaller sizes, and more amenable to future development without radical redesign. A qualitative consideration of these characteristics leads to the following assessment:

1. The multicavity design is not inherently safer, but it has some characteristics under accident conditions, resulting from the solid, unpenetrated concrete vessel lower head directly beneath the refractory core support structure, and the absence of a separate core support floor, which further analysis might show to have significant advantage.
2. The 1000-Mw version of the PSC reactor with scaled-up steam generators has some potential seismic problems. The horizontal accelerations at the top of the tall vessel will affect the design of the refueling machinery and similar components. The current designs for the PSC-type steam generators do not require lateral support. Such support may be necessary with the increased height of the scaled-up units under seismic loads; however, it may be difficult to provide. In the multicavity design lateral support for the top ends of the steam generators can be accommodated fairly readily.
3. In the multicavity design the circulators and steam generators are removed directly upward, working from the relatively clear operating floor, directly under the building crane. For the PSC-type, special equipment is provided for circulator removal into the space under the reactor vessel.
4. The multicavity design has significantly fewer internal components than the PSC design, which has additional components such as core support floor, flex columns, primary coolant bellows, core support floor and columns, steam generator inlet ducts, and clamps.
5. The multicavity design allows somewhat more flexibility in the choice of component dimensions, parameters, and numbers of modules. A change from the rather flat reference design core to one with a height that is more equal to the diameter would result in expensive space wastage in the PSC design, but would probably reduce the vessel cost for the multicavity. Any increase in steam generator dimensions from those considered in these comparisons is likely to be more expensive to the PSC-type than the multicavity design.
6. The multicavity vessel seems to offer some construction advantages, because it would simplify installing and checking out the steam generators, circulators, and pipework, all of which go on simultaneously, and in the case of the PSC design, largely in the same area under the vessel. The elimination of many of the internal components mentioned before has construction advantages. However, the liner and penetrations, which need to be designed and procured early in the construction schedule, are significantly more complicated and

expensive for the multicavity vessel. The need for prepacked aggregate construction of the PCRV bottom head appears to be eliminated in the multicavity design, because of the elimination of many of the penetrations under the bottom head liner. This might result in some cost saving.

7. More advanced reactor designs, such as one employing gas turbine circulators or complete gas turbine power cycles, fit conveniently into the multicavity layout and could form a later stage of a logical development of this arrangement. It is not apparent how such developments could be incorporated into the PSC type of arrangement.

ADVANCED FUEL ELEMENT DESIGN

Detailed analysis of core thermal performance and coolant-flow pressure drop continued this period for the 1000-Mw(e) HTGR orificed backup design. Fuel element geometry, thermal conductivities, and a two-zone cosine axial power profile were all assumed to be identical to the 330-Mw(e) reactor (PSC). Hot-spot factors and the effects of crossflow were neglected. These analyses incorporated the following estimates:

Side reflector power	20 Mw
Side reflector bypass flow	2.5% of total
Control rod flow	4.0% of total
External region gap bypass flow	2.5% of total

The interface loss coefficient (0.07) used was based on data from off-set loss coefficient tests. The value of 0.07 corresponds to a coolant hole eccentricity at fuel element interfaces of 0.070 in., based on current fuel element tolerances. The surface roughness value ($\epsilon/D = 0.001$) used is based upon conservative friction-factor data determined from air-flow tests with deep-drilled graphite elements. The fuel conductivity was assumed to be 1.2 Btu/hr-ft-°F.

The total calculated pressure loss across the core, including the orifice, is 7.4 psi. This is the difference between the upper plenum total pressure (700 psia) and the static pressure at the outlet of the bottom reflector transition element (692.6 psia) in a maximum-powered region ($P/P_{\text{core}} = 1.44$). The velocity head at this exit location is 0.586 psi. The Δp of 7.4 psi does not assume any pressure recovery.

The thermal results are shown in Figs. 1.3 through 1.7. Fuel volume fraction and active core graphite surface area fraction data are presented in Figs. 1.3 and 1.4, respectively. Figures 1.5, 1.6, and 1.7 are axial fuel, graphite, and coolant temperature distribution for maximum, average, and minimum-powered regions, respectively. The peak fuel centerline temperature is 2185°F at the axial midplane of the maximum-powered region, while the corresponding mean fuel temperature ($\int T dA/A$) is 1905°F.

Fig. 1.3--Fuel volume fraction above a given temperature

Fig. 1.4--Active core graphite surface area fraction above a given temperature

Fig. 1.5--Axial temperature distributions for the maximum-powered region

Fig. 1.6--Axial temperature distributions for an average-powered region

Fig. 1.7--Axial temperature distributions for the minimum-powered region

REFUELING STUDIES

During this period work was started on a survey of the prior developments in the field of on-line refueling as far as revealed in the published literature.

The characteristics of existing systems have been tabulated with particular emphasis on those machine characteristics that depend on reactor parameters and the building layout configurations that depend on fuel handling machine design policy.

A refueling head mechanism that appears to be capable of changing a single column of fuel in the 1000-Mw HTGR core configuration has been conceived.

Since the head may involve driven rollers, a survey of the capability of the aircraft actuator industry in this area was initiated. There is good indication that suitable high-temperature rotary pneumatic actuators with dry lubrication are commercially available.

Drawings of the top reflector elements are being prepared to show the configuration the refueling machine would see in the 1000-Mw core with no variable orifice mechanisms and separate keys on all elements. It is of interest to note that the top reflector element in each column can contain a fixed orifice differing from that in an adjacent column. It would thus be possible to have a standard series of top reflector elements, each having a different orifice restriction. These could be changed while the refueling machine is in the nozzle changing fuel in an adjacent column.

The head would have sensors that would be actuated by the presence of coded indentations in the top of the element and so permit automatic simultaneous logging of orifice size and core position.

Task II
PCRV MODEL NO. 1

A review of published data on PCRV Model No. 1 (Ref. 1)* has been completed. The purpose of this review was to further investigate, and possibly explain, observed differences between analytical predictions and test results. The results of the study reported here include previously unpublished data that were formalized in the process of the review work. Reference should be made to Fig. 2.1 for the proper orientation of instrumented planes in Model No. 1.

In summary, the comparison study between Model No. 1 test results and the predictions from an elastic axisymmetric finite-element analysis for loading conditions of prestressing and 500-psig pressurization showed the following:

1. In general, reasonable agreement between test and analytical results is indicated at vessel locations where the assumption of axisymmetry prevails. In local areas where discontinuities are present, such as penetrations and tendon anchor bearing blocks, the comparison is not valid since the analysis did not account for the presence of discontinuities.
2. Differences in the test and analytical strain comparisons at prestressing were attributed to the following:
 - a. Test results include shrinkage and creep effects during the prestressing time span which were not accounted for in the elastic analysis.
 - b. The analytical solution assumed that prestressing forces would be applied uniformly and instantaneously; the actual prestressing sequence can therefore affect the comparison.
3. Good agreement is indicated for strain comparison at pressure loading.
4. A comparison of test and analytical strains for the liner indicated that differences may be attributed to the local bending of the liner, which is partly due to the yielding of the protective material coating of the gages adjacent to the concrete.
5. For prediction of vessel pressure that would initiate concrete cracking, the use of the maximum strain theory compared well with the observed pressure values for Model No. 1. The use of the maximum stress theory gave conservative estimates of the cracking pressures at the critical areas of the vessel.

*References will be listed at the back of each section.

Fig. 2.1--Model No. 1 prestressed concrete reactor vessel

HOOP STRAIN AS MEASURED ON REINFORCING BAR

The hoop strain data for both the fully prestressed case and the pressure case (computed and measured) are plotted in Fig. 2.2. In the heads, the data were taken at a radius of 25 in., and in the barrel section, at a radius of 52.5 in. near the inside surface. In the top head, the hoop bars circumscribe the array of small nozzles and pass between this array and the three large 18-in. penetrations (Fig. 2.1).

With the exception of the gages near the large penetration which could not be accounted for in the analysis, the pressure data matches prediction quite closely. The prestressing data is not as predictable because of the sequential method of prestressing used during construction and the time span involved.

In the top half of the barrel section agreement is good, but in the bottom half the bulk of the data shows a decidedly greater compressive strain than predicted. The two factors that may cause the prestressing strain to exceed the computed value are creep and shrinkage, both of which occur during the prestressing time span and cause the negative strain to increase with time. However, most of the shrinkage occurred during the 3-month period prior to prestressing the vessel. There is also no reasonable explanation why creep should affect the lower half of the barrel section more than at other places.

The prestressing data match prediction quite closely in the heads (again, with the exception of gages near the large penetrations). The three data points on planes 11, 12, and 13 near the 18-in. penetrations show the strong influence of this discontinuity. The strain is greater at all three locations during both prestressing and pressurization by more than a factor of 2. This result is significant as it shows that the penetration has an acceptable effect on the strain. The prestressing strain values are reasonable ($-449 \mu\text{in./in.}$ maximum) and these increases actually overcompensate for the corresponding increase in tensile strain caused by pressure. After pressure is applied, this area is still in net compression at approximately $-294 \mu\text{in./in.}$

To show hoop strain response during prestressing and at all pressures, Figs. 2.3 through 2.8 have been constructed. These six plots are of three gages (194, 196, and 251) for both prestress and pressure. Gage 196 is a head gage, gage 251 is a barrel section gage (at vessel midsection), and gage 194 was chosen to show the strain history in the area near a large 18-in. penetration. The data for this gage are expected to deviate from prediction. In Fig. 2.2, gage 194 is represented by the circle data points at elevation 144.5, and, as Figs. 2.3 and 2.6 show, deviation from predictions is continuous and regular during the complete prestressing cycle and at all pressures. For the typical head and barrel section gages (196 and 251), Figs. 2.4 and 2.5 show that strain followed a predictable history during prestressing. Likewise, Figs. 2.7 and 2.8 show that these gages (which were typical as can be seen from Fig. 2.2) follow the predicted pressure-strain relationship up to pressures at which cracking and non-linear behavior occur.

Fig. 2.2--Hoop strain in the cylinder and heads

Fig. 2.3--Hoop strain versus time in the top head at a main nozzle

Fig. 2.4--Hoop strain versus time in the top head away from a main nozzle

Fig. 2.5--Hoop strain versus time in the cylinder

Fig. 2.6--Pressure versus hoop strain in the top head at the main nozzle

Fig. 2.7--Pressure versus hoop strain in the top head away from the main nozzle

Fig. 2.8--Pressure versus hoop strain in the cylinder

AXIAL STRAIN

The axial strain for both the fully prestressed condition and the 500-psig case are plotted for three radial locations in Figs. 2.9, 2.10, and 2.11.

Three types of discrepancy between the computed lines and the data points are evident: (1) the data for prestressing is scattered and in general falls to the left of the computed lines indicating a greater-than-predicted compressive value, (2) 7 out of 71 of the prestressing data points show large tensile values instead of the predicted compressive values, and (3) for pressure data, the strain peaks near the inner vessel surface at the haunch are not as sharp as predicted by analysis.

The data scatter of the prestressing plots can be expected because the nature of the loading produces many local effects. The computed values necessarily assume a relatively uniform application of hoop load, whereas the hoop tendons actually exert forces that are concentrated at many discrete points along the circumference. Also, the predicted strain profile has many sharp peaks and steep slopes indicating a rapidly changing strain field where a slight change in sensor location would cause a large shift in strain value.

The general shift to the left of the prestressing data is probably due to creep not accounted for in the analysis. In the pressure case, creep was cancelled out by using data point samples taken the same day, but this is not possible for prestressing since prestressing took place during a period of more than 3 months.

Of the seven off-scale prestressing data points on Figs. 2.9, 2.10, and 2.11, the +147 circle at the lower haunch on Fig. 2.9, the two points on Fig. 2.10, and the one on Fig. 2.11 are within a few inches of a discontinuity or the bottom head tendon anchor. The other three points on Fig. 2.9 are not near a discontinuity. The two in the top head area are between (not near) the large 18-in. penetrations which are not accounted for in the analysis. The analysis accounted for all of the other geometric discontinuities (haunches and anchors), and in general the data was scattered and to the left as in other areas. No special effect on axial strain can be attributed to the 18-in. holes, and no general explanation can be given for the few off-scale data points. Local bending of the rebar into the void which was created for the strain gage is an unsubstantiated possibility. It is noted that the seven points exhibit linear and predictable response to pressure loads.

All the data for the pressure condition showed good agreement with analysis. Even in the aforementioned deviations in the haunch regions of Fig. 2.9, the difference is less than 50 $\mu\text{in./in.}$

STRAIN IN THE LINER - BARREL SECTION

Figure 2.12 shows both prestressing and pressure data for the hoop strain in the liner and Fig. 2.13 shows the same for the liner axial strain. Three types of anomalies between the computed lines and the data points are

Fig. 2.9--Axial strain in the cylinder and heads (52.5-in. radius)

Fig. 2.10--Axial strain in the cylinder and heads (60-in. radius)

Fig. 2.11--Axial strain in the cylinder and heads (67-in. radius)

Fig. 2.12--Hoop strain in the liner cylinder

Fig. 2.13--Axial strain in the liner cylinder

evident: (1) all prestressing data are scattered, (2) the hoop prestressing data falls to the right of the computed line, and (3) for both the hoop and axial pressure cases, a certain group of data points shows a significantly greater tensile value than predicted, with some points falling off scale.

The data scatter for the prestressed condition can be expected for the reasons outlined in the preceding section. In addition, liner data may experience a greater degree of scatter since local bending is possible due to lack of 100% liner bonding to the concrete. An inspection of the interior liner surface after prestressing and prior to pressurization revealed small ripples and bending.

In Figs. 2.12 and 2.13 the pressure data appears to be erratic, unlike the excellent agreement noted for the rebar data. In Fig. 2.12 for hoop strain, 12 data points are near the computed line and 9 fall significantly to the right indicating relatively large positive strains due to pressure. In Fig. 2.13 for axial strain, 9 points fall near the computed line while 10 fall significantly to the right. Of the 19 points that fall significantly to the right, 15 are on the concrete side of the liner, which must have some significance in causing this type of anomaly. It is suggested that the increase in positive strain is due to the bending of the liner into the void created by the epoxy around the strain gage on the outside of the liner. If one assumes a circular steel plate 0.162 in. in thickness and 1 in. in diameter, clamped at the edges with no other support, a pressure of 500 psig on one side causes a positive strain of 465 $\mu\text{in./in.}$ on the opposite side. This value is close to the 3 off-scale points of Fig. 2.12. On the actual liner, this affected area at each gage is not a true circle of 1 in., the edges are not rigidly clamped, and the hard epoxy will provide some degree of support. However, the example illustrates the large deviation that a small area of reduced support may create. In view of the fact that the gages showing largest discrepancy are on the concrete side of the liner, this becomes a plausible explanation.

To illustrate the behavior of both a hoop and an axial gage on the concrete side of the liner during prestressing and pressurization, gages 287 (axial) and 288 (hoop) were selected for individual plotting. On Figs. 2.12 and 2.13 these gages are represented by the circle and tail at vessel mid-section (elevation 74.25 in.). Figures 2.14 and 2.15 show these axial and hoop values during the prestressing sequence, and as in Figs. 2.12 and 2.13, agreement is satisfactory. Axial gage 287 (Fig. 2.16) shows that for pressurization the increase in positive strain was quite uniform up to 500 psig and beyond, even though a small negative strain was predicted as noted on Fig. 2.13. Figure 2.16 also shows an example of a liner strain gage undergoing strain adjustment at high pressures (1200 to 1300 psig). Hoop gage 288 on Fig. 2.17 shows that the actual strain is larger than the predicted value.

HOOP AND AXIAL STRAIN CONTOURS

Figures 2.18 through 2.21 are strain contour plots similar to those given in Section 2 of the PCR Model No. 1 report (GA-7097, Ref. 1), but are

Fig. 2.14--Computer plot of an axial gage on the liner

Fig. 2.15--Computer plot of a hoop gage on the liner

Fig. 2.16--Pressure versus axial strain on the outside of liner at vessel midsection

Fig. 2.17--Pressure versus hoop strain on the outside of liner at vessel midsection

Fig. 2.18--Hoop strain contour, full prestressing

Fig. 2.19--Axial strain contour, full prestressing

Fig. 2.20--Hoop strain contour, 500-psig internal pressure

Fig. 2.21--Axial strain contour, 500-psig internal pressure

of strain instead of stress. The hoop and axial contours for both full prestressing and 500 psig are given. This analytical data was used for comparison with test gage data.

Figure 2.18 shows the hoop strain throughout the vessel after full prestressing has been applied. The fact that there are relatively few contours reveals that hoop strain is quite uniform with the maximum value of 190 $\mu\text{in./in.}$ at bottom center. Figure 2.19 is the axial strain for the same case. The maximum value of 503 $\mu\text{in./in.}$ occurs on the vessel exterior under a head tendon anchor. The large number of contours show the rapidly changing strain field at tendon anchors, along the curved head tendon, and at the cylinder-head interface. A rather surprising result is the total positive axial strain in the heads along the vessel centerline. This is caused by the Poisson effect of the hoop loading. These figures show the strain as it is actually predicted to exist prior to vessel pressurization.

Figures 2.20 and 2.21 are the hoop and axial strain contours for the 500 psig case. These values, of course, never exist because before pressurization the strain due to prestressing is superimposed on the vessel. The contours show the strain values caused by 500 psig pressure and reveal that these strains are low everywhere. In hoop, the maximum of 160 $\mu\text{in./in.}$ occurs on the inside wall at the vessel midheight. In the axial case, the maximum strain of 198 $\mu\text{in./in.}$ occurs near the upper haunch. By comparing Fig. 2.18 with Fig. 2.19 and Fig. 2.20 with Fig. 2.21, it is evident that 500 psig represents a rather modest pressure for this vessel. Net negative strain occurs everywhere.

PREDICTION OF ONSET OF CRACKING

A study conducted to predict the pressures at which various types of cracks would appear was reported in GA-7097 (Ref. 1). The results predicted a haunch crack at 650 psig, a horizontal crack on the exterior surface at vessel midsection at 750 psig, and a vertical crack at 800 psig. These results were based on the maximum stress theory which predicts failure (cracking) when the maximum principal stress reaches the value at failure in a simple tension test. Assuming this value to be 500 lb/in.² tension, the above predicted values were calculated.

There is no way to visually detect a haunch crack. Horizontal cracking, however, was first observed at a pressure of 920 psig. Vertical cracking was evident in the data at about 950 to 1000 psig, though the vertical cracks were not visually detectable until the pressure exceeded 1000 psig.

Cracking pressures seem to be significantly higher than predicted. For concrete, the ratio of tensile to compressive strength is around 0.083, which is less than Poisson's ratio at 0.15, and a compressive load should therefore cause failure by transverse tension. For this reason, the maximum strain theory would be more applicable, i.e., cracking is imminent when this strain (not the stress) reaches the value of strain that occurred in a simple uniaxial tension test. Predicted values of pressure for onset of cracking are shown in Table 2.1

Table 2.1
PREDICTED AND OBSERVED CRACKING PRESSURES

	Cracking Pressures (psig)		
	Vertical Crack	Horizontal Crack	Haunch Crack
Max. stress theory	800	750	650
Max. strain theory	1084	886	795
Max. strain theory, with 10% tendon increase	1145	942	717
Test observation	>1000	920	Not visible

The values for the maximum strain theory are much closer to the observed values. The actual initial tendon loads were 10% in excess of the values used in the analysis. This was done to account for a 10% reduction as a result of long-term creep, but the vessel was not tested long term. Therefore, the cracking pressures predicted by the maximum strain theory for a tendon load at 10% increase over the originally assumed loading is also included in the table. These values fall very close to the actual pressure at which cracking was observed. The use of the maximum stress theory will give a conservative estimate of vessel cracking pressures.

References

1. Prestressed Concrete Reactor Vessel Model 1, USAEC report GA-7097, General Atomic, October 25, 1966.

Task III
ANALYTICAL METHODS DEVELOPMENT

THREE-DIMENSIONAL HEAT-TRANSFER CODE

As part of the work on the three-dimensional heat-transfer code RAT-3D, the calculational methods are being investigated as a means of improving the over-all efficiency of the code. It was previously reported that a study was underway to compare a new calculational algorithm (Ref. 1) with the one originally used (Ref. 2) in the code. The new algorithm appeared to have significant advantages over the original one, and a major part of the effort during the reporting period was devoted to comparing the two.

The new algorithm uses explicit relations (new values are based on known old values) but achieves stability by a special calculational method. The commonly used explicit methods are not stable and, if time steps are chosen to be larger than an easily derived criterion, lead to diverging numerical results. The implicit method (new values are related to current values), as presently used in the RAT code, may be shown to be inherently stable. The stability properties for explicit and implicit methods may be determined only for linear problems. The general nonlinear problem is not amenable to a stability analysis, and experience has shown that nonlinear problems suffer from numerical instabilities when the time step is too large. The maximum allowable time step for stability is therefore a useful criterion for comparing calculational heat-transfer methods.

The new and original algorithms were compared by determining the maximum stable time step for both a two-dimensional problem and a three-dimensional problem. The results showed that the original computational algorithm in the RAT code is better than the new method tested by a factor of about 70 for the two-dimensional problem and a factor of about 5 for the three-dimensional problem in terms of the maximum stable time step. Although the new algorithm might have allowed an increase in the size of the RAT code, its use was abandoned because the computational efficiency of the code would have been drastically reduced.

The effort on the heat-transfer code has now been directed toward obtaining a two-dimensional version having the capability of treating internal coolant flows as in the present three-dimensional version.

GAMMA BUILDUP IN HETEROGENEOUS MEDIA

Transport theory is being utilized to calculate gamma buildup factors in materials not included in NYO-3075 (Ref. 3), such as graphite and concrete, and in homogeneous and heterogeneous mixtures, such as iron/concrete and iron/

graphite. It is hoped that the results can be generalized at a later date in order to make the data more tractable for shield engineering purposes.

During the preceding quarter, buildup factors were calculated for heterogeneous mixtures of iron and concrete, for homogeneous graphite, and for a Co^{60} source in water.

The result that buildup was greater for some combinations of iron and concrete than for concrete alone, and in other cases less than for iron alone, appeared anomalous. Since these runs were based on actual iron and concrete densities, it was theorized that the abrupt change of density from one region to the next could be causing these unexpected results. Transport runs were then made on several configurations of iron alone, as shown in Table 3.1.

Table 3.1
EXPLORATORY TRANSPORT RUNS ON VARIOUS IRON COMBINATIONS

Source Description	First Layer		Second Layer		Calculated Energy Buildup Factor at 20 mfp
	Thickness (cm)	Iron Density (g/cm^3)	Thickness (cm)	Iron Density (g/cm^3)	
0.5-Mev isotropic point source	25	2.42	25	7.25	37.8
	25	7.25	25	2.42	53.5
	50	4.84	none	--	47.2
0.5-Mev isotropic plane source	25	2.42	25	7.25	40.7
	25	7.25	25	2.42	40.7
	50	4.84	none	--	41.3

It can be seen that in the case of a point source and spherical geometry, the density distribution of the material has a pronounced effect upon the calculated buildup factor, whereas in the plane case no effect is observed. The attenuation of the 0.5-Mev source group did not vary by more than $\pm 5\%$ in the spherical runs. Therefore, the observed variation of buildup factor must be attributed to the behavior of the collided fluxes. Qualitatively, the effect can be explained with reference to Fig. 3.1.

In Case I, the buildup (scattered) radiation suffers considerably more attenuation than in Case II, because path B'C passes through high-density material; consequently, one would expect the buildup factor for Case I to be smaller than that for Case II, as borne out by the table. Furthermore, the results for homogenized Case III lie between those for the two heterogeneous cases. In the case of slab geometry, AB no longer equals AB', so that the extra attenuation along B'C in Case I would be completely counterbalanced by extra attenuation along AB' in Case II. A comparison of the energy

Fig. 3.1--Variation of buildup factor

buildup factors is also shown in Fig. 3.2; note the rapid increase in buildup in the outer spherical region of low density (Curve A).

The results clearly indicated the need for additional calculations of buildup in heterogeneous iron/concrete mixtures, this time with a uniform cross section in each material. Such calculations were performed, as before, only for a 0.5-Mev isotropic point source. Both sets of calculations are summarized in Figs. 3.3 through 3.5

It is apparent that the appropriate buildup factor to utilize for a practical shielding problem will depend upon the actual source-to-detector geometry. It is just as apparent that this geometry is not usually known in advance for every source point and detector location. Consequently, it is planned to perform future calculations on heterogeneous media only for a uniform cross section throughout each configuration.

A plot of energy-absorption buildup factors in a heterogeneous configuration is interesting (see Fig. 3.6). In the case where a high-Z material follows a low-Z material, a large peak in B_a occurs near the interface, owing to the abrupt absorption of low-energy gammas built up in the low-Z material. In the configuration with 5 mfp iron following 15 mfp concrete, B_a reaches a peak of 300.

In ORNL-3198 (Ref. 4) the following equations are suggested for calculating the energy-absorption buildup factor for radiation passing through layers of two or more different materials:

$$B_a = \{ [B_{a_I}(\mu_1 r_1 + \mu_2 r_2 + \mu_3 r_3)]^k [B_{a_{II}}(\mu_1 r_1 + \mu_2 r_2 + \mu_3 r_3)]^{1-k} \}^m$$

$$[B_{a_{III}}(\mu_1 r_1 + \mu_2 r_2 + \mu_3 r_3)]^{1-m} \quad , \quad (1)$$

Fig. 3.2--Effect of density distribution on iron energy buildup factors calculated by 1DF transport code

Fig. 3.3--Gamma energy buildup factors in iron and concrete

Fig. 3.4--Gamma energy buildup factors in concrete and iron

Fig. 3.5--Gamma energy buildup factors in iron and concrete

Fig. 3.6--Gamma energy-absorption buildup factors in iron and concrete

or

$$B_a = (\{B_{a_I} [\mu_1(r_1 + r_2 + r_3)]\}^k \{B_{a_{II}} [\mu_2(r_1 + r_2 + r_3)]\}^{1-k})^m \{B_{a_{III}} [\mu_3(r_1 + r_2 + r_3)]\}^{1-m}, \quad (2)$$

where $k = e^{-\mu_2 r_2}$ and $m = e^{-\mu_3 r_3}$. A series of calculations was made using these equations. The values obtained with Eq. (2) were very high. A comparison is made between results obtained from Eq. (1) and transport theory for some of the cases thus far run for heterogeneous iron/concrete mixtures in Table 3.2 for uniform density.

Table 3.2
COMPARISON OF ENERGY-ABSORPTION BUILDUP FACTORS
IN IRON/CONCRETE CONFIGURATIONS

Configuration (0.5-Mev point source)	Energy-Absorption Buildup Factor (B_a)	
	Eq. (1)	Transport Theory
5 mfp concrete followed by 15 mfp iron	87.2	89 - 94.5 ^a
10 mfp concrete followed by 10 mfp iron	87.2	95 - 108 ^a
15 mfp concrete followed by 5 mfp iron	87.2	110 - 132 ^a
5 mfp iron followed by 15 mfp concrete	112	102
10 mfp iron followed by 10 mfp concrete	112	93 - 90.5 ^a
15 mfp iron followed by 5 mfp concrete	112	78 - 76 ^a
2.5 mfp concrete, 15 mfp iron, 2.5 mfp concrete	110	67.3
5 mfp concrete, 10 mfp iron, 5 mfp concrete	112	81.5 - 80.4 ^a
7.5 mfp concrete, 5 mfp iron, 7.5 mfp concrete	112	94.9

^aDependent on outer boundary conditions.

Calculations of buildup in homogeneous graphite were repeated, utilizing a somewhat more sophisticated code arrangement to alleviate the need for paired runs. The results (Fig. 3.7) are little changed from those already reported in the previous quarterly progress report (GA-7801). They are given in Table 3.3.

Fig. 3.7--Gamma dose buildup factors in graphite and comparisons with other data

Table 3.3
CALCULATED GAMMA GUILDUP FACTORS IN GRAPHITE

$\mu_0 r$	$E_0 = 0.5 \text{ Mev}$	$E_0 = 1.0 \text{ Mev}$	$E_0 = 2.0 \text{ Mev}$
I. Energy Buildup Factor (B_E)			
1	2.55	2.10	1.73
2	5.20	3.79	2.73
4	14.3	8.59	5.03
7	38.0	18.6	8.92
10	73.9	31.4	13.1
15	167.0	59.5	20.2
20	301.0	92.0	26.6
II. Dose Buildup Factor (B_R)			
1	2.48	2.16	1.88
2	5.21	4.03	3.08
4	15.4	9.60	5.89
7	49.6	22.8	11.0
10	106.0	40.5	16.6
15	252.0	77.5	25.8
20	463.0	121.0	34.2
III. Energy-absorption Buildup Factors (B_a)			
1	2.39	2.08	1.85
2	4.73	3.73	2.98
4	12.8	8.45	5.56
7	36.4	18.9	10.3
10	74.0	32.3	15.2
15	170.0	61.1	23.6
20	308.0	95.3	31.4

Homogeneous mixtures of iron and graphite resulted in the curves shown in Figs. 3.8 through 3.10, in which dose buildup factor is plotted against volume percent iron (or graphite) in the mixture. A typical spectral distribution is presented in Fig. 3.11; it can be seen that a very substantial low-energy peak exists at about 40 kev in pure graphite, which is completely suppressed by the addition of only 10% iron. This low-energy peak contributes substantially to the dose buildup factor in graphite, because of the accompanying rise in the dose conversion factor at these energies (compare B_R and B_E at 0.5 Mev and 20 mfp).

The literature contains many recipes for the interpolation or extrapolation of buildup data to elements not calculated by Goldstein and Wilkins (Ref. 3). One of these is presented in ORNL-3198. In Table B.4 of that report are listed the parameters A, B, and C for energy-absorption buildup in the equation

Fig. 3.8--Gamma dose buildup factors in homogeneous mixtures of iron and graphite

Fig. 3.9--Gamma dose buildup factors in homogeneous mixtures of iron and graphite

Fig. 3.10--Gamma dose buildup factors in homogeneous mixtures of iron and graphite

Fig. 3.11--Gamma spectra from 0.5-MeV point source in mixtures of carbon and iron

$$B_a = 1.0 + A(\mu_0 r) + B(\mu_0 r)^2 + C(\mu_0 r)^3$$

for a point isotropic source. Values of these parameters are given in Z down to 2.

Using the values of A, B, and C listed for Z = 6, a comparison was made with the values of B_a for graphite calculated by transport theory (Fig. 3.12). The data from ORNL-3198 is 35% low at this point. It is expected that the discrepancies for an element like B_a (Z = 4) would be even greater.

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Fig. 3.12--Gamma energy-absorption buildup factors in graphite

Task IV
FISSION-PRODUCT MECHANISMS

POSTIRRADIATION EXAMINATION OF THE GAIL IV FUEL ELEMENT

Gamma-counting of about 400 cored samples from the GAIL IV fuel element has been completed. The gamma spectra have been processed by the 1108 computer, and plotting of the concentration profiles for each isotope has begun.

Radiochemical analyses and beta-counting techniques to determine Sr^{89} and Sr^{90} concentrations across sections of the fuel element are in progress. Beta-counting of Sr^{89} is possible in the absence of Ce^{144} , since it is then the highest energy beta emitter present.

EVALUATION OF PHOTOELECTRIC CROSS SECTIONS FOR THE LITHIUM-DRIFTED GERMANIUM DETECTOR

Several isotopes of known disintegration rates have been used to evaluate photoelectric cross sections for the 15-cm³ lithium-drifted germanium detector now in use. The energy range covered is from 100 to 1330 kev. This evaluation is necessary in order to calibrate the detector for a determination of the absolute disintegration rate of unknown samples. The determined values have been compared to theoretical born approximations, and rather good agreement has been obtained.

An automatic insertion device for counting and processing large numbers of samples is being assembled. Manual counting operations currently in use involve changing samples, making necessary changes in analyzer settings, accumulating data, and recording data on magnetic tape for computer processing.

MATHEMATICAL TREATMENT OF MASS TRANSPORT

An attempt has been made to calculate the simultaneous release of different strontium isotopes at high temperatures in the framework of the one-dimensional diffusion equation. Convergent results have not yet been obtained because of the strong effect of the temperature on the fuel particle release rates and on the transport coefficients. Consequently, there is a rapid change in the fission product concentration with time and with space coordinates. This problem should be solved by a further debugging effort. A problem that remains is the strong effect of the boundary conditions for the current and for pressure (both are continuous in nature) on the concentration and current profiles.

For use with the FIPER code, a routine called SLIDER (Spherical Layer Isotopic Diffusionally Evaluated Release Model) is being constructed.

Although fission product release from particles has previously been considered using release constants, it is believed that release is controlled by diffusion through the kernel or the coating. A numerical code routine is being written in which diffusion is limiting in release. This code will serve as an aid in analyzing fission product release experiments and as a fission-product source term in FIPER calculations. The code is necessarily complex because of the multizone diffusion model presented by a spherical coated particle. The problem, therefore, is more amenable to numerical solution in a manner similar to the FIPER code than to analytical solution.

FISSION PRODUCT VAPORIZATION AND SORPTION STUDIES

Vaporization of Metallic Fission Products from Graphite

Studies of fission product vaporization from graphite using the mass spectrometric Knudsen cell method have concentrated on diagnostic experiments designed to determine the degree of sorption by Knudsen cell walls. These experiments consist of stopping a normal metal effusion experiment (described in the previous quarterly report) after effusing about 95% of the metal from the graphite. After opening the cell and removing the graphite, a new cover with the same size orifice is put on and the cell is replaced in the Knudsen cell mass spectrometer furnace. The cell is heated, and the loss of the sorbed metal from the cell walls is followed. At the conclusion of the experiment, the integral of the ion signal multiplied by the square root of the absolute temperature over both effusion periods is determined and the amount of metal associated with the graphite determined. The conversion of the ion signal to pressure is accomplished using a single sensitivity factor for the two effusion periods. A mass balance is used to determine the amount of metal associated with the wall. In these experiments, a considerable amount of strontium was associated with the tantalum cell walls; the problem is not so severe with barium and cesium.

Tantalum is the only material that has been used for Knudsen cells in these studies. It is possible that some other material would be less reactive, and other materials are being investigated. The requirements for the Knudsen cell material are (1) to maintain integrity to about 2500°K, (2) to contribute a negligible amount of metal loading to the graphite, and (3) to absorb a negligible amount of metallic elements, which should be associated only with the graphite.

An investigation of cell materials was made in a strontium vaporization experiment by including candidate cell materials with the loaded graphite powder. At the conclusion of the experiment, strontium associated with each of the tested materials (platinum, tantalum, molybdenum, and tungsten) was determined by neutron activation analysis. In this experiment, the respective sorption of the various metal foil samples (0.002 in.) was determined to be 70.2, 0.076, 0.0096, and 0.0011 mg Sr/g metal for platinum, tantalum, molybdenum, and tungsten. Tantalum, molybdenum, and tungsten should satisfy the integrity requirement, and as metals they should not quickly load the graphite unless the systems are at oxygen pressures at which MoO_3 or WO_3

are important gaseous species. The results of this experiment strongly recommend tungsten as a Knudsen cell material and fabrication of such a cell is being initiated.

Differences between the pseudo-isopiestic measurements and the Knudsen cell measurements of Cs sorption have been the subject of study. To investigate this problem, graphite has been loaded with Cs using the pseudo-isopiestic vapor loading technique instead of creating sorbed Cs metal by graphite reduction of CsNO_3 . Studies of the unloading rate will be made and analyzed according to the Knudsen equation to evaluate cell vapor pressures versus graphite loading.

Sorption of Cesium on Graphite

Cesium sorption measurements on SX4 graphite are in progress using the pseudo-isopiestic method described in the previous quarterly report. This graphite has been selected for use in thermal diffusion (Soret effect) experiments because of its "near-isotropic" structure. Consequently, it is of interest to measure the cesium sorption characteristics for comparison with HLM-85 and TS-688 graphites, for which cesium sorption data are available.

Cesium sorption runs at 900° and 1000°C have been completed and an additional run at 1100°C is in progress. Table 4.1 shows a comparison of the sorption isotherms for SX4, HLM-85, and TS-688 graphites.

Table 4.1
CESIUM SORPTION DATA FOR SX4, HLM-85, AND TS-688 GRAPHITES

Graphite ^a	Temp. (°C)	Cesium Loading (mg Cs/g C)			
		$P_{\text{Cs}} =$ 1×10^{-4} atm	$P_{\text{Cs}} =$ 1×10^{-5} atm	$P_{\text{Cs}} =$ 1×10^{-6} atm	$P_{\text{Cs}} =$ 1×10^{-7} atm
SX4	900	1.26	0.92	0.67	0.49
HLM-85	900	1.10	0.71	0.47	0.31
TS-688	900	0.82	0.56	0.38	0.25
SX4	1000	0.88	0.59	0.39	0.26
HLM-85	1000	0.69	0.39	0.22	0.12
TS-688	1000	0.69	0.41	0.24	0.15

^aComparative BET (nitrogen adsorption) surface areas for the 6 to 10 mesh samples used in these studies are SX4 = 0.48 m²/g C, HLM-85 = 0.70 m²/g C, and TS-688 = 0.82 m²/g C.

Additional cesium sorption measurements will be made in an attempt to resolve discrepancies in sorption data obtained by the pseudo-isopiestic method and by the mass spectrometric Knudsen cell method. The TS-688 graphite samples used in these studies varied considerably in surface areas and

particle sizes, e.g., 2000 to 3300 μ , 0.82 $\text{m}^2/\text{g C}$ (pseudo-isopiestic method); and 37 to 74 μ , 5.87 $\text{m}^2/\text{g C}$ (mass spectrometric method). Cesium pressures at 1100°C obtained by the mass spectrometric method are about a factor of 10 higher than those observed using the pseudo-isopiestic method at the same cesium concentration (surface area basis). This variation cannot be explained merely by the difference in surface areas of the two samples (about a factor of 7).

In an attempt to resolve the apparent discrepancy between the two experimental methods, a run is in progress whereby two Knudsen cells, each containing about 0.4 g of the 37- to 74- μ graphite, have been placed in the pseudo-isopiestic sorption apparatus and are being loaded with stable cesium at 1000°C. After about 10 days, when the system should be at equilibrium, the cells will be removed. One cell will be used to obtain sorption data using the mass spectrometer, and the other will be cut open to determine the cesium concentration in the graphite.

Study of the Cesium-graphite System at Low Concentrations

Sorption measurements are in progress to study the cesium-graphite (TS-688) system at 1000°C over a cesium concentration range of about 10^{-3} to 1 mg Cs/g C. Previous sorption data show that Freundlich isotherm behavior is observed over a concentration range of about 0.08 to 0.8 mg Cs/g C. The purpose of the current study is to determine the behavior of the system at the lower cesium loadings and vapor pressures that would be found in an HTGR. A sorption run at 1000°C was terminated after 71 days when it became apparent that further reductions* in the cesium concentration in the graphite could not be detected. The high level of activity on the steel furnace tube completely obscured the activity in the specimen.

Determination of the cesium in the specimen at the conclusion of the run showed a concentration of 0.052 mg Cs/g C (0.41 $\mu\text{mole Cs}/\text{m}^2$ BET area). An extrapolation of a previously 1000°C sorption isotherm obtained indicated an equilibrium cesium pressure of 6×10^{-10} atm at this equivalent cesium loading. (The last equilibrium pressure measurement observed before termination of the present run was 6.5×10^{-10} atm.) It appears, therefore, that this run yielded data that agree quite well with previous sorption data for TS-688 graphite at 1000°C and verified that Freundlich behavior is observed over a cesium concentration range of about 0.05 to 0.9 mg Cs/g C. (Assuming a surface area of about 0.5 $\text{m}^2/\text{g C}$ for bulk graphite, these values would correspond to about 0.025 to 0.45 mg Cs/g of bulk graphite.)

Another run is in progress to extend the sorption data to lower cesium concentrations and pressures at 1000°C. The experimental procedure has been modified in an attempt to reduce the background level of activity on the steel furnace tube.

*The specimen was first loaded with Cs at a relatively high pressure. This is the normal procedure to reduce run time.

FISSION PRODUCT DIFFUSION STUDIES

Diffusion of Metallic Fission Products in Graphite

The experimental technique used in thermal diffusion (Soret) experiments has been modified by encapsulating the graphite specimen to reduce or eliminate the effects of vapor transport of the metallic species. Several methods of encapsulating or coating the graphite were tried but only one, the vapor deposition of tungsten, proved to be successful. To test the encapsulation, a tungsten-coated graphite rod that had been loaded with BaC_2 prior to coating was kept in a uniform heat zone at about 1500°C for 100 hr. No barium permeated the tungsten coating.

Two graphite rods (extruded SX4 graphite, 9 cm long and 1.55 cm in diameter; one machined in the extrusion direction, the other 90° to the extrusion direction) were loaded with about $10\text{ }\mu\text{g}$ of Ba^{133} carbide by placing the BaC_2 in a hole in the specimen and closing the opening with a threaded graphite plug. The specimens were coated with tungsten and kept in a uniform heat zone at $1500 \pm 10^\circ\text{C}$ for about 400 hr. At intervals, the samples were removed from the furnace and the diffusion of barium in the axial direction was measured by gamma spectrometry. Concentration profiles were obtained as a function of time, and barium diffusion coefficients are being calculated. The specimens are now being held at temperatures above 1600°C until a uniform concentration profile has been established. They will then be subjected to a temperature gradient to study the Soret (thermal diffusion) effect.

The optimal conditions for producing oxygen-free barium have been established. BaO was heated with graphite in a graphite Knudsen cell with a very small orifice. The reaction was carried out at 1100°C and pressures of about 10^{-5} atm for 3 hr. About 20% of the barium originally present was lost. Completion of the reaction was established by X-ray analysis. Following this procedure, about 2 mc of $\text{Ba}^{133}(\text{NO}_3)$ were converted into BaC_2 . Experiments studying the optimal conditions for producing oxygen-free strontium are under way.

Diffusion of Metallic Fission Products in Pyrolytic Carbon

Basic studies of the diffusion of metallic fission products in pyrolytic carbon are being carried out to obtain an understanding of the mechanism that governs the release of these elements from pyrolytic-carbon-coated fuel particles.

In these studies, $100\text{-}\mu$ layers of pyrocarbon are deposited on small graphite wafers (5/16 in. in diameter by 1/16 in. thick) by the same techniques used to coat fuel particles. For the diffusion anneal, the wafers are packed firmly into a bed of graphite powder impregnated with the element of interest (tagged with a suitable radioactive tracer). In this manner, a constant chemical potential source of the desired level is attained.

An alternative approach has been developed using a self-diffusion technique. With this procedure, samples of pyrocarbon are uniformly loaded with

a predetermined concentration of a fission product metal. Self-diffusion is then measured using a radioactive tracer. This procedure eliminates the effect of concentration on diffusivity. Metals that diffuse readily, such as strontium, are amenable to this approach. Diffusion coefficients determined in this manner should be more fundamental and useful for determining the mobility of metallic fission products in pyrolytic carbon coatings.

Barium Self-diffusion Experiments. Self-diffusion of barium in pyrolytic carbon was measured at a loading of 10^{-5} g Ba/g C. Pyrolytic carbon coatings were first saturated with barium at the desired loading, using Ba^{133} as a tracer to verify that saturation was achieved. Self-diffusion was then measured using a constant chemical potential source of 1.0×10^{-5} g Ba/g C using Ba^{140} as a tracer. Self-diffusion coefficients are shown in Table 4.2.

Table 4.2
SELF-DIFFUSION OF BARIUM IN PYROLYTIC CARBON^a

Temp. (°C)	D (cm ² /sec)
1000	2.7×10^{-11}
1200	4.2×10^{-10}
1400	4.3×10^{-9}
1600	2.7×10^{-8}
1700	7.4×10^{-8}

^aAt a loading of 10^{-5} g Ba/g C.

An Arrhenius plot (D versus $1/T$ °K) yielded the following equation for the self-diffusion of barium in pyrolytic carbon:

$$D_{\text{Ba}} = 9.5 \times 10^{-2} \exp \left(-\frac{55,800}{RT} \right) \text{ cm}^2/\text{sec} \quad (1)$$

The temperature dependence ($-55,800/RT$) of the self-diffusion coefficient differs considerably from that obtained for chemical diffusion of barium in pyrolytic carbon from a constant chemical potential source of 1.0×10^{-5} g Ba/g C, which is given by

$$D_{\text{Ba}} = 3.3 \times 10^{-4} \exp \left(-\frac{37,000}{RT} \right) \text{ cm}^2/\text{sec} \quad (2)$$

The data for both studies are shown in Fig. 4.1. Reasons for the differences are not clearly understood but may be within the precision of the data. Since the self-diffusion coefficient approaches the chemical diffusion coefficient as a limit at equal concentrations, it is difficult to

Fig. 4.1--Diffusion of barium in isotropic pyrolytic carbon with a constant chemical potential source of 1.0×10^{-5} g Ba/g C

understand how the self-diffusion coefficient can exceed the chemical diffusion coefficient.

Barium Chemical Diffusion Experiments. Measurement of the diffusion of barium into pyrolytic carbon from a constant chemical potential source of 1.0×10^{-2} g Ba/g C has revealed diffusion rates about two orders of magnitude higher than those from a source of 1.0×10^{-5} g Ba/g C (Fig. 4.2). Chemical diffusion coefficients measured over the temperature range 800° to 1200°C are listed in Table 4.3.

Table 4.3
CHEMICAL DIFFUSION COEFFICIENTS FOR BARIUM
IN PYROLYTIC CARBON^a

Temp. (°C)	Diffusion Coefficients (cm ² /sec)
800	1.6×10^{-12}
900	5.7×10^{-10}
1000	1.1×10^{-8}
1200	9.9×10^{-8}

^aFrom a constant chemical potential source of 1.0×10^{-2} g Ba/g C.

An Arrhenius plot (D versus $1/T$ °K) yielded the equation

$$D_{\text{Ba}} = 1.2 \times 10^6 \exp \left(- \frac{85,000}{RT} \right) \text{ cm}^2/\text{sec} .$$

In contrast to the data at lower loadings, a self-diffusion run using the same constant chemical potential source of 1.0×10^{-2} g Ba/g C at 1000°C yielded the diffusion coefficient 1.2×10^{-8} cm²/sec, which is almost identical with that of 1.1×10^{-8} cm²/sec obtained for chemical diffusion.

Europium Diffusion Experiments. Both chemical diffusion and self-diffusion of europium in pyrolytic carbon have been measured at a level of 1.9×10^{-3} g Eu/g C over the temperature range 800° to 1200°C. The diffusion coefficients obtained are shown in Table 4.4.

Comparison of these data shows that, within experimental error, the chemical diffusion and self-diffusion coefficients for this element in pyrolytic carbon are the same at this loading. An Arrhenius plot of the self-diffusion data yields the equation

$$D_{\text{Eu}} = 0.54 \exp \left(- \frac{45,700}{RT} \right) \text{ cm}^2/\text{sec} .$$

Fig. 4.2--Chemical diffusion of barium in isotropic pyrolytic carbon

Table 4.4
DIFFUSION OF EUROPIUM IN PYROLYTIC CARBON^a

Temp. (°C)	Diffusion Coefficient (cm ² /sec)	
	Chemical Diffusion	Self-diffusion
800	1.6×10^{-10}	---
1000	5.4×10^{-9}	6.7×10^{-9}
1100	4.4×10^{-8}	6.7×10^{-8}
1200	6.6×10^{-8}	7.5×10^{-8}

^aUsing a constant potential source of
 1.9×10^{-3} g Eu/g C.

Diffusion anneals are currently in progress to determine diffusion coefficients using a constant potential source of 6.0×10^{-7} g Eu/g C. In the initial experiment, a chemical diffusion coefficient of 6.0×10^{-9} cm²/sec has been obtained at 1250°C.

Application of Diffusion Equations to Fission Product Release Data

Attempts are being made to apply diffusion equations to fuel particle fission-product release data. It has been found that, in general, the release data can be fitted to the diffusion equations for a sphere. An example of this fit is shown in Fig. 4.3. The points shown in Fig. 4.3 are measured values for the release of Sr⁸⁹ from pyrocarbon-coated particles (lot 2813-141E) irradiated in the P11 capsule. The curves represent solutions of the diffusion equation for a sphere with constant diffusivity. The high fractional release values are fitted by an exponential time dependence, and the low fractional release values are fitted by a square-root-of-time dependence. The results are given in Table 4.5, in which D is the diffusion coefficient and r is the radius of the kernel.

Table 4.5
DIFFUSION-CONTROLLED RELEASE OF
FISSION PRODUCTS FROM FUEL PARTICLES

<u>T (°K)</u>	<u>D/r² (sec⁻¹)</u>
2073	1.5×10^{-6}
1773	2.4×10^{-7}
1573	7.8×10^{-9}
1373	2.1×10^{-9}

Figure 4.4 is an Arrhenius plot of the diffusion data. The curve was fitted to the data in Table 4.5 by the least-squares method. The activation energy is 56 kcal/mole.

Fig. 4.3--Fit of diffusion equations to experimental data for Sr^{89} release from pyrocarbon-coated particles. The curves represent solutions to the diffusion equation and the points are experimental data

Fig. 4.4--Arrhenius plot for diffusion-limited release of Sr^{89} by pyrocarbon-coated particles

Using a value of 1.5×10^{-2} cm for the radius of the particles in the release experiment, one derives diffusion coefficients that are lower by about 10^3 than diffusion coefficients obtained earlier in studies of the diffusion of strontium in pyrolytic carbon. Possible explanations for the difference in the two sets of diffusivities are (1) concentrations of strontium in the pyrocarbon may have been greatly different in the two cases, and (2) release of strontium from the particles may have been controlled by diffusion in the carbide kernel rather than diffusion in the pyrocarbon coating. The latter explanation is consistent with fission-product release studies.

Prediction of Diffusion Coefficients for Transport of Fission Products in Isotropic Pyrocarbon

Diffusion coefficients of several fission product nuclides have been measured in isotropic pyrocarbon as an aid in evaluating fission-product release from HTGR fuel elements. With these data, it is worthwhile to consider the possibility of predicting diffusion coefficients for the transport of certain nuclides in fuel coating materials. This is especially important for nuclides that are difficult to study experimentally (short-lived or low-permeability nuclides).

Diffusion data have been obtained for the transport of Cs, Sr, Ba, Eu, and Th in isotropic pyrocarbon, and these data can be used to establish a procedure for the prediction of unknown diffusion coefficients. The first four nuclides were studied using a constant chemical potential source, while the Th was studied using the plane source technique. In all cases, the matrix was vapor-deposited on graphite disks.

The experimental diffusion data are fitted to the Arrhenius equation,

$$D = D_0 \exp(-E^*/RT) \quad , \quad (1)$$

where D = the diffusion coefficient (cm^2/sec)

D_0 = an empirical constant (cm^2/sec)

E^* = the activation energy for diffusion (cal/mole)

R = the gas constant ($\text{cal/mole-}^\circ\text{K}$)

T = the temperature ($^\circ\text{K}$).

Equation (1) may be rewritten as

$$\log_{10} D = A - BT^{-1} \quad , \quad (2)$$

where $A = \log_{10} D_0$

$B = E^*/2.303R$.

The objective of this study is to evaluate methods, either empirical or based on theory, of predicting the constants A and B for a particular nuclide. The bases for this approach are the Arrhenius equation, the "compensation law," and an empirical linear relation between the activation energy E^* and the reciprocal atomic radius r^{-1} .

The compensation law states that, for many kinetically controlled processes that satisfy the Arrhenius temperature dependence, the constants A and B are linearly related. From absolute reaction rate theory, A and B are essentially entropy and enthalpy terms,

$$A = C + \Delta S^*/R \quad \text{and}$$

$$B = \frac{\Delta H^* - p\Delta V^*}{2.303R} \quad ,$$

where C is a "constant," ΔS^* is the activation entropy, ΔH^* is the activation enthalpy, and $p\Delta V^*$ is the activation work, which may be assumed to be zero for diffusion in solids. Although no relation between A and B can be derived from thermodynamics, an atomistic or phenomenological consideration can yield such a relation. This question has been reviewed by Ruetschi (Ref. 1). For the purpose of the present study, the compensation law may be written as

$$A = a + bB \quad , \quad (3)$$

where a and b are empirical constants. The use of the compensation law to describe the diffusion of various nuclides in molten silicates has been discussed by Norman and Winchell (Ref. 2).

The third relation used is a linear relation between B and the reciprocal atomic radius r^{-1} of the diffusion species,

$$B = c + dr^{-1} \quad , \quad (4)$$

where r is in angstroms units and c and d are empirical constants. The effectiveness of such a relation in describing the entropy (or energy) term for diffusion of a number of ions in molten silicates has been demonstrated by Norman and Winchell (Ref. 2). Equation (4) is not easily justified on theoretical grounds although the ionic potential Zr^{-1} (Z = charge, r = radius) has been used to describe bond strengths in molten salts and minerals.

Employing Eq. (4) as an empirical relation and substituting Eqs. (2) and (3), one obtains

$$\log_{10} D = m + nr^{-1} - (c + dr^{-1}) T^{-1} \quad , \quad (5)$$

where $m = a + bc$ and $n = bd$. Equation (5) is used to predict the diffusion coefficients of several species in isotropic pyrocarbon. Initially, only the data for Cs, Ba, and Sr are considered, and the coefficients of Eq. (5) are obtained from the Arrhenius equations describing the experimental data. Diffusion coefficients calculated from Eq. (5) are then compared with those obtained from Eq. (1). The excellent fit obtained is shown in Table 4.6.

Table 4.6

COMPARISON OF EXPERIMENTAL DIFFUSION COEFFICIENTS
OBTAINED FROM EQ. (1) AND THOSE OBTAINED FROM EQ. (5)

Species	Temp. (°K)	Diffusion Coefficient (cm ² /sec)	
		Eq. (5)	Eq. (1)
Sr	1273	3.8×10^{-10}	2.5×10^{-10}
Sr	1573	9.8×10^{-9}	7.1×10^{-9}
Sr	1973	1.6×10^{-7}	1.6×10^{-7}
Ba	1273	9.3×10^{-11}	1.4×10^{-10}
Ba	1573	2.3×10^{-9}	2.3×10^{-9}
Ba	1973	3.8×10^{-8}	2.6×10^{-8}
Cs	1273	6.2×10^{-14}	6.0×10^{-14}
Cs	1573	1.3×10^{-12}	1.4×10^{-12}
Cs	1973	1.9×10^{-11}	2.1×10^{-11}

Over the temperature range 1273° to 1973°K, Eq. (5) adequately describes the data reported for Sr, Ba, and Cs within a maximum uncertainty of 52% in the diffusion coefficient. The constants in Eq. (5) are then used to predict diffusion coefficients for Eu. In Table 4.7 the predicted values are compared with the experimental values, and the agreement is seen to be excellent.

Table 4.7

DIFFUSION COEFFICIENTS OF EUROPIUM

Species	Temp. (°K)	Predicted Value	Experimental Value
Eu	1273	4.3×10^{-9}	5.4×10^{-9}
Eu	1473	4.5×10^{-8}	6.6×10^{-8}

The coefficients in Eq. (5) are then refined by including the experimental data for Eu, and the values of the coefficients thus obtained are given in Table 4.8.

Table 4.8

CONSTANTS IN EQ. (5) OBTAINED FROM EXPERIMENTAL DATA

Coefficient	Value
m	-25.9
n	52.3 Å
c	3846 °K
d	13230 Å °K

The data for Cs, Sr, Ba, and Eu appear to be reasonably described by Eq. (5) using the coefficients given in Table 4.8. A comparison of the experimental and calculated values is shown in Fig. 4.5. As more data becomes available, the coefficients in Table 4.8 will be refined.

The constants in Table 4.8 may now be used to predict unknown diffusion coefficients, and this has been done for the transport of Ag and Ce in isotropic pyrocarbon. The predicted diffusion coefficients at three temperatures are given in Table 4.9.

Table 4.9
PREDICTED DIFFUSION COEFFICIENTS OF SILVER
AND CERIUM IN ISOTROPIC PYROCARBON

Temp. (°K)	Predicted Value	
	Ag	Ce
1073	2.0×10^{-2}	4.0×10^{-8}
1273	1.6	1.6×10^{-6}
1473	40 ^a	2.5×10^{-4}

^aThere is some uncertainty associated with such a large extrapolated value. It appears that a change in mechanism might occur before a value of this magnitude could be attained.

Diffusion coefficients calculated for Th transport in isotropic pyrocarbon do not agree with those obtained experimentally. At present, this disagreement cannot be readily explained. It is possible that the diffusion mechanism for Th is different from that of the other four nuclides. It is also possible that the constant chemical potential source can yield results which differ from those obtained from the instantaneous plane source (Ref. 3). This problem will be considered further as more data become available.

Phenomenology of the Release of Ba and Sr from PyC-coated Particles

An understanding of the mechanism of fuel particles fission-product release is of importance in estimating the value of various coating materials. Accordingly, studies have been undertaken to analyze some of the release data for isotropic pyrolytic carbon-coated fuel particles in order to clarify the release mechanisms.

Earlier studies indicate that the release of Sr by pyrolytic carbon-coated fuel particles is governed by diffusion in the (Th,U)C₂ kernel rather than by diffusion through the coating. The results of the present study support this mechanism for both Sr and Ba release.

The data used in this study are from a previous experiment on Ba and Sr release from BISO (2200°C) particles. The release is assumed to be diffusion limited, either by the fuel matrix or by the isotropic pyrocarbon

Fig. 4.5--Comparison of calculated and experimental diffusion coefficients for transport in isotropic pyrocarbon

coating. The buffer region is not considered to be a diffusion barrier. Thus, the model is that of a sphere bounded by a spherical shell. The dimensions of the fuel particles and coatings are such that the mathematical treatment of the "equivalent slab" is a close approximation to the spherical shell (Ref. 4).

The release data are fitted by the least squares method to the empirical relation

$$f = 1 - (C/C_0) = a + b\sqrt{t} \quad , \quad (6)$$

where f is the fractional release, t is the time (sec), and a and b are constants. The constant a represents a lag period that was observed in plots of the data. The results of the least-squares treatment are shown in Table 4.10 and in Figs. 4.6, 4.7, and 4.8, where f is plotted against $\sqrt{t}$.

The lag period observed for both Ba and Sr is of considerable phenomenological interest. It can be interpreted as resulting from the lag inherent in diffusion through the spherical shell (or equivalent slab) (Ref. 5). From the coating thicknesses and lag periods, it is possible to calculate approximate values of diffusion coefficients using the equation

$$D \approx x^2/4t \quad , \quad (7)$$

where D is the diffusion coefficient (cm^2/sec) and x is the coating thickness (cm). These results are compared in Table 4.11 with experimental diffusion coefficients for Ba and Sr in isotropic pyrolytic carbon at 1400°C . There is good agreement between the calculated and reported diffusion coefficients in both cases.

For longer times, the fractional release data are then interpreted to be diffusion-limited by the fuel matrix. From the data given in Table 4.10, diffusion coefficients are calculated for the transport of Ba and Sr in the fuel matrix at 1400°C using the equation

$$f = 1 - (\bar{C}/C_0) \approx \frac{6}{R} \left(\frac{Dt}{\pi} \right)^{1/2} \quad , \quad (8)$$

where the radius R is taken as 2×10^{-2} cm. This equation is expected to govern the low-level release of these nuclides from the spherical kernel. The results are given in Table 4.12.

The average value found for Ba ($D = 1.4 \times 10^{-12}$) is higher than that reported for the transport of Ba in monogranular UC_2 particles at 1400°C ($D = 8.0 \times 10^{-14}$) (Ref. 6). The difference is large but it is reasonable considering the multigranular nature of the $(\text{Th,U})\text{C}_2$ matrix used in release experiments.

Fig. 4.6--Fractional release from PyC-coated fuel particles (lot 3222-145E) at 1400°C

Fig. 4.7--Fractional release from PyC-coated fuel particles (lot 3363-147E) at 1400°C

Fig. 4.8--Fractional release from PyC-coated fuel particles (lot 3363-149E) at 1400°C

Table 4.10

DIFFUSION-LIMITED RELEASE OF Ba AND Sr FROM FUEL PARTICLES

Particle Lot No.	Fission Product	Lag (sec)	Slope $\times 10^4$	Coating Thickness (μ)
3222-145E	Ba	2400	2.35	67
3222-145E	Sr	0	8.11	67
3363-147E	Ba	7040	1.06	86
3363-147E	Sr	1155	8.22	86
3363-149E	Ba	5700	2.38	67
3363-149E	Sr	625	10.57	67

Table 4.11

COMPARISON OF CALCULATED AND REPORTED DIFFUSION COEFFICIENTS D AT 1400°C FOR ISOTROPIC PYROCARBON

Particle Lot No.	Fission Product	Diffusion Coefficient (cm ² /sec)	
		Calculated	Experimental
3222-145E	Ba	4.7x10 ⁻⁹	---
3222-145E	Sr	---	---
3363-147E	Ba	3.2x10 ⁻⁹	---
3363-147E	Sr	9.7x10 ⁻⁹	---
3363-149E	Ba	1.6x10 ⁻⁹	---
3363-149E	Sr	2.9x10 ⁻⁸	---
Avg	Ba	3.2x10 ⁻⁹	4.8x10 ⁻⁹
Avg	Sr	2.3x10 ⁻⁸	2.3x10 ⁻⁸

Table 4.12

CALCULATED DIFFUSION COEFFICIENTS FOR TRANSPORT OF Ba AND Sr IN (Th,U)C₂ AT 1400°C

Particle Lot No.	Fission Product	Diffusion Coefficient (cm ² /sec)
3222-145E	Ba	1.9x10 ⁻¹²
3222-145E	Sr	2.3x10 ⁻¹¹
3363-147E	Ba	0.4x10 ⁻¹²
3363-147E	Sr	2.4x10 ⁻¹¹
3363-149E	Ba	2.0x10 ⁻¹²
3363-149E	Sr	3.9x10 ⁻¹¹
Avg	Ba	1.4x10 ⁻¹²
Avg	Sr	2.9x10 ⁻¹¹

Calculation of Diffusion Coefficients for Transport of Sr^{89} in $(\text{Th,U})\text{C}_2$ Fuel Matrix

Attempts have been made recently to treat experimental fission-product release data according to theoretically derived relations in an effort to evaluate the mechanism of release and to provide a means of predicting the release of a given sample a priori. The following discussion compares the effectiveness of "release constant" treatment of fission product release data with a "diffusion-limited" treatment.

The data reported for the release of fission products from fuel particles are frequently treated according to the equation

$$f = 1 - \exp(-Rt) \quad , \quad (9)$$

where f is the fractional release, R is the release constant (hr^{-1}) and t is the time (hr). With respect to fractional retention ($1 - f = e^{-Rt}$), R is analogous to a first-order reaction rate constant and would be expected to exhibit Arrhenius temperature dependence. Two other analogs are Newton's law of cooling and the discharge of a simple RC circuit.

Recent considerations of fission-product release data have shown that these data may also be interpreted by assuming diffusion-limited transport in the fuel matrix or in the isotropic pyrocarbon coatings. Although the release constant treatment accurately describes the experimental data for large values of f , the accuracy becomes poor for low and intermediate values of f . In comparison, release data are accurately described over the entire range of f when diffusion equations are employed to treat the data. An example of this is shown in Fig. 4.9, where isothermal data for Sr^{89} release from particles have been treated according to the release constant method as shown in Eq. (9), and also by application of the diffusion equation to the release from a sphere. For small values of f ,

$$f \approx (6/R') (Dt/\pi)^{1/2} \quad , \quad (10)$$

where R' is the radius (cm) and D is the diffusion coefficient (cm^2/sec). The two approaches should be compared on the basis of the departure of the experimental data from the predicted linear relations. Equation (10) accurately describes the experimental data over the entire experimental time range. However, the experimental data, when treated using Eq. (9), show nonlinearity that increases rapidly with increasing time. The slope of the solid line is the value of R ($9.4 \times 10^{-3} \text{ hr}^{-1}$), which was obtained by evaluating this constant at 20 hr using the experimental data and Eq. (9). The two methods are also compared in Table 4.13.

The diffusion equation approach appears to have more merit than the release constant approach, not only because it provides more accurate representation, but also because the diffusion mechanism (fuel matrix or coating release limitation) can often be inferred. Although fitting of

Fig. 4.9--Comparison between the release constant treatment (solid circles) and the diffusion equation treatment (open circles) of experimental data for the release of Sr^{89} from fuel particles at 1250°C

Table 4.13

COMPARISON OF THE RELEASE CONSTANT AND THE DIFFUSION EQUATION TREATMENTS FOR RELEASE OF Sr^{89} BY $(\text{Th},\text{U})\text{C}_2$ AT 1250°C AFTER 80 HR

Description	f	Error (%)
Release constant	0.52	85
Diffusion equation	0.27	4
Experimental data	0.28	--

data to diffusion equations does not constitute a proof that diffusion is the rate-limiting process, the evidence that this is indeed the case is supported by the following arguments:

1. In all cases attempted, experimental release data have been amenable to treatment using diffusion equations.
2. Diffusion equations can be successfully applied to the data over the entire range of f, which requires two very different equations for the diffusion from a sphere, an exponential time dependence for large values of f and a square-root-of-time dependence for small values of f.
3. Diffusion coefficients in isotropic pyrocarbon obtained from release data for isotropic pyrocarbon-coated particles at early times or for low values of f are in good agreement with values obtained by direct measurement.
4. The extrapolation of experimental data exhibiting exponential time dependence at large values of f (Eq. (3)) yields reasonable intercepts at zero time.

Diffusion coefficients for transport of Sr^{89} in the $(\text{Th},\text{U})\text{C}_2$ matrix have been obtained from a diffusion equation treatment of recent data on the release of Sr^{89} from isotropic pyrocarbon-coated particles. The experimental data are compared with the results from application of the diffusion equations in Fig. 4.10. In processing these data, it was found that the 1100°C isothermal data could be attributed entirely to contamination in the PyC coating and that diffusion of fission products from the kernel is lower by an order of magnitude. Using the isotropic pyrocarbon thickness and the release data, the value of $5.9 \times 10^{-10} \text{ cm}^2/\text{sec}$ is calculated for the diffusion coefficient of Sr in isotropic pyrocarbon at 1100°C , which is in good agreement with the experimentally determined value of 9.9×10^{-10} at the same temperature (Ref. 3). The isothermal releases at 1250° , 1500° , and 2000°C , can only be interpreted as diffusion-limited transport in the $(\text{Th},\text{U})\text{C}_2$ fuel matrix. A diffusion coefficient for Sr in a similar matrix at 1400°C was obtained in another study, and this value has been included in the data used for least-squares fitting to the Arrhenius equation. These results, which are shown in Fig. 4.11, are summarized by the equation

Fig. 4.10--Fit of fractional release data to diffusion equations for release of Sr^{89} from isotropic pyrocarbon-coated particles. Curves represent solutions of the diffusion equations and points are experimental data

Fig. 4.11--Diffusion coefficients for Sr^{89} transport in $(\text{Th},\text{U})\text{C}_2$ fuel

$$(\log_{10} D) \pm 0.21 = -3.51 - (1.14 \times 10^4) T^{-1} \quad (11)$$

It is emphasized that these diffusion coefficients must depend somewhat on the polygranular nature of the fuel matrix and that Eq. (11) should be used only as a guide until this effect is understood. The activation energy calculated from Eq. (11) is 52 kcal/mole, which is in fair agreement with the value of 56 kcal/mole obtained from an earlier study of Sr release from isotropic pyrocarbon-coated particles.

MICROPROBE EXAMINATION OF IRRADIATED FUEL PARTICLES

Electron microprobe examination of irradiated coated fuel particles have been initiated. Intact particles that have attained burnups of 37% FIFA at 1300°C and 65% FIFA at 900°C have been selected for preliminary investigation. At this burnup level the fuel particles will have generated 1 to 2 wt-% of each of several stable fission elements of interest, for example, Cs, Ba, La, Sr, Mo, Zr, and Ce. In addition, there should be several tenths of one percent of the neutron-produced heavy elements Pu, Np, and Pa.

Since an appreciable amount of effort has been invested in the preirradiation examination of HTGR fuel particles, it is felt that a postirradiation probe examination of these particles might lead to a better understanding of the behavior of the particles under irradiation. Probe examination of irradiated particles offers a direct approach to the migration behavior of the heavy elements and those fission elements of interest.

Tentative results from the examination of one set of particles appear promising. Thorium and uranium values have been obtained. While the material balances appear low with respect to unirradiated material, the difference may be made up in part by the fission elements (the composite contribution could be about 10%). Other effects that might contribute to this are surface preparation, porosity, and irradiation effects. Elements that have been detected above background and line interferences are Zr, Mo, Ce, and possibly I.

Electron microprobe analysis of irradiated fuel particles is apparently feasible and promising. Additional efforts will be directed toward the refinement of thorium-uranium analysis, and an extensive attempt will be made to detect such other stable fission elements as Sr, Cs, Ba, and La. However, the probe is unshielded and perhaps the signal for the elements may be masked by the increase in background radiation. Future sample analyses will be performed with older, cooler particles to help reduce some of the variables associated with the hot particles. The instrument is currently undergoing overhaul and readjustment, which may further enhance elemental sensitivity.

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Task V
HTGR RECYCLE STUDIES

THE EFFECTS OF FUEL ELEMENT DESIGN ON REPROCESSING

Reprocessing Facility for Bonded Particle Fuel Elements

Preliminary costs and process details have been determined for a central reprocessing facility that can treat 0.5 tonne/day of heavy metal in hexagonal-block fuel elements containing bonded fuel particles. Two head-end methods, selective oxidation (chemical method) and brush abrasion (mechanical method), were considered. The purification and recovery of uranium and thorium is accomplished by conventional solvent extraction (thorex). The capital costs associated with the head-end operations are nearly identical within the accuracy of the study for both the chemical and the mechanical methods. The total reprocessing cost (which includes fuel receiving and storage, fuel particle debonding, fuel particle separation, fertile particle leaching, fissile particle storage, fission-product decontamination, uranium and thorium recovery, and waste storage charges, for a central facility incorporating either of the head-end methods) is \$61/kg of heavy metal. These costs, which are very preliminary in nature, assume a 75% reprocessing plant operating factor. In addition, the total plant capital cost and the startup cost are amortized over 15 years, the waste storage tanks being amortized over 3 years.

This cost compares favorably with one obtained in a previous study (\$62/kg heavy metal) for a 0.5-tonne/day throughput central facility for processing fuel elements with unbonded particles. Because of the accuracy of this and former cost studies, however, only a limited importance should be given to this agreement. Although the costly addition of a debonding step to release the fissile and fertile particles from the matrix binder of a fuel block was encountered in this study, a reassessment of the process requirements and equipment costs for the head-end operations resulted in a 0.5-tonne/day plant with a slightly less expensive plant capital cost.

Binder Removal Studies

Debonding studies are presently directed toward the evaluation of gaseous oxidation and mechanical debonding processes. Candidate processes are evaluated by comparing the rate of yield of debonded particles, the extent of cross-contamination between the fissile and fertile sieve fractions (although processes other than sieving might be used to separate debonded particles, e.g., elutriation), and the general practicality of the process.

Gaseous Debonding. One of the most promising methods for recovering coated particles from a bonded bed in a graphite fuel element is a gaseous process employing O_2 , air, or steam. Selective oxidation of the carbonaceous

bonding material allows the coated particles to be separated from the fuel element without damage to the particle coatings.

In this process the gas is introduced into a bonded bed by means of a follower nozzle or into a general reaction chamber where there is no attempt to localize the gas flow. The sample is heated by an external heater. The sample is positioned open-end down, so that the particles fall into a cooled area as they are loosened.

In previous experiments that did not employ a gas injection nozzle, air, O_2 , and steam were used over a total range of 550° to 1000°C on beds 1/2-in. in diameter by 1-in. long held in 1-in.-diameter TS-688 graphite tubes. The matrix of the samples contained furfuryl alcohol and BC-135 charcoal. The bonded beds were carbonized to 900°C in all tests. Using O_2 , air, and steam in sequence required successively higher reaction temperatures, resulted in successively lower yield rates, and resulted in successively larger amounts of unoxidized binder and agglomerates of coated particles mixed with the debonded particles. The highest debonding rate yielding clean, undamaged particles was 0.23 g/min (equivalent to 0.036 in. of fuel hole per minute), which was achieved in O_2 at 600°C.

In another early experiment, the bonded bed was heated in air with an O_2 inlet tube held in close proximity to the receding surface of the bonded bed (forced circulation). The O_2 flow was 2000 cm³/min over the 1/2-in.-diameter by 6-in.-long bonded bed. The coated particle yields were determined at various temperatures. The highest debonding rate yielding clean, undamaged particles was 1.03 g/min (equivalent to 0.16 in. of fuel hole per minute) at 600°C. The use of a jet nozzle to direct the gas into the hole increased time efficiency by a large factor, but a non-nozzle system might be used at the expense of greater time.

Instead of heating the block, the gas may be preheated, or they both could be heated. This variable has been investigated.

During the past quarter, the effects of O_2 flow rate and the use of different binder systems were evaluated. A follower nozzle was used in all tests.

The effects of O_2 flow rate were evaluated using specimens with a matrix containing carbonized furfuryl alcohol and BC-135 charcoal. Two tests were conducted at 600°C in addition to the previous test described above. The results are summarized in Table 5.1. The debonding rate was essentially unaffected in going from 300 to 2000 cm³/min of O_2 . Using a flow rate of 3000 cm³/min doubled the debonding rate.

Debonding rates have been determined for four different binder systems in air and O_2 . The investigation of furfuryl alcohol and R72S phenolic resin in O_2 has been discussed in previous quarterly reports. During this quarter, R72S in air, an epoxy, dicyclopentadiene dioxide (DCPO₂) in air and O_2 , 5023 phenolic resin in air, and furfuryl alcohol in air have been investigated (see Table 5.2).

Table 5.1
THE EFFECTS OF O₂ FLOW RATE ON DEBONDING RATE

O ₂ Flow Rate (cm ³ /min)	Debonding Rate	
	(g/min)	(in. of fuel hole/min)
300	1.2	0.2
	0.82	0.12
2000	1.03	0.16
3000	2.3	0.4

NOTE: Specimen matrix contained carbonized furfuryl alcohol and BC-135 charcoal. Specimen was heated to 600°C in air; O₂ injected through a follower nozzle. Bonded bed was 1/2 in. diam, 1 in. long. Specimens contained in TS-688 graphite tubes.

It was found that with a phenolic or DCPO₂ binder, oxidation in O₂ caused a large evolution of heat, which caused extensive damage to the coated particles even at minimum debonding temperatures. Using R72S phenolic resin, the temperature could not be controlled at temperatures where debonding occurred, and fire caused the sample temperature to increase to 900° to 1200°C depending on O₂ flow rate. Using air with these systems allowed a controlled debonding at temperatures where no damage was done to the coated particles. The temperature at which debonding rate was measured was generally an optimum debonding temperature. Operation significantly below this temperature resulted in larger amounts of unoxidized matrix and agglomerates of coated particles and lower debonding rates. Operation significantly above this temperature caused damage to the coated particles. The reaction of the furfuryl alcohol system in O₂ began at the same temperature in experiments with and without the follower nozzle. However, when air was used, debonding began at 675°C without the nozzle and at 450°C with the nozzle. The test results in air with the nozzle appear to be incompatible with the other tests for unknown reasons.

The debonding rate was highest for DCPO₂ in air; however, the sample was uncontained, and a large surface area was exposed to oxidant. The presence of graphite flour in the 5023 system probably caused the low debonding rate.

It is considered that the general feasibility of gaseous oxidation to selectively oxidize binder matrix without attacking either the coated particles or the parent graphite has been demonstrated. All three gases examined appear to be effective for a furfuryl alcohol system, but O₂ appears most effective and would give much less residue gas than air. Air is apparently the best oxidizer for the other systems investigated, because of the rapid evolution of heat when O₂ is used and the consequent damage to the coated particle.

Table 5.2
EFFECT OF BINDER TYPE ON GASEOUS DEBONDING PROCESS

Binder (Carbonized)	Filler	Gas	Flow Rate (cm ³ /min)	Temperature for Initia- tion of De- bonding (°C)	Temperature at which Debonding Rate is Measured (°C)	Debonding Rate		Remarks
						(g/min)	(in. of fuel hole/min)	
Furfuryl alcohol ^a	BC-135	O ₂	300	500	600	1.2 0.82	0.2 0.13	No damaged particles; very little matrix
		Air	500	450 ^b	550 ^b	0.28 ^b	0.043 ^b	No damaged particles; very little matrix
R72S Phenolic resin ^a	BC-135	O ₂	500	460	---	---	---	Temperature uncontrolla- ble above 460°C; fire caused temperature to 1200°C; particles broken
		Air	750	460	500-540	0.17	0.026	No damaged particles; very little matrix
DCPO ₂ Epoxy ^c	BC-135	O ₂	300	400	---	---	---	Burning particles debond- ing rapidly; many bro- ken particles at 430°C
		Air	800	420	430-460	1.3	0.2	No damaged particles; very little matrix
5023 Phenolic resin ^c	GP-38 Graphite- flour	Air	800	500	520-600	0.077	0.012	No damaged particles; very little matrix

NOTE: O₂ was injected through a follower nozzle. Bonded bed dimensions: 1/2 in. diam, 1 to 1-1/2 in. long.

^a-Sample contained in TS-688 graphite tube.

^b-Incompatible with results without follower nozzle where debonding began at 675°C. Sample may have been incompletely carbonized or may have contained a phenolic.

^c-Sample uncontained (fuel stick).

Mechanical Debonding. Debonding methods involving mechanical abrasion, erosion, or crushing have been investigated. The separation of bonded particles using mechanical methods is based upon a relatively weak or friable matrix or the lack of a strong bond to the surface of the coated particles. The techniques tried include crushing in a press, high velocity water erosion, abrasion with a wire brush, shock depressurization of a fluid in the pores of the matrix (intended to fracture the pores of the matrix in tension). The parameters of importance to the evaluation of these techniques include the complexity of the technique, the extent of coated particle breakage, and the successful debonding of individual particles (lack of agglomerates). In addition, these methods do not provide for matrix removal as in gaseous debonding. It is therefore necessary to determine whether a matrix removal step is required to allow effective separation of the fissile and fertile fractions.

Investigations of the wire brush and shock depressurization methods were concluded during the previous quarter. Bonded beds were exposed to a motor-driven, rotary wire brush. The samples contained both furfuryl alcohol and R72S phenolic resin binders combined with BC-135 charcoal. The samples were readily debonded with undetectable coated particle breakage. The fissile and fertile fractions separated by sieving had very low cross-contamination levels.

The fluids used in the shock depressurization tests included water, He, and a saturated solution of CO_2 in water. The bonded beds contained furfuryl alcohol and BC-135 charcoal. The samples were pressurized to 11,000 psi in the fluid long enough to ensure full diffusion into the sample pores. The sample was then fully depressurized in ≤ 1.5 sec. No debonding action was observed, but the CO_2 -water system caused a noticeable degradation of the mechanical integrity of the bonded bed to a depth of about 1/2 in.

Bonded beds (1 in. long) contained in graphite tubes were debonded by crushing in a press. The specimens contained furfuryl alcohol and BC-135 charcoal. In three tests, coated particle breakage as determined by leaching ranged from 0.2% to 0.5%. One sample was separated into fertile-fissile fractions by sieving. Each fraction had 4% contamination from the other.

Studies on debonding by high-velocity water erosion were concluded with a determination of the effects of pressure and nozzle size on debonding rate. Water pressure in a large sump was allowed to decay in 1000 psi increments through the nozzle, beginning with 9500 psi. The coated particle yields are summarized in Table 5.3 using a 2-in.-long furfuryl alcohol-charcoal bonded bed located 0.25 in. from the nozzle. The debonding action was localized to a channel about 1/4 in. in diameter due to the narrow water stream.

Methods for Destroying SiC Coatings on Fuel Particles

During the past quarter, experiments continued using HF-O_2 gas mixtures to destroy the SiC coatings on fuel particles. As discussed in the previous quarterly report, hydrogen fluoride gas is an effective chemical reagent when used in conjunction with oxygen. However, HF alone appears to remove silicon as SiF_4 , leaving behind a carbon coating that inhibits further reaction between the SiC and HF. The oxygen is therefore necessary for the removal of this carbon coating.

Table 5.3
YIELD OF DEBONDED COATED PARTICLES USING EROSION
BY HIGH-VELOCITY WATER

Nozzle Inside Diameter (in.)	Water Pressure (psi)	Yield		Debonding Time (sec)	Debonding Rate (in./min)
		(g)	(Inches of fuel hole)		
0.015	9500-8500	0.0395	0.0061	3.75	0.099
	8500-7500	0.0409	0.0063	4.00	0.095
0.025	9500-8500	0.9203	0.142	1.25	6.82
	8500-7500	0.3582	0.055	1.30	2.54
	6500-5500	0.1558	0.024	1.7	0.85
0.031	9500-8500	0.350	0.054	0.65	4.98
	8500-7500	0.3268	0.050	1.60	1.87
	7500-6500	0.1520	0.023	1.70	0.81
	6500-5500	0.3336	0.051	1.25	2.45
0.035	9500-8500	1.0264	0.158	0.70	13.5
	7500-6500	2.3752	0.365	1.20	18.3

Static bed experiments have been conducted on SiC powder (200 mesh), SiC-coated disks, and fuel particles at temperatures varying from 400° to 800°C, as shown in Table 5.4. As can be seen, HF alone has a minimal effect on SiC in any form, while the 1:1 mixture of HF-O₂ is effective at 600° to 800°C.

Table 5.4
REACTION OF SiC WITH HF AND O₂ IN A STATIC BED
AT VARYING TEMPERATURES

Material	Weight Loss						Condition of Material after Reaction with HF-O ₂ at 800°C
	at 400°C (%)		at 600°C (%)		at 800°C (%)		
	HF	HF-O ₂	HF	HF-O ₂	HF	HF-O ₂	
SiC-coated disks	0	0.2	0	18	1.3	25	Analysis showed no SiC remaining
SiC-coated particles	0	1	0	26	6	29	>80% destroyed
SiC powder	2	5	3	80	13	100	All material vaporized

Corrosion-resistant flow meters have been received for use with HF in experiments designed to determine the effects of varying HF-O₂ ratios on the rate of attack.

Since the over-all objective is to develop a method whereby the SiC coating can be removed in a fluidized bed, a Monel fluidized bed tube furnace was constructed for use with the HF-O₂ gas mixtures. As expected, the initial experiments indicated a more rapid rate of attack of SiC than that observed with the static bed system. After fluidizing for 1 hr at 600°C without supporting bed material, nothing remained of a sample of SiC-coated fuel particles except some particulate matter on the exit filter. Several experiments that were conducted using 1:1 HF-O₂ gas mixtures and varying temperature and reaction times had the same end result; i.e., the particles were completely destroyed and reaction products had collected on the filter.

Stability of (Th,U)O₂ Fuel Particles Under Oxidizing Conditions

Advanced HTGR reactors are expected to use fueling schemes that will permit a separation of the fissile and fertile particles during the head-end reprocessing step. One proposed fueling method uses PyC-coated ThC₂ fertile particles and PyC-coated (Th,U)O₂ fissile particles. For this fueling scheme the separation process depends on the difference in behavior of the oxide and carbide kernels under oxidizing conditions. If the oxide kernels remain intact during the course of irradiation and during the oxidation of the fuel element, then it should be possible, in principle, to separate the two types of particles by screening after they are discharged from the fluidized-bed oxidizer. The purpose of these studies is to determine the behavior of Sol-Gel thorium-uranium oxides under oxidizing conditions in a fluidized bed.

Data on the behavior, in a fluidized bed under oxidizing conditions, of three samples of thoria and thoria-urania particles prepared by the Sol-Gel process were reported previously (Ref. 1). Surprisingly, it was found that Th_{0.8}U_{0.2}O₂ particles remained intact in a fluidized bed at temperatures up to 1000°C for 24 hr. In contrast, Th_{0.95}U_{0.05}O₂ and pure thoria particles fractured at various degrees. The data were viewed with some skepticism, since the genealogy of these particles was, to a large extent, unknown. Recent examination of these particles has shown that particles that cracked open into hemispheres were partially hollow. This was suspected on the basis of the appearance of the particles in Fig. 5.5 of Ref. 1.

In view of the questionable nature of the starting material used in the experiments discussed above, it was necessary to repeat these studies with well-characterized oxide microspheres, supplied by the Chemical Technology Division of the Oak Ridge National Laboratory.

Five batches of particles were annealed in a fluidized bed with 60-mesh Al₂O₃ for 24 hr. Each batch was annealed four times, once each at 400°, 600°, 800°, and 1000°C. Oxygen flow through the bed was ~4 liters/min. Each batch of particles was microscopically examined and photographed before annealing and after each run. Table 5.5 presents a description of the particles and summarizes the results of the test.

Table 5.5

BEHAVIOR OF THORIA AND THORIA-URANIA PARTICLES UNDER OXIDIZING CONDITIONS^a

Batch No.	400°C	600°C	800°C	1000°C
1. ThO ₂ -UO ₂ , GA No. 54-4, 45-50 mesh, 3.53 wt-% U, 84.4 wt-% Th, nat. U from ORNL.	100% stable (no breakage)	100% stable (no breakage)	100% stable (no breakage)	100% stable (no breakage)
2. ThO ₂ -UO ₂ , GA No. 54-9, 45-50 mesh, 8.01 wt-% U, 79.9 wt-% Th, nat. U from ORNL.	100% stable (no breakage)	100% stable (no breakage)	100% stable (no breakage)	60% stable (40% breakage)
3. ThO ₂ -UO ₂ , GA No. 54-9, 50-70 mesh, 8.01 wt-% Th, 79.9 wt-% U, nat. U from ORNL.	100% stable (no breakage)	100% stable (no breakage)	100% stable (no breakage)	100% stable (no breakage)
4. ThO ₂ -UO ₂ , GA No. 54-4, 50-70 mesh, 3.53 wt-% U, 84.4 wt-% Th, nat. U from ORNL.	100% stable (no breakage)	100% stable (no breakage)	95% stable (5% breakage)	95% stable ^b
5. ThO ₂ spheres, 87.8 wt-% Th from ORNL, 50-60 mesh.	100% stable (no breakage)	100% stable (no breakage)	100% stable (no breakage)	100% stable (no breakage)

^a24-hr anneal.^bNo more breakage occurred at 1000°C than at 800°C.

In the current experiments, all of the breakage in the particles was into large, irregular shapes. That is, none of the particles were split into hemispheres (as has happened with other particles) and none were broken into small fragments.

The data reported herein were obtained on unirradiated particles. The radiation damage associated with high burnup can be expected to result in a considerable degree of destruction of the fuel kernels, which may be fragile and porous when removed from an HTGR reactor. Thus, experiments such as those reported above should be carried out on irradiated oxide kernels to determine whether the damage is too extensive to permit use of this method for particle separation.

SPENT FUEL SHIPPING METHODS

The costs of shipping spent fuel from a single 1000-Mw(e) HTGR have been determined. Costs were developed for shipping the fuel elements from hypothetical reactor sites to hypothetical reprocessing plant sites in four different shipping casks. These four casks varied in size from a 3-element-capacity cask which could be hauled by truck to a 49-element-capacity cask which could be transported by rail. All casks were postulated to be constructed solely of steel.

Table 5.6 presents the annual cost and cost per kilogram of heavy metals for shipping fuel from a single 1000-Mw(e) reactor a distance of 1000 miles. Included are (1) freight cost, (2) insurance cost, (3) handling costs, and (4) amortized cost of casks. These costs include the cost of returning the empty shipping casks to the reactor plant.

Table 5.6
SUMMARY OF SHIPPING COSTS FOR SPENT HTGR FUEL^a

Shipping Cask		Shipping Method	Annual Shipping Cost (\$)	Cost/kg of Heavy Metals (\$)
Capacity (fuel elements)	Weight Loaded (lb)			
49	226,630	rail	259,250	22.50
21	170,220	rail	440,620	39.65
7	84,460	rail	687,220	61.75
3	43,100	rail	857,590	77.66
7	84,460	truck	942,070	85.40
3	43,100	truck	785,370	71.12

^a1000-Mw(e) reactor, 1000 miles shipping distance, 15,000 Mw(e) HTGR economy.

The major conclusions which may be drawn from Table 5.6 are:

1. The lowest shipping cost is realized by shipping the fuel in the largest cask.
2. Truck shipping is not competitive with rail shipping unless the shipments are to be made in the three-element size cask.
3. Truck shipping the seven-element cask is more expensive than truck shipping the three-element cask, because the weights of the seven-element cask and the tractor-trailer combination exceeds most state-established weight units. Any truck shipment that exceeds these limits is subject to regulations for special hauling permits, longer routing, and restricted travel hours.

CENTRAL HTGR FUEL REPROCESSING PLANT

Work continued on a study of a central plant for reprocessing spent HTGR fuels. The study is based on reprocessing hexagonal-block graphite fuels containing SiC-coated UO_2 fissile particles and PyC-coated ThO_2 fertile particles from a 15,000-Mw(e) HTGR economy. A burn-leach head-end process is used to prepare the fuel components for solvent extraction decontamination and separation. The bred U^{233} is decontaminated and purified to the extent necessary for refabrication into new fuel elements; partially decontaminated thorium is stored for Th^{228} decay and eventual reuse; and the fissile particles are packaged and stored for possible future reprocessing. Radioactive wastes are solidified--high-level waste by calcination and intermediate-level waste by incorporation into asphalt.

Drafting work has been completed on the block flow diagrams, the material balances, and the layout criteria drawings for the major process equipment pieces. Plant layout work has been initiated.

REFABRICATION

Work was initiated on the study of refabrication methods for recycle HTGR fuels. Simplified block flow diagrams are in preparation for a process to produce fuel elements containing U^{233} . The Sol-Gel process using amine extraction methods is used for U^{233} microsphere preparation. Other operations will include particle coating and hexagonal-fuel-block assembly. In the amine extraction route for sol preparation, a solution of uranyl nitrate is chemically reduced to U(IV) uranium and the nitrate removed by a solvent extraction process using an amine in a hydrocarbon diluent. The process appears very adaptable to remote operation.

HTGR FUEL STORAGE, REPROCESSING, AND SHIPPING COSTS

A summary of costs developed thus far on HTGR fuel storage, reprocessing, and shipping is shown in Table 5.7 for six cases of fuel management situations for a 15,000-Mw(e) HTGR economy. Included are single-purpose

Table 5.7

SUMMARY OF PRELIMINARY HTGR FUEL STORAGE, REPROCESSING AND SHIPPING COST VARIATIONS
(Reprocessing costs may vary $\pm 25\%$ from values shown)

Description	\$/kg Heavy Metals ^a					
	Case I	Case II	Case III	Case IV	Case V	Case VI
	0.5 T/D Central Plant, PyC, SiC-coated Particles	0.5 T/D Central Plant, SiC Coating, Both Particles	0.5 T/D Central Plant, Reactor Site 5-yr Fuel Storage	Head-end on NFS Plant (0.5 T/D, 130 d/yr) PyC, SiC-coated Particles	Head-end on NFS Plant (0.5 T/D, 130 d/yr) SiC Coating both Particles	Integral Reactor Head-end - 0.5 T/D Central Plant Unbonded, PyC-Coated Particles
Long-term fuel storage	0	0	57	---	---	---
Fuel shipping	18	18	18	18	18	6
Head-end	}	}	}	}	}	106
Uranium recovery						75
Other	---	2 ^b	---	---	9	---
TOTAL	82	84	139	137	156	187

^aCosts are based on 15-yr amortization of capital for central plant facilities (Cases I, II, III). Amortization of investment for head-end facilities at NFS is about 7 to 8 yr in Cases IV and V. The existing NFS plant is amortized over 15 yr. Head-end investment for Case VI is amortized over 30 yr.

^bProcessing to expose kernel of SiC-coated particles.

central plant reprocessing, multipurpose central plant reprocessing, integral reactor head-end reprocessing with central plant uranium recovery, and interim fuel storage at a reactor with delayed reprocessing at a central plant.

Of the combinations reviewed, central plant reprocessing with no interim storage is the least costly: \$82/kg of heavy metals at a 0.5 tonne/day plant (including 1000-mile fuel element shipping). At a plant with capacity of 3.0 tonne/day, this cost will decrease to about \$45/kg. These costs are preliminary in nature.

PRELIMINARY COST ESTIMATE FOR AQUEOUS REPROCESSING PLANTS

Pending development of more refined cost estimates, preliminary estimates of capital and reprocessing costs have been developed. The estimates are based on extrapolations and adjustments to earlier studies.

For this rough estimate, reprocessing of a hexagonal-block, bonded-particle type of fuel element is assumed. A burn-leach head-end process is used. Only the pyrocarbon-coated fertile particles are processed through solvent extraction: uranium is recovered, and thorium is stored for future recovery. No dollar value has been assigned to the recovered thorium. Silicon carbide coated fissile particles (containing undesirable U^{236}) are placed in long-term storage.

Capital costs for 0.5, 1.0, and 3.0 tonne/day central plants were estimated at 26, 34, and 58 million dollars, respectively. These costs include initial (three year) aqueous waste and thorium storage tankage costs and ninety day fuel storage facility costs. Unit reprocessing costs are estimated at 62, 43, and 27 \$/kg of heavy metals, respectively, for 0.5, 1.0, and 3.0 tonne/day plants.

Further estimates based on the above data indicate that about 70% of total capital costs are associated with the head-end and solvent extraction operations.

SOL-GEL STUDIES

During the present reporting period, liaison with ORNL on the Thorium-Uranium Recycle Facility (TURF) and Sol-Gel developments resulted in the following information:

1. TURF has been delayed about 1 year. It is now scheduled for cold operation in mid 1969 and hot operation in early 1970.
2. Most of the design of the Sol-Gel microsphere formation equipment (from sol through microsphere preparation) has been finalized.
3. About 10% of the development and design effort on coating, bonded bed, and fuel element loading equipment has been completed.

4. Amine-extraction has recently been chosen as the reference TURF method for preparing UO_2 sols.

References

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Task VI
PHYSICS METHODS

DIFFUSION THEORY CODES

Two-dimensional Diffusion Codes

GAMBLE-1 (Ref. 1), a two-dimensional multigroup diffusion theory code utilizing an alternating direction method that works very effectively for cell problems and for reactor problems with a highly nonuniform mesh spacing, has been modified to accept the standard cross-section format as well as the old format. The standard cross-section format can now be used for all two-dimensional and for most of the one-dimensional diffusion codes operable on the 1108 computer.

GAMBLE-5 is the latest version of a series of two-dimensional multigroup diffusion theory codes (Ref. 2) (number of energy groups ≤ 10 , number of mesh points $\leq 20,000$, arbitrary scattering matrix). The coarse-mesh rebalancing technique presently being incorporated in GAMBLE-5 is nearing its final debugging stages. The final version will include an alternating column-row rebalancing scheme that has been tested in a two-dimensional test code, as well as a more general scheme allowing coarse-mesh intervals for the two space variables and for the energy variable.

The incorporation of the coarse-mesh rebalancing scheme in GAMBLE-5 has been somewhat delayed by a general investigation of the most appropriate calculational models in finding the power distribution for large HTGR reactors. The large HTGR power reactors have an hexagonal core structure and a circular outer reflector boundary. The question therefore arises as to how the material structure can best be expressed in rectangular (X,Y) geometry. There is no doubt that a sufficiently accurate representation is possible for a large number of mesh points, but there remains the question of how many mesh points are necessary. The HTGR core with a graded fuel cycle and a scatter loading does not in general have any symmetry planes. However, in a large HTGR core, a 60° rotation of the core materials should result in a power distribution that is rotated 60° but otherwise unchanged. For a calculational model with about 6800 mesh points, representing in detail all the heterogeneities at the core-reflector interface and the outer boundary of the reflector, a rotation of the rectangular mesh grid gave discrepancies in the average region powers of up to 12%, an unacceptable situation. It was discovered that the reflector did not have rotational symmetry, and these calculations were redone in order to find out whether the wrong reflector treatment was responsible for most of the discrepancy. With the appropriate reflector treatment, the maximum discrepancy in the average region power was reduced to 3.6%. This work is continuing.

TRANSPORT THEORY CODES

An anisotropic distributed-source option was incorporated in 1DF, the one-dimensional multigroup transport theory code (Ref. 3). The addition permits the specification by group and space point of an arbitrary number of Legendre components of a fixed source.

A "white" boundary condition option (isotropic flux return) for cell calculations was incorporated in 1DF and satisfactorily tested.

The code 1DF has three levels of iteration: (1) inner iterations for within-group flux convergence, (2) outer iterations for fission and upscatter source convergence, and (3) iterations for implicit eigenvalues, e.g., critical thicknesses, isotopic concentrations. In cell calculations an additional convergence requirement is imposed on angular flux boundary values during the first level of iteration. In the first version of 1DF obtained from Los Alamos, convergence of HTGR cell problems was very slow. This poor convergence rate was caused by lack of an upscatter convergence acceleration scheme to accelerate outer iterations and by an error in the code affecting cell calculations. The error, detected and corrected early in the quarter, did not affect the results, but it caused a great deal of unnecessary inner iteration during each outer iteration.

As noted in the previous quarterly report, a GAPLSN-like iteration scheme was installed in 1DF because of the poor convergence of the original 1DF version. The GAPLSN-type scheme bypassed the unnecessary iterations caused by the error noted above and thus materially reduced computation time. However, the GAPLSN scheme did not at first ensure convergence of the boundary angular fluxes in cell calculation. But because there was no upscatter acceleration procedure, the GAPLSN scheme required many outer iterations, and in effect, obtained converged boundary fluxes in spite of the absence of a specific criterion on this point.

After the error in the GAPLSN scheme was detected, a version of 1DF containing upscatter acceleration procedures was revised to integrate all convergence procedures. A very satisfactory reduction of running times was obtained for most problems, and all convergence requirements are now met by both the standard and GAPLSN convergence options.

The GAPLSN convergence scheme is essentially an automated method for tightening inner iteration convergence as the calculation progresses and is based on the assumption that very precise convergence of within-group fluxes is not necessary to adjust fission or upscatter sources. However, the scheme is ad hoc, as is much of the strategy used in transport codes. More fundamental convergence acceleration procedures are described below.

The existing 1DF code uses only a group (inner) and system-wide (outer) neutron conserving rebalancing scheme to accelerate convergence. Theoretically, there is no reason that Chebyshev acceleration procedures, so successful in diffusion theory, cannot be used to accelerate both the inner and outer iteration processes in both the 1DF and 2DF transport codes. However, even though a great deal is known about the nature of the iteration matrices

in diffusion theory, very little is known about their structure in transport theory. A program of experimental numerical testing has therefore been undertaken to determine whether such acceleration is feasible and, if it is, to determine the optimum acceleration strategy. Preliminary results with 1DF are most encouraging.

In the first stage of the investigation, an experimental version of 1DF was prepared to permit Chebyshev acceleration of the inner iterations. In cell calculations, acceleration procedures are applied to the within-group scalar flux and the boundary angular flux simultaneously. In a one-group problem representative of HTGR cell calculations the following results were obtained:

<u>Type of Scheme</u>	<u>Number of Inner Iterations</u>
Standard 1DF	77
Chebyshev:	
a = 1.1	26
a = 1.15	22
a = 1.2	20
a = 1.3	27
a = 1.4	31

The "a" is an adjustable parameter in the Chebyshev acceleration procedure. The standard 1DF procedure involves two types of passes in each iteration cycle. One pass is made with normal sources, and then another pass is made with a boundary source. The results of these two passes are combined and the sequence repeated. The figure 77 represents 39 passes of the first type and 38 of the second. In the Chebyshev scheme, only passes of the first type are made. Hence, if the Chebyshev scheme proves more generally successful, sizable reductions can be made in 1DF storage requirements by eliminating the two-pass treatment.

Additional results from the above problem indicate that the Chebyshev procedure is less sensitive to the initial flux guess than the standard procedure.

The work described above is continuing. The parameter "a" should be chosen automatically, but in a reasonably optimum fashion. In calculations in which the material properties or the geometric configurations do not change, the parameter for each group needs to be estimated only once. Once this problem is solved, an investigation of outer iteration acceleration will be undertaken.

The greatest potential saving in computer time due to Chebyshev acceleration is in two-dimensional transport calculations. Work elsewhere indicates that the process is not so straightforward. Indications are that optimum acceleration depends on optimum spatial differencing. Work is planned in this latter area.

DEPLETION CODES

The extension of FEVER-7 (Ref. 4), a one-dimensional depletion code, to an arbitrary scattering matrix has been completed. The maximum number of groups remains unchanged at seven. The Wielandt iteration procedure has been included.

GAMBLE-5 and a dimension-independent burnup program have been combined to form one computer program (BURNUP-GAMBLE) called BUG. This program has been checked out and is operational for production runs.

An improvement in the use of auxiliary storage capacity in the 1108 computer has resulted in a 30% reduction in running time in GARGOYLE (Ref. 5). GARGOYLE is a zero-dimensional depletion code frequently used in fuel cycle analyses to study the time-dependent core composition and reactivity during the approach to equilibrium. Further savings are anticipated by the incorporation of an improved burnup routine.

A code (DIP) has been written to assist in the analysis of fuel element irradiation samples. The principal reason for the program was to obtain good estimates of the contribution of U^{233} fission to the final exposure of fuel particles that are initially composed of only Th^{232} and U^{235} . The differential equations describing the burnup relations in the heavy isotope decay chain Th^{232} to U^{236} were solved analytically in terms of fast and thermal fluxes. Using the initial isotopic ratios, the initial Th/U^{235} ratio, and values of cross-section ratios (e.g., σ^{23}/σ^{25} , σ^{24}/σ^{25} , σ^{26}/σ^{25} , etc.), values of a fast and thermal flux are calculated such that over a prescribed irradiation cycle the final measured isotope ratios are matched with the calculated values. The total fission density due to both U^{235} and U^{233} can then be calculated. The program is in its final debugging stage.

THE TIME-DEPENDENT MULTIGROUP DIFFUSION EQUATION

Development work with GAKIN, a multigroup one-dimensional kinetics code, has proceeded slowly. The possibility of taking directly into account the linear time dependence of the cross sections by using a modified GAKIN operator is being explored. This operator has the same stability properties as the original GAKIN operator.

CROSS SECTIONS AND CROSS-SECTION CODES

A revised set of neutron cross sections for U^{235} has been prepared for the GAM-II and GATHER-II libraries. These data have been obtained from a recent evaluation of the U^{235} cross sections for the ENDF/B data file at KAPL (Ref. 5). The major differences between this new set of data and the GA-5944 set previously used for the cross section libraries (Ref. 6) are in the epithermal energy range. A summary of the differences between these two sets of data is given in Table 6.1. The resonance integrals calculated from this new set of data are in good agreement with experimentally measured values (Ref. 7).

U²³³ Cross Sections

A revised set of neutron cross sections for U²³³ has been obtained from Oak Ridge National Laboratory (Ref. 8). The values are similar to the "GA Best Fit" set described by Drake (Ref. 9), except for the neutron energy region between 0.5 and 1.0 kev. In this energy region the fission and capture cross sections have been replaced by data obtained in recent differential measurements made at Rensselaer Polytechnic Institute by ORNL and RPI scientists. A comparison between the new and old data is shown in Table 6.2.

Neutron-cross-section Library

The master neutron-cross-section libraries contain data for about 220 different elements and/or nuclides. The thermal-neutron cross sections and resonance integrals for a few materials of interest to HTGR programs are summarized in Table 6.3.

Table 6.1
U²³⁵ CROSS-SECTION DATA

KAPL ENDF/B Set Lubitz & Szydluk, KAPL (Ref. 5)		Joanou & Drake (Ref. 6)
2200 m/sec Values		
$\sigma_{n,\lambda}$	100.5 barns	101.0 barns
$\sigma_{n,f}$	579.5 barns	581.0 barns
α	0.1734	0.1738
η	2.071	2.070
ν	2.43	2.430
Epithermal Values		
Capture integral ^a	142.5	147.8
Fission integral ^a	280.0	263.2
α_{epi}	0.509	0.558

^aFor a cutoff energy of 0.5 ev.

Table 6.2
U²³³ CROSS-SECTION DATA

	ORNL-RPI Set	GA Best Fit Set
2200 m/sec Values		
$\sigma_{n,\lambda}$	48.2	48.2
$\sigma_{n,f}$	525	525
α	0.0918	0.0918
η	2.293	2.293
ν	2.503	2.503
Epithermal Values (0.5 ev cutoff)		
Capture integral	140	135
Fission integral	770	775
α_{epi}	0.182	0.175

Table 6.3
SUMMARY OF NEUTRON CROSS SECTIONS

Material	σ_{2200} m/sec		Resonance Integrals ^a		
	Capture	Fission	Capture	Fission	Absorption
Iron	2.62	---	1.29	---	1.44
Gold	98.8	---	1618.4	---	1618.4
Th ²³²	7.45	---	85.7	0.46	86.3
Pa ²³¹	201.0	---	505.7	3.62	509.4
Pa ²³³	45.0	---	820.2	3.62	823.8
U ²³²	77.4	81.6	224.6	318.0	557.2
U ²³³ (ORNL-RPI) ^b	48.2	525.0	140.0	769.8	909.8
U ²³⁴	104.9	---	667.4	5.28	672.7
U ²³⁵ (KAPL-ENDF/B) ^b	100.5	579.5	142.5	280.0	422.5
U ²³⁶	6.97	---	321.0	2.81	323.8
U ²³⁸	2.70	---	275.5	1.64	277.2
Np ^{237c}	170.0	---	738.2	5.65	743.8
Pu ²³⁹	274.1	740.9	178.1	278.2	456.3
Pu ²⁴⁰	279.7	0.05	8498.6	7.16	8505.8
Pu ²⁴¹	395.4	962.0	140.0	573.0	712.7
Pu ^{241d}	351.0	1019.5	131.6	592.4	724.0
Pu ²⁴²	17.76	---	1099.6	5.75	1105.3

^aCalculated for a cutoff energy of 0.5 ev.

^bThese data are very recent, as described in this report.

^cCutoff energy of 0.414 ev.

^dRevised set of data for the ENDF/B data file. Data normalized to σ_{2200} m/sec values recommended by Westcott, et al. (Ref. 10).

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Task VII
EXPERIMENTAL PHYSICS

The experimental measurements on the C/U-859 core that were made during the previous quarter are presented in this report along with the measurements on the C/U-432 core made this quarter. The data appear in general to be of excellent quality. The measurements on the C/U-432 core were most difficult because of the small size, hard spectrum, and perturbing effects of control and safety rods.

Phase 1 of the calculational program is nearly complete. Phase 1 refers to the most straightforward analysis of experiments using cross sections in use at General Atomic. Phase 2 of the analysis is under way. In phase 2, an analysis of the comparative results is being made, and improved sets of cross sections are being developed and evaluated.

The calculational program to date has resulted in the adoption of improved sets of U^{235} and U^{238} cross-section sets for HTGR analysis.

EXPERIMENTS WITH THE C/U-859 CORE

Core Specifications

The C/U-859 core, shown in Fig. 7.1, consists of four explicit regions: the central or exact region containing fuel elements with a C/ U^{235} ratio of about 859; the adjacent heterogeneous region containing one fuel element whose C/U ratio is about 432 for every graphite column; the reflector region; and the outermost region of aluminum honeycomb material. The material densities contained in these different regions are summarized in Table 7.1.

Table 7.1
MATERIAL DENSITIES OF C/U-859 CORE

Core Region	Isotopic Mass Loading, $\bar{M}$ (g/cm ³) ^a				
	Carbon	Aluminum ^b	U^{235}	U^{238}	Oxygen
Exact, C/U-859	1.393	0.3073	3.1706-2	2.314-3	0.523-2
Heterogeneous, C/U-859	1.443	0.2526	3.1795-2	2.320-3	0.524-2
Reflector	1.493	0.1980	---	---	---
Voided Aluminum Honeycomb	---	0.1980	---	---	---

^a $\bar{M}$ is the isotopic mass per element divided by the volume of the honeycomb unit cell where the area of the honeycomb cell is 15.452 cm².

^bHoneycomb and fuel tube.

Fig. 7.1--C/U-859 core configuration

Shutdown Mechanisms

The differential control rod worth was measured by the period technique. The total integrated worths for the four rods ranged from 0.238 to 0.25.

The total worth of a single safety rod was obtained by the subcritical source multiplication technique using a previously calibrated regulating rod to generate the necessary inverse source multiplication curve. The worths of the eight rods ranged from 0.85 to 0.89.

The worth of a nuclear fuse was determined by the inverse source multiplication technique. A fired fuse was inserted in place of a good fuse giving a reactivity decrease of \$0.54.

Excess Reactivity

To facilitate comparisons with calculations, it is desirable to make experimental corrections to the actual core excess reactivity for the locations taken up by control rods, safety rods, nuclear fuses, etc. These corrections were made by removing graphite columns or fuel elements at the same radial distance as the component under investigation and substituting a stainless steel sleeve (in the case of the rods) similar to the control and safety rod guide sleeves. The resultant reactivity loss was then measured using a calibrated control rod. These results are shown in Table 7.2.

Table 7.2

CORRECTED C/U-859 CORE EXCESS REACTIVITY

Measured core excess reactivity	\$ 0.475
Correction for eight safety rod locations (graphite out, stainless steel in)	1.029
Correction for four control rod locations (C/U-859 fuel element out, stainless steel in)	0.835
Correction for two source tubes	0.051
Correction for two nuclear fuses	0.445
Honeycomb correction	<u>-1.074</u>
Net corrected excess reactivity	<u>\$ 1.761</u>

The honeycomb correction refers to the reactivity gain resulting from the reflective effect of the aluminum honeycomb in the region beyond the graphite reflector. The correction was made by inserting empty aluminum fuel element tubes into one quadrant of one half of the honeycomb in the region beyond the graphite reflector. The resulting reactivity increase was then measured. Multiplying this result by a factor of eight and applying a correction for the difference in weight between honeycomb and fuel element tubes gave the correction indicated above.

Reactivity Coefficients of Special Elements

The reactivity coefficients of all special elements were measured in the central honeycomb location (26,29). The measurements were performed

by first determining the control rod position required to make the assembly supercritical by about \$0.002 with the central location occupied by a special C/U-859 fuel element. The actual supercritical amount was determined by the drift technique, in which the linear component of the exponential increase for periods greater than several thousand seconds is measured by a linear strip recorder with a high-gain differential input. The special C/U-859 element was then replaced by the special element under investigation and the new control rod position required to make the core just slightly supercritical determined. In cases where the special element differed in worth from the special C/U-859 element by more than \$0.50 (the maximum allowable excess reactivity) intermediate steps were taken in which reflector graphite elements were added or removed to adjust the total excess reactivity. The reactivity worth of these reflector changes was again measured on the control rods. Several of the control rod calibration points were rechecked when the reflector was changed, and it was found that the calibration had not changed. Table 7.3 shows the results obtained and the ratio of experimental to calculated results.

Table 7.3
CENTRAL REACTIVITY COEFFICIENTS OF SPECIAL ELEMENTS
IN THE C/U-859 CORE

Element Description	Measured Worth ^a (\$)	Calculated Worth ^a (\$)	Ratio of Exp. to Calc.
Boron 0.10	-0.055	-0.047	1.170
Boron 0.20	-0.083	-0.075	1.107
Boron 0.45	-0.149	-0.148	1.007
Boron 0.55	-0.185	-0.177	1.045
Boron 1.00	-0.302	-0.302	1.000
Boron 1.50	-0.409	-0.431	0.995
Boron 1.90	-0.523	-0.531	0.985
Thorium 50	-0.085	-0.079	1.076
Thorium 100	-0.121	-0.124	0.976
Thorium 200	-0.188	-0.194	0.969
Thorium 300	-0.241	-0.252	0.956
Thorium 500	-0.325	-0.342	0.950
Depleted U 50	-0.125	-0.132	0.947
Depleted U 100	-0.172	-0.194	0.887
Depleted U 200	-0.257	-0.275	0.935
Depleted U 300	-0.315	-0.336	0.938
Depleted U 500	-0.398	-0.426	0.934
U ²³³ C/U-432 ^b	+0.214	+0.254	0.843
U ²³³ C/U-1718	+0.050	+0.065	0.769
U ²³³ C/U-2500	+0.004	+0.012	---
U ²³³ C/U-432 ^c	---	+0.251	0.853
U ²³³ C/U-1718	---	+0.063	0.79
U ²³³ C/U-2500	---	+0.010	---

Table 7.3 (continued)

Element Description	Measured Worth ^a (\$)	Calculated Worth ^a (\$)	Ratio of Exp. to Calc.
U ²³⁶ C/U-432	-0.039	-0.073	0.534
U ²³⁶ C/U-1718	-0.034	-0.034	1.000
U ²³⁶ C/U-2500	-0.033	-0.027	1.222
U ²³⁵ C/U-432	+0.024	+0.016	1.500
U ²³⁵ C/U-859	0.0	0.0	---
U ²³⁵ C/U-1718	-0.010	-0.008	1.250
U ²³⁵ C/U-2500	-0.015	-0.011	1.364
U ²³⁵ C/U-5000	-0.019	-0.015	1.267
Au 5 (new resonance GAM)	-0.117	-0.116	1.009
Au 15 (new resonance GAM)	-0.248	-0.240	1.008
Au CN (45) (new resonance GAM)	-0.466	-0.457	1.020
Np ²³⁷ 5 ^d	-0.093	-0.082	1.134
Np ²³⁷ 15	-0.215	-0.179	1.201
Np ²³⁷ 55	-0.618	-0.609	1.015
PuH 240 A ^e	-0.280	---	---
PuH 240 B	-0.160	---	---
PuH 240 C	-0.087	---	---
PuL 240 A ^e	-0.090	---	---
PuL 240 B	-0.054	---	---
PuL 240 C	-0.038	---	---
Carbon, ref. density	-0.029	-0.022	1.318
Carbon, 0.9 ref. density	-0.040	-0.347	1.175
Aluminum rod	-0.330	-0.347	0.951

^a-With respect to special C/U-859 element.

^b-U²³³ cross-section set from M. K. Drake, Neutron Cross Sections for U²³³, USAEC report GA-7076, General Atomic, September 15, 1966.

^c-U²³³ ORNL-RPI set.

^d-Np²³⁷ cross-section set as reevaluated by General Atomic, December 1966, to be published.

^e-Some calculational results for plutonium samples are discussed in this report. The analytical methods in computing the worth of plutonium require more refinements because of the large low-lying resonance in Pu²⁴⁰.

During the course of this analysis a number of substantial changes were made in the basic data. The analytical results shown in Table 7.3 are based on the same cross section sets as were used in the preceding two quarterly reports. The new analytical results are discussed later.

Pulsed-neutron Measurement of $\beta_{\text{eff}}/\Lambda$

The C/U-859 core was pulsed and the prompt decay constant measured. Using the expression

$$\frac{\beta_{\text{eff}}}{\Lambda} = \frac{\alpha}{\rho - 1} ,$$

where ρ is the reactivity in dollars that the core is sub-delayed critical, Λ is the prompt generation time, and α is the prompt decay constant, a value of

$$\beta_{\text{eff}}/\Lambda = 94.9$$

was obtained for $\rho = \$0.20$. The calibrated control rod was used to make the reactor supercritical by just this amount.

EXPERIMENTS WITH THE C/U-432 CORE

Core Specifications

The C/U-432 core, shown in Fig. 7.2 consists of four explicit regions: the central or exact region containing fuel elements with a C/U²³⁵ ratio of about 432; the adjacent region containing fuel elements with a C/U ratio of about 859; the reflector region; and the outermost region of aluminum honeycomb material. The material densities contained in these different regions are summarized in Table 7.4.

Table 7.4
MATERIAL DENSITIES OF C/U-432 CORE

Core Region	Isotopic Mass Loading, $\bar{M}$ (g/cm ³) ^a				
	Carbon	Aluminum ^b	U ²³⁵	U ²³⁸	Oxygen
Exact, C/U-432	1.395	0.3073	6.3590-2	4.641-3	1.049-2
C/U-859	1.393	0.3073	3.1706-2	2.314-3	0.523-2
Reflector	1.493	0.1980	---	---	---
Voided Aluminum Honeycomb	---	0.1980	---	---	---

^a $\bar{M}$ is the isotopic mass per element divided by the volume of the honeycomb unit cell where the area of the honeycomb cell is 15.452 cm².

^bHoneycomb and fuel tube.

Fig. 7.2--C/U-432 core configuration

Shutdown Mechanisms

The differential control rod worth was measured by the period technique. The total integrated worths are: rod 1, \$0.161; rod 2, \$0.47; rod 3, \$0.472; and rod 4, \$0.156. The larger worths of rods 2 and 3 are the result of adding a cartridge containing B_4C to these rods.

The total worth of a single safety rod was obtained by the subcritical source multiplication technique using a previously calibrated regulating rod to generate the necessary inverse source multiplication curve. The eight safety rods were worth approximately \$0.87 each.

The worth of a nuclear fuse was determined by the inverse source multiplication technique. A fired fuse was inserted in place of a good fuse giving a reactivity decrease of \$0.31.

Excess Reactivity

Experimental corrections to the actual core excess reactivity for the locations taken up by control rods, safety rods, and nuclear fuses were made again. These corrections were obtained by removing graphite columns or fuel elements at the same radial distance as the component under investigation and substituting a stainless steel sleeve (in the case of the rods) similar to the control and safety rod guide sleeves. The resultant reactivity loss was then measured using a calibrated control rod. These results are shown in Table 7.5.

Table 7.5

CORRECTED C/U-432 CORE EXCESS REACTIVITY

Measured core excess reactivity	\$ 0.432
Correction for eight safety rod locations (graphite out, stainless steel in)	1.180
Correction for four control rod locations (C/U-859 fuel element out, stainless steel in)	0.809
Correction for two source tubes	0.042
Correction for two nuclear fuses	0.874
Honeycomb correction	<u>-1.062</u>
Net corrected excess reactivity	<u>\$ 2.275</u>

Reactivity Coefficients of Special Elements

The reactivity coefficients of all special elements were measured in the central honeycomb location (26,29) in the same manner as described for the C/U-859 core. The results are shown in Table 7.6.

Pulsed-neutron Measurement of β_{eff}/Λ

Using the same procedure as described before, a measured value of $\beta_{eff}/\Lambda = 150.8$ was obtained for the C/U-432 core.

Table 7.6
SPECIAL REACTIVITY COEFFICIENTS OF SPECIAL ELEMENTS
IN THE C/U-432 CORE

Element Description	Measured Worth ^a (\$)	Calculated Worth ^a (\$)	Ratio of Exp. to Calc.
U ²³⁵ C/U-432	0.000	0.000	---
U ²³⁵ C/U-859	-0.11	-0.008	1.375
U ²³⁵ C/U-1718	---	-0.010	---
U ²³⁵ C/U-2500	---	---	---
U ²³⁵ C/U-5000	---	---	---
Au 5 (new resonance GAM)	-0.085	---	---
Au 15 (new resonance GAM)	-0.187	---	---
Au CN (45) (new resonance GAM)	-0.345	---	---
Np ²³⁷ 5 ^b	-0.067	-0.058	1.155
Np ²³⁷ 15	-0.158	-0.124	1.274
Np ²³⁷ 55	-0.449	-0.544	0.825
PuH 240 A ^c	-0.205	---	---
PuH 240 B	-0.115	---	---
PuH 240 C	-0.062	---	---
PuL 240 A ^c	-0.068	---	---
PuL 240 B	-0.041	---	---
PuL 240 C	-0.028	---	---
Carbon, ref. density	-0.021	-0.017	1.235
Carbon, 0.9 ref. density	-0.039	-0.034	1.147
Aluminum rod	-0.256	-0.264	0.970
Boron 0.10	---	---	---
Boron 0.20	---	-0.038	---
Boron 0.45	-0.083	-0.074	1.122
Boron 0.55	-0.100	-0.087	1.149
Boron 1.00	-0.163	-0.147	1.109
Boron 1.50	-0.233	-0.209	1.115
Boron 1.90	-0.288	-0.259	1.112
Thorium 50	-0.070	---	---
Thorium 100	-0.102	-0.102	1.000
Thorium 200	-0.161	-0.160	1.006
Thorium 300	-0.207	-0.208	0.995
Thorium 500	-0.280	-0.282	0.993

Table 7.6 (continued)

Element Description	Measured Worth ^a (\$)	Calculated Worth ^a (\$)	Ratio of Exp. to Calc.
Depleted U 50	-0.101	-0.102	0.990
Depleted U 100	-0.150	-0.152	0.987
Depleted U 200	-0.216	-0.217	0.995
Depleted U 300	-0.266	-0.267	0.996
Depleted U 500	-0.340	-0.340	1.000
U ²³³ C/U-432 ^d	+0.142	0.146	0.973
U ²³³ C/U-1718	+0.030	0.033	0.909
U ²³³ C/U-2500	---	0.003	---
U ²³³ C/U-432 ^e	+0.142	0.144	0.986
U ²³³ C/U-1718	+0.030	0.031	0.968
U ²³³ C/U-2500	---	---	---
U ²³⁶ C/U-432	-0.048	-0.057	0.842
U ²³⁶ C/U-1718	-0.032	-0.030	1.067
U ²³⁶ C/U-2500	-0.027	-0.022	1.227

^aWith respect to special C/U-432 element.

^bNp²³⁷ cross-section set as reevaluated by General Atomic, December 1966, to be published.

^cSome calculational results for plutonium samples are discussed in this report. The analytical methods in computing the worth of plutonium require more refinements because of the large low-lying resonance in Pu²⁴⁰.

^dU²³³ cross-section set from M. K. Drake, Neutron Cross Sections for U²³³, USAEC report GA-7076, General Atomic, September 15, 1966.

^eU²³³ ORNL-RPI set, L. W. Weston, ORNL-TM-1751, to be released.

ANALYSIS OF EXPERIMENTAL DATA

The phase 1 analysis of the C/U-859 core results was based on the same U²³⁵ cross sections as were used in the analyses of the C/U-2500 and -1718 cores.

About midway through the analysis of the C/U-859 core, a new set of U²³⁵ cross sections became available. This was the KAPL ENDF/B set, nuclide number 1044. This set is characterized by an epithermal α of 0.509 as compared with an epithermal α of 0.558 for the General Atomic U²³⁵ cross-section set. Because of existing discrepancies in the U²³⁵ reactivity worths as well as the core eigenvalues, a decision was made to first complete the analysis of the C/U-859 results using the "older" General Atomic U²³⁵ cross

sections and then to reevaluate certain results obtained in the C/U-2500, -1718, and -859 cores using the KAPL ENDF/B U^{235} cross sections. In view of the favorable results obtained, the analysis of the C/U-432 core was based exclusively on the ENDF/B U^{235} data.

Broad-group Cross Sections

The effective broad-group cross sections used in analyzing the C/U-859 and -432 cores were obtained in the manner described in the two previous quarterly reports for the earlier core assemblies. If the geometric buckling for the critical "as built" core assembly is used in the spectrum calculations, the calculated neutron energy spectrum over which the broad-group cross sections are averaged is within a few percent of the spectrum obtained from the GAZE diffusion calculation in the region of the core where the special elements are inserted. This was done for the final analysis of both the C/U-432 and -859 core assemblies. A 30-group diffusion theory analysis was made for both assemblies.*

As previously indicated, the C/U-432 core assembly had a significant number of C/U-859 fuel elements in the outer region of the core. In analyzing this assembly, the broad-group cross sections for the central region and for all of the special element materials were averaged over the C/U-432 neutron spectrum. The broad-group cross sections in the C/U-859 core region and the thin radial reflector region were averaged over a spectrum characteristic of the 859 carbon-to-uranium ratio.

Eigenvalue Calculations

The measured core excess reactivity for both the C/U-859 and -432 assemblies was experimentally corrected to include the effect of voids at the core locations used for control rods, safety rods, and nuclear fuses as well as the reflective effect of the empty aluminum honeycomb surrounding the core assembly. The effective multiplication constant, k_{eff} , for both the C/U-859 and -432 cores was calculated using a one-dimensional multigroup diffusion theory code with the three explicit radial core and reflector regions shown in Figs. 7.1 and 7.2, respectively. The experimentally measured water content reported in the previous quarterly report was included in each region. The group-dependent axial buckling was obtained by iterating between radial and axial problems.

The calculated results for both U^{235} cross-section sets are shown in Table 7.7. In addition, the calculated values of the effective multiplication factors for the C/U-2500 and -1718 cores are shown as computed with the new ENDF/B U^{235} cross section data. Very good agreement for all of the cores is obtained when the new U^{235} data are used.

* A discrepancy in the graphite scattering kernel was also found. At 1 ev, where the "free gas" and "crystalline" approximations are joined, a discontinuity in the computed spectrum was observed. An improved kernel was generated but the results were not significantly affected.

Table 7.7
COMPARISON BETWEEN THE CALCULATED AND MEASURED
VALUES OF k_{eff}

Core	GA U^{235} Cross Sections	KAPL ENDF/B U^{235} Cross Sections	Measured k_{eff}
C/U-2500	1.009	1.017	1.012 ^a
C/U-1718	1.003	1.013	1.013
C/U-859	0.986	1.012	1.013
C/U-432	0.997	1.019	1.016

^aAn extrapolated value for the worth of the empty aluminum honeycomb assembly surrounding the core was applied to the previously measured value.

In addition to the analysis of the four HTGR core assemblies, one-dimensional diffusion theory calculations were run on a UCRL critical experiment that had been studied at General Atomic Several years ago. The UCRL assembly had a C/ U^{235} ratio of 297, core dimensions of 42.5 x 42.5 x 47.1 in., and was just critical. These calculations, which utilized the same 30-group broad-group structure and methods discussed above, resulted in an eigenvalue of 0.9925 compared with 0.962 obtained with the old cross sections. The improvement in the results with the new U^{235} data is impressive.

Reactivity Coefficient Calculations

The reactivity coefficients of the special elements in the central cell of the core assemblies have been calculated using the 30-group diffusion theory model. They are summarized in Tables 7.3 and 7.6 for the C/U-859 and -432 cores, respectively, as well as in Figs. 7.3 through 7.12.

The calculations for the C/U-859 core were done originally with the General Atomic U^{235} cross-section data (Table 7.3) and then selected calculations were made using the ENDF/B U^{235} cross-section data. For the C/U-432 core, all of the calculations were done with the latter U^{235} data. With the exception of the reactivity coefficient for U^{235} , the use of the new data in the surrounding core for the C/U-2500, -1718, and -859 cores had little effect on the previously calculated reactivity coefficients for the special materials. Slight changes were made and these are summarized in Table 7.8. The major effect of using the KAPL-ENDF/B U^{235} data was in the effective multiplication constant for the core and in the reactivity worth of the U^{235} special elements.

Several cross section sets for U^{233} have been used during the analysis of the reactivity worth of U^{233} in the four core assemblies. The calculated worths as shown in Tables 7.3 and 7.6 for the C/U-859 and C/U-432 cores, respectively, are given for the two most recent cross-section sets. The General Atomic best fit set, and the ORNL-RPI set. Data based on the ORNL-RPI set is also shown in the curves of Fig. 7.5.

Fig. 7.3--Boron content (10^{-4} g/cm^3)

Fig. 7.4-- U^{235} content (g/cm³)

Fig. 7.5-- U^{233} content (g/cm³)

Fig. 7.6--U²³⁸ content (g/cm³)

Fig. 7.7--Thorium (g/cm³)

Fig. 7.8--Gold concentration (g/cm^3)

Fig. 7.9-- U^{236} content (g/cm³)

Fig. 7.10--Neptunium concentration (g/cm³)

Fig. 7.11-- Pu^{239} content (g/cm^3)

Fig. 7.12--Pu²³⁹ content (g/cm³)

Table 7.8
SENSITIVITY OF RESULTS TO U²³⁵ CROSS SECTIONS
(Related to Carbon)

Description Core and Parameter	Calculated Value (\$)		Measured Value (\$)
	GA U ²³⁵ Cross Sections	KAPL-ENDF/B U ²³⁵ Cross Sections	
Worth of 432 U ²³⁵ fuel relative to all carbon:			
C/U-2500	0.326	0.338	0.332
C/U-1718	0.165	0.179	0.185
C/U-859	0.038	0.051	0.053
C/U-432	---	0.017	0.021
Worth of 859 U ²³⁵ fuel element relative to all carbon:			
C/U-2500	0.188	0.194	0.196
C/U-1718	0.094	0.102	0.104
C/U-859	0.022	0.028	0.029
C/U-432	---	0.008	0.011
β_{eff} :			
C/U-2500	0.00694	0.00689	---
C/U-1718	0.00714	0.00707	---
C/U-859	0.00731	0.00719	---
C/U-432	---	0.00708	---
β/Λ :			
C/U-2500	32.6	33.2	32.6±0.3
C/U-1718	46.0	48.6	46.3±0.3
C/U-859	102.3	105.4	94.9
C/U-432	---	191.0	150.8
Worth of 1.9 g boron ele- ment relative to all carbon:			
C/U-2500	-1.051	-1.039	-1.031
C/U-1718	-0.853	-0.837	-0.817
C/U-859	-0.501	-0.494	-0.494
C/U-432	---	-0.242	-0.267
Worth of 1.5 g boron ele- ment relative to all carbon:			
C/U-2500	-0.858	-0.849	-0.853
C/U-1718	-0.692	-0.680	-0.668
C/U-859	-0.409	---	-0.400
C/U-432	---	-0.192	-0.212

Table 7.8 (continued)

Description Core and Parameter	Calculated Value (\$)		Measured Value (\$)
	GA U ²³⁵ Cross Sections	KAPL-ENDF/B U ²³⁵ Cross Sections	
Worth of 1.0 g boron element relative to all carbon:			
C/U-2500	-0.596	-0.590	-0.593
C/U-1718	-0.478	-0.470	-0.461
C/U-859	-0.280	-0.272	-0.273
C/U-432	---	-0.130	-0.142
Worth of 432 U ²³³ element relative to carbon ^a :			
C/U-2500	0.601	0.593	0.565
C/U-1718	0.446	0.439	0.415
C/U-859	0.276	0.265	0.243
C/U-432	---	0.163	0.163
Same as above with ORNL-RPI U ²³³ data:			
C/U-2500	---	0.584	0.565
C/U-1718	---	0.431	0.415
C/U-859	---	0.263	0.243
C/U-432	---	0.161	0.163
Worth of 500 g Th ²³² element relative to carbon:			
C/U-2500	-0.349	-0.348	-0.342
C/U-1718	-0.347	-0.348	-0.331
C/U-859	-0.320	-0.317	-0.296
C/U-432	---	-0.265	-0.259
Worth of 500 g depleted uranium element relative to carbon:			
C/U-2500	-0.382	-0.382	-0.370
C/U-1718	-0.412	-0.412	-0.389
C/U-859	-0.404	-0.398	-0.369
C/U-432	---	-0.323	-0.319

^aGeneral Atomic best-fit U²³³ cross sections used.

Calculation of β_{eff} and Neutron Lifetime

Using the methods outlined in the previous quarterly report, β_{eff} and the neutron lifetime were calculated for both the C/U-859 and -432 core assemblies. These results as well as the results for the C/U-2500 and -1718 cores are summarized in Table 7.8. The agreement between measurement and calculation for these cores is poor for the C/U-432 core, but the lifetime for that core is extremely small, 46.9 μsec . The calculated effective delayed neutron fraction for the C/U-432 core is smaller than that for the C/U-859 core even though the core itself is physically smaller. This is caused by the increased water content in the C/U-432 core, which reduces the total neutron leakage.

Reactivity Worth of Plutonium Elements

The treatment of the space-energy shielding with a lumped resonance absorber embedded in another material is best handled by methods (such as Nordheim's) incorporated in the GAM-II code. However, the principal resonances of Pu^{239} (at 0.296 eV) and Pu^{240} (at 1.057 eV) lie below the GAM code cutoff for graphite-moderated reactors and in the region of neutron upscattering where a theoretical treatment of resonance space-energy shielding is not available. Therefore, the calculational approach to date to the plutonium reactivity coefficient has been to investigate various approximations to the more exact treatments, e.g., a many-group transport theory calculation using the S_n approximation of Carlson. Table 7.9 shows the results of successively improved diffusion theory and transport theory calculations for the various plutonium special elements in the C/U-2500 critical assembly. The thermal group structures that were used with additional groups provided finer energy detail around the very large Pu^{240} resonance at 1.057 eV.

As can be seen from Table 7.9, the major experimental-analytical discrepancies exist in the high Pu^{240} content samples. As the number of thermal groups increases from 8 to 16 in the diffusion theory, a large improvement in agreement was achieved. However, little improvement resulted from using P1-S4 transport theory. Improving the transport theory approximation from S4 to S8 (which increases the number of segments in the unit sphere from 8 to 24) gave a significant improvement. An important cause of the discrepancy in the high Pu^{240} samples may be due to self-shielding in the individual plutonium oxide grains. The grains were less than 44 μ in diameter, but the exact grain size distribution is not known. However, 44- μ grains could give a significant self-shielding effect.

Based on the results shown in Table 7.9, higher order approximations to the exact treatment did not appear fruitful and were not pursued, especially since the grain size was not known. Analyses of results obtained in other HTGR critical assemblies are in progress. Basic improvements in the methods of treating lumped thermal absorbers are also under study.

Summary of Results to Date

The following general statements can be made with respect to the analysis made of the experiments performed in the four different core assemblies to date.

Table 7.9

MEASURED AND CALCULATED PLUTONIUM REACTIVITY COEFFICIENTS IN THE C/U-2500 ASSEMBLY

Plutonium Sample	Measured Reactivity Coefficient (\$)	Calculated Reactivity Coefficients (\$)				
		Diffusion Theory		Transport Theory		
		30 Groups (8 thermal)	38 Groups (16 thermal)	38 Groups (P_1S_1)	38 Groups (P_1S_8)	44 Groups (22 thermal) (P_1S_4)
PuL-240 A ^a 18.85 g Pu/element	-0.0014	-0.0049	-0.0051	-0.0067	---	---
PuL-240 B ^a 7.59 g Pu/element	-0.045	---	-0.042	---	---	---
PuL-240 C ^a 3.11 g Pu/element	-0.064	-0.061	-0.060	---	---	---
PuH-240 A ^b 23 g Pu/element	-0.225	-0.352	-0.270	-0.264	-0.249	-0.262
PuH-240 B ^b 9.25 g Pu/element	-0.163	-0.212	-0.186	-0.183	-0.166	---
PuH-240 C ^b 3.86 g Pu/element	-0.118	---	-0.127	---	---	---

^aSamples contain 95.62% Pu²³⁹, 4.10% Pu²⁴⁰, 0.27% Pu²⁴¹, and 0.01% Pu²⁴².

^bSamples contain 72.34% Pu²³⁹, 22.19% Pu²⁴⁰, 4.69% Pu²⁴¹, and 0.78% Pu²⁴².

The effective multiplication constant, k_{eff} , reactivity worth of the boron and U^{235} special elements, as well as the reactivity worth of the carbon in the lower density carbon element, agree very well with the measured values if the KAPL-ENDF/B U^{235} cross section data is used in the analysis. The agreement in the case of the C/U-432 core is not generally as good as for the other cores; however, there were more uncertainties, both experimentally and analytically, in the C/U-432 core, such as the proximity of the rods and nuclear fuses to the cell in which the measurements were made, as well as the greater water content and its high reactivity worth.

There are several cross-section sets available for U^{233} . With any of these sets, the calculated reactivity worths for the U^{233} special elements differ from the measured values. Generally, the smallest differences occur when the ORNL-RPI cross-section set is used. As indicated in Fig. 7.5, the calculated values are about 5% to 8% too high.

For the resonance absorbers, thorium and U^{238} , the reactivity worth as calculated is generally greater than the measured values by up to 8%. The deviation in all cases is larger for the harder spectrum cores. In the lumped resonance calculation the effective broad-group cross sections for the resonance region are calculated with the GAM code using the asymptotic approximation to describe the scattering source due to the carbon in the lump. Improvements in this theory are in progress that slightly change the effective resonance integral.

The analysis of the reactivity coefficients of plutonium, U^{236} , and Np^{237} is unsatisfactory to date, and a larger effort with these isotopes is planned for the future.

HTGR OFFSITE PROGRAM SURVEILLANCE

General Atomic is studying physics research related to HTGR safety being performed at various sites with the following objectives:

1. To review and coordinate temperature coefficient measurements to be done on HTGR lattices in the High Temperature Lattice Test Reactor (HTLTR) at Battelle Northwest Laboratory.
2. To evaluate the usefulness of transient experiments using the UHTREX facility at Los Alamos.
3. To analyze the feasibility of installing and using a GREET (Ref. 1) type reactor in the EBOR (Ref. 2) facility.
4. To prepare a program for the testing of HTGR fuel particles in transient burst ranges characteristic of accidents.

Both the HTLTR and UHTREX facility are considered to be engaged in the HTGR Base Program. The above work is expected to establish experimental guidelines, to assure that the offsite programs are useful to the Base Program and to explore some alternative methods.

Some proposed experimental lattices for the HTLTR facility were analyzed in comparison with an HTGR assembly. The analysis consisted of infinite medium fast- and thermal-spectra calculations. The temperature dependence of the resonance escape probability, the thermal utilization, the infinite medium multiplication factor, and the effective resonance integral were calculated for four proposed lattices and for an HTGR assembly. The results are shown in Table 7.10.

The four HTLTR lattices were to be composed on 1-in. diameter rods in a carbon moderator. The rods were to be composed of either 1.5% or 3.0% U^{233} oxide mixed with thorium oxide. As can be seen from Table 7.10, the proposed C/Th and C/U ratios bracket the nominal values of the HTGR equilibrium core; however, it is apparent that there are sizable differences in the magnitude and structure of the temperature coefficient between the HTLTR and the nominal HTGR values. These differences stem primarily from the low values of the effective resonance integral for thorium in the HTLTR cores. The high self-shielding in the HTLTR lattices is caused both by the large (1-in. diameter) rods and by the relatively high concentration of thorium in the rods.

As a result of this analysis, revisions to the HTLTR experimental plans have been suggested in order to more closely duplicate the actual HTGR lattice, and a continuing review of the experiments will be performed.

References

1. Stern, M. O., et al., Gas-cooled Reactor Experimental Excursion Test, Final Report on Feasibility Studies, Supplements 1 and 2, USAEC report GA-6178, General Atomic, March 15, 1965.
2. Final Safety Analysis Report for the Experimental Beryllium Oxide Reactor, USAEC report GA-5000, General Atomic, November 30, 1964.

Table 7.10
TEMPERATURE COEFFICIENT DATA FOR VARIOUS LATTICES
(Temperature Interval from 300° to 900°K)

Lattice	p (300°K)	$\frac{1}{p} \frac{dp}{dt}$ (10 ⁻⁵ °C ⁻¹)	f (300°K)	$\frac{1}{f} \frac{df}{dt}$ (10 ⁻⁵ °C ⁻¹)	k _∞ (300°K)	$\frac{1}{k_{\infty}} \frac{dk_{\infty}}{dt}$ (10 ⁻⁵ °C ⁻¹)	I _{Th} 300°K (b)	$\frac{1}{I} \frac{dI}{dt}$ (10 ⁻⁵ °C ⁻¹)
Typical HTGR equilibrium core (no burnable poison), C/Th=225, C/U=5000.	0.6725	-6.92	0.6774	+2.13	1.1137	-5.45	29.424	+26.7
HTLTR No. 1, 7.5-in. pitch, 1.5% U ²³³ , 1-in. rods, C/Th=370, C/U=24,000.	0.9343	-1.02	0.5124	+3.51	1.1417	+2.19	11.689	+22.1
HTLTR No. 2, 3.75-in. pitch, 1.5% U ²³³ , 1-in. rods, C/Th=90, C/U=5800.	0.7583	-4.27	0.5813	+2.99	1.1728	-1.56	10.675	+20.4
HTLTR No. 3, 7.5-in. pitch, 3.0% U ²³³ , 1-in. rods, C/Th=370, C/U=12,000.	0.9124	-1.04	0.6833	+2.25	1.5164	+0.90	11.636	+22.1
HTLTR No. 4, 3.75-in. pitch, 3.0% U ²³³ , 1-in. rods, C/Th=90, C/U=2900.	0.6881	-4.24	0.7535	+1.78	1.4969	-2.64	10.484	+20.4

Task VIII
PHYSICS ANALYSES AND FUEL MANAGEMENT

ADVANCED CONVERTER EVALUATION

Mass flows for both the reference design (on-stream refueling) and the backup design (annual refueling) were supplied to ORNL during the previous quarter. During the current quarter, additional mass flows have been developed using U^{233} makeup in place of U^{235} . This has been done on the assumption that the HTGR could use U^{233} produced in other reactors such as the HWOCR or the blanket of fast breeders. Table 8.1 shows the mass flows with bimonthly refueling at 80% capacity. All cases reported including that shown in Table 8.1 have been based on an assumed efficiency of 40%. The efficiency of the backup design is 40.7%, and thus the core is capable of 1018 Mw(e); the efficiency of the reference design is 43.1%, and thus the core is capable of 1078 Mw(e). The results have additional conservatism in that:

1. A minimum of 0.5% Δk excess reactivity is present at shutdown.
2. A conservative set of U^{235} cross sections was used.
3. The calculational model used for the reference design approximated on-stream refueling with bimonthly refueling.

FEED-BREED FUEL CYCLE

The feed-breed fuel cycle studies have been divided into two parts. In the first, batch refueling of the breed zones was assumed; that is, all breed zones were replaced simultaneously. As discussed in the previous quarterly report, the object of this approach was to simplify the requirements of on-stream refueling. The second part of the study considers periodically replacing a fraction of the breed region (grading the breed).

For survey purposes, fuel cycle calculations were performed using the GAFFOX code, a one-dimensional equilibrium-cycle diffusion-theory depletion program that determines the fuel requirements. This model overestimates the fuel requirements, in that the shutdown k_{eff} , which must be satisfied, occurs only at the end of the breed residence time. Because of the fission-product inventory in the breed at this time, the resulting uranium requirements are overestimated for all other times in the cycle. For survey purposes however, this limitation was not regarded as abrogating the convenience of the program.

The feed-breed cycle has an additional degree of freedom relative to the reference HTGR fuel cycle as the fertile and fissile material can have different residence times. For this survey, cycle times of from 0.5 to 2.0 yr for the feed region and from 6 to 10 yr for the breed regions were considered.

Table 8.1
MASS FLOWS FOR 1000-MW(E) HTGR^a

Time (yr)	Th Charged (tonnes)	U ²³³ Charged (kg)	U ²³⁵ Recycled (kg)	Total U Charged (kg)	Th Dischg. (kg)	U ²³³ Dischg. (kg)	U ²³⁵ Dischg. (kg)	Total U Dischg. (kg)	U ²³⁵ Retired (kg)	Total U Retired (kg)
0-1.5	40.0	0	1711	1839	---	---	---	---	--	---
1.5-2.5	10.0	378	0	378	9678	182	2.4	202	98	165
2.5-3.5	10.0	397	2.4	417	9501	210	5.6	249	53	131
3.5-4.5	10.0	376	5.7	415	9323	219	9.4	273	27	123
4.5-5.5	10.0	346	9.3	401	9149	221	13.0	293	15	111
5.5-6.5	10.0	370	12.9	439	9282	230	16.8	320	0	0
6.5-7.5	10.0	366	16.3	454	9278	231	20.1	330	0	0
7.5-8.5	10.0	371	19.9	470	9276	233	22.1	342	0	0
8.5-9.5	10.0	371	22.1	478	9282	234	23.7	353	0	0
9.5-10.5	10.0	366	23.2	480	9282	234	25.4	356	0	0
10.5-11.5	10.0	372	24.5	485	9281	235	26.5	360	0	0
11.5-12.5	10.0	371	26.2	487	9282	237	27.2	363	0	0
12.5-13.5	10.0	373	27.1	490	9281	238	29.0	365	0	0
13.5-14.5	10.0	375	29.0	494	9280	238	30.5	368	0	0
14.5-15.5	10.0	374	30.5	493	9280	240	32.0	372	0	0

^aC/Th = 200, τ = 4 years, on-stream refueling, U²³³ makeup.

The moderator-to-fertile-material atom ratio in the core was varied from 150 to 240. Figures 8.1 and 8.2 show the variation of fuel cycle costs and cell peaking factors as a function of these three independent parameters. The minimum cost point observed occurs for $C/Th = 237$, $\tau_{\text{fissile}} = 2$ years, and $\tau_{\text{fertile}} = 6$ years.

To determine the magnitude of the overestimated fuel requirements calculated by GAFFOX, one case was evaluated in detail using the FEVER-7 program. Table 8.2 compares some of the pertinent physics characteristics and the fuel cycle costs as obtained from the two models. It is seen that the GAFFOX model overestimates the fuel cycle costs by about 10%.

Table 8.2
COMPARISON OF FUEL CYCLE RESULTS FROM GAFFOX AND FEVER-7 CALCULATIONS

Parameters	GAFFOX	FEVER-7
C/Th, core	175	175
Feed residence time, years	1	1.5 ^a
Breed residence time, years	6	6
Net U ²³⁵ requirements, kg/breed residence time	1515	1385
Maximum peaking factor	1.69	1.57
Fuel cycle costs, mills/kw-hr:		
Fabrication	0.24	0.20
Depletion	0.41	0.36
Recovery	0.10	0.09
Fab working capital	0.09	0.07
Fuel inventory	0.48	0.48
Recovery working capital	-0.02	-0.02
TOTAL	1.30	1.18

^aIt appears that no new feed elements are required for the first year of operation after replacing the breed regions. Thus the average life of the feed elements is 1.5 years.

Use of a graded breed in the feed-breed fuel management has several advantages relative to batch reloading of the breed. A graded breed has a smaller fission-product inventory at end of cycle, which reduces the fissile feed requirements, thereby reducing the power peaking. The GAFFOX model is directly applicable to this type of fuel management. The survey has not been completed, but some of the available results are given in Table 8.3, which indicates that the graded breed cycle optimizes at a somewhat longer exposure time than with batch breed. Also, the graded breed cycle shows fuel cycle costs 0.1 mill/kw-hr lower than are obtainable with batch refueling of the breed.

Fig. 8.1--Fuel cycle costs

Fig. 8.2--Cell peaking factor

Table 8.3
CHARACTERISTICS OF FEED-BREED WITH GRADED BREED

C/Th	175	175	237	237
τ_{feed} , years	1	2	1	2
τ_{breed} , years	6	12	6	12
Conversion ratio	0.80	0.71	0.74	0.65
Cell peaking	1.30	2.18	1.68	2.73
Fuel cycle costs, mills/kw-hr:				
Fabrication	0.23	0.12	0.22	0.12
Depletion	0.31	0.41	0.40	0.54
Recovery	0.09	0.04	0.07	0.03
Fab working capital	0.09	0.07	0.09	0.06
Fuel inventory	0.41	0.36	0.33	0.28
Recovery working capital	<u>-0.01</u>	<u>-0.01</u>	<u>-0.01</u>	<u>-0.01</u>
TOTAL	1.12	0.99	1.10	1.02

As indicated in Fig. 8.2 and Table 8.3, the power peaking factors associated with the feed-breed cycle are quite high, particularly those with attractive fuel cycle costs. Some preliminary calculations have been performed to determine the maximum peaking that can be tolerated. The feed-breed cycle has two inherent advantages over the reference HTGR fuel cycle in terms of thermal characteristics. Thermal conductivity of graphite decreases with exposure, so by virtue of the short residence time of the feed regions and the fact that the maximum peaking occurs just after insertion of the feed, higher conductivities can be used. Secondly, since the feed regions contain only uranium particles, the volume required for fuel holes is significantly reduced and a larger volume fraction can be used for coolant channels. With the standard HTGR fuel element, a peaking factor of 2.5 can be tolerated for feed-breed fuel management. Increasing the number of coolant holes per fuel hole from two to three increases the acceptable peaking factors to 2.8. None of the cases of interest studied to date have peaking factors in excess of 2.8. However, the fuel volume requirements are small enough to allow four fuel holes per fuel channel if it becomes necessary. The criteria used in evaluating the maximum allowable peaking factors were (1) maximum core pressure drop, 8.5 psi, (2) outlet gas temperature, 1500°F, and (3) maximum fuel temperature, 1500°C. In all cases, the outlet gas temperature determined the temperature limit.

EVALUATION OF FUEL AND MODERATOR RODS FOR REACTIVITY CONTROL

The reactivity changes due to fuel and burnable poison depletion in the HTGR are on the order of 0.02 to 0.03 $\Delta k/k$. This is small enough to encourage the consideration of fuel- or moderator-bearing control rods for this shim control. Their use offers the prospect of reduced average fuel loadings and increased conversion ratios, thereby reducing fuel cycle costs. Detailed calculations have not supported initial optimism in this regard, however, and

neither fuel rods nor moderator rods look attractive for HTGR applications at this time. A more detailed discussion is presented below.

It is sometimes convenient to separate the reactivity changes during normal operation into four components:

1. The temperature defect, typically 0.04 to 0.07 $\Delta k/k$ for HTGR's.
2. The xenon and samarium defect, typically 0.02 to 0.04 $\Delta k/k$.
3. The Pa^{233} defect, typically 0.02 to 0.03 $\Delta k/k$.
4. The reactivity loss due to fuel and burnable poison depletion and fission-product accumulation, other than xenon and samarium, typically 0.02 to 0.03 $\Delta k/k$.

The temperature defect is generally characteristic of the core and its fuel element. Temperature coefficient requirements (from a safety viewpoint) at operating conditions lead to temperature defects from 0.04 to 0.07 $\Delta k/k$ for the HTGR. The mode of control of this reactivity does not affect core conversion ratio, fuel load, or fuel cycle cost. The same thing can be said for the xenon and samarium defects, and to a large extent, also, for the Pa^{233} defect. The reactivity changes occur over a time interval that is a small fraction of the time between reloadings.

The depletion of fuel and burnable poison leads to long-term reactivity changes whose mode of control could influence fuel cycle costs. If control could be achieved without an increase in parasitic captures, by a decrease in the Th^{232} absorptions, benefits could accrue because of reduced fuel load requirements and, perhaps, an increased conversion ratio. Rods composed essentially of fissile material or moderator material instead of poison offer the prospect of achieving these benefits and controlling 0.02 to 0.03 $\Delta k/k$. Results are summarized in Tables 8.4 and 8.5.

Unit cell calculations were utilized in the analysis of these control concepts. The unit cell corresponded to a 7-column refueling region whose properties are shown in Table 8.6.

Table 8.4
COMPARISON OF REACTIVITY WORTHS

Percent of Core Occupied by Rod	Fuel Rods, 1 Rod/Refueling Region	Moderator Rods, 1 Rod/Refueling Region	Standard Poison Rod-Pair
1.5	0.006	---	0.18
4.0	0.015	0.005	---
14.5	0.058	0.010	---

Table 8.5
COMPARISON OF ROD SYSTEM PERFORMANCE

	Fuel Rods	Moderator Rods	Poison Rods
Reactivity controlled by rods (assumed), $\Delta k/k$	0.015	0.015	0.015
Number of rods per refueling region	1	3	1
Percent of core occupied by rods	4	12	$\ll 1$
Rod diameter, in.	8.0	8.0	0.5
Excess reactivity BOC ^a , $\Delta k/k$	0.015	0.015	0.015
Rod position EOC ^a	out	out	in
Relative fuel load in core BOC	0.93	0.85	1.0
Relative fuel density in core	0.97	0.97	1.0
In-core conversion ratio BOC:			
Critical case ($k = 1.0$)	CR +0.020	CR +0.020	CR
Most reactive case ($k = 1.015$)	CR _o -0.010	CR _o -0.040	CR _o +0.005
Excess reactivity EOC, $\Delta k/k$	0	0	0
Rod position EOC	in	in	out
Relative fuel load in core EOC	0.97	0.85	1.0
Relative fuel density in core	0.97	0.85	1.0
In-core conversion ratio EOC	CR _o -0.015	CR _o -0.045	CR _o
Relative in-core power density, kw/l:			
Beginning of cycle	1.04	1.12	1.0
End of cycle	1.00	1.12	1.0
Relative in-core specific power, kw(t)/kg (fissile):			
Beginning of cycle	1.07	1.15	1.0
End of cycle	1.03	1.15	1.0
Relative fuel cycle cost	≥ 0.99	----	1.0

^aBOC = at beginning of cycle; EOC = at end of cycle.

Table 8.6

CORE CHARACTERISTICS FOR FUEL AND MODERATOR ROD STUDY

C/U	4600
C/Th	225
Nuclei/b-cm of B ¹⁰	0.68×10^{-6}
Nuclei/b-cm of C	0.065
Nuclei/b-cm of Th ²³²	2.89×10^{-4}
Nuclei/b-cm of U ²³⁵	1.40×10^{-5}
Nuclei/b-cm of H in ZrH _{1.6}	0.05824
Nuclei/b-cm of Zr in ZrH _{1.6}	0.0364
Thermal utilization	0.615
Eta	2.03
Fraction of neutrons absorbed in Th ²³²	0.31
K _∞	1.01

Fuel Rods

The control rods composed of fuel were assumed to be of the same composition as the rest of the core, without Th^{232} . A comparison of the fuel and poison rods is shown in Tables 8.4 and 8.5. An 8-in.-diameter fuel rod giving a reactivity worth of $0.015 \Delta k/k$ seems a reasonable compromise between rod size and rod worth compared with a small poison rod, also worth $0.015 \Delta k/k$. It was assumed that the initial core was critical with the poison rod fully inserted. If fuel rods were used, they would have been fully withdrawn. To restore criticality, the fuel load would have to be reduced by 3%. Since the fuel rods comprise 4% of the core, the total in-core fuel load would be 7% less with fuel rods than with poison rods. With the reduced fuel load, the in-core conversion ratio would be 0.02 higher than that of the poison rod core.

At the end of the cycle, poison rods would be completely withdrawn, and fuel rods would be fully inserted. The conversion ratio with the fuel rods completely inserted would be less than in the base (poison rod) core because of the reduced Th^{232} load in the former. The total in-core fuel load in the fuel rod core would be about 3% less than in the base core.

The improvement of fuel cycle cost appears small considering that such a large fraction of the core volume is involved. In addition, the use of fuel rods would lead to problems in the following areas:

1. Extremely large rods are apparently necessary to control a small reactivity defect. A heavier mechanism may be necessary.
2. Rod maintenance would be complicated.
3. Rod cooling when in the least reactive position (withdrawn) might be difficult.
4. The fuel rod would probably be encased in a steel can, sizeably reducing the small advantage it has over the poison rod.
5. The cost of a fuel rod would probably be significantly larger than that of a poison rod.

Finally, there would still be a need for a large number of conventional poison rods to control the temperature, xenon, and Pa^{233} defects.

Moderator Rods

The moderator rods were assumed to be $\text{ZrH}_{1.6}$. This high-hydrogen-content material will result in an increase in the resonance escape probability when introduced into the reactor, giving a potentially large reactivity worth. However, as shown on Tables 8.4 and 8.5, the reactivity realistically available with moderator rods is small. For a 15-in.-diameter rod (15% of the total core) only about $0.01 \Delta k/k$ can be controlled and this requires a $\text{ZrH}_{1.6}$ volume fraction of only 3% in the rod. An 8-in.-diameter rod with a $\text{ZrH}_{1.6}$ volume fraction of 6% gives $0.005 \Delta k/k$ in reactivity. About $0.015 \Delta k/k$ could be achieved by using three 8-in.-diameter rods, in which case the rods would occupy about 12% of the core volume.

At the beginning of cycle, with the moderator rod fully withdrawn and with the same fuel density as existed in the base poison rod core, the core would be 0.015 $\Delta k/k$ supercritical. To restore criticality, 3% of the fuel should be removed, which when considering the core volume occupied by the moderator rods, would result in a relative fuel load of 0.85, a relative fuel density of 0.97, a relative power density of 1.12 kw/l, and a relative in-core specific power of 1.12 to 1.15. The initial conversion ratio would be about 0.02 higher than in the base poison rod core.

During operation the moderator rod would move gradually into the core, and the conversion ratio would decrease significantly as the Th^{232} absorptions were reduced.

The power density is about 12% higher than that for the base core. With a reduced fuel load, this also increases the specific power. This "gain" is illusory, however, since if a higher power density were acceptable in the moderator rod core it would be acceptable in the base poison rod core, also.

A high power spike of 1.35 exists at the core-rod interface and this would have to be suppressed, further reducing the rod worth. Rod cladding, a necessity for $\text{ZrH}_{1.6}$, would also reduce rod effectiveness.

These factors, when combined with problems of rod weight, etc., make moderator rods unattractive for HTGR applications. A more attractive application of such control concepts may be possible in a core more heavily loaded in Th^{232} than was assumed in these studies ($\text{C/Th} \approx 225$). If future HTGR designs tend in this direction, these alternative control concepts should be reexamined.

FUEL CYCLE COST PENALTY OF SiC COATINGS

Recent development work with particle coatings has shown the attractiveness of SiC coatings with respect to the metallic fission-product retention. A thin layer of SiC in this instance is applied around the buffer coating, and this in turn is surrounded by a thin layer of high-density isotropic graphite. The effect of using SiC coatings on the fuel cycle cost of a 1000-Mw(e) HTGR plant has been assessed. Since silicon has a higher neutron capture cross section than graphite, the insertion of silicon into the core results in a reduction in the conversion ratio and an increase in the fissile inventory, and consequently a fuel cycle cost penalty.

Using the assumptions given in Table 8.7, the fuel cycle cost increase is 0.038 mill/kw-hr for a 30- μ SiC coating, and 0.017 mill/kw-hr for a 15- μ coating is relative to a reference fuel cycle in which only BISO-coated particles are used. The cost penalty is directly proportional to the amount of SiC in the core. Hence, if the SiC coating thickness varies, or if the kernel diameter or buffer thickness vary, the cost penalty can be scaled directly. The above values assumed that all particles are coated with SiC. If only the fissile particle is coated with SiC, the penalty is nearly halved.

Table 8.7

ASSUMPTIONS USED IN EVALUATING SiC COATINGS

Density of fissile particle kernels, g/cm ³	8.6
Density of fertile particle kernels, g/cm ³	8.3
Density of SiC material, g/cm ³	3.0
Amount of Si in SiC, wt-%	70
Diameter of fissile particle kernel, μ	200
Diameter of fertile particle kernel, μ	350
Buffer thickness, μ	30
Metal-to-carbide weight ratio	1/1.12
Th/U ratio in fissile particle	Inert diluent
Recycle of U ²³³	Yes
Fuel lifetime, years	4
C/Th ratio, equilibrium cycle	200
Reload interval, years	1
Average conversion ratio, equilibrium core	0.76
Average specific power, equilibrium core, kw/kg	1890
Equilibrium cycle loadings:	
Thorium, kg/yr	10,000
Uranium, kg/yr	550

Task IX
ADVANCED COATED PARTICLE FUELS

BARRIER COATING DEVELOPMENT

Thermal Stability Testing

Two samples of SiC-coated particles were tested at 1400°C for one year (8765 hr) in vacuum. Particles of both samples contained a UC₂ core with successive layers of laminar carbon, SiC, and laminar carbon. All particles were intact following the examination. One sample showed an average migration distance of 1 μ , and the other a distance of 16 μ . Testing of these samples is continuing, and has reached 14,800 hr.

A second test of four particle samples with a SiC layer and two particle samples with a ZrC layer has reached 3400 hr at 1400°C. An examination at 2400 hr indicated no migration in four of the samples; uranium migration through the buffer layer in one third of the particles in one of the ZrC samples; and uranium migration through the buffer layer in 9% of the particles in one of the SiC samples.

Buffer Layer Thickness

A study was made of an apparent decrease in buffer thickness following the application of an SiC overcoat. (The same results are noticed with PyC overcoats.) Following the application of a porous buffer layer to core material, the thickness and the density of the buffer layer are determined. An overcoat of SiC is then applied, and the thickness of all coating layers can be determined routinely using radiographic techniques. When the buffer layer thicknesses are compared, it is often observed that the measured thickness with the overcoat in place is less than the former value. Table 9.1 presents two instances where the decrease in thickness averages 15%, corresponding to a volume decrease of 20%. If accepted at face value, this would indicate that there is 20% less void volume available for fission gas pressure, which must be compensated for in particle design.

Various factors could account for this apparent decrease in thickness. Firstly, it is not known whether or not the effect is real. The curvature of the SiC overcoat could shadow the radiographic plate causing the SiC layer to be read too thick and the buffer layer too thin. Supporting the supposition that there is no penetration of the buffer layer is the fact that the deposition of the very porous buffer layer is immediately followed by the application of a dense, thin, carbon layer to act as a shell, making the buffered particles easier to handle in subsequent operations. This shell is seldom observed, and is perhaps 2 μ thick. It is this dense surface that would be affected by subsequent overcoats, not a soft, spongy layer. The second thickness reading is never greater than the first, and is usually less. The decrease may be related to the buffer density, although the dense shell layer would seem to negate this supposition.

Table 9.1
BUFFER THICKNESS OF SiC-COATED PARTICLES

Buffered Material	Overcoated Material	Buffer Thickness (μ)			Decrease in Avg Thickness (%)	Buffer Density (g/cm^3)
		Max	Min	Avg		
3592-21E	---	62	37	44	--	1.55
	3592-23E	62	35	44	0	--
	3592-27E	57	29	44	0	--
3592-33E	---	60	24	39	--	1.38
	3592-35E	43	25	32	18	--
	3592-45E	53	23	35	10	--
3583-61E	---	111	29	51	--	1.21
	3592-69E	68	27	45	12	--
	3592-71E	51	29	41	20	--

In perspective, this discussion does not point to a problem area of concern; it is an observation that has been made. The trend will be checked occasionally, but no serious study is foreseen.

Zirconium Carbide Coatings

Effort is continuing on zirconium carbide barrier coatings, as zirconium carbide in thicknesses from 10 to 15 μ is well worth continued study for coated particle development. It is relatively easy to apply, it is a barrier for fission-product release, and it allows a separation of particles in a two-particle fuel system. There is a penalty to be paid in the neutron cross section of zirconium, but a 10- μ -layer of ZrC is approximately equivalent to a 30- μ -layer of SiC, which is also being evaluated.

Deposition of ZrC coatings on fuel particles has been demonstrated, and the few neutron activation tests that have been made have indicated very low fission-product release. However, these coatings, which are 10 to 12 μ in thickness, are not massive enough for other physical property measurements. Equipment and procedures have been modified in an attempt to prepare coatings 50 μ thick, but reproducibility has not yet been proven. The preparation of the vapor phase of the zirconium appears to be the problem. Zirconium tetrachloride is the raw material used, and a parameter study involving the temperature of the material and the total flow through the material is in progress. The rate of zirconium vapor production decreases with time, and this prevents the establishment of good process control.

Preliminary results indicate that the best conditions are a vaporization temperature of 230°C and a hydrogen flow of 1000 to 1500 cm^3/min through the ZrCl_4 bed.

Properties of Metal Carbide Coatings

Electrical Resistivity of Vapor-deposited SiC. Since silicon carbide is a semiconductor, its electrical resistivity is very sensitive to the presence of impurities and to irradiation damage. Measurements of electrical resistivity are therefore of interest as indicators of impurity levels, and resistivity changes may be studied to provide information on the neutron irradiation damage. In particular, the temperature at which irradiation-induced resistivity changes start to appear during postirradiation annealing may be an indication of the irradiation temperature. The magnitude of the change is more difficult to interpret because it is affected by such factors as the change in doping level by neutron-induced transmutations in the impurities.

The resistivities of a series of silicon carbide strips deposited from a methyltrichlorosilane-hydrogen mixture at 1400°C were measured at room temperature. The strips measured 6 x 1 x 0.1 mm and the surfaces were diamond-lapped but unetched. The measuring apparatus consisted of a four-probe arrangement in which the samples were clamped between copper contacts. The voltage was then measured via gold-plated probes spaced 2.5 mm apart. The current flow was set at 5 μ amp and the probe voltages measured 20 v.

The resistance values were independent of the current flow direction, indicating that contact rectification was not a problem. The readings were also the same regardless of whether the current contacts were applied to the inside of the deposit, the outside of the deposit, or both sides of the deposit. A series of measurements with the current increasing from 1 to 10 μ amp showed that the conductivity was ohmic. There was a considerable variation in resistivity from sample to sample. The mean value was 1050 ± 240 Ω -cm, which compares with published values of 200 to 300 Ω -cm for pyrolytic silicon carbide deposited on a stationary substrate (Ref. 1) and 10 to 30 Ω -cm for commercial porous silicon carbide (Ref. 2). The measured samples were companion samples to those which are being irradiated in capsule P13J.

Hot-probe measurements of the polarity of the Seebeck coefficient showed that the material is an n-type semiconductor.

Thermal Conductivity of Vapor-deposited SiC. The use of the "flash" technique (Ref. 3) to measure the thermal conductivity of pyrolytic silicon carbide was described in the previous quarterly report. Because the measured values (about 0.1 cal/cm-sec-°C) were low in view of the purity of the material, it is considered possible that the results were affected by the interface resistance between the carbide and the layer of nickel applied to facilitate thermocouple contact on the rear face of the sample and to render the front face opaque. In order to test this possibility further, composite samples of silicon carbide and polycrystalline graphite were measured and the conductivities calculated using a two-layer analysis (Ref. 4). The graphite layer was then removed from the silicon carbide and the carbide samples were cut into three pieces. The pieces then had one face coated with evaporated silver, a thin layer of colloidal silver paint, and a graphite "smear". Preliminary results indicate that the conductivity calculated for

the silicon carbide layer is independent of the material used for the conductive backup layer. This suggests that interface resistance is unimportant and the data obtained previously are valid.

Irradiation Effects in Vapor Deposited SiC. Samples of SiC and ZrC were included in the irradiation capsule P13J. The samples were divided between two crucibles; the anticipated sample temperatures are 500° and 1000°C, and the exposures will be 2.5 to 5×10^{21} nvt and 3 to 6×10^{21} nvt, respectively. The crucibles contain 17 diamond-lapped 1-mm-wide SiC strips, four 2.5-mm-wide SiC strips, and five ZrC slivers. The narrow SiC strips will be used to measure changes in dimensions, density, mechanical properties, and electrical resistivity. The wide SiC strips will be used for measurements of thermal conductivity. The ZrC samples are being included as a preliminary test to discover whether vapor-deposited ZrC retains its integrity under irradiation. This test is important because it has been reported (Ref. 5) that hot-pressed and slip-cast-sintered ZrC can fracture under some conditions of irradiation. If the pyrolytic ZrC samples survive irradiation, their dimensional changes will be measured.

Deposition of Zr and ZrC. The conditions under which Zr or ZrC may be deposited pyrolytically in a graphite tube from their halides are determined by the temperature and pressure of the halide. At a given temperature there is one "limiting pressure" below which the metal is deposited. As the pressure is increased, formation of the carbide is favored until a second limiting pressure is reached where the carbide is attacked. The parameters for calculating limiting pressures for the deposition of zirconium and zirconium carbide from zirconium chloride and zirconium iodide have been given by Powell (Ref. 6).

A study of the pressure and temperature regions in which each material is deposited, based on Powell's parameters, shows that ZrCl_4 is a suitable starting material for the deposition of ZrC in the temperature range 1700° to 2200°C, but that the deposition of Zr would require such low pressures of ZrCl_4 (about 0.1 torr) that deposition rates would be very low. Zirconium iodide is a better starting material for the deposition of Zr, which is formed at pressures of several torr at around 1500°C.

Coating Contamination Analysis

The presence of fission products resulting from coatings with contaminating uranium has hampered the interpretation of fission-product release data. It is postulated that uranium is trapped in the coatings during the coating process and diffuses outward from the source. The results of alpha-counting analysis for uranium before and after anneal of the coated particle cannot be used to evaluate the merits of diffusion barrier layers, since the source of the contaminant is unknown. An experiment was carried out to determine the following data:

1. The effect of coating temperature on the degree of uranium and thorium contamination.
2. The effect of the Th:U ratio on the contamination at the various temperatures.
3. The effectiveness of alpha analysis of the degree of contamination as compared with activation analysis.

In the first series, 50 g of depleted UC_2 uncoated particles (surface area $\approx 1400 \text{ cm}^2$) were run at 1200° , 1600° , and 2200°C . In each run fifteen graphite disks (17.2 cm^2 surface area) were added to absorb the contamination present during the operation. Only carrier gas was used and no actual coating was done. The disks were then alpha counted for uranium activity. Following the alpha analysis, the disks were activated in the TRIGA reactor and the amount of uranium contaminant was determined by gamma counting.

In the second series, a 50-g charge of depleted $(Th,U)C_2$ uncoated particles with a Th:U ratio of 5:1 was used and temperatures of 20° , 1200° , 1400° , and 1600°C were investigated.

The results of the two series are tabulated in Table 9.2. The following trends seem evident:

1. The amount of free uranium in the coating chamber and adsorbed on the disks decreases with increasing coating temperature.
2. There is considerable transfer of both thorium and uranium at room temperature, due only to mechanical agitation in the 5:1 fuel.
3. The transferred thorium varies considerably from stoichiometric release as the temperature varies.
4. The uranium release from the 5:1 fuel varies only slightly over the temperature range used.

It should be noted that the same charge of fuel was used across the temperature range in each series. A fuel particle surface depletion effect may be in operation in this instance. On the other hand, diffusion within the particles may offset the effect. These possibilities can be evaluated in a duplication of the run using a new fuel charge for each temperature. At the conclusion of each series, the degree of contamination of the coating tubes was checked by running new disks without fuel across the temperature range. The results, shown on Table 9.2, indicate that the contamination on the tube used in the UC_2 runs is released at higher temperatures. The contamination from the tube used in the 5:1 runs, both thorium and uranium, showed higher release at 1200°C .

The table shows that it is difficult to analyze contamination via alpha analysis in $(Th,U)C_2$ fuel. More U^{23+} (enriched fuel) would increase the uranium alpha activity, but the presence of Th^{228} and its volatile alpha-emitting daughters virtually masks the uranium-produced alpha activity.

DILUENT PARTICLE DEVELOPMENT

Previous work has indicated that void, carbon, and to a lesser extent beryllium are the most promising potential diluents for HTGR fuels. Since void most nearly represents an ideal diluent from a theoretical standpoint, work during the current reporting period has been directed toward determining the feasibility of preparing porous fuel particles. In addition, studies directed toward optimizing the design of diluent particles using an analytical model were continued.

Table 9.2
CONTAMINATION ANALYSIS

Sample	Fuel	Coating Temp. (°C)	Coating Time (min)	Alpha Analysis		Activation Analysis ^a	
				α_1 /hr	Delay 48 hr α_2 /hr	Total U(μ g)	Total Th(μ g)
1	UC ₂	1200	15	1500	1530	493	0
2	UC ₂	1600	15	950	990	449	0
3	UC ₂	2200	15	204	202	117	0
4	UC ₂ tube	1200	15	6	---	16	0
5	UC ₂ tube	1600	15	0	---	7	0
6	UC ₂ tube	2200	15	10	---	37	0
7	5:1	20	15	826	152	78	202
8	5:1	1200	15	9800	1600	113	915
9	5:1	1400	15	1.27x10 ⁵	19,700	69	552
10	5:1	1600	15	1.29x10 ⁵	20,000	64	230
11	5:1 tube	20	15	240	38	5	8
12	5:1 tube	1200	15	442	72	19	15
13	5:1 tube	1400	15	380	52	2	7
14	5:1 tube	1600	15	---	122	16	4

^aU as Np²³⁹, Th as Pa²³³.

Preparation of Spherical Uranium Oxide - Carbon Particles

Powder-agglomeration Method. In the powder-agglomeration method, a UO₂-carbon mixture is blended with binder and formed into granules which are spheroidized by rolling in a planetary mill. Initial experiments using this process indicated that the type and amount of binder, the size and method of granule preparation, the amount of granules loaded into the bowl of the planetary mill, its rotation (milling) speed, and time are all important variables in the spheroidization process. Further investigations conducted this quarter have shown that the method of granule preparation is one of the most important variables in the spheroidization process. Therefore extensive investigations of various methods of granule preparation have been conducted. These include the following:

1. Compaction granulation (compaction of the powder-binder mix followed by crushing and sieving, as described in the previous quarterly report).

2. Crushing and sieving the powder-binder mixture.
3. Extrusion sieving (forcing the powder-binder mixture through sieves by hand).
4. Mechanical-extrusion sieving (forcing the powder-binder mixture through a colander sieve using a Hobart mixer).
5. "Meat-grinder" extrusion (feeding a powder-binder mixture through a conventional meat grinder).

Methods 1 and 2 above result in granules with a length-to-diameter ratio of about 1, while methods 3, 4, and 5 result in needle-like granules with a length-to-diameter ratio greater than 2.

Results of the studies mentioned above indicate that spheroidization can be accomplished more reproducibly, in less time, and with less sensitivity to charge weight, milling speed, and surface area of the starting powders, when granules are formed using methods 3, 4, and 5. (For example, the spheroidization time was decreased from 15 min to 3 min by using these methods rather than method 1. Method 2 resulted in granules which powdered during spheroidization. At the same time the charge weight of granules loaded into the planetary mill was increased from 20 to 100 g. In general, these studies indicated that the most promising granulation techniques are methods 4 and 5. Therefore further powder-agglomeration work will concentrate on these methods.

In addition to the above experiments, the mineral oil content of the powder mixture was varied in an effort to increase the plasticity of the granules, thereby increasing the ease of spheroidization. Mineral oil contents ranging between 10 and 25 wt-% were used. By using a binder content of 21 wt-% (rather than 17 wt-%) the spheroidization time for granules prepared by mechanical extrusion sieving was lowered from 3 min to 5 sec.

Results of all studies conducted thus far indicate that the following is the preferred powder-agglomeration particle preparation procedure:

1. Granulation by any of the three extrusion techniques (methods 3, 4, and 5 listed above).
2. Use of 17 wt-% mineral oil binder.
3. A maximum planetary mill charge weight of 100 to 150 g per bowl.
4. Spheroidization for 3 min at about 250 rpm.

The conditions listed above are dependent upon the surface area of the starting powders. For the experiments described above the surface area of the UO_2 and carbon powders was about 7 m^2/g as determined by BET N_2 adsorption.

Sol-Gel Method. Investigation of spherical oxide-carbon particle preparation utilizing a Sol-Gel method continued. Current studies have been concerned with the expansion of previous work done on a basic uranium (VI) sol prepared by peptization of uranium peroxide ($\text{UO}_4 \cdot 2\text{H}_2\text{O}$) with tetraethylammonium hydroxide (TEAH).

Uranium peroxide was prepared for sol preparation by two methods: UO_2 digestion and $\text{UO}_2(\text{NO}_3)_2$ precipitation. In UO_2 digestion a quantity of UO_2 powder was digested in a 5% formic acid - 20% hydrogen peroxide - 75% water solution (numbers are volume percent) at 70°C . The black uranium dioxide powder was rapidly replaced by the yellow-white uranium peroxide precipitate. In the $\text{UO}_2(\text{NO}_3)_2$ precipitation method the uranium peroxide was precipitated from an acidic uranyl nitrate solution by addition of an excess of hydrogen peroxide.

Sols were successfully prepared from both the UO_2 digestion product and the $\text{UO}_2(\text{NO}_3)_2$ precipitation product by peptization with TEAH. The UO_2 digestion sol was definitely basic ($\text{pH} \approx 10$) while the $\text{UO}_2(\text{NO}_3)_2$ precipitation sol was more of a neutral sol ($\text{pH} \approx 7$). This effect was thought to be the result of the presence of excess nitrate, since no precautions were taken to efficiently wash the uranium peroxide obtained from the $\text{UO}_2(\text{NO}_3)_2$ precipitation method. These sols also have a tendency to gel at room temperature in air when mixed with a minimum of water. The addition of more water prevents this. Work is continuing on the comparison of the two sols.

No attempts were made to produce spherical particles. However, some observations of the gelling characteristics of sphere-like material produced by spraying sol into a dehydrating agent with a hypodermic needle and syringe were made. From this it became evident that acidic 2-ethylhexanol was an applicable gelling and dehydrating agent. Much work still needs to be done on acid concentration, gelling medium temperature, and other such parameters necessary for most efficient application.

Subsintering Conversion

Subsintering conversion is being investigated as a method of preparing void-diluted UC_2 fuel particles. It involves converting below sintering temperature to ensure that voids, present before and formed during conversion, are retained. Studies have been directed toward investigation of conversion temperatures and evaluation of the resulting carbide particles.

Properties of porous UC_2 particles prepared at various conversion temperatures in vacuo are shown in Table 9.3. Photomicrographs of typical particles are shown in Fig. 9.1. X-ray diffraction indicated the particle material to be UC_2 with some C and UC present. The range of density values reported in Table 9.3 is assumed to be the result of slightly different conversion and spherical particle preparation procedures. Further work is in progress to define the shape of the density versus conversion temperature curve. In general, however, the data indicate the following:

1. Densities in the range of interest (about 50% of theoretical density) can be reproducibly obtained.
2. Density is a function of the surface area of the graphite flour used as well as conversion temperature (e.g., coarser graphite gives higher density).
3. No change in density is found for conversion temperatures in the range 1700° to 2100°C .

Table 9.3

PROPERTIES OF POROUS UC₂ PARTICLES PREPARED BY SUBSINTERING CONVERSION

Sample Number	Carbon Type	Conversion Conditions		Uranium (%)	Carbon (%)	Density (g/cm ³)	% of T.D. ^a
		Time (hr)	Temp. (°C)				
JT5-127-60	Fine ^b	} 31	1400	(d)	(d)	5.1	44
JT5-127-68	Fine ^b			(d)	(d)	5.0	43
JT5-77-1	Fine ^b	} 3	1500	(d)	(d)	5.0	43
JT5-77-2	Coarse ^c			(d)	(d)	8.6	74
JT5-77-3	Coarse ^c			(d)	(d)	7.9	68
JT5-77-4	Coarse ^c			(d)	(d)	8.2	70
JT5-93-46	Coarse ^c			(d)	(d)	9.0	77
JT5-101-64	Fine ^b	} 3	1600	88.4	11.1	5.1	44
JT5-114-66	Fine ^b			(d)	(d)	5.8	50
JT5-48-1	Fine ^b	} 3	1600	90.3	10.5	5.6	48
JT5-48-2	Fine ^b			90.2	10.6	6.0	51
JT5-54-1	Fine ^b			90.2	10.2	5.5	47
JT5-54-2	Fine ^b			89.6	10.7	5.5	47
JT5-72-1	Fine ^b	} 1-1/2	1600	(d)	(d)	5.6	48
JT5-72-2	Coarse ^c			(d)	(d)	9.3	80
JT5-72-3	Coarse ^c			(d)	(d)	8.9	76
JT5-72-4	Coarse ^c			(d)	(d)	8.9	76
JT5-104-66	Fine ^b	} 1	1700	(d)	(d)	5.7	48
JT5-106-66	Fine ^b			(d)	(d)	6.3	54
JT5-108-66	Fine ^b	1	1900	(d)	(d)	6.2	53
JT5-51-1	Fine ^b	} 1	2100	90.2	10.4	5.9	50
JT5-51-2	Fine ^b			90.0	10.6	6.0	51
JT5-57-1	Fine ^b			(d)	(d)	6.5	56
JT5-57-2	Fine ^b			(d)	(d)	6.5	56
JT5-57-4	Coarse ^c			(d)	(d)	9.4	--
JT5-112-66	Fine ^b	} 1	2300	(d)	(d)	6.5	56
JT5-47	Fine ^b			89.7	10.8	6.8	80

^aCalculated using 11.7 g/cm³ as a theoretical density for UC₂. Numbers are rounded off to the nearest percent.

^bParticle size nominal 1 μ ; surface area by BET N₂ adsorption 7 m²/g.

^cParticle size <75 μ ; surface area by BET N₂ adsorption 3.5 m²/g.

^dAnalysis in progress.

(a)

(b)

Fig. 9.1--Photomicrographs of porous UC_2 particles prepared by sub-sintering conversion at 1500°C in vacuo; (a) macrophotograph (22X), (b) cross-sectional metallograph (200X), density = 5.1 g/cm^2 .

4. 1500°C is the lowest temperature for effecting complete conversion in a reasonable time (31 hr at 1400°C was necessary for complete conversion while 3 to 6 hr at 1500°C was sufficient).

The dependence of the final density on the surface area of the graphite powder used in the powder-agglomeration process is presumed to be due to the fact that a less homogeneous $\text{UO}_2\text{-C}$ mixture results when a coarser graphite is used. This would allow more contact between UC_2 grains as they are formed, thereby increasing the possibility of sintering at lower temperatures. Further investigation of this effect is in progress.

Diluent Particle Design Studies

Methods of predicting coated particle irradiation performance through the use of a mathematical model (Ref. 7) are being investigated in an attempt to optimize the design of diluent particles. These studies are directed toward maximizing the uranium loading for any given coated particle design parameter (core diameter, total coating thickness, core density, etc.). Results of initial studies indicated that significant increases (a factor of 2) in uranium loading may be possible by optimizing design parameters. Further calculations performed during this reporting period have demonstrated that increases in uranium loading of 1.5 to 2 may be possible if void is used as the diluent. These design studies are incomplete.

CHEMISTRY STUDIES

Postactivation Release Studies

Annealing tests were conducted at 1400°C on samples of SiC-coated particles irradiated in the TRIGA reactor to about 10^{14} fissions. The Ba^{140} , Sr^{91} , and Xe^{135} release data are given in Table 9.4.

Postirradiation Release Studies

The release of metallic fission products from SiC- and PyC-coated particles is being compared in long-term annealing tests. The particles selected for these experiments are from lots 3332-139E (300- to 400- μ (Th,U) C_2 kernels, Th:U = 12:1; coatings, 46- μ buffer, 32- μ SiC, 34- μ isotropic PyC) and 3222-71E (150- to 250- μ (Th,U) C_2 kernels, Th:U = 3:1; coatings BISO-2200, 29- μ buffer, 68- μ isotropic PyC) irradiated at 1300°C in the P13F capsule. These experiments will consist of (1) initial release measurements, (2) long-term (about 200 hr), high temperature (about 1400°C) anneals, and (3) additional release measurements. The releases of these samples were initially measured at 1400°C for 140 hr. The fractions of cesium, cerium, and strontium released were measured at approximately 20-hr intervals. The cesium and cerium release data are given in Figs. 9.2 and 9.3. The strontium data are not yet available. The samples were then transferred to a tantalum crucible containing activated charcoal, which should act as a fission-product sink, preventing back-diffusion of the fission products. The tantalum crucibles were then sealed by welding. These particles are being annealed at 1400°C. Initially, weekly release measurements on the particles will be made.

Table 9.4

FRACTIONAL RELEASE FROM SiC-COATED FUEL PARTICLES

Lot and Kernel Description	Coating Thickness (μ)				Anneal		Fractional Release		
	Buffer	SiC	Isotropic	Total	Time (hr)	Temp. ($^{\circ}$ C)	Xe ¹³⁵	Ba ¹⁴⁰	Sr ⁹¹
3592-35E (Th,U)C ₂ 30.1% U 59.9% Th 150-250 μ	39	15	10	64	Recoil ^a	1400	2.0x10 ⁻⁵	---	---
					1		---	1.3x10 ⁻⁶	4.6x10 ⁻⁵
					3		---	5.9x10 ⁻⁶	1.1x10 ⁻⁴
					5		2.0x10 ⁻⁶	1.4x10 ⁻⁴	3.2x10 ⁻⁴
					8		---	3.0x10 ⁻⁴	4.0x10 ⁻⁴
3592-53E (Th,U)C ₂ 30.1% U 59.9% Th 150-250 μ	39	10	52	101	Recoil ^a	1400	1.9x10 ⁻⁵	---	---
					1		---	4.9x10 ⁻⁶	6.1x10 ⁻⁵
					3		---	8.2x10 ⁻⁶	4.4x10 ⁻⁴
					5		<1.0x10 ⁻⁷	5.1x10 ⁻⁵	6.4x10 ⁻⁴
					8		---	1.1x10 ⁻⁴	1.5x10 ⁻³
3592-69E (Th,U)C ₂ 30.1% U 59.9% Th 150-250 μ	51	29	10	90	Recoil ^a	1400	<5.2x10 ⁻⁷	---	---
					1		---	<2.8x10 ⁻⁷	3.3x10 ⁻⁷
					3		---	---	2.6x10 ⁻⁶
					5		4.2x10 ⁻⁷	---	6.1x10 ⁻⁶
					8		---	<3.0x10 ⁻⁷	1.1x10 ⁻⁵
3592-71E (Th,U)C ₂ 30.1% U 59.9% Th 150-250 μ	51	35	10	96	Recoil ^a	1400	3.8x10 ⁻⁸	---	---
					1		---	<7.8x10 ⁻⁷	2.3x10 ⁻⁷
					3		---	---	1.2x10 ⁻⁶
					5		4.5x10 ⁻⁸	---	3.8x10 ⁻⁶
					8		---	<8.0x10 ⁻⁷	7.4x10 ⁻⁶

^aRecoil release as determined by Xe¹³⁵ present in the irradiation chamber.

Fig. 9.2-- Cs^{137} release at 1400°C from particles from lots 3332-139E and 3222-71E irradiated at 1300°C in the P13F capsule

Fig. 9.3-- Ce^{144} release at 1400°C from particles from lots 3332-139E and 3222-71E irradiated at 1300°C in the P13F capsule

The cesium release data for the PyC- and SiC-coated particles were essentially the same for about 70 hr. After this, the PyC-coated sample showed a marked increase in release (Fig. 9.2). The Kr^{85} released from both samples was measured and remained low (fractional release less than 10^{-4} , the detection limit under experimental conditions), thus establishing the integrity of the coatings. It was noted that the cesium release continued to increase for the PyC-coated particles throughout the remaining 140 hr of annealing, whereas the release from the SiC-coated particles remained essentially constant after 70 hr of annealing. The cerium release from the PyC-coated particles was a factor of 5000 higher than the cerium release from the SiC-coated particles at the end of 140 hr of annealing (Fig. 9.3).

Fission-product Release Measurements in the High-level Furnace

A furnace designed to be operated with samples reading approximately 200 r/hr has recently been placed in operation. Improved precision in release measurements from irradiated particles can be attained by the use of samples containing a larger number of particles than have been used in the past in these experiments. Fission-product release measurements on 100 high-burnup fuel particles from lot 3332-149E (150- to 250- μ (Th,U) C_2 kernels, Th:U = 3:1; coatings, 23- μ buffer, 40- μ isotropic PyC, 22- μ SiC, 6- μ isotropic PyC) have been initiated. The cesium and cerium release data for this sample at 1000°C are given in Fig. 9.4. Two additional samples will be annealed at 1200° and 1400°C, respectively, to determine the activation energies for release of the metallic fission products.

Studies of Uranium Contamination in Coatings

The release of the metallic fission-product nuclides barium and strontium is substantially lower from SiC-coated particles than from PyC-coated particles. However, at least some of the barium and strontium released from SiC-coated particles appears to originate in the outer isotropic PyC layer. The uranium contamination of this layer apparently occurs during the coating operation. Several approaches for the determination of the level of contamination have been suggested and are being pursued.

Studies were initiated this quarter to determine the level of uranium contamination in the outer isotropic PyC layer of SiC-coated particles. Samples containing approximately 0.5 g of particles each from lots 3332-101E, 3332-139E, 3332-147E, and 3332-153E, which were used in the Pl3F capsule, were given light irradiations (10^{13} fissions) in the TRIGA reactor. The outer isotropic PyC layer was subsequently burned off at 1000°C in 1.5 hr in a 10% oxygen atmosphere. The Xe^{135} released was collected in a charcoal trap at liquid-nitrogen temperature. The metallic fission products that plated out on the furnace tube were removed with concentrated nitric acid. Gamma spectroscopy of these solutions revealed that substantial amounts of the fission products were present. In some cases the fractions of the fission products present in these solutions were higher than one would expect from simple uranium contamination of the outer isotropic PyC layer. Where the acid solutions contained high fractions of the fission product (fraction $> 5 \times 10^{-4}$), microscopic examination of the particles revealed that broken particles were present in these samples. These experiments will be repeated, and the oxidation will be lowered to 600°C in an attempt to prevent the particle breakage.

Fig. 9.4--Fission product release from lot 3332-149E at 1000°C of P13F capsule (100 particles)

The outer pyrocarbon coatings of high-burnup P13F capsule particles were burned off in experiments similar to that described above. Ten particle samples from lots 3332-101E (150- to 250- μ (Th,U) C_2 kernels, Th:U = 3:1; coatings, 58- μ buffer, 13- μ SiC, 40- μ isotropic PyC; burnup 12% FIMA at 900°C) and 3332-149E (150- to 250- μ (Th,U) C_2 kernels, Th:U = 3:1; coatings, 23- μ buffer, 40- μ isotropic PyC, 22- μ SiC, 6- μ isotropic PyC; burnup 12% FIMA at 1300°C) were annealed at 1000°C for 1.5 hr in a 10% oxygen atmosphere. The only fission products present in the effluent gas in detectable amounts were Ru^{103} and $Zr^{95}-Nb^{95}$. Calculations from these data indicated that about $6.0 \times 10^{-4}\%$ of the total uranium present was in the outer layer of the coating. In these preliminary studies the absence of Ce^{144} , Cs^{137} , and Ba^{140} indicated that a substantial loss of fission products occurred during the irradiation in the ETR and that transport of the species from the SiC layer to the outer PyC layer is extremely small. Microscopic examination of these particles revealed no broken coatings.

To differentiate between release resulting from contamination in the outer PyC coatings and that resulting from migration through the SiC coatings, postirradiation fission-product release measurements were conducted at 1200°C on the two samples that no longer had outer pyrocarbon coatings. The cesium and cerium release data for these experiments are given in Table 9.5. These preliminary data indicate that fission-product release from the SiC barrier layer is low.

Table 9.5
CESIUM AND CERIUM RELEASE DATA

Sample Lot No.	Anneal Time (hr)	Fractional Release at 1200°C	
		Cs^{137}	Ce^{144}
3332-101E	22.6	---	1.2×10^{-6}
	45.1	---	3.6×10^{-6}
	87.1	$< 4.7 \times 10^{-6}$	5.8×10^{-6}
3332-149E	22.6	$< 6.7 \times 10^{-6}$	1.6×10^{-6}

Diffusion of Beryllium in Pyrolytic Carbon

Attempts to measure the diffusion of beryllium in pyrolytic carbon by using a source of pure Be_2C powder as a constant chemical potential source have been futile. No diffusion occurs below 1250°C, and above this temperature a hard layer of Be_2C is formed on the surface of the pyrolytic carbon. A possible explanation is given by the reaction of Be_2C with the tantalum container which results in the high activity of the excess beryllium. Experiments have been performed, therefore, in which an excess of carbon is present along with the beryllium.

Attempts to diffuse beryllium into pyrolytic carbon using a constant chemical potential source of 1.0×10^{-3} g Be/g C by annealing for 40 hr at

1700°C were unsuccessful. No penetration occurred. Also, during the anneal most of the beryllium was absorbed on the inner surfaces of the screw-top tantalum container, leaving the constant chemical potential source of 6.7×10^{-5} g Be/g C at the end of the run. When the experiment was repeated using a graphite container, all the beryllium diffused out of the container.

Another experiment was performed using a higher-level source of 5.0×10^{-2} g Be/g C and a tantalum container that was sealed by welding, and a similar result was obtained. Annealing for 90 hr at 1700°C yielded no penetration into the pyrolytic carbon and, again, most of the beryllium was absorbed on the inner walls of the tantalum container, leaving the constant chemical potential source with a loading of 2.5×10^{-3} g Be/g C.

It has been found that a pyrolytic-carbon-coated wafer exposed to a Be_2C powder source at 1250°C will form a hard shell of what is presumed to be dense Be_2C . The physical appearance of the pyrolytic carbon coating is unchanged. The specimen cannot be lapped with the usual 4/0 emery paper; only a high polish is produced on the surface. One such specimen produced by annealing a wafer in Be_2C powder (Be^7 -tagged) at 1250°C for 400 hr was used as a diffusion source by pressing it between two pyrolytic-carbon-coated wafers. The wafers were stacked inside a screw-top graphite container along with some uncoated disks that were used as spacers. The screw top was tightened against the stacked wafers to maintain good contact with the source. Annealing was performed for 19 hr at 1700°C. At the end of the run the two fresh wafers were stuck to the source wafer. It was possible to free one of the wafers and lapping of the surface that had been exposed to the Be_2C -coated wafer yielded a concentration profile from which a diffusion coefficient of 8.7×10^{-10} was obtained. Inspection of the exposed surface before lapping indicated that it had begun to fuse in several places with the Be_2C -coated source wafer, and it may be that the Be_2C penetration in this interaction was deep enough to produce what appeared to be a diffusion profile.

In summary, beryllium diffusion in pyrolytic carbon is slow, and unequivocal diffusion data have not yet been obtained. Pyrolytic carbon is relatively impervious to low levels of beryllium vapor at temperatures up to 1700°C. Physical contact with Be_2C powder at temperatures of 1400° to 1700°C results in degradation of the pyrolytic carbon coatings. Exposure to Be_2C powder at 1250°C produces an extremely hard layer of what is presumably Be_2C without any apparent degradation.

A set of pyrolytic-carbon-coated wafers will be annealed in Be^7 -tagged Be_2C to produce samples with the hard layer of Be_2C , which can then be used to characterize this material. A sample of pyrolytic-carbon-coated fuel particles will be treated similarly to produce an outer coating of Be_2C . These will then be tested for retention of metallic fission products.

COATED PARTICLE IRRADIATIONS

A series of capsule irradiations is being conducted to evaluate advanced types of coated particles under potential HTGR irradiation environments. Current tests are directed toward the evaluation of metal carbide barrier

coatings to guide the development of coated particles with improved fission-product retention. Future tests will also include irradiation evaluation of fuel particles containing diluents other than thorium. Additional irradiation experiments in support of the HTGR program are being conducted at ORNL. These include several high-burnup tests of advanced coated particles and tests of SiC barrier-coated particles.

General Atomic Capsules

Capsule P13H. Capsule P13H was a high-fast-flux test of SiC- and ZrC-barrier-coated particles. The nine samples tested included various metal carbide coating thicknesses and layer combinations with PyC on fissile and fertile particles. Emphasis was placed on evaluating the relative merits of the SiC layer versus the PyC layer for directly bearing the pressure load resulting from internal fission gas accumulation. Three of the samples were tested at two different irradiation temperatures (1000° and 1300°C) to evaluate possible thermal effects on stability. One sample of BISO-coated particles is also included for comparison. A detailed description of the coated particle samples, including deposition conditions and microstructural parameters, was presented in an earlier Base Program quarterly report, GA-7553.

The P13H capsule has completed its five-cycle irradiation in the ETR and has been returned for postirradiation examination. It operated successfully with near-design fuel temperatures of 1150° to 1325°C in cells two through five and 925° to 1000°C in cell one. The fuel reached estimated burnups of 27% FIMA for fissile particles and 10% FIMA for fertile. The design burnups were 20% and 6% FIMA, respectively, in four cycles. The fast-neutron exposure is estimated as 4.0×10^{21} nvt. Design exposure was 3 to 6×10^{21} nvt.

Capsule P13J. Capsule P13J is a test of SiC-barrier coated particles designed to evaluate the limits of barrier coating stability. The design of the coated particle samples has been guided by an analytical model of coated particle performance under irradiation (Ref. 7). Some of the samples are designed to survive and other are designed to fail. The results will aid in evaluating the accuracy of the model. Another objective of the P13J experiment is to determine the effect of irradiation temperature on the stability of SiC barrier coatings. Irradiation data given in the previous quarterly report from the P13F experiment on unrestrained SiC strips have shown that the saturated linear expansion of SiC increases with decreasing irradiation temperature. Thus, several samples are being tested in triplicate at design irradiation temperatures of 1300°, 1000°, and 600°C. A third objective is to compare the relative stabilities of SiC-barrier and BISO coatings.

The nine samples being tested include seven with SiC-barrier coatings and two with BISO coatings. Three of the barrier-coated samples are designed to evaluate the effect of SiC layer thickness on stability with the SiC as the internal pressure load-bearing layer. These have SiC layers of 15, 25, and 37 μ with constant buffer thickness (39 μ) and density (1.4 g/cm³), and all are overlaid with thin (8 to 10 μ) PyC layers. A second group of samples has thick outer PyC (31 and 52 μ) as the load-bearing layer with a thin (10 μ) inner SiC layer for fission-product retention and the same buffer

thickness (39 μ). A third group is designed to evaluate the effect of buffer thickness and density on stability with thicker (51 μ), lower density (1.2 g/cm³) buffer layers. These are overlaid with SiC layers of 29 μ and 35 μ and thin (10 μ) PyC overcoats. This group will give a direct comparison with the first group of SiC load-bearing samples on the effects of buffer structure. Finally, two BISO-coated samples have been included, one with a low-temperature isotropic PyC layer and the other with a high-temperature isotropic layer. These have different ratios of buffer-to-isotropic thickness. One is designed to give a direct comparison of the relative stabilities of BISO and SiC barrier coatings, since it has the same buffer thickness (51 μ) and total outer "pressure vessel" layer strength as one of the barrier coated samples. All samples are on 200- μ fissile-size (Th,U)C₂ cores with a Th:U ratio of 2:1. Detailed descriptions of the coated particle samples are given in Table 9.6.

The capsule is designed to be irradiated for three to six ETR cycles at fuel temperatures of 1300° (three cells), 1000°, and 600°C. Design fuel burnup is 20% FIMA in four cycles. The fast-neutron dose will be 3 to 6 x 10²¹ nvt. The capsule has been inserted in the ETR in the 113SE core position and is operating satisfactorily very close to design temperatures.

Cooperative Irradiation Programs

ORNL Capsule HIBUP-2. Capsule HIBUP-2 is the second very-high-burnup irradiation experiment at ORNL. The objectives of this experiment are:

1. To optimize coating designs on fuel particles by an analytical model based on the strength of a shell surrounding a void volume that can be filled with fission gases.
2. To test such particles to about 50% burnup.
3. To compare both SiC and PyC coatings under these conditions.

The four General Atomic samples include two with SiC-barrier coatings and two with BISO coatings. Coating thicknesses were selected with the analytical model such that one BISO and one SiC would be on the threshold of failure after 50% burnup at 1300°C, whereas the other two would have an arbitrary 50% margin of safety. Detailed descriptions of the four General Atomic samples were given in the previous quarterly report. Several ORNL samples are also being tested in a different section of the capsule.

The capsule began irradiation in November in the ORR. The fuel has reached burnups of 25% to 30% FIMA to date at temperatures of about 1250°C. The fission-gas release rates have continued to increase and are currently very high (Xe¹³³ R/B $\approx$ 8 x 10⁻²). These release rates are monitored on a single purge stream from the capsule, so it is not possible to know which of the four General Atomic samples or five ORNL samples are contributing to the high release.

ORNL Capsule B9-34. Capsule B9-34 is an additional ORNL test of one of the General Atomic SiC-barrier coated samples from HIBUP-2 to 20% to 25% FIMA burnup at 1300°C. This sample (lot 3425-57E) has a 40- μ buffer and a

Table 9.6
DEPOSITION CONDITIONS AND DESCRIPTION OF
COATED PARTICLE SAMPLES^a IN CAPSULE P13J

Lot No.	Coating Type	Metallographic Classification	Temp (°C)	Deposition Rate (μ/min)	Deposition Gas	Density (g/cm ³)	Thickness (μ)	Design Irrad ^b Temp (°C)
3592-35E	SiC barrier	Buffer	1100	6.50	C ₂ H ₂	1.38	39	1300, 1000, 600
		Silicon carbide	1400	0.80	CH ₃ SiCl ₃	(c)	13	
		Isotropic	1300	5.56	C ₃ H ₈	(c)	10	
3592-37E	SiC barrier	Buffer	1100	6.50	C ₂ H ₂	1.38	39	
		Silicon carbide	1400	0.66	CH ₃ SiCl ₃	(c)	10	
		Isotropic	1300	5.00	C ₃ H ₈	(c)	74	
3592-45E	SiC barrier	Buffer	1100	6.50	C ₂ H ₂	1.38	39	
		Silicon carbide	1400	0.62	CH ₃ SiCl ₃	(c)	37	
		Isotropic	1300	4.00	C ₃ H ₈	(c)	8	
3592-47E	SiC barrier	Buffer	1100	6.50	C ₂ H ₂	1.38	39	
		Silicon carbide	1400	0.74	CH ₃ SiCl ₃	(c)	10	
		Isotropic	1300	5.17	C ₃ H ₈	2.03	31	
3592-53E	SiC barrier	Buffer	1100	6.50	C ₂ H ₂	1.38	39	
		Silicon carbide	1400	0.74	CH ₃ SiCl ₃	(c)	10	
		Isotropic	1300	4.56	C ₃ H ₈	2.03	52	
3592-69E	SiC barrier	Buffer	1000	10.20	C ₂ H ₂	1.21	51	1300
		Silicon carbide	1400	0.60	CH ₃ SiCl ₃	(c)	29	
		Isotropic	1300	4.00	C ₃ H ₈	(c)	10	
3592-71E	SiC barrier	Buffer	1000	10.20	C ₂ H ₂	1.21	51	
		Silicon carbide	1400	0.58	CH ₃ SiCl ₃	(c)	35	
		Isotropic	1300	3.85	C ₃ H ₈	(c)	10	
3592-73E	BISO(2200)	Buffer	1000	10.00	C ₂ H ₂	1.11	80	
		Isotropic	2200	5.60	CH ₄	2.05	38	
3583-67E	BISO(1300)	Buffer	1000	10.20	C ₂ H ₂	1.21	51	
		Isotropic	1300	3.82	C ₃ H ₈	2.04	65	

^a200-μ (Th,U)C₂ particles with a Th/U ratio of 2; all uranium is fully enriched. The Bacon anisotropy factor of the isotropic coating on lot 3592-73E is 1.03. All other Bacon anisotropy factors and the crystallite sizes (L_c) have not been determined.

^bDesign burnup is 20% FIMA in four ETR cycles; design fast-neutron dose is 3 to 6 x 10²¹ nvt.

^cNot determined.

28- μ SiC layer with no PyC overcoat. The objective of this experiment is to monitor the fission-gas release rates from a single sample of SiC-barrier coated particles taken to a high burnup.

The capsule began irradiation in the ORR in March. The fuel has reached 6% to 8% FIMA burnup to date at 1300°C. The fission-gas release rates were initially very low (Kr^{88} R/B $\approx 10^{-7}$) but have increased gradually with increasing burnup (current Kr^{88} R/B $\approx 10^{-5}$). The experiment is scheduled to continue irradiation until June.

ORNL Capsule 41-28. Capsule 41-28 was an ORNL high-burnup test of three General Atomic BISO- and Triplex-coated carbides along with three ORNL samples. The capsule completed irradiation after three cycles in the ETR and is awaiting postirradiation examination. Each coated particle sample was being tested in quadruplicate in different axial flux locations so that the estimated burnups for each range from 15% to 31% FIMA. The estimated maximum fuel temperature was 1200°C.

PLUTONIUM DIFFUSION STUDIES

Diffusion Studies of Plutonium in Pyrolytic Carbon

Preliminary diffusion studies are being conducted using U^{232} to ensure the reliability of the technique adopted for the study of plutonium diffusion in pyrocarbon. All equipment problems have been resolved. Currently, a solid-state battery-operated alpha counter is being used in conjunction with an NMC gas proportional counting chamber.

Diffusion anneals have been conducted at 1700° and 1900°C using U^{232} . The calculated diffusion coefficients for the room-temperature blank and 1900°C anneal are in good agreement with those previously obtained. Because of uncertainties in the sectioning procedure, the anneal at 1700°C will be repeated, along with an anneal at 1500°C. Plutonium diffusion anneals will be undertaken shortly.

Determination of Plutonium in Irradiated Capsule Components

Efforts directed toward the determination of plutonium in irradiated P-capsule components (thimbles and crucibles) have continued. A control run was made for the separation of plutonium using a double ion-exchange method. A 2- μg sample of plutonium was processed through the individual ion-exchange columns. The various eluted fractions are awaiting identification and quantitative recovery via neutron activation analysis. This operation requires an amendment to the technical specification for the TRIGA Mark I reactor to permit the irradiation of elements heavier than uranium.

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Task X
ADVANCED MATRIX-TYPE FUELS

FM-1 CAPSULE

Irradiation of the BeO fuel matrix pellets in capsule FM-1 was started on March 26. The purpose of this irradiation is to evaluate the retention capabilities of the fuel as a function of irradiation temperature, burnup, and fast neutron exposure.

The fuel was prepared by hot-pressing a dispersion of 215- μ -diameter UO₂ particles in a matrix of BeO. The formed pellets were approximately 1/2 in. in diameter and 1.2 in. long with a 1.8-in.-diameter hole. The fueled portion of the matrix was clad with unfueled BeO to a thickness of approximately 0.04 in. The pellets were hot-pressed in a graphite die at 1530°C which resulted in matrix densities that exceeded 99% of theoretical density for BeO.

The FM-1 capsule consists of four cells with five pellets in each cell. At the initial startup, the capsule performed excellently, as the four cells reached their designed temperatures of 800°, 1100°, 1400°, and 1600°C. These values were obtained through the use of a double-annulus control system in which helium and argon can be mixed to attain the proper temperature. The temperature spectrum was selected to encompass the potential usable range of the BeO fuel. It is conjectured that if the fuel fails at the lower temperatures it will be because of BeO matrix fracturing due to fast neutron effects when the exposure exceeds 10^{21} nvt. Because the capsule is designed to receive 3×10^{21} nvt this effect will be evaluated. At the upper end of the temperature scale it is possible that the fuel will not be retentive to fission products due to the low strength of the BeO matrix. This would permit voids in the matrix due to the generation of fission gases and helium. It is expected that this capsule will operate to a burnup of 50% for the enriched particles. Thus, examination of the microstructure should determine whether the matrix has indeed become porous at these elevated temperatures. It is also possible the fast neutron effect may cause the BeO grains to expand into any voids or pores that may be generated. This effect might be enhanced because of the plastic nature of the BeO at 1100°C and above.

The fuel integrity within each cell is being evaluated continuously by monitoring the gross activity of the heat transfer gas which passes over the samples. The gases used as a heat transfer medium in the inner or primary capsule are slowly passed over the samples. This sweeps the xenon and krypton gases from the fuel pellet out of the capsule and into the fission product traps outside of the reactor. The gases from each cell pass over radiation meters before they enter the traps. Thus, the relative gross activity from the fuel within each cell can be determined as a function of

time and exposure. No absolute value can be obtained as the gases cannot be sampled for individual isotopes. However, this system of monitoring the activity is already providing worthwhile information.

During approximately nine days of full-power operation of the ETR, the capsule reached an estimated burnup between 2% and 3% and a fast-flux exposure of approximately 2×10^{20} nvt. During this period there were 11 thermal cycles of the fuel, eight of which were caused by reactor scrams and three by manipulations of the conduction gas within the capsule. During each cycle the fuel temperatures dropped by 400° to 1500°C in several minutes. As a result, there were exceedingly steep thermal gradients across the fuel. Despite this severe treatment, the activity or release of fission products from the pellets only increased 2 to 4 times. Preirradiation evaluations of this type of fuel indicate that the initial R/B is 10^{-6} . If one uses this value, then the fuel is currently operating with a release fraction of approximately 2 to 4×10^{-6} . In general, the ratio of the release value after nine days of exposure to the initial release value is the same for each of the four irradiation temperatures.

Thermal tests of the FM-1 type of fuel are being continued in the laboratory at 800° and 1000°C. These control samples will be examined metallographically after they have been exposed for a period equivalent to that for the fuel in the irradiation capsule. This evaluation will determine the influence of temperature on the grain integrity as well as growth and stability between the fuel particles and the BeO matrix. Thirty days of thermal exposure have now been accumulated, and these tests will continue while the capsule is being irradiated.

F7 CAPSULE

BeO-matrix fuel has been irradiated at approximately 1400°C in capsule F7. This fuel was hot-pressed at 1900°C to a density greater than 99% theoretical. The 250- μ -diameter UO₂ fuel particles were dispersed in a pellet whose geometry was similar to that described for the FM-1 capsule.

The irradiation occurred in the MTR at approximately 1400°C to a burnup of $3.27 \pm 0.05\%$ and a fast-neutron exposure of $1.87 \pm 0.05 \times 10^{19}$ nvt ($E > 0.18$ Mev). At the beginning of this irradiation, the fuel was inadvertently overtemperated to 1800°C for 4 hr and to 1600°C for three weeks. The capsule was then relocated in the reactor to permit the fuel to operate at the design temperature of 1400°C for the remaining seven cycles. Postirradiation studies showed the fuel to be cracked as a result of thermal stress, and it was concluded that this was caused by the initial overtemperature. In spite of these cracks the R/B was approximately 1×10^{-4} for Kr⁸⁹, 8×10^{-4} for Xe¹³⁵, and 9×10^{-3} for Xe¹³³.

The capsule contained five fuel pellets, which are numbered sequentially from 1 through 5, and on postirradiation evaluation, each pellet was found to be cracked as a result of thermal stress. This effect is shown in Fig. 10.1, where a diametral section of pellet 5 shows a circumferential cracking. It appears that the crack started at the outer surface at approximately the

midplane of the pellet and progressed to the inside diameter, where it branched. During the overtemperated condition the pellet produced more than 450 watts of energy per centimeter. Subsequent laboratory thermal stress tests have shown that the pellets will crack at 130 w/cm. Other tests have indicated that such cracks will reduce the retention capability of the fuel by a factor of 10 to 100.

The effect of neutron irradiation and temperature was also evaluated with respect to microstructure by placing samples of unfueled BeO at either end of the fuel pellets. The microstructure of one of these BeO samples, or thermal barriers, is shown in Fig. 10.2. It was adjacent to the end fuel pellet, No. 5. In general, the microstructure shows the BeO to be very dense. Its grain size averaged $9\ \mu$ with a range of values from 3 to $20\ \mu$. This grain size and distribution is approximately the same as the control sample. The irradiation temperature for the sample was between 680° and 690°C . There were no cracks, pores, or pull-outs while the sample was being polished.

The coolest fuel pellet to be evaluated was No. 5. The outer portion, cladding, contained no fuel and had an appearance similar to the thermal barrier mentioned above. It is calculated that the irradiation temperature of this cladding was between 1290° and 1370°C . The average grain size was $8\ \mu$ with a range in size from 2 to $15\ \mu$. In contrast, the control sample grain size was between 3 and $5\ \mu$. Therefore, some grain growth had occurred. No cracks or pullout was observed in the cladding.

Inside the cladding, the fuel matrix of pellet 5 was irradiated between 1335° and 1400°C . An unetched section of the matrix is shown in Fig. 10.3. The average grain size was $13\ \mu$ with a range from 8 to $30\ \mu$. Thus, some grain growth did occur as the control sample grain size was $8\ \mu$. The grains appear to be dense and do not contain pores, but there did appear to be 1- to $3\text{-}\mu$ -diameter pores at the intersection of grain barriers. Detailed examination indicates that these may be either pores or places where extremely shallow, small grains have been pulled out during polishing. No cracks or spalls were observed, but some apparent porosity was observed when control samples were polished similarly to the irradiated samples, as may be seen in Fig. 10.4, which shows an unirradiated control specimen. It would thus appear that the irradiated samples may have a slightly higher susceptibility to pull-out or pore density, but that in general, irradiated and control samples were very similar in appearance.

The UO_2 particles appear to be unaffected by the irradiation. In both the control and irradiated sample there is a slight indication of porosity and scalloping of the edges.

The centermost pellet, No. 3, received the highest exposure and temperature. The cladding operated between 1400° and 1420°C and the resulting grain size was $8\ \mu$ with a range from 5 to $28\ \mu$. Two-micron pores or areas of pull-out were also observed at the grain boundaries. Thus, the claddings for both pellets have approximately the same average grain sizes, though the range of values is larger for pellet 5.

Fig. 10.1--Circumferential cracking of a diametral section of pellet 5 (6x)

Fig. 10.2--Microstructure of BeO sample adjacent to fuel pellet 5; average grain size $9\ \mu$, irradiation temperature between 680° and 690°C , etched (270x)

Fig. 10.3--Unetched section of fuel matrix of pellet 5 irradiated between 1335° and 1400°C; average grain size 13 μ (270x)

Fig. 10.4--Unirradiated control specimen 12 (270x)

The fuel matrix of pellet No. 3 operated between 1420° and 1490°C. The grains of BeO were dense, but their size was an average of 16 μ with a range between 10 and 30 μ . The structure of the matrix is shown in Fig. 10.5, where it can be seen that there was a fair degree of pull-out and apparent porosity at the intersection of grain boundaries. In some regions there was excessive pull-out, as can be seen in Fig. 10.6. Here is shown a thermal-stress crack that propagated through a fuel particle. This type of cracking, which was observed in numerous locations, appears to have occurred during the initial startup of the capsule where the fuel was at a temperature where the particle would still be elastic. At full capsule power, the particle would have been at 1400°C and plastic, which would not have permitted the fuel particle to fracture. This type of grain structure and pull-out was also observed for unirradiated control samples that were polished in the hot cell (see Fig. 10.7). It will be noted that the fuel particles are also scalloped, and it would thus seem that such an effect was produced as a result of hot pressing and not irradiation. A more typical structure of the matrix is shown in Fig. 10.8, where the fuel particles have been etched. It can be seen that there appears to be a rim or zone at the interface of the BeO and fuel particle.

In all cases the particles were still as dense-appearing as the control-sample fuel particles. It is believed that this high density was the result of the matrix preventing the particles from swelling during irradiation. A swelling phenomenon was observed in an earlier capsule, F5, where there was a void between the particle and matrix. This void permitted the particle to expand, thereby lowering the density of the fuel for a burnup of only 2%.

Although the burnup was low, it is felt that this study indicates the influence of the matrix in maintaining the integrity of the particle. Furthermore, there does not seem to be microcracking of the matrix either at the fuel particle-matrix interface or within the matrix. There was grain growth by a factor of about two for the matrix and cladding after about 300 hr at 1400°C and 500 hr at 1600°C. Also some porosity or pull-out was noted at the intersection of the grain boundaries. No porosity or cracking was observed within the grains.

Irradiation capsule FM-1 discussed above will provide additional data on the type of fuel that was contained in capsule F7.

Fig. 10.5--Structure of pellet 3 fuel matrix, with fair degree of pull-out and apparent porosity, etched (270x)

Fig. 10.6--Region of pellet 3 showing excessive pull-out and a thermal-stress crack through the fuel particle (270x)

Fig. 10.7--Unirradiated control specimen 12 polished in the hot cell showing cracking and pull-out (270x)

Fig. 10.8--Typical matrix structure, etched (270x)

Task XI

GRAPHITE

GRAPHITE IRRADIATIONS

Graphite irradiation studies on the HTGR Base Program are directed toward obtaining more definitive data on the irradiation behavior of conventional anisotropic and isotropic graphites or model carbons in the range 300° to 1500°C. The data on model carbons are being used to aid in the interpretation of the behavior of polycrystalline graphites. The graphite data are being related to current theories that attempt to explain the bulk dimensional and crystallite changes and property changes.

Fuel Element Graphite

A number of conventional needle-coke or Texas-coke graphites, upgraded in density and strength by impregnation, and near-isotropic graphites, prepared by a pressure-baking method, have been irradiated in capsule G10 in the temperature range 300° to 1500°C to fast-neutron exposures from 1.70 to 3.25×10^{21} n/cm² ($E > 0.18$ Mev). Changes in linear dimensions, coefficient of thermal expansion, and apparent crystallite height were measured.

When anisotropic graphites are irradiated in the range 880° to 1270°C to doses of 2 to 3×10^{21} n/cm² ($E > 0.18$ Mev), linear contraction starts to saturate in the direction normal to the layer planes. The isotropic graphites do not show this characteristic when irradiated at the same temperatures and doses. The apparent crystallite height, L_c , saturates at a common value of 150 to 250 Å for all graphites studied regardless of their original values or irradiation temperatures, except for samples irradiated at 615°C, which were in the range 300 to 400 Å.

Changes in bulk thermal expansion coefficients were small or nonexistent, except for graphites made with Texas-coke filler and samples irradiated at 300° to 400°C. All graphites studied exhibited a single linear relationship between thermal expansion and bulk dimensional changes in the range 320° to 615°C. In the range 840° to 1490°C, a separate linear relationship was observed for anisotropic or isotropic graphites. At high temperatures the linear relationship is shifted toward higher contraction for the isotropic graphites.

The presence of a nongraphitizing impregnant carbon in the pores of an anisotropic needle-coke graphite increased the volume contraction, reduced the anisotropy of contraction, and retarded "turnaround" in the perpendicular direction when irradiated in the range 880° to 1015°C to a dose of 3.25×10^{21} n/cm² ($E > 0.18$ Mev).

Thermal conductivity was measured in the perpendicular direction from 25° to 2000°C on impregnated graphites (CHN and H-315-A), which were irradiated at 300°, 950°, and 1230°C to doses in the range 1.1 to 3.1×10^{21} n/cm² ($E > 0.18$ Mev). Two methods of measuring thermal conductivity at 25° to 800°C and 800° to 2000°C were described in the previous quarterly report.

The thermal conductivities as a function of temperature for H-315-A graphite (a near-isotropic graphite made with Gilsonite coke and impregnated with coal-tar pitch) and CHN graphite (a needle-coke graphite that has been heavily impregnated with a nongraphitizing carbon) irradiated at 300°, 950°, and 1230°C are shown in Figs. 11.1 and 11.2, respectively. Figure 11.2 also includes data for 780-S graphite (a Texas-coke graphite impregnated with coal-tar pitch), which had been irradiated at ORNL in the range 1300° to 1500°C.

The thermal conductivities for all graphites measured at 25°C increase with increasing irradiation temperature. This indicates that a higher proportion of neutron-induced lattice defects remain in the samples irradiated at 300°C than in those irradiated at higher temperatures. However, a considerable amount of damage still remains after irradiation at 1200° to 1300°C. These defects are partially annealed in the range 1100° to 1400°C during measurement. After heating to 2000°C, the conductivity returns to the unirradiated values when measured at 800°C. However, remeasurement of the annealed samples at room temperature still shows a conductivity lower than the pre-irradiation value. The near-isotropic graphite H-315-A contained enough defects after annealing at 2000°C to show a 15% fractional change in thermal conductivity at room temperature. In contrast, the CHN graphite showed only a 3% fractional change at 25°C after annealing at 1500°C.

Pyrolytic Carbons

A model has been developed to describe the dimensional behavior of pyrolytic carbon during irradiation by fast neutrons. The model uses crystallite-averaging methods to describe the anisotropic distortion of the bulk in terms of its preferred orientation and the change in shape of the individual crystallites. The densification, a simple first-order rate process, is assumed to be independent of the shape change and is superimposed isotropically on it. The predictions of the model agree with experimental observations.

Strain rates calculated from the model are used to estimate the suitability of various carbon structures for use as coatings on an unyielding support. For example, using a creep constant of 1×10^{-27} (nvt-psi)⁻¹ at 1200°C, the model predicts that a carbon with a Bacon anisotropy factor (BAF) of 1.2 and apparent crystallite heights, L_c , in the range 120 to 155 Å would not fail as a result of neutron-induced dimensional changes if its original density were greater than 2.0 g/cm³. Similarly, carbons with the same crystallite size but with a BAF of 1.0 would not fail under the same conditions if their original densities were greater than 1.7 g/cm³.

Results from the carbons irradiated show that in the temperature range 500° to 1000°C the densification constant, k_d , for carbons with L_c parameters

Fig. 11.1--Thermal conductivities as functions of temperature for H-315-A and CHN graphites

Fig. 11.2--Thermal conductivities as functions of temperature for H-315-A, CHN, and 780-S graphites

in the range 25 to 45 Å is independent of temperature and equal to $3.3 \times 10^{-22} \text{ nvt}^{-1}$. When L_c is relatively large (155 to 175 Å), k_d is also independent of temperature and equal to $1.1 \times 10^{-22} \text{ nvt}^{-1}$. The value of k_d increases with temperature for carbons with intermediate values of L_c .

General Atomic-BNWL Joint Graphite Irradiation Program

A graphite irradiation capsule experiment designated GA-H-13-13 (formerly GE-H-13-12) in which the space is shared between General Atomic and BNWL is under way in the ETR.

The objectives and a description of the experiments and specimens have been presented in previous quarterly reports. Graphites previously irradiated in capsule G10, an orientation series of hot-worked graphite and of massive pyrolytic carbon, and model carbons that represent a wide range of structures were included. The capsule was damaged while cold at the conclusion of the second cycle and was withdrawn. It had completed two cycles in the ETR and had operated in the approximate range of 800° to 1300°C to doses of 0.5 to $2 \times 10^{21} \text{ n/cm}^2$ ($E > 0.18 \text{ Mev}$). Contamination was introduced into the capsule from an unknown source, and a number of the specimens were contaminated. However, the bulk of the specimens were not contaminated and have been shipped to General Atomic for examination.

After examination, the irradiated samples will be shipped to BNWL, along with replacement samples for those found contaminated, for reencapsulation in GE-H-13-15, which is scheduled to begin irradiation in the ETR in early June. Capsule GE-H-13-15 is scheduled for four cycles and approximately $4 \times 10^{21} \text{ n/cm}^2$, which will extend the doses on the original samples from G10 to about 7 to $9 \times 10^{21} \text{ n/cm}^2$ in the range 800° to 1300°C.

Dounreay Cooperative Graphite Experiment

A single graphite irradiation vehicle or rig sponsored by the United States is being planned for irradiation in the Dounreay Fast Reactor. The objective of the experiment is to extend experimental data on graphites that are of interest to advanced HTGR's to doses of about $1 \times 10^{22} \text{ n/cm}^2$ at 600° to 700°C and 1000° to 1100°C. Details of the experiment and materials were discussed in previous quarterly reports. Primarily, the rig will contain graphite or pyrocarbon samples, though a few cans will contain coated fuel particles. The rig will be shared among General Atomic, BNWL, and ORNL and will be irradiated in DFR, core position Pitch 10, beginning about June 1, 1967, and will continue for 55 days. ORNL is acting as liaison with the Dounreay site.

All specimens have been prepared, measured, and loaded into niobium cans that were supplied by the Dounreay reactor site. The cans were shipped to ORNL, where the final sealing operation (welding the lids) will be done. ORNL will ship the sealed cans to the Dounreay reactor site.

A second Dounreay experiment that will contain one cell for graphite and one for PyC is scheduled for insertion in August or September. This experiment will operate at 1000°C for approximately 110 days, or to about $18 \times 10^{21} \text{ n/cm}^2$. Preparations for this experiment are proceeding.

RADIATION DAMAGE MECHANISMS IN GRAPHITE

Theoretical Analysis

Work on the mathematical study of the growth of defect concentrations in irradiated graphite during the quarter was directed toward the problem of continuing the numerical integration to longer times. Two different methods of increasing the time step without increasing the numerical error by large amounts are being considered. Both of these methods make use of the fact that the integrations performed to date indicate that the time rate of change of n_1 (the concentration of interstitials) and n_2 (di-interstitials) becomes very small with increasing time. If the time derivatives of n_1 and n_2 are set equal to zero, an algebraic (not a differential) equation for n_1 and n_2 is obtained. The other method is to compute the derivatives numerically by taking an average of the derivatives computed at the previous time point and the next time point. This takes advantage of the fact that any error introduced at one point tends to generate an error of the opposite sign at the next point. This method is in essence an iterative one since the values of the functions needed to find the derivative at the next point are not known until the derivatives themselves are known.

Carbon Ion Bombardment

The experimental work during the quarter has concentrated on carrying out a series of thirty-seven bombardments of single crystal specimens.

The apparatus used for the bombardments was described in the previous quarterly report. The carbon ions used were 60 kev C^+ ions, which have a range of approximately 1400 Å. The specimens were contained in a holder that could be heated to 1200°C. The temperature of a specimen heated to less than 850°C was determined by a thermocouple whose hot junction was spot welded to the heater strip. Above 800°C an optical pyrometer was used to measure temperatures through a viewing port in the specimen holder. In the overlap region the thermocouple temperature agreed with the pyrometer reading to within 5°C. It is estimated that throughout the range 23° to 1200°C the specimen temperature is known to within 20°C.

In carrying out a bombardment, a flake mounted in a double Ti grid, as described in the previous quarterly report (GA-7801), was placed in the specimen holder, and the holder was mounted in the target assembly of the atomic beam apparatus. The chamber was then evacuated by means of an Hg diffusion pump to 1 to 3×10^{-6} torr. Then the current to be delivered to the specimen was adjusted, with a beam flag shielding it from the carbon ions. A bombardment was started by shifting this flag to allow the ion beam to impinge on the carbon flake in its holder; it was terminated after the prescribed period by resetting the beam flag. The ion current was rechecked periodically throughout the bombardment. After a bombardment was terminated, the heater current was turned off. The specimen was then left in the vacuum chamber for about one-half hour to cool and finally was removed from the apparatus.

The identification number of each specimen and the conditions of the bombardment are shown in Table 11.1. In the table the ion exposure in $\mu\text{amp-min/cm}^2$ is correlated with neutron exposure in n/cm^2 and the ion current density in $\mu\text{amp/cm}^2$ with neutron fluxes. From the paper of Thompson and Wright (Ref. 1), it is possible to find the displacement rate for lattice atoms as a function of carbon ion current density and also as a function of neutron flux in a given neutron energy spectrum (PLUTO). The results are

$$G_0 t = 2.64 \times 10^{-3} g \left. \frac{dE}{dx} \right|_{\text{disp.}} \quad (\text{for ions}) = 1.31 \times 10^{-21} \gamma \quad (\text{for reactors}) ,$$

where $G_0 t$ is given in displacements per lattice site, g is the ion exposure in $\mu\text{amp-min/cm}^2$, and γ is the neutron exposure in n/cm^2 . For this case dE/dx was taken to be $10 \text{ ev/\AA}$.

The bombardment phase of this project has been completed, and evaluation of the results has just begun. The specimens are to be examined in the electron microscope in an effort to identify bombardment-induced interstitial and vacancy clusters. These clusters can be described as dislocation loops, largely with Burger's vectors $\pm c/2$ (001). To see them in dark field situations, it is necessary to use a spot (hkl) with $l \neq 0$. Figure 11.3 shows two pictures of an area of a flake bombarded for $10 \mu\text{amp-min}$ at 1100°C at an ion current of $1 \mu\text{amp/cm}^2$, equivalent to a neutron exposure of $2 \times 10^{20} \text{ n/cm}^2$ at 1100°C at a neutron flux of $3.4 \times 10^{17} \text{ n/cm}^2\text{-sec}$. It was established by selected area diffraction that the dark field picture shows the (112) diffracted beam. This is the case even though the specimen stage was not tilted. Apparently the section of flake being examined was sufficiently tilted to bring in this spot. The pictures show small features similar to those seen by Thrower (Ref. 2) in neutron-irradiated specimens, but no extensive interpretation of the electron microscope pictures has yet been undertaken.

MECHANICAL STRESSES IN GRAPHITE

Test Apparatus

Work on the isolator platform described in previous reports was concluded. The installation was completed by cross bracing, which resulted in the desired increase of the effective stiffness coefficient. The resonant frequency of the base structure when excited by microseismic, or man-made, noise has now been increased to greater than 50 cps. At this frequency sufficient decoupling from the specimen resonance is achieved.

The isolation from subcycle noise has not been pursued. For the class of measurements started during this report period and planned for the near future, noise interference in this frequency domain has negligible effect. This will not be the case when very-high-sensitivity, low-amplitude stress-strain loops will be determined. A limited effort on this problem is therefore being continued.

Table 11.1
CARBON ION BOMBARDMENT CONDITIONS

Ion Flux ($\mu\text{amp}/\text{cm}^2$)	Equivalent Neutron Flux ($\text{n}/\text{cm}^2\text{-sec}$)	Ion Dose ($\mu\text{amp-min}/\text{cm}^2$)	Equivalent Neutron Dose (n/cm^2)	Specimen No.						
				at 23°C	at 150°C	at 300°C	at 600°C	at 800°C	at 900°C	at 1100°C
0.1	3.4×10^{16}	1	2×10^{19}	---	---	C40	---	---	---	---
		10	2×10^{20}	C11	C19	C20	C21	C15	C14	C25
		100	2×10^{21}	---	---	---	---	C16	---	---
0.5	1.7×10^{17}	1	2×10^{19}	---	---	C38	---	---	---	---
		10	2×10^{20}	C9	C17	C18	C23	C1	C6	C7
		100	2×10^{21}	C10	C39	C42	C22	C13	C12	C8
5.0	1.7×10^{18}	1	2×10^{14}	---	---	C33	---	---	---	---
		10	2×10^{20}	---	C30	C31	C32	C3	C35	C37
		100	2×10^{21}	---	C27	C26	C28	C29	C36	C34

Fig. 11.3--Two views of the same area of a graphite flake bombarded for $10 \mu\text{amp-min/cm}^2$ at 1100°C . The upper photo is a bright field shot and the lower is a (112) dark field picture (50,000X)

The apparatus in which the micromechanical measurements are made is essentially a very advanced and versatile inverted torsion pendulum (Ref. 3). In essence, a specimen is twisted from its relaxed position. From the necessary torque to do this and the resulting angular deflection, shear stress and shear strain can be determined. From the measurement of the input power dissipated during stress-strain cycles, which is related to the internal friction, internal loss mechanisms can be inferred. If the specimen is set into oscillation, one obtains information from the resulting frequency relating to the internal restoring forces that determine the modulus. The apparatus therefore has provisions to twist a specimen and measure the input power, strain angle, and period of oscillation with high precision. An important feature of the equipment is that the specimen temperature can be changed over a wide range for this type of measurement; presently, operation is over a 300°K span.

Measurement Methods

Internal friction and modulus (Ref. 4) defect measurements have been used extensively in metals research, alloy development, and the field of polymers (Ref. 5). The particular approach being used is in the so-called low-frequency regime as contrasted with flexural, longitudinal, or echo techniques, which go from the audio frequencies through ultrasonics into the megacycles range. One of the most important advantages of operating in the subsonic region is that the measurement frequency can be varied without a change or disturbance of the specimen. This is of prime importance for materials that are structure sensitive.

The internal friction can be determined in a number of ways. The standard method consists of setting a specimen into oscillation and then observing the decrease of oscillating amplitude with time. From the amplitude decay, called free decay, the logarithmic decrement can be obtained, and this is proportional to the internal friction. The shear modulus can be derived from a simultaneous measurement of the period of oscillation.

More important than the determination of these quantities by themselves is the ability to measure any small changes that occur. The more obvious changes are brought about by externally applied mechanical stresses, temperature cycling, exposure to different ambient gases or liquids, exposure to irradiation, etc.

Instrumentation was therefore developed to explore more advanced modes of operation, which are schematically illustrated in Fig. 11.4. The envelope of successive strain amplitude maxima is plotted against time. The free decay mode is used for calibration and reference whenever possible. However, with a real specimen under various operating conditions, the mechanical response can range widely. For very low internal friction, it is preferable to employ the "brake" mode, since low internal friction leads to long decay times. The deliberate introduction of an additional component in series with the specimen's internal friction makes for more efficient, improved measurements. This is accomplished by electrically simulating a constant, known amount of "external friction" acting like a brake. At the other end of the scale is the case of high internal friction (very high damping--rapid decay)

Fig. 11.4--Envelope of oscillating amplitude versus time for various decay modes as applied to a specimen

often associated also with high background internal friction even at low strain amplitudes. In this instance, a constant, known amount of energy that will ultimately sustain the oscillations at a certain level is supplied during the decay. This inhibits the decay and hence improves the measurement resolution by increasing the number of oscillations within a given amplitude range. This technique is called "restrained" mode and was specifically developed for this experimental program.

In the measurement approaches described above the specimen is first brought to a preselected oscillating strain level and the measurements are then carried out as the amplitude decays. If the specimen shows a very large range of internal friction for the amplitudes of interest, the use of the "buildup" mode shown in Fig. 11.5 is preferable. In this technique the maximum internal friction is preselected, and the resulting amplitude increase with time is observed. This allows measurement resolution for a wide dynamic range, whereas in a corresponding decay method either the high or low regime would be emphasized.

It is essential to know if the specimen's over-all mechanical state changes. This can be detected by monitoring the time dependence of the modulus and internal friction. Often this is combined with amplitude dependence measurements by using the "driven" mode, one form of which is schematically indicated in Fig. 11.6.

All techniques discussed above can be used either isothermally, with the specimen at one or more constant temperatures, or with the temperature changing in a definite way with time, e.g., linearly, as shown in Fig. 11.7. If the time to measure internal friction and modulus is short, and the magnitude of these quantities does not change much in the corresponding temperature interval, then this manner of determining the internal friction spectrum is very powerful. This can be seen in Fig. 11.7, in which continuously repeated amplitude decay curves are indicated on the same time scale. Between the maximum and minimum oscillating strain amplitude, a large number of numerical values from which to obtain the desired information can be selected. Thus from a single sweep through a large temperature range, the amplitude dependence for any given temperature can be extracted by cross-plot analysis, as illustrated in Fig. 11.8.

Measurements and Results

A pyrolytic graphite specimen, PG-JB-11, was mounted in a small pendulum that was placed, using a special adaptor, in the regular instrument. The specimen gauge section was 200 mils long, 63 mils wide, and 22 mils thick with layer planes along the largest dimension. The starting material had a slight bow that persisted through cleaving to this thickness. The gauge section was machined as described in the previous quarterly report. This specimen was used for preliminary measurements of internal friction and period at low oscillating strain amplitudes at room temperature to check out the instrumentation. The specimen was quite lossy and showed considerable internal friction even at low strain amplitudes ($Q^{-1} \approx 1300 \times 10^{-5}$).

Fig. 11.5--Buildup mode of operation

Fig. 11.6--Driven mode of operation

Fig. 11.7--Amplitude dependence taken during specimen heating

Fig. 11.8--Cross-plot analysis of temperature dependence of intense friction

Later another specimen, PGA-JB-11, was installed in the torsion pendulum at the end of the drive tube for a series of temperature-dependence measurements. This dumbbell-shaped specimen had a gauge section 350 mils long, 84 mils wide, and 63 mils thick with layer planes along the torque axis.

Room-temperature measurements showed that even though the internal friction range could be handled by the then available measurement approach and data reduction, it would be essential to develop a new technique. The "restrained" mode described above was selected, and the technique was checked by using the free decay mode as calibration.

A temperature-dependence run starting at about 50°K was attempted. The specimen response showed a large amount of noise and erratic behavior and the run was aborted. The problem appeared to be associated with the grip and the intrinsically noisy resonant frequency of about 28 cps. The specimen was remounted, and the inertia of the resonant system was changed to produce a resonant frequency of about 16 cps. No grip problems developed in subsequent experiments.

An isothermal amplitude-dependence run was made at room temperature to a maximum oscillating angular strain of 50×10^{-5} radian for specimen PGA-JB-11 to determine whether this would result in any observable state change. The internal friction is plotted versus the logarithm of the strain angle in Fig. 11.9, which also includes data from tests on PG-JB-11. It appears that the amplitude-dependence experiment has not materially changed the mechanical state of the specimen. This is deduced from (1) the lack of time dependence, (2) the constancy of the low-amplitude modulus, and (3) the reproducibility of the internal friction spectrum before and after room-temperature oscillations.

Various temperature-dependence runs were made mostly from liquid-nitrogen temperature to room temperature. Both internal friction and period were determined by the method shown in Fig. 11.7. The internal friction as a function of temperature is shown in Fig. 11.10. Angular strain amplitude was used as parameter, but for clarity only the values indicated in the figure have been plotted, and experimental points are shown for only one amplitude. A prominent increase in internal friction occurs at approximately 200°K, which is also reflected in the corresponding period versus temperature plots (not shown here). Below this temperature there appears to be a broad peak. Further, one notes that near room temperature, for the high amplitudes, a downtrend in internal friction develops that may be indicative of another set of processes.

The results indicate, first of all, that reproducible internal friction and period measurements can be made in pyrolytic graphite by the methods outlined earlier. There exists, in the temperature region from about 50° to 300°K, structure in the internal friction spectrum. The micromechanical state of these specimens is sufficiently fixed that the oscillating measurement strain amplitudes used here do not alter the response of the samples.

Fig. 11.9--Amplitude dependence of internal friction

Fig. 11.10--Internal friction spectrum for specimen PGA-JB-11

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