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Gulf General Atomic
Incorporated

AEC RESEARCH AND
DEVELOPMENT REPORT

GA-10088

HTGR BASE PROGRAM
QUARTERLY PROGRESS REPORT
FOR THE PERIOD ENDING
MAY 31, 1970

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

June 30, 1970

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GA-9372-March, 1969, through May, 1969
GA-9660-June, 1969, through August, 1969
GA-9815-September, 1969, through November, 1969
GA-9944-December, 1969, through February, 1970

INTRODUCTION

This report covers the work performed by Gulf General Atomic Incorporated under U.S. Atomic Energy Commission Contract AT(04-3)-167, Project Agreement No. 17. This Project Agreement calls for preliminary design and related exploratory development of a 1000-Mw(e), gas-cooled, nuclear power reactor incorporating advanced technology, and a 150-300 Mw(e) prototype. This program is based on the concept of the High-temperature Gas-cooled Reactor (HTGR) system developed by 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 the prototype at an earlier date. The plant would demonstrate such new design features as:

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

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

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Task IV

FISSION-PRODUCT MECHANISMS

FISSION-PRODUCT TRANSPORT IN HTGR SYSTEMS

A study and reassessment of fission-product transport in HTGR systems was made. The intent of the study was (1) to examine the main features of fission-product behavior in HTGR systems, (2) to summarize recent developments in the analysis of fission-product release and plateout, and (3) to summarize the results of recent investigations of fission-product release.

Noble Gas Release

To understand more fully the noble-gas release process, the end-of-life $\text{Kr}^{85\text{m}}$ R/B values at 1300°C for a number of ORNL, Dragon, and Gulf General Atomic experiments were assembled in roughly chronological order as shown in Fig. 4.1. The first point on the graph is the result obtained by GGA (Ref. 1) for an uncoated carbide and the next two are the results obtained by ORNL for an uncoated carbide (Ref. 2) and a mechanically uncoated carbide (Ref. 3). The next point is a Pluto IIIA result obtained by the Dragon Project (Ref. 4). This result was multiplied by a factor of five to account for the 20% particle failure. The next six points are early results on coated particles obtained from the GA 309 sweep capsule series (Ref. 1) irradiated in 1960 through 1961. The next two are results obtained from 1962 to 1964 for the GAIL III-A and III-B in-pile loop experiments (Refs. 5, 6). The next three are results from the three-compartment 6F2 sweep capsule (Ref. 7) irradiated in 1963. The first compartment of this experiment had fuel coated with multiple layers of silicon carbide and pyrolytic carbon. Since only about one-fourth of the particles failed during the irradiation, the $\text{Kr}^{85\text{m}}$ R/B value was arbitrarily multiplied by a factor of four to indicate release from the kernels alone.

R/B OF Kr-85m AT ~1300°C

Fig. 4.1. Kr-85m R/B values corrected to 1300°C

The next point is from the GAIL IV experiment (Ref. 8) irradiated from 1964 to 1966. Since it was known to have a particle failure of only 0.8% at the end of life, this value was arbitrarily multiplied by 125. The last point is from Peach Bottom Core I results (Ref. 9). This result was increased by a factor of two to account for the lower temperatures. This temperature correction is obtained by assuming that the release rate has an exponential temperature dependence with an activation energy of approximately 30 kcal/g mole (Ref. 1).

It can be seen from Fig. 4.1 that all of the end-of-life $\text{Kr}^{85\text{m}}$ R/B values are approximately the same if corrections are made for the amount of failed particles. It is therefore likely that the end-of-life release is from exposed kernels rather than from diffusion out of contaminated coatings. Also, it was noted that particle damage ranged from minor distortion to gross distortion, with the final kernel volume more than twice the original kernel volume. Therefore, it is probable that the characteristic dimension that governs diffusional release from the kernel is much smaller than the radius of the kernel.

The similarity of the releases from all of these experiments indicates that there is a noble-gas release rate characteristic of failed fuel, and that the rate is relatively independent of the initial material or of the degree of failure. The reactor designer can therefore establish the design noble-gas release levels by determining the amount of fuel failure to be expected and using a release curve obtained from an appropriate experiment having approximately fully failed fuel. If one ignores precursor contributions, the number of noble gas atoms in the coolant stream is then governed by the equation

$$\frac{dn}{dt} = fB(R/B)_{\text{failed}} - n\lambda - nF \quad ,$$

where n is the concentration of noble gas in the coolant stream, t is the time, f is the fraction of fissions occurring in failed fuel, R/B is the instantaneous release rate divided by the birth rate, λ is the decay constant, and F is the fractional removal rate by the purification system.

FIPER Code Analyses

Cesium activity profiles measured on the Peach Bottom DO6-01 fuel element and the GAIL IV fuel element are shown in Figs. 4.2 and 4.3 (Ref. 10). Each set of profiles is from a different elevation along the length of the fuel elements. It can be seen from these steeply sloping profiles that the direct loss of cesium in these experiments was negligible.

The curves in Figs. 4.2 and 4.3 are the results of an attempt to fit the data with the FIPER code (Ref. 11), which was developed under the Base Program. The cesium diffusion coefficient that appeared to give the best fit to the data was the same for both of these experiments and, for some reason, appeared to be very small and to have the extremely low activation energy of 3 kcal/g mole. Over the temperature range investigated, the temperature dependence of the diffusion coefficient can actually be approximated by a temperature to the 3/2-power dependence, indicating that the rate-limiting step is possibly gas-phase transport at the low concentration levels of these experiments. (It should be noted that these data cover three different types of graphite, two of which were impregnated.) These curves should be considered an illustration of the transient FIPER code results rather than a confirmation of the diffusion process; it is hoped that this difficulty can be resolved by the analysis of data obtained from examination of additional Peach Bottom elements.

Figure 4.4 shows the results of a series of out-of-pile strontium diffusion experiments performed at Gulf General Atomic. The solid lines on these plots are the results of a FIPER analysis using the GGA reference design diffusion coefficient.* It can be seen that a reasonable fit to the data has been obtained, considering the experimental difficulties and uncertainties.

DIFFUSION OF STRONTIUM IN GRAPHITE

Work on the diffusion of strontium in graphite consists of (1) permeation experiments to determine effective diffusion coefficients for strontium in

*The reference diffusion coefficient data for strontium in graphite are described by the equation $\ln D = 5.7 - (3.0 \times 10^4/T)$.

Fig. 4.2. Cesium profiles in Peach Bottom DO6-01 fuel element

Fig. 4.3. Cesium concentration in GAIL IV (Type 57KBLC particles were closer to GETR core)

Fig. 4.4. Strontium diffusion profiles at 1000°C

graphite and (2) autoradiographic experiments to study the mode of diffusion of strontium in graphite.

Permeation Experiments

Strontium permeation experiments 4370-143 and 4370-144 have been temporarily discontinued. However, plans are under way to modify the apparatus so that concentration profiles in the graphite can be determined periodically during an experiment. It is desirable to follow the strontium concentration profile in a permeation experiment as a function of time to ensure that steady-state release has been reached. To achieve this objective, the graphite crucibles will be periodically cored with an appropriate drill having a diameter of about 0.25 in., and the concentration profile in the cored material will be measured. The resulting hole will be closed with a graphite plug and the experiment continued. A larger crucible is necessary to keep the unperturbed diffusion area large.

Autoradiographic Studies

Further development of the autoradiographic method of study of diffusion of strontium in graphite is being made, establishment of an appropriate source for these studies having been one of the chief objectives during the current reporting period. To date, powders have received the principal consideration.

Powdered furfuryl alcohol coke, which had been doped with strontium containing Sr^{89} as a tracer, was placed in an H-327 graphite crucible and heated to 800°C for 30 min under vacuum. The part of the crucible that had been near the coke was sectioned into thin pieces using a diamond wire saw. The four sections nearest the coke were cleaned in benzene and then autoradiographed. In general, although the film was slightly darkened only by the surfaces that were adjacent to the powder, there were many spots from the sectioned pieces that indicated high concentrations of Sr^{89} . The spots are thought to have been caused by powder migration rather than by diffusion of the strontium.

In order to find a cleaner source material than the furfuryl alcohol coke powder, a solid sample of furfuryl alcohol coke doped with Sr^{89} was placed in a tantalum crucible with a sample of POCO carbon and a sample of the coke powder. The crucible was heated to 1500°C for 5-1/2 hr. The samples were then removed from the crucible and their relative specific activities were found to be: powdered coke, 1; solid coke, 0.419; and POCO carbon, 22.8.

According to these data, it would appear that the POCO carbon would make a better source matrix than the powdered furfuryl alcohol coke. However, it was found to dust slightly, and its dust, which was very active, caused even more problems than the coke dust. It is now believed that the dust constitutes a problem in autoradiographic studies, and for this reason a gas source is being investigated for the diffusion studies.

VAPORIZATION OF STRONTIUM FROM GRAPHITE

Some further thought has been given to the data concerning the effect of CO on strontium partial pressures over graphite (see earlier quarterly report GA-9815). Under some circumstances, a very strong depression in strontium vapor pressure was achieved. However, under experimental conditions that more closely approached reactor conditions, only a minor reduction of about 10% was observed. Extrapolation of the data to reactor conditions indicates that no significant dependence of strontium partial pressure on CO pressure would be expected, and no further work is planned in this area.

The sorption behavior of 37- to 74- μm grain size H-327 graphite has been remeasured in a single experiment. The pressures at a given concentration differ somewhat from those previously reported (see earlier quarterly report GA-9372), being somewhat lower. It is believed that the difference is due to kinetic effects, and experiments are planned to investigate this question. These studies should lead to improved reliability of vapor pressure measurements.

Diffusion samples were prepared for irradiation in the Peach Bottom fuel test element in order to provide important information on the diffusion of strontium, cesium, barium, and samarium in graphite under reactor conditions. The study will also provide information on the effect of getter materials (sulfur and magnesium fluoride) on the migration of the metals.

The diffusion samples consist of graphite crucibles containing metal carbide sources distributed uniformly on coke material. The graphite crucibles are 1.25 in. long, 1.00 in. in outer diameter, and have a 0.2-in. wall thickness. Each crucible has a graphite cylinder in the center. The crucibles are coated with polystyrene to avoid hydrolysis of the metal carbides during shipping. The crucibles will be irradiated at temperatures between 800° and 1150°C. After irradiation, each graphite crucible and inner graphite cylinder will be sectioned, and metal concentrations in the graphite will be established. The source material will be analyzed to determine the total metal loss from the source. From the total metal losses and the concentration gradients, migration rates and diffusion coefficients will be calculated. Having the graphite cylinder in the center assures concentration profiles for slow migration where no measurements can be made of the fractional release.

In preparation of the source material, metals were sorbed on furfuryl alcohol coke, converted to the carbides, and loaded into graphite crucibles. The metals used were natural cesium, Sr^{40} (80% enriched with Sr^{84}), Sm^{152} and barium tagged with 7.2-y Ba^{133} . Natural cesium consists of Cs^{133} that activates to 2.1-y Cs^{134} during irradiation; Sr^{84} activates to 65-d Sr^{85} ; and Sm^{152} activates to 47-h Sm^{153} . Mixtures of the metals were used in some samples, and selected samples contained magnesium fluoride and sulfur for use in studying the effectiveness of these materials as getters for the metals.

FISSION-PRODUCT RELEASE STUDIES IN FUEL TEST ELEMENT

Fuel samples have been prepared for irradiation in the Peach Bottom fuel test element to study the release of fission-product metals from coated

fuel particles. The objective of this work is to relate fission-product release behavior with particle characteristics, burnup, time, and temperature.

The samples consist of a mixture of coke material and coated fuel particles contained in a graphite crucible. The coke material and the graphite crucible are sorbent materials for the released metal fission products. After irradiation in the reactor, the particles, coke material, and crucibles will be separated and analyzed to determine the release levels for important metallic fission products such as Cs^{137} , Sr^{90} , and Ba^{140} .

Each sample contains approximately 500 fuel particles coated with either TRISO (silicon carbide type) or BISO (pyrolytic carbon type) coatings. The particles were carefully selected using X-ray radiography to ensure that all particles had intact coatings, were spherical, and were of similar size.

Most of the samples were held in an outer graphite crucible; however, some of the samples were encapsulated in containers made of Nb-1% Zr alloy. Encapsulation of the samples in a metal (barrier-type) container was necessary in order to determine release values below about 5%. This is because of cross-contamination from other samples and from the reactor fuel. The Nb-1% Zr alloy was chosen as the container material because of its nuclear properties and low carbide formation rate. Calculations indicate that crucibles made of this alloy with wall thicknesses of approximately 0.25 in. will survive the time-temperature conditions in the reactor core.

Work on preparing fission-product release samples for use in the fuel test element has been completed.

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Task V
RECYCLE FUEL STUDIES

RECYCLE TEST ELEMENTS

Test Element Design

The basic test element design was described in the previous quarterly report (GA-9944) and has remained essentially unchanged. The loadings described in Table 5.1 of GA-9944 have been modified slightly, and these are reflected here in Table 5.1. Table 5.2 describes the various combinations of fissile and fertile particles noted in Table 5.1.

The detail design of the recycle test elements is complete. Details of both thermal analysis and the mechanical design are presented in Task VIII of this report, since the recycle test elements have the same general design as the fuel test element. The recycle test elements are not instrumented.

Each recycle test element has been assigned a position in the Peach Bottom core, and these are shown in Table 5.3. With the location selected, the required uranium loadings were determined. The axial temperature profiles for the various elements are shown on Figs. 5.1 through 5.4. The radial temperature profiles are shown as Fig. 5.5 for a body with bonded rods and Fig. 5.6 for a body with blended beds.

Test Element Fabrication

Fabrication is in progress on the eight recycle test elements. All of the graphite components are ready for assembly. The blended bed fuel bodies are complete. Bonded rod fabrication is in progress for the bonded rod body assemblies.

Table 5.1
RTE LOADING COMBINATIONS^a

Position of Fuel Body in Element	Centerline Temp Range (°C)	First Discharge		Second Discharge		Final Discharge			
		RTE 1 ^b	RTE 2	RTE 3 ^b	RTE 4	RTE 5 ^b	RTE 6	RTE 7 ^b	RTE 8
6 (top of reactor)	2050-1950	bdei	f	bdei	e*	bdei	g	acfg	f
5	2250-2050	behd	a*	behd	d	behd	f	acfg	i
4 (max flux)	2250-2300	acfg	e	acfg	f	acfg	f	begd	d
3 (max flux)	2050-2250	acfg	f*	acfg	a*	acfg	d	begi	e
2	1650-2050	acfg	d	acfg	f*	acfg	f	begi	i
1 (bottom of reactor)	1050-1650	acfg	e*	acfg	e	acfg	e	begi	f*

^aLoading compositions are indicated by letters a through j as defined in Table 5.2. The fuel compositions marked with an asterisk (*) are blended beds; all others are bonded rods.

^bIn these elements, each pair of adjacent fuel holes contains one of the four fuel compositions indicated.

Table 5.2
RTE LOADING COMPOSITIONS^a

Composition	Fissile	Fertile
a	(Th,U)O ₂ ^b	None
b	UO ₂	ThC ₂
c	(Th,U)O ₂ ^c	ThO ₂
d	(Th,U)O ₂ ^c	ThC ₂
e	UC ₂	ThC ₂
f	UC ₂ TRISO	ThC ₂
g	UO ₂	ThO ₂
h	UC ₂	ThO ₂
i	UC ₂ TRISO	ThC ₂ TRISO

^aUnless otherwise indicated, all particles are BISO coated.

^bTh/U = 4.2.

^cTh/U = 2.0.

Table 5.3
CORE LOCATION FOR RECYCLE TEST ELEMENTS

<u>Recycle Test Element</u>	<u>Core Location</u>
1	B07-06
2	F07-06
3	A10-06
4	B10-06
5	C10-06
6	D10-06
7	E10-06
8	F10-06

Fig. 5.1. Axial temperature profile at five full-power days for recycle test element 1, core location B07-06, 95% power, 105% flow

Fig. 5.2. Axial temperature profile at five full-power days for recycle test element 2, core location F07-06, 95% power, 105% flow

Fig. 5.3. Axial temperature profile at five full-power days for recycle test elements 3, 5, 6, 7, and 8, core location A-, C-, D-, E-, and F10-06, 95% power, 105% flow

Fig. 5.4. Axial temperature profile at five full-power days for recycle test element 4, core location B10-06, 95% power, 105% flow

Fig. 5.5. Radial temperature profile at point of peak axial temperature for a body with bonded rods

Fig. 5.6. Radial temperature profile at point of peak axial temperature for a body with blended beds

The blended bed is a uniform blend of fissile, fertile, and coke particles. The particular fissile and fertile particles are defined on Table 5.2. One type of coke is used in all the blends. It is a calcined petroleum coke with a particle size of 60 to 125 μm with less than 1% sulphur content. There were 6.20 g of coke used in each fuel hole of the recycle test element bodies. Photos of a typical blended bed are shown in Figs. 5.7 and 5.8. The procedure developed and used at Gulf General Atomic for loading the blended beds is discussed below.

Every 15.5-in.-long graphite body is inspected to ensure that the requirements of the drawing are met. A typical machined body is shown on Fig. 5.9. Qualified bodies are vacuumed and cleaned as required, and the fuel body serial number and graphite log number are engraved on the bottom of the body with a vibratool.

The fuel body assembly drawing specifies the uranium and coke loading per fuel hole in each body. It also specifies the fuel particle combination shown on Table 5.2. The quantities of fissile and fertile particles required are calculated from the actual particle data sheets. These quantities are weighed out to an accuracy greater than ± 0.01 g.

The blending equipment used to load the particles in the body is shown in Fig. 5.10. Each particle loading weighed out for a given element is run through the blender individually and timed. The orifices of the blender are adjusted to obtain the loading time for each particle type within 0.2 sec of the other two particle types. An effort was made to have the blender empty within the 0.2 sec in the following sequence: fissile, coke, and fertile particles.

An aluminum sampling tube having the same internal diameter as the fuel hole is loaded for each fuel combination. The tube is then gamma scanned to check the uranium uniformity along the length of the rod (gamma scan equipment is shown in Fig. 5.11). This gamma scan plus visual observation of each blended bed loading is the basis for determining loading uniformity.

7X

20X

Fig. 5.7. Cross section of 27-1/2-in. blended bed 15 in. from bottom, showing at right the smaller coke particles intermixed with fuel particles

7X

20X

Fig. 5.8. Axial midplane of 27-1/2-in. blended bed 21 in. from bottom, showing at right the smaller coke particles intermixed with fuel particles

Fig. 5.9. Typical recycle test element fuel body

Fig. 5.10. Apparatus for loading blended beds

Fig. 5.11. Gamma scan equipment

Each body was loaded as shown on Fig. 5.12. A sliding gate valve, which is solenoid operated, covers all three orifice openings. When energized, all three orifices open simultaneously, and the particles flow directly into a graphite funnel striking the inclined surface of the funnel. This provides a mixing of the particles before they enter the fuel hole and minimizes radial segregation of the fissile and fertile particles.

The fuel body is tamped vertically during the loading and for 1 hr after all eight holes are loaded (the vertical tamping mechanism is shown in Fig. 5.10). A cam raises the support plate approximately 0.04 in., and then drops it causing impact loading on the blended bed. Occasional side tamping is also used to accelerate the packing of the bed. Aluminum rods were placed on the top of each bed during the tamping to prevent particle segregation in the top layers of the bed.

Upon completion of tamping, the top 1 to 2 in. of the blended bed is bonded to the inside surface of the fuel hole with a liquid thermosetting polystyrene solution to prevent segregation during shipment. The polystyrene volatilizes at reactor temperatures.

The fuel hole caps are then installed, and the bottom center hole cap is cemented and cured in place as shown in Fig. 5.13. The center spine is loaded into the body and the top center hole cap is installed. The body assembly is then ready for loading in the fuel element sleeve at its desired axial position.

HTGR FUEL RECYCLE PLANT STUDY

Work continued on plant designs and cost estimates for a combined HTGR fuel reprocessing-refabrication (fuel recycle) facility sized for handling fuel from and for HTGR economies totaling 15,000, 30,000, and 45,000 MW(e). When the study was initiated, the reference fuel particles were pryocarbon coated (BISO) and loaded into the graphite fuel blocks as blended beds of fresh fissile (U^{235}), fertile (Th), and recycle mixed (Th- U^{233}) particles. The design of the fuel recycle plant proceeded during the previous

Fig. 5.12. Views showing (left) loading of fuel particles into blender (gate valve closed) and (right) loading of blended bed in fuel body

Fig. 5.13. Curing of bottom cap cement with heating tapes

quarter to the stage of block process flow diagrams, material balances, and partial equipment sizing. The reprocessing plant process flow diagrams are: (1) head end and graphite disposal (spent fuel receiving and storage, particle and graphite separation, and graphite burning); (2) U^{233} and Th recovery (particle burning, U^{233} and Th decontamination and separation by solvent extraction, U^{233} purification and transfer to the refabrication facility, and thorium storage); (3) U^{235} recovery (particle burning, U^{235} decontamination and purification by solvent extraction and silica gel treatment, and loading for shipment); and (4) waste processing (solid, liquid, and gaseous waste collection, concentration or cleanup and disposal). Process flow diagrams for the refabrication plant are: (1) U^{233} sol production (U^{233} and thorium solutions conversion to a sol), (2) U^{233} gel production (gel and kernel production), (3) fuel element assembly (purchased graphite blocks and BISO coated thorium particles are combined with the Th- U^{233} particles into fuel blocks and stored until shipped to reactors), and (4) scrap treatment and waste disposal (fuel components are recovered or concentrated for disposal).

During the current reporting period, a redefinition of the reference fuel was made because of improvements in the design of advanced HTGR fuel. Accordingly the work done on the designs and cost estimates for the recycle plant was reviewed, and adjustment of the process flow diagrams and material balances was started. The reference fuel was redefined, because irradiation tests on fuel rods consisting of close-packed beds of coated particles bonded with a carbonaceous matrix have shown that these rods have excellent stability under irradiation. In addition, bonded rods will have lower thermal gradients than blended beds.

The newly defined fuel element compositions are shown in Table 5.4. These compositions are based on advanced reactors having fuel loading with carbon-to-thorium ratios of 270, average burnups of about 98,000 MWd/ton and with a silicon carbide coating included on U^{235} fuel kernels. Bred U^{233} and U^{235} particles are loaded into separate types of fuel blocks. The refabrication plant is thus designed to handle only U^{233} -particle fuel elements. The U^{235} -containing fuel elements are made by the "fresh" fuel fabrication plant under "cold" conditions. The advanced HTGR fuel loading system is shown in

Table 5.4

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	10.856	8.880	9.530
SiC	0	1.938	1.300
Kernel oxygen + carbon	1.069	0.901	0.956
Thorium	7.946	7.946	7.401
U233 mix	1.111	0	0.365
U235 mix	0	0.758	0.135
Pu	0	0	0.001
Fission products (Approx.)	0	0	0.913
Total Weight	121.782	121.223	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	11.400	9.253	
SiC	0	2.130	
Kernel oxygen + carbon	1.093	0.909	
Thorium	7.946	7.946	
U233 mix	1.221	0	
U235 mix	0	0.834	
Total Weight	122.460	121.872	

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

Table 5.5. Only recycle (U^{233}) fuel is considered in this study. Both the recycle and initial core consist of elements requiring no particle separation (Type A) and those requiring particle separation (Type B). Type A elements have only BISO coatings and either contain only U^{233} or only small amounts of U^{236} with U^{235} that can be recycled to the reactor. Type B elements contain TRISO coated U^{235} particles, which prevents mixing of the U^{235} and U^{233} during reprocessing if Type A and B blocks should become mixed.

The fuel design change has but little effect on the recycle facility process flow diagrams. These minor effects involve the handling of the silicon carbide-coated fuel particles in the head-end section of the fuel reprocessing plant and the making of fuel sticks in the fuel refabrication plant. Because the quantitative makeup of the fuel elements is altered, a recalculation of the material balances is required and some equipment pieces must be resized.

NATIONAL HTGR RECYCLE DEVELOPMENT PROGRAM PLAN

Review and revision of the National HTGR Recycle Program Plan were started, primarily in the areas of schedules and costs. This is being done to make the program reflect the changes made this quarter in the reference fuel element design and to update the program timing. The experimental program changes are expected to be minor, since the reference fuel of the first program had all TRISO coated particles, whereas in the new reference fuel, the particles that contain U^{233} after irradiation will be BISO coated.

HTGR REFERENCE FUEL REPROCESSING PLANT

A conceptual HTGR fuel reprocessing plant design and unit fuel reprocessing charges have been produced by the AEC Division of Production. During this quarter, the conceptual plant design criteria, the plant design, and capital and operating costs, and methods of obtaining unit reprocessing charges were reviewed. Comparisons were made with the Gulf General Atomic HTGR fuel reprocessing plant design and costs. The evaluation of the AEC plant study was made for the AEC.

Table 5.5
ADVANCED HTGR FUEL LOADING SYSTEM

	Initial Core	Recycle Core
Type A element, no particle separation required	U ²³⁵ C ₂ BISO ThC ₂ BISO	(Th,U ²³⁵)O ₂ BISO ThC ₂ BISO
Type B element, particle separation required	U ²³⁵ C ₂ TRISO ThC ₂ BISO	U ²³⁵ C ₂ TRISO ThC ₂ BISO

The conceptual plant design criteria chosen by the Division of Production are that the reprocessing plant should be a single-purpose plant using a burn-leach head end, a modified acid-Thorex solvent extraction system for recovery of heavy metal from fertile particles, storage of thorium solutions, and calcination of radioactive waste. Essentially the same criteria have been used by Gulf General Atomic in independent HTGR fuel reprocessing plant studies. These are also the bases of the process selected for further development by the National HTGR Recycle Development Program.

HEAD-END PROCESSING

Fluid Bed Burning

Continuous fluid bed burning with alumina-free TRISO particle beds (TRISO particles burned down to the SiC coating) continues to show promising results.

Several modifications in the 4-in. fluid bed have been completed, including control equipment that will give continuous feed and continuous product withdrawal, a water-cooled heat exchanger on the product line, a new base cone and in-bed CO burner, an off-gas monitoring system consisting of gas chromatographic analysis plus continuous oxygen analysis, and off-gas cleaning system that includes cyclone fines separation, metal blow-back filters, and absolute filters.

Continuous operation of the fluid bed burner has been achieved in which a crushed particle-graphite mixture (-1/4-in.) is continuously removed from a storage hopper and fed through a screw feeder to the burner. Since it is advantageous to have a carbon-free product, burning is maintained with a minimum carbon inventory in the bed. The bed, after ignition, is built up to the operating level and the excess carbon is burned. As the bed becomes carbon free, the oxygen concentration in the off-gas increases; the addition of feed increases the burnable carbon in the bed, and the oxygen concentration decreases. By controlling the feed rate (using the indicated off-gas oxygen concentration as a guide), a minimum amount of carbon in the product is obtained with a 98% to 99% utilization of the oxygen. (This same oxygen utilization is obtained within a wide range of air-oxygen mixtures.) Particles with less than 1% carbon can be continuously withdrawn. The product passes through the heat exchanger and is stored in a bin. Near quantitative recovery of particles is obtained, indicating that little particle breakage occurs in the fluid bed.

The centerline temperature of the bed is maintained at about 950°C during normal continuous operation. If the temperature is allowed to drop to 750°C, combustion cannot be maintained. All efforts to revive combustion by feeding more or less oxygen or carbon have failed. Above 900°C, combustion can easily be controlled at constant temperature by balancing the carbon feed rate against the oxygen flow. Between 750° and 900°C, combustion may be revived, but there is a chance it may not.

Bed depth affects oxygen utilization. The deeper beds studied to date (about 24 in.) have coincided with higher concentration of oxygen in the off-gas. This is probably due to slugging in the deeper beds and is probably a function of depth/diameter ratio.

Runs 20 and 21 were completed before the feed control system was installed. Loose TRISO fertile particles were burned with no particle agglomeration, and no operational problems were encountered.

Runs 22 and 23 were conducted to obtain data for energy balance calculations. Graphite was burned in alumina at an average rate of 21.2 g C/min. Coolant flow rates and temperatures including surface temperatures were measured. Figure 5.14 shows the heat loss distribution at equilibrium for Run 23. These tests indicate that scaleup predictions of energy removal requirements can be satisfactorily obtained from theoretical calculations.

Basic data for Runs 24 through 32 are shown in Table 5.6. These runs were conducted primarily to study the phenomenon of particle agglomeration, which occurs when resin-bonded fuel rods are burned. Results from these tests showed that fluidization with high gas velocities delays but does not eliminate agglomeration. Loose particles and pitch-bonded rods have been burned with no particle agglomeration.

Runs 24 through 28 were attempts to burn resin-bonded fuel rods (crushed and screened to $-1/4$ -in.). In each case, particle agglomeration terminated the run. Loose TRISO particles were burned on a continuous basis during Run 29. Here, particle agglomeration did not occur, and near quantitative recovery of particles was obtained.

Runs 30 through 31 were two additional attempts to burn resin-bonded rods on a continuous basis. Each run lasted only about 2 hr before being terminated by lump formation.

Crushed graphite screened to $-1/4$ in. was fluidized and burned during Run 31A (which is not included in Table 5.6). No inert material was present, and a burning rate of 31.3 g/min was obtained. The oxygen utilization was 99.8% with an air-to-pure-oxygen ratio of more than three. An inlet gas flow rate of 200 l/min was required to keep the porous bed fluidized.

Run 32 was a continuous burning of pitch-bonded fuel rods; there was no particle agglomeration. Five pitch-bonded fuel rods were fabricated for burner feed. Each rod weighed about 3.3 kg and was about $2-3/4$ in. in diameter and 15 in. long. The rods were carbonized and crushed in a jaw crusher with the maximum opening of $3/4$ in., then recycled with a maximum

Fig. 5.14. Heat loss distribution for Run 23, burning rate 21.2 g/min

Table 5.6
BURNING PARTICLE-GRAPHITE MIXTURES IN THE 4-IN. FLUID BED

Run	Feed (g)		Product and Final Bed (g)		Burning Time (min)	Burning Rate (g/min)	Approximate Particle Recovery (%)	Average Oxygen Utilization (%)	Remarks
	TRISO Particles ^a	Burnable Carbon	TRISO Particles ^b	Burnable Carbon					
F4B24	6195.0	2891.0	5981.3	280.8	125	20.9	96.6	98.0	Agglomeration
F4B25	5738.2	1708.9	5407.3	301.0	97	14.5	94.5	99.8	Agglomeration
F4B26	6751.3	2354.8	6438.2	349.1	112	17.9	96.5	97.0	Agglomeration
F4B27	5705.1	2445.0	5846.1	276.0	84	25.8	100+	97.0	Agglomeration
F4B28	9902.1	3870.4	9826.6	736.4	113	27.7	99.3	98.5	Agglomeration
F4B29	7221.0	3484.0	7222.1	282.4	168	19.1	100+	92.0	No agglomeration
F4B30	8643.5	3208.6	8432.8	411.7	130	21.5	97.7	98.0	Agglomeration
F4B31	8243.3	3201.7	8206.3	397.3	125	22.5	99.7	98.0	Agglomeration
F4B32	8277.9	6667.5	7975.0	492.7	269	22.9	96.4	99.0	No agglomeration

^aWeight of particles in SiC coats, calculated from a preliminary burning test.

^bWeight of particles in SiC coats recovered.

opening of 3/8 in. Approximately 8.2% particle breakage occurred during rod preparation, carbonizing, and crushing. Another 4.3% particle breakage was noted after screen-tumbling for 20 min. This large fraction of broken particles is not regarded as a problem at present because the diameter of the rods (2-3/4 in.) is much larger than the crusher jaw gap (3/4 in.).

The screen-tumbling was done with a 1/4-in. screen opening; 83.8% of the material came through in 2 min and 92.5% was through after 20 min. There were 12 kg of the -1/4-in. rod material fed to the fluid bed burner during continuous operation. Another 1.7 kg of the -1/4-in. rod material was fed with an equal weight of crushed -1/4-in. graphite. After nearly 5 hr of continuous operation, the bed was allowed to burn out. Near quantitative recovery of particles was obtained, with generally less than 2% burnable carbon in the product samples. An interesting result from this experiment was the behavior of the -1/4-in. graphite when fed along with the crushed rods. Product samples taken during this phase of the run had less than 0.3% burnable carbon in the fertile particle size range. The carbon fines had burned first, leaving the remaining burnable carbon about +869 μm in size. These graphite "lumps" could be recycled to continue burning.

Fines entrainment in the off-gas is a common situation associated with fluid bed operation. Figure 5.15 shows the characteristics of solids entrainment for fluid bed burning in a bed of alumina-free TRISO particles. An approach being studied is to use cyclone and metal blow-back filter separation of the solids. The fines will be recycled and fed to the bottom of the burner. Because of the large ratio of surface area to mass for fines, the carbon should burn before reaching the top of the bed.

Another area being studied is the tendency of the crushed graphite and fuel rods to plug in the line between the hopper and the screw feeder, and modifications in the present equipment are in progress. Also, 100 lb of crushed graphite, screened to -1/4 in., will be sent to Vibra Screw Inc. to be tested in their hopper-feeder combinations.

Fig. 5.15. Solids entrainment, 4-in. fluid bed

Tests are in progress to determine the effect of alumina on particle agglomeration when burning resin-bonded fuel rods. Also, pitch-bonded TRISO-BISO fuel rods (BISO fertile) are being prepared for carbonization and subsequent fluid bed burning.

The 8-in. fluid bed burner has arrived, and installation should soon be completed.

Leaching

The objectives of the leaching studies are to define the principles of operation of the leacher, which is being fed with Thorex liquor and ThO_2 , and to specify the burner product. To date, three glass leachers have been built and operated.

The first leacher (No. 1) (illustrated in Fig. 5.16) was batch operated and utilized recycling of the solution. Operation was as follows: 1 liter of Thorex liquor* was charged into the leacher and the temperature raised to about 115°C . The air-lift pump, which recycled solution at about 300 ml/min, was started. A weighed amount of solute was then added through the top powder feed inlet, and the experiment was terminated when the whole solution became transparent. Samples of solution were taken at various times to determine the reaction rate. After the solution had been taken from the apparatus, it was filtered to remove undissolved solids. The solution volume was measured, and the solids were washed and dried. Both the solution and solids were analysed for thorium.

Run 1L1 showed that while the -325 mesh solids (burned, crushed TRISO fertile particles) was being added, the solution boiled vigorously and the rate of addition had to be restricted to prevent the solution from boiling over. The solution became dark brown and copious NO_2 fumes were evolved. The results are given in Table 5.7.

*Thorex liquor is 37.5 g $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$; 10 ml of 48% HF; 70% nitric acid added to make 1 liter solution.

Fig. 5.16. Leacher No. 1 which used recycling of solution

Table 5.7
RESULTS OBTAINED FROM LEACHER NO. 1
(Temp, 115°-120°C; Volume of solution, ~1 liter)

Description of Feed Solids	Wt. of Solid Feed (g)	Wt. of Th (g)	Time taken to add Feed (min)	Samples		Wt. of Th in Final Solution (g)	Residue								
				Time (min)	Conc. (g Th/l)		Wt. (g)	Th Content (g)							
Run 1L1															
TRISO fertile carbide particles burned to SiC coat, crushed, and burned in a trav; -325 mesh.	137.73	104	12	30	82	102	12.2	2.8							
				89	73										
				128	89										
				166	101										
				220	79										
				273	90										
Run 1L2															
As in 1L1, but +325 mesh.	240.79	~140	15	30	144	149	62.7	0.4							
				60	145										
				90	148										
				150	148										
				210	150										
				270	150										
				330	149										
				390	147										
Run 1L3															
As in 1L1. Sieve Analysis Mesh wt-% +60 1.4 -60+120 28.6 -120+140 13.1 -140+200 12.2 -200+230 9.3 -230+325 10.7 -325 24.7	268	~200	30	10	215	194	38	0.2							
				20	195										
				30	208										
				50	210										
				70	204										
				130	206										
				Run 1L4											
				Burned BISO fertile carbide particles; 750°C. Sieve Analysis Mesh wt-% +60 0.1 -60+120 59.0 -120+140 15.4 -140+200 9.0 -200+230 4.4 -230+325 4.3 -325 7.8	265				~232	58	No samples		~232	0	0

Run 1L2 was a repeat of 1L1, except that +325-mesh feed was used. Again, the solution became brown and NO_2 was evolved. The results are shown in Table 5.7

It was concluded from these two runs that carbon that had not been completely oxidized in the feed solid had been attacked by nitric acid giving rise to NO_2 . The cause of the brown color is not known.

In run 1L3, the feed solids were oxidized thoroughly to ensure the complete removal of carbon, and the solution remained colorless. No NO_2 was evolved. The solution boiled vigorously despite the fact that the initial temperature was 97°C (i.e., well below the boiling point of 120°C). The results are shown in Table 5.7. It was concluded that boiling is caused by evolution of heat as the ThO_2 dissolves.

In Run 1L4, the feed was burned (750°C) BISO fertile carbide particles. The solids were dumped into the solution in one mass and, although a vigorous reaction occurred, most of the solids settled at the bottom and the air-lift was unable to force solution through it. The sludge was removed and fed back continuously as a slurry. All the solids were dissolved in 58 min (see Table 5.7). The solids dissolved so vigorously that the banging of collapsing steam bubbles could be heard. This phenomenon necessitated restricting the rate of solids addition to prevent the leacher from breaking. It was concluded that the dissolution rate capability was greater than the solids addition rate.

In Run 1L5, solids addition was batch continuous and each batch was of known weight. Each batch was added when the previous batch was observed to have dissolved, i.e., when the solution became transparent. The results are plotted on Fig. 5.17. It was seen, after the addition of a batch, that a large fraction of the solids dissolved very quickly, whereas a small fraction of the larger particulates took longer. Each solids batch addition was about 12 g, so if the average size of heel in the leacher is taken to be 6 g, then the rate of dissolution is $1.4 \text{ g ThO}_2/\text{min}$ per gram of heel. It was concluded that the dissolution rate of carbide fertile particles burned at 750°C is fairly high.

Fig. 5.17. Dissolution of ThO₂ in Thorex liquor at 120°C, Run 1L5, leacher No. 1; ThO₂ obtained by burning BISO fertile particles at 750°C

Run 1L6 was a repeat of 1L5, but the BISO carbide fertile particles were burned at 850°C. The results are shown in Fig. 5.18. In a similar calculation as in 1L7, the maximum dissolution rate is 1.9 g ThO₂/min per gram of heel. It was concluded that the burning temperatures of the BISO particles does not have a significant effect on the dissolution rate.

Run 1L7 was operated in a similar manner to 1L6, but the feed was sintered ThO₂ spheres produced by the sol-gel process at ORNL. The results are shown in Fig. 5.19. The high density of the spheres caused a high pressure drop across the fluidized bed at the bottom of the leacher, and the air-lift pump was unable to force solution through at the required rate. The calculated rate of dissolution (0.03 g ThO₂/min per gram of heel) is therefore lower than it otherwise would have been. It was concluded that burned carbide particles dissolve more rapidly than oxide particles because the burned carbide material has a high specific surface area compared with sintered oxide spheres.

Run 1L8 was done to observe the effect of the fluoride ion and the effect of Th concentration on the dissolution rate. The leacher was charged with one liter of Thorex liquor, without the HF being added. The temperature was raised to 100°C, the air-lift was started, and the first batch of ThO₂ was added. No reaction occurred for 57 min. The temperature was raised to about 120°C, the HF was introduced, and further batches of solids were added as the previous batch was consumed. The results are shown in Fig. 5.20. When the concentration reached 320 g ThO₂/l, a sample of solution was cooled to find the crystallization temperature; it was 25°C. This was done again at 428 g ThO₂/l, and the crystallization temperature was found to be 52°C. It can be seen that the rate of reaction falls with increasing concentration, but that between 0 and 264 g ThO₂/l, the rate remains fairly high. The dissolution rate after the introduction of HF increases dramatically. The crystallization temperatures at the two concentrations show that there is a fair degree of freedom in using a flowsheet concentration of 264 g ThO₂/l in that the solution is far from depositing crystals that could lead to blocked piping.

Fig. 5.18. Dissolution of ThO_2 in Thorex liquor at 120°C, Run 1L6; ThO_2 obtained by burning BISO fertile particles at 850°C

Fig. 5.19. Dissolution of sol-gel ThO₂ spheres in Thorex liquor, Run 1L7

Fig. 5.20. Effect of HF and solution concentration on the rate of dissolution of ThO₂ in Thorex liquor; ThO₂ obtained by burning BISO fertile particles at 850°C

Leacher 1 was retired and leacher 2 was brought into service. It was continuously operated with fresh feed of both hot Thorex liquor and ThO_2 . The product was allowed to spill through the top cock. The solids were fed down a tube to a point near the bottom of the wide chamber. If the solids had been added to the top of the solution, some undissolved material would have been collected in the product. Leacher 2 is shown in Fig. 5.21.

A trial experiment showed that when ThO_2 was fed down the long tube, blockages were readily caused. The solids were therefore mixed with nitric acid and fed as a slurry.

From Run 2L1 onward, the ThO_2 used was that supplied for the manufacture of ThC_2 kernels. It was considered that operation of the leacher would be very expensive if burned particles were used continually.

Operation was as follows: one liter of water was poured into the leacher and heated to 110°C , and nitric acid from a head tank was piped through a furnace and heated to about 80°C . ThO_2 slurry was introduced down the feed tube and the solution became cloudy at the bottom. Both feeds were maintained so that only clear solution spilled over as product.

Run 2L2 was carried out and the results are shown on Fig. 5.22. Calculations from the curves indicate that the final ThO_2 rate was 7.8 g/min and the solution flow rate was 12 ml/min. These conditions produced a very concentrated solution of 650 g ThO_2 /l at the operating temperature of about 115°C . It was concluded that because no obvious difficulty was experienced operating under these conditions, the production of solution with a concentration of 264 g ThO_2 /l should be relatively easy.

Run 2L3 was carried out to operate for a longer period of time, i.e., 150 min, and the results are shown in Fig. 5.23. The average concentration of the product was 273 g ThO_2 /l; this was produced at an average rate of 22 ml/min. It was observed that if the solids feed was interrupted, the flow of nitric acid into the bottom of the reactor would mix with the solution

fig. 5.21. Leacher No. 2

Fig. 5.22. Dissolution of ThO_2 in Thorex liquor, Run 2L2; ThO_2 purchased for ThC_2 production and is calcined from Th oxalate; average particle size, $1\ \mu\text{m}$

Fig. 5.23. ThO_2 dissolution in Thorex liquor, Run 2L3; ThO_2 from oxalate calcination

at the bottom and reduce the density to less than the solution higher up in the leacher. This resulted in a spontaneous reversal of position. The heavier solution would flow to the bottom and the lighter to the top. If the lighter one contained slurry, slurry would be carried into the product. It was concluded from this run, that a once-through nonstirred reactor was of no practical use, so leacher 3 was designed and ordered.

Run 2L4 was almost identical to run 2L3, except that the flow rate was increased. The result showed that the apparatus was capable of producing solution at 35 ml/min at a concentration of 345 g ThO_2 /l.

Run 2L5 was an attempt to dissolve sol-gel ThO_2 spheres. The attempt failed because the Thorex liquor head was insufficient to fluidize the dense slurry.

In order to determine whether the production rate in run 2L4 was near the maximum for the leacher, run 2L6 was carried out. A product rate of 66 ml/min was maintained for 24 min, the average concentration being 272 g ThO_2 /l. Even under these conditions, the slurry had dissolved well below the level of the spillover cock. It was concluded that if irradiated fuel has a dissolution rate similar to this ThO_2 , the commercial leacher size would not be unduly large.

Leacher 2 was retired and leacher 3 (shown in Fig. 5.24) was installed. It was a two-stage, continuous type that recycles the solution in the first stage. Operation was as follows: 1 liter of Thorex liquor was charged into the first stage, the temperature raised to about 115°C, and the air-lift pump started. A slurry of nitric acid and ThO_2 was prepared (about 140 g ThO_2 /80 ml nitric acid) and fed at the appropriate rate into the recycle solution. Thorex liquor was fed at the appropriate rate from a head tank, preheated, and introduced below the recycle injection point. It was seen that the slurry filled the volume of the recycled solution, and the volume above the air-lift pump intake was clear solution. The interface between the slurry and clear solution was quite sharp. The clear solution spilled

Fig. 5.24. Leacher No. 3

over into the second stage which was also provided with a heater to maintain the solution at about 115°C. Product spilled over the second stage.

Run 3L1 was started with the first stage temperature at 75°C and continued for 55 min. The Thorex liquor flow rate was maintained at 25 ml/min. The temperature was then raised to 110°C, and the run continued for another 70 min. The results are shown on Fig. 5.25. Although the product was a clear solution at all times when operating at 75°C, slurry was carried into stage 2. During operation at 110°C, the slurry was always at the bottom of the first stage. The final solution concentration was 287 g ThO_2 /l, and the density was 1.65 g/ml. It was concluded that it is advantageous to operate at a temperature of about 110°C.

Run 3L2 was carried out at 110°C, and the acid feed rate was maintained at 25 ml/min. The slurry feed was prepared with ThO_2 and water (130 g ThO_2 /100 ml water) and fed at a uniform rate. The final solution concentration was 250 g ThO_2 /l, and the rate was calculated to be 12.8 g ThO_2 /min. It was concluded from runs 3L1 and 3L2 that, with two stages, the leacher will produce clear product solution at 110°C, at a feed rate of about 12 g ThO_2 /min and can be used to test-dissolve product from the burner. Also, the concentration of acid in the Thorex liquor could obviously be reduced.

A stirred leacher is being constructed with the objective of having a back-up facility in case leacher 3 should prove to be undesirable when further experiments are performed.

A summary of the conclusions drawn to date are as follows:

1. Carbon or carbide causes NO_2 and brown solutions.
2. Reaction heat evolution can cause boiling.
3. ThO_2 derived from burned carbide kernels is extremely reactive.

Fig. 5.25. ThO₂ dissolution in Thorex liquor, Run 3L1; ThO₂ from oxalate calcination

4. ThO_2 produced by burning carbide kernels at 850°C appears to have about the same high reactivity as that burned at 750°C .
5. Sol-gel, as may be expected, has a very much lower reaction rate than burned carbide.
6. The dissolution rate of the reactive burned carbide material is very low if the acid does not contain fluoride ions.
7. The desired concentration of 264 g ThO_2 /l is well below the crystallization concentration at 25°C .
8. Solids fed to the leacher caused blockages in the feed line. A slurry feed eliminated this difficulty.
9. A once-through nonstirred leacher suffered from the disadvantage that spontaneous stirring could occur and the product thus become contaminated with solids.
10. It is advantageous to operate at about 110°C . At this temperature, the reaction rate is high with virtually no condensate. At 75°C the reaction rate is noticeably lower.
11. The slurry feed can be prepared using water without any obvious change in dissolution rate in the acid.
12. The acid concentration in the Thorex liquor could be reduced and still give acceptable dissolution rates.

The most important factors still to be investigated are:

1. The effect of burnup and temperature on dissolution rate.
2. The effect of carbon fines in the feed.

3. The efficiency with which U and Th are leached from the insoluble residue.
4. The reaction rate of the most unreactive feed material.
5. The effect of nitric acid concentration.
6. The effect of temperature.

Task VIII
PHYSICS AND FUEL MANAGEMENT

HTLTR SUPPORT ACTIVITIES

Two sets of basic calculations for HTGR Lattice 1 ($C/Th=200$ and $C/U^{235}=6000$) in the HTLTR have been completed. The first set was completed with the cross sections and methods used in the reference HTGR design library. The second set was completed with the latest ENDF/B cross-section data and updated version of GGC-5. Calculations were performed at 300° , 600° , 900° , 1200° , and $1500^\circ K$; and $K_\infty(T)$, the Doppler coefficient, and the temperature coefficient were determined. The experimental data from this lattice are not yet available.

Measurements on HTGR Lattice 2 ($C/Th=200$, $C/U^{233}=1100$) in the HTLTR have been completed and analysis of the data is under way. GGC-5 calculations as a function of temperature are now being performed using the latest ENDF/B cross sections. These results will be compared to the experimental data for this lattice when it becomes available.

Battelle has started loading HTGR Lattice 3 ($C/Th=300$, U^{233}). Preliminary GGC-4 calculations for several U^{233} -Th loadings have been sent to Battelle to aid them in their plans for this lattice. Plans are under way to calculate the nuclear behavior of this lattice as a function of temperature and compare the results with the experimental data when they become available.

Some additional calculations have been performed to determine the effects of various options and definitions in the latest version of GGC-5 on HTGR Lattice 1 at room temperature. Specifically, the effects of the Dancoff factor, the definition of $(\sigma_m)_{eff}$, the GAROL/GAMNIT resonance option, and the inclusion of GAROL U^{238} and U^{235} resonance integrals in the GAM spectrum calculations have been evaluated. Table 8.1 shows the results of representative calculations to date.

Table 8.1
REPRESENTATIVE CALCULATIONS FOR HTGR LATTICE 1

Run	Temp (°K)	RRI (a)	URI (b)	E>2.38 eV			E<2.38 eV	K _∞
				Σ_{a1}	Σ_{s1}	$\nu\Sigma_{f1}$	ηf	
1 (c)	300	30.92	1.91	1.126×10^{-3}	2.710×10^{-3}	0.361×10^{-3}	1.41601	1.0945
2 (d)	300	33.11	1.17	1.141×10^{-3}	2.734×10^{-3}	0.371×10^{-3}	1.41688	1.0955

(a) ${}_{\text{Th}}^{232}$ resolved resonance integral.

(b) ${}_{\text{Th}}^{232}$ unresolved resonance integral.

(c) Run 1 analysis using HTGR reference cross sections and methods.

(d) Run 2 analysis using latest ENDF/B cross sections.

FUEL TEST ELEMENT

Design work on the fuel test element for irradiation in the Peach Bottom reactor was completed during the quarter. The element is designed to be irradiated for three years and to subsequently receive postirradiation examination at Gulf General Atomic.

The major purpose of this element is to evaluate the performance of advanced fuels over a wide range of operating conditions and lifetimes. The time-at-temperature lifetimes of various types of fuel will be determined, and data will be obtained to identify causes of particle failure in order to improve fuel performance and lifetimes. The element will also make possible controlled irradiation experiments on:

1. Fuel particle thermal stability.
2. Metallic diffusion rates in an HTGR environment.
3. Fission-product release of various types of fuel particles.
4. Boronated graphite properties.
5. Various fuel rod fabrication concepts.

In addition, the long-term in-pile performance of bonded rods under HTGR conditions will be evaluated.

The general design of the element was described in detail in the previous quarterly progress report (GA-9944). Figures 8.1 and 8.2 show the fuel test element assembly and fuel body details. A more extensive discussion of the thermal and mechanical design will be covered in the following sections. A description of the fuel and spine samples is also included.

Thermal Analysis

Calculational Methods and Physical Property Data. The thermal performance calculations reported here were carried out with the use of general purpose one- and two-dimensional heat-transfer computer codes. TAC-2D, a two-dimensional

Fig. 8.1. Fuel test element assembly

Fig. 8.2. Fuel body details

heat-transfer code, was used to analyze radial temperature distributions of the test element in selected axial planes. COPE, a multichannel flow and thermal analysis code, was used for the analysis of axial temperature profiles along the length of the element. In these codes material properties that are a function of temperature are evaluated locally for each spatial node point within the analytical model. Also, local film and bulk fluid temperatures are evaluated and used to compute temperature-dependent fluid properties, Reynolds numbers, and heat-transfer coefficients.

The average heat-transfer coefficient correlation used here is the same as used in the Core 2 fuel element design. Thermal conductivity values of each material used in the calculations are shown in Table 8.2.

Table 8.2
MATERIAL THERMAL CONDUCTIVITY VALUES

Material	K (Btu/hr-ft-°F)
Helium	$1.29 \times 10^{-3} T^{0.674} (T \text{ in } ^\circ R)^a$
Sleeve graphite	16 (perpendicular) ^a
Fuel body graphite	16 (perpendicular)
Bonded rod fuel	4.0

^aSame values used for the Core 2 fuel elements.

Temperature Analysis. Table 8.3 summarizes the Core 2 power and flow conditions used as the basis for the thermal analysis of the test elements.

The maximum power of the test element is 124 kw(t), which is 88% of an average Core 2 fuel element (140 kw(t)) and 67% of the end-of-life peak power in the hottest Core 2 fuel element (186 kw(t)).

The fully enriched uranium loading in the test element is 216 g, which is 87% of an inner (Type 1, 2, or 3) Core 2 fuel element (249.6 g).

Table 8.3

CORE 2 POWER AND COOLANT FLOW CONDITIONS

<u>Parameter</u>	<u>Value</u>
Thermal Power, MW:	
Generated in fuel elements	112.65
Generated in reactor internals other than core	<u>2.85</u>
Total.....	115.50
Reactor Coolant Flow, lb/hr:	
Total active core and bypass flow	474,000
Through active core tricuspid channels	426,600
Miscellaneous bypass flows	47,400
Coolant Temperatures, °F:	
Reactor vessel inlet	633
Reactor vessel outlet	1302
Core inlet	652
Core outlet:	
Average tricuspid	1382
Maximum tricuspid	1700
Core Pressure Loss, psi	2.7
Core Position (0-900 EFPD)	C02-01

The actual power produced in a test element is determined by the test element uranium loading and the radial peaking factor of the six surrounding Core 2 elements. For example, an element containing the maximum test element loading of 216 g of U would have to be placed in a core location where the radial power peaking factor is 0.99, to produce the maximum 124 kw(t) of power. Test elements containing a lower uranium loading could be placed in core locations having a higher average radial power peaking factor to generate the same test element power.

The maximum fuel temperature in the test elements is not a design criteria for the element, since it is desirable to irradiate fuel beds at various time-temperature conditions to determine the limits of successful fuel bed performance. The expected maximum fuel bed temperature for the test elements is about 2200°F (at 95% power and 105% coolant flow). The test element will be placed in a core location where the fuel temperature will remain relatively constant throughout their irradiation.

Figure 8.3 shows the axial temperature distribution. Figure 8.4 shows the radial temperature distribution through the same element at the plane of maximum fuel temperature.

Hot-spot Analysis. The temperatures of the test element will deviate from the calculated temperatures if the pertinent parameters of the element have different values from those used in the calculations. Such differences arise from uncertainty in material property values and from dimensional manufacturing tolerances. Dimensional variations causing temperature deviations are: Sleeve inside diameter variation, fuel body outside diameter variations, and eccentric location of the body in the sleeve. These variations result in deviations in the clearance between the body and the sleeve. Temperature deviations are also caused by variations in fuel rod and fuel hole diameters, power deviations from tolerances in uranium concentrations, and deviations in the radial and axial power factors. Uncertainties in material properties include radiation-induced dimensional changes in graphite parts, which result in dimensional changes of clearances and irradiation damage effects, which cause variations in graphite and bed thermal conductivities.

Fig. 8.3. Axial temperature profile at five full-power days for fuel test element, 95% power, 105% flow

Fig. 8.4. Radial temperature through fuel test element at plane of maximum fuel temperature

An estimate of the deviation in maximum fuel temperature resulting from any of the individual variations is listed in Table 8.4. The individual deviations lead to a total root-sum-square temperature variation of 280°F in the maximum fuel temperature at design conditions.

Mechanical Performance

Core-2 Components. The sleeve, bottom connector, and upper reflector will perform in the same manner as a Core 2 fuel element.

Fuel Body Dimensional Changes. The fuel bodies are machined from extruded logs of anisotropic H-327 needle coke graphite with the long axis of the fuel body parallel to the extrusion axis of the graphite log. During core operation, the graphite will be exposed to fast-neutron irradiation ($E > 0.18$ MeV), which will induce dimensional changes in the graphite. Due to the anisotropy of the graphite material, dimensional changes in the direction parallel to the extrusion axis will be greater than those in the direction perpendicular to the extrusion axis. For the temperature and fast fluence range of the fuel bodies up to 900 equivalent full-power days (EFPD), the irradiation-induced dimensional changes will result in a net contraction of the graphite fuel bodies. In no case is irradiation-induced expansion predicted.

The fuel body dimensional change behavior is dominated by the irradiation-induced graphite contraction and results in a net contraction in both the axial and radial directions. After 900 EFPD, corresponding to a maximum fast fluence greater than 4.5×10^{21} n/cm², the maximum fuel body contractions are approximately 2.6% axially and 0.7% radially at the locations of highest fast fluence and temperature. The entire fuel body experiences an axial contraction averaging about 2.1% over the whole 31-in. length in 900 EFPD. Thus, the length of the 31-in. fuel body will decrease by up to 0.7 in. in 900 EFPD. Similarly, the outer diameter of the fuel body will shrink a maximum of 0.017 in. in 900 EFPD. An individual fuel hole will decrease about 0.003 in. in diameter in 900 EFPD.

Table 8.4
FUEL TEST ELEMENT PEAK FUEL TEMPERATURE INCREASE RESULTING
FROM INDIVIDUALLY APPLIED HOT-SPOT FACTORS AT
POINT OF MAXIMUM FUEL TEMPERATURE
(End of Life)

	Temp (°C)
Manufacturing tolerances:	
Sleeve ID (+0.002 in.)	65
Body OD (-0.002 in.)	65
Body eccentricity (0.007 in. local gap increase).....	220
Fuel rod diameter (-0.001 in.) ^(a)	15
Fuel rod hole (+0.001 in.) ^(a)	15
Local uranium concentration (+25%)	50
Material properties (measurement technique error):	
Graphite thermal conductivity (-10%)	70
Fuel bed conductivity (-10%)	25
Irradiation-induced changes in graphite dimensions (0.002-in. sleeve gap increase)	65
Calculational variables	
Axial power factor (+5%)	10
Radial power factor (+5%)	95
Circumferential variation of film coefficient	<u>10</u>
Total root-sum-square deviation (from all of the above)	280

^(a) For fuel rods only.

Sleeve/Fuel Body Gap. The thermal expansion in the H-327 graphite fuel body is larger than the thermal expansion in the HLM-85 graphite sleeve because of the different graphite thermal expansion properties and lower sleeve temperatures. This reduces the sleeve/fuel body beginning-of-life shutdown diametral gap of 0.013 to 0.008 in. at operating conditions.

The fuel body contracts a little faster than the sleeve owing to fast-neutron irradiation up to a fast fluence of about 1.5×10^{21} n/cm². After this fluence they both contract at approximately the same linear rate, leading to a slight decrease in the sleeve body gap. Diametral clearance versus fast fluence is plotted in Fig. 8.5, for both operating and shutdown conditions and is also presented in Table 8.5.

Fuel Body/Center Sample Clearance. The center hole will contain irradiation samples enclosed in a graphite container or a niobium can. In all cases the outer diameter will be a maximum of 1.00 in., leaving a minimum 0.095-in. diametral clearance between the fuel body and the spine or container. This clearance is equivalent to 9% of the center hole diameter, and is more than adequate clearance to accommodate any unexpected differential radial dimensional changes between the fuel body and center hole graphite.

The minimum axial clearance between the stack of center hole crucibles or spines and the bottom of the threaded plug at the top of the center hole is 0.50 in. This clearance is equivalent to about 1.7% of the 31.0-in. body length and about 3.4% of the 15.5-in. body length.

The fuel body, spine, and sample containers are all fabricated from H-327 graphite and have the same orientation with respect to the extrusion axis. The fuel body and center graphite will have the same fast-fluence history, and the temperature of the center hole graphite will be the same as, or higher than, the fuel body center hole wall temperature. Therefore, the radial and axial graphite contraction of the center hole graphite will be the same as, or greater than, the fuel body and the beginning-of-life radial and axial minimum clearances will therefore remain approximately the same.

Fig. 8.5. Diametral clearance between sleeve and fuel body versus fast fluence

Table 8.5
SLEEVE/FUEL BODY CLEARANCE

Fast Fluence (n/cm ²)	Nominal Diametral Clearance (in.)	
	Operating Condition	Shutdown Condition
BOL	0.008±0.003	0.013±0.003
1x10 ²¹	0.010±0.003	0.015±0.003
2x10 ²¹	0.010±0.003	0.015±0.003
3x10 ²¹	0.009±0.003	0.014±0.003
4x10 ²¹	0.008±0.003	0.013±0.003
5x10 ²¹	0.007±0.003	0.012±0.003

Fast fluences for the 900 EFPD lifetime will not be sufficient to produce any contraction turn-around in the center hole graphite.

Fuel Body and Sleeve Stresses. Analyses of the stresses in the fuel bodies and sleeves have been made for fast fluences up to 5×10^{21} n/cm², which corresponds to more than 900 EFPD. Stresses in these graphite components can arise in the following ways:

1. Stresses that result from external loading, such as a load that one component might apply to a neighboring component.
2. Internal stresses in the graphite structure owing to differential thermal expansion, differential irradiation-induced dimensional changes, and possible fuel bed/graphite interactions.

The test element is so designed that a gap always exists between the sleeve and fuel body. Thus, no interaction between the sleeve and fuel body will occur that would result in a mutual application of load.

The maximum combined thermal and irradiation-induced contraction axial and in-plane fuel body stresses for full-power operation and reactor shutdown are shown in Table 8.6. A plot of the local maximum and body-average maximum axial stresses versus fast fluence throughout core life is

shown in Fig. 8.6. Figures 8.7 and 8.8 show axial stress distributions across the fuel body at maximum stress conditions (fast fluence = 3.5×10^{21} n/cm²) for operation and shutdown, respectively.

The maximum local axial and in-plane tensile stresses in the graphite body shown in Table 8.6 are well below the measured tensile strength of the H-327 graphite. When the reactor is shut down, the reverse thermal strains add to the current operational contraction strains and produce the maximum stresses in the fuel body at this time. When the reactor returns to full operation, the thermal strains are relieved and the stresses are again reduced.

Table 8.6
FUEL BODY INTERNAL STRESSES

Direction	Time	Local Maximum Tensile Stress (psi)
Axial	Operation	530
Axial	Shutdown (thermal)	770
Axial	Shutdown (total)	1300
In-plane	Operation	230
In-plane	Shutdown (total)	270

The maximum shutdown tensile stresses occur away from the ends of the fuel body and are located at the point of maximum power density along the length of the body. At all planes where the power density (or linear heat rating) is less, the stresses are also less. At reactor shutdown, the reverse thermal strain produces a maximum axial thermal stress shutdown component of 770 psi. The maximum axial shutdown stress (1300 psi) is the sum of the maximum operating axial stress and the maximum axial shutdown thermal stress. The thermal shutdown stresses are based on an elastic analysis, and thus have only a small uncertainty of 5% maximum. The operating stress is conservatively calculated using the SAFE-GRAFITE code and has a maximum uncertainty of 25%. The sum of the uncertainties could contribute an

Fig. 8.6. Maximum axial tensile stresses in graphite fuel body versus fast fluence

Fig. 8.7. Axial stress distribution maximum operating case; fast fluence, $3.5 \times 10^{20} \text{ n/cm}^2$

Fig. 8.8. Axial stress distribution maximum shutdown case; fast fluence, $3.5 \times 10^{20} \text{ n/cm}^2$

additional 170 psi to the nominal maximum value. Consequently, the maximum axial tensile shutdown stress is about 1300 ± 170 psi compared to the mean axial tensile strength of the graphite of about 2000 ± 350 psi. The maximum in-plane shutdown stress of 270 psi is also far less than the mean transverse tensile strength of about 1000 ± 120 psi.

Fuel Beds

The loadings of the fuel beds in the fuel test element, which are different from those previously planned, are shown in Fig. 8.9. Each bed will be loaded with fuel rods 0.490 in. in diameter and 2 in. long, and the combinations used are relevant to large HTGRs. All the fuel rods will be made by hot-injection molding in production equipment, and will be bonded with a matrix of natural flake graphite plus pitch.

The distribution of the fuel rods between the eight holes is also shown in Fig. 8.10. All three bodies will be identical, and the loading of fissile and fertile particles in each rod type is dictated by the over-all loading of 216 g of U. After irradiation, representative samples of each of the types of fuel rods will be examined visually and metallographically to determine the effect of irradiation dose and temperature.

The maximum burnups achieved will be 20% FIMA in the 2:1 (Th,U)C fissile particles and 60% FIMA in the buffer-diluted UC fissile particles. The maximum fast fluence at the end of three years should be 4.2×10^{21} n/cm² --about half that to be accumulated during the core lifetime of a large HTGR. Maximum fuel bed temperatures will be 2200°F.

Central Irradiation Samples

Experimental samples will be contained in the central 1.10-in.-diameter hole of the element. In general, these samples can be divided into five types:

UPPER END

BODY
NO. 3

BODY
NO. 2

BODY
NO. 1

ALL BODIES IDENTICAL

N.F. = NATURAL FLAKE FILLER + PITCH BINDER

Fig. 8.9. Fuel rod loadings in outer beds of fuel test element

Fig. 8.10. Distribution of spine samples in fuel test element

1. Thermal stability (fuel particles).
2. Fission-product release (fuel particles).
3. Metallic diffusion.
4. Fuel rods.
5. Boronated graphite.

The designs of the specimens are different, as shown in Figs. 8.11 through 8.17, but they will all be contained within cylindrical H-327 graphite crucibles having sufficient clearance not to interfere with the graphite body at any time during irradiation. By using these crucibles, all the center samples are in effect triply contained, and the more critical experiments (thermal stability and fission-product release) employ smaller crucibles inside these containments. The purpose and description of each type of experimental sample has been listed in the previous quarterly report (GA-9944). Also, the planned postirradiation examination of each sample type has been reported previously. The distributions of the spine samples in the various fuel bodies are shown diagrammatically in Fig. 8.10. These positions have been selected based on the temperature and fluence requirements for the different experiments.

LC81077

Fig. 8.11. Design of Thermal Stability Spine Samples (type 1)

Fig. 8.12. Thermal stability center hole sample, alternative design

LC81083

Fig. 8.13. Design of fission-product release spine samples

LC81084

Fig. 8.14. Design of alternative fission-product release spine samples

LC81085

Fig. 8.15. Design of diffusion spine samples

Fig. 8.16. Fuel rod center hole sample

Fig. 8.17. Boronated graphite center sample

Task IX
ADVANCED COATED PARTICLE FUELS

COATED PARTICLE IRRADIATIONS

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

Capsule P13L

Capsule P13L is the second test of coated diluent particles and the first to full exposure. Both carbide and oxide diluent and fertile particles are being tested, with 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 are of the buffer-diluted design. Some of the samples are 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 in this series (GA-9227 and GA-9815).

The capsule has completed its fifth cycle of irradiation in the ETR. Most samples in the high-temperature cells continue to operate considerably below their design temperatures due to fuel depletion (along with thermocouple decalibration). Temperatures in the 1500°C cell (cell 1) are 885° to 1090°C; and temperatures in the 1300°C cells (cells 2 through 4) are 735° to 1065°C. Samples in the 900°C cell (cell 5), however, are close to design temperature. The capsule has received an estimated fast fluence of $7.0 \times 10^{21} \text{ n/cm}^2$ ($E > 0.18 \text{ MeV}$) to date, with estimated burnups of 75% and 25%

for the UC_2 and $(Th,U)C_2$ particles, respectively. The capsule is scheduled to be irradiated for seven cycles in the ETR to a fast fluence of 8×10^{21} n/cm² and a burnup of 75% FIMA for the diluent particles.

Capsule P13M

Capsule P13M will be the second full-exposure test of coated diluent particles, and the first test of this type fuel made in production equipment. Prototype coated oxide recycle fuel is also included. Many of the samples will be tested in pitch-bonded fuel rods to properly evaluate the full range of interactive stresses experienced by initial and recycle core fuel for a large HTGR. Many of the samples tested will also be duplicates of those in the fuel test elements (see Task VIII), and the P13M capsule will provide full exposure data on these important materials. (The maximum exposure received in the test elements will be 4.2×10^{21} n/cm² after 900 EFPD.)

The P13M samples will be irradiated at 1500°, 1350°, and 1000°C to burnups of 75% FIMA for diluent fissile particles, 20% FIMA for recycle fissile particles, and 7% FIMA for fertile particles (full HTGR burnups for all three types of particles). The design fast fluence is 8×10^{21} n/cm² (full HTGR exposure).

Capsule fabrication and sample preparation are proceeding for insertion in ETR.

Task XI
GRAPHITE RESEARCH

CARBON COATING IRRADIATION

Irradiations of pure and silicon-alloyed isotropic carbons (deposited below 1500°C) at 900° to 1300°C to fast-neutron fluences in the range 1.2 to 4.2×10^{21} n/cm² ($E > 0.18$ MeV) show that silicon concentration above about 18 wt-% are effective in reducing the irradiation-induced dimensional changes. For example, at 900°C the lineal shrinkages caused by irradiation are reduced about a factor of three by an addition of 34 wt-% silicon. At 1250° to 1300°C the corresponding reduction is about a factor of two. At 900°C, irradiation creep strains as high as 6% per 10^{21} n/cm² were measured for the unalloyed carbons. At 1250° to 1300°C total creep strains of 10% were observed for unalloyed carbons, and 8% were observed for the alloyed coatings. Since none of the creep specimens fractured, these are not limiting values.

Comparison of the data for isotropic carbons deposited below 1500°C (low-temperature isotropic carbons) with those for isotropic carbons deposited above 1500°C (high-temperature isotropic carbons) show that at high fluences the LTI carbons expand at a rate that is substantially less than the corresponding rate for HTI carbons.

Several significant conclusions can be drawn:

1. Silicon co-deposited with isotropic carbon at temperatures below about 1500°C enhances the dimensional stability during irradiation at 900° to 1300°C when the concentration is above about 18 wt-% Si. For lower silicon concentrations the effects are small.
2. During irradiation in the range 900° to 1300°C, low-temperature isotropic carbons expand less rapidly at high fluences than high-temperature isotropic carbons.

3. At 900°C, creep strains as high as 6% per 10^{21} n/cm² can be sustained in unalloyed LTI carbons without fracture. This value is higher than values reported previously for either LTI or HTI carbons. Since none of the restrained coatings fractured, this is not a limiting value.
4. At irradiation temperatures of 1250° to 1300°C, total creep strains of at least 10% can be accumulated in unalloyed LTI carbon; the corresponding value for alloyed carbons is 8%. Since none of the creep specimens fractured, these values are also not limiting.

A more detailed discussion of these findings may be found in a topical report that has been prepared on this subject (Ref. 1).

CONTROL MATERIALS IRRADIATION

Capsule BG-2

The primary purpose of the BG-2 experiments (GA-19-2) is to measure the effects of neutron irradiation on boronated (B or B₄C plus graphite) and hafnated (HfC plus graphite) compacts. Dimensional changes in boron fibers and density changes in loose granules of B₄C and HfC will also be determined in piggyback type samples to enhance knowledge of irradiation effects in these poison materials in the absence of a bonding matrix material. A better understanding of irradiation effects in boronated or hafnated graphite should result from this investigation.

Irradiation-induced dimensional change is the phenomenon of primary interest; however, there will also be correlation of thermal expansion coefficient, strength, and microstructure with neutron exposure. Some specimens irradiated previously in capsule BG-1 (reported in earlier quarterly report GA-9372) continue irradiation exposure in BG-2. A detailed summary of samples being tested in BG-2 was given in earlier quarterly report GA-9660.

The capsule was inserted in the ETR and started irradiation in July 1969 at temperatures of 600° to 700°C.

During this report period, BG-2 completed its four-cycle irradiation in the ETR and was discharged. The capsule received an estimated fast fluence of about 5.5×10^{21} n/cm² (E > 0.18 MeV).

References

1. Bokros, J. C., D. W. Stevens, and R. J. Akins, "Irradiation-induced Dimensional Changes in Co-deposited Carbon-Silicon Coatings," Gulf General Atomic report GA-10000, to be submitted for publication in J. Am. Ceram. Soc.