MASTER

GULF GENERAL ATOMIC

AEC RESEARCH AND
DEVELOPMENT REPORT

GA-10399

HTGR BASE PROGRAM
QUARTERLY PROGRESS REPORT
FOR THE PERIOD ENDING
NOVEMBER 30, 1970

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

December 31, 1970

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GULF GENERAL ATOMIC COMPANY, P.O. BOX 608, SAN DIEGO, CALIFORNIA 92112

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Gulf General Atomic Project 317

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GA-9815-September, 1969, through November, 1969
GA-9944-December, 1969, through February, 1970
GA-10088-March, 1970, through May, 1970
GA-10288-June, 1970, through August, 1970

INTRODUCTION

This report covers the work performed by Gulf General Atomic 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. This program is based on the concept of the High-temperature Gas-cooled Reactor (HTGR) system developed by Gulf 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 a 330-MW(e) prototype in the early 1970's. The plant would demonstrate such new design features as:

1. High conversion rates 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.

Physics, metallurgical, chemical, electronic, and analytical design and development engineering programs support the primary reference design objective.

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TASK IV
FISSION-PRODUCT MECHANISMS

STRONTIUM DEPOSITION LOOP

A simple strontium deposition loop is being assembled to study the nature, distribution, and adherence of strontium deposits formed under typical reactor conditions. The primary objective of this work is to obtain plateout distribution data for use in testing a mass-transport code developed at Gulf General Atomic. This code is used to predict the fission-product plateout distribution in a reactor circuit.

The deposition loop, which is shown in Fig. 4.1, is designed to circulate 350-psia helium at reactor Reynolds numbers with a typical reactor temperature distribution of 600° to 1400°F. This temperature distribution will be maintained by inserting the entire loop into a 5-ft, high-pressure, high-temperature autoclave. The thick-walled piping normally associated with high-pressure loops is being avoided by having the autoclave operate at the same pressures as the deposition loop. Thus, the loop can be fabricated with a simple blower and with thin-walled tubing that can be readily bent, cut, or otherwise adjusted to fit the needs of the experimental program. A pneumatic control system for the autoclave has been designed that incorporates fail-safe controls on the primary heat and pressure systems as well as the critical support systems.

A porous graphite annular cylinder containing a Sr-85-impregnated matrix material will serve as the source of strontium. The cylinder will be heated by conduction through the wall of the loop. Since there is essentially no pressure difference across the loop wall, temperatures of approximately 1000°C can be attained without using special materials; consequently, it should be somewhat easier to obtain significant vapor pressures in this tubing loop than in normal thick-walled loops. In addition, the experimental flexibility of a simple tubing loop will allow modifications to be made relatively easily.

Fig. 4.1. Diagram of strontium deposition loop

A prototype centrifugal blower has been designed and built to establish the performance characteristics of such a unit within the confines of the autoclave chamber. A GGA-TFM flowmeter was equipped with a calibrated bypass tube to permit measurement of the high volume flows required for the loop, and a water manometer was set up to measure head pressure and pressure drop in the test system. A series of tests was run in which the effects of blower speed, intake porting, exhaust porting, and loop configurations were recorded. These tests led to a final blower housing design which, at a standard 3450-rpm blower speed, should give flow conditions well into the turbulent region. The as-built model and associated loop will be further tested and calibrated for pressure drop to provide performance characteristics that can be monitored from outside the autoclave during actual loop operations.

The undisturbed Sr-85 plateout profile at the end of an experiment will be measured by gamma-scanning the tubing after the loop is removed from the autoclave. At the conclusion of a run, the loop tubing can be sectioned with a parting tool so that the deposits can be investigated and simple blowoff experiments performed.

VAPORIZATION OF CESIUM FROM GRAPHITE

In preparation for a mass spectrometric study of the vaporization behavior of strontium and cesium cosorbed on H-327 graphite, mass spectrometric measurements of adsorption isotherms of cesium on H-327 graphite are being carried out. The purpose of these measurements is to determine the correspondence between cesium vapor pressure data measured using the mass spectrometer and data measured using the pseudoisopiestic method. Extensive cesium vapor pressure measurements have been previously performed using the pseudoisopiestic method (see earlier quarterly report GA-8662).

The sample preparation for the mass spectrometric studies involved adding the graphite sample to an aqueous CsNO_3 solution, drying, and heating at 1150°C for 3 hr in a sealed tantalum crucible. The sample was then placed in the mass spectrometer Knudsen cell.

Data obtained by the two methods are somewhat different. The pseudo-isopiestic measurements, using 2- to 3-mm grain-size graphite, yielded data for each graphite studied (including H-327) which nicely fit a Freundlich isotherm. For TS-688 graphite an abrupt transition to Langmuir behavior was observed for small loadings (see earlier quarterly report GA-8356). By contrast, the mass spectrometric isotherms are not linear on a log P versus log C plot. The isotherms are curved and give the appearance of being in a broad transition region between Freundlich and Langmuir behavior (see Fig. 4.2). The reproducibility between two mass spectrometric experiments using 37- to 74- μ m grain samples was quite good.

The cesium partial pressures measured by the mass spectrometric method are generally lower than those measured by the pseudoisopiestic method at equal loadings, but the two sets of data tend to converge at lower loadings.

A possible reason for the discrepancy between the two methods was thought to be the sample grain size. A mass spectrometric experiment was therefore run on a sample of 2- to 3-mm grain H-327 graphite that had earlier been prepared for some pseudoisopiestic experiments but had not been used. The isotherms obtained duplicated those that had been obtained for the 37- to 74- μ m grain samples, indicating the sorptive capacities of the two sample size preparations to be the same. While this result tends to indicate that the experiments were not subject to kinetically limiting processes, it is not in agreement with earlier pseudoisopiestic measurements in which the sorptive capacity of a 37- to 74- μ m grain TS-688 graphite sample was measured to be 3.3 times as great as for a 2- to 3-mm grain sample of the same graphite when expressed on a mg Cs/g graphite basis (see earlier quarterly reports GA-8356 and GA-8530).

Simple annealing (isopiestic) experiments were performed in which samples of both grain size fractions were annealed together, but physically separated, in a sealed tantalum container. The samples were prepared in the same way as for the mass spectrometric experiments. In one case the cesium was initially loaded onto 2- to 3-mm grains, the vapor-phase transport of Cs occurred during a 1150°C 8-hr anneal, such that the powder-to-bulk

Fig. 4.2. Vaporization of Cs from H-327 graphite (37 to 74 μm grain)

Cs ratio was 1.7. To check for equilibration, the experiment was repeated, this time pre-loading the 37- to 74- μm grains instead. For the second experiment, the ratio of the final concentration of Cs on the two samples was 2.6. The differences in values apparently indicate that equilibrium was not achieved. The results are adequate to indicate that the sorptive capacity (mg Cs/g graphite) of the 37- to 74- μm grains is about two times that of the 2- to 3-mm grains.

FIPER CODE DEVELOPMENT

In the application of the two-dimensional FIPER code to an experimental situation determined by the micro (or structural) geometry of nuclear graphite, the diffusion problem is formulated in the vicinity of a corner. This is because nuclear graphite is permeated by pores that are important for the release of metallic (as well as gaseous) fission products. The pore geometry is indicated in the following sketch.

The connection between an outer surface (which could represent the surface of a fuel hole) and the inner surface of a pore forms a corner. The physical parameters (diffusion coefficients, distribution ratios, etc.) for outer and inner surfaces are the same. The objective is to describe the physical and chemical parameters near a corner by reasonably smooth functions. A sketch of such a corner, with notations of the relevant coordinates, is given below.

The corner is at $z = z^*$, $r = r^*$. A function $f(r, z)$ is sought with the following properties:

$$\begin{aligned} f(r, z) &= 0, & \text{for } r > r^*, z > z^* \\ f(r, z) &= 1, & \text{for } r < r^*, z \text{ arbitrary} \\ & & z < z^*, r \text{ arbitrary} \end{aligned}$$

The function has the form of a Fermi-Dirac distribution, and is given by

$$f(r, z) = \frac{1}{\exp\left(\frac{rz - r^*z^*}{r_0 z_0}\right) + 1}, \quad r \leq r^*, z \leq z^*; \quad (\text{region 1})$$

$$f(r, z) = \frac{\exp\left(\frac{z^*(r - r^*)}{r_0 z_0}\right) + \exp\left(\frac{r^*(z - z^*)}{r_0 z_0}\right)}{\left[\exp\left(\frac{z^*(r - r^*)}{r_0 z_0}\right) + 1\right] \left[\exp\left(\frac{r^*(z - z^*)}{r_0 z_0}\right) + 1\right]}, \quad r \geq r^*, z \geq z^*; \quad (\text{region 2})$$

$$f(r, z) = \frac{1}{\exp\left(\frac{z^*(r - r^*)}{r_0 z_0}\right) + 1}, \quad z \leq z^*, r \geq r^*; \quad (\text{region 3})$$

$$f(r,z) = \frac{1}{\exp\left(\frac{r^*(z - z^*)}{r_0 z_0}\right) + 1} , \quad z \leq z^* ; \quad r \geq r^* .$$

(region 4)

Here, r_0 and z_0 are parameters that determine the "smoothness" of the transition from region 1 to region 2, etc.

An example for an application of this function is given by a diffusion coefficient that has a corner at r^*, z^* :

$$\begin{aligned} D(r,z) &= D_1 \text{ in region 2,} \\ &= D_2 \text{ in regions 1, 3, and 4.} \end{aligned}$$

The analytic expression for $D(r,z)$ used by the computer is

$$D(r,z) = D_1 f(r,z) + D_2 [1-f(r,z)] .$$

A physical application of this formula to a real case occurs in the diffusion problem described in the previous quarterly report (GA-10288), if we assume that the inside path 1 plays the role of an outer surface.

PROGRAM PLAN FOR WORK TO ESTABLISH FISSION-PRODUCT BEHAVIOR

Updating of the National Program Plan for Work to Establish Fission-product Behavior in HTGR Systems is nearly complete. The purpose of this plan is to present (1) an overall discussion of the technical aspects of fission-product behavior in HTGR systems and (2) an overall program for further development work to characterize fission-product behavior in HTGR systems. The first edition of the plan was completed in June 1969.

During the updating of the plan, it was expanded considerably to include a more thorough and comprehensive discussion of known information and sources of information related to all aspects of fission-product behavior in HTGR systems. In addition to discussing the technical aspects of fission-product

behavior, the plan also describes needed information on fission-product behavior and outlines a program of work for obtaining the needed information.

DIFFUSION OF STRONTIUM IN GRAPHITE

Strontium permeation experiments 4370-143 and 4370-144 with initial source concentrations of 20.3 mg Sr/g matrix (source) material have now accumulated 2860 hr at 850°C and 3430 hr at 1000°C, respectively. Preliminary data for these experiments are given in earlier quarterly report GA-9944, and a description of the apparatus is given in earlier quarterly report GA-9660. Fractional releases at the present time are 2.3×10^{-6} and 8.5×10^{-6} , and release rates are 1×10^{-8} and 5×10^{-8} mg Sr/hr for the respective experiments.

The primary objective of these experiments is to measure the temperature dependence of permeation coefficients. Another objective is to continue the experiments for a sufficient length of time to be assured that steady state is reached. This will make it possible to determine whether diffusion in the bulk graphite becomes important after some long time period. Earlier results suggested that diffusion in paths of high diffusivity (pores) is predominant at all times over diffusion in the bulk graphite (see earlier quarterly report GA-9815).

It should be noted that in earlier quarterly reports (see for example GA-9815) the coefficient measured in permeation experiments was designated as "an effective diffusion coefficient." However, the designation "permeation coefficient" more clearly distinguishes this coefficient from the fundamental (Fick's law) diffusion coefficient, which applies in both steady-state and transient situations. The permeation coefficient is measured under steady-state conditions and applies, without qualification, only to a steady-state situation.

EFFECT OF STEAM OXIDATION ON THE STRENGTH OF GRAPHITE

Studies of the effect of steam oxidation on the strength of H-327 graphite are being conducted. Tensile specimens are presently being exposed to 3% steam in helium at 900°C. The specimens are 1/2-in. diameter rods cut from graphite logs both perpendicular and parallel to the direction of extrusion. The perpendicular-cut rods are 1-1/2-in. long and the parallel-cut rods are 3 in. long.

Twelve specimens are being exposed at one time, two parallel-cut specimens and two perpendicular-cut specimens from each of three production logs. The samples are held in a circular array around a central perforated gas tube. They are supported between two graphite disks by depressions in the disks. The perpendicular-cut samples are in one array of six and the parallel-cut samples in a contiguous but separate array of six. A small amount of gas is also allowed to pass over the perimeter of the sample holders.

Following exposure, the samples will be subjected to standard uniaxial tensile strength tests. Samples subjected to various degrees of burnoff will be tested.

TASK V

RECYCLE FUEL STUDIES

RECYCLE TEST ELEMENTS

Test Element Fabrication

Fuel particles for the last recycle test element have been ordered. The schedule for loading the element in Peach Bottom has been changed from 150 to 300 EFPD, since no reactor outage is scheduled for 150 EFPD.

Test Element Irradiations

Six recycle test elements are currently in operation in the Peach Bottom reactor. The reactor was raised to power on July 14, 1970. Full power (95%) was reached at approximately 2200 hr, and as of October 30, 1970 the recycle test elements had accumulated approximately 94 EFPD. Three reactor outages have been experienced to date since start up. Based on data obtained from instrumented test elements, it is estimated that all of the recycle test elements are operating in the range of 2000° to 2200°F peak fuel temperature.

HTGR FUEL RECYCLE PLANT STUDY

Cost estimates of installed equipment were completed for the head-end steps to the reprocessing section of the combined reprocessing-refabrication facility design and economic study. The equipment sizing and costs for the crush, burn, leach head-end are developed for facility designs of 15,000-, 30,000-, and 45,000-MW(e) HTGR installed economies. The head-end steps include the graphite dispal flowsheet operations and the feed preparation operations shown on the uranium and thorium recovery flowsheet (the process flowsheets were presented in the previous quarterly report, GA-10288).

New material balance sheets are being prepared to incorporate revisions in the fuel element definition with respect to the equilibrium recycle fuel element composition. Table 5.1 shows the averaged fuel element composition and replaces the data reported in earlier quarterly report GA-10088, p. 33. The basis for the revised composition remains unchanged (i.e., fuel loading with carbon-to-thorium ratio of 270, average burnup of about 98,000 MWd/ton, and silicon carbide coating on the U-235 fissile fuel particles).

Material balance recalculations are in progress where necessitated by the small change in the average recycle fuel element composition. Equipment sizing and cost estimates are in progress for the remaining steps of the reprocessing section and for the refabrication section.

NATIONAL HTGR RECYCLE DEVELOPMENT PLAN

Discussions with ORNL representatives were held to review ORNL requirements for the recycle development program and to review Gulf General Atomic head-end experimental results. The fabrication and postirradiation examination of the Peach Bottom reactor Recycle Test Elements were reviewed. As a consequence of the discussions,

1. The average 1000-MW(e) HTGR fuel element composition and equilibrium recycle core data were reviewed, revised, and sent to ORNL.
2. Drawings of the clean fuel loading tool used in the 330-MW(e) prototype were reviewed and given to ORNL personnel to aid in their development of similar spent fuel handling equipment.

HEAD-END REPROCESSING

Crushing and Tumbling

Pitch-bonded rods now being made have proven stronger and more resistant to attrition than Rezolin-bonded rods, and because of this, three alternative head-end schemes are being considered:

Table 5.1
AVERAGED FUEL ELEMENT COMPOSITION
(C/Th = 270, T = 4 yr, BU = 98,000 MWD/T)

	Block Type A (Unirradiated)	Block Type B (Unirradiated)	Average A + B (Irradiated)
Reprocessed Fuel			
Quantity/1000 MW(e) HTGR segment	284	580	864
Kilograms/Fuel Element:			
Block graphite	98.0	98.0	98.0
Filler coke	2.8	2.8	2.8
Coating carbon	10.852	8.873	9.525
SiC	0	1.935	1.299
Kernel oxygen + carbon	1.071	0.900	0.956
Thorium	7.946	7.946	7.401
U233 mix	1.113	0	0.365
U235 mix	0	0.757	0.135
Pu	0	0	0.001
Fission products (Approx.)	0	0	0.918
Total Weight	121.782	121.211	121.401
Refabricated Fuel (a)			
Quantity/1000 MW(e) HTGR segment	281	583	
Kilograms/Fuel Element:			
Block graphite	98.0	98.0	
Filler coke	2.8	2.8	
Coating carbon	10.847	8.873	
SiC	0	1.935	
Kernel oxygen + carbon	1.070	0.900	
Thorium	7.946	7.946	
U233 mix	1.111	0	
U235 mix	0	0.757	
Total Weight	121.774	121.211	

(a) Type B element not produced in refabrication facility; it is shown here for information only.

1. Crush and tumble the fuel element and separate the product into graphite-rich and particle-rich streams. Both streams are comminuted to $-3/16$ -in. and fed to two separate fluidized bed burners. The objective of this scheme is to burn the graphite more rapidly than when it is mixed with fuel particles.
2. Crush the fuel element and comminute the fuel particle and graphite mixture to $-3/16$ -in. and feed the product to one type of fluidized bed burner. This scheme is being considered because it may not be possible to separate the graphite from the particles without breaking an unacceptably large fraction of fuel particles.
3. Crush the fuel element and feed the lumps to a modified fluidized bed burner. This scheme may be necessary if it is found that comminution to $-3/16$ in. breaks an unacceptably large fraction of fuel particles.

Two process feed streams to the head-end plant are possible. The first system, which consists of TRISO fissile/TRISO fertile fuel particles, is now being used in the fuel elements for the 330-MW(e) prototype reactor, and the need to know how to separate the fissile from the fertile particles is not urgent at this time because it is planned to feed the bred fuel (U-233) and the small fraction of U-236 to the 1100-MW(e) reactors.

The second system, which utilizes TRISO fissile/BISO fertile fuel particles, is the reference fuel for the 1100-MW(e) reactor, and it is vital to know how to separate the fissile from the fertile particles in order that the U-236 can be retired from the system when its concentration has reached the permissible maximum. Separation must be accomplished by recovering the fissile particles with only minimum breakage.

Crushing tests were carried out on four 12-rod Rezolin-bonded fuel elements using the 5-in. jaw crusher and the results are reported in Table 5.2. It was concluded that a gap of less than 0.475 in. causes large fraction

TABLE 5.2
RESULTS OF CRUSHING 12-ROD FUEL ELEMENTS^(a)

Run No. 5C	1st Jaw Gap (in.)	% Broken ^(b) Particles in the -3/16-in. Material From 1st Jaw Gap Product	2nd ^(c) Jaw Gap (in.)	% Broken ^(b) Particles in the -3/16-in. Material From 2nd Jaw Gap Product	3rd ^(c) Jaw Gap (in.)	% Broken ^(b) Particles in the -3/16-in. Material From 3rd Jaw Gap Product
1	0.475 ^(d)	3.4	5/32	4.7	--	--
2	3/4	1.2	0.475	1.8	5/32	8.2
3	1	1.10	0.475	1.6	--	--
4	1-1/2	0.97	0.475	2.9	--	--

(a) Fuel Rods: TRISO fissile/TRISO fertile, Rezolin bonded. (5-in. jaw crusher with 5/32-in. swing.)

(b) Broken particles in -3/16-in. fraction is the weight fraction that passes through a 250- μ m screen after burning the sample.

(c) Feed is the +3/16-in. material obtained from the previous jaw gap product.

(d) A jaw gap of 0.475-in. is certain to crush a fuel rod, which has a diameter of 0.492-in.

of broken particles. A further disadvantage of this small jaw gap is the low throughput. Table 5.3 shows the throughput at various jaw gaps.

Two 12-rod fuel elements were crushed and tumbled. The results are given in the flowsheets on Figs. 5.1 and 5.2. Particle breakage was about 2% in each case. The overall mass balance is only moderately good because a considerable amount of fines is lost to the ducting during tumbling. The mass balance of fertile and fissile particles is not as good as was expected, and it cannot be concluded where the discrepancy lies (in the expected results, which were calculated from values given by the manufacturer, or in the experimental results). In an effort to resolve this discrepancy, a single fuel rod, from the same batch as the 12 rods, was divided into three pieces and each burned in a tray. The results of the test are shown in Table 5.4, together with those of the 12-rod fuel element values. It appears that the rod loading is not constant along the length of the rod and is probably not constant from rod to rod.

Fluid Bed Burning

2-in. Fluid Bed. Six runs have been carried out with the 2-in. fluid bed, and the results are given in Table 5.5. Run 29 was performed to test the hypothesis that the $-420\text{ }\mu\text{m}$ TRISO/BISO product (containing about 36% carbon) from the 4-in. fluid bed could be burned in a fluid bed that was supplied with heat from a furnace. No difficulties were encountered. The final bed was used in a leaching experiment.

Run 30 was a repeat of 29, except that there was much less carbon in the feed. No difficulties were encountered.

Run 32 was a trial of burning ground fertile particles without a fluidizing medium present. The result was that a hole was burned in the reactor wall. It was concluded that the SiC shells are no substitute for a fluidizing medium.

Run 33 was carried out to collect some SiC-coated fertile particles.

TABLE 5.3
CRUSHER THROUGHPUT AT VARIOUS JAW GAPS
(5-in. Jaw Crusher)

Jaw Gap Open Side (in.)	Feed	Throughput (a) (Kg/min-in.)
0.475	12-rod fuel element	~0.2
3/4	12-rod fuel element	0.43
1	12-rod fuel element	0.63
1-1/2	12-rod fuel element	0.94
0.475	+3/16 in. after crushing at 3/4-in. gap	0.4
0.475	+3/16 in. after crushing at 1-in. gap	0.4
0.475	+3/16 in. after crushing at 1-1/2-in. gap	0.4
0.156 ^(b)	+3/16 in. after crushing at 1/2-in. gap	0.08

(a) The inch refers to the length of the jaw perpendicular to the jaw gap.

(b) This gap creates a product which is 99% -3/16-in. size.

Fig. 5.1. Results of crushing and tumbling a 12-rod fuel element; 1-in. jaw gap (run 5C-3)

Fig. 5.2. Results of crushing and tumbling a 12-rod fuel element; 1-1/2-in. jaw gap (run 5C-4)

TABLE 5.4
RESULTS OF BURNING A SINGLE FUEL ROD COMPARED WITH RUN 5C-3
AND WITH THOSE EXPECTED

Weight: ~100 g
Length: 14 in.
Diameter: 0.492-in.

	Fertile Particles		Fissile Particles		Ash	
	Weight (g)	(%)	Weight (g)	(%)	Weight (g)	(%)
Sample Position:						
Top	15.55	73.65	5.48	25.95	0.083	0.04
Middle	17.32	71.36	6.87	28.31	0.081	0.03
Bottom	14.87	69.59	6.41	29.97	0.100	0.04
Run 5C-3	593.0	75.45	178.9	22.76	14.0	1.78
Expected ^(a)	605.3	74.79	203.7	25.17	0.3	0.04

^(a) By calculation from manufacturer's data.

TABLE 5.5
RESULTS OF OPERATING THE 2-IN. FLUID BED BURNER^(a)

Run F2B-	Feed	Feed Weight (g)	TRISO Weight to SiC (g)	Final Bed Weight (g)	Rate of Combustion (g C/min)	O ₂ in Offgas (%)	C in Final Bed (%)
29	TRISO/BISO crushed pitch- bonded rods -420 μm	3181	~1150	1695	13.0	~25	~0.2
30	TRISO/BISO crushed pitch- bonded rods -420 μm	1423	~936	1380	0.7	~25	0.5
31	Very poor mass balance	--	--	--	--	--	--
32	Ground fertile particles without inert fluidizing	Hole burned in the reactor.				~18	7.9
33	TRISO fertile material with carbon, +420 μm	972	~870	875	0.8	~20	0.6
34	Ground fertile particles with ZrO ₂ as fluidizing medium.	Parti- cles: 270, ZrO ₂ : 510	--	Oxide: 181, ZrO ₂ : 509	1.0	~3	0.7

(a) Unit heated by means of a furnace; batch operation; burner temperature, ~750°C.

Run 34 was a trial of burning ground fertile particles using zirconia as the inert fluidizing medium. No difficulties were encountered, and virtually all the zirconia was separated from the thorium by screening.

The results of these runs show that:

1. The -420 μm material from a continuous exothermic* burner can be batch-burned without difficulty in an endothermic[†] burner.
2. TRISO particles behave quite well as an inert fluidizing medium when BISO particles are being burned.
3. The combustion of ground fertile particles cannot be achieved without a fluidizing medium present.
4. Zirconia, obtainable in spherical form, is a satisfactory fluidizing medium. The spheres, being smooth, can be screened without difficulty.

It is surprising that the burning rate is so low, in most cases, despite the large excess of oxygen being fed to the reactor. This aspect will be investigated further using the 4-in. reactor instead of the 2-in.

The final carbon concentration appears to be about 0.5%, and it appears that it may be time-consuming and oxygen-wasting to get below this value.

4-in. Exothermic Fluid Bed. Operation of the 4-in. exothermic burner was continued using graphite as feed. Runs 69 through 79 were carried out with the objective of recycling carbon fines that were collected in a cyclone, while feeding -3/16-in. graphite on top of the bed. To do this, an eductor was fitted to the apparatus as shown on Fig. 5.3. The results of these runs showed that the cyclone fines blocked the eductor. It was concluded that this

*Exothermic in this context means that sufficient heat is generated in combustion to maintain the temperature.

†Endothermic means that insufficient heat is generated to maintain the temperature and heat has to be supplied.

Fig. 5.3. Arrangement of eductor for recycling fines

method of fines recycle is not satisfactory, and the eductor was therefore discarded.

Run 78 was an experiment in which, in addition to trying to recycling the fines, a high throughput was achieved. The average burning rate was 72 g/min for 37 min.

Runs 80 through 82 were attempts to recycle the fines by mixing it with the -3/16-in. feed graphite and feeding the mixture in at the bottom of the reactor by means of a screw feeder. These three attempts failed because the mixture would not flow. It was concluded that the fines cannot be recycled by mixing it with the feed graphite.

Run 83 was carried out to see whether the fines alone could be fed to the reactor using the screw feeder. The result of the run showed that there was no difficulty in doing this. The run was started using 707- μ m coke, and then 3500 g of cyclone fines from previous runs was fed in at the bottom. Operation was continued without difficulty until combustion ceased through lack of fuel. The final bed consisted of 305 g of unburned fines. The duration was 60 min, and the average burning rate was 47 g/min. It was concluded that if the fines must be recycled, they must be fed alone, using a screw feeder.

Runs 85 through 90 were carried out using a Type 304 stainless steel grate that was drilled out with 3/16-in. holes. The apparatus is illustrated on Fig. 5.4. The objective was to determine under what conditions nonfluidizable (i.e., lump) graphite would burn. It was shown that if the concentration of carbon below the grate is sufficient to consume the oxygen, the carbon above the grate was attacked by the CO_2 very slowly. If the carbon below the grate is allowed to burn out, combustion proceeds above the grate. The danger of having nonfluidized carbon on the grate with oxygen coming through the grate is that high local temperatures can be created with the real possibility of damage to the reactor wall, grate, and thermowell. From past experience, the bed must not be allowed to defluidize while a high concentration of oxygen is being fed. It was concluded from the results of these runs that a grate can be safely used only if the size range of carbon feed to the reactor is

Fig. 5.4. Diagram showing grate in reactor

such that some -3/16-in. material will pass through the grate and burn in a fluidized state so that the concentration of oxygen going up through the grate is reduced to the point where high temperatures are not created in the nonfluidized bed. When a grate is used in the reactor, it is necessary to operate with a fluidizing medium. The grate was removed after Run 90.

Runs 91 through 95 were carried out to determine the operating conditions while burning carbon in the presence of a fluidizing medium. Coke (-707 μm) and alumina (-60 mesh) mixture was fed to the reactor until the bed consisted of 3.0 kg of alumina. Coke was then fed in at the bottom until a total of 2500 g was charged. When combustion ceased, the final bed contained a concentration of 1.1% carbon. The experiment was repeated, but using zirconia as the inert fluidizing medium, and the final concentration was 0.7% carbon. Both runs presented no difficulties.

It was noticed that in each case, the oxygen concentration in the offgas started increasing about 6 min before the temperature started dropping because of the shortage of fuel. This phenomenon indicates that it should be easier to control the reaction by regulating the carbon feed according to the oxygen concentration in the offgas than by the temperature. Two more runs were carried out in a similar manner, but -3/16-in. graphite was fed instead of coke. Both runs started well but ran into difficulties when the temperature began to decrease despite efforts to maintain operation at 980°C. When combustion ceased, the final alumina bed contained 15.7% and the zirconia bed, 21.3% carbon. It was concluded from these runs that the final concentration of carbon depends on surface area available for reaction. If the carbon is coarse, the surface area required to balance the heat loss from the reactor is provided only by a large amount in the fluidizing mediums, but if it is fine, a small amount of carbon is required.

4-in. Endothermic Fluid Bed. This reactor is being constructed and will be operating in the near future. The wall of the bottom of the reactor is surrounded by a furnace that is capable of reaching 1000°C. The objective of this unit is to gain experience on the combustion of carbon mixed with thorium and SiC-coated fissile particles.

8-in. Fluid Bed. The 8-in. fluid bed has been operated seven times. The first six runs enabled the operator to gain experience in starting up. This is done by means of a CO/O₂ flame injected into the bed. The startup time is about 20 min, which is about the same as for the 4-in. fluid bed reactor. These runs showed that the heat radiation was considerable and that some modifications were necessary to prevent ancillary equipment from being damaged. These modifications have been completed and the seventh startup carried out. This trial showed that the reactor can now be safely operated. The burning rate was 113 g/min for 48 min.

Leaching

The final bed from the 2-in. fluid bed (Run 29) was divided into eleven portions, and each portion was successively leached in one liter of Thorex liquor at 115°C. The operation was as follows: the first portion of solids was added to the liquor and, at the end of 10 min, the leacher was drained, the solids separated from the solution, a sample of solution taken, and the solids and liquor returned to the leacher for further leaching. This sequence was carried out four times. After the fourth time out, the liquor was returned to the leacher with the next portion of solids to be leached. This was treated in the same way as the first portion. In this way, all eleven portions were each leached for 40 min. The solutions, which were colorless and clear, were analyzed, and the solid residue was burned and analyzed for thorium. The results of the thorium extraction are shown in Fig. 5.5, where the thorium in solution, measured by analysis, is compared with the weight loss of extracted solids as a function of time.

The results are scattered, but it can be seen that most of the extraction occurs in the first 10 min of leaching. The dissolution rate is maintained up to the end of the experiment when 542 g ThO₂ had dissolved. The residue was found to contain 0.16% of carbon, based on the weight of feed to the leacher, and 1.3 g Th. The latter value indicates that 99.6% of the thorium was extracted in an average of 40 min. It was concluded that there should be no difficulties in leaching burned BISO particles. It is too early to

Fig. 5.5. Dissolution of burned TRISO/BISO pitch-bonded fuel; full-strength Thorex liquor, 115°C

decide that carbon does not interfere with thorium extraction, because the 0.4% unleached thorium may have been retained by the 0.16% carbon.

Some leaching trials were carried out on samples of spherical zirconia particles. The results, given in Fig. 5.6, show that the material is extremely resistant to Thorex liquor attack and may be a contender as an inert fluidizing medium.

Discussion

The unit processes being studied fit into the three alternative schemes shown on Figs. 5.7, 5.8, and 5.9.

Gulf General Atomic is now making pitch-bonded fuel rods, and these are much stronger and more resistant to attrition than the old Rezolin-bonded ones. This leads to the possibility that tumbling may not be an efficient method of separating graphite from fuel particles as shown on Fig. 5.7. Hence, other means of separation are being studied.

In the event that separation of graphite from fuel particles is proved to be impracticable, it will be necessary to consider the comminution of the whole fuel block to provide feed to one type of fluid bed burner. This scheme is shown on Fig. 5.8.

If comminuting the whole fuel block to fluidizable size proves to be impracticable, it will be necessary to feed lumps to the fluidized bed. This scheme is shown in Fig. 5.9. The fluidized bed in this case has to be modified to cope with the lumps. The modification first being considered is the use of grates in the bed.

Fig. 5.6. Dissolution of zirconia in full-strength Thorex liquor, 115°C

Fig. 5.7. Head-end conceptual flowsheet; graphite/particle separation; TRISO fissile/BISO fertile

Fig. 5.8. Head-end conceptual flowsheet; no graphite/particle separation; TRISO fissile/BISO fertile

Fig. 5.9. Head-end conceptual flowsheet; no graphite/particle separation and no comminution; TRISO fissile/BISO fertile

TASK VIII
PHYSICS AND FUEL MANAGEMENT

FUEL TEST ELEMENT

Test Element Program

The test element schedule was modified during the quarter. The entire test element program includes Phase 1, Phase 2, and Phase 3 portions, with Phase 1 consisting of the test elements inserted into the Peach Bottom reactor in June 1970. Phase 2 includes three fuel test elements that are to be fabricated and inserted into the Peach Bottom reactor core prior to June 30, 1971. Phase 3 of the program includes two additional fuel test elements that are to be designed, licensed, and fabricated before June 30, 1971. Insertion of these elements will take place at the next refueling shutdown. A schedule of the revised program is shown in Fig. 8.1.

Element Design

The Phase 2 Base Program fuel test element design parameters were established during the quarter, and these are given in Table 8.1. The parameters defined as approximate require detail engineering effort.

The Phase 2 Base Program elements are of the Fuel Test Element type and are designated FTE-3, FTE-4, and FTE-6. The fuel beds are rods, and there are several possible fuel combinations that may be used in the quartered elements:

1. $(\text{Th,U})\text{C}_2$ BISO + ThC_2 BISO.
2. $(\text{Th,U})\text{C}_2$ TRISO + ThC_2 BISO.

TASK	FISCAL YEAR 1971											
	JUL	AUG	SEP	OCT	NOV	DEC	JAN	FEB	MAR	APR	MAY	JUN
PHASE 2												
1. PLANNING												
2. ENGINEERING												
3. STANDARD COMPONENT PROCUREMENT	COMPLETE											
4. FUEL PRODUCTION												
5. CENTER SAMPLE FABRICATION	COMPLETE											
6. ELEMENT ASSEMBLY												
7. DATA PACKAGE												
8. SHIPMENT												
9. REACTOR LOADING AND IRRADIATION												
PHASE 3												
1. PLANNING												
2. ENGINEERING												
3. STANDARD COMPONENT PROCUREMENT												
4. FUEL PRODUCTION												
5. CENTER SAMPLE FABRICATION												
6. ELEMENT ASSEMBLY												
7. SAFETY DOCUMENT (IF REQUIRED)												
8. DATA PACKAGE												

Fig. 8.1. Base program test element schedule

TABLE 8.1
PHASE 2 BASE PROGRAM TEST ELEMENT DESIGN PARAMETERS

Mixed fissile particle Th/U Ratio	2.75
Fuel kernel size, μm :	
Fissile B (mixed Th/U)	175-275
Fissile (U)	70-130
Fertile (Th)	300-450
Fuel bed temperature, $^{\circ}\text{F}$:	
Maximum	2300
Volume median	2025
Maximum gradient	250
Linear heat rating, Kw/ft per rod:	
Peak	3.2
Average	2.4
Fast-neutron fluence, 10^{21} n/cm^2 :	
FTE-4 (300 EFPD)	
Maximum	~ 1.3
Average	~ 1.0
FTE-3, -6 (600 EFPD)	
Maximum	~ 2.6
Average	~ 2.0
Maximum fuel particle burnup, % FIMA:	
FTE-4 Average	~ 4.0
FTE-3, -6 Average	~ 8.0
Fuel bed R/B ($\text{Kr}^{85\text{m}}$) at 1200°C	$\leq 1 \times 10^{-4}$

3. UC_2 TRISO + ThC_2 BISO.
4. $(\text{Th,U})\text{C}_2$ TRISO + ThC_2 TRISO.
5. $(\text{Th,U})\text{O}_2$ TRISO + ThO_2 BISO.
6. UO_2 TRISO + ThO_2 BISO.

The fuel test elements will contain center samples. They will each be instrumented with two thermocouples in order to measure (1) the temperature of the fuel body inside diameter at the thermal peak axial position and (2) the temperature of the fuel body at a point outside the ring of fuel holes at the same axial position. There will be no provision for individual purge sampling.

The design parameters for the Phase 3 test elements are being determined.

Thermal Design

An analysis was made to determine representative temperature distributions for the fuel in the test elements; the results are presented in Figs. 8.2 through 8.5. These figures show typical integrated and derived distributions for elements with blended bed and bonded rod fuel.

Figures 8.2 and 8.4 present the temperature of the entire volume of fuel within a typical test element that is designed to have a peak fuel temperature of 2300°F. Elements with a different peak fuel temperature can be compared by sliding the T coordinate to the specific peak temperature in question.

Figures 8.3 and 8.5 are expansions of Figs. 8.2 and 8.4, respectively. The actual percentages of fuel, DF, in a specific temperature range can be determined. DF is equal to the area under the curves shown in Figs. 8.3 and 8.5 between a specific temperature range. The total area under these curves is equal to 1 or 100%. Specific DFs have been calculated for different temperature ranges and are summarized in Table 8.2.

Fig. 8.2. Integrated temperature distribution of test element fuel, blended bed with conductivity of 1.5 Btu/hr-ft-°F

Fig. 8.3. Temperature distribution of test element fuel, blended bed with conductivity of 1.5 Btu/hr-ft-°F

Fig. 8.4. Integrated temperature distribution of test element fuel, bonded rod with conductivity of 4.0 Btu/hr-ft-°F

Fig. 8.5. Temperature distribution of test element fuel, bonded rod with conductivity of 4.0 Btu/hr-ft-°F

TABLE 8.2
TEMPERATURE DISTRIBUTION

Temp Range (°F)	Fuel in Range, DF (%)	
	Blended Bed	Bonded Rod
2300 to 2200	9	8
2200 to 2100	21	23
2100 to 2000	26	23
2000 to 1900	14	12
1900 to 1800	6	5
1800 to 1700	5	5

From the table it can be seen that the conductivity of the fuel (1.5 Btu/hr-ft-°F for the blended bed versus 4.0 Btu/hr-ft-°F for the bonded rods) does not influence the temperature distribution to any meaningful extent.

Element Fabrication

Fuel particles requirements for the phase 2 test elements have been determined and production of these particles is in progress. As reported in the previous quarterly report (GA-10288), all other components, including center samples, are ready for assembly.

HTLTR SUPPORT ACTIVITIES

Summary

The GGA calculations of the variation of k_{∞} with temperature for HTGR lattice 1 (C/Th = 200, C/U²³⁵ = 6000) were reviewed and new calculations performed by iterating between zero-dimensional spectrum calculations and multigroup one-dimension diffusion theory calculations until converged bucklings were obtained. Particle self-shielding effects were included as well as as-built U-234, U-236, and N-14 concentrations. The calculated 300° to 1200°K temperature defect of $\Delta k_{\infty}' = -0.059$ decreased significantly compared to previous results based on spectrum calculations with zero buckling.

A new derivation of the theory behind the correction of the experimental data for leakage and flux mismatch effects was carried out. This derivation of leakage and flux mismatch terms agrees with a recently revised BNWL derivation, although there is apparently some disagreement as to just what buckling should be used. A review of all of the corrections to the experimental data indicated that the most probable source of any remaining uncertainties in the HTLTR results for HTGR lattice 1 are due to temperature dependence of the increased reflection encountered when the test cell is withdrawn or to the correction for the nonfueled "end cap" regions located at the ends of each test block. Each of these corrections amounts to about 20% of the measured reactivity change.

Two-energy-group, two-dimensional transport theory calculations were performed to assess the temperature dependence of the increased reflection encountered when the HTGR test cell was withdrawn. The reactivity worth of the cluster of four protruding HTGR fuel blocks was found to decrease by about 4% between 300° and 1200°K, with an estimated uncertainty of about 4% in the computed temperature dependence due to convergence limitations. If no temperature dependence for this correction is assumed, a revised experimental temperature defect of -0.067 in k_{∞}' is obtained between 300° and 1200°K compared to -0.075 in k_{∞}' obtained with the previously assumed temperature dependence.

Nine-energy-group, two-dimensional transport theory calculations for a small unit cell containing one fuel rod were performed to assess the reactivity perturbations caused by the HTGR fuel block ends. The results were qualitatively in agreement with the experimental corrections made by Battelle (about 0.02 in $\Delta k/k$). It is not clear that such an experimental correction should be made, since normal reactor cell calculations must always be capable of treating such effects, and all of the graphite in the test region must be accounted for in order to obtain the correct two-group cross sections for the calculation of the leakage and flux mismatch corrections to the HTLTR experiments.

Thermal group absorption cross sections for copper were computed and compared with those used by Battelle. The Gulf General Atomic values were very close in both magnitude (<1.7% difference) and temperature dependence (<0.2% difference between 300° to 1200°K ratios) to the BNWL values.

Data regarding the ThO_2 and UC_2 particle size distributions obtained from Battelle in August indicated that no changes from this source are required in the Gulf General Atomic calculations.

Table 8.3 summarizes the best presently available Gulf General Atomic calculations for HTGR Lattice 1 ($\text{C/Th} = 200$, $\text{C/U}^{235} = 6000$) as well as experimental results revised (by Gulf General Atomic) for zero temperature dependence of the extended block correction.

TABLE 8.3
CALCULATIONS FOR LATTICE 1

Temp (°K)	GGA	Revised BNWL Experiment	Difference
$k'_{\infty}(300^\circ\text{K})$	1.0825	1.1008	0.0183
$k'_{\infty}(1200^\circ\text{K})$	1.0231	1.0336	0.0105
$\Delta k'_{\infty}(300^\circ \rightarrow 1200^\circ\text{K})$	0.0594	0.0672	0.0078

Derivation of Revised Formulas for Analysis of HTLTR Experiments

1. Derivation of the Basic Formulas

A multiregion assembly V_o is considered with an internal cavity V , as shown in Fig. 8.6.

Fig. 8.6. Multiregion Assembly V_o

V_o is assumed to be bounded by the internal surface S and the external surface S_o .

It is intended to measure reactivities and reactivity coefficients for certain samples of multiplying media by placing these probes into the cavity V.

It is assumed that the unperturbed situation, with a void in V, can be described in a two group diffusion theory model. The two group diffusion equations in the usual notation are:

$$-\nabla(D_1 \nabla \phi_1) + (\Sigma_1^a + \Sigma_{1 \rightarrow 2}) \phi_1 = \frac{1}{k} (\nu \Sigma_1^f \phi_1 + \nu \Sigma_2^f \phi_2) , \quad (1)$$

$$-\nabla(D_2 \nabla \phi_2) + \Sigma_2^a \phi_2 - \Sigma_{1 \rightarrow 2} \phi_1 = 0 .$$

The adjoint two group diffusion equations have the form

$$-\nabla(D_1 \nabla \phi_1^+) + (\Sigma_1^a + \Sigma_{1 \rightarrow 2}) \phi_1^+ = \frac{1}{k} \nu \Sigma_1^f \phi_1^+ \quad (2)$$

$$-\nabla(D_2 \nabla \phi_2^+) + \Sigma_2^a \phi_2^+ = \frac{1}{k} \nu \Sigma_2^f \phi_1^+ .$$

In operator form these equations can be written as follows:

$$A \phi = \frac{1}{k} B \phi , \quad (3)$$

$$A^+ \phi^+ = \frac{1}{k} B^+ \phi^+ . \quad (4)$$

The perturbed equations, with some material probe in the cavity V, can be written in the following form

$$A_c \phi_c = \frac{1}{k_c} B_c \phi_c , \quad (5)$$

$$A_c^+ \phi_c^+ = \frac{1}{k_c} B_c^+ \phi_c^+ . \quad (6)$$

If equation (5) is multiplied (inner product) by ϕ^+ and equation (4) by ϕ_c and if these equations are subtracted and integrated over V_o , then

$$\int_{V_o} [\phi^+, A_c \phi_c] - [\phi_c, A^+ \phi^+] dV = \frac{1}{k_c} \int_{V_o} [\phi^+, B_c \phi_c] dV - \frac{1}{k} \int_{V_o} [\phi_c, B^+ \phi^+] dV . \quad (7)$$

Within V_o one has

$$A_c = A \quad \text{and} \quad B_c = B .$$

Therefore, equation (7) can be simplified in the following way:

$$\begin{aligned} - \int_{V_o} \left\{ \phi_1^+ \nabla D_1 \nabla \phi_{c,1} + \phi_2^+ \nabla D_2 \nabla \phi_{c,2} - \phi_{c,1} \nabla D_1 \nabla \phi_1^+ - \phi_{c,2} \nabla D_2 \nabla \phi_2^+ \right\} dV \\ = \left(\frac{1}{k_c} - \frac{1}{k} \right) \int_{V_o} \left\{ \nu \Sigma_1^f \phi_1^+ \phi_{c,1} + \nu \Sigma_2^f \phi_1^+ \phi_{c,2} \right\} dV . \quad (8) \end{aligned}$$

The left side of equation (8) can be further simplified by making use of the identity

$$\nabla \cdot (\psi D \nabla \psi) = \nabla \psi D \nabla \psi + \psi (\nabla D \nabla \psi) \quad (9)$$

and by using Gauss' Theorem to transform the volume integral into a surface integral ($\underline{\sigma}$ is the outward directed normal)

$$\begin{aligned} - \int_{S+S_o} \left\{ \phi_1^+ D_1 \nabla \phi_{c,1} + \phi_2^+ D_2 \nabla \phi_{c,2} - \phi_{c,1} D_1 \nabla \phi_1^+ - \phi_{c,2} D_2 \nabla \phi_2^+ \right\} \underline{d\sigma} \\ = \left(\frac{1}{k_c} - \frac{1}{k} \right) \int_{V_o} \left\{ \nu \Sigma_1^f \phi_1^+ \phi_{c,1} + \nu \Sigma_2^f \phi_1^+ \phi_{c,2} \right\} dV . \quad (10) \end{aligned}$$

The quantities ϕ_g^+ and $\phi_{c,g}$ are zero on S_o and $\nabla\phi_g^+$ is zero on S . This gives

$$\int_S \{ \phi_1^+ D_1 \nabla \phi_{c,1} + \phi_2^+ D_2 \nabla \phi_{c,2} \} d\sigma = +\Delta\rho_c \int_{V_o} \phi_1^+ (\nu \Sigma_1^f \phi_{c,1} + \nu \Sigma_2^f \phi_{c,2}) dV, \quad (11)$$

where

$$-\Delta\rho_c = \frac{1}{k_c} - \frac{1}{k}.$$

If we observe that the ϕ_g^+ are constant within V , then we can transform equation (11) into the following expression:

$$\int_V \{ \phi_1^+ \nabla D_1 \nabla \phi_{c,1} + \phi_2^+ \nabla D_2 \nabla \phi_{c,2} \} dV = \Delta\rho_c \int_{V_o} \phi_1^+ (\nu \Sigma_1^f \phi_{c,1} + \nu \Sigma_2^f \phi_{c,2}) dV. \quad (12)$$

With

$$\int_V \nabla D_g \nabla \phi_{c,g} dV = -D_g B_{c,g}^2 \bar{\phi}_{c,g} V$$

one obtains

$$\Delta\rho_c \int_{V_o} \phi_1^+ (\nu \Sigma_1^f \phi_{c,1} + \nu \Sigma_2^f \phi_{c,2}) dV = +(\phi_1^+ B_{c,1}^2 \phi_{c,1} + \phi_2^+ D_2 B_{c,2}^2 \phi_{c,2}) V. \quad (13)$$

Another, sometimes more convenient, formulation is

$$\begin{aligned} \Delta\rho_c \int_{V_o} \phi_1^+ (\nu \Sigma_1^f \phi_{c,1} + \nu \Sigma_2^f \phi_{c,2}) dV = & - \int_V \left\{ \phi_1^+ \left[(\Sigma_1^a + \Sigma_{1 \rightarrow 2}) \phi_{c,1} - \frac{1}{k_c} (\nu \Sigma_1^f \phi_{c,1} + \nu \Sigma_2^f \phi_{c,2}) \right] \right. \\ & \left. + \phi_2^+ \left[\Sigma_2^a \phi_{c,2} - \Sigma_{1 \rightarrow 2} \phi_{c,1} \right] \right\} dV. \end{aligned} \quad (14)$$

For a pure absorber, designated by the index p , one obtains

$$-\Delta\rho_p \int_{V_o} \phi_1^+ (\nu \Sigma_1^f \phi_{p,1} + \nu \Sigma_2^f \phi_{p,2}) dV = \int_V \left\{ \phi_1^+ \Sigma_1^p \phi_{p,1} + \phi_2^+ \Sigma_2^p \phi_{p,2} \right\} dV. \quad (15)$$

If we assume that the fluxes $\phi_{c,g}$ and $\phi_{p,g}$ are normalized so that

$$\int_{V_o} \phi_1^+ (\nu \Sigma_1^f \phi_{c,1} + \nu \Sigma_2^f \phi_{c,2}) dV = \int_{V_o} \phi_1^+ (\nu \Sigma_1^f \phi_{p,1} + \nu \Sigma_2^f \phi_{p,2}) dV,$$

then by introducing average fluxes we obtain the following equation from (14) and (15):

$$\bar{\phi}_{c,g} = \frac{1}{V} \int_V \phi_{c,g} dV; \quad \bar{\phi}_{p,g} = \frac{1}{V} \int_V \phi_{p,g} dV,$$

$$\frac{\Delta\rho_c}{\Delta\rho_p} = \left(\frac{V_c}{V_p} \right) \cdot \frac{\phi_1^+ \left[\left(\Sigma_1^a + \Sigma_{1 \rightarrow 2} \right) \bar{\phi}_{c,1} - \frac{1}{k_c} \left(\nu \Sigma_1^f \bar{\phi}_{c,1} + \nu \Sigma_2^f \bar{\phi}_{c,2} \right) \right] + \phi_2^+ \left[\Sigma_2^a \bar{\phi}_{c,2} - \Sigma_{1 \rightarrow 2} \bar{\phi}_{c,1} \right]}{\phi_1^+ \Sigma_1^p \bar{\phi}_{p,1} + \phi_2^+ \Sigma_2^p \bar{\phi}_{p,2}}. \quad (16)$$

If bucklings are used one obtains*

$$\frac{\Delta\rho_c}{\Delta\rho_p} = - \left(\frac{V_c}{V_p} \right) \frac{\phi_1^+ D_{1c,1}^2 \bar{\phi}_{c,1} + \phi_2^+ D_{2c,2}^2 \bar{\phi}_{c,2}}{\phi_1^+ \Sigma_1^p \bar{\phi}_{p,1} + \phi_2^+ \Sigma_2^p \bar{\phi}_{p,2}}. \quad (17)$$

*Equation (21) gives the proper definition of these bucklings.

If we define

$$k_{\infty}^c = \frac{v \Sigma_1^{\bar{f}} \phi_{c,1} + v \Sigma_2^{\bar{f}} \phi_{c,2}}{\Sigma_1^{\bar{a}} \phi_{c,1} + \Sigma_2^{\bar{a}} \phi_{c,2}} \quad (18)$$

then equation (16) can be written

$$\frac{\Delta \rho_c}{\Delta \rho_p} = \left(\frac{v_c}{v_p} \right) \cdot \frac{\phi_1^+ \left[\left(\Sigma_1^{\bar{a}} + \Sigma_{1 \rightarrow 2} \right) \bar{\phi}_{c,1} - \frac{k_{\infty}^c}{k_c} \left(\Sigma_1^{\bar{a}} \phi_{c,1} + \Sigma_2^{\bar{a}} \phi_{c,2} \right) \right] + \phi_2^+ \left[\Sigma_2^{\bar{a}} \phi_{c,2} - \Sigma_{1 \rightarrow 2} \phi_{c,1} \right]}{\phi_1^+ \Sigma_1^{\bar{p}} \phi_{p,1} + \phi_2^+ \Sigma_2^{\bar{p}} \phi_{p,2}} .$$

The last equation can be modified in the following way:

$$\begin{aligned} & \frac{\Delta \rho_c}{\Delta \rho_p} \left(\frac{v_p}{v_c} \right) \left(\frac{\Sigma_2^{\bar{p}} \phi_{p,2}}{\Sigma_2^{\bar{a}} \phi_{c,2}} \right) \left[1 + \frac{\phi_1^+}{\phi_2^+} \left(\frac{\Sigma_1^{\bar{p}} \phi_{p,1}}{\Sigma_2^{\bar{p}} \phi_{p,2}} \right) \right] \\ &= \frac{\phi_1^+}{\phi_2^+} \left(\frac{\Sigma_1^{\bar{a}} + \Sigma_{1 \rightarrow 2}}{\Sigma_2^{\bar{a}}} \right) \frac{\bar{\phi}_{c,1}}{\bar{\phi}_{c,2}} - \frac{\phi_1^+}{\phi_2^+} \frac{k_{\infty}^c}{k_c} \left[\left(\frac{\Sigma_1^{\bar{a}} \phi_{c,1}}{\Sigma_2^{\bar{a}} \phi_{c,2}} \right) + 1 \right] + 1 - \frac{\Sigma_{1 \rightarrow 2} \phi_{c,1}}{\Sigma_2^{\bar{a}} \bar{\phi}_{c,2}} \\ &= 1 - \frac{k_{\infty}^c}{k_c} \frac{\phi_1^+}{\phi_2^+} \left[1 + \frac{\Sigma_1^{\bar{a}} \phi_{c,1}}{\Sigma_2^{\bar{a}} \phi_{c,2}} \right] + \frac{\phi_1^+ \Sigma_1^{\bar{a}} \phi_{c,1}}{\phi_2^+ \Sigma_2^{\bar{a}} \phi_{c,2}} + \left(\frac{\phi_1^+}{\phi_2^+} - 1 \right) \left(\frac{\Sigma_{1 \rightarrow 2} \bar{\phi}_{c,1}}{\Sigma_2^{\bar{a}} \phi_{c,2}} \right) . \quad (19) \end{aligned}$$

This gives, for k_{∞}^c , the expression

$$k_{\infty}^c = k_c \frac{\phi_2^+}{\phi_1^+} \left[1 + \frac{\Sigma_{1c,1}^{a-}}{\Sigma_{2c,2}^{a-}} \right]^{-1} \left\{ 1 - \frac{\Delta \rho_c}{\Delta \rho_p} \left(\frac{V_p \Sigma_{2p,2}^{p-}}{V_c \Sigma_{2c,2}^{a-}} \right) \left[1 + \frac{\phi_1^+ \Sigma_{1p,1}^{p-}}{\phi_2^+ \Sigma_{2p,2}^{p-}} \right] \right. \\ \left. + \frac{\phi_1^+ \Sigma_{1c,1}^{a-}}{\phi_2^+ \Sigma_{2c,2}^{a-}} + \left(\frac{\phi_1^+}{\phi_2^+} - 1 \right) \left(\frac{\Sigma_{1 \rightarrow 2c,1}^{a-}}{\Sigma_{2c,2}^{a-}} \right) \right\} \quad (20)$$

This formula can easily be generalized to multigroup diffusion theory; this is done in Section 3.

Equations (16), (17), and (20) are exact under the assumption that the unperturbed and the perturbed reactor can be described by the two-group Eqs. (3) through (6). In particular, it is assumed that the two-group cross sections in V_0 are not affected by the perturbations in V . These equations contain only quantities related to the unperturbed and perturbed reactor configurations. Equation (20) [or the equivalent multigroup Eq. (65)] avoid the introduction of a somewhat artificial bare critical system and do not require flux mismatch terms.

2. An Alternate Derivation of the Old Formulas

In this section, Eq. (17) is expressed in terms of a bare critical system composed of the material that was put into cavity V in order to obtain equations of the same forms as those obtained by Heineman (Ref. 1) and Lippincott (Ref. 2).

If one uses Equation (17) one has to keep in mind that the bucklings $B_{c,g}^2$ and the fluxes $\bar{\phi}_{c,g}$ are defined as follows:

$$-D_g B_{c,g}^2 \bar{\phi}_{c,g}^V = \int_V \nabla D_g \nabla \phi_{c,g} dV, \quad g=1,2 \quad (21)$$

$$\bar{\phi}_{c,g} = \frac{1}{V} \int_V \phi_{c,g} dV, \quad g=1,2. \quad (22)$$

Equation (17) can now be written in the following way:

$$-\frac{\Delta \rho_c}{\Delta \rho_p} \left(\frac{V \Sigma_2^p \bar{\phi}_{2,p}}{V \Sigma_2^a \phi_{c,2}} \right) \left[1 + \frac{\phi_1^+ \Sigma_1^p \bar{\phi}_{p,1}}{\phi_2^+ \Sigma_2^p \bar{\phi}_{p,2}} \right] = \left\{ \frac{\phi_1^+}{\phi_2^+} \left(\frac{D_1 B_{c,1}^2}{\Sigma_2^a} \right) \frac{\phi_{c,1}}{\phi_{c,2}} + \frac{D_2 B_{c,2}^2}{\Sigma_2^a} \right\}. \quad (23)$$

If one considers a bare critical assembly for the material which was put into the cavity V then one obtains another set of two group equations (the bare critical assembly is designated by an index "b")

$$\begin{aligned} D_1 B_{b,1}^2 \bar{\phi}_{b,1} + (\Sigma_1^a + \Sigma_{1 \rightarrow 2}) \bar{\phi}_{b,1} &= \nu \Sigma_1^f \bar{\phi}_{b,1} + \nu \Sigma_2^f \bar{\phi}_{b,2} \\ D_2 B_{b,2}^2 \bar{\phi}_{b,2} + \Sigma_2^a \bar{\phi}_{b,2} - \Sigma_{1 \rightarrow 2} \bar{\phi}_{b,1} &= 0 \end{aligned} \quad (24)$$

$$\begin{aligned}
D_1 B_{b,1}^2 \frac{\overline{\phi}_{b,1}^+}{\Sigma_1^a + \Sigma_{1 \rightarrow 2}} - \Sigma_{1 \rightarrow 2} \overline{\phi}_{b,2}^+ &= v \Sigma_1^f \overline{\phi}_{b,1}^+ \\
D_2 B_{b,2}^2 \frac{\overline{\phi}_{b,2}^+}{\Sigma_2^a + \Sigma_{2 \rightarrow 1}} &= v \Sigma_2^f \overline{\phi}_{b,1}^+ .
\end{aligned} \tag{25}$$

We will use the following abbreviations:

$$\tau = \frac{D_1}{\Sigma_1^a + \Sigma_{1 \rightarrow 2}} , \quad L^2 = \frac{D_2}{\Sigma_2^a} , \quad p = \frac{\Sigma_{1 \rightarrow 2}}{\Sigma_{1 \rightarrow 2} + \Sigma_1^a} , \tag{26}$$

$$\eta_1^{f_1} = \frac{v \Sigma_1^f}{\Sigma_1^a} , \quad \eta_2^{f_2} = \frac{v \Sigma_2^f}{\Sigma_2^a} , \tag{27}$$

$$k_\infty^b = \frac{v \Sigma_1^f \overline{\phi}_{b,1}^+ + v \Sigma_2^f \overline{\phi}_{b,2}^+}{\Sigma_1^a \overline{\phi}_{b,1}^+ + \Sigma_2^a \overline{\phi}_{b,2}^+} , \quad k_\infty^b = \frac{(1+\tau B_{b,1}^2)(1+L^2 B_{b,2}^2)}{1 + (1-p)L^2 B_{b,2}^2} . \tag{28}$$

The flux quotients can be expressed as follows:

$$\frac{\phi_{b,1}}{\phi_{b,2}} = \frac{D_2 B_{b,2}^2 + \Sigma_2^a}{\Sigma_{1 \rightarrow 2}} , \tag{29}$$

$$\frac{\phi_{b,1}^+}{\phi_{b,2}^+} = \frac{\Sigma_{1 \rightarrow 2}}{D_1 B_{b,1}^2 + \Sigma_1^a + \Sigma_{1 \rightarrow 2} - v \Sigma_1^f} = \frac{D_2 B_{b,2}^2 + \Sigma_2^a}{v \Sigma_2^f} . \tag{30}$$

The right side of Eq. (24)

$$\left\{ \right\} = \frac{\phi_1^+}{\phi_2^+} \left(\frac{D_1 B_{c,1}^2}{\Sigma_2^a} \right) \left(\frac{\bar{\phi}_{c,1}}{\bar{\phi}_{c,2}} \right) + \frac{D_2 B_{c,2}^2}{\Sigma_2^a}$$

can be written in another form if one uses the following relations:

$$B_{c,1}^2 = \frac{1}{D_1} \left[\frac{1}{k_c} \left(v_{\Sigma_1^f} + v_{\Sigma_2^f} \frac{\bar{\phi}_{c,2}}{\bar{\phi}_{c,1}} \right) - \left(\Sigma_1^a + \Sigma_{1 \rightarrow 2} \right) \right]$$

$$B_{c,2}^2 = \frac{1}{D_2} \left(\Sigma_{1 \rightarrow 2} \frac{\bar{\phi}_{c,1}}{\bar{\phi}_{c,2}} - \Sigma_2^a \right) \quad (31)$$

$$\Delta \phi^+ = \frac{\phi_1^+}{\phi_2^+} - \frac{\bar{\phi}_{b,1}^+}{\bar{\phi}_{b,2}^+}, \quad \Delta \phi = \frac{\bar{\phi}_{c,1}}{\bar{\phi}_{c,2}} - \frac{\bar{\phi}_{b,1}}{\bar{\phi}_{b,2}}. \quad (32)$$

The new form for the right side of Eq. 24 is

$$\left\{ \right\} = \left(\frac{\phi_1^+}{\phi_2^+} \right) \left(\frac{1}{k_c} \left(\frac{v_{\Sigma_1^f}}{\Sigma_2^a} + \frac{v_{\Sigma_2^f}}{\Sigma_2^a} \left(\frac{\bar{\phi}_{c,2}}{\bar{\phi}_{c,1}} \right) \right) - \frac{\Sigma_1^a + \Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \right) \left(\frac{\bar{\phi}_{c,1}}{\bar{\phi}_{c,2}} \right) + \frac{\Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \left(\frac{\phi_{c,1}}{\phi_{c,2}} \right) - 1 \quad (33)$$

or

$$= \left(\frac{\phi_1^+}{\phi_2^+} \right) \left[\left(\frac{v_{\Sigma_1^f}}{k_c \Sigma_2^a} - \frac{\Sigma_1^a + \Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \right) \left(\frac{\bar{\phi}_{c,1}}{\bar{\phi}_{c,2}} \right) + \frac{v_{\Sigma_2^f}}{k_c \Sigma_2^a} \right] + \frac{\Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \left(\frac{\bar{\phi}_{1,c}}{\bar{\phi}_{2,c}} \right) - 1 \quad (34)$$

or somewhat simplified

$$\left\{ \right\} = \left(\Delta\phi^+ + \frac{\overline{\phi}_{b,1}^+}{\overline{\phi}_{b,2}^+} \right) \left[\left(\frac{v\Sigma_1^f}{k_c \Sigma_2^a} - \frac{\Sigma_1^a + \Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \right) \left(\Delta\phi + \left(\frac{\overline{\phi}_{b,1}}{\overline{\phi}_{b,2}} \right) \right) + \frac{v\Sigma_2^f}{k_c \Sigma_2^a} \right] + \frac{\Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \left(\Delta\phi + \frac{\overline{\phi}_{b,1}}{\overline{\phi}_{b,2}} \right) - 1. \quad (35)$$

By using the notation

$$\delta\phi^+ = \frac{\Delta\phi^+}{\left(\frac{\overline{\phi}_{b,1}^+}{\overline{\phi}_{b,2}^+} \right)}, \quad \delta\phi = \frac{\Delta\phi}{\left(\frac{\overline{\phi}_{b,1}}{\overline{\phi}_{b,2}} \right)} \quad (36)$$

we obtain

$$\begin{aligned} \left\{ \right\} &= \left(\frac{\overline{\phi}_{b,1}^+}{\overline{\phi}_{b,2}^+} \right) \left[\left(\frac{v\Sigma_1^f}{k_c \Sigma_2^a} - \frac{\Sigma_1^a + \Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \right) \left(\frac{\overline{\phi}_{b,1}}{\overline{\phi}_{b,2}} \right) + \frac{v\Sigma_2^f}{k_c \Sigma_2^a} \right] + \frac{\Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \frac{\overline{\phi}_{b,1}}{\overline{\phi}_{b,2}} - 1 \\ &+ \delta\phi^+ \delta\phi \left[\left(\frac{\overline{\phi}_{b,1}^+}{\overline{\phi}_{b,2}^+} \right) \left(\frac{\overline{\phi}_{b,1}}{\overline{\phi}_{b,2}} \right) \left(\frac{v\Sigma_1^f}{k_c \Sigma_2^a} - \frac{\Sigma_1^a + \Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \right) \right] \\ &+ \delta\phi^+ \left[\left(\frac{\overline{\phi}_{b,1}^+}{\overline{\phi}_{b,2}^+} \right) \left(\frac{\overline{\phi}_{b,1}}{\overline{\phi}_{b,2}} \right) \left(\frac{v\Sigma_1^f}{k_c \Sigma_2^a} - \frac{\Sigma_1^a + \Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \right) + \frac{v\Sigma_2^f}{k_c \Sigma_2^a} \left(\frac{\overline{\phi}_{b,1}^+}{\overline{\phi}_{b,2}^+} \right) \right] \\ &+ \delta\phi \left[\left(\frac{\overline{\phi}_{b,1}}{\overline{\phi}_{b,2}} \right) \left(\frac{\overline{\phi}_{b,1}}{\overline{\phi}_{b,2}} \right) \left(\frac{v\Sigma_1^f}{k_c \Sigma_2^a} - \frac{\Sigma_1^a + \Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \right) + \frac{\Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \left(\frac{\overline{\phi}_{b,1}}{\overline{\phi}_{b,2}} \right) \right] \end{aligned} \quad (37)$$

By using equations (29) and (30) and the relations

$$\frac{\Sigma_{1 \rightarrow 2}}{D_1 B_{b,1}^2 + \Sigma_1^a + \Sigma_{1 \rightarrow 2} - v \Sigma_1^f} \cdot \left(\frac{v \Sigma_1^f}{k_c \Sigma_2^a} - \frac{\Sigma_1^a + \Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \right) \left(\frac{D_2 B_{b,2}^2 + \Sigma_2^a}{\Sigma_{1 \rightarrow 2}} \right)$$

$$= \frac{1}{1 + \tau B_{b,1}^2 - (1-p)\eta_1 f_1} \left(\frac{(1-p)\eta_1 f_1}{k_c} - 1 \right) \left(1 + L^2 B_{b,2}^2 \right)$$

$$\frac{D_2 B_{b,2}^2 + \Sigma_2^a}{v \Sigma_2^f} \cdot \frac{v \Sigma_2^f}{k_c \Sigma_2^a} = \frac{1}{k_c} \left(1 + L^2 B_{b,2}^2 \right)$$

$$\frac{\Sigma_{1 \rightarrow 2}}{\Sigma_2^a} \frac{D_2 B_{b,2}^2 + \Sigma_2^a}{\Sigma_{1 \rightarrow 2}} = 1 + L^2 B_{b,2}^2$$

$$\frac{1}{k_c} \cdot \frac{1 + L^2 B_{b,2}^2}{1 + \tau B_{b,1}^2 - (1-p)\eta_1 f_1} \left((1-p)\eta_1 f_1 - k_c + 1 + \tau B_{b,1}^2 - (1-p)\eta_1 f_1 \right)$$

$$= \frac{1}{k_c} \frac{1 + L^2 B_{b,2}^2}{1 + \tau B_{b,1}^2 - (1-p)\eta_1 f_1} \left(1 - k_c + \tau B_{b,1}^2 \right)$$

we obtain the following relation

$$\begin{aligned}
\left\{ \right\} &= \left(\frac{1}{k_c} \right) \left(\frac{1 + L^2 B_{b,2}^2}{1 + \tau_{B_{b,1}}^2 - (1-p)\eta_1 f_1} \right) \left(1 - k_c + \tau_{B_{b,2}}^2 \right) + \left(1 + L^2 B_{b,2}^2 \right) \\
&+ \delta\phi^+ \delta\phi \left[\frac{1 + L^2 B_{b,2}^2}{1 + \tau_{B_{b,1}}^2 - (1-p)\eta_1 f_1} \left(\frac{(1-p)\eta_1 f_1}{k_c} \right) \right] \\
&+ \delta\phi^+ \left[\frac{1 + L^2 B_{b,2}^2}{1 + \tau_{B_{b,1}}^2 - (1-p)\eta_1 f_1} \left(\frac{1 - k_c}{k_c} + \frac{\tau_{B_{b,1}}^2}{k_c} \right) \right] \\
&+ \delta\phi \left[\frac{1 + L^2 B_{b,2}^2}{1 + \tau_{B_{b,1}}^2 - (1-p)\eta_1 f_1} \left(\frac{1 - k_c}{k_c} (1-p)\eta_1 f_1 + \tau_{B_{b,1}}^2 \right) \right]. \quad (38)
\end{aligned}$$

or by using (28) we have

$$\begin{aligned}
\left\{ \right\} &= + k_\infty^b - 1 \\
&- \left[\frac{(1 + \tau_{B_{b,1}}^2)(1 + L^2 B_{b,2}^2)}{1 + (1-p)L^2 B_{b,2}^2} - (1 + \tau_{B_{b,1}}^2)(1 + L^2 B_{b,2}^2) \right] \\
&+ \left[\left(\frac{1 - k_c + \tau_{B_{b,1}}^2}{k_c} \right) \left(\frac{1 + L^2 B_{b,2}^2}{1 + \tau_{B_{b,1}}^2 - (1-p)\eta_1 f_1} \right) + (1 + L^2 B_{b,2}^2) \right. \\
&\quad \left. (1 + \tau_{B_{b,1}}^2)(1 + L^2 B_{b,2}^2) \right] \\
&+ \frac{1 + L^2 B_{b,2}^2}{1 + \tau_{B_{b,1}}^2 - (1-p)\eta_1 f_1} \left[\left(\frac{(1-p)\eta_1 f_1}{k_c} - 1 \right) \delta\phi^+ \delta\phi \right. \\
&\quad + \left(\frac{1 - k_c}{k_c} + \frac{\tau_{B_{b,1}}^2}{k_c} \right) \delta\phi^+ \\
&\quad \left. + \left(\frac{1 - k_c}{k_c} (1-p)\eta_1 f_1 + \tau_{B_{b,1}}^2 \right) \delta\phi \right]. \quad (39)
\end{aligned}$$

By making use of

$$\begin{aligned}
& \left(\frac{1 - k_c + \tau_{b,1}^2}{k_c} \right) \left(\frac{1 + L_{b,2}^2}{1 + \tau_{b,1}^2 - (1-p)\eta_1 f_1} \right) - (1 + L_{b,2}^2) \tau_{b,1}^2 \\
&= \frac{1}{k_c} \cdot \frac{1 + L_{b,2}^2}{1 + \tau_{b,1}^2 - (1-p)\eta_1 f_1} \left(1 - k_c + \tau_{b,1}^2 - k_c \tau_{b,1}^2 - k_c^2 \tau_{b,1}^4 + (1-p)\eta_1 f_1 \tau_{b,1}^2 k_c \right) \\
&= \frac{1 + L_{b,2}^2}{1 + \tau_{b,1}^2 - (1-p)\eta_1 f_1} \left(\frac{1-k_c}{k_c} + \frac{1-k_c}{k_c} \tau_{b,1}^2 - \tau_{b,1}^4 + (1-p)\eta_1 f_1 \tau_{b,1}^2 \right) \quad (40)
\end{aligned}$$

we finally obtain the formula

$$\begin{aligned}
k_\infty^b &= 1 - \left(\frac{\Delta \rho_c}{\Delta \rho_p} \right) \left(\frac{V_{p,2}^{\Sigma \phi_{p,2}}}{V_{c,2}^{\Sigma \phi_{c,2}}} \right) \left[1 + \left(\frac{\phi_1^+}{\phi_2^+} \right) \left(\frac{\Sigma_{p,1}^{\phi_{p,1}}}{\Sigma_{p,2}^{\phi_{p,2}}} \right) \right] \\
&\quad - \frac{(1 + \tau_{b,1}^2)(1 + L_{b,2}^2)[(1-p)L_{b,2}^2]}{1 + (1-p)L_{b,2}^2} \\
&\quad + \frac{1 + L_{b,2}^2}{1 + \tau_{b,1}^2 - (1-p)\eta_1 f_1} \left(\tau_{b,1}^4 - (1-p)\eta_1 f_1 \tau_{b,1}^2 - \frac{1-k_c}{k_c} (1 + \tau_{b,1}^2) \right) \\
&\quad - \frac{1 + L_{b,2}^2}{1 + \tau_{b,1}^2 - (1-p)\eta_1 f_1} \left[\left(\frac{(1-p)\eta_1 f_1}{k_c} - 1 \right) \delta \phi^+ \delta \phi \right. \\
&\quad \quad \left. + \left(\frac{1-k_c}{k_c} + \frac{\tau_{b,1}^2}{k_c} \right) \delta \phi^+ \right. \\
&\quad \quad \left. + \left(\frac{1-k_c}{k_c} (1-p)\eta_1 f_1 + \tau_{b,1}^2 \right) \delta \phi \right] \quad (41)
\end{aligned}$$

For $k_c = 1$ and $B_{b,1}^2 = B_{b,2}^2 = B_b^2$ we have

$$\begin{aligned}
 k_{\infty}^b = & 1 - \left(\frac{\Delta \rho_c}{\Delta \rho_p} \right) \left(\frac{V_{p,2} \Sigma_{p,2}^{\overline{p\phi}}}{V_{c,2} \Sigma_{c,2}^{\overline{a\phi}}} \right) \left[1 + \frac{\phi_1^+ \Sigma_{p,1}^{\overline{p\phi}}}{\phi_2^+ \Sigma_{p,2}^{\overline{p\phi}}} \right] \\
 & - \frac{(1 + \tau B_b^2)(1 + L^2 B_b^2)}{1 + (1-p)L^2 B_b^2} \left((1-p)L^2 B_b^2 \right) \\
 & + \frac{(1 + L^2 B_b^2)\tau B_b^2}{1 + \tau B_b^2 - (1-p)\eta_1 f_1} \left(\tau B_b^2 - (1-p)\eta_1 f_1 \right) \\
 & - \frac{1 + L^2 B_b^2}{1 + \tau B_b^2 - (1-p)\eta_1 f_1} \left[\left[(1-p)\eta_1 f_1 - 1 \right] \delta \phi^+ \delta \phi \right. \\
 & \quad \left. + \tau B_b^2 \delta \phi^+ \right. \\
 & \quad \left. + \tau B_b^2 \delta \phi \right]
 \end{aligned} \tag{42}$$

Equation (42) agrees with the new BNWL derivation (Ref. 3) indicating somewhat different flux mismatch terms than previous derivations. It should be noted that k_c , the reactivity of the reactor with the inserted HTGR lattice, is never exactly equal to unity at all temperatures because the reactivity of the unperturbed situation (reactor with cavity) is also assumed to be unity.

3. A Multigroup Formulation

The multigroup diffusion theory equations can be written

$$-\nabla(D_g \nabla \phi_g) + \left(\Sigma_g^a + \sum_{\substack{g'=1 \\ g' \neq g}}^G \Sigma_{g \rightarrow g'}^s \right) \phi_g - \sum_{\substack{g'=1 \\ g' \neq g}}^G \Sigma_{g' \rightarrow g}^s \phi_{g'} = \frac{\chi_g}{k} \sum_{g'=1}^G \nu \Sigma_{g'}^f \phi_{g'}, \quad (43)$$

$$g=1,2,\dots,G.$$

The adjoint equations can be written

$$-\nabla(D_g \nabla \phi_g^+) + \left(\Sigma_g^a + \sum_{\substack{g'=1 \\ g' \neq g}}^G \Sigma_{g \rightarrow g'}^s \right) \phi_g^+ - \sum_{\substack{g'=1 \\ g' \neq g}}^G \Sigma_{g' \rightarrow g}^s \phi_{g'}^+ = \frac{\nu \Sigma_g^f}{k} \sum_{g'=1}^G \chi_{g'} \phi_{g'}^+, \quad (44)$$

$$g=1,2,\dots,G.$$

In operator form these equations can be written as follows:

$$A\phi = \frac{1}{k} B\phi, \quad (45)$$

$$A^+\phi^+ = \frac{1}{k} B^+\phi^+. \quad (46)$$

The perturbed equations, with some material in the cavity V , can be written in the following form:

$$A_c \phi_c = \frac{1}{k_c} B_c \phi_c, \quad (47)$$

$$A_c^+ \phi_c^+ = \frac{1}{k_c} B_c^+ \phi_c^+. \quad (48)$$

If equation (47) is multiplied (inner product) by ϕ^+ and equation (46) by ϕ_c and if these equations are subtracted and integrated over V_o , then

$$\int_{V_o} [\phi^+, A_c \phi_c] - [\phi_c, A^+ \phi^+] dV = \frac{1}{k_c} \int_{V_o} [\phi^+, B_c \phi_c] dV - \frac{1}{k} \int_{V_o} [\phi_c, B^+ \phi^+] dV. \quad (49)$$

Within V_o one has

$$A_c = A \quad \text{and} \quad B_c = B.$$

Equation (49) can be written in the following way:

$$\begin{aligned} & \int_{V_o} \left[\sum_{g=1}^G \phi_g^+ \left\{ -\nabla (D_g \nabla \phi_{c,g}) + \left(\Sigma_g^a + \sum_{\substack{g'=1 \\ g' \neq g}}^G \Sigma_{g \rightarrow g'}^s \right) \phi_{c,g} - \sum_{\substack{g'=1 \\ g' \neq g}}^G \Sigma_{g' \rightarrow g}^s \phi_{c,g'} \right\} \right. \\ & \left. - \sum_{g=1}^G \phi_{c,g} \left\{ -\nabla (D_g \nabla \phi_g^+) + \left(\Sigma_g^a + \sum_{\substack{g'=1 \\ g' \neq g}}^G \Sigma_{g \rightarrow g'}^s \right) \phi_g^+ - \sum_{\substack{g'=1 \\ g' \neq g}}^G \Sigma_{g \rightarrow g'}^s \phi_{g'}^+ \right\} \right] dV \\ & = \frac{1}{k_c} \int_{V_o} \sum_{g=1}^G \phi_g^+ \left\{ \chi_g \sum_{g'=1}^G \nu \Sigma_{g \rightarrow g'}^f \phi_{c,g'} \right\} dV - \frac{1}{k} \int_{V_o} \sum_{g=1}^G \phi_{c,g} \left\{ \nu \Sigma_g^f \sum_{g'=1}^G \chi_{g'} \phi_{g'}^+ \right\} dV. \quad (50) \end{aligned}$$

By using $A_c = A$ and $B_c = B$ in V_o formula (50) can be simplified to

$$\begin{aligned} & - \int_{V_o} \sum_{g=1}^G \left\{ \phi_g^+ (\nabla D_g \nabla \phi_{c,g}) - \phi_{c,g} (\nabla D_g \nabla \phi_g^+) \right\} \\ & = \left(\frac{1}{k_c} - \frac{1}{k} \right) \int_{V_o} \sum_{g=1}^G \sum_{g'=1}^G \phi_g^+ \chi_g \nu \Sigma_{g \rightarrow g'}^f \phi_{c,g'} dV. \quad (51) \end{aligned}$$

The left side of equation (51) can be further simplified by making use of the identity

$$\nabla \cdot (\psi D \nabla \psi) = \nabla \psi \cdot D \nabla \psi + \psi (\nabla D \nabla \psi) \quad (52)$$

and by using Gauss' theorem to transform the volume integral into a surface integral ($\underline{\sigma}$ is the outward directed normal)

$$\begin{aligned} - \int_{S+S_0} \sum_{g=1}^G \left\{ \phi_g^+ D_g \nabla \phi_{c,g} - \phi_{c,g} D_g \nabla \phi_g^+ \right\} d\underline{\sigma} \\ = \left(\frac{1}{k_c} - \frac{1}{k} \right) \int_{V_0} \sum_{g=1}^G \sum_{g'=1}^G \phi_g^+ \chi_g \nabla \Sigma_g^f \phi_{c,g'} dV . \quad (53) \end{aligned}$$

The quantities ϕ_g^+ and $\phi_{c,g}$ are zero on S_0 and $\nabla \phi_g^+$ is zero on S .

This gives

$$\int_S \sum_{g=1}^G \left\{ \phi_g^+ D_g \nabla \phi_{c,g} \right\} d\underline{\sigma} = \Delta \rho_c \int_{V_0} \sum_{g=1}^G \sum_{g'=1}^G \phi_g^+ \chi_g \nabla \Sigma_g^f \phi_{c,g'} dV \quad (54)$$

where

$$\Delta \rho_c = \frac{1}{k} - \frac{1}{k_c} .$$

If we observe that ϕ_g^+ are constant within V , then we can transform equation (11) into the following expression:

$$- \int_V \sum_{g=1}^G \left\{ \phi_g^+ \nabla (D_g \nabla \phi_{c,g}) \right\} dV = \Delta \rho_c \int_{V_0} \sum_{g=1}^G \sum_{g'=1}^G \phi_g^+ \chi_g \nabla \Sigma_g^f \phi_{c,g'} dV . \quad (55)$$

With

$$\int_V \nabla D_g \nabla \phi_{c,g} dV = -D_g B_c^2 \phi_{c,g}^V$$

one obtains

$$\Delta \rho_c \int_V \sum_{g=1}^G \sum_{g'=1}^G \phi_g^+ \chi_{g,g'} \nu \Sigma_{g'}^f \phi_{c,g'} dV = \sum_{g=1}^G \phi_g^+ D_g B_c^2 \phi_{c,g}^V. \quad (56)$$

Another formulation we will prefer here is

$$\begin{aligned} \Delta \rho_c \int_V \sum_{g=1}^G \sum_{g'=1}^G \phi_g^+ \chi_{g,g'} \nu \Sigma_{g'}^f \phi_{c,g'} dV \\ = - \sum_{g=1}^G \phi_g^+ \left\{ \int_V (\Sigma_g^a + \sum_{\substack{g'=1 \\ g' \neq g}}^G \Sigma_{g \rightarrow g'}^s) \phi_{c,g} - \sum_{\substack{g'=1 \\ g' \neq g}}^G \Sigma_{g' \rightarrow g}^s \phi_{c,g'} \right. \\ \left. - \frac{\chi_g}{k_c} \sum_{g'=1}^G \nu \Sigma_{g'}^f \phi_{c,g'} dV \right\}. \end{aligned} \quad (57)$$

For a pure absorber, designated by an index p , one obtains

$$\begin{aligned} \Delta \rho_p \int_V \sum_{g=1}^G \sum_{g'=1}^G \phi_g^+ \chi_{g,g'} \nu \Sigma_{g'}^f \phi_{p,g'} dV \\ = - \sum_{g=1}^G \phi_g^+ \left\{ \int_V \Sigma_g^p \phi_{p,g} dV \right\}. \end{aligned} \quad (58)$$

If we assume that the fluxes $\phi_{c,g}$ and $\phi_{p,g}$ are normalized so that

$$\int_{V_0} \sum_{g=1}^G \sum_{g'=1}^G \phi_g^+ \chi_{gg'} \nu \Sigma_{g'}^f \phi_{c,g'} dV = \int_{V_0} \sum_{g=1}^G \sum_{g'=1}^G \phi_g^+ \chi_{gg'} \nu \Sigma_{g'}^f \phi_{p,g'} dV, \quad (59)$$

then we obtain from (57) and (58) by introducing average fluxes

$$\frac{\Delta \rho_c}{\Delta \rho_p} = \left(\frac{V_c}{V_p} \right) \frac{\sum_{g=1}^G \phi_g^+ \left\{ \left(\Sigma_g^a + \sum_{g' \neq g} \Sigma_{g \rightarrow g'}^s \right) \bar{\phi}_{c,g} - \sum_{g' \neq g} \Sigma_{g' \rightarrow g}^s \bar{\phi}_{c,g'} - \frac{\chi_g}{k_c} \sum_{g'=1}^G \nu \Sigma_{g'}^f \bar{\phi}_{c,g'} \right\}}{\sum_{g=1}^G \phi_g^+ \left\{ \Sigma_g^p \bar{\phi}_{p,g} \right\}} \quad (60)$$

If a buckling is used instead, one obtains

$$\frac{\Delta \rho_c}{\Delta \rho_p} = - \left(\frac{V_c}{V_p} \right) \frac{\sum_{g=1}^G \phi_g^+ D_{g,c} B_c^2 \phi_{c,g}}{\sum_{g=1}^G \phi_g^+ \Sigma_g^p \phi_{p,g}} \quad (61)$$

With the definition

$$k_{\infty}^c = \frac{\sum_{g=1}^G \nu \Sigma_g^f \bar{\phi}_{c,g}}{\sum_{g=1}^G \Sigma_g^a \bar{\phi}_{c,g}} \quad (62)$$

the equation (60) can be written in the following form:

$$\frac{\Delta \rho_c}{\Delta \rho_p} = \left(\frac{v_c}{v_p} \right) \frac{\sum_{g=1}^G \phi_g^+ \left\{ (\Sigma_g^a + \sum_{g' \neq g} \Sigma_{g \rightarrow g'}^s) \bar{\phi}_{c,g} - \sum_{g' \neq g} \Sigma_{g' \rightarrow g}^s \bar{\phi}_{c,g'} - k_\infty^c \frac{\chi_g}{k_c} \sum_{g'=1}^G \Sigma_{g'}^a \bar{\phi}_{c,g'} \right\}}{\sum_{g=1}^G \phi_g^+ \Sigma_g^p \bar{\phi}_{p,g}} \quad (63)$$

$$k_\infty^c = \frac{\sum_{g=1}^G \phi_g^+ \Sigma_g^a \bar{\phi}_{c,g} - \sum_{g=1}^G \phi_g^+ \sum_{g' \neq g} \left\{ \Sigma_{g \rightarrow g'}^s \bar{\phi}_{c,g} - \Sigma_{g' \rightarrow g}^s \bar{\phi}_{c,g'} \right\}}{\frac{1}{k_c} \sum_{g=1}^G \phi_g^+ \chi_g \sum_{g=1}^G \Sigma_g^a \bar{\phi}_{c,g}} - \frac{\sum_{g=1}^G \phi_g^+ \Sigma_g^p \bar{\phi}_{p,g}}{\frac{1}{k_c} \sum_{g=1}^G \phi_g^+ \chi_g \sum_{g=1}^G \Sigma_g^a \bar{\phi}_{c,g}} \left(\frac{v_p}{v_c} \right) \frac{\Delta \rho_c}{\Delta \rho_p} \quad (64)$$

$$k_\infty^c = k_c \left[\frac{\sum_{g=1}^G \phi_g^+ \Sigma_g^a \bar{\phi}_{c,g}}{\left(\sum_{g=1}^G \phi_g^+ \chi_g \right) \left(\sum_{g=1}^G \Sigma_g^a \bar{\phi}_{c,g} \right)} - \frac{\sum_{g=1}^G \sum_{g'=1}^G \left(\Sigma_{g \rightarrow g'}^s \bar{\phi}_{c,g} \right) \left(\phi_{g'}^+ - \phi_g^+ \right)}{\left(\sum_{g=1}^G \phi_g^+ \chi_g \right) \left(\sum_{g=1}^G \Sigma_g^a \bar{\phi}_{c,g} \right)} - \frac{\sum_{g=1}^G \phi_g^+ \Sigma_g^p \bar{\phi}_{p,g}}{\left(\sum_{g=1}^G \phi_g^+ \chi_g \right) \left(\sum_{g=1}^G \Sigma_g^a \bar{\phi}_{c,g} \right)} \left(\frac{v_p}{v_c} \right) \frac{\Delta \rho_c}{\Delta \rho_p} \right] \quad (65)$$

Discussion

Equation (20) [or the equivalent multigroup Eq. (65)] is recommended for the analysis of the experiments. These formulas avoid the introduction of a somewhat artificial bare critical system, and they do not require flux mismatch correction terms.

The old formulas rederived in Section 2 were generalized to the case of different fast- and thermal-energy-group bucklings. The resulting formulas agree with the newly derived formulas by E. Lippencott of BNWL, with one small difference: Lippencott assumes that k_c , the reactivity of the reactor with the inserted HTGR lattice, is equal to one, which is not exactly the case, because the reactivity of the unperturbed situation (reactor with cavity) is also assumed to be one.

The reasons for recommending Eq. (20) [or Eq. (65)] instead of the formulas of Section 2 are given in the following discussion.

Equations (16), (17), and (20) are exact, assuming that the unperturbed and the perturbed reactor can be described by the two-group Eqs. (3) through (6). Especially, it is assumed that the two-group cross sections in V_0 are not affected by the perturbations in V . It is interesting to note that these equations contain only quantities related to the unperturbed and perturbed reactor configurations; none of the quantities is related to a bare critical assembly.

Equation (23), which follows directly from Eq. (17), also contains only quantities related to the unperturbed and perturbed reactor configurations. If several quantities in Eq. (23), e.g., $\Delta\rho_c/\Delta\rho_p$, are measured, then Eq. (23) is no longer exactly fulfilled, and this discrepancy is exactly the error between experiment and theory (including cross sections, two group model, etc.).

For the derivation of Eq. (41), a bare critical assembly is introduced. It is assumed that the bare critical assembly can be described by two-group

equations with the same cross sections that apply to the lattice introduced into region V. This is an additional assumption, because the cross sections are dependent on the buckling. Now the derivation of Eq. (41) is straightforward: the quantity

$$k_{\infty}^b = \frac{(1 + \tau B_{b,1}^2)(1 + L^2 B_{b,2}^2)}{1 + (1-p) L^2 B_{b,2}^2} \quad (66)$$

is added to and subtracted from the right side of Eq. (23), the bucklings $B_{c,1}^2$ and $B_{c,2}^2$ are expressed by the identities of Eq. (31), flux mismatch terms are introduced, and after some algebraic manipulations Eq. (41) is obtained. This consideration shows that there is no chance to find a k_{∞}^b that fulfills Eq. (41) and Eq. 66 simultaneously, if Eq. (23) was not exactly fulfilled in the first place.

Another consideration is that the cross sections to be used in Eq. (41) should be evaluated using the bucklings $B_{c,1}^2$ and $B_{c,2}^2$ introduced in Eq. (17) and defined in Eq. (21). Thus, even though one might argue that the introduction of a bare critical assembly would allow the calculations to proceed without performing detailed calculations for the complete HTLTR assembly, it is apparent that such calculations will be required to evaluate $B_{c,1}^2$ and $B_{c,2}^2$ for the test region as well as the temperature dependence of the fast to thermal flux ratios.

In summary one should use Eqs. (41) or (42) cross sections evaluated for $B_c^2 (\approx B_{c,1}^2 \approx B_{c,2}^2)$ and a B_b^2 corresponding to a bare critical assembly for a two-group model with the cross sections of the lattice that was introduced into the test region. It should be noted that all of the graphite end caps, etc., that were actually inserted should be included in the homogenized two-group cross sections used for the hypothetical bare critical assembly calculation.

References

1. Heineman, R.E., "Reactivity Measurements on Samples of Multiplying Media," USAEC Report BNWL-SA-263, Battelle-Northwest, 1965.
2. Lippincott, E. P., C. R. Richey and D. D. Lanning, "Reactor Physics Quarterly Report, July, August, September, 1969," USAEC Report BNWL-1240, p. 2.3, Battelle-Northwest, 1969.
3. Lippincott, E. P., private communication, September 1970.

TASK IX
FUEL MATERIALS DEVELOPMENT

COATED PARTICLE IRRADIATIONS

A series of capsule irradiations is being conducted to evaluate both startup and recycle fuel systems for a large HTGR under the expected irradiation environments. These tests are directed toward the evaluation of fuel particles that contain only uranium (diluent particles) and toward evaluation of coated oxide fuel. Additional emphasis has been placed on irradiation testing at temperatures above the maximum in a large HTGR (1350°C) to better define long-term behavior and for safety analysis applications.

Capsule P13L

Capsule P13L was discharged from the ETR after completing its scheduled seven cycles of irradiation. It is currently undergoing postirradiation examination in the Gulf General Atomic hot-cell facility. Capsule P13L was the second test of coated diluent particles. Both carbide and oxide diluent and fertile particles were being tested, with both BISO and TRISO coatings on the diluent particles and BISO coatings on the fertile particles. (Samples of reference TRISO coated fissile and fertile carbide particles are included for comparison.) All diluent samples were of the buffer-diluted design. Some of the samples were designed to evaluate the effect of higher fuel loadings in coated particles (i.e., thinner coatings). Details of the sample selection were given in earlier quarterly reports (see GA-9227 and GA-9815).

Dosimeter wire analyses yielded the following fast neutron fluences ($E > 0.18$ MeV) for the capsule:

Cell 1	7.1 x 10 ²¹	n/cm ²
Cell 2	7.8 x 10 ²¹	n/cm ²
Cell 3	7.8 x 10 ²¹	n/cm ²
Cell 4	7.2 x 10 ²¹	n/cm ²
Cell 5	5.8 x 10 ²¹	n/cm ²

Metallographic examination of the fuel particles is currently being performed; the initial results from this and the macroscopic examination are given below and are summarized in Table 9.1:

1. All fissile TRISO-LTI samples appeared from the macroscopic examination to have survived irradiation at 1300° and 1500°C with the exception of one high BAF sample. The fertile-B TRISO-LTI sample had 16% failure of the O-PyC layer.
2. The majority of the fissile BISO carbide samples that were irradiated at 1300° and 1500°C had particles bonded to the graphite thimbles. The BISO oxide particles did not show this behavior.
3. Metallographic examination of the two fissile BISO carbide samples irradiated at 1500°C showed almost total failure of the isotropic layer. Nearly all the particles from these two batches were bonded together, and it appeared that metallic fission products had diffused from the kernel and accumulated on the particle and thimble wall surfaces, which presumably caused the bonding. Also, there was considerable erosion of the outer surface of the isotropic coating, and this erosion is inferred to have resulted from reactions with the fission products.
4. The fertile BISO carbide survived while the fertile BISO oxide failed.
5. All TRISO and BISO samples irradiated at 900°C survived except the UC₂ BISO-HTI sample.

TABLE 9.1
SUMMARY DESCRIPTION OF THE COATED PARTICLE SAMPLES TESTED IN CAPSULE P13L

Batch No.	Coated Particle (a)		Coating										Fuel Kernel	Irradiation Conditions				Postirradiation Examination			
			Buffer		Inner Isotropic (LTI)		SiC		Outer Isotropic		Total Coating Thickness (μm)	Type		Size (μm)	Burnup (% FIMA)	Temp (°C)	Fast Dose (10 ²¹ n/cm ²)	Failure (%)			
																		Macro		Micro	
	Type	PyC	Thickness (μm)	Density (g/cm ³)	Thickness (μm)	Density (g/cm ³)	Thickness (μm)	Density (g/cm ³)	Thickness (μm)	Density (g/cm ³)	BAF	OPyC	SiC	OPyC	SiC						
3516-47E	BISO	HTI	62	1.23	--	--	--	--	48	1.82	1.00	110	UC ₂	106	75	1500	7.1	(d)	--	98	--
4403-113E	BISO	LTI	56	1.27	--	--	--	--	45	1.88	1.04	101	UC ₂	106	75	1500	7.1	(d)	--	85	--
4413-5E	TRISO	LTI	53	1.30	24	1.83	20	3.20	23	1.80	1.08	120	UC ₂	106	75	1500	7.1	0.1	0		
4413-67E	TRISO	LTI	46	1.26	22	1.85	20	3.16	22	1.75	1.01	110	UC ₂	124	75	1500	7.1	0.2	0.2		
4263-75E	TRISO	LTI	48	1.23	22	1.87	19	3.18	30	1.83	1.00	119	(Th,U)C ₂ (c)	200	26	1500	7.1	0	0		
4413-35E	BISO	LTI	57	1.23	--	--	--	--	57	1.85	1.00	114	UO ₂	124	75	1500	7.1	6	--	--	
3516-39E	TRISO	HTI	55	1.34	16	1.83	20	3.20	19	1.86	1.09	110	UC ₂	106	75	1300	7.8	88	4		
4403-113E	BISO	LTI	56	1.27	--	--	--	--	45	1.88	1.04	101	UC ₂	106	75	1300	7.8	30	--		
4403-143E	TRISO	LTI	53	1.30	24	1.83	20	3/20	26	1.90	1.05	123	UC ₂	106	75	1300	7.8	0.1	0.1		
3516-35E	TRISO	HTI	46	1.29	14	1.87	16	3.20	20	1.73	1.00	96	UC ₂	106	75	1300	7.8	2.5	2.2		
4413-7E	TRISO	LTI	46	1.29	14	1.87	16	3.20	22	1.85	1.12	98	UC ₂	106	75	1300	7.8	4.0	3.7		
4413-21E	TRISO	LTI	44	1.22	21	1.78	21	3.17	19	1.91	1.00	105	UC ₂	106	75	1300	7.8	0.3	0.2		
3516-33E	TRISO	HTI	49	1.33	17	1.91	20	3.20	22	1.72	1.00	108	UC ₂	106	75	1300	7.8	1.3	1.0		
4403-81E	BISO	LTI	56	1.17	--	--	--	--	52	1.83	1.06	108	UC ₂	106	75	1300	7.8	0.2	--		
4413-5E	TRISO	LTI	53	1.30	24	1.83	20	3.20	23	1.80	1.08	120	UC ₂	106	75	1300	7.8	0.1	0.1		
4413-35E	BISO	LTI	57	1.23	--	--	--	--	57	1.85	1.00	114	UO ₂	124	75	1300	7.8	5.5	--		
4413-67E	TRISO	LTI	46	1.26	22	1.85	20	3.16	22	1.75	1.01	110	UO ₂	124	75	1300	7.8	0.6	0.6		
3516-47E	BISO	HTI	62	1.23	--	--	--	--	48	1.82	1.00	110	UC ₂	106	75	1300	7.8	1.2	--		
4403-145E	BISO	LTI	57	1.29	--	--	--	--	20	1.90	1.11	77	UC ₂	106	75	1300	7.2	(d)	--		
3516-57E	BISO	HTI	59	1.24	--	--	--	--	21	1.82	0.96	80	UC ₂	106	75	1300	7.2	(d)	--		
4413-29E	BISO	LTI	59	1.24	--	--	--	--	24	1.82	1.00	83	UC ₂	106	75	1300	7.2	(d)	--		
4423-67	TRISO	LTI	59	1.22	20	(b)	21	3.22	22	1.90	1.02	122	ThC ₂	530	7	800	7.2	1.6	4		
4423-89	BISO	LTI	64	--	--	--	--	--	89	1.72	0.97	153	ThC ₂	530	7	800	7.2	0	--		
4413-75	BISO	LTI	69	--	--	--	--	--	85	1.89	1.02	154	ThO ₂	576	7	800	7.2	14	--		
3516-47E	BISO	HTI	62	1.23	--	--	--	--	48	1.82	1.00	110	UC ₂	106	60	900	5.8	2.9	--		
4403-113E	BISO	LTI	56	1.27	--	--	--	--	45	1.88	1.04	101	UC ₂	106	60	900	5.8	0.1	--		
4413-5E	TRISO	LTI	53	1.30	24	1.83	20	3.20	23	1.80	1.08	120	UC ₂	106	60	900	5.8	0	0		
4263-75E	TRISO	LTI	48	1.23	22	1.87	19	3.18	30	1.83	1.00	119	(Th,U)C ₂ (c)	200	20	900	5.8	0.2	0		
4413-67E	TRISO	LTI	46	1.26	22	1.85	20	3.16	22	1.75	1.01	110	UO ₂	124	60	900	5.8	0.1	0.1		
4413-35E	BISO	LTI	57	1.23	--	--	--	--	57	1.85	1.00	114	UO ₂	124	60	900	5.8	1	--		

- (a) BISO denotes a coating sequence of buffer and LTI or HTI isotropic PyC. TRISO denotes the coating sequence of buffer, inner isotropic (LTI), SiC, and outer isotropic LTI or HTI PyC.
- (b) This batch was prepared in production coaters out to, and including, the SiC layer; inner isotropic density was not reported.
- (c) Ratio of thorium to uranium was 2.25.
- (d) No failure estimate: particles remained stuck in graphite container.
- (e) Some particle sticking observed.

Capsule P13M

Capsule P13M will be the second full-exposure test of candidate 1100-MW(e) HTGR initial and recycle core fuel and the first test of this type of fuel made in production equipment. Many of the samples will be tested in pitch-bonded fuel rods to properly evaluate the full range of interactive stresses. Some of the samples will be duplicates of those to be tested in the Phase II Peach Bottom test elements (see Task VIII), and the P13M capsule will provide full-exposure data on these important materials. (The maximum exposure received in the Phase II test elements will be approximately 3×10^{21} n/cm².)

The P13M capsule will contain two fuel rod cells, each holding six rods, which will operate at 1000°C to 8×10^{21} n/cm² and 1350°C to 4 to 5×10^{21} n/cm², respectively. There will also be one cell containing coated particle thimble samples that will operate at 1350°C to 6 to 7×10^{21} n/cm².

In addition, due to the importance of obtaining thermocouple decalibration data to apply to the operation of subsequent capsules and to the analysis of the completed experiments, P13M will contain two thermocouple decalibration cells. One cell will operate at 950°C to about 8×10^{21} n/cm², and the other will be at 1250°C to about 6×10^{21} n/cm². The only decalibration data available to date are at 950°C to 2.8×10^{21} n/cm² from the BG-2 capsule. The results are presented in a report that is being prepared for publication (see Ref. 1).

Work on capsule P13M is progressing satisfactorily, and thermal analysis data obtained from previous capsules are being applied to the thermal design of this one. Problems have been encountered in developing a satisfactory design for the 1350°C thermocouple decalibration cell, but efforts are being continued. The fuel rod samples to be irradiated have been selected, and the coated particles required have been ordered.

The capsule is presently scheduled for insertion in the ETR (I13 core position) in cycle 110B (January 1971). Investigations of higher flux ETR

core positions are under way, but it appears that several months would be required for characterizing these new positions to the point of being able to design a capsule that would operate successfully. The P13M capsule will therefore be inserted in the present I13 position to avoid delay.

References

1. Carpenter, F.D., N. L. Sandefur, R. J. Grenda, and J. S. Steibel, "EMF Stability of Chromel-Alumel and Tungsten-3% Rhenium versus Tungsten-25% Rhenium Sheathed Thermocouples in a Neutron Environment," USAEC Report GA-10384, Gulf General Atomic, to be published.

TASK XI
GRAPHITE RESEARCH

CONTROL MATERIALS IRRADIATIONS

Capsule BG-2

The primary purpose of the BG-2 experiment (GA-19-2) is to measure the effects of neutron irradiation on boronated (B or B_4C plus graphite) and hafnated (HfC plus graphite) compacts. Dimensional changes in boron fibers and density changes in loose granules of B_4C and HfC were also determined in piggyback specimens to enhance the knowledge of irradiation effects in these poison materials in the absence of a bonding matrix material. A better understanding of irradiation effects in boronated and hafnated graphite resulted from this investigation.

Irradiation-induced dimensional change was the phenomenon of primary interest; however, a correlation of thermal expansion coefficient, strength, and microstructure with neutron exposure was planned. Some specimens irradiated previously in capsule BG-1 (reported in earlier quarterly report GA-9372) continued irradiation exposure in BG-2. A detailed summary of samples being tested in BG-2 was given in earlier quarterly report GA-9660. The design of BG-2 was basically the same as BG-1 as described in Ref. 1.

The capsule was inserted in the ETR and started irradiation in July 1969 (cycle 103) at temperatures of 600° to 700°C. The capsule remained in the ETR through cycle 106 and was removed in May 1970. The capsule was opened at the GGA hot-cell facility and initial observations were reported in the previous quarterly report (GA-10299). During this report period the post-irradiation examination of specimens has continued.

Postirradiation Examination of Specimens

Specimens of boronated and hafnated graphite were examined to determine changes in dimensions, crush strength, microstructure, and coefficient of thermal expansion. A summary of the irradiation conditions and material properties including results of previous irradiation (capsule BG-1) is shown in Table 11.1.

BORONATED GRAPHITE

Microstructure

Metallographic examination of boronated graphite specimens has not been completed. Preliminary results show microstructure features which were also observed in BG-1 specimens and reported in (Ref. 1). The B^{10} burnup gradient which should exist in the specimens is not evident in the microstructure. Some cracking of B_4C granules appears to occur during irradiation, but it was as prevalent on the side exposed to thermal neutrons as on the shielded side. An example of boronated graphite microstructure before and after irradiation is shown in Fig. 11.1. A high magnification view of a cracked B_4C granule is shown in Fig. 11.2. The cracking and swelling of B_4C granules during irradiation probably contributes to the net dimensional change observed in irradiated boronated graphites.

Dimensional Change

The extruded boronated graphite (3912-34) contracted slightly in the parallel direction and expanded perpendicular to the extrusion direction as shown in Fig. 11.3. The change in dimension was linear with fast-neutron exposure when only specimens irradiated in both BG-1 and BG-2 are considered. The perpendicular expansion of the specimens irradiated only in BG-2 was significantly less than the specimens continuing on from BG-1 at the same fast-neutron exposure. The explanation of this may be related to differences in B^{10} burnup gradients, which are yet to be determined. However, these specimens were tightly held in the graphite crucibles at the end of irradiation,

M29982-6

(a)

(75x)

M367-142

(b)

(75x)

Fig. 11.1. Microstructure of boronated graphite (B_4C in graphite), material 4465-42; (a) before irradiation, (b) after irradiation to 4.80 to 5.87×10^{21} n/cm^2 ($E > 0.18$ MeV) at $700 \pm 50^\circ C$ in thermal-neutron window of capsule BG-2. Note wide variation in B_4C microstructure before irradiation; after irradiation some B_4C granules were fractured

M367-143

(500x)

Fig. 11.2. Fracture pattern typically observed in B_4C granules of boronated graphite at high B-10 burnup; material 4465-42; fast-neutron exposure 4.80 to 5.87×10^{21} n/cm² ($E > 0.18$ MeV) at $700 \pm 50^\circ\text{C}$ in capsule BG-2

M367-143

(500x)

1. Micrograph pattern showing the effect of 1000 rads of beta radiation on the structure of the material. The material is a mixture of boron and graphite at 1000 rads; water at 400-42; fast-neutron exposure 4.81 to 5.87 x 10²¹ n/cm² (0.1 MeV) at 700-50 in capsule BG-2.

Fig. 11.3. Irradiation-induced dimensional change in extruded boronated graphite baked at 2200°C and irradiated at 700° ± 50°C

so the possibility of some restraint on the specimens by the crucible cannot be ruled out. This observation is receiving further study.

The warm-pressed boronated graphites with a 2000°C bake temperature changed dimensions as shown in Fig. 11.4. Expansion in both the parallel and perpendicular direction was observed. The boronated graphite sample from a FSV HTGR absorber (4465-42) was the most isotropic of materials tested and showed less parallel expansion but greater perpendicular expansion than material made in the laboratory. Warm pressed nonboronated graphite made from graphite fillers and pitch binder similar to those used in the boronated graphites showed less irradiation-induced parallel expansion and more perpendicular contraction as shown in Fig. 11.5. Not all of the difference in irradiation performance of nonboronated and boronated graphite can be ascribed to the B_4C content. Some of the difference in performance was due to differences in the degree of preferred orientation (anisotropy) of graphite crystallites as revealed by differences in the coefficient of thermal expansion in the two principle directions. The anisotropy in the nonboronated graphite was greater than in the boronated graphites as shown in Table 11.1.

Bake temperature affects the irradiation-induced dimensional change of boronated graphite as shown in Fig. 11.6. A final bake temperature of 2000°C resulted in significantly less expansion in both directions than observed in boronated graphite baked at 1000°C.

The effect of B^{10} content in graphite on irradiation-induced dimensional change was explored by irradiating graphites containing B_4C or boron powder made from B^{10} depleted, and B^{10} enriched boron powders (see Table 11.1). With the B^{10} -depleted B_4C , the total boron content of boronated graphite was kept about the same as in boronated specimens containing natural boron. The dimensional changes which took place upon irradiation are shown in Fig. 11.7. The boronated graphite containing B^{10} depleted B_4C underwent much less expansion in both directions than did graphite containing natural boron. In fact, the parallel direction dimensional change was similar for nonboronated and the B^{10} -depleted boronated graphite. In the perpendicular direction the

Fig. 11.4. Irradiation-induced dimensional change in warm-pressed boronated graphite baked at 2000°C and irradiated at $700^\circ \pm 50^\circ\text{C}$

Fig. 11.5. Irradiation-induced dimensional change in warm-pressed boronated and nonboronated graphite baked 2 hr at 2000°C and irradiated at 700° ± 50°C; dashed lines are drawn through laboratory-produced boronated graphite shown in Fig. 11.4

Fig. 11.6. Effect of bake temperature on irradiation-induced dimensional change of boronated graphite; data points for material with 1000°C bake show consistently more expansion than that baked at 2000°C (shown in Fig. 11.4 and shown here by dashed line)

Fig. 11.7. Irradiation-induced dimensional change in warm-pressed B-10-depleted, nonboronated, and natural boronated graphite baked at 1000°C; irradiated at 550° to 750°C (graphitized coke filler C₀ 6.694 to 6.750 Å, L_c 730 to 2000 Å)

TABLE 11.1
SUMMARY OF MATERIAL PROPERTIES AND IRRADIATION EFFECTS IN BORONATED GRAPHITE
IN THE BG-2 EXPERIMENT INCLUDING PRIOR IRRADIATION IN THE BG-1 EXPERIMENT

	Extruded and Baked						Warm Pressed and Baked																		
	Peach Bottom HTGR Control Rod Absorber						Non-Boronated Graphite Control Samples						Unleached Commercial B ₄ C in Graphite		Leached Commercial B ₄ C in Graphite				B-10 Depleted Boron Laboratory B ₄ C in Graphite		B-10 Enriched Boron in Graphite (91% B-10 Enriched)	B-10 Enriched Boron Laboratory B ₄ C (91% Enriched)	Fort St. Vrain HTGR Control Rod Absorber (Commercial)	Hafnated Graphite	
																								Low Temp Bake	High Temp Bake
Material No.	3912-34						3685-118-1000		4465-21-2000		3685-111-1000		3685-124-1000		3685-124-2000		3685-120-1000		4465-24	4465-26	4465-42	4065-132-2000		4065-132-2600	
Bake Temperature (2 hours), °C	2200 (30 min)						1000		2000		1000		2000		2000		1000		2000	2000	2000	2000		2600	
Boron Content, wt-%	28						0		0		22		34		34		22 ^(e)		7.1	7.5	43	0		0	
Boron Density, g B/cm ³	0.45						0		0		0.38		0.52		0.52		0.38		0.11	0.14	0.69	0		0	
Hafnium Content (wt-%)	0						0		0		0		0		0		0		0	0	0	90		90	
Hafnium Density (gm Hf/cm ³)	0						0		0		0		0		0		0		0	0	0	24		5.19	
Bulk Density, g/cm ³	1.60						1.72		1.98		1.77		1.68		1.68		1.72		1.70	1.91	1.60	5.81		5.76	
Irradiation Experiment	BG-1	BG-2	BG-2	BG-1	BG-2	BG-2	BG-1	BG-2	BG-1	BG-2	BG-1	BG-2	BG-1	BG-2	BG-2	BG-2	BG-2	BG-2	BG-2	BG-2	BG-2	BG-2	BG-2		
No of Specimens:																									
Prismatic 0.2 x 0.2 x 0.58 in.	0	0	0	6	0	0	8	4	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0		
Prismatic 0.2 x 0.4 x 0.58 in.	3	3	0	3	2	0	4	0	4	2	4	3	4	2	0	0	0	0	0	0	2	0	3		
Cylindrical 9.4 DIA x 0.58 in.	0	0	2	0	0	2	0	0	0	0	0	0	0	0	3	3	6	6	6	6	6	6	6		
Total Fast Fluence (E>0.18 Mev), 10 ²¹ n/cm ²	2.0-2.5	5.6-6.1	6.0	3.5-4.0	8.25	5.5	2.5-3.5	7.5	2.5-3.0	7.3	3.0-3.5	7.0-7.3	3.5-4.0	8.8	4.36-4.66	3.76-4.19	4.80-5.87	6.0	5.5-6.0	6.0	5.5-6.0	6.0	5.5-6.0		
Total Thermal Fluence (E<0.4 ev), 10 ²¹ n/cm ²	1.5	3.5	3.0	2.5	4.5	2.8	1.5	3.7	1.5	3.5	1.5	3.5	2.5	4.8	2.0	1.8	2.0-2.7	3.0	2.7-3.0	3.0	2.7-3.0	3.0	2.7-3.0		
Specimen Irradiation Temp, °C:																									
Minimum	600(a)			550(a)			625		675		675(a)		675(a)		675(a)		725	725	725	725	725	725	725		
Maximum	675(a)	675	790	600(a)	725	725	750	700	850	725	850(a)	725	700(a)	625	725	725	730	700	700	700	700	700	700		
Average	650(a)	650	700	550(a)	700	700	675	675	750	700	750(a)	700	675(a)	575	700	700	700	700	700	700	700	700	700		
B-10 Isotope Burnup, % Depletion																									
At Neutron Window	80	(i)	(i)	0	(b)	(b)	80	(i)	66	(i)	76	(i)	(f)	(b)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Mid Specimen	(b)	(i)	(i)	0	(b)	(b)	30	(i)	35(d)	(i)	35(d)	(i)	(f)	(b)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Opposite Neutron Window	37	(i)	(i)	0	(b)	(b)	53	(i)	38	(i)	48	(i)	(f)	(b)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Dimension Change During Irrad. Relative to Forming Direction, %:																									
Parallel Minimum	-0.52	-0.51	-0.21	+1.23	+4.14	+1.76	+2.35	+5.21	+1.76	+5.34	+0.31	+4.10	+1.05	+4.29	+9.24	+6.08	+1.72	+1.41	+0.87	+1.41	+0.87	+1.41	+0.87		
Parallel Maximum	-0.03	-0.35	-0.52	+1.73	+4.30	+1.93	+2.81	+7.57	+2.67	+6.10	+2.23	+4.65	+1.87	+4.63	+13.61	+6.42	+2.17	+1.69	+1.29	+1.69	+1.29	+1.69	+1.29		
Parallel Average	-0.23	-0.05	-0.37	+1.40	+4.22	+1.84	+2.51	+6.65	+2.20	+5.72	+1.26	+4.29	+1.42	+4.46	+11.32	+6.22	+1.88	+1.55	+1.09	+1.55	+1.09	+1.55	+1.09		
Perpendicular Minimum	-0.13	+1.43	+0.62	-1.21	-1.34	-2.78	-0.58	+1.53	-0.43	(h)	-1.09	+0.418	-0.68	+0.255	-1.50	-0.13	+0.37	+0.25	+0.25	+0.25	+0.25	+0.25	+0.25		
Perpendicular Maximum	+0.86	+1.76	+0.62	-0.41	-1.67	-2.78	+0.91	+3.04	+1.54	(h)	+0.91	+0.585	+0.31	+0.587	-2.25	-1.01	+1.00	+0.25	+0.37	+0.25	+0.37	+0.25	+0.37		
Perpendicular Average	+0.69	+1.57	+0.62	-0.80	-1.50	-2.78	+0.24	+2.41	+0.44	+1.60	0.00	+0.502	-0.16	+0.421	-1.88	-0.543	+0.68	+0.25	+0.29	+0.25	+0.29	+0.25	+0.29		
Graphite Matrix Crystallite Dimension, Å:																									
Preirradiation L _c	580	(i)	(i)	850	(i)	(i)	490	(i)	675	(i)	580	(i)	850	(i)	(i)	(i)	540	(i)	(i)	(i)	(i)	(i)	(i)		
Preirradiation C ₀	6.729	(i)	(i)	6.715	(i)	(i)	6.719	(i)	6.715	(i)	6.726	(i)	6.715	(i)	(i)	(i)	6.730	(i)	(i)	(i)	(i)	(i)	(i)		
Postirradiation L _c	103	(i)	(i)	195	(i)	(i)	200	(i)	210	(i)	111	(i)	170	(i)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Postirradiation C ₀	6.820	(i)	(i)	6.775	(i)	(i)	6.736	(i)	6.729	(i)	6.758	(i)	6.742	(i)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Coef. of Thermal Exp. Relative to Forming Direction (22° to 700°C), 10 ⁻⁶ in./in. °C:																									
Preirradiation Parallel (α _{pa})	3.68	3.68	3.68	9.37	9.37	8.01	5.57	5.57	6.32	6.32	5.30	5.30	6.80	6.80	8.84	7.76	4.83	5.19	5.09	5.19	5.09	5.19	5.09		
Preirradiation Perpendicular (α _{pe})	4.70	4.70	4.70	4.19	4.19	4.19	4.29	4.29	4.70	4.70	3.82	3.82	4.26	4.26	2.11	2.23	4.58	4.01	4.52	4.01	4.52	4.01	4.52		
Ratio (α _{pa} /α _{pe})	1.3(c)	1.3(c)	1.3(c)	2.2	2.2	3.5	1.3	1.3	1.3	1.3	1.4	1.4	1.6	1.6	4.2	3.5	1.1	1.3	1.1	1.3	1.1	1.3	1.1		
Postirradiation Parallel (α _{pa})	(b)	5.04	5.34	10.41	12.32	12.41	8.17	8.54	(b)	7.14	(b)	9.01	(b)	8.98	12.49	10.02	6.78	(b)	(b)	(b)	(b)	(b)	(b)		
Postirradiation Perpendicular (α _{pe})	(b)	6.62	5.70	(b)	5.87	2.87	5.25	3.76	(b)	4.60	(b)	4.84	(b)	5.71	3.93	3.09	6.56	(b)	(b)	(b)	(b)	(b)	(b)		
Ratio (α _{pa} /α _{pe})	(b)	1.3(c)	1.1(c)	(b)	2.1	4.3	1.6	2.3	(b)	1.5	(b)	1.9	(b)	1.6	3.2	3.2	1.0	(b)	(b)	(b)	(b)	(b)	(b)		
Thermal Conductivity Perpendicular to Forming Direction, cal/cm-sec-°C:																									
Preirradiation 25°C	0.120	0.120	0.120	0.170	0.170	(i)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Preirradiation 400°C	0.090	0.090	0.090	0.110	0.110	(i)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Preirradiation 800°C	0.065	0.065	0.065	0.090	0.090	(i)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Postirradiation 25°C	(b)	(i)	(i)	0.027	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Postirradiation 400°C	(b)	(i)	(i)	0.033	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Postirradiation 800°C	(b)	(i)	(i)	0.035	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(i)	(i)	(i)	(b)	(b)	(b)	(b)	(b)	(b)		
Room Temperature Crush Strength, Loaded Parallel to Forming Direction, psi:																									
Preirradiation	(b)	(b)	5500	(b)	(b)	5400	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	4050	7050	6000	3780	4780	3780	4780	3780	4780		
Postirradiation	(b)	(b)	5120	(b)	(b)	8050	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(b)	2630	4225	3807	(i)	(i)	(i)	(i)	(i)	(i)		

(a) Estimated temperatures based on indicated temperatures of adjacent specimens.
(b) Not measured.
(c) Ratio (α_{pa}/α_{pe}) for extruded specimens.
(d) Estimated B-10 burnup based on measured burnup of adjacent specimens.
(e) Boron depleted in B-10 (1.45 at % B-10; remainder B-11).

(f) Burnup of B-10 not detectable.
(g) An 11 hour period was spent at ~1175°C because of temperature control error.
(h) Only one specimen suitable for + measurement.
(i) Measurement in progress.

B^{10} -depleted ($B^{10}/B^{11} = 0.015$) graphite showed slight swelling, whereas the nonboronated graphite underwent shrinkage of 1.5% at 8.25×10^{21} n/cm² ($E > 0.18$ Mev). Therefore, it appears that the incorporation of B_4C granules into graphite results in increased volume expansion during irradiation. However, the volume expansion is reduced if the B_4C is relatively inert by virtue of being depleted in the B^{10} isotope.

Another series of graphites contained B^{10} -enriched ($B^{10}/B^{11} = 9.2$) boron either as boron powder or as B_4C made from the B^{10} -enriched boron. These were baked at 2000°C and irradiated in BG-2. They showed a marked tendency to bow. The bow was caused by the accelerated axial expansion of that portion of the specimen exposed to thermal neutrons. For example the specimens made with B^{10} powder (see 4465-24, Table 11.1) expanded axially about 13% on the surface in the thermal-neutron window and 9% on the opposite shielded side. This effect was present but less pronounced in the specimens made with B^{10} -enriched B_4C . They expanded 7% on the surface exposed in the thermal-neutron window and 5% on the shielded side. A photograph of one specimen showing the bowing effect is shown in Fig. 11.8. The average dimensional changes due to irradiation of these specimens is shown in Fig. 11.9. The graphite containing B^{10} -enriched B_4C or boron powder showed much longer dimensional changes than were observed with nonboronated or boronated graphite containing natural or B^{10} -depleted boron. The dimensional changes in the B^{10} -enriched boronated graphite showed the same anisotropic character as the nonboronated material.

This observation suggests that the dimensional change in the boronated graphite was probably controlled by accelerated change in the graphite matrix rather than swelling of randomly oriented B_4C or boron particles. Boron atoms in solution in graphite have been shown to produce accelerated dimensional change during irradiation (Ref. 2). However, swelling of B_4C granules during irradiation has also been demonstrated (Ref. 3). The observed dimensional changes in boronated graphite containing a greater volume load of B_4C than the B^{10} enriched materials show anisotropy in dimensional change but expansion usually takes place in both directions.

M33626-1

SIDE

M33626-2

TOP

Fig. 11.8. Boronated graphite made with B-10 enriched (91% B-10) boron powder 0.11 g B/cm^3 (shielding equivalence $\sim 0.55 \text{ g natural B/cm}^3$); irradiated at $700 \pm 50^\circ\text{C}$ to a fast-neutron exposure of $4.5 \times 10^{21} \text{ n/cm}^3$ ($E > 0.18 \text{ Mev}$). Note the specimen bow showing greater expansion on the side with the highest B-10 burnup; material 4465-24 specimen 4-4 (scale 0.1 in./div).

Fig. 11.9. Irradiation-induced dimensional change in B-10-enriched and nonboronated graphite baked at 2000°C and irradiated at 700° ± 50°C (filler GP-38 graphite powder <48 μm, C₀ 6.73 Å, L_c 770 Å)

These observations suggest that at least two mechanisms for dimensional change in boronated graphite compete. While these conclusions are preliminary, it would seem that dimensional change in boronated graphite can be minimized by fabrication processes which minimize both the quantity of B^{10} atoms in solution in the graphite matrix and the volume loading of B_4C . For example, saturating the matrix with B^{11} atoms prior to the introduction of B^{10} -enriched B_4C might be a process capable of yielding a desirable control material with irradiation properties very similar to nonboronated graphite.

HAFNATED GRAPHITE

The hafnated graphite specimens were unchanged in appearance by the irradiation. Dimensional change measurements were made on the specimens in the GGA hot-cell facility because of the high radioactivity of the specimens (~ 250 R, $B + \gamma$ at 6 in.). While the appearance of the specimens was good the strength was relatively low and under the pressure of the micrometer anvil some chipping and crumbling was observed. The results of the dimensional change measurements are shown in Fig. 11.10. It can be seen that the higher temperature bake had a slight effect in reducing axial expansion during irradiation. The hafnated graphites expanded axially less than the nonhafnated graphite. However, the hafnated graphites experienced a net volume expansion, whereas the nonhafnated graphite showed a net volume contraction. A complicating feature in comparing performance of the hafnated and nonhafnated graphite is the difference in anisotropy as shown by the coefficient of thermal expansion. The nonhafnated graphite had a much more anisotropic structure than the hafnated graphite and that condition as well as the addition of HfC probably influenced the relative dimensional changes during irradiation.

IRRADIATION EFFECTS IN CARBON AND GRAPHITE

Recent measurements obtained from piggyback specimens of LTI carbons irradiated in capsules P22 and F28 are reported below. The irradiated density as a function of the initial density for the various positions in the capsules are shown in Figs. 11.11 through 11.16. Dimensional change measurements as

Fig. 11.10. Irradiation-induced dimensional change in hafnated (HfC + graphite) and nonboronated graphite baked at 2000° or 2600°C and irradiated at 700° ± 50°C

Fig. 11.11. Density measurements of LTI piggyback specimens in cell 1 of capsule P22; irradiated at 700°C to 6×10^{21} nvt

Fig. 11.12. Density measurements of LTI piggyback specimens in cell 4 of capsule P22; irradiated at 800°C to 6.6×10^{21} nvt

Fig. 11.13. Density measurements of LTI piggyback specimens in cell 5 of capsule P22; irradiated at 750°C to 5.2×10^{21} nvt

Fig. 11.14. Density measurements of LTI piggyback specimens in hole 2-3 of capsule F28; irradiated at 900° to 650°C to 3.7×10^{21} nvt

Fig. 11.15. Density measurements of LTI piggyback specimens in hole 3-1 of capsule F28; irradiated at 900° to 650°C to 3.7×10^{21} nvt

Fig. 11.16. Density measurements of LTI piggyback specimens in thermocouple holes of capsule F-28; irradiated at 1250° to 900°C to 3.7×10^{21} nvt

a function of density for the various positions in the capsules are shown in Figs. 11.17 through 11.22. The dimensional changes for the design density of about 1.85 g/cm^3 are shown in Table 11.2. As can be seen in Figs. 11.14, 11.15, 11.20, and 11.21, annealing at 1400°C for 2 weeks leads to a reduction in the dimensional changes, especially at the higher densities where the anneal causes larger structural changes. The effect is about equal to the dimensional change which occurs during annealing and might well be explained by an annealing effect on the unannealed specimens in-pile. Under these conditions an accelerated dimensional change due to annealing was not observed.

TABLE 11.2
DIMENSIONAL CHANGES FOR A DENSITY OF 1.85 G/CM^3

Temp ($^\circ\text{C}$)	Fluence	Parallel Dimensional Change	Perpendicular Dimensional Change
900-650	3.7×10^{21} nvt	-0.05	-0.01
1250-900	3.7×10^{21} nvt	-0.055	+0.005
800	6.6×10^{21} nvt	-0.075	+0.055
750	5.2×10^{21} nvt	-0.06	+0.03
700	6.0×10^{21} nvt	-0.065	+0.025

References

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2. Mayer, R. M., "Influence of Boron on the Nucleation of Radiation Damage in Graphite," Presented at the 8th Biennial Carbon Conference, Buffalo, New York, June 1967; Abstract published in Carbon **6**, 211 (1968).
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Fig. 11.17. Dimensional change of specimens in cell 1 of capsule P22; irradiated at 700°C to 6.0×10^{21} nvt

Fig. 11.18. Dimensional change of specimens in cell 5 of capsule P22; irradiated at 750°C to 5.2×10^{21} nvt

Fig. 11.19. Dimensional change of specimens in cell 4 of capsule P22; irradiated at 800°C to 6.6×10^{21} nvt

Fig. 11.20. Dimensional change of specimens in hole 2-3 of capsule F28; irradiated at 900° to 650°C to 3.7×10^{21} nvt

Fig. 11.21. Dimensional change of specimens in hole 3-1 of capsule F28; irradiated at 900° to 650°C to 3.7×10^{21} nvt

Fig. 11.22. Dimensional change of specimens in thermocouple holes of capsule F28; irradiated at 1250° to 900°C to 3.7×10^{21} nvt