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GAS-COOLED REACTOR PROGRAMS

HIGH-TEMPERATURE GAS-COOLED REACTOR BASE-TECHNOLOGY PROGRAM
ANNUAL PROGRESS REPORT FOR PERIOD ENDING DECEMBER 31, 1979

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SUMMARY

1. HTGR CHEMISTRY

Effect of Steam Corrosion on the Strength of Graphite

Samples of graphite grades H451, Stackpole 2020, and purified Stackpole 2020 have been corroded in a wide range of helium-steam-corrosion product mixtures at 900 and 1000°C. Both rod [12.77- and 19-mm-diam (1/2- and 3/4-in.)] and brittle-ring specimens are being used. Burnoffs ranged between 2 and 18% in corrosion campaigns of from one to four weeks in duration. Purified Stackpole 2020 was tested because large inclusions of calcium (a catalytic agent) were observed in the as-received material. Determination of strength degradation as a function of burnoff for the brittle-ring samples is in its preliminary stage. Studies on effect of burnoff on compressive strength, internal surface area, and pore-size distribution will begin soon.

Iodine Sorption and Desorption

Iodine sorption and desorption tests are being conducted on a low-alloy steel (alloy T-22) and on grade H451 graphite. Iodine concentrations and specimen temperatures in these tests have thus far ranged from 10^{-7} to 10^{-11} mol fraction and from 400 to 600°C. Our intention is to go as low as possible in iodine concentration; however, the estimated lower concentration limit of 10^{-12} mol fraction has not yet been achieved.

Diffusion of Fission Products on Graphite

Recovery has been completed of the data on diffusion of cesium in graphite from experiments performed by the late R. B. Evans III. His notebooks have been scanned, and diffusion data on five graphite grades are being published.

Experiments on the rate of silver diffusion in H451 graphite are in progress under subcontract with the University of Virginia. Thus far, interpretation of concentration profiles indicate a value of the diffusion coefficient between 2×10^{-5} cm²/s at 700°C and 6×10^{-7} cm²/s at 500°C.

Carbon-14 Production and Distribution in the Peach Bottom HTGR

An analysis of ¹⁴C levels in Peach Bottom HTGR core components has been completed and is being published.

2. FUELED GRAPHITE DEVELOPMENT

The Fueled Graphite Development Program at ORNL continues as part of a national HTGR fuel development effort being conducted in cooperation with the General Atomic Company (GA). The ORNL portion of the program centers around irradiation testing and specialized postirradiation examination procedures. GA's portion of the program includes interface with core designers, responsibility for the fuel-performance model development, and interface with the licensing activity.

The major focus of the HTGR fuel development program for the past two years has been qualification of fuel for the medium-enriched uranium (MEU) fuel cycle. The first irradiation capsules containing MEU fuel (HRB-14 and HRB-15b) completed irradiation in January 1979, and a major portion of the work during this reporting period was associated with examination of fuel from those capsules.

In addition to the cooperation with GA under the national HTGR fuel development program, a cooperation with the German Nuclear Research Center at Jülich (Kernforschungsanlage Jülich) is in progress. The cooperation with Kernforschungsanlage (KFA) is part of the formal agreement between the U.S. and German governments for gas-cooled reactor development. Areas of work under the ORNL/KFA cooperation include fuel performance assessment, postirradiation examination techniques and procedures, and coating development and characterization. The ORNL/KFA cooperation includes an exchange of technical personnel. A report of work performed during this reporting period by the ORNL assignee to KFA is included in Chap. 2 of this document.

The Fueled Graphite Development Program continues to be organized around five subtasks: (1) Irradiation Testing, (2) Development Work, (3) Irradiation Support Activities, (4) Project Work Statement Activities (ORNL/KFA Cooperative Program), and (5) Equipment Development and Support Activities. Highlights from this reporting period are listed below:

- Completion and documentation of studies on PyC permeability.
- Development of a two-step process for the fabrication of dense UCO kernels, which permits control of the kernel density and of phase composition at sintering temperatures in the range of 1550°C. This development is significant because work being done elsewhere features sintering temperatures as high as 1900°C.
- Completion of PyC coating studies in large-scale equipment by using CO₂ in the diluent gas. Processes have been developed that produce coatings of high density and low BAF₀ (optical Bacon anisotropy factor) in production-scale equipment by using propylene diluted with CO₂ and argon. Irradiation testing will determine whether or not such coatings match the very favorable performance observed with batch OR 2013-T, which was produced in laboratory-scale coaters by using propylene and CO₂.

- Completion of out-of-reactor thermal-gradient heating studies by using Triso-coated doped kernels. These studies were designed to measure the fission product-SiC attack rates for a variety of fission product species.
- Collection of the first data from the postirradiation gas analyzer (PGA) system. This system was designed to measure the content of fission gases and reaction gases in irradiated coated particles by breaking the particles in an evacuated chamber coupled to a time-of-flight mass spectrometer.
- Completion of irradiated microsphere gamma analyzer (IMGA) analysis on fuel particles irradiated in a number of irradiation experiments.
- Completion of the experimental work with PWS FD-12 and -13 on fuel of German origin.
- Design and installation of an x-radiographic apparatus for use with irradiated coated particles.

3. PRESTRESSED CONCRETE PRESSURE VESSEL DEVELOPMENT

Under the analytical methods task area, techniques are being developed that can be incorporated into existing modern finite-element computer programs to improve the accuracy and efficiency of those codes. Two techniques currently under investigation are an improved constitutive model and a method for efficiently modeling the reinforcing steel contained in multicavity prestressed concrete reactor vessels (PCRVs).

The University of Tennessee, under a subcontract, completed a study in which they compared (1) the results of finite-element analyses of concrete structures with test data and (2) the predicted with the measured responses of homogeneous concrete test specimens subjected to uniaxial, biaxial, and triaxial states of stress. A total of 32 data files were used in the evaluation. Results revealed good agreement between the analysis results and experimental data for concretes of higher strengths, but the agreement was not as good for the lower strength concretes.

Under the liner and penetration studies, a report presenting results on chemical analysis, metallography, and tensile and Charpy impact testing of three plate steels and one forging steel has been completed. Micro-hardness, Charpy impact, and fracture toughness results are presented for the heat-affected zone (HAZ) and weld metal of weldment W-3.

A study to appraise the applicability of grouted tendon prestressing systems to PCRVs was completed. Results obtained indicate that (1) the grouted tendon flexure specimens exhibit improved performance relative to nongrouted members, (2) the polymer-silica and fibrous concrete material systems exhibit potential for applications to PCRVs, (3) the corrosion-inhibiting ability of grout in the environments investigated is equivalent to that of the organic petrolatum-based commercial corrosion inhibitors,

(4) acoustic emission (AE) is capable of monitoring plain concrete and simple concrete structures, and (5) techniques are available for grouting large prestressing tendon systems such as would be encountered in a PCR. V.

A review of candidate material systems that may be potentially cost and performance effective for fabrication of reactor containment structures was completed. As a result of this review fibrous concrete was selected for evaluation. A study has been initiated to test structural models fabricated from fibrous concrete to demonstrate its applicability to PCRVs for effecting reduced construction costs.

A study conducted at the University of Illinois to investigate the strength and behavior of flat end-slabs of cylindrical prestressed concrete nuclear reactor vessels was completed. During the investigation ten small-scale cylindrical vessels with flat end-slabs were tested to failure under internal pressure. The vessels were prestressed circumferentially and longitudinally. Experimental variables were end-slab thickness, size and arrangement of penetrations in the end-slab, and concrete strength. Results indicated that the failure pressures were proportional to end-slab thickness and concrete strength but were relatively insensitive to changes in the amount of cross section removed by penetrations. An axisymmetric nonlinear finite-element model was developed to simulate the behavior of the physical models.

Design of an enclosure for a test system to obtain properties of concrete under multiaxial loadings at both room and elevated temperatures was completed, and construction specifications were developed. Preliminary discussions were held with several potential vendors to discuss problem areas associated with the uniqueness of such a test system. Development of a conceptual design for the test system has been initiated.

Development of the capability to monitor the structural integrity of complex elements has continued through an evaluation of AE techniques. Background data has been obtained on both concrete and steel structural members. Results obtained indicate that (1) for plain concrete specimens loaded to failure in one cycle, there was good correlation between AE and stress-strain curves relative to AE activity and extent of cracking; (2) for specimens cyclically loaded to failure, there is evidence that the Kaiser effect is applicable to concrete; (3) for flexure specimens whose tensile stress-strain relationship is essentially linear to failure, there was an absence of AE activity until just prior to failure; and (4) for the prestressed concrete beams, there was good correlation between AE source location data where only one or possibly two cracks form between transducers, but, as the number of cracks increase, there is a "smearing" of AE source location data.

4. STRUCTURAL MATERIALS

The ORNL work on gas-cooled reactor (GCR) structural materials provides assistance in the national program of development of materials,

materials technology, and materials data in support of design and licensing of advanced GCR systems. Highlights of this year's efforts follow.

Mechanical Properties Subtask

Tensile and creep tests of weldments have demonstrated that the filler metals and welding techniques used for joining Hastelloy X, Incoloy 802, Inconel 617, and HD 556 produce satisfactory weldments. Creep tests on 2 1/4 Cr-1 Mo steel and Hastelloy X base metal have reached times in excess of 25,000 h.

Determination of the tensile, creep, and impact properties of Hastelloy X and Inconel 617 base metal and weldments aged for up to 20,000 h in inert atmospheres and in GCR helium has begun. Fracture ductilities and impact strength are reduced significantly by the aging. There are indications that aging at the highest temperatures in GCR helium is more detrimental than in the inert atmosphere.

Crack propagation studies dealt mostly with Hastelloy X during the past year. We found that the fatigue crack-growth rate in GCR helium is lower than that in air. Fatigue crack growth proceeded at a lower rate in weld metal than in the base metal.

Work on the low-cycle fatigue behavior of 2 1/4 Cr-1 Mo steel was completed and reported. Continuous cycling fatigue tests of both solution-treated and aged specimens of Hastelloy X were initiated. Thermal aging reduces cyclic life slightly.

Literature on the creep and stress relaxation behavior of structural ceramics was reviewed. Facilities for mechanical testing of ceramics were designed and constructed.

Corrosion Subtask

The carburization kinetics of Hastelloy X and Incoloy 800H in a simulated GCR helium were determined through 4000 h at temperatures from 750 to 1000°C. Both alloys experience "breakaway" in carburization rate after an incubation period, which lengthens as temperature is decreased. Surface carbon concentrations and carbon gradient profiles were also determined and analyzed.

Experiments were conducted on the dimensional stability of several alloys in simulated GCR helium. Both Hastelloy X and Alloy 800H were found to dilate up to a maximum of 0.8% after 2000 h at 1000°C. This result is explained on the basis of carburization (carbon gradient profile) induced stresses.

A report describing the results of the first 2500 h of operation of the Inconel 617 steam corrosion loop was completed. All materials

exhibited parabolic oxidation kinetics during the first year and subsequently have oxidized at low constant rates. Tests of 2 1/4 Cr-1 Mo steel in superheated steam under high heat flux have demonstrated a significant effect of heat flux on corrosion rate.

Joining Subtask

Evaluation of the weldability of Incoloy 802, Inconel 618E, and HD 556 was completed. The spot-Varestraint testing procedure was used to rank the hot cracking susceptibility of these alloys relative to Inconel 617 and Hastelloy X. Incoloy 802 was found to be the least hot-crack sensitive of the group tested, but the behavior of all alloys was satisfactory.

Ceramics Subtask

The characterization of two fibrous insulations formed from slurries of Kaowool ceramic fiber, Saffil alumina fiber, and organic binders was completed. Two types of structural silicon nitride were also characterized.

A system for exposing structural ceramics to a simulated GCR helium atmosphere at elevated temperatures was designed and constructed. Design was completed of a system for studying the effects of rapid depressurization on the integrity of ceramics and fibrous insulations.

5. HTGR GRAPHITE STUDIES

The Graphite Characterization Program sponsored five irradiation capsules during the year. The most complex of these was creep capsule OC-3, which extended the 900°C creep data to a damage fluence of 3.2×10^{25} neutrons/m². This capsule also demonstrated that secondary creep is volume conserving and verified that primary creep is recoverable. The irradiation of the German graphites was carried to full life or beyond, but a significant temperature error was uncovered. This error was corrected for the capsule inserted to determine the graphite life for the N-reactor at Hanford.

6. EVALUATION OF PEBBLE-BED HTR

The purpose of this effort is to examine the design and operation of the pebble-bed high-temperature reactor (PBR) concept. The work performed supports two major efforts: (1) analysis of the PBR under a technical information exchange umbrella agreement with the Federal Republic of Germany (FRG) and (2) a comparison of the PBR and the prismatic HTGR.

During this past year, results obtained by ORNL regarding projected ore requirements, fuel cycle costs, and flexibility in application of the PBR were compared with prior analysis at KFA. General agreement was found concerning the superior ore utilization of the PBR compared with light-water reactors (LWR) for all applications (throwaway, recycle, near breeder) at a fuel cost no greater than that of LWRs. Details of the ORNL results compared favorably with KFA for similar reactor modeling. Cooperation with KFA has continued this year through a joint analysis of well-defined benchmark problems for comparison of methods and data.

Basic evaluation of the PBR has been directed toward more detailed studies of reactor operation, particularly reactor control and stability.

Development of analysis methods continues under this program, in the areas of cross-section processing for double heterogeneous systems, temperature feedback effects on cross sections, stability analysis, thermal-hydraulic analysis capability, and fuel management with selective recycle.

A comparison of the PBR with the prismatic HTGR was initiated during the past year. The work performed at ORNL consists of an overall assessment as well as technical input in the areas of reactor control and stability, ore requirements and fuel cycle costs, PCRV costs, fission product release, fabrication and reprocessing unit costs, material and fuel requirements, graphite reflector damage, instrumentation requirements, seismic effects on safety, estimated reactor availability, and overall capital costs.

1. HTGR CHEMISTRY

R. P. Wichner

1.1 EFFECT OF STEAM CORROSION ON THE STRENGTH OF GRAPHITE — R. P. Wichner and W. P. Eatherly

1.1.1 Corrosion of Graphite Specimens

1.1.1.1 Steam Corrosion Tests — R. P. Wichner, L. L. Fairchild, and J. D. Kintigh

Specimens of graphite grades H451, standard Stackpole 2020, and heat-treated Stackpole 2020 were corroded in a pair of circulation flow rigs described previously.¹ The nominal corrosion conditions and burn-offs are summarized in Tables 1.1 and 1.2. Gas samples for all runs except an initial few were acquired on at least a daily basis and analyzed for hydrogen, carbon monoxide, carbon dioxide, methane, moisture, nitrogen, and oxygen by gas chromatography.

The gas chromatograph has not functioned as well as we had hoped it would with the sampling system. Moisture readings are generally lower than expected from mass balance determinations, evidently caused

Table 1.1. Nominal Corrosion Conditions for Grade H451
Cylindrical Specimens

Diameter [cm (in.)]	Temperature (°C)	Nominal concentrations			
		H ₂ (vpm) ^a	H ₂ O (vpm)	Duration (weeks)	Burnoff (%)
1.3 (0.5) ↓ ↓	1,000	16,000	7,400	2	10.0
	1,000	15,000	7,900	1	4.8
	900	11,000	13,000	2	6.7
	900	8,300	15,000	3	7.9
	900	4,300	7,800	3	4.0
1.9 (0.75) ↓ ↓	1,000	17,000	6,000	4	9.8
	1,000	25,000		2	7.1
	900	6,300	5,600	4	3.6
1.3 (0.5) ↓ ↓	900	3,300	8,800	2	2.1
	1,000	13,000	10,000	1	4.2
	1,000	7,900	4,300	1	2.3
1.9 (0.75)	900	10,000	13,000	2	3.0
1.3 (0.5)	↓	3,000	5,500	3	2.9
1.3 (0.5)		1,900	2,400	4	2.4
1.9 (0.75)		3,900	4,600	4	2.2
1.9 (0.75)		2,400	1,800	6	2.1

^avpm = volume parts per million.

Table 1.2. Nominal Corrosion Conditions for Grade Stackpole 2020 Cylindrical Specimens

Diameter [cm (in.)]	Temperature (°C)	Nominal concentrations			
		H ₂ (vpm) ^a	H ₂ O (vpm)	Duration (weeks)	Burnoff (%)
1.3 (0.5)	1,000	19,000	6,200	2	8.4
	900	5,100	8,200	3	3.2
	1,000	16,000	7,900	1	5.0
	1,000	17,000	7,000	3	16.8
1.9 (0.75)	1,000	18,000	5,500	2	4.8
1.3 (0.5) ^b	1,000	18,000	2,400	2	7.9

^avpm = volume parts per million.^bSignifies heat-treated specimen.

by adsorptive losses in the sampling line. Proper moisture-level determination requires either short, heated sampling lines or an in situ determination method. We have found hydrogen determinations to be generally reliable, although imprecise with the thermal conductance detection method employed. Hydrogen concentration variation in helium carrier has only a slight effect on the thermal conductivity of the carrier; hence, the sensitivity is low. Carbon monoxide readings are generally the most reliable and precise of the group. Carbon dioxide determinations, although potentially as reliable and precise as those for carbon monoxide, are occasionally inaccurate because of an unidentified impurity that is seemingly introduced by the carrier gas purifier. Unfortunately, this inaccuracy was not recognized until late in the experiment; consequently, CO₂ determinations in many of the earlier runs exhibit a concentration peak shape that has been proved to be incorrect.

Therefore, gas analyses and corrosion rate determinations for each run were made by means appropriate for each case. For example, the following procedure was employed for run H451-10 in which the carbon dioxide determinations were incorrect because of contamination.

The two reactions of primary interest are

and

(We have found no detectable levels of methane in any of the runs.)

Mass balances for H_2 , CO, CO_2 , and H_2O yield, respectively,

$$R_1 + R_2 = Q[H_2] , \quad (3)$$

$$R_1 = R_2 = Q[CO] , \quad (4)$$

$$R_2 = Q[CO_2] , \quad (5)$$

$$Q[H_2O] = Q[H_2O]_i - R , \quad (6)$$

where, Q is the volumetric flowrate of the makeup gas containing $[H_2O]_i$, concentration of moisture, and R_1 and R_2 are the rates of reactions (1) and (2). The quantities in the brackets are the respective concentrations of H_2 , CO, CO_2 , and H_2O in the test apparatus.

As readily seen, reliable determination of H_2 , CO, and CO_2 (knowing $[H_2O]_i$) allows determination of the unknown rates R_1 and R_2 and the moisture level in the rig $[H_2O]$ and permits cross-checking for internal consistency. With no CO_2 level determination, the calculation may still be made, but no cross-check is possible. However, the calculated R_1 as a function of time could be compared with the time-averaged value obtained from the measured specimen mass loss.

Figure 1.1 shows the H_2 and CO gas chromatographic determinations during run H451-10. From these values, Eqs. (3) and (4) may be used to determine the rates R_1 and R_2 ; hence, the CO_2 concentrations may be calculated by using Eq. (5), as shown by the lower curve in Fig. 1.1.

The calculated rates of reactions (1) and (2) are plotted in Fig. 1.2 as a function of time. As shown, the principal corrosion reaction rate [Eq. (1)] increased with time during this run, but the rate of reaction (2) stayed fairly level. The time-averaged value of R_1 , determined from specimen mass loss and shown in Fig. 1.2 as the horizontal line, compares well with the time-averaged value of R_1 calculated from gas chromatographic determinations of H_2 and CO.

Figure 1.3 shows the comparison of moisture levels determined from the gas chromatograph with values predicted from the mass balance of

Fig. 1.1. Measured H_2 and CO Concentrations and Calculated CO_2 Concentration During Run H451-10.

Fig. 1.2. Calculated Reaction Rates R_1 and R_2 During Run H451-10.

Fig. 1.3. Moisture Levels During Run H451-10.

Eqs. (3), (4), and (5) by using measured values for $[H_2]$, $[CO]$, and the known inlet concentration of moisture. The result shown is typical for all runs, in that measured moisture levels invariably fall below anticipated levels due to losses on the sampling line.

1.1.1.2 Mass Loss Data — C. R. Kennedy

After removal from the oxidation apparatus, the samples were degassed at 750°C for 1 h before weight-loss measurements. Because the water vapor was discontinued in the oxidation apparatus at temperature, such degassing after oxidation is probably not necessary.

We discuss the grade 2020 graphite weight losses first, because the situation is simpler. Representative weight-loss data for the 2020 rods are presented in Fig. 1.4. The data are reasonably uniform as a function

Fig. 1.4. Axial Density Profiles for Grade 2020 Compression Specimens.

of furnace position, with the exception of rod number 4 with the longest residence time. We have no conclusive explanation for the increased weight loss at the exit end of this rod, although it must be pointed out that the graphite temperatures were not directly measured; hence, this may be an indication of some cooling of the rod by the incoming gas. In Fig. 1.5, data of this type are replotted as a function of residence time in the furnace. It will be noted that rod 4 lies at 500 h and 15.7% weight loss based on the entire rod weight. Favoring only the points at rod positions of 20 to 36 cm (8 to 14 in.) will not bring this point on the 1000°C curve.

Figure 1.6 presents both data on grade 2020 brittle-ring samples as a function of exposure time and the above results for the rods. The sampling plan first denotes a sample measurement by x_{ij} , with the index i representing radial position and j representing axial position in the billet. Slabs were cut at five different axial heights from the original billet (i.e., $j = 1, 2, \dots, 5$). From each slab, five sample positions were used: the center (axis), halfway to the cylindrical billet edge, and three adjacent positions at the cylindrical edge ($i = 1, 2, \dots, 5$). A sample set consisted of 25, one from each possible value of i and j .

Fig. 1.5. Burnoff as a Function of Exposure Time for Grade 2020 Rod Samples.

Fig. 1.6. Burnoff for Grade 2020 Brittle-Ring Specimens.

For a given oxidation run, the set of five with running i was adjacent, but each set with fixed j was dispersed equidistantly along the furnace axis. The ordering by the index j was altered from run to run of the furnace. This dispersion of sample positions led to the following conclusions:

- No difference was observed within the sets of three circumferential samples ($i = 3, 4, 5$).
- No difference was observed from the various axial positions j with fixed i .
- A significant tendency was observed for samples on the axis (center, $i = 1$) to oxidize more rapidly than those intermediate ($i = 2$) or on the edge ($i = 3, 4, 5$). This tendency is indicated in Fig. 1.6.
- A slight, but discernible, tendency was observed for samples at the exit end to oxidize more slowly (the opposite effect from that noted in Fig. 1.4).

The following observations were also noted: the ring samples oxidize more slowly than do the rod samples up to 400 h. The upswing at 500 h appears to be real. The furnace held 50 samples; hence, two sample sets of 25 were oxidized simultaneously. The 500-h samples were accompanied by sets at 170 and 330 h so that the upswing noted at 500 h does not represent a unique furnace run, but all furnace runs.

For a description of the microstructural appearance of the 2020 oxidations, see Sect. 1.1.2. All samples indicated catalytic oxidation associated with an identifiable "high-Z" inclusion, that is, one with an electron density much above the carbon atom. This impurity is almost certainly calcium, with possibly some barium. Conversely, the uniform dispersion of silicon in the sample indicates that its role under these conditions of oxidation is either passive or suppressive.

The results for H451 graphite at 1000°C were "normal," whereas results at 900°C were unexpected. Such data are shown in Fig. 1.7 for the rods at 900°C. These end anomalies in weight loss were not apparent from the external surface. Sectioning and radiography revealed that a tear-drop shaped void and/or low-density region tailing toward the axial

center was the cause of the high weight loss. In general, then, these regions represented a surviving "tube" with a central void. The regions near the surface appear radiographically to be at or near the original graphite density. Eliminating these end anomalous regions from the data base yields the curves of Fig. 1.8. Ring data are shown in Fig. 1.9, superimposed on the rod data. Finally, the rod densities as a function

Fig. 1.7. Axial Variation of Density of Oxidized H451 Rods.

Fig. 1.8. Overall Burnoff of H451 Rod Samples, Excluding 900°C End Specimens.

of axial position at 1000°C are given in Fig. 1.10; they exhibit none of the anomalies seen at 900°C.

1.1.2 Purification of Stackpole 2020 Graphite by Heat Treatment — W. P. Eatherly

A method for reducing the impurity content of grade 2020 graphite appeared desirable, at least for testing purposes. The major impurities observed in this grade were calcium and barium, both known to catalyze

Fig. 1.9. Comparison of Degree of Burnoff for Rod and Ring Specimens, 900°C.

Fig. 1.10. Burnoff Versus Axial Location for H451 Rod Specimens Oxidized at 1000°C.

steam oxidation of graphite. A number of small samples were thermally purified by heat treating in an argon atmosphere of from 2600 to 3200°C, during which it became obvious that the samples had not reached temperatures in excess of about 2500°C during manufacture. This fact has since been verified by the manufacturer.² These preliminary experiments established also that the calcium and barium are essentially removed by heat treatment to 2800°C, although iron, as expected, remains present at temperatures to 3300°C. The results of a semiquantitative optical spectrochemical analysis are given in Table 1.3. Flexural strength measurements indicated no change in ultimate strength or displacement after heat treatment.

To provide a more realistic test, a "billet" of 2020 graphite 9 cm (3.5 in.) in diameter by 36 cm (14 in.) was heat treated, and samples were extracted at various points throughout this cylinder as well as adjacent material from the unpurified mother billet. Preliminary x-ray fluorescence data substantiate the data of Table 1.3, and more detailed spark-source mass spectrometric analyses are in progress.

An indication of the catalyzed oxidation in unpurified grade 2020 graphite is apparent in Fig. 1.11. The x-radiographs are of a broken brittle-ring sample after oxidation at 1000°C for 500 h. The sample

Table 1.3. Optical Emission Spectrochemical Analysis of Heat-Treated Stackpole 2020 Graphite

Element	Detection limit	As received	Temperature (°C)			
			2600	2800	3000	3200
Al	<10	50	<i>a</i>			
B		2	2	2	2	2
Ba	<5	15				
Ca	<2	250		<10	4	
Cu		8	2	6	3	3
Fe		120	120	430	110	90
Mg	<1	2				
Pb	<5	15				
Si		230	25	20	35	30
Ti	<5	150	90	<20	<10	5
V	<5	30				
Zn	<20	140	20			

^aMissing entries in the table are at the detection limit.

Y-165311

AS OXIDIZED, 1000°C - 500 HRS.

HEAT TREATED 2800°C

Fig. 1.11. X-ray Radiographs of Oxidized 2020 Graphite Before and After Heat-Treatment at 2800°C. Dark areas in upper radiograph represent impurities diffused about a void. Lower figure shows voids after evaporation of impurity.

is 0.64 cm (0.250 in.) thick, normal to the photographic film; the photographs are positives. In the upper photograph, the dark areas represent a high-Z impurity, which could be visually identified as a void. The high-Z impurity diffused as the void formed and before oxidation would have appeared as a fine point mass. After this sample was heat treated at 2800°C, the impurity evaporated and the voids clearly appear as low-density regions. A one-to-one correlation of voids to impurity concentrations clearly exists. The correlation is not quite one-to-one in the other direction; that is, high-impurity regions occur that did not produce a void, presumably because of inaccess to steam.

We reach the following conclusions:

- Under the conditions utilized in these oxidation experiments, catalysis is the major mechanism of weight loss in 2020 graphite.
- The probable catalytic agent is calcium or possibly barium; iron is not a dominant catalytic agent.
- Grade 2020 graphite may be "cleaned-up" by a simple heat treatment at 2800°C. This graphitization temperature is not uncommon in the graphite industry.

1.1.3 Methods for Physicochemical Characterization — W. P. Eatherly

1.1.3.1 Surface Area and Gas Porosimetry Measurements — J. D. Kintigh

The automated BET-type surface apparatus,³ designed and built to measure the surface areas and helium densities of the samples, encountered a number of minor technical difficulties. One of the pressure transducers had to be returned to the manufacturer because of electrical malfunction. The major difficulty, however, resulted from an unacceptable temperature rise in the gas manifold on activation of any of the manifold solenoid valves. The apparatus had to be redesigned and rebuilt, using pneumatic valves. This situation offered the opportunity to make a number of additional modifications to improve the flexibility and efficiency of the system. To improve pumping efficiency, the gas manifold was reconstructed with 0.635-cm-OD (1/4-in.) copper

tubing instead of the original 0.318-cm-OD (1/8-in.) copper tubing. The overall length of the manifold was shortened to keep the manifold volume the same (about 20 cm³) in both designs. The system was also redesigned to eliminate the need for the series of five calibrated volumes connected to the manifold. This change was accomplished by using the manifold as the calibrated volume and controlling the introduction of gas to, or the removal of gas from, the manifold by monitoring the pressure in the manifold as the gas was added or removed. The apparatus is now capable of measuring surface areas by adsorption or desorption, a capability that was not available in the original design but is necessary for porosity measurements and for active site studies. The system has been interfaced to a Hewlett-Packard 9835 desk-top computer for system control and data acquisition. The computer system will also be used to calculate the surface areas and to plot the isotherms on a graphics plotter.

In its current configuration, the apparatus can handle both powder and solid samples up to 1.2 cm in diameter by 2.5 cm in length; both argon and krypton gas can be employed. We have assisted in setting up a "round-robin" experiment on graphite surface areas under the auspices of ASTM, and both Great Lakes H451 and Stackpole 2020 grades have been supplied by us for these experiments. The first sample set will be returned to ORNL for measurement in March 1980.

1.1.3.2 Mercury Porosimetry — J. D. Kintigh

The macroporosimetry of the graphites is being determined by using an Aminco porosimeter, model 5-2171B. The apparatus was modified by the replacement of the original pressure gauges with 30-cm (12-in.) precision Heise gauges. Experiments were conducted to determine the maximum pressures to which the samples can be submitted before micro-structural damage. In the case of H451 graphite, this pressure appears to be about 1.3 MPa (2000 psi). Experiments were also performed to determine the ability to remove mercury after intrusion; temperatures of 900°C were found necessary to reduce the residual mercury contamination to less than 5 ppm. Although no change in pore spectrum occurred on repeated pore spectra at applied pressures below 1.3 MPa (2000 psi),

strength measurements are currently in progress to verify completely that no microstructural damage occurred.

1.1.3.3 X-Ray Densitometry — B. E. Foster

The use of x-ray densitometry to determine bulk density has been established. A number of calibration standards have been constructed in the density range of 1.4 to 1.9 Mg/m³, utilizing fine-grained graphites of varying density or thickness. Absolute densities of about 0.5% can be determined over this entire range by exposing simultaneously the unknown samples and the standards. To establish sensitivity (relative density), step wedges were constructed from a highly uniform aerospace graphite. At optimal exposure for the density region being investigated, relative density changes of about 0.2% can be detected. These accuracies and precisions are quite adequate for current purposes.

The two graphites of current interest, H451 and 2020, are relatively coarse grained, particularly H451. In the 0.6-cm (1/4-in.) specimens being studied, this granularity can largely mask gross density gradients caused by oxidation, unless integration is performed over an extended area of the cross section. The x-ray films of the samples are read on an optical densitometer that provides a linear motion of the film, the resulting measurements being displayed on an X-Y recorder. To accommodate the flat disk samples provided by the oxidation experiments, this linear motion is transformed to a combined rotating plus linear motion by a precision drive train. The resulting output is then displayed on the recorder as the mean density as a function of radius. This drive train has been assembled and is currently being installed on the optical densitometer.

In addition to the routine gross density measurements made on slabs of graphite prior to sample machining, a number of postoxidation samples have been radiographed. The dominant effect noted to date has been catalyzed oxidation resulting from impurities. These data are discussed elsewhere in this report.

1.1.4 Results of Mechanical Property and Related Tests — W. P. Eatherly and C. R. Kennedy

1.1.4.1 Billet Characterization and Sample Preparation — C. R. Kennedy

The graphite used in these studies was purchased jointly with the General Atomic Company (GA) and to its general specification. In the case of the moderator graphite, Great Lakes grade H451, split billets were shipped to ORNL after acceptance testing at GA. For the support post material, Stackpole grade 2020, the billets were shipped to, and sectioned by, GA; portions of each billet were used by both GA and ORNL.

The two graphites are quite different: H451 is a moderately coarse-grained extruded graphite that has been heat treated to 2800°C and gas purified, whereas grade 2020 is a fine-grained isostatically molded material whose heat-treatment temperature appears to have been about 2500°C. At ORNL, both materials were nondestructively tested to select regions of the billets to be used for sample extraction. Because of the intensive effort to be expended on each specimen, such preliminary testing was warranted.

The nondestructive evaluation consisted of eddy-current mapping, ultrasonics, and both direct and x-radiographic densitometry. The electrical conductivity and its orientation was calculated from the eddy-current measurements, and mechanical properties, from the sonic tests. On the basis of these data, the H451 graphite was divided into three classes, briefly characterized as low-, medium-, and high-bulk density. In addition, one split billet was rejected in its entirety; this billet showed an abnormal structure and gross defects caused by its being held in the extrusion press nozzle as the mud-chamber was reloaded. The 2020 graphite was quite uniform and was therefore treated as a unit rather than being subclassified. Mean values from the results of the nondestructive testing data are given in Tables 1.4 and 1.5.

All samples for oxidation were machined without lubricants and were handled with gloves. In cases requiring trace chemical analysis, machining was performed with carbide tooling. After machining, the specimens were subjected to a standardized sonic dedusting, were heated to 750°C to remove all sorbed water, and preoxidation weights were

Table 1.4. Average Properties of Grade 2020 Billet Slabs

Block No. Section No.	249			252			254			297			301		
	B	D	E	B	D	E	B	D	E	B	D	E	B	D	E
Electrical resistance ^a															
W/G, ^b $\mu\Omega\text{-m}$	17.4	17.9	18.3	17.7	17.1	17.2	18.4	17.7	17.0	17.3	17.1	17.3	18.4	17.7	17.6
A/G, ^c $\mu\Omega\text{-m}$	17.4	19.8	21.7	21.8	18.7	18.4	21.3	19.7	18.4	20.4	18.2	18.9	25.0	22.5	22.5
Elastic constants ^d															
$1/S_{33}$, ^e GPa	10.3	9.9	9.3	9.4	9.7	9.2	9.6	9.7	10.4	9.8	9.6	9.5	10.1	10.1	10.5
C_{44} , ^f GPa	4.5	4.3	4.1	4.1	4.2	3.9	4.2	4.1	4.9	4.3	4.1	4.1	4.4	4.4	4.8
μ , ^g	0.15	0.15	0.12	0.15	0.16	0.18	0.15	0.18	0.06	0.13	0.17	0.16	0.15	0.15	0.08

^aDetermined by eddy-current measurements.^bWith grain.^cAgainst grain.^dDetermined by sonic measurements.^eYoung's modulus in the axial direction.^fShear modulus.^gPoisson's ratio.

Table 1.5. Average Properties of Grade H451 Billet Slabs

	High densities	Medium densities	Low densities
Electrical resistance ^a			
W/G ^b ($\mu\Omega$ -m)	7.6	8.0	8.1
A/G ^c ($\mu\Omega$ -m)	7.9	8.3	8.4
Elastic constants ^d			
1/S ₃₃ , ^e GPa	9.8	9.6	9.3
1/S ₁₁ , GPa	8.5	7.8	7.0
C ₄₄ , ^f GPa	4.2	3.9	3.5
ν ^g	0.13	0.15	0.15

^aDetermined by eddy-current measurements.

^bWith grain.

^cAgainst grain.

^dDetermined by sonic measurements.

^eYoung's modulus in the axial direction.

^fShear modulus.

^gPoisson's ratio.

determined. Oxidations were performed as described above. Sample weights were determined on a balance calibrated to ± 0.1 mg, and dimensions were determined by a supermicrometer that was accurate to better than 0.1 mil, when such accuracy was required.

Compression samples were machined from the oxidized rods subsequent to oxidation. In addition to the compression strength samples, short 0.635-cm (0.250-in.) disks were also removed at various axial positions along the rod for cross-section x-ray densitometry. Sonic velocity and eddy-current measurements were also made on the intact oxidized rods. Brittle-ring strength specimens were oxidized in their final geometry and could be radiographed as such.

Nondestructive test data on large slabs, cut at various locations from the billets of grades 2020 and H451, are given in Tables 1.4 and 1.5.

1.1.4.2 Strength Loss and Other Related Physical Property Data — C. R. Kennedy

The direct measurements of mechanical properties were made on four types of samples: brittle ring, derived C-ring, four-point flexural,

and simple compression. Because of equipment limitations, flexural samples were employed only for preoxidation data. Brittle-ring samples have been utilized for a number of reasons: a large number of samples can be oxidized at once because of their economy of space, two to three measurements can be made per ring, the stress volume (as defined by Weibull statistics) can be easily varied over a factor of 10, and the shape lends itself directly to a variety of other measurements. Compression samples were included as a measurement of a uniaxial stress.

Three sizes of brittle-ring specimens were included, with outside diameters of 38.1, 25.4, and 19.0 mm (1.5, 1.0, and 0.75 in.). All three types had a wall thickness of 5.1 mm (0.2 in.) and an axial thickness of 6.3 mm (0.25 in.). These selections of relative geometry minimize the effect of surface flaws. Fracture could be initiated at one point, the ring could be rotated 45° to place this original fracture at a point of new neutral forces, and a second fracture could be initiated. Finally, the minor section between the two fractures could be removed to yield a C-ring sample for a third fracture. This procedure is indicated in Fig. 1.12. Stresses were calculated by using formulas that allow for shift in the neutral plane.⁴

The four-point bend specimen 5.2- by 0.64-mm (0.125- by 0.250-in.) cross-section was third-point loaded on 12.7-mm (0.5-in.) spans. A center-span deflectometer was used for strain measurement.

Compression samples were right circular cylinders with a 2/1 length/diameter ratio. Two diameters were employed, 12.7 and 19.0 mm (0.5 and 0.75 in.). Testing was performed with a Baldwin compression jig. Simple deflection was measured, which was too insensitive to measure moduli but did provide a reasonable estimate of the ultimate strain at fracture.

Data on the testing of unoxidized 2020 graphite are summarized in Table 1.6 with their coefficients of variation (standard deviations expressed as percentages). Equivalent data on H451 present us with a problem in that the graphite is sufficiently nonuniform to require a correction for density, or equivalently, porosity. This correction is usually expressed in the form of a Knudsen (exponential) relationship such as

$$\sigma_f = \sigma_0 \exp(-\alpha P) , \quad (7)$$

Fig. 1.12. Method of Testing Brittle-Ring Specimens.

where P is the porosity, σ_f the ultimate strength, and σ_0 and α experimentally determined parameters. The data on unoxidized H451 are presented in Table 1.7, with the additional Knudsen relationships:

$$\begin{aligned}
 E &= E_0 \exp(-\beta P) , \\
 C &= C_0 \exp(-\gamma P) , \\
 \epsilon_f &= \epsilon_0 \exp(-\delta P)
 \end{aligned}
 \tag{8}$$

for Young's modulus, sonic attenuation, and ultimate strain at fracture, respectively.

Table 1.6. Results of Testing 2020 Graphite Before Oxidation

(CV = coefficient of variation; see text for explanation of other symbols)

	Ultimate strength		Young's modulus		Ultimate strain	
	$\bar{\sigma}_f$ (MPa)	CV (%)	$\bar{E}$ (GPa)	CV (%)	$\bar{\epsilon}_f$ (%)	CV (%)
3.8-cm (1.5-in.) diam						
O-ring	39.16	4.38	10.43	3.10	0.498	4.38
C-ring	33.52	6.58	10.39	2.57	0.495	10.20
2.5-cm (1.0-in.) diam						
O-ring	36.67	7.75	10.40	2.65	0.488	4.29
C-ring	32.49	7.71	10.09	4.01	0.466	7.43
1.9-cm (0.75-in.) diam						
O-ring	34.16	7.63	9.67	5.56	0.479	6.24
C-ring	32.95	7.36	10.26	4.70	0.468	8.10
Bending						
With grain	33.95	4.71	10.25	4.29	0.46	4.3
Against grain	35.38	4.55	10.26	5.95	0.47	6.7
Av		6.50		4.10		6.77
Compression						
1.3-cm (0.5-in.) diam against grain	87.50	1.26	10.3 (sonic)		1.98	8.59

The large coefficients of variation noted for the H451 graphite as contrasted to 2020 graphite are not unexpected. The Weibull statistical model relates the fracture stress to material and stress field parameters by

$$\sigma_f = S_0 \left(\frac{V}{V_0} \right)^{1/m} \quad (9)$$

where S_0 (a "stress") and V_0 (a volume) are constants, and V is the volume under the stress σ_f . The exponent m is the so-called Weibull constant. In the case of nonuniform stress fields, an effective volume

Table 1.7. Results of Testing H451 Graphite Before Oxidation

(CV = coefficient of variation; see text for explanation of other symbols)

	σ_0 (MPa)	α	CV (%)	E_0 (GPa)	β	CV (%)	C_0 (dB/mm)	γ	CV (%)	E_{f_0} (%)	δ	CV (%)
3.8-cm (1.5-in.) diam												
O-ring	165	9.97	15.23	63.6	11.0	6.63				2.60	9.43	11.3
C-ring	333	14.90	16.31	36.0	8.15	6.92				1.32	6.25	32.4
2.5-cm (1.0-in.) diam												
O-ring	235	11.11	7.69	17.2	16.1	4.43				1.57	6.64	7.05
C-ring	428	15.84	14.27	47.0	9.4	5.96				1.68	7.08	14.2
1.9-cm (0.75-in.) diam												
O-ring	140	7.79	6.86	55.3	10.4	5.86				1.44	5.89	7.3
C-ring	410	15.92	13.51	23.6	6.1	9.26				4.31	12.20	15.3
All							0.33	13.54	6.27			
Av		12.59	12.33		10.20	6.53					7.92	14.59
	$\bar{\sigma}$ (MPa)	S (MPa)					$\bar{C}$ (dB/mm)	S (dB/mm)				
Compression												
High density, 1.765 g/cm ³	59.3	4.10	6.92				2.63	0.30	11.5	3.30	0.33	10.1
Medium density, 1.742 g/cm ³	56.49	1.73	3.07				2.74	0.11	4.16	2.83	0.22	7.88
Low density, 1.726 g/cm ³	48.67	0.93	1.94				3.35	0.05	1.68	2.55	0.22	8.67

V must be defined or an integration over the stress field performed. Bortz et al.,⁵ have successfully used a "95% maximum stress" to define an effective volume for the brittle ring and have experimentally justified its use. With this definition, we have plotted the fracture stresses for the various sample geometries, shown in Fig. 1.13. For Weibull statistics, the coefficient of variation (CV) is a function of the exponent m only; hence, m can be calculated also from the CV. This calculation is shown in Table 1.8. The agreement is reasonable and consistent with past studies yielding low m (or high CV) values for extruded graphites in comparison with molded grades. It should also be noted that the compression test results had a much lower CV for both grades, implying a reduced effect of stress volume on compression strength.

Fig. 1.13. Weibull Statistics as Derived from Brittle-Ring, C-Ring, and Flexure Strength Samples.

Table 1.8. Calculated and Observed Values of the Weibull m -Value

Graphite grade	CV (%)	m -Value from CV	m -Value from Fig. 1.13
2020	6.50	18.5	16.1
H451	12.33	9.0	6.0

We have many hundreds of oxidized samples but make no attempt here to reproduce that data. Rather, we will summarize the information in the form of empirical relationships and their correlation on the basis of fracture mechanics.

1.1.4.3 Correlation of Data: Use of Fracture Mechanics — C. R. Kennedy

In our work on specialty graphites for aerospace applications and, more recently, on radiation damage in graphite, the concepts of fracture mechanics have proved most useful in understanding the mechanical properties of graphite and their relationships to microstructure. Simply stated, the Griffith-Irwin⁶ crack theory for elastic materials produces a

relationship between fracture strength and other more fundamental but observable properties of the material:

$$\sigma_f \propto \left(\frac{EG_{ic}}{\pi a} \right)^{1/2} \quad (10)$$

where

G_{ic} = strain energy release rate, that is, the work to create new surfaces within the material,

E = Young's modulus,

a = a measure of the critical defect size.

In the case of oxidized graphite, it is tempting to assume that the oxidation predominantly increases the size of preexisting defects (voids) in the graphite structure, thus resulting in a decrease in strength. As we shall see, both the defect size and Young's modulus are accessible parameters by nondestructive sonic measurements; from these, a prediction of strength should be possible. If so, valid statistical data on the mechanical behavior of oxidized graphites can be readily obtained from relatively simple tests, such as brittle rings. We now examine these hypotheses.

In our graphite fabrication studies, performed mainly for the Department of Defense, we have produced controlled series of samples by systematically varying the particle sizing and the sulfur content of the precursor cokes. These sample sets have been subjected to detailed fracture mechanics testing of both smooth and notched beams; the defect size calculated thereby has been demonstrated to be proportional to the sonic attenuation. That is, the sonic waves appear to be predominantly scattered from voids within the graphite, which is a measure of the number and size of these voids. If oxidation does indeed represent only an increasing size of preexisting voids in the graphite, sonic attenuation is proportional to the "defect size." Conversely, the quantity G_{ic} may be assumed to be only mildly, if at all, affected by oxidation as long as the fracture mechanism remains unchanged. These conclusions

immediately suggest a relationship between the sonic modulus E , sonic attenuation C , and fracture strength σ_f :

$$\sigma_f \propto \left(\frac{E}{C} \right)^{1/2} \quad (11)$$

from the Griffith-Irwin relation, Eq. (10). This relationship is tested in Fig. 1.14 for 2020 graphite, and the proportionality is excellent.

In the preceding section, particularly Table 1.7, we alluded to the dependence of properties on porosity through an exponential-type relationship (the Knudson equations). The introduction here of the sonic attenuation as a fundamental property requires us to demonstrate unequivocally that it also satisfies such an empirical correlation, which is shown in Fig. 1.15, again for oxidized and unoxidized H451. The goodness of fit is obvious.

Fig. 1.14. Compressive Strength of Oxidized 2020 Graphite as a Function of the Parameter (Young's Modulus/Attenuation) $^{1/2}$.

Fig. 1.15. O-Ring Attenuation Versus Porosity. Dashed line is 95% confidence level on individual points.

We have seen already an indication that G_{ic} is not strongly affected by oxidation. A sharper test of this constancy of G_{ic} may be obtained by inserting the Knudson relationships, Eqs. (7) and (8), into the Griffith-Irwin equation, with the tentative assumption that G_{ic} is not dependent on porosity. This test leads to a relationship between the exponential coefficients

$$\alpha = (1/2)(\beta + \gamma) . \quad (12)$$

The data to test this relationship are given in Tables 1.9 and 1.10; the results are summarized in Table 1.11 for the various mechanical tests

Table 1.11. A Comparison of Observed Knudson
Coefficients with Their Theoretical
Interrelationship
[See Eq. (12)]

Grade	α	β	$\frac{\beta + \gamma}{2}$
2020 ($\gamma = 8.8$)			
O-ring	10.7	9.9	9.4
C-ring	8.4	7.2	8.0
Compression	7.8		
H451 ($\gamma = 7.72$)			
O-ring	6.6	9.0	8.4
C-ring	8.0	6.8	7.3
Compression	7.8		

employed here. The agreement between the exponents is reasonably good and certainly encourages further study.

These early results on Stackpole 2020 must be viewed with reservation. The analysis assumes that the defect population remains the same before and after oxidation and only changes character, that is, the defects enlarge. This situation appears to be the case for H451. For 2020, however, we are apparently observing marked catalytic effects, which implies an entirely new defect population not present in the unoxidized material.

The above analyses must still be considered tentative. Further data and a more careful statistical treatment are required. Moreover, the Griffith-Irwin equation is an oversimplification when applied to a pseudoplastic material such as graphite. Nevertheless, we present the following as working hypotheses:

- The Griffith-Irwin equation applies to graphite at least in principle.
- The Weibull statistical model applies to these graphite grades and may not involve a change in m -value as oxidation proceeds.
- The energy to create new surfaces by fracture is in first order not affected by oxidation.

- Oxidation alters both the texture and the amount of void volume in the bulk material, which may be quantitatively estimated by sonic attenuation.

1.1.5 Anelastic Analysis of a Ring Under Diametral Compression — H. Cords*

1.1.5.1 Introduction

The ring geometry in fracture tests is favorable for a number of reasons. Both irradiation capsules and oxidation furnaces used in graphite corrosion experiments preferably have cylindrical shapes. Thus, in studying the effects of both neutron irradiation and chemically corrosive atmospheres on graphite, the convenience of cylindrical specimens is apparent. The subsequent strength tests on the affected materials can be performed best as diametral compression tests. This method precludes the removal of excessive material that would otherwise have unnecessarily occupied the reactor or that would have possibly required costly cutting operations in the hot cells. Also, in the case of corrosion experiments, shaping of the specimens into new forms might interfere with the study of surface effects.

Use of the brittle-ring test (or the so-called "Carneiro test") is widespread; an evaluation of strength values from the applied critical loads is described by Timoshenko.⁷ Nevertheless, the calculated strength values depend on additional simplifying assumptions used for the stress analysis. The assumptions are beyond the specifications of the theory of elasticity and are made to reduce the number of dimensions involved in the problem. Furthermore, as compared with the uniaxial tensile test specimen, the ring geometry is more difficult to understand: the material fails under near-bending conditions, and the relationship between bending strength and uniaxial tensile strength has been a notorious problem. A statistical explanation is available,⁸ but the relationship to strength of the ring is not obvious. Because the results from uniaxial tensile tests are basic information that is used,

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for instance, as input to all finite-element stress codes, it is desirable to have a means of deducing the mechanical response of a tensile test specimen by use of ring data and vice versa.

This work aims at establishing such a relationship for an anelastic material. Similar methods have been described by Nadai;⁹ however, as explained earlier, the degree of accuracy applied is marginal. Our approach is based on an analytical, elastic analysis of the ring geometry in plane stress that provides all components of the stress tensor.

The anelastic analysis retains the elastically calculated normal forces and bending moments integrated over transverse cross sections (Fig. 1.16). Transverse cross sections that were originally plane are not required to remain so after load application (nonplanar deformation). The interrelating influence of both normal stress components σ_{rr} and $\sigma_{\theta\theta}$ have been accounted for. However, only the tangential stress distribution is redistributed in compliance with the condition that the radially integrated forces F and the radially integrated moments M be conserved. The anelastic tangential strain distribution is essentially composed of four parts:

- an elastic part, employing an average Young's modulus;
- a part that establishes the influence of the stress component σ_{rr} by lateral contraction;
- a part that, like the elastic part, effects a nonplanar deformation; and

Fig. 1.16. Symmetry Section of a Diametrically Loaded Ring.

- a part that effects a plane deformation and that is comparable to the solution used in pure bending.

The four contributions are evaluated as a function of an incrementally applied load and are integrated with respect to the load variable to obtain a total strain function.

The analysis has been implemented in the form of a FORTRAN computer program, and preliminary results that are based on an assumed nonlinear stress-strain relationship are discussed.

1.1.5.2 Linear Elastic Analysis of a Ring

The linear elastic analysis has been performed and described by Timoshenko and Goodier,¹⁰ in two steps: First, the two stress distributions of the diametrically applied point loads and the stress distribution resulting from an external hydraulic tension are superposed to provide an elastic solution of a diametrically loaded solid disk. Second, a fourth stress distribution is superposed to yield the solution for the hollow-ring problem. This last step involves a Fourier analysis of the radial and shear stresses of the disk at the location of the inner-ring boundary. It also involves a match of this Fourier series with the regular (i.e., nonsingular) general solution of Airy's stress function in cylindrical coordinates, to meet the boundary conditions on the inner ring surface. The displacement field is evaluated by numerically integrating the elastically derived strains. The total forces F and total bending moments M , acting on transverse planes, are computed as a function of the arc between 0 and 90°:

$$F(\theta) = \int_{r_1}^{r_2} (\sigma_{\theta\theta} - \nu\sigma_{rr}) dr \quad (13)$$

$$M(\theta) = \int_{r_1}^{r_2} r(\sigma_{\theta\theta} - \nu\sigma_{rr}) dr \quad (14)$$

These values serve as an input to the subsequent anelastic analysis.

1.1.5.3 Anelastic Analysis

The ring fails under critical load conditions because the tangential stress $\sigma_{\theta\theta}$ on the inner ring surface at $\theta = 0^\circ$ exceeds critical values. Apart from the point of load application, both the radial stress components σ_{rr} and the shear stresses $\sigma_{r\theta}$ are relatively small. Therefore, only the tangential stresses are redistributed.

The problem of an anelastic analysis is essentially a problem of redistribution of stresses, leaving the integral values F and M unchanged. The difficulty can be phrased in the following way: If one were simply to evaluate strains from the nonlinear stress-strain relationship without further restrictions, the strains would not comply with the symmetry conditions. For the case of the ring, the two invariant symmetry planes at 0 and 90° would deform into arbitrarily shaped surfaces.

To solve the problem, it is advantageous to proceed in the opposite way; that is, rather than immediately to calculate strains from stresses, first devise a strain distribution that complies with the symmetry conditions and contains some internal degrees of freedom (i.e., arbitrary constants to be fixed). These degrees of freedom are eliminated by using the stress-strain relationship to obtain the corresponding stress distribution and then imposing, among others, the conditions of conservation of the total forces and moments. As mentioned before, the initial determination of the shape of the tangential strain distribution is always subject, first, to the symmetry conditions; that is, the transverse section on the symmetry planes at $\theta = 0^\circ$ and $\theta = 90^\circ$ are permitted to undergo only a radial translation, not a deformation or a rotation.

In general, other conditions must be met and could be imposed initially, such as the conservation of shear forces or the requirement that the nonlinear solution does not appreciably deviate from the elastic solution except in regions of high stress. It is only necessary that the number of integral conditions imposed on the stresses be equal to the number of degrees of freedom initially inserted into the strain distribution function. This latter number is subject to considerable personal judgment as to how much freedom of deformation is in the material. The freedom

of deformation is certainly different in the case of diametral compression from that of the same material used in a uniaxial tensile specimen. Therefore, the results of the current analysis depend on the chosen number of degrees of freedom in the strain distribution and, hence, may differ from results of another analysis that might impose other integral conditions in a somewhat different manner.

For example, the usual approach used in the theory of pure bending¹¹ is to demand that all transverse planes remain plane after deformation; then, specifically, the planes at $\theta = 0^\circ$ and $\theta = 90^\circ$ remain plane, which leads to a simple hyperbolic strain distribution ϵ as a function of radius:

$$\epsilon = c'[1 - (r'_n/r)] . \quad (15)$$

The approach assumes a neutral fiber at the radius r'_n and allows the amount of strain to be adjusted by means of the parameter c' . The two degrees of freedom involved are necessary and sufficient to meet the condition that the total forces and moments are conserved. The two degrees of freedom are counted as pertaining to each transversal cross section.

1.1.5.3.1 Elastic Contribution. In this analysis more than two degrees of freedom are involved. The amount of admixture of an elastic solution is a degree of freedom, which is independently and initially fixed by the premise that the elastic part of the response is characterized by an average value of the modulus of elasticity. This and two additional degrees of freedom are assigned to the whole ring. Other additional degrees of freedom of the type used in Eq. (15) pertain, two each, to every transverse cross section. The linear elastic solution fully complies with the imposed symmetry conditions; predominantly, at low load values, the mechanical response is expected to be nearly elastic. Therefore, depending on the applied load, a portion of the elastic solution is included in the total anelastic solution. The amount of the elastic admixture $\epsilon_{\theta\theta}^{(1)}$ in the tangential strain distribution is determined by an average Young's modulus, which is in turn determined by the incrementally load-induced strain distribution.

1.1.5.3.2 Contribution from Lateral Contraction. There is a contribution of strains which originate from the radial stress distribution via lateral contraction:

$$\epsilon_{\theta\theta}^{(2)} = \exp\left(-\int_{r'}^r \frac{\alpha(s)}{s} ds\right) \int_{r'}^r \frac{1-\nu^2}{\rho} \frac{\sigma_{\rho\rho}}{E(\epsilon_{\rho\rho})} \exp\left(\int_{r'}^{\rho} \frac{\alpha(s)}{s} ds\right) d\rho, \quad (16)$$

where ρ and s (radii) are dummy integration variables, and the radial function $\alpha(r)$ is given by

$$\alpha(r) = \{1 + \nu[E(\epsilon_{\theta\theta})/E(\epsilon_{rr})]\}, \quad (17)$$

and the Poisson's ratio ν and Young's moduli E are variables evaluated at strain values $\epsilon_{\theta\theta}$ and ϵ_{rr} . Obviously, there would be no contribution $\epsilon_{\theta\theta}^{(2)}$ from Eq. (16) if the radial stress distribution were to be neglected; that is, $\sigma_{rr} = 0$.

1.1.5.3.3 Redistribution as a Function of Both Coordinates r and θ . As explained in Sect. 1.5.2, the elastic solution is composed of two parts: a singular (disk) contribution containing the effects of the point loads and a regular contribution (ring) to satisfy the inner-ring boundary conditions. The latter component is a Fourier series derived from Airy's stress function (see Timoshenko and Goodier)¹² and, as such, complies with the symmetry conditions. The lowest term in the Fourier series was chosen to contribute to a redistribution of tangential stresses. This term and another, which is introduced to account temporarily for the simultaneous influence of the fourth contribution, provides a strain distribution, $\epsilon'_{\theta\theta}$:

$$\epsilon'_{\theta\theta} = c''' \exp\left(-\int_{r_1}^r \frac{\alpha(\rho)}{\rho} d\rho\right) + [d + e(r/\bar{r})^{-4}] \cos 2\theta, \quad (18)$$

with $\bar{r} = (r_1 + r_2)/2$ being the average radius. The three constants c''' , d , and e are determined in the following way: Initially, these constants are required to produce a stress distribution $\sigma_{\theta\theta}$ with resi-

dual forces $F^{(3)}$, moments $M^{(3)}$, and a stress distribution that represents a close fit to the elastic stress distribution. In this manner, the constants are determined in sets of three for each transverse cross section. Then, because the constants $\bar{d}$ and $\bar{e}$ must be true constants for all transversal cross sections, the average values $\bar{d}$ and $\bar{e}$ are employed to form a third contribution:

$$\epsilon_{\theta\theta}^{(3)} = [\bar{d} + \bar{e}(r/\bar{r})^{-4}] \cos 2\theta. \quad (19)$$

The influence of the first term in Eq. (18) is finally assessed in the fourth and last contribution to the strain function, to which we now turn our attention.

1.1.5.3.4 Redistribution as a Function of the Radius Only. All residual forces $F^{(4)}$ and moments $M^{(4)}$ are taken into account by the fourth contribution to the tangential strains:

$$\epsilon_{\theta\theta}^{(4)} = \exp\left(-\int_{r_1}^r \frac{a(\rho)}{\rho} d\rho\right) \left[c - \epsilon_{\theta\theta}^0 \int_{r_1}^r \rho^{-1} \exp\left(\int_{r_1}^{\rho} \frac{a(s)}{s} ds\right) d\rho \right] + \epsilon_{\theta\theta}^0. \quad (20)$$

The constants c and $\epsilon_{\theta\theta}^0$ are determined by the condition that $\epsilon_{\theta\theta}^{(4)}$ is related to a stress distribution through the force $F^{(4)}$ and the moment $M^{(4)}$ for each transverse cross section. The constant $\epsilon_{\theta\theta}^0$ has the significance of a constant strain contribution for each transverse cross section.

1.1.5.3.5 General Remarks. It may be useful to point out a few relationships in order to understand the current approach as compared with the well-known procedure applied in pure bending.

In returning to pure bending conditions, no normal forces would be acting on the transverse planes; consequently, the constant strain contributions $\epsilon_{\theta\theta}^0$ are identically zero, and Eq. (20) assumes the same form as the first term of Eq. (18). This explains why the first term in

Eq. (18) is only initially used and is subsequently apparently neglected. In fact, the first term in Eq. (18) serves as a first-order approximation to the fourth contribution $\epsilon_{\theta\theta}^{(4)}$, for which the latter is eventually substituted; it was temporarily introduced to determine the constants $\bar{d}$ and $\bar{c}$. Returning to a linear stress-strain relationship, the radial function $\alpha(r)$ becomes a constant $\alpha = 1 + \nu$, and Eq. (20) reduces to

$$\epsilon_{\theta\theta}^{(4)} = c' [1 - (r'_n/r)^\alpha] , \quad (21)$$

where c' and r'_n are constants related to the constants c and $\epsilon_{\theta\theta}^0$ and where $\epsilon_{\theta\theta}^{(4)}$ incorporates $\epsilon_{\theta\theta}^{(0)}$.

Furthermore, if the influence of the lateral forces is also neglected, the value for α would be 1 and Eq. (21) assumes the form of Eq. (15).

1.1.5.3.6 Completion of the Stress and Strain (Displacement) Tensors.

After establishing an anelastic tangential strain distribution $\epsilon_{\theta\theta}$, it is relatively easy to determine the remaining components of the stress and strain tensors:

$$\sigma = \begin{pmatrix} \sigma_{rr} \\ \sigma_{\theta\theta} \\ \sigma_{r\theta} \end{pmatrix} \quad \epsilon = \begin{pmatrix} \epsilon_{rr} \\ \epsilon_{\theta\theta} \\ \epsilon_{r\theta} \end{pmatrix} . \quad (22)$$

The components are calculated as follows:

(1) The elastically calculated radial stresses σ_{rr} are considered to be small in value. They were not subjected to a redistribution process, and the original values are retained.

(2) By employing the nonlinear stress-strain relationship, it is possible to calculate the tangential stresses $\sigma_{\theta\theta}$ from $\epsilon_{\theta\theta}$ and σ_{rr} :

$$\epsilon_{\theta\theta} E(\epsilon_{\theta\theta}) = \sigma_{\theta\theta} - \nu \sigma_{rr} . \quad (23)$$

(3) The radial strains ϵ_{rr} are also determined by employing the nonlinear stress-strain relationship

$$\epsilon_{rr} E(\epsilon_{rr}) = \sigma_{rr} - \nu \sigma_{\theta\theta} . \quad (24)$$

(4) Like the radial stress σ_{rr} , the elastically calculated shear stresses are not subjected to a redistribution and retain their original values.

(5) Shear strains can be evaluated from the stress-strain relationship as follows:

$$\epsilon_{r\theta} E(\epsilon_{r\theta}) = 2(1 + \nu) \sigma_{r\theta} . \quad (25)$$

Displacements are obtained by integrating the normal strains, employing the usual expressions for cylindrical coordinates.¹² The evaluation of displacements serves as a good check on the computer program and the algorithm. As initially required, the two symmetry planes at $\theta = 0$ and 90° should be merely radially displaced and neither deformed nor rotated. In these calculations this check was positive for all locations, excluding the singular point at the load application.

An alternative way of determining the shear strains does not involve the stress-strain relationship, that is, from the displacements u and v .

$$\epsilon_{r\theta} = \frac{\partial u}{r \partial \theta} + \frac{\partial v}{\partial r} - \frac{v}{r} . \quad (26)$$

These shear strains would be generally somewhat different from the ones obtained from Eq. (25) because the compatibility relationship was not enforced except at the inner-ring boundary, where $\epsilon_{r\theta} = 0$.

1.1.5.4 Numerical Results

To explore the typical properties of our approach to calculate anelastically the stresses in a diametrically compressed ring, a nonlinear stress-strain relationship was assumed (Fig. 1.17). The figure

Fig. 1.17. Nonlinear Stress-Strain Relationship.

shows that the tensile part of the curve is continued beyond the yield point, which was done to study the influence of a possible softening process prior to a crack opening. No experimental results have yet been obtained on this aspect.

A load of 47.244 N per millimeter of thickness was applied to a 3.18-mm-ID, 10.16-mm-OD ring. As a result, an elastically calculated tangential stress distribution $\sigma_{\theta\theta}$ was obtained (Fig. 1.18). The highest stress value is reached on the inner-ring surface at $\theta = 0^\circ$ and amounts to 30.3 MPa, which is 87% of the critical stress value (35 MPa) that may be obtained from the termination point of the tensile stress-strain curve (Fig. 1.17). In approaching the outer-ring surface close to the point of load application, the curves of Fig. 1.18 are influenced by the presence of the nonphysical singularity.

It is of interest to compare the elastically calculated stress distribution with that obtained by using the theory of pure bending of curved bars (Fig. 1.19). The highest value in stress amounts to

Fig. 1.18. Elastically Calculated Tangential Stress Distribution.

Fig. 1.19. Tangential Stress Distribution According to the Theory of Pure Bending of Curved Bars.

34.8 MPa. Elsewhere the solution is not in good agreement with the exact elastic solution (Fig. 1.18). In performing the anelastic calculations, a highest stress value of 66% of the critical stress value is obtained (Fig. 1.20).

In another calculation the load was increased to twice its original value. As a result (Fig. 1.21), the highest value in stress attained is 99% of the critical value. The calculations disclose to what extent the highest stresses are reduced if the material is allowed to react anelastically.

Figure 1.22 shows how the tangential stresses at three selected points are increased as the load is increased. Obviously, as a function of load, the degree of nonlinearity is different for different locations.

Fig. 1.20. Anelastically Calculated Tangential Stress Distribution.

Fig. 1.21. Anelastically Calculated Tangential Stress Distribution (Double Load as Compared with Fig. 1.20).

Fig. 1.22. Tangential Stresses at Selected Points as a Function of Load.

This statement also applies to the radial displacements of those points shown in Figs. 1.23 and 1.24. The displacements of the inner and outer surface at $\theta = 90^\circ$ are shown in Fig. 1.23. The curves are nonlinear; however, on the inner-ring surface at $\theta = 0^\circ$, the nonlinearity in the radial displacement component is not obvious. If the calculations are in accordance with the experimentally observed response, the measurements of displacements only at this location can lead to a false conclusion that the material behaves linearly.

1.1.5.5 Summary and Conclusions

The analysis described here of an anelastic ring is different from the usual approach, which is commonly based on the theory of pure bending

Fig. 1.23. Radial Displacements of the Inner and Outer Surface at $\theta = 90^\circ$.

Fig. 1.24. Radial Displacement at the Location of Highest Stress.

of curved bars. The current anelastic analysis utilized a preceding linear elastic calculation. Also, because there is more than one way to redistribute the elastically derived stresses, this method is not unique in its algebraic form. Therefore, it remains to be shown whether or not the predicted mechanical response is in satisfactory agreement with experimentally derived values. Such experiments would include the measurement both of uniaxial stress-strain curves and of a number of displacement curves for the ring geometry. The objective is to show that these displacement curves can be obtained by using the uniaxial stress-strain data as input.

Another and ultimate goal of the current analysis is also to describe the mechanical response of a nonuniformly oxidized graphite

ring. Such an analysis would differ from the solution for a ring of homogeneous material by an input of a number of uniaxial stress-strain curves that describe the various levels of oxidation in the ring. In other words, the stress-strain curve should be provided with at least one, and possibly two, additional space variables, that is, the coordinates r and θ . We hope that such a function can be determined by initially measuring the density distribution of the oxidized ring. The density distribution could then be correlated to a corresponding distribution of Young's modulus, and, by retaining the same degree of nonlinearity throughout the oxidized material, the required uniaxial stress-strain curves could be established.

The described algorithm was implemented in the form of a FORTRAN computer program. A more detailed description of the program will be provided in a future publication.

1.1.5.6 Acknowledgment

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1.1.6 Fracture Propagation and Modeling — J. D. Buch and D. Swanson, Aerospace Corporation

1.1.6.1 Purpose/Methodology

The purpose of this portion of the HTGR program is to define and demonstrate the mechanics of oxidation-induced structural degradation and to provide analytical correlations for life prediction.

The methodology is a balance between theoretical studies and direct observations of crack propagation in the as-received and the oxidized materials.

1.1.6.2 Theoretical Modeling

In many respects, the major progress to date has been the development of theoretical concepts that lead to new insights of how to characterize experimentally and subsequently to understand the results of

oxidation. Two areas of theoretical modeling being studied are the effects of porosity and pore geometry on (1) fracture path and fractography and (2) elastic modulus. These modeling efforts bring out the reasons why density and/or weight losses by themselves may inadequately represent the loss of material strength.

1.1.6.3 Elastic Modulus Model

The elastic modulus and fracture surface energy are basic to all fracture mechanics model concepts. The pores were assumed to be strain-relief centers that yield field compliance contributions, which later were computed by analogies to elliptical planar cracks. By incrementally considering all porosity contributions, an explicit Knudsen exponential relationship was derived:

$$E = E_0 \exp -[BP] . \quad (27)$$

The Knudsen relationship has long been a commonly used empirical data fit. The current work represents its first purely analytical derivation. The theory also provides an explicit relationship between the coefficient B and the pore/aspect ratio (a/c). Pore geometries currently treated are ellipsoids of revolution and cylinders.

An example of the results is given in Fig. 1.25, which displays the sensitivity of modulus to porosity and pore geometry. Overplotted on Fig. 1.25 are results on PGA graphite obtained under differing oxidation conditions. The apparent disparity between the different atmospheric oxidation conditions can be rationalized by the present theory as being the results of differing pore structures. Unfortunately pore geometries are rarely reported in an experimental program; thus, appropriate measurements should be taken in this direction for any future work.

1.1.6.4 Fracture Path Model

An idealized fracture path model was constructed for a two-dimensional material with equispaced noncircular pores. The model's contribution of pores to the fracture surface was found to be considerably in excess of

Fig. 1.25. Theoretical Predictions of Porosity and Pore Shape Effects on Modulus and Data from PGA Experiments in Various Atmospheres.

the porosity for essentially all pore geometries. The essential point was to emphasize the potential use of detailed information on pore shape, and confirming experiments were conducted.

1.1.6.5 Experimental

A considerable effort was devoted to equipment development to overcome some previously unnoticed deficiencies in the high-force mechanical loading stage for the scanning electron microscope as well as to provide the size capability to test full-scale brittle rings that form the basis of the ORNL data base. These developments were all successfully completed.

The improved capabilities were used to provide direct real-time observations of crack propagation as well as to form the basis for

quantitative fractography in conjunction with the theory. For unoxidized materials, the theory and the experimental results are given in Fig. 1.26. As noted, the pores occupy a significantly greater fraction of the fracture path than would be anticipated on the basis of porosity. The theory appears to have the capability of rationalizing the observed differences between the graphite grades. More important, the theory has the capability to indicate the role of oxidation-induced porosity from both the aspects of porosity and pore shape changes. The raw data, of course, are the crack propagation studies as given in Fig. 1.27.

In addition to the main body of the effort described above, support was provided to ORNL on the nature of the local impurity contamination of the observed localized pitting and wormhole formation in oxidation experiments. These findings have been incorporated into Sect. 1.1.2.

1.1.6.6 Conclusions

The program results are encouraging in that they demonstrate an ability to deal with the details of porosity changes other than simple bulk density. In fact, the results demonstrate the absolute need to characterize fully the geometry of the porosity to (1) understand the available direct observations of crack propagation, (2) rationalize conflicting data on oxidation effects (as presented in Fig. 1.25), and

Fig. 1.26. Contribution of Porosity to Fracture Surface as Functions of Porosity and Pore Shape (Theory and Experiment).

Fig. 1.27. Observations of Fracture Path in 2020 Graphite as Raw Data for Quantitative Analysis (500 $\times$).

(3) determine the applicability of short-term laboratory data to long-term structural performance.

1.2 IODINE SORPTION AND DESORPTION ON LOW-ALLOY STEEL — M. F. Osborne and R. B. Briggs*

As in all reactor systems, iodine is a potential radiation hazard in the HTGR under both normal operation and accident conditions. Although efficiently contained by the pyrocarbon coatings on the fuel particles, a small fraction is released to the helium coolant and distributed around the coolant circuit. The objectives of this task are

* Consultant.

(1) to determine equilibrium loadings of iodine on metal surfaces under relevant conditions and (2) to measure rates of desorption under conditions that may be related to accident situations. These data are needed as input for computer calculations to evaluate the hazards of iodine release as related to both reactor licensing and operation.¹³

Applicable literature data in the range of interest for the HTGR are limited. Although the partial pressure of iodine P_I is expected to be $<10^{-10}$ atm, most previous work had been conducted at much higher P_I ($>10^{-7}$ atm), where experimental measurements were correspondingly simpler to obtain. However, as recognized by Hoinkis,¹⁴ the behavior of significant gaseous iodine forms (I, FeI_2 , and HI) varies considerably over this range of P_I .

The computer program SOLGASMIX-PV¹⁵ was used to calculate the equilibrium partial pressures of the principal iodine species in an isothermal system at typical HTGR conditions: temperatures of 200 to 800°C, a helium pressure of 50 atm, a hydrogen partial pressure of 5×10^{-4} atm, and 10^{-10} to 10 mol of iodine in the system. The results of these calculations at an intermediate temperature, 400°C, are plotted in Fig. 1.28. The dominant gaseous species in all cases was hydrogen iodide. As the system loading was increased above 1 mol, most of the iodine condensed as $\text{FeI}_2(\text{s})$, thereby limiting the partial pressures of gaseous species. At lower system temperatures, this FeI_2 condensation point was correspondingly lower. Therefore, in a real coolant circuit with a range of temperature regions, FeI_2 would tend to deposit in the lower-temperature regions, and the partial pressures of all species in the gas phase would be reduced.

An experimental apparatus and procedure were developed to measure the deposition of iodine on metal specimens from flowing helium.¹⁶ Purified helium passed through a low-temperature saturator containing elemental iodine traced with ^{131}I . This stream was diluted by a larger stream of helium before it passed across metal specimens mounted in a quartz furnace tube. The metal specimens were composed of thin plates in a rectangular array to facilitate counting by shielded sodium iodide detectors.

Fig. 1.28. Partial Pressures of the Principal Iodine Species in Equilibrium with Iron at 400°C in an Atmosphere of He + 500 ppm H₂.

A series of experiments was conducted at 400°C, either in purified helium or in helium containing a small fraction of hydrogen. Equilibrium loadings with iodine burdens ($P_I + 2P_{I_2} + P_{HI} + 2P_{FeI_2}$) in the range 10^{-10} to 10^{-7} were determined, and the results are shown in Fig. 1.29. Data from other studies¹⁷⁻²⁰ of iodine sorption on low-alloy steels, generally at higher iodine burdens, are shown for comparison. These plotted data are based on the geometric surface area. The real surface

Fig. 1.29. Iodine Sorption on Low-Alloy Steel at 400°C.

area of our specimens, as determined by the stylus technique, is about 6.8 times greater; presumably, the specimens used by other investigators would have comparable correction factors. Based on a previous analysis²¹ that assumed an ionic radius for the iodine atom of 2.16 Å and an arrangement to form iodine-to-iron bonds, one monolayer should be about 0.086 $\mu\text{g I}/\text{cm}^2$ (true surface area). Thus, to compare with the plotted data, one monolayer should be about 0.5 $\mu\text{g I}/\text{cm}^2$. In any event, the larger loadings ($\sim 10 \mu\text{g}/\text{cm}^2$) would be expected to be largely condensed FeI_2 .

Varying surface conditions are thought to be the reason for most of the scatter in the data. We have observed approximately 10 times higher loadings under reducing, compared with slightly oxidizing conditions. The presence of surface oxides should reduce the number of sites available for iodine-to-iron chemisorption.

Desorption from these same specimens has been measured in a few experiments. Typical desorption behavior under reducing conditions is

Fig. 1.30. Iodine Deposition to Equilibrium Loading, Followed by Thermal Desorption.

illustrated in Fig. 1.30; following a temperature increase of from 400 to 600°C, about 75% of the iodine desorbed, and an equilibrium loading at the higher temperature was approached. However, desorption from slightly oxidized specimens proceeded much more slowly, perhaps indicating that the iodine was located within or below the oxide layer.

1.3 ADSORPTION OF IODINE ON GRAPHITE — R. A. Lorenz and R. Towns

Experimental apparatus was assembled, tested, and used to measure the adsorption of iodine at low partial pressures on type H451 graphite. A simplified diagram of the apparatus is shown in Fig. 1.31. By adjusting purified helium flow rates through the saturator tube (iodine

Fig. 1.31. Apparatus for Iodine Sorption on Graphite.

source) and dilution stream and by varying the saturator temperature between -35 and -70°C , iodine partial pressures between 10^{-7} and 10^{-9} atm can be obtained easily. Two runs were made primarily for apparatus checkout. Run 3 was made to test the sorption of iodine on H451 graphite heated to 428°C in dry purified helium. Details of the conduct of run 3 follow.

1.3.1 Preparation of the Iodine Source

Radioactive carrier-free ^{131}I as NaI in 0.7 ml H_2O was obtained from ORNL Isotope Sales on September 12, 1979. We measured the activity to be 7.11 mCi referenced to September 12, 1979, at 0800, very close to the 7 mCi requested. The addition of carrier and conversion to the I_2 chemical form was performed essentially as described in Appendix A of ORNL/TM-5094.²² The completed source contained 1630 μg of iodine and was divided as follows: standard HS-1 contained 71.2 μCi (all activities are referenced to 9-12-79 at 0800); standard HS-2, 68.7 μCi ;

and standard HS-3, 71.2 μCi . The standards were separated from the stock NaI solution after the carrier was added but before conversion to I_2 . The I_2 source H-1 contained 3520 μCi ^{131}I (807 μg ^{131}I) and was used by M. F. Osborne to measure the sorption of iodine on steel; I_2 source H-2, containing 3500 μCi ^{131}I (802 μg ^{131}I), was used in sorption studies on H451 graphite.

1.3.2 Description of Procedure for Run 3

The quartz and Pyrex components were cleaned with both a basic rinse and a nitric-acid rinse. Previously used components were checked to be certain that no traces of ^{131}I activity remained from runs 1 and 2. A background count rate of 1.28 c/s (counts per second) was measured on the single-channel analyzer. The graphite specimen (Fig. 1.32) was installed and held in place against a quartz retainer by a stainless steel coil spring located in the large ball joint, applying pressure through the charcoal filter cartridge and a quartz retainer tube. Source H-2 was fused into the system, and purified helium was used to purge entrapped air. A mechanical vacuum pump was used to pull a vacuum of about 10 μm Hg while the graphite was heated to 250°C.

ORNL DWG 80-480

Fig. 1.32. Closed-End Cylinder of H451 Graphite (12 Units Needed).

The zirconium chip purifier was heated to 500°C, helium flow started at 1 atm and 30 cm³/min for about 24 h. The procedure for transferring iodine from the source tube to the saturator tube was as follows: The dilution flow was reduced to 10 cm³/min, and the bypass valve was opened to allow flow to a charcoal trap located in front of a hole in the shielding of the counting crystal. The outlet valve from the furnace was closed. With the saturator refrigerator at maximum cooling (-84°C), the saturator flow was adjusted to 6 cm³/min, and the break-seal on the source tube was broken. Heating tape was used to raise the iodine source to about 120°C for 5 h. A torch was used to seal off and remove the source tube.

Volatile iodine components were purged out of the saturator tube by raising the temperature to -50°C and allowing the helium flow to continue for about 4 h. The bypass charcoal filter was replaced, and iodine transport rate followed, using the counting crystal and its measured efficiency (150 c/s, μ Ci), until the rate of transport decreased to the level expected for a pure I₂ source at -50°C. The bypass filters collected 1.7 μ g I during the loading operation at -84°C and 4.9 μ g while purging at -50°C.

The saturator temperature was then reduced to -65°C in preparation for a test run at an iodine partial pressure of 10⁻⁹ atm (10⁻¹⁰ MPa). Fresh bypass filters were used to monitor the iodine partial pressure. When equilibrium was reached at the target iodine pressure, flow was changed from the bypass stream to the furnace. Sorption equilibrium was reached on the graphite sample in 3.5 d. At that time, the radiation detector signal was diverted from the single-channel analyzer to a TP-5000 analyzer system to obtain a more accurate count. This change was necessary because the background was increased as a result of the presence of scattered radiation from iodine deposited on the backup filter and on the cool ends of the quartz furnace tube.

Similar procedures were used to make sorption tests at 10⁻⁸ and 10⁻⁷ atm iodine partial pressure. A run was attempted at 3 $\times$ 10⁻⁷ atm, but the saturator became depleted of iodine before sorption equilibrium was attained. The saturator flow was then turned off and cooled to -84°C, and dilution flow at 50 cm³/min was used to desorb iodine from the graphite.

1.3.3 Run 3 Results

Results of run 3 are summarized in Table 1.12, and a plot of the sorption isotherms calculated from the data is shown in Fig. 1.33. An iodine mass balance for run 3 is itemized in Table 1.13 and shows that 94% of the iodine was accounted for at the end of the run. The sorption isotherm obtained in this test at 400°C for H451 graphite shows the same iodine pressure dependence measured for H327 graphite powder.²²

1.4 DIFFUSION OF FISSION PRODUCTS IN GRAPHITE

1.4.1 Diffusion of Cesium in Graphite — R. B. Evans III, W. Davis, Jr., and A. L. Sutton, Jr.

Recovery has been completed of the data on diffusion of cesium in graphite from experiments performed by the late R. B. Evans III (prior to January 1977) and A. L. Sutton, Jr. A topical report presents the data, the mathematics derived by Evans, and partial data analysis. The report has been reviewed by three staff members at ORNL and is now being edited prior to publication. The abstract of the report is as follows:

During the two years prior to his death in January 1977, R. B. Evans III, in part-time collaboration with A. L. Sutton, Jr., performed experiments on diffusion of ^{137}Cs in five types of graphite, namely, Hawker-Siddeley HS-1-1; Great Lakes Carbon Company H327, H451, HLM85; and National Carbon Company ATJ. At the time of this death, Evans had written about one-half of a topical report; he had not yet analyzed or summarized the data. The present document provides a completion of the report that Evans had started and includes a presentation of all of the diffusion data, previously unpublished, obtained by Evans and Sutton.

Except for data on mass transfer of ^{137}Cs in the Hawker-Siddeley graphite, analyses of experimental results were initiated but not completed. The mass transfer process of cesium in HS-1-1 graphite at 600 to 1000°C in a helium atmosphere is essentially pure diffusion wherein values of $(D/\epsilon)_0$ and ΔE of the equation

$$D/\epsilon = (D/\epsilon)_0 \exp[-\Delta E/RT]$$

are about $4 \times 10^{-2} \text{ cm}^2/\text{s}$ and 30 kcal/mol, respectively.

Table 1.12. Summary of Results for Iodine Sorption in Run 3

Saturator temp.	Iodine partial pressure in saturator	Saturator flow rate (cm ³ /min)	Dilution flow rate (cm ³ /min)	Iodine partial pressure at specimen	Iodine on graphite at steady state		Time to reach steady state (s)	Total time run at pressure (s)	Volume of He flowed through saturator at this temp. (liter)	Estimated mass of I ₂ transported (μg)
					(μg) ^a	(μg/g) ^b				
-67	4.4×10^{-9}	9.6	34	0.96×10^{-9}	0.13	0.0203	2.8×10^5	5.4×10^5	124	5.7
-58.3	2.0×10^{-8}	9.6	34	4.30×10^{-9}	0.157 ^c	0.025 ^c	Not reached	1.32×10^5	30	6.3
-53.2	4.6×10^{-8}	9.6	35	0.98×10^{-8}	0.41	0.065	7.26×10^5	1.05×10^6	240	117
-38.0	4.2×10^{-8}	9.7	35	0.91×10^{-7}	1.23	0.190	3.45×10^5	1.76×10^5	41	182
		3.1	35	0.91×10^{-7}				1.76×10^5	13	58
-29.8	1.3×10^{-6}	3.0	11	2.79×10^{-7}			Not reached	1.40×10^5	10	135
-29.8	1.3×10^{-6}	3.0	11	2.79×10^{-7}			Not reached	2.54×10^5	17	(245) ^d

^a Measured on TP-5000 multichannel analyzer.^b Mass of specimen was 6.04 g.^c Estimated.^d Actual amount transported was probably less because decreasing activity on H451 graphite suggested depletion of iodine in the saturator.

Fig. 1.33. Partial Iodine Isotherm on H451 Graphite at 400°C.

Table 1.13. Observed Mass Balance of Iodine
in Run 3

Location	Mass (μg)
Source tube residue	44
H451 sample	0.68
Furnace tube	22
Sample retainer tube (mostly at unheated end)	133
Charcoal filter No. 1	444
Retaining spring	3
Charcoal filter No. 2	13
Socket joint	1
All bypass filters used to expel original iodine and to calibrate pressure	8
Amount remaining in saturator	<u>67</u>
Total	735
Percentage of original mass	94

Evans has presented a model of diffusion coupled with irreversible first-order fixation of cesium to explain data on mass transfer of ^{137}Cs in some of the other graphites. We tested this model by use of nonlinear least squares to estimate diffusion and reaction rate parameters of three sets of data, one on ATJ and two on H327 graphites. Although the computer program did calculate these parameters, the fit of data to model was not good. The inadequacy of the model is its inability to account for a very steep ^{137}Cs concentration gradient at very small penetration distances. Further analyses of the data will clearly require the use of numerical techniques that permit a more complicated mechanism for cesium trapping than that given by irreversible first-order kinetics.

Three experiments were performed to test for vapor-phase transport of cesium in H451 graphite at 635 to 680°C. In each case, such transport was less than the limits of measurement.

1.4.2 Silver Transport in HTGR Core Graphite -- Rion Causey*

1.4.2.1 Procedures and Results

Diffusion profiles for ^{110m}Ag in H451 graphite have been measured. The graphite samples were hollow cylinders, with both ends sealed. The seal on one end consisted of a screw cap with a graphon gasket. The source of silver inside the cylinder was silver-laden graphite powder. This powder was prepared by decomposing AgNO_3 on a graphite disk with subsequent crushing.

The graphite samples were isothermally annealed in a vacuum of approximately 50 mtorr. At the end of each anneal, the samples were opened and the powder removed. The ends of the hollow cylinders were removed to avoid corrections for axial diffusion. The cylinders were then sectioned radially in 10- to 20-mil layers by using a Unimat III lathe with an attached precision indicator. The graphite was caught during sectioning by a filtered suction line. After each section, the filter was removed and gamma counted to give differential count rates as a function of radial distance.

The results are shown in Figs. 1.34 and 1.35. The maximum concentration of silver in graphite did not exceed 10^{-9} g/g in any of the

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Fig. 1.34. Radial Diffusion Profile of ^{110m}Ag in H451 Graphite.

Fig. 1.35. Radial Diffusion Profile of ^{110m}Ag in H451 Graphite.

experiments. Figure 1.36 shows a comparison of experiments by using samples cut "with" and "across" the line of extrusion. Because the experiments measured radial diffusion profiles, the samples labeled "with" actually measured the diffusion across the line of extrusion. The samples labeled "across" measured diffusion both with and across the line of extrusion.

An Arrhenius plot of the diffusion data is shown in Fig. 1.37. The values for the diffusion coefficients were determined by the simple expression $D = x^2/4t$, where x is the point at which the concentration goes to zero and t is the duration of the isothermal anneal. Initial results from computer analysis of a least-squares fit to the analytical solution give values that are consistently lower by about one order of magnitude.

Fig. 1.36. Radial Diffusion Profile of ^{110m}Ag in H451 Graphite.

Fig. 1.37. Arrhenius Plot for ^{110m}Ag in H451 Graphite.

1.4.2.2 Discussion

The diffusion profile technique appears to be a viable means of measuring the silver diffusion coefficients in graphite. Experimental duplication showed the results to be consistently reproducible.

The most surprising results so far are the very high values measured for the apparent diffusion coefficients. Even if the computer results show the values to be lower by an order magnitude, they will still be high for the temperatures at which measurements have been performed. These results lead to the conclusion that diffusion is controlled by silver migration through the graphite pores.

Although diffusion may be controlled by the pores, the diffusion profiles do not deviate greatly from results expected from Fickian or classical diffusion. This can be explained by a model in which the silver is assumed to be adsorbed on the pore surface. The rate at which the silver atoms break free from the adsorption sites is analogous to the Fickian jump frequency. The mean distance that the atoms travel between adsorption sites would then be analogous to the diffusional mean free path. For such a model to hold there would have to be no migration into the bulk and little closed porosity.

Results for diffusion with an across-the-line extrusion are almost identical, which should not have been totally unexpected because the porosity in the graphite is not significantly anisotropic. The permeability may be affected to a greater extent because of the amount of material transported being directly proportional to the number of open pores traveling in the direction of interest.

Experiments are currently under way to measure desorption isotherms of silver from graphite. This data should help to explain the mechanisms of diffusion. Problems are currently being encountered with metal sample tubes having a greater affinity for silver than does the graphite.

1.5 CARBON-14 PRODUCTION AND DISTRIBUTION IN THE PEACH BOTTOM HTGR CORE — R. P. Wichner and F. F. Dyer

A study of the inventory of ^{14}C in the Peach Bottom HTGR core has been completed, and a final report²³ is being prepared. The abstract of this report follows.

Carbon-14 concentrations were measured in a variety of core components during the course of the Peach Bottom Surveillance Program. Five fuel element sleeves and spines were dissected to yield radial concentration profiles at four axial locations each. The profiles show that the major part of the ^{14}C in these graphite components characteristically showed a flat radial distribution across the interior of the member; the balance was found in concentration peaks at each exposed surface. The source of these surface peaks is not fully understood.

Concentrations were also determined in five fuel element fission product traps, in one removable radial reflector block, and in 12 fuel particle pairs taken from a number of axial

locations in one fuel element. Estimates of whole core inventory were made from these concentration determinations. Most surprising was the comparatively high ^{14}C inventory associated with fuel particles, which must be ascribed to nitrogen contamination of the fuel. Also unexpectedly high was the level found in the fuel element charcoal traps, which, despite its small mass, contained almost as much ^{14}C as all the graphite sleeves and spines. On a per-unit-energy basis, an estimated 113 Ci/GW(t)-year of ^{14}C was produced in the Peach Bottom HTGR, which is about 30% higher than predictions based on the ORIGEN code. The difference seems due mainly to the assumed level of nitrogen contamination in the fuel.

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2. FUELED GRAPHITE DEVELOPMENT

F. J. Homan

2.1 INTRODUCTION

The transition discussed in earlier annual reports^{1,2} is completed, and the ORNL program is fully integrated into the national effort to develop and qualify fresh and makeup fuel for the HTGR. The emphasis of the ORNL program centers on irradiation testing, specific development activities associated with fissile kernels and PyC and SiC coatings, equipment and procedure development for postirradiation examination (PIE) and performance assessment, and specific areas of work under the U.S.-Federal Republic of Germany (FRG) cooperative program on fuel development. Significant accomplishments during the reporting period include:

- Completion and documentation of studies on PyC permeability.
- Development of a two-step process for the fabrication of dense UCO kernels, which permits control of the kernel density and phase composition, at sintering temperatures in the range of 1550°C. This development is significant because work being done elsewhere features sintering temperatures as high as 1900°C.
- Completion of studies of pyrocarbon coating in large-scale equipment by using CO₂ in the diluent gas. Processes have been developed that produce coatings of high density and low BAF_O (optical Bacon anisotropy factor) in production-scale equipment by using propylene diluted with CO₂ and argon. Irradiation testing will determine whether or not such coatings match the very favorable performance observed with batch OR 2013-T, which was produced in laboratory-scale coaters by using propylene and CO₂.
- Completion of out-of-reactor thermal-gradient heating studies by using Triso-coated doped kernels. These studies were designed to measure the fission product-SiC attack rates for a variety of fission product species.

- Collection of the first data from the postirradiation gas analyzer (PGA) system. This system was designed to measure the content of fission and reaction gases in irradiated coated particles by breaking the particles in an evacuated chamber coupled to a time-of-flight mass spectrometer.
- Completion of irradiated microsphere gamma analyzer (IMGA) analysis on fuel particles irradiated in a number of experiments.
- Completion of the experimental work with PWS FD-12 and -13 on fuel of German origin.
- Design and installation of an x-radiographic apparatus for use with irradiated coated particles.

2.2 IRRADIATION EXPERIMENTS — F. J. Homan

No new capsules were built during this reporting period; PIE of capsules HRB-14 and -15b was the major activity. In addition, IMGA analysis on particles irradiated in HRB-13, -11, and -12 and HT-34 was accomplished. This work, especially the examinations on HT-34 particles, has provided a wealth of data for the fuel performance models. Because of budget reductions, the size of the irradiation program has been reduced considerably over the past few years. The next capsules to be irradiated will be HRB-15a (scheduled for May 1980) and HT-35 (scheduled for June 1980). No ORR capsules will be scheduled until 1982. The Fort St. Vrain test elements were loaded into the reactor with segment seven in April 1979.

2.2.1 High-Flux Isotope Reactor Target Capsules — F. J. Homan

The routine PIE capsule HT-34 was completed last year,³ and IMGA analysis of particles irradiated in HT-34 was accomplished during this reporting period. The HT-31 and -33 final reports were published, and the HT-32 final report was written. A final report for capsule HT-34 will be completed in March 1980. The next HT capsule will be HT-35, which is scheduled to begin irradiation in June 1980. A test plan has

been completed for this capsule, and a request for approval in principle has been submitted to the Department of Energy (DOE).

2.2.1.1 Capsule HT-32 — E. L. Long, Jr.

The design, objectives, operating history, and postirradiation results of capsule HT-32 have been reported previously.⁴⁻⁶ During this reporting period a final topical report⁷ on this experiment was completed, the abstract of which follows.

The HT-32 experiment was an uninstrumented capsule irradiated for four cycles in the target position of the High-Flux Isotope Reactor (HFIR). The experiment was designed to:

- provide supplemental simulated fuel rods for thermal transport and expansion measurements,
- test fertile kernels fabricated in Germany with Al_2O_3 and SiO_2 additives for improved fission product retention,
- study the stability and permeability of low-temperature isotropic (LTI) pyrocarbon coatings,
- test Bisso- and Triso-coatings derived in a large (0.24-m-diam) coating furnace with a fritted-plate gas distributor,
- investigate the performance of particles with an outer layer of SiC , both as loose particles and as resin-bonded fuel rods, and
- evaluate high-density alumina as a potential high-temperature thermometry sheathing material.

The simulated fuel rods were recovered and transferred to the Physical Properties Group of the Metals and Ceramics Division; the thermal transport and expansion measurements have been reported separately.⁸ The results from the analysis of the kernel additive experiment showed that bare kernels from failed particles retained less than 10% of their ^{137}Cs inventory and that particles with cracked coatings retained between 60 and 80% of their ^{137}Cs inventory. The intact particles that contained reference ThO_2 and ThO_2 plus additives performed equally well in retaining the theoretical quantities of ^{137}Cs produced during irradiation. The LTI pyrocarbon stability and permeability study was not completed because of a material compatibility problem within the capsule.

Visual examination revealed that between 5 and 6% of the Bisso- and Triso-coated particles prepared in the 0.24-m-diam coater failed. The Bisso-coated particles experienced

pressure-vessel failures, whereas the outer LTI layers of the Triso-coated particles failed from fast-neutron damage, which led to failure of the exposed SiC layer through evaporation. Gamma analysis of the intact Triso-coated particles showed complete retention of their ^{137}Cs inventory for some, but 65% had lost significant amounts of ^{137}Cs . Gamma analysis of the intact Biso-coated particles showed a loss of about 70% of their ^{137}Cs inventory, with no particle retaining its entire ^{137}Cs .

This experiment showed that the presence of the outer LTI layer on Triso-coated particles was not required for producing warm-molded, resin-bonded fuel rods that retained their integrity in both the high- and low-temperature magazines of this capsule. Finally, high-purity alumina showed promise as a high-temperature sheathing material for HTGR conditions but requires additional investigation.

2.2.1.2 Capsule HT-34 — T. N. Tiegs

The fabrication and irradiation of capsule HT-34 has been described previously.^{9,10} Analysis on the Biso-coated ThO_2 particles in the capsule by the IMGA system was reported in the last annual report.¹¹ By the end of December 1979, analysis of the ORNL Triso-coated ThO_2 and weak-acid resin (WAR) driver particles containing 7% enriched uranium had been completed.

2.2.1.2.1 Triso-Coated ThO_2 — T. N. Tiegs. A summary of results on the $^{137}\text{Cs}/^{95}\text{Zr}$ ratio is given in Table 2.1. As shown, failures occurred in rods at positions 28, 34, and 37, which contained particles from batches A-834, A-837, and A-836, respectively. Particles from the same batches experienced no failures in other rods irradiated at lower temperatures and heavy-metal burnups. Particles from batch A-835 showed no failures in either the low- or high-temperature and burnup regions. The SiC deposition rate for batch A-835 was the highest (0.45 $\mu\text{m}/\text{min}$) of the four Triso-coated batches in the experiment.

We plan to use the PGA (Sect. 2.6.2) system with the intact Triso-coated ThO_2 particles to measure the release of the fission gases from the kernel during irradiation. Because of the Triso-coating, we can assume that no gas was lost from the particle during irradiation. The fractional gas release from the kernel is an important parameter in the fuel-performance models used in safety studies.

Table 2.1. Summary of Results of $^{137}\text{Cs}/^{95}\text{Zr}$ Ratios from Gamma Analyses of Triso-Coated ThO_2 Particles from HT-34

Position	$^{137}\text{Cs}/^{95}\text{Zr}$ ratios							Percentage failed ^a
	Particle batch	Minimum	Maximum	Mean	Standard deviation (%)	Theoretical ratio	Mean/theoretical ratios (%)	
28	A-834	2.782×10^{-3}	9.857×10^{-3}	7.985×10^{-3}	33.47	9.476×10^{-3}	84.3	35.5 (11/31)
31	A-835	9.548×10^{-3}	9.819×10^{-3}	9.682×10^{-3}	0.58	9.477×10^{-3}	102.2	0 (0/33)
34	A-837	2.775×10^{-3}	9.851×10^{-3}	9.228×10^{-3}	19.07	9.477×10^{-3}	97.4	8.6 (3/35)
37	A-836	2.695×10^{-3}	1.006×10^{-2}	9.586×10^{-3}	15.06	9.478×10^{-3}	101.1	4.0 (1/25)
41	A-834	9.427×10^{-3}	9.737×10^{-3}	9.579×10^{-3}	0.87	9.491×10^{-3}	100.9	0 (0/24)
44	A-835	9.633×10^{-3}	9.830×10^{-3}	9.709×10^{-3}	0.59	9.502×10^{-3}	102.2	0 (0/14)
47	A-837	9.539×10^{-3}	9.794×10^{-3}	9.622×10^{-3}	0.61	9.521×10^{-3}	101.1	0 (0/22)
50	A-836	9.723×10^{-3}	9.895×10^{-3}	9.809×10^{-3}	0.54	9.532×10^{-3}	102.9	0 (0/22)

^aNumbers in parentheses (failed particles/total particles examined) are based on $^{137}\text{Cs}/^{95}\text{Zr}$ ratios of individual particles. Failures are particles greater than three standard deviations away from the mean.

2.2.1.2.2 Low-Enriched Uranium Driver Particles — T. N. Tiegs. In addition to the Triso-coated ThO_2 particles irradiated in HT-34, Triso-coated WAR driver particles (batch OR-2576H) were included in the same positions. The driver particles are of interest because they contained 7% enriched uranium and represent the only low-enriched uranium (LEU) fissile particles available at ORNL for examination. A summary of the gamma analysis results on the $^{137}\text{Cs}/^{95}\text{Zr}$ and $^{110\text{m}}\text{Ag}/^{95}\text{Zr}$ ratios is shown in Table 2.2. Only one particle at the highest burnup and neutron-fluence position lost an appreciable amount of ^{137}Cs during irradiation. On the other hand, $^{110\text{m}}\text{Ag}$ was released to some extent from all particles. The results indicate that, although the SiC released $^{110\text{m}}\text{Ag}$, its ability to retain ^{137}Cs was not impaired. Metallography on these particles is planned, to examine the extent of the fission product palladium-SiC interaction.

Gamma analysis has not been used extensively in the past to determine the behavior of $^{110\text{m}}\text{Ag}$ in irradiated fuel because of interference between the main gamma peak of $^{110\text{m}}\text{Ag}$ at 657 keV and a ^{137}Cs gamma peak at 661 keV. In the current study, the gamma peak of $^{110\text{m}}\text{Ag}$ at 884 keV was used, and no interference problems were encountered. The branching ratios between the 657- and the 884-keV peaks are 0.94336 and 0.75098, respectively, so that there is only about a 20% difference in gamma activity between the two peaks. Because the yield of $^{110\text{m}}\text{Ag}$ is so low, long counting times were required. Typically, each particle was counted for 300 to 500 s, detector live time (500 to 850 s, actual time), which resulted in counting errors for the 884-keV peak in the range of 8 to 12%.

2.2.1.2.3 Bare ThO_2 Kernels. During irradiation some Biso-coated ThO_2 particles failed. The kernels were visually picked out and analyzed by the IMGA system. Although it is unknown when the coatings were lost, analysis of the kernels gives some insight into fission product releases into the buffer during irradiation.

Summaries of the $^{137}\text{Cs}/^{95}\text{Zr}$ ratio and $^{134}\text{Cs}/^{137}\text{Cs}$ ratio are given in Tables 2.3 and 2.4. As expected, ^{137}Cs release from the kernels was observed, which was very temperature and burnup dependent.

Table 2.2. Summary of Results from $^{137}\text{Cs}/^{95}\text{Zr}$ and $^{110m}\text{Ag}/^{95}\text{Zr}$ Ratios of Low-Enriched Uranium Driver Particles from HT-34

Capsule position	$^{137}\text{Cs}/^{95}\text{Zr}$ ratio			$^{110m}\text{Ag}/^{95}\text{Zr}$ ratio ^a			Failure fraction ^{b,c} (%)
	Mean	Standard deviation (%)	Mean fraction of theoretical ratio (%)	Mean	Standard deviation (%)	Mean fraction of theoretical ratio (%)	
28	1.631×10^{-2}	7.40	105.3	1.080×10^{-3}	29.85	32.0	0.6 (1/149)
31	1.610×10^{-2}	0.95	104.1	0.505×10^{-3}	56.76	15.4	0 (0/165)
34	1.628×10^{-2}	0.78	105.4	1.626×10^{-3}	21.53	51.2	0 (0/174)
37	1.712×10^{-2}	1.08	111.0	1.605×10^{-3}	16.94	52.7	0 (0/141)
41	1.590×10^{-2}	1.53	103.9	1.857×10^{-3}	14.23	72.6	0 (0/76)
44	1.590×10^{-2}	1.36	104.7	1.787×10^{-3}	15.76	81.9	0 (0/66)
47	1.580×10^{-2}	1.45	104.4	1.636×10^{-3}	13.81	80.6	0 (0/85)
50	1.649×10^{-2}	1.14	109.6	1.345×10^{-3}	13.12	76.6	0 (0/76)

^a ^{110m}Ag based on gamma peak at 884 keV.

^bDetermined by $^{137}\text{Cs}/^{95}\text{Zr}$ ratio.

^cNumbers in parentheses are (number of failed particles/total number of particles).

Table 2.3. Summary of $^{137}\text{Cs}/^{95}\text{Zr}$ Ratios for ThO_2 Kernels from Failed Particles in HT-34

Capsule position	Mean $^{137}\text{Cs}/^{95}\text{Zr}$ ratio	Standard deviation (%)	Fraction of ^{137}Cs inventory retained ^a (%)	Time-averaged particle temp. (%)	Burnup ^b (% FIMA)
27	9.035×10^{-4}	4.96	9.3	1345	12.2
39-1	2.828×10^{-3}	44.75	29.0	1288	10.3
42	8.033×10^{-3}	27.77	81.9	1161	8.6
46	8.641×10^{-3}	7.91	85.5	1146	7.3

^aObserved $^{137}\text{Cs}/^{95}\text{Zr}$ ratio for kernels divided by observed $^{137}\text{Cs}/^{95}\text{Zr}$ ratio for intact particles in similar positions.

^bFIMA = fissions per initial (heavy) metal atom.

Table 2.4. Summary of $^{134}\text{Cs}/^{137}\text{Cs}$ Ratios for ThO_2 Kernels from Failed Particles from HT-34

Capsule position	Mean $^{134}\text{Cs}/^{137}\text{Cs}$ ratio	Standard deviation (%)	Fraction ^{133}Xe retained ^a (%)
27	0.3958	30.67	22.6
39-1	0.7431	27.32	51.9
42	1.328	4.18	95.4
46	1.268	2.94	98.3

^aObserved $^{134}\text{Cs}/^{137}\text{Cs}$ ratio for kernels divided by observed $^{134}\text{Cs}/^{137}\text{Cs}$ ratio for intact particles in similar positions.

We have previously shown that the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio can be used to determine the behavior of ^{133}Xe , which is a precursor to ^{134}Cs (ref. 12). In examining the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio we must assume (1) ^{133}Cs , ^{134}Cs , and ^{137}Cs diffuse at the same rate so that the only difference from the theoretical ratio is due to ^{133}Xe loss, (2) if sufficient time were provided, there would be total release of the rare gases from the kernel, (3) the amount of ^{133}Xe released is the amount that has not decayed to ^{133}Cs , and (4) the effect of ^{133}Xe release by fission recoil is undeterminable. Therefore, the effective release time required for ^{133}Xe release from a ThO_2 kernel is given by:

$$D_t = -(\ln {}^{133}\text{Xe}_r)K^{-1}$$

where

D_t = effective release time (days),

$^{133}\text{Xe}_r$ = (100-fraction of ^{133}Xe retained)/100 = fraction of ^{133}Xe released,

K = decay constant for ^{133}Xe = 0.1315.

Fitting the release time to the following equation gave a good fit to the data:

$$D_t = a_0 + a_1 e^{[1/T(K)]} + a_2(F)$$

where

$$a_0 = 9.5747 \times 10^4,$$

$$a_1 = 9.5746 \times 10^4,$$

$$a_2 = -4.4633,$$

T = temperature (K),

F = burnup (% FIMA),

Accuracy of fit = 0.96 (1.0 is perfect).

The results are illustrated for a range of burnups in Fig. 2.1, which shows that, as temperature and burnup increase, the time it takes for ^{133}Xe to be released from the kernel becomes smaller. At high burnups and temperatures the ^{133}Xe is therefore released almost instantaneously.

2.2.1.3 Capsule HT-35 — E. L. Long, Jr.

A tentative experimental plan for the next in the series of HFIR target region capsules, HT-35, has been submitted for approval by DOE. This experiment will be a cooperative effort between ORNL and General Atomic Company (GA). Specimens prepared at ORNL will occupy about 75% of the available test space for fertile particles; the remainder of the test space for fertile particles will be occupied by GA specimens.

Fig. 2.1. Effective ^{133}Xe Release Times from ThO_2 Kernels as a Function of Temperature and Burnup.

The tentative experimental plan covers both ORNL and GA portions of the irradiation experiment. Brief objectives for HT-35 are to:

- provide an assessment of the irradiation performance of LTI pyrocarbon coatings derived by using standard coating gases diluted with CO_2 and prepared in a 0.13-m coating furnace;
- investigate the influence of fabrication process variables, coating properties, and postcoating annealing on the overall performance of Biso-coated ThO_2 particles;

- investigate ^{110m}Ag retention and solid fission product interaction with SiC and to compare results with deposition conditions;
- perform a detailed study of fast-neutron damage in SiC by both transmission electron microcopy (TEM) and small-angle x-ray scattering (SAXS) measurements;
- assess the irradiation performance of Triso-coated dense $\text{UC}_{x,y}\text{O}_2$ kernels, if available in time for this experiment, and to compare with results from Triso-coated WAR-derived kernels; and
- test the performance of candidate silicon Biso- and Triso-coated fertile (ThO_2) particles prepared at GA and to provide specimens for postirradiation heating tests at GA.

The HT-35 capsule will be irradiated for four cycles. The fertile particles will be irradiated loose in small graphite crucibles; the fissile particles (7% enriched) will be irradiated in small warm-molded rods.

Each of the above objectives are justified on the following bases:

Biso-coated particles prepared in a small coater by using CO_2 -diluted propylene and irradiated in HT-18, -19, and -34 were outstanding: no failures were observed even in the more severe test in which the HTGR design fluence was exceeded by 75% at a maximum particle-surface temperature of 1575°C . Consequently, additional coating development work has been conducted, using a larger (0.13-m-diam) coating furnace to produce several batches of reference design particles for irradiation testing. Capsule HT-35 will provide us with our first opportunity to irradiation test LTI pyrocarbon that is derived in a large coating furnace by using CO_2 as a diluent gas.

Differences in PyC microstructures resulting from variations in fabrication process variables strongly influence the coating's resistance to fast-neutron damage and its ability to retain fission gases. Previous experiments, HT-12 through -15, have shown that Biso-coatings produced at high coating rates (about $20\text{ }\mu\text{m}/\text{min}$) are resistant to fast-neutron damage but are susceptible to fission-gas premeability. Other experiments, HT-28, -29, and -30, have shown that coatings produced at low

to moderate coating rates (2 to 5 $\mu\text{m}/\text{min}$) and under various process variables are resistant to both neutron damage and fission-gas permeability. Particles were produced through a systematic variation of fabrication process variables for testing in HT-34 (as loose particles) and in HRB-14 (as fuel rods). Particles contained in HT-34 were irradiated in the as-coated condition, because coatings produced by using a frit gas distributor do not require the 1800°C annealing step from a coating-strength standpoint. Elimination of the annealing step apparently resulted in high failure fractions in HT-34, especially in a low-temperature magazine. Thus, we feel that it is appropriate to irradiate in HT-35 a select number of the particles from batches used in HT-34.

The interaction of SiC with metallic fission products has received much attention recently. Utilization of a medium-enriched uranium (MEU) fuel system will result in significant increases in the inventory of both silver and palladium in the fissile particles, as compared with the highly enriched uranium (HEU) fuel. Although there are indications that the retention properties of SiC to metallic fission products are affected by deposition conditions, additional investigations are required to establish a correlation.

Capsule HT-35 offers an opportunity to provide samples of SiC that have received fast-neutron damage for detailed evaluation by such techniques as TEM examinations and SAXS measurements. These studies will be performed on SiC layers that will be deposited on inert carbon kernels, which will avoid the complications that would be introduced by the presence of fission products. Also, these samples would be placed in the end plugs of the magazines at no expense to the experimental test space.

Based on favorable experience with WAR-derived fissile fuel, dense UC_xO_y , with a controlled O/U ratio, would appear to be an attractive alternative for the reference UC_2 fissile kernel for MEU application. Although High-Flux Isotope Reactor (HFIR) target capsules are not instrumented and do not have the flexibility of temperature regulation, they nevertheless provide a vehicle for performing screening tests on fissile fuel of low ($\sim 7\%$) enrichment. If the UC_xO_y kernels are available in

time, this experiment will provide the first opportunity to test reference-geometry MEU fissile fuel particles prepared at ORNL in a large (0.13-m) coating furnace. This experiment will permit a direct comparison of the performance of dense $UC_{x_y}O$ with WAR fuel.

To assess the fission product retention properties of coating designs, GA conducts heating tests on irradiated particles as a part of its HTGR fuel development program. Two of the more promising designs for reducing fission product losses from 600- and 500- μ m-diam fertile kernels are Triso- and silicon Biso-coatings, respectively. This capsule would provide GA with a supply of particles of the above designs with which to perform postirradiation heating (PIH) experiments.

A brief description of the experiment is as follows:

Target capsule HT-35 will consist of four graphite magazines, each containing 13 fuel positions and two end caps. The magazines adjacent to the reactor horizontal midplane will be designed to operate at a specimen surface temperature of 1250°C and in an estimated fast fluence range of from 8.5×10^{25} to 9.7×10^{25} neutrons/m² ($E > 29$ fJ). The upper and lower magazines will be designed to operate at a specimen surface temperature of 900°C and in an estimated fast fluence range of from 4.6×10^{25} to 7.8×10^{25} ($E > 29$ fJ). The proposed loading diagram for the HT-35 capsule is shown in Table 2.5. This table shows both the ORNL and GA portions of the capsule. Heavy-metal loadings for both the ORNL and GA portions of the capsule remain to be calculated, but, based on previous experience with HFIR target capsules, the total fuel loadings for HT-35 should be about 0.3 g of uranium (7% enriched) and about 1.1 g of thorium.

Five types of ORNL coated particles are planned for this irradiation experiment:

- Biso-coatings applied in a large coater, using CO₂ as the diluent gas,
- Biso-coatings prepared under a variety of coating conditions in a large coater to investigate fast-neutron induced permeability,
- Triso-coatings deposited with a large coater on dense $UC_{x_y}O$ kernels to compare performance to low-density, WAR-derived $UC_{x_y}O$,

Table 2.5. Proposed Location of Various Particle Types in HT-35

Capsule position	Particle source and type ^a	Kernel composition	Coating design	Estimated fast fluence 29 fJ (neutrons/m ²)	Surface design temp. (°C)
End plug	OR-I	C	Triso	4.6 × 10 ²⁵	900
1	OR-F	UCO	Triso	4.7	
2	GA-B	ThO ₂	Triso	4.9	
3	OR-F	UCO	Triso	5.3	
4	OR-E	ThO ₂	Biso	5.5	
5	OR-E	ThO ₂	Biso	5.8	
6	OR-F	UCO	Triso	6.1	
7	GA-A	ThO ₂	Triso	6.3	
8	GA-A	ThO ₂	Triso	6.6	
9	OR-F	UCO	Triso	6.8	
10	OR-E	ThO ₂	Biso	7.1	
11	OR-E	ThO ₂	Biso	7.3	
12	OR-F	UCO	Triso	7.6	
13	OR-D	ThO ₂	Biso	7.7	
End plug	OR-I	C	Triso	7.8	
End plug	OR-I	C	Triso	8.5	1250
14	OR-F	UCO	Triso	8.6	
15	GA-C	ThO ₂	SiBiso	8.8	
16	OR-F	UCO	Triso	9.0	
17	OR-D	ThO ₂	Biso	9.1	
18	OR-D	ThO ₂	Biso	9.2	
19	OR-F	UCO	Triso	9.3	
20	OR-D	ThO ₂	Biso	9.4	
21	OR-D	ThO ₂	Biso	9.5	
22	OR-F	UCO	Triso	9.6	
23	OR-D	ThO ₂	Biso	9.6	
24	OR-D	ThO ₂	Biso	9.6	
25	OR-F	UCO	Triso	9.7	
26	OR-D	ThO ₂	Biso	9.7	
End plug	OR-I	C	Triso	9.7	
End plug	OR-I	C	Triso	9.7	1250
27	OR-D	ThO ₂	Biso	9.7	
28	OR-F	UCO	Triso	9.7	
29	OR-E	ThO ₂	Biso	9.6	
30	OR-E	ThO ₂	Biso	9.6	
31	OR-F	UCO	Triso	9.6	
32	OR-E	ThO ₂	Biso	9.5	
33	OR-E	ThO ₂	Biso	9.4	
34	OR-F	UCO	Triso	9.3	
35	OR-Ref	ThO ₂	Biso	9.2	
36	GA-C	ThO ₂	SiBiso	9.1	
37	OR-F	UCO	Triso	9.0	
38	GA-C	ThO ₂	SiBiso	8.8	
39	OR-F	UCO	Triso	8.6	
End plug	OR-I	C	Triso	8.5	
End plug	OR-I	C	Triso	7.8	900
40	OR-D	ThO ₂	Biso	7.7	
41	OR-F	UCO	Triso	7.6	
42	OR-D	ThO ₂	Biso	7.3	
43	OR-D	ThO ₂	Biso	7.1	
44	OR-F	UCO	Triso	6.8	
45	OR-D	ThO ₂	Biso	6.6	
46	GA-A	ThO ₂	Triso	6.3	
47	OR-F	UCO	Triso	6.1	
48	OR-D	ThO ₂	Biso	5.8	
49	OR-D	ThO ₂	Biso	5.5	
50	OR-F	UCO	Triso	5.3	
51	GA-B	ThO ₂	Triso	4.9	
52	OR-F	UCO	Triso	4.7	
End plug	OR-I	C	Triso	4.6	

^aGA - General Atomic Company; OR - Oak Ridge National Laboratory; A - Triso-coated, 600-μm-diam ThO₂ kernels, low burnup; B - Triso-coated, 600-μm-diam ThO₂ kernels, high burnup; C - silicon Biso-coated, 500-μm-diam kernels, high burnup; D - Biso-coatings prepared by using CO₂ diluent; E - Biso-coatings for fast-neutron-induced permeability study (HT-34); F - Triso-coated WAR kernels; F' - Triso-coated dense UCO kernels; I - Triso-coated inert kernels; and Ref - reference Biso-coated ThO₂ kernels.

- Triso-coated WAR kernels prepared with a small coater to study ^{110m}Ag retention and fission product interaction with SiC, and
- Triso-coated inert kernels for fast-neutron-damage study on SiC.

General Atomic Company plans to include two types of coated fertile particles in this experiment: (1) Triso-coated, 600- μm -diam ThO_2 kernels for use in PIH experiments at GA and (2) silicon Biso-coated, 500- μm -diam ThO_2 kernels for use in PIH experiments at GA.

The tentative schedule calls for insertion of HT-35 in HFIR in June 1980 and for removal in September 1980, followed by PIEs, beginning in October 1980.

2.2.1.4 Capsules HT-31 and HT-33 — K. R. Thoms

When capsules HT-31 and HT-33 were removed from the reactor, the burnup and fluence data were reported, based on the best information available at that time. We have since been able to incorporate more up-to-date data and to use the CACA-2 computer program¹³ for target capsule analysis. Target capsule HT-34 was the first capsule to be designed and to have burnup data reported by using these techniques. Because target capsules HT-31 and HT-33 are part of the same series as is HT-34, the fluence and burnup data were recalculated so that all three capsules should now be comparable. The revised fast-fluence and burnup data for capsules HT-31 and HT-33 are presented in Tables 2.6 and 2.7, respectively.

The total and fast flux and the effective total cross sections used in the analysis of all three capsules have been reported.¹⁴

2.2.2 HRB Capsules — F. J. Homan

Routine postirradiation examination was completed during the last period¹⁵ for capsules HRB-11 through -13. Examination by IMGA of particles from these capsules was delayed until the current reporting period because of the large backlog of work for the IMGA system. The IMGA examinations for these capsules is now complete, and evaluation of the results is in progress. Routine PIE for HRB-14 and -15b was completed during this reporting period, with IMGA examination of fuels

Table 2.6. Revised Fast-Fluence and Burnup Data for Fuel Specimens in Capsule HT-31

Specimen number	Fast fluence (neutrons/cm ²) ($E > 29$ fJ) HTGR	Burnup (% FIMA) ^a		
		²³⁵ U	²³⁸ U	²³² Th
U-16	7.4×10^{21}	85.7	14.7	
U-17	7.5	85.7	14.9	
LASL-1	7.7	85.8	15.3	7.9
LASL-2	7.9	85.8	15.8	8.2
LASL-3	8.1	85.9	16.2	8.5
27	8.1			8.6
28	8.1	85.9	16.2	
29	8.1			8.4
30	8.0			8.3
31	7.9	85.8	15.8	
32	7.8			8.1
33	7.8			8.0
34	7.7	85.8	15.3	
35	7.6			7.7
36	7.5			7.6
37	7.4	85.7	14.7	
38	7.3			7.3
39	7.2	85.6	14.3	
40	6.5			6.2
41	6.3	85.3	12.3	
42	6.1			5.8
43	5.9			5.5
44	5.7	85.2	11.2	
45	5.4			5.1
46	5.2			4.9
47	5.0	84.9	10.1	
48	4.8			4.4
49	4.5			4.2
50	4.3	84.6	8.9	
51	4.0			3.8
52	3.8	84.3	8.1	

^aFissions per initial (heavy) metal atom.

Table 2.7. Revised Fast-Fluence and Burnup Data for Fuel Specimens in Capsule HT-33

Specimen number	Fast fluence (neutrons/cm ²) ($E > 29$ fJ) HTGR	Burnup (% FIMA) ^a		
		²³⁵ U	²³⁸ U	²³² Th
1, 52	5.37×10^{21}	85.1	10.8	
2, 51	5.74			5.9
3, 50	6.11	85.3	11.8	
4, 49	6.47			6.6
5, 48	6.82			6.9
6, 47	7.16	85.5	13.3	
7, 46	7.48			7.5
8, 45	7.78			7.8
9, 44	8.12	85.8	14.7	
10, 43	8.44			8.4
11, 42	8.74			8.7
12, 41	9.03	86.0	16.0	
13, 40	9.26			9.3
14, 39	10.3	86.3	18.4	
15, 38	10.5			10.9
16, 37	10.6	86.4	18.9	
17, 36	10.7			11.3
18, 35	10.9			11.4
19, 34	11.0	86.5	19.7	
20, 33	11.1			11.7
21, 32	11.2			11.9
22, 31	11.3	86.6	20.3	
23, 30	11.4			12.1
24, 29	11.5			12.3
25, 28	11.6	86.7	20.8	
26, 27	11.7			12.5

^aFissions per initial (heavy) metal atom.

from these capsules scheduled for mid-1980. The term "routine examination" applies to the procedures in use before the IMGA system was built. Included are visual examination, dimensional measurements, metallography, and microprobe examinations. Gamma scanning of the fuel rod and the empty graphite sleeve is also included. Because metallographic and microprobe examinations are destructive in nature, they are now done

after the particles have been through the IMGA system. The next HRB capsule (HRB-15a) is scheduled to begin irradiation in May 1980. This capsule will have no fuel from the ORNL program but will be operated as a service to GA. The test plan and the request for approval in principle have been prepared by GA.

2.2.2.1 Capsule HRB-13 — T. N. Tiegs

Descriptions of the capsule design, fabrication, operation, and initial PIE have been reported.¹⁶⁻¹⁸ Selected fuel rods were electrolytically deconsolidated and analyzed with the IMGA system.

A summary of results on the $^{137}\text{Cs}/^{95}\text{Zr}$ ratio of the fissile particles is given in Table 2.8. These particles were WAR derived from Amberlite and Duolite resins with varying degrees of sphericity. The purpose of the irradiation was to test the performance of misshapen particles. Reference spherical particles were also included as standards. As shown in Table 2.8, the particles generally showed high failure fractions. A plot of the failure fraction versus the degree of sphericity (Fig. 2.2) shows increasing failures as the particles deviate from spherical shape. The results also indicate that the Duolite resin appeared to perform better than did the Amberlite resin.

The particles classified as failed were submitted for metallographic examination. Those results showed numerous SiC coating failures for both particle batches at all shape levels. These failures were associated with corrosion of the SiC. The most severe corrosion was observed with the particle batch made with Amberlite resin (OR-2549H). Gamma analysis showed that these same particles also had the highest failure fractions and ^{137}Cs release.

Selected samples will be analyzed with the shielded electron microprobe to determine the cause of the SiC corrosion.

2.2.2.2 Capsule HRB-14 — T. N. Tiegs, E. L. Long, Jr., and K. R. Thoms

Descriptions of the fabrication and initial operation of capsule HRB-14 have been reported.^{19,20}

Table 2.8. Summary of Results on $^{137}\text{Cs}/^{95}\text{Zr}$ Ratios of Fissile Particles from HRB-13

Position in capsule	$^{137}\text{Cs}/^{95}\text{Zr}$ ratio					Mean percentage of theoretical	Percentage failed ^b	Deviation from sphericity ^c (%)
	Minimum	Maximum	Mean	Standard deviation (%)	Theoretical ^a			
2 ^d	7.907×10^{-4}	8.697×10^{-2}	7.102×10^{-2}	19.20	8.198×10^{-2}	77.9	9.2 (31/337)	13.3
10 ^d	2.665×10^{-4}	1.246×10^{-1}	5.174×10^{-2}	79.14	1.045×10^{-1}	49.5	30.8 (69/224)	14.6
15 ^d	7.162×10^{-4}	1.095×10^{-1}	5.425×10^{-2}	76.10	1.094×10^{-1}	49.6	31.2 (119/381)	9.6
17 ^d	1.743×10^{-4}	1.087×10^{-1}	4.545×10^{-2}	94.95	1.103×10^{-1}	41.2	50.4 (60/119)	29.7
18 ^d	6.130×10^{-5}	1.073×10^{-1}	3.981×10^{-2}	106.75	1.107×10^{-1}	37.2	57.3 (67/117)	29.7
19 ^d	4.107×10^{-4}	1.112×10^{-1}	5.387×10^{-2}	78.12	1.109×10^{-1}	48.6	30.9 (103/333)	13.3
20 ^d	7.060×10^{-4}	1.101×10^{-1}	5.522×10^{-2}	77.81	1.113×10^{-1}	49.6	32.2 (93/289)	13.3
21 ^e	6.809×10^{-5}	1.149×10^{-1}	8.607×10^{-2}	40.22	1.112×10^{-1}	77.4	14.2 (25/176)	12.1
22 ^e	3.046×10^{-5}	1.143×10^{-1}	8.166×10^{-2}	47.76	1.112×10^{-1}	73.4	19.6 (38/194)	12.1
23 ^e	2.147×10^{-4}	1.155×10^{-1}	7.577×10^{-2}	55.88	1.111×10^{-1}	68.2	23.3 (31/133)	15.8
24 ^e	5.107×10^{-5}	1.111×10^{-1}	7.169×10^{-2}	57.64	1.107×10^{-1}	64.8	25.7 (57/222)	15.8
25 ^e	1.017×10^{-3}	1.136×10^{-1}	9.302×10^{-2}	28.12	1.101×10^{-1}	84.5	9.4 (16/171)	9.6
26 ^e	4.648×10^{-3}	1.104×10^{-1}	9.497×10^{-2}	19.45	1.096×10^{-1}	86.7	4.7 (9/193)	9.6
31 ^e	4.072×10^{-4}	1.069×10^{-1}	9.195×10^{-2}	20.74	1.050×10^{-1}	87.6	10.6 (19/179)	13.3

^aTheoretical ratio calculated with computer code: E. J. Allen, CACA-2, ORNL/TM-5266 (February 1976).

^bNumbers in parentheses are: (number failed particles/total number particles); failed particles were those more than one standard deviation from the mean.

^cDeviation from sphericity (%) = $\{1/4\pi - [\text{cross-sectional area of kernel}/(\text{perimeter of kernel})^2]\} \cdot 4\pi(100)$.

^dMade from Amberlite resin.

^eMade from Duolite resin.

Fig. 2.2. Failure Fraction as a Function of Deviation from Spherical Shape for WAR-Derived Fissile Particles Irradiated in Capsule HRB-13.

Capsule HRB-14 completed its scheduled ten-cycle irradiation period on January 4, 1980, at 5:50 PM. The capsule was stored in the HFIR pool for a week to allow the short-lived fission products to decay to an acceptable level before transferring to the High-Radiation-Level Examination Laboratory (HRLEL) hot cells for PIE. The capsule was normal in appearance, with no unusual features.

The capsule operated for an equivalent 5124 h at 100-MW reactor power, resulting in a peak fast fluence of 8.3×10^{25} neutrons/m² HTGR ($E > 29$ fJ). The preliminary temperature histories of the specimens located at the same elevation as the thermocouples are shown in Figs. 2.3, 2.4, and 2.5. A summary of the fission-gas release/birth rate ratio (R/B) is presented in Table 2.9. The calculated burnup and damage fluence for each specimen is presented in Table 2.10. A summary of the reactor operating history for HRB-14 is shown in Table 2.11.

A GA representative was here to observe the removal of its fuel rods from the lower half of the HRB-14 magazine. All 20 of the rods

(a)

HRB-14, 2-14G

(b)

HRB-14, 6-14D

Fig. 2.3. Time Temperature Histories for ORNL Planchets: (a) 2-14G and (b) 6-14D in HRB-14. Top, middle, and bottom lines represent maximum fuel, minimum fuel, and midwall graphite sleeve temperatures, respectively.

Fig. 2.4. Time Temperature Histories for (a) ORNL Planchet 9-5D and (b) General Atomic Company Fuel Rod 2 in HRB-14. Top, middle, and bottom lines represent maximum fuel, minimum fuel, and midwall graphite sleeve temperatures, respectively.

(a)

HRB-14 ROD-10

(b)

HRB-14 ROD-17

Fig. 2.5. Time Temperature Histories for General Atomic Company Fuel Rods in HRB-14 for (a) Rod 10 and (b) Rod 17. Top, middle, and bottom lines represent maximum fuel, minimum fuel, and midwall graphite sleeve temperatures, respectively.

Table 2.9. Capsule HRB-14 R/B Data^a

Sample	Date	Accumulated irradiation time (h)	Fission gas R/B ($\times 10^{-5}$)				
			^{85m} Kr	⁸⁷ Kr	⁸⁸ Kr	¹³⁵ Xe	¹³³ Xe
1	5/26/78	154	0.22	0.13	0.13	0.05	0.18
2	6/02/78	320	0.35	0.19	0.16	0.09	0.43
3	6/08/78	467	0.50	0.27	0.28	0.13	0.68
4	6/15/78	598	0.69	0.39	0.38	0.25	0.89
5	6/23/78	785	1.04	0.49	0.56	0.37	1.41
6	6/30/78	955	1.25	0.76	0.84	0.38	1.54
7	7/13/78	1225	1.41	0.74	0.79	0.49	1.69
8	7/21/78	1416	2.49	1.14	1.38	0.91	3.29
9	8/04/78	1668	2.84	1.31	1.60	1.60	6.56
10	8/15/78	1932	2.48	1.13	1.69	1.24	5.94
11	9/12/78	2583	4.04	2.85	2.75	2.09	18.40
12	9/20/78	2715	58.10	72.10	63.00	0	261.00
13	9/26/78	2860	77.80	114.00	111.00	0.50	278.00
14	10/04/78	3048	90.40	172.00	133.00	59.80	335.00
15	10/11/78	3195	53.80	47.70	67.80	34.80	222.00
16	10/17/78	3337	57.70	47.00	65.60	36.20	209.00
17	10/26/78	3550	72.40	72.00	77.80	46.80	313.00
18	11/01/78	3676	70.20	64.10	66.10	40.40	257.00
19	11/07/78	3820	78.50	74.90	78.70	43.60	234.00
20	11/28/78	4100	149.00	131.00	135.00	96.30	614.00
21	12/04/78	4395	88.90	92.30	87.30	52.50	350.00
22	12/13/78	4612	128.00	115.00	101.00	76.60	582.00
23	12/21/78	4781	94.30	95.30	106.00	59.20	356.00
24	12/28/78	4950	114.00	110.00	100.00	66.70	532.00
25	1/04/79	5116	168.00	151.00	153.00	101.00	815.00

^aR/B = release/birth rate ratio.

were recovered, visually inspected, and shipped to GA for detailed postirradiation examination. All of the ORNL specimens have been successfully recovered from capsule HRB-14. All of the planchets were in excellent condition, except for five that were severely debonded. Stereoexamination of these five planchets revealed that the Biso-coated particles had failed as a result of fast-neutron damage; the failed particles came from a batch of particles (A-770) with a coating that had a

Table 2.10. Calculated Burnup and Damage Fluence for Specimens in Capsule HRB-14

	Calculated burnup (% FIMA) ^a			Fluence (neutrons/m ²) (E > 29 fJ) HTGR
	²³⁵ U	²³⁸ U	²³² Th	
ORNL loading zone number				
1	83.0	10.4	3.5	5.2 × 10 ²⁵
2	83.8	12.0	4.1	5.7
3	84.3	13.6	4.8	6.2
4	84.7	15.1	5.4	6.7
5	85.0	16.3	5.9	7.1
6	85.3	17.5	6.4	7.5
7	85.4	18.2	6.7	7.8
8	85.6	18.8	7.0	8.0
9	85.6	19.3	7.2	8.2
10	85.7	19.5	7.3	8.3
General Atomic Company fuel rod number				
1	0	0	7.4	8.3
2	85.7	19.5	7.3	8.2
3	0	0	7.2	8.1
4	85.6	19.2	7.2	8.1
5	0	0	7.1	8.0
6	85.5	18.8	7.0	7.8
7	0	0	6.8	7.6
8	85.4	18.1	6.7	7.5
9	0	0	6.6	7.3
10	85.2	17.4	6.4	7.1
11	0	0	6.1	6.9
12	85.0	16.1	5.8	6.6
13	0	0	5.6	6.4
14	84.7	15.0	5.3	6.2
15	0	0	5.0	5.9
16	84.3	13.5	4.7	5.6
17	0	0	4.3	5.3
18	83.7	11.7	4.0	5.0
19	0	0	3.6	4.8
20	82.8	10.0	3.3	4.5

^aFIMA = fissions per initial (heavy) metal atom.

Table 2.11. HRB-14 Reactor Operating History

HFIR cycle	Cycle schedule				Irradiation time ^a (h)	
	Begin		End		In cycle	Total accumulated
	Date	Time	Date	Time		
162	5/20/78	0240	6/11/78	0157	527	527
163	6/12/78	1533	7/04/78	2020	531	1058
164 ^b	7/06/78	1615	7/30/78	2350	515	1573
165	7/31/78	1752	8/22/78	0100	510	2083
166	8/22/78	1909	9/13/78	0400	513	2596
167	9/15/78	1824	10/06/78	2115	506	3102
168	10/07/78	1650	10/29/78	0000	511	3613
169 ^c	10/29/78	1735	11/22/78	0225	502	4115
170	11/22/78	2147	12/14/78	0140	507	4622
171	12/14/78	1945	1/04/79	1759	502	5124

^aIrradiation time is given in equivalent hours at 100 MW.

^bReactor was shut down from 7/24/78 2100 h to 7/27/78 1408 h. The reactor also operated at reduced power on numerous occasions during this cycle because of weather conditions.

^cReactor was shut down from 11/13/78 0815 h to 11/15/78 1830 h.

BAF_O of 1.107. These particles were part of a series to establish the upper limit for BAF_O and were expected to fail.

Metrology for all of the ORNL fuel specimens and graphite components has been completed. The raw data have been corrected and will provide input for the final thermal analysis.

Gamma scans were made of the HRB-14 capsule with all components in place and of the empty graphite sleeve after disassembly and are summarized in Figs. 2.6 and 2.7, respectively. As shown in Fig. 2.6, the ORNL fuel planchets (~2 mm thick) and the GA fuel rods are easily recognizable. The gamma scan of the empty sleeve, Fig. 2.7, showed high gamma activity in the region corresponding to the ORNL portion of the sleeve. Detailed spectral scans showed the majority of the activity in the ORNL portion was caused by ¹³⁷Cs, ¹³⁴Cs, ^{110m}Ag, ¹⁴⁰La, ¹⁴⁰Ba, and ¹⁵⁶Eu. Spectral scans of the GA portion of the empty sleeve showed a small amount of activity caused by ¹³⁷Cs and ¹³⁴Cs.

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Fig. 2.6. Gamma Scan of HRB-14 Capsule (0.55 to 0.75 MeV). ORNL plachets on left and General Atomic Company fuel rods on right.

Fig. 2.7. Gamma Scan of HRB-14 Empty Graphite Sleeve (0.55 to 0.75 MeV). High activity observed in ORNL half of sleeve.

Electrolytic deconsolidation of the planchets was begun in 1979 and is expected to be completed in early 1980. The IMGA examination of the fuel particles is scheduled to begin in March 1980.

2.2.2.3 Capsule HRB-15b — T. N. Tiegs, W. E. Thomas, and J. A. Conlin

The description and objectives of capsule HRB-15b have been reported.²¹ The capsule resided in HFIR facility RB-5 for eight cycles. First-cycle operation began July 6, 1978, at 4:15 PM (EDS), and the last cycle ended at 5:59 PM (EST), January 5, 1979. The experiment was in the reactor for a total of 182.11 calendar days. The number of cumulative full-power days of operation was 169.42, that is, the equivalent calendar days with HFIR operating at rated power [100 MW(t)]. See Tables 2.12 and 2.13 for operating history and R/B data, respectively.

Table 2.12. Reactor Operating History During Operation of HRB-14 and HRB-15b

HFIR cycle	Cycle schedule				Irradiation time ^a (h)		
	Begin		End		In cycle	Total accumulated	
	Date	Time	Date	Time		HRB-14	HRB-15b
162	5/20/78	0240	6/11/78	0157	527	527	
163	6/12/78	1533	7/04/78	2020	531	1058	
164 ^b	7/06/78	1615	7/30/78	2350	515	1573	515
165	7/31/78	1752	8/22/78	0100	510	2083	1025
166	8/22/78	1909	9/13/78	0400	513	2596	1538
167	9/15/78	1824	10/06/78	2115	506	3102	2044
168	10/07/78	1650	10/29/78	0000	511	3613	2555
169 ^c	10/29/78	1735	11/22/78	0225	502	4115	3057
170	11/22/78	2147	12/14/78	0140	507	4622	3564
171	12/14/78	1945	1/04/79	1759	502	5124	4066

^a Irradiation time is given in equivalent hours at 100 MW.

^b Reactor was shut down from 7/24/78 2100 h to 7/27/78 1408 h. The reactor also operated at reduced power on numerous occasions during this cycle because of weather conditions.

^c Reactor was shut down from 11/13/78 0815 h to 11/15/78 1830 h.

Table 2.13. Capsule HRB-15b R/B Data^a

Sample number	Date	Accumulated irradiation time (h)	Fission gas R/B ($\times 10^7$)				
			^{85m} Kr	⁸⁷ Kr	⁸⁸ Kr	¹³⁵ Xe	¹³³ Xe
1	7/13/78	167	0.29	0.21	0.17		
2	7/21/78	358	0.40	0.31	0.18	0.46	0.63
3	8/04/78	609	0.85	0.47	0.45	0.80	1.74
4	8/15/78	874	0.83	0.48	0.44	0.94	2.08
5	9/12/78	1521	0.72	0.57	0.40	0.97	2.16
6	9/20/78	1654	1.00	0.69	0.57		5.24
7	9/26/78	1800	0.80	0.56	0.45		2.12
8	10/04/78	1986	0.77	0.52	0.45	1.09	2.11
9	10/11/78	2135	0.92	0.60	0.57	1.06	2.38
10	10/17/78	2278	1.03	0.58	0.62	1.07	2.86
11	10/26/78	2491	1.16	0.53	0.63	1.02	5.08
12	11/01/78	2619	2.03	0.86	1.05		
13	11/07/78	2763	3.75	1.30	1.75	0.96	17.9
14	11/21/78	3040	6.08	1.90	2.94	1.06	23.3
15	12/04/78	3335	10.3	3.25	4.61	1.14	41.1
16	12/13/78	3553	12.0	3.55	5.29	1.22	64.0
17	12/21/78	3724	20.6	5.28	8.85	0.92	138.0
18	12/28/78	3892	36.4	8.46	17.6	1.43	176.0
19	1/04/79	4055	50.8	10.2	24.0	1.13	238.0

^aR/B = release/birth rate ratio.

A report was prepared on the thermal analysis and neutronics calculations associated with the capsule;²² an abstract of the report follows.

Candidate high-temperature gas-cooled reactor fissile and fertile particles were irradiated in the High Flux Isotope Reactor removable beryllium reflector facility (RB-5) for eight reactor cycles. The experiment contained 18 different "types" of fissile particles and five different types of fertile particles. All uranium was 20% enriched in ²³⁵U. The loose particles were loaded into "trays" that resemble flat graphite washers, each having 116 drilled holes in one face. One hundred eighty-four trays were stacked in columns in an alternate fertile particle-fissile particle sequence. The particles were irradiated for 169.4 full-power days. Peak

fuel burnup in percentage fissions per initial heavy-metal atom (FIMA) was as follows: ~20.5% for mixed-oxide fissile particles, ~33.5% for the other fissile particles, and ~7% for fertile particles. Maximum fast neutron fluence ($E > 183$ keV) at the end of the test was $\sim 7 \times 10^{25}$ neutrons/ m^2 . The report discusses methods used to specify particle loadings and contains thermal and neutronics results applicable to the irradiation test period.

The capsule was disassembled with a representative from GA present for the entire operation. Of more than 17,000 loose particles in the capsule, less than 1% were mixed with other batches. However, because the trays alternated between fissile and fertile particle types, even these particles can be retrieved by using the gamma-analysis system. The ORNL particles were visually examined, and all appeared to be in excellent condition. These particles are awaiting IMGA analysis, which is scheduled in the summer of 1980.

2.2.2.4 Capsule HRB-15a — E. L. Long, Jr.

Capsule HRB-15a, which will contain specimens supplied by GA, will be assembled and irradiated at ORNL. We performed the thermal analyses and supplied GA with the required heavy-metal loadings for fuel specimen preparation (see Sect. 2.4.1 of this report for details). This capsule, which is a part of the HTGR Generic Technology Program, will contain fuel rods, unbonded particles, and bonded wafers and is scheduled for insertion in HFIR in May 1980.

2.3 DEVELOPMENT WORK — F. J. Homan

The work described in this section is aimed at product improvement, preirradiation and postirradiation characterization of fuels, and techniques for performance assessment of particle coatings. Four areas are included:

- PyC permeability studies centering on both the techniques for measurement of permeability and the correlations of measured permeability with PyC properties,

- coating studies using CO₂ as a diluent gas in attempting to produce PyC coatings in the large coater having the same outstanding properties as batch OR 2013-T, which was produced in the laboratory-scale coater,
- sintering and conversion studies on dense UCO fuel for use with the MEU fuel cycle, and
- developing an improved SiC coating that can resist both attack by the fission product palladium and diffusion of the fission product silver.

2.3.1 Permeability Studies — D. P. Stinton

The results from several years of detailed study of the complex problem of PyC coating permeability have been summarized in a report.²³ The abstract from this report follows.

Fuel for High-Temperature Gas-Cooled Reactors (HTGRs) consists of dense uranium carbide and thoria microspheres coated with layers of PyC and SiC. The PyC coatings must be impermeable to fission gases to function properly during irradiation. Therefore, particles must be carefully characterized to determine as-produced permeability. The experiment described here compares several methods of permeability measurement and evaluates their value in predicting irradiation stability.

Sixty Bisco-coated particle batches with coatings deposited in either a 13- or 24-cm-diam coater were studied in this work. These batches were carefully characterized for permeability by Ne/He intrusion, long-term chlorination followed by radiography, and fission-gas release. These methods of permeability measurement were compared and correlated with deposition conditions and with PyC properties. The results from several irradiation tests were also used to evaluate the validity of the permeability measurements.

Very clear correlations were found to exist between the Ne/He and long-term chlorination techniques. Neon/helium values below 0.3 were found to be gastight by the chlorination procedure, and Ne/He values above 0.4 were found to be permeable by the chlorination technique. The fission gas release technique could not distinguish between slightly permeable coatings and gastight ones. Therefore, the Ne/He and long-term chlorination procedures are preferred over the fission-gas-release technique. Results from several irradiation tests verified that coatings with Ne/He values below 0.3 are gastight, and coatings with Ne/He values above about 0.4 were permeable.

2.3.2 Pyrocarbon Coating Using CO₂ Dilution — D. P. Stinton

During the past year we examined the effect of using CO₂ as carrier gas during PyC coating operations in the 0.13-m coater. This work was started because one batch (OR-2013-T) of particles prepared in a small coater produced good irradiation results in several different irradiation tests. Another potential advantage of CO₂ dilution is that CO₂ would oxidize fine soot produced during coating and thus might reduce the problems associated with off-gas cleanup.

The first CO₂-diluted coating (OR-2013-T) was deposited by using 33% propylene and 67% CO₂. Studies performed in the large coater examined several different ratios of coating gas to CO₂ (Table 2.14). The initial runs were made from 50% propylene and 50% CO₂. Severe oxidation of graphite components within the furnace occurred after only a couple of runs. Deposition of PyC coatings from 50% CO₂ and 50% mixed gas (54% acetylene and 46% propylene) also attacked the graphite components by oxidation. Several coatings were deposited after reducing the CO₂ to 33%. This ratio of CO₂ to coating gas did not attack the graphite parts within the furnace; however, other problems occurred. At the desired coating rates of 3 to 4 $\mu\text{m}/\text{min}$, the total gas flow was insufficient to produce adequate fluidization, which resulted in particles sticking to the crucible. Finally coatings were deposited by using 50% coating gas, 25% CO₂, and 25% argon. This mixture produced adequate

Table 2.14. Coating Gas Compositions Used in CO₂ Dilution Studies

33% Propylene, 67% CO ₂ — OR-2013-T		
50% Propylene, 50% CO ₂	{	Caused oxidation of graphite components
50% Mixed gas, 50% CO ₂		
67% Propylene, 33% CO ₂	{	Produced inadequate fluidization and sticking
67% Mixed gas, 33% CO ₂		
50% Propylene, 25% CO ₂ , 25% Ar	{	Produced adequate fluidization with no evidence of oxidation
50% Mixed gas, 25% CO ₂ , 25% Ar		

fluidization to prevent particle sticking and did not oxidize the graphite furnace components.

All batches produced during this study were carefully characterized in terms of density, permeability, and anisotropy. Batches were characterized for density by measuring a liquid gradient density of coating fragments as well as mercury porosimetry of whole particles to determine the volume percent open porosity. A bulk density of the coating referred to as the corrected gradient density²⁴ was then calculated by using the gradient density value and the volume percent open porosity. The fraction of defective particles (either cracked coatings or bare kernels) was measured by using a 2-h chlorination.²⁵ Coating permeability was measured by two techniques: one was the Ne/He measurement,²⁶ and the second was an 18-h chlorination, followed by an examination by radiography.²⁷ All coatings in this study were produced at coating rates that normally yielded nearly isotropic coatings. Bacon anisotropy factors (BAFs) of the coatings are currently being measured but are expected to be in an acceptable range (1.03 to 1.04 BAF₀ units).

After several coating runs, we observed that CO₂-diluted coatings had unusually large amounts of open porosity. Previous work with conventional diluents showed that high-quality coatings had gradient densities of about 1.94 g/cm³ and 3% open porosity (Table 2.15). This work produced a coating with a corrected gradient density of 1.88 g/cm³

Table 2.15. Characterization of CO₂-Diluted Coatings Showing Increased Amounts of Open Porosity

	Gradient density (g/cm ³)	Open porosity (%)	Corrected gradient density (g/cm ³)	Ne/He measurement	Chlorination (18 h)
Optimum coatings (conventional diluents)	1.94	3.0	1.88	0.30	Gastight
OR-2013-T	2.01	3.7	1.94	0.14	Gastight
50% Propylene- 25% CO ₂ -25% Ar	1.92	4.2	1.84	0.25	Gastight
50% Mixed gas- 25% CO ₂ -25% Ar	1.92	7.0	1.79	0.35	Permeable

and resulted in a gastight coating (Ne/He values below 0.30). Coatings produced with CO₂ dilution had open porosities greater than 3%. Batch OR-2013-T, which has proved several times to be gastight, had an open porosity of 3.7%. Coatings produced with 50% propylene, 25% CO₂, and 25% argon had open porosities of about 4.2%, which resulted in a lower corrected gradient density of 1.84 g/cm³. However, even with a lower corrected gradient density, the Ne/He measurement and the 18-h chlorination showed the coating to be gastight. Coatings produced with 50% mixed gas, 25% CO₂, and 25% argon had open porosities of about 7%, resulting in a very low corrected gradient density of 1.75 g/cm³. However, these coatings were frequently determined to be permeable both by the Ne/He technique and by the 18-h chlorination. Because mixed gas frequently produced permeable coatings, the majority of this study dealt with coatings produced by using 50% propylene, 25% CO₂, and 25% argon.

A careful study was performed, using 50% propylene, 25% CO₂, and 25% argon, to examine the effect of deposition temperature and coating rate on various properties of the LTI layer (Table 2.16). After examining Table 2.16, it is clear that the deposition temperature controls both the gradient density and the percent open porosity. Higher deposition temperatures produced lower gradient densities and higher percent porosities. Because the gradient density and porosity were controlled by the deposition temperature, the corrected gradient density was also controlled by the deposition temperature. As shown in Fig. 2.8, low deposition temperatures produced high corrected gradient densities. From previous experience, we expected the corrected gradient density to control the Ne/He ratio. This was again true for CO₂-diluted coatings (Table 2.16). Deposition temperatures of 1300°C or below produced Ne/He ratios of 0.25 or below. These values are all extremely low and, in fact, lower than most Ne/He ratios produced by conventional coating processes. Figure 2.9 shows a plot of Ne/He versus corrected gradient density for CO₂-diluted coatings and coatings being irradiation tested in HRB-14. It is clear that CO₂ dilution produced more gastight coatings than did dilution with helium, argon, or hydrogen.

Table 2.16. Complete Characterization of CO₂-Diluted Coatings^a

Run number	Deposition temp. (°C)	Low-temp. isotropic coating rate (μm/min)	Gradient density (Mg/m ³)	Open porosity (%)	Corrected gradient density (Mg/m ³)	Ne/He ratio
A-898	1275	3.2	1.954	4.1	1.874	0.21
A-892	1275	3.5	1.981	3.6	1.910	0.16
A-899	1300	3.0	1.953	4.1	1.874	0.21
A-914	1300	3.6	1.889	4.5	1.804	0.26
A-915	1300	4.1	1.928	4.4	1.844	0.23
A-920	1325	3.3	1.864	5.5	1.761	0.34
A-919	1325	4.0	1.891	4.9	1.798	0.31
A-908	1350	4.5	1.833	6.7	1.710	0.40

^aAll coatings deposited by using 50% propylene, 25% CO₂, and 25% argon. Fraction defective (Cl₂ leach) of all coatings $\sim 1 \times 10^{-5}$.

Fig. 2.8. Low Deposition Temperatures Produce Coatings with High Corrected Gradient Densities.

Fig. 2.9. The As-Produced Permeability of CO_2 -Diluted Coatings Is Generally Lower than Coatings Produced by Conventional Diluents.

The deposition of coatings by CO_2 dilution is slightly more difficult than by conventional diluents because multiple diluents (argon and CO_2) are required to produce good coatings. The CO_2 is also a more hazardous gas because it could potentially destroy the furnace by oxidation if incorrect flows were used. From our brief study, the use of CO_2 did not appear to reduce the volume of soot produced. In fact, a problem occurred during this work that may be attributable to the use of CO_2 . Very fine soot was carried beyond the scrubber system and deposited in the off-gas lines. The continual buildup of soot eventually caused blockage of the lines. It is not clear at this time if the soot carryover was caused entirely by the use of CO_2 .

The conclusions drawn from this work are that CO_2 seems to yield more gastight coatings (as-produced) than do conventional diluents. Much work remains to be done to determine if the irradiation behavior of CO_2 -diluted coatings is superior to the behavior of conventional coatings. Work of this nature has already begun. Nine CO_2 -diluted coatings will be irradiation tested in HT-35. The results from this test will not be known for many months. Until the results from this

test verify the superiority of CO_2 -diluted coatings, routine coatings will be produced by using conventional diluents. Currently, no additional development of CO_2 -diluted coatings is planned.

2.3.3 Dense UCO Fuel Kernel Development — D. P. Stinton

A potential fissile particle for the HTGR is a highly dense microsphere with a composition of UO_2 plus UC_2 . Microspheres with a composition of UO_3 plus carbon are reacted and sintered to produce these highly dense UO_2 plus UC_2 kernels. Carbon is mixed into the solution of UO_3 by using ultrasonic dispersion and a dispersing agent. This procedure distributes the carbon in a uniform fashion in the as-dried microspheres.

During calcination (heating to $\sim 450^\circ\text{C}$ to remove volatiles), a considerable amount of carbon is lost from the microspheres. We determined that carbon from near the outer surface of the microsphere reacts with oxygen liberated from the UO_3 during reduction. A microsphere exhibiting this behavior is shown in Fig. 2.10. This sintered

Fig. 2.10. A Microsphere Showing Loss of Carbon from the Outer Surface. The white phase is UC_2 , and the gray phase is UO_2 .

microsphere consists of UO_2 (gray phase) distributed evenly throughout the microsphere and UC_2 (white phase), which is present in the interior of the particle but absent from the outer surface. Careful examination of the calcining procedure showed that from 15 to 37% of the carbon was lost during calcination, depending on the atmosphere used (Table 2.17).

After several sintering runs were made, we determined that additional carbon was being lost during the sintering operation. Partial pressures of carbon monoxide were varied within the furnace to control the amount of carbon lost. The atmospheres used were Ar-3% CO, Ar-2% CO, Ar-1.5% CO, Ar-1% CO, and Ar-4% H_2 . The final percent carbon in the desired 15 to 30% converted microsphere is from 1.3 to 2.7 wt %. As shown in Table 2.18, this level was obtained when using Ar-1% CO or

Table 2.17. Loss of Carbon During Calcination,
Using Bottled Gases

Calcining atmosphere	C/U ratio after sintering	Carbon retained (%)
Ar-0.5% CO	0.0335	62.9
Ar-1.0% CO	0.0414	77.7
Ar-2.0% CO	0.0451	84.6
Ar-4% H_2	0.0432	81.1

Table 2.18. Characterization of Sintered UO_2
Plus Carbon Microspheres^a

Atmosphere	Mercury density (Mg/m^3)	Carbon (wt %)
Ar-3% CO	7.5	2.69
Ar-2% CO	8.0	2.97
Ar-1.5% CO	10.3	1.93
Ar-1% CO	10.9	1.47
Ar-4% H_2	11.0	1.13

^aAll runs were sintered at 1550°C for 4 h and originated from one batch of material that contained 5.17% C.

Ar-1.5% CO. The table also shows that high densities were obtained only by using less than 1% CO.

The desired composition for potential HTGR applications is from 15 to 30% UC_2 , with the remainder UO_2 as shown in the phase diagram of the UCO system (Fig. 2.11). Uranium oxycarbide (UC_xO_y) is undesirable because it will react with the carbon of the buffer layer and alter the kernel's composition. Therefore, the composition must be precisely on the line presented in Fig. 2.11 and not off the line in any direction. The batches shown in Table 2.18 were examined to determine the phases present after sintering. We learned that the thermodynamic equilibrium

ORNL-DWG 79-12542R

Fig. 2.11. Sintering at 1550°C in Ar-1% CO and Then Ar-3% CO Produces $UO_2 + UC_2$ with Densities of About 97%. The results when sintering in Ar-1% CO, Ar-4% H_2 , and Ar-3% CO are also presented.

shifts in this region of CO partial pressures. At 1550°C, UO_2 plus UC_2 are stable only when the sintering atmosphere contains from 1.3 to 2.0% CO. If the atmosphere contained less than 1.3% CO, the oxycarbide phase and UO_2 were the equilibrium phases. If the atmosphere contained more than about 2% CO, only the oxide and carbon were stable. From this we concluded that high densities were obtained when Ar-1% CO or Ar-4% H_2 was used but that the oxycarbide phase was produced (see Fig. 2.11). If Ar-1.5 or 2.0% CO were used, the proper phases of UO_2 and UC_2 were produced, but low densities resulted. If greater than 2.0% CO were used, the reaction did not occur and very low density UO_2 and carbon were the equilibrium phases (see Fig. 2.11 when using Ar-3% CO).

A procedure was developed to sinter UO_3 plus carbon microspheres, produced by internal gelation to the desired composition at temperatures as low as 1550°C. After calcination (as described earlier) kernels are sintered at 1550°C in Ar-1% CO for 4 h to yield a high-density microsphere with a composition of UO_2 plus UC_xO_y . These kernels are then sintered at 1550°C for an additional 4 h in Ar-3% CO to shift the thermodynamic equilibrium from $\text{UO}_2 + \text{UC}_x\text{O}_y$ to $\text{UO}_2 + \text{UC}_2$. The progression of the sintering process may be followed in Fig. 2.11. Material is first sintered in Ar-1% CO because the sintered densities decrease rapidly as the percentage of CO increases. Ar-1% CO is preferred over Ar-4% H_2 because more carbon is retained in the microspheres. Four batches with varying amounts of carbon have been successfully sintered to produce highly dense UO_2 plus UC_2 (see Table 2.19). Work has begun to shorten

Table 2.19. Characterization of Sintered $\text{UO}_3 + \text{C}$

Run	Initial carbon (%)	Mercury density (Mg/m^3)	Theoretical density (%)	Final carbon (%)	Final composition	
					% UO_2	% UC_2
S-270	2.77	10.29	93.6	1.01	88.9	11.1
S-268	3.71	10.22	93.0	1.48	83.5	16.5
S-269	4.46	10.72	97.2	1.79	79.7	20.3
S-262	5.17	10.64	96.5	2.03	77.5	22.5

the sintering schedule but to retain the high density and proper composition and to scale up the process from 10-g quantities to 100-g quantities.

2.3.4 Silicon Carbide Characterization and Testing — R. J. Lauf and D. N. Braski

The current experimental plan involves 12 SiC coating types, which were deposited under systematically varied conditions to yield a range of microstructures. Each coating type was deposited on three types of Biso-coated kernels: UO_2 , $\text{UO}_2 + \text{Ag}$, and $\text{UO}_2 + \text{Pd}$. The coatings are being characterized by several techniques, including TEM. At the same time, coated particles containing the simulated fission products silver and palladium are being annealed in a thermal gradient of $27.5^\circ\text{C}/\text{mm}$ at typical HTGR operating temperatures. Examination during and after annealing will allow the different SiC coatings to be evaluated with respect to fission product transport or retention. By combining these results with those of characterization efforts and comparisons of microstructural differences, we will gain a better understanding of the modes of interaction between SiC and noble metal fission products.

2.3.4.1 Silicon Carbide Microstructure Characterization

A topical report covering the characterization of SiC coatings²⁸ was issued in August 1979. The abstract of this report follows.

Fuel particles for the HTGR contain a layer of pyrolytic silicon carbide to act as a pressure vessel and fission product barrier. The SiC is deposited by the thermal decomposition of methyltrichlorosilane (CH_3SiCl_3 or MTS) in an excess of hydrogen. Coatings deposited at temperatures from 1500 to 1700°C and coating rates from 0.4 to 1.2 $\mu\text{m}/\text{min}$ have been studied by optical microscopy, x-ray diffraction, electron microscopy, and density measurements. Transmission electron microscopy (TEM) has shown the microstructural features to be extremely complex and much finer than the ability of optical microscopy to resolve. X-ray diffraction has detected traces of α -SiC in some coatings, and those conditions of deposition temperature and coating rate that give rise to this phase have been determined. Lattice fringe images of 1.5-nm spacing indicate that the α -SiC observed in TEM is of the 6H polytype. Previously, the coatings were assumed to be all cubic β -SiC. Optical microscopy has indicated

that the SiC coatings are optically anisotropic. This result is unexpected in a cubic material and is not completely understood at this time. Coating density varied with deposition temperature and coating rate, in agreement with earlier work.

2.3.4.2 Correlation of Silicon Carbide Microstructures with Deposition Parameters

A report on the relationship between SiC coating properties and the corresponding deposition conditions²⁹ has been written. The abstract of this report follows.

Twelve silicon carbide coating batches have been deposited on Biso-coated UO_2 microspheres under systematically varied conditions to yield a variety of microstructures. These coatings have been characterized by several techniques to determine their properties and to study the effect of deposition parameters on coating characteristics. The grain size and shape depend on coating rate and deposition temperature. The fine structure of the coating, observed at the submicron level by TEM, has been found to vary in the same way as does the general microstructure observed optically. X-ray diffraction has shown the presence of one or more polytypes of α -SiC in coatings deposited at relatively low temperatures and/or relatively high coating rates. The coating density and surface morphology are strongly dependent on deposition temperature and coating rate. The best surface appearance is observed in those coatings that have the highest density.

2.3.4.3 Interaction with Simulated Fission Products

Microspheres of UO_2 containing the simulated fission products silver and palladium were fabricated by the following method: Sol-gel microspheres were sintered to approximately 60% theoretical density (T.D.). The spheres were then immersed in an aqueous solution of AgNO_3 or PdCl_2 . After drying at about 90°C in air, the spheres were heated to 600°C in flowing hydrogen to reduce the silver or palladium to the metallic state. As Fig. 2.12 shows, the result was a uniform dispersion of small noble metal particles.

Each of the 12 SiC batch types was deposited on doped spheres and on pure UO_2 spheres. Samples of each have been annealed at temperatures of 1300 to 1500°C in the thermal-gradient furnace. The particles were radiographed after 400 and 1024 h of annealing. These radiographs are

PHOTO Y-169718

Fig. 2.12. Biso-Coated UO_2 Particle Containing a Fine Dispersion of Silver as a Simulated Fission Product.

being studied to determine any evidence of attack by the silver or palladium. Selected specimens will be examined metallographically and by TEM to attempt to identify possible reaction products and diffusion paths. Studying the microstructures of coatings deposited on undoped UO_2 particles that were also annealed will enable us to observe any changes caused by annealing alone.

2.3.4.4 Irradiation Performance

Irradiated coatings from fuel rod HRB-13-9 were examined by TEM. These coatings received a fast fluence of about 7×10^{25} neutrons/ m^2 at an average temperature of 1050°C . Because the coatings were deposited on inert (carbon) microspheres, the complicating effects of fission product attack are not present.

Figure 2.13 shows a representative micrograph of an irradiated coating at about 140,000 $\times$. From TEM studies, several observations can be made:

- The stacking fault density has decreased remarkably, and those faults that remain are not as sharply defined as are those observed in unirradiated samples. This result is probably caused by the diffusion of point defects to the faults, which act as a sink for vacancies and/or interstitials and are partly consumed by them.

PHOTO E-21810

Fig. 2.13. Electron micrograph of an Irradiated SiC Coating, Showing the Small Voids and the Nearly Eliminated Stacking Faults; 140,000 $\times$; 33% Reduction.

- Numerous small voids (20 to 50 Å) are present. Stereopair electron micrographs are being analyzed to obtain a value for the void density (voids per cubic angstrom), from which the approximate swelling can be calculated.
- The voids appear to lie in planes parallel to the remaining stacking faults in some cases. Most grain-boundary areas appear to have fewer voids per unit volume than are found in the interiors of the grains.
- Electron diffraction patterns have indicated the possible presence of one or more additional phases in some grains. At least one pattern can be indexed as a polytype of alpha-SiC, but another set of diffraction spots cannot. This result could indicate another polytype of SiC or could perhaps indicate that free silicon is present. The second phase particles that give rise to these extra diffraction spots have been photographed and are found to be approximately 50 to 300 Å in diameter.

Selected SiC coating samples from the current experimental plan are tentatively scheduled to be included in an irradiation capsule (HT-35) for insertion in HFIR in June 1980.

2.4 IRRADIATION SUPPORT ACTIVITIES — F. J. Homan

This work is associated with both supplementing and improving the quality of information developed through irradiation testing. In the past, work associated with characterizing the irradiation facilities (temperatures, flux distributions, gamma heating measurements, etc.) was included. No work has been done during the current reporting period on capsule thermometry, flux mapping, or gamma-heating characterization. The loading calculations for capsule HRB-15a are described. This capsule is scheduled to begin irradiation in May 1980. The thermal analysis of specimens irradiated in capsule HRB-15b is presented. This capsule completed irradiation in January 1979. The out-of-reactor, thermal-gradient heating experiments to measure fission product-SiC coating interaction rates are also reported.

2.4.1 Nuclear Evaluation and Thermal Analysis

The work during this reporting period is associated with two in-reactor experiments, HRB-15a and HRB-15b. Fuel loadings have been calculated for HRB-15a, and a report was issued on HRB-15b. Work related to each experiment is discussed separately.

2.4.1.1 Experiment HRB-15a

The problem here is to specify fuel loadings for experiment HRB-15a, which is scheduled to begin irradiation in May 1980. There are 18 bonded-bed fuel specimens in the experiment, each containing both fissile particles and fertile particles. The experiment is to be loaded such that the maximum fuel centerline temperature will not exceed 1350°C. Fuel temperature is controlled by varying the composition of the sweep gas, which is present in a gap (or annulus) between the graphite sleeve and the primary containment vessel and is also in the clearance between the fuel rods and the inside radius of the graphite sleeve. To allow some latitude in temperature control, it is desirable that the experiment be loaded such that the maximum fuel temperature is obtained with a helium-neon gas mixture containing not less than approximately 20% neon.

The experiment will reside in the removable beryllium reflector of the HFIR. The HFIR control rods (which are physically two concentric cylinders) are positioned between the experiment and the reactor core. The movement, or management, of the control rods during a cycle will introduce considerable variation in the neutron flux in the experiment. Experimentally determined axial position-dependent, time-dependent neutron fluxes were used in the calculations.

The procedure that we used to specify the loadings is straightforward. The first step was to compute specific powers for both fissile material (^{235}U) and fertile material (^{232}Th) as a function of axial position and fuel residence time. Specific power is defined as the power (watts) at a particular time in the history of the experiment per initial amount (grams) of material. The fuel depletion code was used to obtain these data.

The contribution to the volumetric heat sources from fission is maximum near the reactor horizontal midplane (HMP) early in the first cycle of operation. At that time almost all of the heat source from fission occurs in the fissile particles. Knowing the specific power, the maximum loading (fissile particles) for specimen 10 was determined. The thermal analysis code was used to determine the loading (i.e., to determine the amount of fissile material required for specimen 10 to produce a fuel centerline temperature of 1350°C when using a sweep gas composition of 80% He-20% Ne).

The next step was to determine the relative fissile particle loadings for the remaining 17 fuel rod specimens in the experiment. Throughout first-cycle operation, most of the volumetric heat source from fission is contributed by the fissile particles. At a particular time, the relative specific power in fissile particles of each specimen is obtained by dividing by the specific power for specimen 10. The inverse of the relative specific powers gives the relative fissile particle loadings at a particular time during the cycle. Arrays of relative fissile-particle loadings were computed at increments of one full-power day during the 21 reactor operating days of cycle one. HFIR control rod management will cause considerable temperature variation in the fuel specimens during the cycle. The relative fissile particle loadings were time-averaged to minimize the temperature variation within each of the 18 fuel rod specimens. Multiplying the time-averaged relative fissile-particle loadings by the previously calculated loading for specimen 10 gave the loadings for fissile particles in each specimen of the experiment.

During first-cycle operation, the contribution to the volumetric heat source from fission in bred uranium within fertile particles is very small. However, during the history of the experiment, the amount of bred uranium increases, and, during the last few cycles, the amount of bred uranium in the experiment is significant. Conversion ratios for fuel specimens in the experiment are less than unity. If no fertile material were present in the fuel specimen, the power produced from fission would become progressively lower at corresponding times in each subsequent cycle. The inclusion of fertile material tends to flatten

the power curve. The fertile/fissile ratio can be computed by considering the power produced in each specimen at times early and late in the history of the experiment. The procedure is as follows: Let P_{11} be the specific power in fissile particles early in the history of the experiment and P_{12} be the specific power in fissile particles late in the history of the experiment; P_{21} and P_{22} are, respectively, specific power from bred uranium in fertile particles at the same times. Let c be the ratio of total power produced in the specimen near the end of the irradiation period to the total power produced in the specimen early in the first cycle of operation. The fertile/fissile ratio R can then be computed from the equation:

$$R = (P_{12} - cP_{11}) / (cP_{21} - P_{22}) .$$

The fertile loadings are obtained by multiplying the previously determined fissile loadings by the fertile/fissile ratios. The fertile/fissile ratios increase with increasing values for c ; that is, the more fertile material that can be loaded in a specimen the higher the total specimen power will be. The maximum fertile loadings are determined by the particle packing fraction. Fuel loadings shown in Table 2.20 are based on the assumption that the particles cannot occupy more than 35% of the fuel specimen volume. The particle packing fractions were based on the nominal particle dimensions shown in the table.

The final step in the analysis was to test the proposed fuel loadings with the thermal analysis code. These calculations include all contributions to the regional volumetric heat source (i.e., from fission in the particles plus gamma-ray heating in the graphite and the stainless steel vessels). Thermal expansion; dimensional stability; temperature-dependent and, if applicable, fluence-dependent thermal properties for the fuel specimen; the graphite sleeve; and the stainless steel vessels are considered in these calculations.

Calculated fuel centerline temperatures during first-cycle operation for the loadings in Table 2.20 are presented in Table 2.21.

The temperature variation with time is a characteristic of the HFIR reflector facility and is caused by control rod management. Note

Table 2.20. Fissile and Fertile Loadings for Experiment HRB-15a

Specimen	Fissile particles				Fertile particles			
	Material ^a	Kernel ^b	Particle ^b	Loading ²³⁵ U (g/m)	Material ^a	Kernel ^b	Particle ^b	Loading ²³² Th (g/m)
1	1	350	870	1.925	4	450	800	37.257
2	1		800	1.581		450	800	30.455
3	1		800	1.358		600	1038	26.838
4	2		800	1.205		600	910	24.486
5	2		800	1.097		600	940	22.794
6	1		880	1.021		500	704	21.523
7	3		810	0.967		500	704	20.567
8	1		800	0.932		600	1038	19.880
9	1		800	0.912		600	1038	19.450
10	3		800	0.909		450	800	19.364
11	3		800	0.908		450	800	19.287
12	1		800	0.920		450	800	19.460
13	1		870	0.950		450	800	19.987
14	3		800	1.001		500	704	20.941
15	3		800	1.078		500	704	22.422
16	2		800	1.189		450	800	24.541
17	2		800	1.529		450	800	30.198
18	2		800	1.805		450	800	32.840

^a1 = UO₂, 2 = UC_{0.3}O_{1.6}, 3 = UC₂, and 4 = ThO₂.

^bNominal diameter, μm.

Table 2.21. Estimated Specimen Operating Temperatures and Sweep Gas Compositions for Capsule HRB-15a

Specimen	Specimen temperature (°C)			Sweep gas composition
	1	10	18	
AT 1 FPD ^a	1200	1350	1170	80 He-20 Ne
AT 14 FPD	1270	1330	1290	60 He-40 Ne
AT 21 FPD	1360	1210	1370	80 He-20 Ne

^aFull-power days of reactor operation.

that there is an approximately 200°C variation in fuel centerline temperatures during the cycle, even with optimum fuel loadings.

2.4.1.2 Experiment HRB-15b

The irradiation period for this experiment ended January 4, 1979, at 1750 h. The procedure used to specify fuel loading for this experiment, together with data for temperatures, fuel burnups, and fast neutron fluence, is described in a published report.³⁰ The abstract for that report is as follows:

Candidate high-temperature gas-cooled reactor fissile and fertile particles were irradiated in the High-Flux Isotope Reactor removable beryllium reflector facility (RB-5) for eight reactor cycles. The experiment contained 18 different "types" of fissile particles and five different types of fertile particles. All uranium was 20% enriched in ²³⁵U. The loose particles were loaded into "trays" that resemble flat graphite washers, each having 116 drilled holes in one face. One hundred eighty-four trays were stacked in columns in an alternate fertile particle-fissile particle sequence. The particles were irradiated for 169.4 full-power days. Peak fuel burnup in percent fissions per initial heavy-metal atom (FIMA) was as follows: ~20.5% for mixed-oxide fissile particles, ~33.5% for the other fissile particles, and ~7% for fertile particles. Maximum fast neutron fluence ($E > 183$ keV) at the end of the test was $\sim 7 \times 10^{25}$ neutrons/m². The report discusses methods used to specify particle loadings and contains thermal and neutronics results applicable to the irradiation test period.

2.4.2 Fission Product-Silicon Carbide Coating Interaction Studies — R. L. Pearson and T. B. Lindemer

The main objective of this program has been to acquire an understanding of the fuel, fission product, and coating kinetic processes and thermodynamics of the system to the degree that permits identification and quantitative description of failure mechanisms and improved fuel performance by means of kernel chemical modification.

The last scheduled topical report in the fission product-SiC coating interaction studies has been completed;³¹ the abstract follows.

Proliferation issues relating to the use of highly enriched uranium (HEU) have led to an evaluation of the fission product-SiC interaction problems if low enriched uranium (LEU) or medium enriched uranium (MEU) were used as fissile fuel in HTGR systems. Simulated Triso-coated UO_2 , UC_2 , or UO_2/UC_2 particles mixed with varying amounts of molybdenum, ruthenium, rhodium, palladium, silver, and cadmium were prepared. These fission products were chosen because, after full burnup, their concentrations are higher in LEU and MEU fuels than in HEU fuel. The particles were heat treated in the laboratory, then their behavior examined by metallography, scanning electron microscopy, and electron microprobe x-ray analysis. Ruthenium, rhodium, palladium, and silver were shown to interact with the SiC layer. The palladium-SiC interaction was the most severe. The rate of SiC thinning as a function of temperature was determined for rhodium and palladium. Kernel composition did not seem to influence the SiC thinning rate due to the Pd-SiC interaction. Palladium in UO_2 kernels, after it reached the SiC layer, probably formed silicides and a low-melting, palladium-rich eutectic. Palladium in UC_2 or UO_2/UC_2 kernels probably formed UPd_3 in the iLTI and SiC layers.

2.5 PROJECT WORK STATEMENT ACTIVITIES — FUEL DEVELOPMENT — E. L. Long, Jr.

Under the auspices of the umbrella agreement between the U.S. and the FRG governments for high-temperature reactor (HTR) technology development, a number of cooperative tasks have been identified that will require an exchange of information, materials for testing, and results between ORNL and Kernforschungsanlage (KFA), Jülich. This year there were five Project Work Statements in Fuel Development (PWS FD) that received attention: (1) PWS FD-10, Optimization of SiC Microstructure for Retention of Metallic Fission Products; (2) PWS FD-12, Quantification of Irradiation-Induced Permeability in Pyrocarbon Coatings;

(3) PWS FD-13, Comparison of IMGA and Postirradiation Annealing and Autoradiography (PIAA); (4) PWS FD-18, Fuel Chemical Performance Modeling; and (5) PWS FD-20, Evaluation of German Fuel Particles, Using IMGA.

2.5.1 PWS FD-10 — F. J. Homan

Several letters were exchanged during 1979 between technical specialists interested in the characterization and performance of SiC coatings for HTGR fuel particles. It was desired to coordinate the resources of KFA Jülich, GA, and ORNL and to focus these resources on the development of an SiC coating capable of retaining silver but resisting chemical attack of palladium. These two fission products have rather high yields in the LEU and MEU fuel cycles, compared with the HEU cycle. The LEU fuel cycle is currently the reference cycle for HTR introduction in Germany, whereas the MEU cycle is the reference cycle in the United States. Silver diffusion and palladium attack of the SiC may be the performance-limiting features of the LEU and MEU fuel cycles.

A PWS was drafted that features a three-stage approach to SiC microstructural optimization. The stages are (1) in-depth characterization and correlation of microstructure with deposition parameters, (2) out-of-reactor thermal gradient testing with doped (Ag and Pd) particles to identify the microstructures that are most resistant to diffusion/attack, and (3) irradiation testing of the most promising SiC to verify, under irradiation, the trends identified in the second stage.

This work statement must now be approved by the working group for the fuels/graphite/fission product subprogram and submitted to the ad hoc committee. The current draft of PWS FD-10 calls for work to begin in June 1980, assuming approval by that time.

2.5.2 PWS FD-12 — E. L. Long, Jr.

The objective and status of PWS FD-12 were reported previously.³² After completion of the installation of the PGA system this year,³³ work was initiated on analyzing the fission-gas contents of Biso-coated particles from the German experiment BR2-P12. A listing of the results to date are given in Table 2.22; fast-fluence exposures increase from

Table 2.22. Results of Xenon and Krypton Measurements on Fuel Particles from Experiment BR2-P12^a

Position- particle	Quantity of gas (nmol)						
	¹³¹ Xe	¹³² Xe	¹³⁴ Xe	¹³⁶ Xe	⁸³ Kr	⁸⁴ Kr	⁸⁶ Kr
0/5-1	2.2	7.7	9.4	15.0	0.2	0.8	1.5
0/5-2	3.4	12.1	14.7	23.2	0.3	1.3	2.2
0/5-3	2.4	7.8	9.4	15.1	0.2	0.8	1.5
0/5-4	2.0	6.6	7.9	13.1	0.2	0.7	1.2
0/5-5	6.4	24.5	29.6	46.7	0.9	3.5	5.9
0/11-1	10.9	42.2	48.8	69.6	1.4	5.6	9.7
0/11-2	8.7	33.4	40.5	68.6	1.1	4.8	7.2
0/11-3	0.3	0.7	0.9	2.1	0.07	0.07	0.2
0/11-4	0.2	0.4	0.5	1.3	0.02	0.04	0.09
0/11-5	0.6	1.7	2.2	4.1	0.09	0.2	0.4
0/11-6	9.7	36.3	43.7	71.3	1.1	5.1	8.0
0/16-1	8.1	31.8	37.5	64.1	1.0	4.3	6.7
0/16-2	8.5	33.1	39.6	65.3	1.0	4.5	6.8
0/16-3	8.5	32.7	38.7	64.2	0.9	4.4	6.7
0/16-4	8.6	33.7	40.0	65.6	1.0	4.6	7.2
0/16-5	0.3	0.6	0.8	1.7	0.03	0.1	0.1
0/34-1	0.4	1.4	1.8	3.7	0.08	0.1	0.3
0/34-2	0.2	0.8	0.9	1.9		0.06	0.2
0/34-3	0.5	1.4	1.7	3.3	0.07	0.1	0.2
0/34-4	0.7	1.6	1.9	3.7	0.03	0.2	0.3
0/34-5	1.3	2.8	3.3	6.2	0.06	0.3	0.4
0/34-6	1.0	2.9	3.6	6.2	0.2	0.3	0.4

^aAll particles in this series from batch DO-760; nominal dimensions: kernel diameter, 600 μm ; buffer thickness, 80 μm ; LTI thickness, 100 μm .

the top to the bottom of the table. Although the exact fast-fluence values are not available at this time, the particles from position 0/5 were from the lowest fluence region ($\sim 1.2 \times 10^{26}$ neutrons/ m^2 ; $E > 29$ fJ) and 0/34 is the highest fluence region ($\sim 1.4 \times 10^{26}$ neutrons/ m^2 ; $E > 29$ fJ). All of the particles from the lowest fluence region (0/5) were slightly permeable, and the particles from the highest fluence region (0/34) had lost essentially all of their fission-gas inventory. Work is continuing on particles from the other batches with thicker and thinner LTI layers than the particles discussed in this section. Other work remaining to complete this PWS includes measuring the fission-gas inventories of particles irradiated in HT-34³⁴ and HRB-14.³⁵

2.5.3 PWS FD-13 — E. L. Long, Jr.

The objective of PWS FD-13 is to compare PIE techniques used in the United States and in the FRG for the measurement of statistically significant failure fractions of irradiated coated particles. The IMGA system is used at ORNL; a description of this system and the statistical basis for failure-fraction determinations have been reported.^{36,37} Another technique, PIAA, has been developed and is in use at KFA.³⁸

Irradiated coated particles from a KFA capsule (BR2-P21) and an ORNL capsule (OF-2) have been agreed on, and arrangements have been made to exchange the particles. About 1300 Triso-coated fissile particles from OF-2 have been classified with the IMGA system³⁷ and are ready for shipment to KFA for PIAA analysis. About 2000 Triso-coated fertile particles from BR2-P21 have been received at ORNL and are scheduled for IMGA classification. The particles from OF-2 and BR2-P21 will be shipped to KFA for PIAA analysis after the failure fraction has been determined with the IMGA system. After failure fractions have been determined, a report will be prepared comparing the results of the two techniques.

2.5.4 PWS FD-18 — T. B. Lindemer

This PWS is entitled "Fuel Chemical Performance Modeling." The long-term objective is to predict the chemical behavior of fuel, fission products, and containment system (including Fe-Cr-Ni) under normal and hypothetical accident conditions. The first report is essentially written and will be issued as an ORNL report and submitted to the *Journal of Nuclear Materials*. The report assesses the thermodynamic behavior of the alkali metals (A), primarily the fission products cesium and rubidium, and their oxides with the metals and the oxides of other fission products and containment systems used in nuclear technology (SiC, stainless steel, and fusion reactor first-wall materials). A review of the entire alkali-metal family was needed to permit estimation of often unknown 298.15-K enthalpy-of-formation and entropy values for cesium and rubidium species from known values of analogous lithium, sodium, and potassium species. The report contains thermodynamic values, phase diagrams

(several were calculated and were previously unavailable), and Ellingham diagrams ($RT \ln P_{O_2}$ versus T) for the following systems: A-O; A-halides; A-O-H; A-(alkaline earths)-O; A-(Y, lanthanides)-O; A-(Ti,Zr,Hf)-O; A-(Cr,Mo, W)-O; A-Fe-O; A-(V,Nb,Ta)-O; A-Ni-O; A-(Th,U,Pu)-O; A-(S,Se,Te)-O; Cs-Al-Si-O, partial; and Cs-U-Mo-Te-Cr-Fe-H-O.

2.5.5 PWS FD-20 — F. J. Homan

The IMGA system, described in Sect. 2.6.1 of this report, is an advanced analytical tool for statistical measurement of the performance of coated-particle fuels. The automated particle-handling system makes possible the examination automatically of hundreds or thousands of individual coated particles in only a few days. Further, this system measures the parameter of most interest relative to the performance of the coated particles — the fission product inventory. ORNL has invested several hundred thousand dollars and several years of development time in this system, which is unique. Because of the speed and precision available through IMGA for coated-particle performance assessment, it is logical to make this tool available to others interested in HTR fuel development. At this time, "others" includes GA and the FRG. Accordingly, a PWS has been drafted, which would make it possible for German fuel to be examined with IMGA, with the incremental cost associated with IMGA operation charged against the cooperative program. This draft PWS has not yet been approved by the ad hoc committee (a government-level committee with representation from the United States, Germany, France, and Switzerland, which oversees the conduct of the cooperative program under the four-party umbrella agreement). However, it is anticipated that draft PWS FD-20 will be approved by summer 1980, when the first batches of German fuel for examination under this PWS will arrive in the United States.

2.6 EQUIPMENT DEVELOPMENT AND SUPPORT ACTIVITIES — E. L. Long, Jr.

The work described in this section is devoted to improving and increasing our capabilities in the postirradiation assessment of the

performance of irradiated coated-particle fuels. Three areas are included in this section as follows:

- review of the operational history of the IMGA system and listing of particles from various experiments that were classified this year,
- description of the PGA system that was developed at ORNL for measuring the fission and reactive gases contained within individual irradiated coated particles, and
- description and status of the x-ray micrographic facility currently being developed for use with irradiated coated-particle fuels.

2.6.1 Irradiated Microsphere Gamma Analyzer System — T. N. Tiegs

The IMGA system operated about 80% of the time during 1979 (approximately 10 out of 12 months). The system analyzed irradiated particles from experiments HT-34, HRB-13, HRB-11, and HRB-12. The status of the analyses is presented in Table 2.23.

Table 2.23. Status of Experiments Analyzed
by IMGA System

Experiment	Status of analysis
HT-34	Reported in Sect. 2.2.1.2
HRB-13	Reported in Sect. 2.2.2.1
HRB-11	Analysis in progress
HRB-12	Analysis in progress

In addition, the IMGA system analyzed miscellaneous particles prior to analysis by the PGA system (Sect. 2.6.2). All of those particles must be gamma-analyzed to determine the theoretical gas inventory of each individual particle.

2.6.2 Postirradiation Gas Analyzer System — J M Robbins, E. L. Long, Jr., and E. L. Ryan

A PGA system was built at ORNL for measuring quantitatively the fission- and reactive-gas inventories of irradiated HTGR fuel particles. This equipment was described earlier³⁹ along with certain problems encountered during initial checkout and "debugging."

During this reporting period the problems experienced earlier have been eliminated or minimized. Changing from a refrigeration unit to water cooling of the diffusion pump eliminated the vacuum problems and the inconsistencies related to cyclic variation in pump speed. Shifts in the location of mass numbers in the mass selector channels were minimized by adjusting the collimating magnets to prevent spreading of the electron beam as it transversed the ion source. Corrections of other troubles were effected by operational experience and by program changes in collecting and handling data designed to bypass the problem. Debugging of the equipment has progressed to the point that krypton and xenon are being determined on a routine basis. Currently, irradiated particles from the FRG experiment BR2-P12 are being evaluated for krypton and xenon under the FRG/US umbrella agreement (Sect. 2.5.2). Individual particles are broken at ambient temperature and the gases quantitatively analyzed. The measure of coating integrity will be determined by comparing the fission-gas inventories of the irradiated particles with their theoretical yields.

The inert gas permeability of Biso-coatings on HTGR particles is variable, and a measurement of it provides an important clue to particle performance. A method has been developed to measure the permeability of PyC coating by intrusion of light inert gases.⁴⁰ In this method the particles are intruded with a helium-neon mixture at elevated temperatures, broken at room temperature, and analyzed for helium and neon content. The ratio of these two gases in the particles provides a sensitive measure of the permeability of the particle coating. However, problems are still being encountered in devising techniques for analyzing for helium and neon in coated-fuel particles. The amounts of these inert gases of interest are in the picamole range, and accuracy at these low levels is difficult because of the low signal/background noise

ratio. One of the things being considered to improve the signal/noise ratio is the installation of a variable-frequency oscillator (VFO) and a sensitivity enhancer. The VFO would increase the duration of trap current flow with an attendant increase in ions created from the current 30 thousand spectral scans per second to about 100 thousand spectral scans per second. The manufacturer claims that, if used in conjunction with the sensitivity enhancer, an increased sensitivity of about 600% would be attained at a cost of about 20% loss in resolution. Another modification being considered is reducing the overall volume of the system through which the gas sample must be pumped. This would require redesigning the particle-breaking chamber to allow up-close coupling with the time-of-flight, mass-spectrometric (TOFMS) analyzing source.

The high-temperature breaking chamber will be used to measure and equilibrate the active gas (CO) back to the irradiation temperature prior to breakage. Other investigators^{41,42} have shown that the amount of CO generated in the particles during irradiation is important in making thermochemical evaluations of the particles. Data have not been generated to date by using the high-temperature chamber. However, unirradiated coated particles have been heated in the chamber to 2000°C without difficulty, and evaluation of the system with irradiated particles will soon be started.

A paper on the PGA was presented at the ANS Winter Meeting in San Francisco in November 1979; a report on the PGA and its use is being published.⁴³

2.6.3 X-Ray Micrographic Facility — E. L. Long, Jr.

An x-ray micrographic system has been designed and fabricated and is currently being installed in the HRLEL. This system will provide the capability of preparing x-ray micrographs of irradiated coated particles for detecting heavy-metal and fission product redistribution. A schematic of this system is shown in Fig. 2.14. The horizontal sub-assembly contains the film-transfer mechanism and sample holder. The film-transfer mechanism will handle 5- by 5-mm, high-resolution radiographic plates and is hand operated. The sample holder is a 0.13-mm-thick recessed beryllium window that supports the particles and maintains an alpha-tight seal for containment purposes.

Fig. 2.14. Arrangement of the Shielded X-Ray Micrographic System for Examining Irradiated Coated Particles. This equipment is located in the cubicle that is interfaced with a shielded x-ray diffractometer and a shielded electron microprobe.

The vertical subassembly is the entry sleeve that supports the x-ray tube. The x-ray source is located about 0.6 m above the beryllium window, which is sufficiently high to avoid beam divergence problems. Test exposures on unirradiated particles have been made by using a mock-up that supported the assembly in a geometry that simulated the cubicle arrangement. The resolution of the images equaled that of the equipment that is used routinely for quality control. However, these tests were made at very low voltages with exposure times of over 1 h. An optimum set of exposure conditions at higher voltages and shorter times must be established for irradiated particles to minimize film fogging from fission product decay.

To install the horizontal and vertical subassemblies of this system, it was necessary to machine 130-mm-diam penetrations through the wall and roof of the cubicle. The setup used to make the horizontal penetration through the 200-mm-steel wall of the cubicle is shown in Fig. 2.15. A drill press was attached to an electromagnetic base, as shown, and a relatively small (13-mm-diam) hole was drilled through the steel wall. The drill size was increased incrementally until a hole about 50 mm in diameter was produced. The hole was enlarged to its final diameter by using a boring bar with a cutting tool attached; the boring bar was aligned by using a self-centering bearing located on the inside wall of the cubicle. Installation of the equipment has been completed and should be operational in early 1980.

PHOTO Y-165692

Fig. 2.15. Equipment Setup Used to Machine the Horizontal and Vertical Penetrations Through the Shielded Cubicle in Preparation for Installing X-Ray Micrographic Equipment.

2.7 WORK ON FOREIGN ASSIGNMENT — M. J. Kania

The work outlined in this section was carried out while the author was on assignment to the Institut für Reaktorwerkstoffe (IRW) at KFA, Jülich, West Germany. This assignment is part of the U.S.-FRG umbrella agreement on high temperature reactor development.

2.7.1 Low-Enriched Uranium Fuel Evaluation

An evaluation was performed on HOBEG-prepared, Triso-coated LEU fuel particles that centered around two specific particle batches: (1) EO 249/250/251 t and (2) EO 403-405 t. Each of these particle batches was irradiated in one real-time irradiation experiment and in one accelerated test. For batch EO 249/250/251 t, the real-time test was DR-S4 in the Dragon reactor, and the accelerated test was BR2-P15 in the BR2-Mol reactor. For batch EO 403-405 t, DR-S6 was the real-time test in the Dragon reactor, and HFR-M5 was the accelerated test in the HFR-Petten reactor. This evaluation was based on available PIE data from these four irradiation experiments, and a report was issued on the results.⁴⁴ A summary of the conclusions reached in this evaluation is outlined in the following paragraphs.

2.7.1.1 EO 403-405

Irradiation performance of batch EO 403-405 t, a Triso-coated 615- μm -diam, UO_2 kernel with 6.65% enrichment, was excellent in both the real-time DR-S6 and the accelerated HFR-M5 experiments. No particle failures were observed in the fission product analysis performed on DR-S6 compacts after an irradiation of 550 full-power days (FPDs) to a peak burnup of 6% FIMA and a fluence of 4.3×10^{25} neutrons/ m^2 ($E > 16$ fJ). Similarly, no particle failures were indicated by in-reactor $^{85\text{m}}\text{Kr}$ R/B measurements in the HFR-M5 experiment. The R/B for $^{85\text{m}}\text{Kr}$ was 2×10^{-8} after 339 FPDs of irradiation. Peak burnup was near 8% FIMA, with a fluence of 6.3×10^{25} neutrons/ m^2 ($E > 16$ fJ). These results were somewhat surprising, because a banded structure in the SiC coating was noted in the preirradiation metallography. Such a structure would be a good indicator of poor irradiation performance. However, in this case the performance was the opposite.

2.7.1.2 EO 249/250/251

Irradiation performance of batch EO 249/250/251 t, another Triso-coated 616- μm -diam UO_2 kernel but with 9.49% enrichment, was good in the real-time DR-S4 experiment and extremely poor in the accelerated BR2-P15 experiment. No irradiation-induced particle failure was observed in DR-S4 after 656 FPDs with a peak burnup of 7.8% FIMA and a fluence of 8×10^{25} neutrons/ m^2 ($E > 16$ fJ). However, some fuel contamination was apparent, presumably caused by particle breakage during compact fabrication. Gross failure occurred in the BR2-P15 experiment, primarily because of the high fuel temperatures and thermocouple-fuel interaction. The failure mechanism most dominant was fuel kernel migration (Amoeba) with central fuel compact temperatures of $>1300^\circ\text{C}$ and thermal gradients at the compact surfaces of $>500^\circ\text{C}/\text{cm}$.

Recommendations were made for further PIE on the irradiated DR-S6 and HFR-M5 compacts to confirm the irradiation performance described here. These PIEs should include metallography, microprobe analysis, and possibly IMGA measurements for fission product retention and failure fraction.

2.7.2 (Th,U) O_2 Performance Assessment

Part of the development program for the advanced process heat system for nuclear coal gasification (PNP)/power generating HTR with direct circuit helium turbine (HHT) high temperature reactor concepts is the selection of a candidate fuel for the reactor core. This selection is contingent on verification of irradiation performance with a high degree of reliability. For the reference thorium-HEU fuel cycle in Germany, two particle systems are candidates for application to PNP/HHT. One system contains both fissile and fertile fuel, uranium and thorium, respectively, in a single particle and is called the "one-particle system." The other contains separate particles for the fissile and fertile fuels (same as U.S. reference system), the so-called "two-particle system." This performance assessment has as its objective to determine the candidate particle design from the one-particle system that has the best irradiation performance. Two particle candidates of

interest in the one-particle system are (1) the 400- μm -diam (Th,U) O_2 kernel with a high-temperature isotropic (HTI) Biso-coating and (2) the 500- μm -diam (Th,U) O_2 kernel with an LTI Triso-coating.

Initially it was necessary to identify those irradiation experiments in which mixed-oxide fuel of the type listed above was used. Some 37 experiments and nearly 45 different particle types were tentatively selected as having possible importance to this assessment. These large numbers of experiments and particle designs were then screened for the most relevant data. The screening was performed to determine quantity of fuel irradiated, experiment duration and significant operating conditions for PNP/HHT, integrity of experiment, and sufficient PIE available to assess performance.

For the HTI Biso-coated particle, the selected particle batches and their respective irradiation experiments are listed in Table 2.24. Experiments denoted by K, such as DR-K3, are fuel element experiments; whereas those denoted by P, such as BR2-P5, are fuel particle experiments.

Table 2.24. Relevant Particle Batches and Irradiation Experiments Selected for the Performance Assessment of the (Th,U) O_2 One-Particle System

Particle batch	Th/U ratio	Particle design ^a (μm)	Irradiation experiments
(Th,U) O_2 HTI Biso-coated particles			
WO 468 f	5	400-63-32-90	DR-K3, BR2-P5
GO 105 f	5	414-65-32-76	DR-K4, R2-K3, BR2-P9
EO 178	6	415-68-107 ^b	R2-K7, BR2-P10
EO 324/353/354	5	410-75-107 ^b	R2-K9
EO 1232-1234	5	407-77-104 ^b	HFR-K1, FRJ2-P22
EO 1084	10	410-75-107 ^b	R2-K10
(Th,U) O_2 LTI Triso-coated particles			
EO 211	48		R2-K5
EO 819/820	25		BR2-P21, BR2-P22
EO 1607	5		R2-K12, FRJ2-P23

^aKernel diameter, buffer thickness, seal thickness, and HTI thickness.

^bDimensions given for seal and HTI layers together.

They differ mainly in the number of fuel particles irradiated and the configuration of the fuel. A total of six different particle batches, irradiated in 11 irradiation experiments, was selected. Three experiments — HFR-K1, FRJ2-P22, and R2-K10 — are still in PIE. For the LTI Triso-coated particle, the selected batches and irradiation experiments are also listed in Table 2.24. Of those selected only particle batch EO 1607 is representative of the candidate Triso-coated particle for PNP/HHT. The experiments for this particle type are FRJ2-P23 and R2-K12. Experiment FRJ2-P23 is awaiting PIE; it was removed from the reactor in November 1979. Experiment R2-K12 is scheduled to be removed from the reactor in February 1980.

The primary effort has been centered on the HTI Biso-coated particle because of the large amount of data available compared with the Triso-coated particle. The experiments were carried out in a variety of facilities throughout Europe. The following list provides some information about each experiment and the facility in which the experiment was conducted.

<u>Experiment prefix</u>	<u>Facility</u>	<u>Environment</u>	<u>Reactor type</u>
DR-	Dragon HTR	Real-time	HTR
BR2-	BR2-Mol	Accelerated	Materials test LWR
FRJ2-	FRJ2-Jülich	Accelerated	Materials test HWR
HFR-	HFR-Petten	Accelerated	Materials test LWR
R2-	R2-Studsvik	Accelerated	Materials test LWR

In the review of the experiments conducted, it was consistently found that little or no thermal analysis was performed to determine thermal history for the irradiation experiment (exceptions being DR-K4 and HFR-K1, the latter now in progress). The majority of experiments selected in Table 2.24 are fuel element tests. In nearly all, thermocouples were used to monitor and control temperatures during irradiation. These thermocouples were placed in or on the surface of the fuel-free zone of the spherical fuel elements. The elements were then held in place by graphite cups in which the element was placed. Generally the

thermocouples were of the Chromel-Alumel type, which have good irradiation stability for long periods of time. (Fuel elements from the Dragon reactor tests contained no thermocouples on or in the element surfaces.)

A mathematical model was developed to describe the time-temperature behavior for those tests where a thermal analysis was lacking. It is a two-dimensional solution in R and θ (radial and polar angle directions) in spherical coordinates. A computer code based on this model was then written. Basic input data for the code are

- Thermocouple temperatures at two positions on the fuel element surface at various times during each cycle of irradiation (usually at the element pole and equatorial positions).
- Material heating rates, both fission and gamma, at similar times as thermocouple temperature readings. Provisions are also available for the code to perform these calculations when proper neutron fluxes and cross sections are provided.
- Thermal properties of fuel and fuel-free zone materials as a function of fluence, temperature, fabrication route, and particle volume loading.

Sample cases by use of small amounts of averaged input data have been successfully run with the thermal code. However, real-time data are as yet not available to perform a full-scale thermal analysis. Communication with the reactor sites indicates that such data should arrive soon.

Irradiation performance of fuel irradiated in particle experiments BR2-P5, -P9, and -P10 was based on in-reactor ^{133}Xe release measurements and postirradiation visual inspection of the loose particle holders or compacts. The irradiation conditions and specific comments for each particle experiment are described in detail in Table 2.25. The general conclusion that we reached from the loose-particle irradiations for the HTI Bisco-coated particle is that, for irradiation temperatures in the 1000 to 1250°C range, nearly 100% particle failure will occur early in life. This result is indicated by the high ^{133}Xe R/B values of $>10^{-2}$ at fluences of 2.4 to 3.0×10^{25} neutrons/m² ($E > 16$ fJ). In the one

Table 2.25. Irradiation Conditions and Performance of HTI Biso-Coated Particles as Determined from Particle Experiments Containing Loose Particles or Small Compacts

Particle batch	Experiment	Temperature (°C)	Fluence $\times 10^{25}$ neutrons/m ² ($E > 16$ fJ)	¹³³ Xe release (in-reactor R/B)	Comments
WO 468 f	BR2-P5	1150-1250	<2.8	BOL 10^{-6} EOL $>10^{-1}$	Visual inspection of loose particles only; some failed 100%, whereas others showed no failure; capsule R/B inconclusive because of different particle types in capsule
GO 105 f	BR2-P9 (capsules 1 and 2)	800-1250	<3.75	BOL 10^{-6} EOL $>10^{-1}$	Visual inspection on loose particles showed 80 to 100% failure in all positions; at fluence of 2.5, R/B jumped from a $<10^{-4}$ to a $>10^{-1}$ level
	BR2-P9 (capsule 3)	800-900	2.1	BOL 10^{-6} EOL 10^{-5}	Visual inspection on loose particles showed no failure
EO 178	BR2-P10	1000-1150	7.5-8.5	BOL 1.2×10^{-5} EOL 2×10^{-2}	All particles irradiated loose and in compacts, 100% observed failure; after fluence of 3, R/B was $>10^{-1}$ and dropped off at end of irradiation; R/B inconclusive because of different particle types in capsule

capsule where no failure was observed (batch GO 105 f, capsule 3 of BR2-P9) the operating temperatures were 800 to 900°C, and the peak fluence was 2.1×10^{25} neutrons/m² ($E > 16$ fJ). The predominant failure mechanism for the loose particles is fast-neutron-induced cracking of the HTI pyrocarbon layer. Also noted in visual inspection was the pressure-vessel failure mechanism. Both extremes were observed during visual inspection.

For the fuel element experiments, in addition to the in-reactor fission gas release measurements, fuel elements were subjected to detailed PIE at ÖSGAE, Seibersdorf, Austria. Detailed fission product measurements were made at ÖSAGE on the element and deconsolidated particles. In some cases, notably DR-K3 and DR-K4, the fuel elements were annealed near 1000°C at KFA to determine particle breakage in the elements. This annealing was accomplished by heating the elements in a helium-purged furnace and monitoring the released xenon and krypton gases. Irradiation performance for fuel elements is based on the results of defective particle fractions determined by in-reactor ¹³³Xe fission-gas release, ¹³⁷Cs release, and from free uranium determination. The ¹³⁷Cs release and free uranium determination are still in process for the HFR-K1 and R2-K10 experiments.

The defective particle fractions determined by the various methods are outlined below.

2.7.2.1 In-Reactor ¹³³Xe Fission Gas Release

Defective particle fractions based on fission-gas release are determined in the same manner as reported earlier.^{45,46} The procedure is to use the measured in-reactor R/B of a specific fission gas and to divide it by the fractional release of the same fission gas from a single failed particle.

2.7.2.2 Cesium-137 Solid Fission Product Release

Defective particle fractions based on cesium release use data generated in the electrolytic profile boring of fuel elements.⁴⁷ Boring through the diameter of the element usually requires 25 to 30 well-defined

steps to traverse a 60-mm-diam element. Each step contains a mixture of particles, matrix material, and electrolyte solution, which are then analyzed for fission product inventories. A cesium release fraction for each step can be determined by forming a ratio of the cesium inventory of the matrix and electrolyte to the total cesium in the matrix, electrolyte, and particles. Such a release fraction can be determined for each step deconsolidated along the diameter of the fuel element to form a profile. Particles from each step are used to determine the amount of cesium retained by the kernel, which is the internal cesium release fraction, also determined for each step of the boring. A defective particle fraction for each step can then be determined by dividing the cesium release fraction by the internal cesium release:

$$\text{Defective particle fraction} = \frac{{}^{137}\text{Cs release fraction } (A_{\text{free}}/A_{\text{total}})}{\text{Internal } {}^{137}\text{Cs fractional release}} .$$

This procedure assumes that a defective particle is one in which the coating fails to retain any of the cesium released from the kernel.

2.7.2.3 Uranium Determination by Chlorine Leach

Defective particle fractions are based on the amount of free uranium recovered by hot gaseous chlorine (700 to 900°C) leaching of deconsolidated particles and matrix materials from fuel elements. A ratio of the uranium recovered to the total amount of uranium in the sample gives a value for defective fraction:

$$\text{Defective particle fraction} = \frac{\text{Free uranium recovered by } \text{Cl}_2 \text{ leach}}{\text{Total uranium inventory}} .$$

In Table 2.26 the irradiation conditions and defective particle fractions for each element investigated are shown. The defective particle fractions determined by gas release have been assumed to vary by a factor of ± 2 . For the ${}^{137}\text{Cs}$ release defective particle fractions, a range was

Table 2.26. Irradiation Conditions and Performance of HTI Biso-Coated Particles as Determined from Fuel Element Experiments Containing Spherical Fuel elements

Particle batch	Experiment/sphere	Fluence $\times 10^{25}$ neutrons/m ² ($E > 16$ fJ)	¹³³ Xe release ^a (in-reactor R/B)	Defective particle fractions (%)		
				Gas release	¹³⁷ Cs release	Free uranium
WO 468 f	DR-K3/5	3.2		No failure ^b	0.7-2.5	0.1-0.9
GO 105 f	DR-K4/7	6.4		No failure ^b	0.2-0.9	0.02
	DR-K4/11	7.3		Few failures ^b	1.7-8.1	1.5
	R2-K3/1	2.9 ^c	2.3E-6-1.4E-4	0.03		
		5.2 ^c		0.3		
		6.1		>1	0.4-0.9	1.5
		2.7 ^c		0.08		
		6.1 ^c		0.33		
		7.4		>1	0.8-1.6	1.3 ^d
	R2-K3/3		4.0E-6-2.2E-4			
EO 178	R2-K7/3	9.3	5E-7-1E-7	<0.3		1.5
EO 324/353/354	R2-K9/2	8.7	1.6E-5-1.3E-5	<0.3	0.4-1.1	1.75
	R2-K9/3	7.1	1.4E-5-1.0E-5	<0.3		0.2

^aR2-K experiments are cycled at three temperatures - 600, 750, and 900°C; release/birth rate ratio data shown here represent only 900°C cycles.

^bNumber of failed particles determined during annealing test at 1050°C conducted at Kernforschungsanlage.

^cThese data points were selected in the calculation of failure fractions determined by gas-release measurements.

^dData on free uranium shown here were determined on fuel-element matrix material; during chlorination of the particles at 900°C, several were found to have broken due to the environment.

determined based on the internal cesium release of broken versus intact particles. For experiment DR-K4, data were available for the broken and intact particles. The internal ^{137}Cs release fractions, compiled from references 48 through 54, are shown in Fig. 2.16.

For all experiments other than DR-K4 broken-particle internal cesium release fractions were not known. In these cases 100% release was assumed. In Table 2.26, for the ^{137}Cs defective fraction, the lower value represents the calculation based on broken particles releasing all the cesium, and the higher value is based on the intact particle internal cesium release data from Fig. 2.16. For the defective fraction from free uranium determination, only data derived from the chlorination

Fig. 2.16. Internal ^{137}Cs Release Fractions as Compiled from Measurements Made on $(\text{Th},\text{U})\text{O}_2$ Particles from HTI Biso-Coated Particles Obtained in Fuel Element Irradiation Experiments.

on particles are given. In some cases data from the deconsolidated portion were combined with the integral values after proper weighting to arrive at the values given here. In one case, R2-K3 sphere 3, particles burst during the chlorination at 900°C so that the value given was determined on the matrix material.

One other important point about the cesium defective fraction is the manner in which it was determined for the total element, based on the central cylinder along the fuel element diameter. This region is not a representative random sample of fuel from the element. It contains too high a percentage of particles from the center of the element in comparison with those located nearer to the surface of the element. To overcome this situation, the element was divided into five regions, one central spherical region with a radius of 5 mm and four surrounding shells, each with a thickness of 5 mm. Each step from the profile boring was subsequently assigned to one of the regions, based on the dimensions of the top and bottom of the segment removed. In this manner several steps were assigned to each region and could be averaged to find a region-averaged defective particle fraction. The total element defective fraction was found by weighting each region by the fraction of total particles it contained and summing.

A comparison of the defective particle fractions determined by the three methods in Table 2.26 shows good agreement, with the exceptions of sphere 3 of R2-K7 and of sphere 2 of R2-K9. Here the gas-release data indicates a failure rate much lower than indicated by free uranium or by ^{137}Cs release. Investigation into PIE procedures that were applied to these elements before the profile boring was performed revealed that sphere 3 of R2-K7 had been subject to crushing strength tests and that sphere 2 of R2-K9 had been subjected to fall tests several times from a height of 1 m. These PIE procedures on the elements before profile boring may have damaged the HTI coatings of the particles, especially because both experiments were of long duration (both had a fast fluence of near 9×10^{25} neutrons/m²). Figure 2.17 is a plot of the defective particle fractions as determined by ^{137}Cs release for experiments DR-K3, DR-K4, R2-K3, and R2-K9 and as determined by calculations based on in-reactor fission-gas release for experiment R2-K3.

Fig. 2.17. Defective Particle Fraction Determined from ^{137}Cs Release and In-Reactor Fission-Gas Release for High-Temperature Isotropic Biso-Coated Particle Fuel Element Experiments.

The extrapolation of the defective particle fraction determined from cesium release agrees well with that calculated from fission-gas release. This result is particularly true for the R2-Studsvik temperature-cycled experiments. The two spheres where agreement is not so good (DR-K3 sphere 5 and DR-K4 sphere 11) were both high-temperature irradiations in excess of 1200°C sphere-center temperatures. In addition, both spheres were subjected to annealing tests prior to the profile boring investigations, which may have induced some particle breakage.

The data presented in Table 2.25 for the particle experiments and the data in Table 2.26 and Fig. 2.17 for the fuel element experiments suggest the following preliminary conclusions about the irradiation performance of the $(\text{Th,U})\text{O}_2$ 400- μm -diam kernel with an HTI Biso-coating:

- At fuel element average operating temperatures in the 650 to 950°C range and end-of-life fluences $>6 \times 10^{25}$ neutrons/m²

($E > 16$ fJ), the defective particle fractions showed a variation at the lower range of 0.13 to 0.4% up to a maximum range of 1 to 3%.

- At PNP/HHT end-of-life fluences of 4.5×10^{25} neutrons/m² ($E > 16$ fJ), the defective particle fraction ranges from a low of 0.06% to a high of >0.4%. This data compares with target values for PNP/HHT of 0.02 to 0.04%. It is unlikely that further optimization of the HTI Bisco-coated particle can reduce the defective particle fraction to the target specification.
- At elevated fuel element operating temperatures above 1250°C for extended times, the defective particle fraction increases to the 10 to 100% range, as observed in the loose-particle experiments.
- The predominant failure mechanism observed for the HTI Bisco-coating is fast-neutron-induced cracking of the HTI layer. Another failure mechanism, that of pressure-vessel failure, was also observed in the loose particle irradiations.

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3. PRESTRESSED CONCRETE PRESSURE-VESSEL DEVELOPMENT

D. J. Naus

3.1 INTRODUCTION

The Prestressed Concrete Reactor Vessel (PCRV) research and development program consists of generic studies designed to provide technical support for ongoing PCRV design activities, to contribute to the technological data base, and to provide independent review and evaluation of the relevant technology. These studies are identified through direct communication with PCRV designers, participation in ACI-ASME design code activities, and findings of literature reviews and communications with researchers in other countries. The overall program is divided into the following seven task areas: (1) analysis methods, (2) PCRV head failure studies, (3) concrete properties in nuclear environment, (4) prestressing systems, (5) liners and penetrations, (6) instrumentation, and (7) structural models. A paper¹ presented at the Specialists Meeting on Vessel Concepts for Gas-Cooled Reactors in Lausanne, Switzerland, contains an overview of results obtained under the task areas listed above. The abstract of the paper follows.

Under the Prestressed Concrete Nuclear Pressure Vessel Development Program at the Oak Ridge National Laboratory, various aspects of prestressed concrete pressure vessels (PCPVs) are investigated and evaluated with respect to reliability, structural performance, constructability, and economy. Based upon identified needs, analytical and experimental investigations are conducted. Areas of interest include finite-element analysis development, materials and structural behavior tests, instrumentation evaluation and development, and structural model tests. Studies have been recently completed in the following areas: concrete embedment instrumentation systems for PCPVs, grouted-nongrouted prestressing systems, acoustic emission as a technique for structural integrity monitoring, and model tests of steam-generator cavity closure plugs for a Gas-Cooled Fast Reactor (GCFR).^{*} An overview of results will be presented.

^{*} The major portion of the overall effort discussed in this report was supported by the HTGR Base Technology Program. The Gas-Cooled Fast Reactor model studies were funded under a separate program. These studies were conducted to provide verification and support for the PCRV design selected for the 300 MW(e) GCFR demonstration power plant.

Under the instrumentation evaluation task, 13 types of commercially available concrete embedment strain meters were evaluated with respect to general performance, performance in a simulated PCR/V environment, and performance under extreme operating conditions. General conclusions obtained were that (1) meter selection should be based on specific applications, (2) meter calibration factors should be determined for each application, (3) improved materials and sealing techniques are needed, and (4) other promising measurement techniques should be evaluated.

Grouted and nongrouted posttensioning prestressing systems were investigated under the structural models task. The investigation was divided into: relative performance of grouted and nongrouted systems, an evaluation of selected new materials for PCR/V applications, and a limited corrosion study to demonstrate the corrosion-inhibiting capability of grout. Results indicate that: grouted tendon beam elements provide improved crack control, improved ultimate load capacity in flexure, and anchorage conservatism; the polymer-silica cement and fibrous concrete material systems exhibit potential for PCR/V applications; and grout provides corrosion-inhibiting capability equivalent to commercial organic petrolatum-based products.

Acoustic emission (AE) is being evaluated as a technique for monitoring the structural integrity of pressurized systems. Data have been obtained from monitoring of a prestressed concrete beam tested to failure. General results indicate that AE is applicable to prestressed concrete and can be utilized to detect the onset and propagation of flaws, identify prior loading levels, and locate flaws.

Proposed designs of the PCR/V steam-generator cavity and central cavity closure plugs for a prototype GCFR are being evaluated using relatively small scale models. To date, two model tests have been conducted for the steam-generator cavity closure. Results indicate that the designs are overly conservative. Parallel analytical studies have shown the need for improved constitutive equations for concrete in order to provide capability for predicting the characteristic punching shear-type failures of the models.

3.2 ANALYSIS METHODS - D. J. Naus

The objective of the PCR/V analytical effort is to develop techniques that can be incorporated into existing modern finite-element computer programs to improve the accuracy and efficiency of these codes. Two

techniques under investigation are an improved constitutive model and a method for efficiently modeling the reinforcing steel contained in multicavity PCRVs.

3.2.1 Analytical Evaluations of the Endochronic Nonlinear Concrete Model

3.2.1.1 Description of the Endochronic Model

The endochronic model for concrete is an extension of the inelastic endochronic model first introduced by Valanis.² In this model, the inelastic strain accumulation is characterized by a scalar parameter z , called intrinsic time, whose increment is a positive definite function of the strain increment. The principal features introduced by Bazant³ were to include (1) the sensitivity of the intrinsic time increments caused by hydrostatic pressure, (2) the inelastic dilatancy resulting from shear straining, and (3) a description of the strain-softening that occurs after the peak stress has been achieved. The inclusion of strain-softening in the constitutive relations eliminates the necessity for defining separate compressive failure surfaces.

To satisfy the condition of isotropy, the incremental stress-strain relationships are expressed independently in terms of the volumetric and deviatoric components of stress and strain; the resulting relationships are:

$$de_{ij} = (ds_{ij}/2G) + (s_{ij}/2G)dz$$

and

(1)

$$d\varepsilon = d\sigma/3K + d\lambda ,$$

where $d\varepsilon$ and $d\sigma$ are increments of the volumetric strain and hydrostatic stress, and de_{ij} and ds_{ij} are increments of the deviatoric strain and stress; the variable $d\lambda$ is the increment in inelastic volumetric strain, and dz is the increment in intrinsic time. The shear and bulk moduli G and K are taken to be decreasing functions of the inelastic dilatancy given by

$$G = G_0[1 - 0.25(\lambda/\lambda_0)]$$

and (2)

$$K = K_0[1 - 0.25(\lambda/\lambda_0)] ,$$

where G_0 and K_0 are the initial moduli, and λ_0 is a constant. The increment in the inelastic dilatancy is related to the increment in the deviatoric strains by

$$d\lambda = \ell(\lambda) L[J_2(\epsilon), I_1(\sigma), \lambda] \sqrt{J_2(d\epsilon)} , \quad (3)$$

where $\ell(\lambda)$ and $L[J_2(\epsilon), I_1(\sigma), \lambda]$ are functions, $I_1(\sigma)$ is the first invariant of the stress tensor, and $J_2(\epsilon)$ is the second invariant of the strain tensor. The increment in the intrinsic time is related to an increment in the deviatoric strains by

$$dz = |d\eta/f(\eta)|$$

and (4)

$$d\eta = F(\epsilon, \sigma) \sqrt{J_2(\epsilon)} ,$$

where $f(\eta)$ and $F(\epsilon, \sigma)$ are functions of stress and strain invariants. The explicit forms of the functions ℓ , L , f , and F developed by Bazant³ are complicated and will not be included. The functional form developed by Bazant requires the specification of 19 constants. Using previously published experimental data, all 19 constants were related to a single material property — the compressive strength of the particular concrete.

3.2.1.2 Comparisons with Experimental Data

The University of Tennessee, under a subcontract, compared (1) the results of finite-element analyses of concrete structures with test data and (2) predicted and measured responses of homogeneous concrete test specimens subjected to uniaxial, biaxial, and triaxial states of stress.

Comparisons of the results of finite-element analyses were made with available test data on prestressed concrete beam models. The ADINA computer program was used to perform the analyses with two material models for concrete: a model furnished with the ADINA code and an improved endochronic model developed at ORNL. Despite the similarity between the two models with respect to cracking, the endochronic model was found to produce superior results where cracking was the controlling mode of failure in these tests. This finding probably resulted from the different finite-element implementations used by the two models; that is, the ADINA code prohibits equilibrium iteration of the ADINA model, whereas iteration is performed with the user-defined endochronic model. Equilibrium iteration is necessary when an incremental approach is used in solving nonlinear problems. On the other hand, the endochronic parameter approach is handicapped by its inability to account for the direction of inelastic strain or damage; that is, if a specimen is subjected to fatigue loading in one direction and is loaded with a new orientation, the endochronic parameter cannot take this fact into consideration.

The second phase of the study was related to an evaluation of the endochronic model's capability for predicting the stress-strain relationship for concrete through its comparison with existing data on concrete subjected to multiaxial loadings. The procedure followed in performing this evaluation included the following steps:

- documentation of source of data, test description, and specimen characteristics for each reference providing stress-strain data,
- digitization and storing of data in a computer file,
- run refined endochronic model for data digitized and store results in a computer file,
- compare results obtained in step 3 with information from step 2 by plotting results on same graph, and
- write brief evaluation of results obtained.

Thirty-two files were established by using this procedure. The majority of the data were from references used by Brazant et al.³ in formulations of the endochronic model, but 11 files were from other data. These results revealed good agreement for concretes of higher strength, but agreement was not as good for the lower-strength concretes.

Reference 4 presents more details of results obtained in this study.

3.2.2 Embedded Bar Modeling Technique

Most finite-element codes use truss elements to represent reinforcing in PCRVs; however, because of the extensive amount of reinforcement contained in these multicavity vessels, it is impossible to represent each individual rebar with a truss element. Current practice is to model the vessels by use of relatively large isoparametric elements, each of which may contain portions of several rebars. Also the truss element approach requires the rebars to lie only along the edges of the continuum elements; thus, many rebars are omitted from the analysis or are combined to form large single bars.

Currently under development at ORNL is a procedure that arbitrarily permits embedment of oriented rebars in isoparametric continuum elements. If the rebar is straight, its position within the continuum element is defined by the coordinates of the two end points of the bar. When the bar is curved, it is also necessary to input the coordinates of an intermediate point along the bar (curved embedded bars are required to model the complex array of rebars around penetrations in PCRVs). Stress-strain behavior of the rebars may be done by using any of three currently available material models: (1) linear elastic, (2) elastic-plastic with isotropic hardening, and (3) elastic-plastic with kinematic hardening. Typical analyses of reinforced-concrete structures would use the elastic-plastic model with isotropic hardening for modeling the rebars. This procedure has been implemented into the two-dimensional element library of the ADINA finite-element computer program.

Coding for implementation of the technique into the three-dimensional-element library has been virtually completed. Results of trial analysis

indicate that the embedded bar routines are functioning properly. Experimental results obtained from test specimens fabricated by Wiss et al.⁵ to simulate the anchorage zone of posttensioned tendons in secondary containment vessels of nuclear power plants are being used as a reference for checking and debugging the procedure. Figure 3.1 presents the three-dimensional continuum elements that represent the concrete and steel bearing pad of one of these test specimens being considered. The input data file for analysis of this specimen has been completed. Eighty-four elements were used to model the concrete, and five elements were used to model the bearing pad. The input file for the second anchorage block being considered has also been completed. One hundred thirty-six embedded bar elements were required to model the 19 rebars in the specimen. Analyses of these two anchorage blocks have been attempted; however, the solution routine has exhibited poor stability, resulting in divergence. This divergence may be the result of an oversight in the coding of the solution routine, which

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Fig. 3.1. Analytical Model of End Anchor Test Specimen.

was recently discovered after these runs had been made. This oversight has since been corrected, but the runs have not been resubmitted.

3.3 PCRV HEAD FAILURE STUDIES — D. J. Naus

A report was published,⁶ which presents results obtained under a subcontract with the University of Illinois to investigate the behavior and strength of flat end-slabs of cylindrical prestressed concrete nuclear reactor vessels. A summary of the report follows.

The objective of this investigation was the development of a method to permit reliable calculation of the strength of end-slabs of prestressed concrete nuclear reactors.

The experimental study included the tests to failure under internal pressure of ten small-scale cylindrical vessels with flat end-slabs. The vessels were prestressed circumferentially and longitudinally. The end-slab contained no additional reinforcement.

The experimental variables were the end-slab thickness, size and arrangement of penetrations in the end-slab, and concrete strength. The penetrations, which were not reinforced by sleeves or liners, were closed by steel plates at the pressurized surface of the slabs.

Each vessel was pressurized to failure over a period of approximately 3 h. The range of attained maximum pressures was from 12.4 MPa (1800 psi) to 26.0 MPa (3765 psi). End-slabs of all test vessels failed in shear. Failures were violent, despite the use of fluid to develop the internal pressure.

The failure pressures indicated decisively the effects of changes in slab thickness and concrete strength. However, end-slab strength was relatively insensitive to changes in the amount of cross section removed by the penetrations.

An axisymmetric nonlinear finite-element model was developed to simulate the behavior of the physical models. The model was successful in indicating the observed changes in (1) internal stress paths, (2) internal strains, and (3) slab deflections, as well as the observed strength of the test vessels.

A significant feature of the analytical model is that it uses a very simple failure criterion for the concrete (Mohr in compression and limiting strain in tension). Close agreement between experimental and analytical results confirms that over-all nonlinear response phenomena in thick end-slabs of prestressed

concrete pressure vessels may be estimated satisfactorily without the necessity of detailed information on the response of concrete subjected to a three-dimensional stress field.

Shear strength of the test vessels was calculated successfully without "calibration" constants derived from the test results of the vessels. Required input data were limited to information on the geometry of the vessel, effective prestress forces, the compressive strength of the concrete, and type of loading.

3.4 CONCRETE PROPERTIES IN NUCLEAR ENVIRONMENT — D. J. Naus

The availability of data representative of concrete behavior under loadings (multiaxial and thermal) likely to be encountered in a PCRV is quite limited. To provide data that are characteristic of the concrete behavior in a PCRV under both normal and postulated accident conditions, experimental studies are to be conducted. Conducting studies, however, requires development of a relatively unique testing system, to include the capability of loading concrete specimens multiaxially in tension, in compression, or in a combination of tension and compression at either room or elevated temperatures. Activities during this reporting period were directed at developing such a test system. Included in these activities were (1) development of the facility to house the test system, (2) review of facilities that have performed tests on concrete under conditions somewhat representative of those for a PCRV, and (3) informal discussions with potential vendors of such a system.

3.4.1 Test System Enclosure

The design of the enclosure to house the concrete multiaxial testing machine has continued. The enclosure is required both to provide standard laboratory conditions so that data obtained will be meaningful and to protect the complex servohydraulic loading system and peripheral equipment from thermal fluctuations and contaminants that would impair their operation. To date, the facility design has been completed, and the required documentation for construction under a lump-sum contract has been completed. Construction of the enclosure is scheduled for completion during the next reporting period.

3.4.2 Facility Review

As noted during the previous reporting period,⁷ an obvious need exists for experimental data on the multiaxial behavior of concrete for development of more representative constitutive models for input into modern finite-element codes. Despite the lack of data, only a few facilities are involved in this activity. In an effort to assemble as much data as possible, to reduce design time, and to develop an optimum facility at ORNL, organizations that have conducted tests under conditions somewhat representative of those encountered in a PCRV were visited. Discussions were held with representatives of Fried Krupp GMBH, Essen, Federal Republic of Germany (FRG), and of Institut für Baustoffkunde and Stahlbetonbau der Technischen Universität Braunschweig, FRG.

Fried Krupp has been involved in multiaxial property testing of concrete. In these tests 100-mm cube specimens were used. Specimen loading was applied by a three-actuator hydraulic system that applied a programmed loading (constant rate) to the specimens along each of the three axes. Strain gages were contained in the specimens so that load-strain data could be obtained from the test. The Krupp multiaxial test system differs from others in the interfaces between the platens and the specimens. Krupp utilized a sandwich construction of 12 aluminum foil sheets 0.1 mm thick, separated by a molybdenum disulfide lubricant (friction between specimen and platen for this interface was estimated at 1%). The ability to load in tension in one direction was provided by replacing one set of the aluminum foil platens with polycarbonate plates 5 mm thick, containing 3-mm-deep cuts parallel to the edges on 4-mm spacings (brush type platen), which were epoxied to the specimen (friction between specimen and platen for this interface was estimated at 2.5%). Some elevated-temperature work was performed with sealed specimens, in which a plastic seal was used at lower temperatures and a copper foil was used to seal the specimens for higher temperature. The considerable problems of sealing specimens for temperatures greater than 100°C were pointed out; that is, it was estimated that internal tensile stresses on the order of 50 bars are developed at 1000°C. It was suggested that future work might be directed toward evaluating the viscoelastic

behavior of concrete for normal temperature applications of 40 to 50°C and investigating strength development (material performance) for temperatures up to 200°C. It should be noted, however, that these suggestions were made with reference to the German High-Temperature Reactor with Gas Turbine (HHT) plant.

The technical university at Braunschweig was visited to obtain information about its work on concrete behavior under thermal loadings. Its primary program consisted of conducting tests in four areas:

- (1) determination of stress-strain relationships at elevated temperatures,
- (2) determination of restraint forces in heated restrained concrete specimens,
- (3) determination of deformations of heated concrete under constant load, and
- (4) determination of creep values for concrete specimens at elevated temperatures.

The desire to test specimens fabricated from practical concrete mixes (aggregate sizes of up to 16 mm) necessitated the development of special equipment such as a 10^3 -kN hydraulic test machine equipped with an electric furnace. One of the main variables in the Braunschweig testing program was the aggregate material used for the concrete mix; the types investigated included quartz, calcite, hornblende, schist, and sandstone. Rigid-steel, spherically seated platens were used to load the specimens, and strain measurements of up to 900°C were made with a dilatometer-based system, which the Braunschweig staff had developed. Concrete temperatures were recorded from five thermocouples contained in each specimen tested; the average of these readings was used as the test temperature. Associated investigations were also being conducted to evaluate the ability of AE to detect cracking in concrete structures as a function of loading and thermal effects, the effect of temperature ($\leq 900^\circ\text{C}$) on the bond between concrete and reinforcing steel, and the elevated-temperature behavior of fiber-reinforced concrete.

3.4.3 Multiaxial Test System Development

Discussions were held with several potential vendors of a concrete multiaxial material test system. Subjects of discussion included automated testing, hardware-computer configuration, hardware-test station

interface, software fundamentals and applications to testing, fracture-testing applications, and examples of automated testing systems. In addition, problem areas associated with development of a test machine capable of providing concrete property data under multiaxial loadings at either room or elevated temperatures were discussed. Included in these problem areas were the loading platens (interface should not constrain lateral movement of specimen, platen material selected must withstand temperatures up to 316°C , and option to incorporate tensile loadings should be included), instrumentation for determining specimen deformations (access to specimen, proximity versus embedment strain gages), and techniques for specimen heating up to 316°C .

Based on results of these discussions, it was decided that, since such a testing system is relatively unique, especially in this country, the conceptual designs for the test frame, platens, specimen heating device, and specimen deformation instrumentation will be developed in-house. The balance of the system (actuators, power supply, controller, etc.) will be procured from a vendor who meets a performance specification currently under development.

A "first cut" at a conceptual design of a test frame for a testing system that may be utilized for obtaining concrete properties under multiaxial loadings at either room or elevated temperatures was developed. Figure 3.2 presents a view of the system for applying loads in the Z-direction, and Fig. 3.3 presents the system as it might appear for applying loads in the X- and Y-directions. As techniques are developed for solving the problems associated with platen-specimen interface, deformation measurements, and specimen heating, this design will be modified.

3.5 INSTRUMENTATION EVALUATION AND DEVELOPMENT — D. J. Naus

An evaluation of the ability of AE to monitor structural elements, specifically PCRVs, has continued through laboratory studies. To develop background data on AE, both concrete and steel structural elements have been investigated.

Fig. 3.2. Test Frame: Z-Direction Conceptual Loading System.

3.5.1 Prestressed Concrete Test Specimens

Results obtained from AE monitoring of the 0.15-m-wide by 0.30-m-deep by 3.1-m-long grouted and nongrouted tendon prestressed concrete beams tested under the structural models task were examined in more detail. This examination was performed to investigate the relationship between the beam behavior and AE, the actual beam crack locations, and locations derived from AE results.

Figure 3.4 presents centerline deflection, cumulative event count, and cumulative event magnitude data as a function of load for a typical grouted tendon beam. Results indicate that prior to cracking (elastic range) AE activity was generally absent or of very low level. After first cracking (transition range) AE activity occurs at a relatively uniform rate until the onset of failure (plastic range) and a rapid increase in activity.

Fig. 3.3. Test Frame: X- and Y-Direction Conceptual Loading System.

The ability of AE to locate cracks in grouted and nongrouted prestressed concrete beams is presented in Figs. 3.5 and 3.6, respectively. Numbers shown adjacent to the cracks in the beams in these figures are kips of applied load and denote the extent of cracking at that load. Results for the grouted tendon beams in which four or more cracks generally formed indicate that AE can locate cracks in the beams. However, as more than one crack forms between adjacent transducers or as the extent of cracking increases (branching), the concentration of AE activity has a tendency to "smear" because the source-initiated stress wave must travel around cracks from transducer to transducer for source location. Results show that, for nongrouted tendon beams in which only

Fig. 3.4. Acoustic Emission Beam Behavior for a Typical Grouted Tendon Beam. (a) Load versus centerline deflection; (b) load versus cumulative event count; and (c) load versus cumulative event magnitude.

Fig. 3.5. Acoustic Emission Results Obtained for a Grouted Tendon Beam. (a) First cracking: $P = 48.9$ kN (11 kips); (b) secondary crack initiation: $P = 53.4$ kN (12 kips); (c) crack development: $P = 66.7$ kN (15 kips); (d) crack branching: $P = 89.0$ kN (20 kips); (e) failure: $P = 106.8$ kN (24 kips); and (f) failure: $P = 106.8$ kN (24 kips).

Fig. 3.6. Acoustic Emission Results Obtained for a Nongrouted Tendon. (a) First cracking: $P = 129.0$ kN (29 kips); (b) crack branching: $P = 142.3$ kN (32 kips); (c) failure: $P = 204.6$ kN (46 kips); and (d) failure: $P = 204.6$ kN (46 kips).

one or two major cracks occurred during testing to failure, AE-determined crack locations and actual crack locations correlated quite well.

3.5.2 Steel Pressure-Vessel Test Specimens

Steel structural elements were also monitored to develop both a reservoir of data on AE and improved capabilities for monitoring complex structures. These structures were monitored while they were tested under the Heavy-Section Steel Technology Program at ORNL. Based on these

tests three reports were published.⁸⁻¹⁰ A summary of each of these reports follows.

*Acoustic Emission Monitoring of HSST Intermediate Test Vessel V-8:*⁸

An HSST intermediate test vessel V-8 was fabricated from ASTM A533, grade B, class 1, steel plate. The test vessel had an outside diameter of 991 mm and an inside diameter of 686 mm. The test vessel was designed both to evaluate a particular type of weld repair in a thick-walled pressure vessel and to verify that the residual stresses resulting from a nonstress relieved repair weld are important in the transition region where material toughnesses are well below upper-shelf values. Contained in the vessel was a surface flaw located in a half-bead, weld-repair region where residual stresses were high. Acoustic emission data were obtained during both pretest notch pressure cycling, to sharpen a part-circular saw cut that had been placed in the vessel wall in the region of interest, and pressurization to failure.

*Acoustic Emission Monitoring of Intermediate Pressure Vessels Tested Under the ORNL Heavy-Section Steel Technology Program:*⁹

Acoustic emission as a technique for detecting and locating flaws in thick-walled steel pressure vessels is being investigated as part of the Heavy-Section Steel Technology Program at ORNL. Detailed descriptions of the AE monitoring of the most recently completed vessel test, V-7B, and of the fatigue crack sharpening of the flaw for test vessel V-8 are presented. Results obtained during these tests indicate that AE can locate flaws, detect onset of flaw growth, and provide an indication of the rate of crack extension (immediacy of fracture).

*Acoustic Emission Monitoring of Steel and Concrete Structural Elements with Particular Reference to Primary Nuclear Containment Structures:*¹⁰

Primary containments for nuclear reactors are either thick-walled steel pressure vessels (pressurized water reactors) or massive prestressed concrete structures (gas-cooled reactors). The ability to detect and analyze flaws in these structures is important for ensuring structural integrity throughout their 30 to 40-year design life and, thus, for preventing a catastrophic in-service failure in the unlikely event that conditions would be favorable for such an event to occur. One of the techniques being evaluated at ORNL to detect flaws in these structures is AE. To date, AE has been used to monitor two flawed thick-walled steel pressure vessels tested to failure, pressure-fatigue cycling of the flaw contained in one of the above vessels, and tests to failure of both plain and prestressed concrete elements. Results are presented for each of these tests.

Two 152-mm-thick, 991-mm-OD steel pressure vessels have been tested under the Heavy-Section Steel Technology Program. Both vessels, fabricated from ASTM A533, grade B, class 1, steel plate, contained surface flaws. The first vessel contained a flaw formed by hydrogen charging of an electron-beam weld in the heat-affected zone of a previous weld repair, and the second vessel contained a flaw located in a half-bead, weld-repair region where residual stresses were high. Test temperatures for the vessels were 87 and -23°C, respectively. The first vessel was pressurized in two cycles, with the first cycle to approximately 110% design pressure and the second to failure. Vessel two was pressurized to failure, with intermediate pressure holds for instrument scans and after-flaw pop-ins. Acoustic emission data were obtained throughout the testing of both vessels and also during sharpening of the flaw contained in the second vessel. Results indicate that AE can monitor flaw growth in thick-walled pressure vessels, can indicate the relative rate of crack extension, and can readily identify the presence of leaks in hydraulically pressurized structures.

Concrete elements are being monitored by AE to provide data for assessing the feasibility of utilizing this technique to monitor concrete containments. Results are presented for two limited test series that have been conducted: (1) monitoring of plain concrete compression cylinders (152.4 mm diam by 533 mm long) and prismatic flexure members (152.4 × 152.4 × 914 mm) and (2) prestressed concrete beam structural members (0.15 × 0.30 × 3.1 m). Plain concrete members were fabricated from three different strength concretes (19.5, 36.8, and 50.8 MPa) to determine strength effects on AE results. Three of four cylinders cast for each strength concrete were slowly loaded to failure in compression. The fourth cylinder in each set was cycled to failure, with each succeeding load cycle increased. The prismatic plain concrete specimens cast for each strength concrete were tested in flexure (four-point bending). Grouted and nongrouted tendon beam elements were also tested in flexure to failure. Acoustic emission data were obtained throughout each of these tests. Results indicate that, for plain concrete specimens loaded to failure in one cycle, there was good correlation between AE and stress-strain curves relative to AE activity and extent of cracking; for specimens cyclically loaded to failure, there is evidence that the Kaiser effect is applicable to concrete; for flexure specimens whose tensile stress-strain relationship is essentially linear to failure, there was an absence of AE activity until just prior to failure; and, for the prestressed concrete beams, there was good correlation between AE source location data where only one, or possibly two, cracks form between transducers, but, as the number of cracks increases, there is a "smearing" of AE source location data.

3.6 LINER AND PENETRATION STUDIES — R. K. Nanstad and D. A. Canonico

The PCRV for the HTGR contains several steel penetrations and closures, which are part of the liner system. In previous annual reports the requirements of Appendix G to Division 1, Section III of the *ASME Boiler and Pressure Vessel Code* were discussed in detail, in particular, the K_{IR} (reference fracture toughness) curve and the R_{NDT}^{RT} (reference nil-ductility temperature) requirement. The materials of interest to the liner testing program, as well as specimens and test procedures, are also discussed in previous reports. A report entitled *Characterization of PCRV Liner Steels*¹¹ has been completed, which discusses chemical analysis, metallography, and tensile and Charpy impact testing of three plate steels and of one forging steel.

Testing during this reporting period involved weldment W-3. Weldment W-3 was fabricated from A 508, class 1 (forging F1), and A 537, class 2, plate. The A 537 plate used for this weldment is not from the same plate studied in this program; hence, its properties have not been determined. The weld root was deposited with the shielded metal-arc process (10% of the weld). The weld was completed with the flux cored-arc process (90%). The weldment was given a postweld heat treatment at 566 to 621°C for 5 h. A macroscopic view of the weldment cross section is shown in Fig. 3.7. The direction of welding is perpendicular to the photograph. The heat-affected zones (HAZs) are evident on both sides of the weld, and the weld root is pronounced. Because of the relatively narrow weld root, the possibility of carbon dilution (of the base metal) to the weld metal motivated the study of both root- and bulk-weld metal properties. Test specimens are oriented transversely to the direction of welding, and the notch is oriented in the direction of welding and perpendicular to the base metal surfaces.

The chemical compositions of the root weld and bulk weld are shown in Table 3.1. The primary differences between the bulk- and root-weld metals are the copper and nickel contents. The specifications for the shielded metal-arc-weld metal (root) and flux-cored-arc-weld metal (bulk) require nickel contents of 3.0 to 3.75%. The nickel content shown in Table 3.1 for the root-weld metal appears to substantiate the analysis

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Fig. 3.7. Macroscopic Views of Weldment W-3 Cross Section.

Table 3.1. Chemical Compositions of the Root Weld and Bulk Weld

Weld metal	Composition (%)										
	C	Mn	Si	Mo	P	S	Cr	Ni	Cu	V	Al
Bulk	0.069	0.90	0.42	<0.10	0.011	0.022	<0.10	3.39	<0.10	<0.05	0.003
Root	0.080	0.96	0.44	<0.10	0.011	0.020	<0.10	2.45	0.23	<0.05	0.004

in that dilution effects would be expected to reduce the nickel content by at least the amount indicated.

A microhardness traverse across the root area of the weldment was made to define unusually hard areas. Figure 3.8 shows the results of the microhardness tests (10-kg load) in terms of diamond pyramid hardness (DPH) values.

Both HAZs show similar values near the fusion line, about 240 to 245 DPH. The hardest areas in the weld metal are also 240 to 245 DPH, and they exist in the areas close to the HAZs. All areas in the weld metal have hardness values above 220 DPH. A similar survey across an area of the bulk weldment showed some values of from 200 to 220 DPH in the center region of the weld. The hardness traverse pointed out that the center of the root-weld metal is about 20 DPH points higher than the center of the bulk-weld metal. Thus, higher strength will be found in the root-weld area, and this generally corresponds to the higher carbon content because of dilution from the base metal. The carbon content reported in the manufacturer's certificate of analysis was 0.04 to 0.05%. Thus, the measured content of 0.08% in the weldment indicates carbon dilution from the base metal. Additionally, the higher cooling rate in that area (because of lower heat input during welding) could increase the hardness (thus, strength).

Charpy impact test results for the HAZs of the forging and plate are shown in Figs. 3.9 and 3.10. The data are very similar, with the plate HAZ exhibiting slightly higher impact values on the upper shelf.

The forging HAZ Charpy curve is similar to that for the forging-base metal, except at the upper-shelf energy level, which, for the forging, is about 300 J. The plate base metal in W-3 has not been characterized.

Fig. 3.8. Hardness Traverse of Weldment W-3 in the Root Region of the Weld.

Fig. 3.9. Charpy Impact Toughness for Plate Heat-Affected Zone, Weldment W-3.

Fig. 3.10. Charpy Impact Toughness for Forging Heat-Affected Zone, Weldment W-3.

Figures 3.11 and 3.12 show the Charpy impact results from the bulk-weld metal and the root-weld metal, respectively. The transition region of the bulk metal is about 50°C lower than that of the root metal, whereas the upper-shelf energy level is somewhat higher in the bulk than in the root. Thus, the Charpy impact tests indicate that the bulk-weld metal toughness is superior to that of the root-weld metal. This

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Fig. 3.11. Charpy Impact Toughness for Bulk-Weld Metal, Weldment W-3.

difference was anticipated because of the higher carbon content in the root area. Figure 3.13 provides a comparison of Charpy impact behavior of the HAZs and weld metals and shows the superior toughness of the HAZs with regard to transition behavior and upper-shelf energy levels.

Fracture toughness tests to date have been conducted with precracked Charpy specimens under slow-bend and impact loading conditions. Figures 3.14 and 3.15 show the results of slow-bend fracture toughness for the HAZs. The data are quite similar for the entire range of behavior.

Fig. 3.12. Charpy Impact Toughness for Root-Weld Metal, Weldment W-3.

Fig. 3.13. Comparison of Charpy Impact Toughness for Various Regions of Weldment W-3.

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Fig. 3.14. Pre-cracked Charpy Fracture Toughness of Plate Heat-Affected Zone, Weldment W-3.

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Fig. 3.15. Pre-cracked Charpy Fracture Toughness of Forging Heat-Affected Zone, Weldment W-3.

Both HAZs have excellent static fracture toughness, with upper-shelf toughness levels attained at -100°C . Figures 3.16 and 3.17 show the slow-bend results for the bulk- and root-weld metals. Although the number of tests are somewhat scarce for the root-weld metal, the data indicate transition behavior occurs at lower temperatures for the bulk metal, similar to the results shown by Charpy impact tests.

Fig. 3.16. Precracked Charpy Fracture Toughness of Bulk-Weld Metal, Weldment W-3.

The effects of higher loading rates are presented in Figs. 3.18 through 3.21, which show the results of precracked Charpy impact fracture toughness testing. The HAZ toughness curves show a dramatic increase to higher temperatures when subjected to dynamic loading, with a "shift" of about 75 to 100°C . Similarly, Fig. 3.20 shows a shift in transition temperature toughness of about 75°C for the bulk-weld metal. Figure 3.21, however, shows that dynamic loading had much less effect on the toughness of the root-weld metal, with a shift of only about 25°C .

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Fig. 3.17. Pre-cracked Charpy Fracture Toughness of Root-Weld Metal, Weldment W-3.

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Fig. 3.18. Dynamic Pre-cracked Charpy Fracture Toughness of Plate Heat-Affected Zone, Weldment W-3.

Fig. 3.19. Dynamic Precracked Charpy Fracture Toughness of Forging Heat-Affected Zone, Weldment W-3.

Fig. 3.20. Dynamic Precracked Charpy Fracture Toughness of Bulk-Weld Metal, Weldment W-3.

Fig. 3.21. Dynamic Precracked Charpy Fracture Toughness of Root-Weld Metal, Weldment W-3.

Additional dynamic tests will be conducted to define better these preliminary observations.

Testing now under way includes dynamic precracked Charpy testing with weldment W-3, weldment W-5, and weldment W-1 as identified priorities, along with drop-weight NDT and Charpy V-notch impact tests. Following testing of weldments, plans include completion of test schedules for forgings F2, F3, F5, and the 1 1/4 Cr-1/2 Mo steels (forging T2 and weldment T4).

3.7 STRUCTURAL MODEL TESTS — D. J. Naus and C. B. Oland

Activities under this task have been related to completing the grouted-nongrouted prestressing tendon study, overviewing potential new construction material systems for application to PCRVs, and initiating a fibrous concrete study.

3.7.1 Grouted-Nongrouted Tendon Study

A study to appraise the applicability of grouted tendon prestressing systems to PCRVs was completed. the results of this study are contained in three reports;¹²⁻¹⁴ a summary for each of these reports follows.

*Structural Model Testing for Prestressed Concrete Pressure Vessels: A Study of Grouted Versus Nongrouted Posttensioned Prestressing Tendon Systems:*¹²

Nongrouted tendons are predominantly used in this country as the prestressing system for prestressed concrete pressure vessels (PCRVs) because they are more easily surveyed to detect reductions in prestressing level and distress, such as results from corrosion. Grouted tendon systems, however, offer advantages that may make them cost effective for PCPV applications. Literature was reviewed to (1) provide insight on the behavior of grouted tendon systems, (2) establish performance histories for structures utilizing grouted tendons, (3) examine corrosion protection procedures for prestressing tendons, (4) identify arguments for and against using grouted tendons, and (5) aid in the development of the experimental investigation. The experimental investigation was divided into four phases: (1) grouted-nongrouted tendon behavior, (2) evaluation of selected "new" material systems, (3) bench-scale corrosion studies, and (4) preliminary evaluation of acoustic emission techniques for monitoring grouted tendons in PCPVs. The groutability of large tendon systems was also investigated. Results indicate that: grouted tendon flexure specimens exhibit improved performance relative to nongrouted members, the polymer-silica and fibrous concrete material systems exhibit potential for applications to PCPVs, the corrosion-inhibiting ability of grout in the environments investigated is equivalent to that of organic-petrolatum-based commercial corrosion inhibitors, acoustic emission is capable of monitoring plain concrete and simple concrete structures, and techniques are available for grouting large prestressing tendon systems such as would be used in a PCPV.

*An Evaluation of the Effectiveness of Selected Corrosion Inhibitors for Protection of Prestressing Steels in PCPVs:*¹³

The objective of this study was to evaluate the corrosion protection provided prestressing steel by portland cement-based grout in the presence of sulfide, nitrate, and chloride ion environments. Results were compared with those obtained from selected, commercially available petroleum-microcrystalline waxes (petrolatums) compounded with organic corrosion inhibitors. The investigation was conducted in two phases: (1) a review of literature to establish the mechanisms of prestressing steel corrosion, techniques available for protection of prestressing steel in hostile environments, and the performance of structures

that have utilized either nongrouted- or grouted-tendon prestressing systems and (2) a laboratory study to develop relative performance data for portland cement grout and selected commercial petroleum-based greases and waxes containing inhibitors. Conclusions derived from the investigation indicate that (1) sulfide, nitrate, and chloride salts must be excluded from prestressing materials; (2) prestressing materials must be continuously protected from inimical environments; (3) the effectiveness of the protection provided by both the organic- and cement-based corrosion inhibitors is reduced unless the steel is completely covered; and (4) both cement- and organic-based corrosion inhibitors completely protect prestressing materials when properly applied.

*Corrosion of Steel Tendons Used in Prestressed Concrete Pressure Vessels:*¹⁴

The purpose of this investigation was (1) to determine the corrosion behavior of a high-strength steel (ASTM A416-74, grade 270), typical of those used as tensioning tendons in prestressed concrete pressure vessels, in several corrosive environments and (2) to demonstrate the protection afforded by coating the steel with either of two commercial petroleum-based greases or portland cement grout. In addition, the few reported incidents of prestressing steel failures in concrete pressure vessels used for containment of nuclear reactors are reviewed. The susceptibility of the steel to stress corrosion cracking and hydrogen embrittlement and its general corrosion rate were determined in several salt solutions. Wires coated with the greases and grout were soaked for long periods in the same solutions, and changes in their mechanical properties were subsequently determined. All three coatings appeared to give essentially complete protection, but small flaws in the grease coatings were detrimental; flaws or cracks less than 1 mm wide in the grout were without effect.

3.7.2 Potential New Materials for PCRV Application

Reactor containment structures have traditionally been massive, thick-walled, flat headed, essentially right circular cylinders fabricated from conventional reinforced-concrete material systems. Properties of the system, such as low tensile and shear strengths, decreasing strength, and modulus of elasticity with increasing temperature, creep, etc., have led to designs that have produced massive, costly containment structures. Recently several "concrete-type" material systems have been developed to the stage that they may be potentially cost and performance effective for fabrication of reactor containment

structures. These material systems include fiber-reinforced concrete, concrete with polymers, hydrothermal and refractory cements, high-alumina cements, and cements with partial cement replacement by pozzolans.

3.7.2.1 Overview of Candidate Material Systems

3.7.2.1.1 Fiber-Reinforced Concrete. Fiber-reinforced concrete consists of hydraulic cements containing fine, or fine and coarse, aggregates and discontinuous, discrete fibers. The fibers, which are generally steel (although glass, nylon, polypropylene and other materials have been used), are added to the mix in much the same manner as are other constituents and impart improved properties to the concrete. Mechanical and physical properties of fibrous concrete are functions of the fiber volume concentration, fiber aspect ratio, and type of fiber. Typical property improvements that can result from the inclusion of fibers into a concrete matrix include: 250% increase in flexure strength, slightly increased compressive strength; 250% increase in splitting-tensile strength; 175% increase in shear strength; 140% increase in direct tensile strength; an order-of-magnitude increase in toughness (energy input required to produce fracture); and an order-of-magnitude increase in resistance to dynamic loadings resulting from impacts or explosions. The fibrous concrete exhibits improved thermal shock resistance, and the fibers arrest and bridge cracks so that loads may be transmitted across flaws. Material costs for fibrous concrete are a function of type and content of fibers, but, for a mix with 1.5 vol % steel fibers, the material cost is approximately twice that of plain concrete. However, when the material cost is adjusted for improved performance and possibly reduced reinforcement requirements, fibrous concrete may be cost as well as performance effective.

3.7.2.1.2 Concrete with Polymers. Polymers in concrete combine hydraulic cement and polymer technologies to form new and improved construction material systems. Advantages of the material systems relative to the conventional concrete material system include: increased compressive strengths of 50 to 300%, tensile strength increase up to

200%, increased shear strength of 200 to 300%, flexure strength increase up to 200%, increased resistance to aggressive environments, relative impermeability, increased bonding to reinforcements or other materials, 200 to 300% increase in the modulus of elasticity, and rapid strength gain. Several types of concrete with polymers exist, but the polymer concretes and polymer-modified concretes possess the greatest potential for application to structures such as PCRVs.

Polymer concrete material systems consist of a polymeric matrix, such as a polyester or epoxy resin, to which may be added diluents, fine fillers, fine and coarse inorganic aggregates, and reinforcing fibers. By a judicious selection of the matrix and its modifiers, material systems may be developed that exhibit a wide range of material properties. The polymer concretes are batched, mixed, and placed by using essentially the same techniques that were used for the conventional portland cement concrete material systems. Polymerization of the system into a solid structural-grade material is generally by chemical action, heat, or radiation. Despite the advantages of these systems, a number of disadvantages also must be considered: material costs are from 3 to 10 times that of conventional concrete (results from the replacement of the cement by expensive polymeric materials), curing shrinkage is generally in excess of that for concrete, the strength and modulus of elasticity of room-temperature curing systems generally decreases rapidly as the temperature increases,* and exothermic heat buildup during curing may limit the mass of polymer concrete that may be cast at one placement.

Polymer-modified concretes are conventional concrete mixes to which monomers or polymers such as latex or epoxy resins are added during the mix cycle. The effects of the polymer addition are (1) the material system ductility and toughness are increased; (2) compressive strength and modulus of elasticity may be increased or decreased, depending on selection and amount of polymer; and (3) bonding to reinforcement or other materials may possibly be increased. To date, the introduction of polymers in conventional concrete mixes has been only

* Proper selection of curing agents in addition to elevated-temperature curing produces material systems that can retain their room-temperature strengths at temperatures greater than 149°C.

moderately successful, but the polymer-modified concretes may be cast in place, which makes them potentially advantageous for structural applications.

3.7.2.1.3 Hydrothermal and Refractory Cements. Hydrothermal (nonportland) cements have recently been developed for the lining of oil wells, but they are also potentially suitable for other applications in which heat may be deleterious to normal concrete materials. The materials are basically polymer silicates whose cure initiates at an activation temperature that is dependent on material formulation. After curing, the cements are capable of withstanding service temperatures of up to 538°C (up to 1093°C in certain formulations) without alteration of physical or mechanical properties. In addition, the material system can be formulated to obtain: compressive strengths of 68.9 to 137.9 MPa, excellent adhesion to metals except for aluminum, good resistance to aggressive environments, low permeability, and material system costs comparable to those of special portland cements such as Type III high early strength cement. Large placement of the material system is apparently not the problem that it is for polymer-concrete materials because the exothermic heat buildup is very low. Available property data indicates that the hydrothermal cement systems are quite promising as structural materials. However, the data available from the material producer is limited, and more detailed data need to be developed by an independent investigator before use of the hydrothermal cement can be recommended.

Fast-setting refractory cements also possess potential as structural materials for fabrication of PCRVs. The material systems cure at room temperature, set in 4 h, develop up to 41.4 MPa compressive strengths in 24 h, develop excellent bond to either metals or other minerals, resist heat cycling and continuous exposure to 1093°C, have low shrinkage and exotherm, require low water contents, and are fabricated from readily available materials, which makes the material costs relatively low. Limitations are basically the same as for the hydrothermal cements in that only limited data from one source are currently available.

3.7.2.1.4 High-Alumina Cements. Aluminous, or high-alumina, cement is a hydraulic cement used to make concrete in the same manner as normal portland cement. The high-alumina cements exhibit rapid strength gains (up to 96.5 MPa in 24 h and 124.1 MPa in 28 d), are resistant to aggressive environments, may be used as refractory materials at temperatures up to 1816°C when used with special aggregate materials and desiccated, and exhibit creep similar to that of normal concretes loaded to the same stress/strength ratio. The high-alumina cements, however, cost several times more than do normal portland cements; must be protected against water loss during curing; lose strength on exposure to a hot, moist environment unless a rich mix has been used; are generally not compatible with many additives; develop heat on curing approximately 2.5 times that of normal cements, which may produce cracking and strength reductions in thick sections; may lose workability rapidly after mixing; and can contribute to accelerated steel corrosion. We recommend that only rich, strong, high-alumina cement mixes be used for structural applications; thin sections are preferred.

3.7.2.1.5 Cement with Pozzolans. Pozzolans are siliceous materials that react with lime in the presence of moisture to produce a cementitious calcium-silicate compound. The pozzolans occur naturally in forms such as opaline shales and cherts, tuffs, and volcanic ashes or pumicites; or they may be produced artificially as a by-product such as fly ash, which results as a combustion waste product of powdered coal used in the generation of steam. Because they are generally more economical than cements, the pozzolans may be used to replace from 10 to 40 wt % of the cement in concrete. Advantages of pozzolan use include improved workability, lower heat of hydration and thermal shrinkage, decreased permeability, increased resistance to sulphates, reduced adverse alkali reactivity, improved economy, utilization of waste products (fly ash), and reduced bleeding and segregation. Potential disadvantages are rate of strength gain may be reduced (ultimate strength, however, may equal, or be superior to, conventional portland cement), increased drying shrinkage and reduced strength may occur in concrete exposed to unusual drying conditions, and increased water requirements. Because adverse effects on quality of concrete may be

induced if inferior pozzolans or excessive pozzolan contents are used, the pozzolan should be tested in conjunction with the cement and aggregate to assess its suitability.

3.7.2.2 Material System Recommendations

Fiber-reinforced concretes, concretes with polymers, hydrothermal and refractory cements, high-alumina cements, and concretes with partial replacement of the cements by pozzolans have been considered as potential materials for fabrication of PCRVs. Of these material systems, the fiber-reinforced concretes, the hydrothermal and refractory cements (with or without fiber reinforcement), and concretes with pozzolans appear to possess the greatest potential for being cost and performance effective. These systems have been selected because of their favorable costs; improved material properties; and ability to be batched, mixed, and placed by use of conventional concrete techniques. Also, the use of pozzolans is even more attractive because cements are becoming short in supply as a result of the high cost of building new plants and because of the closing of old plants as a result of pollution restrictions.

Polymer concrete materials were found to be generally unsatisfactory because of their high costs, poor hot strengths of room-temperature-cured materials, and the requirement of elevated-temperature curing to produce material systems with favorable hot strengths. The polymer-modified concretes do not produce sufficiently significant improvements in material properties to merit their increased costs. High-alumina cements are relatively expensive, should not be cast in large quantities (which limits their applicability to PCRVs), and may accelerate steel corrosion; but the high-alumina cements may find application as thin thermal barriers.

3.7.3 Fibrous Concrete Study

Based on the previous section, fibrous concrete was selected for additional investigation to evaluate its applicability to PCRVs for effecting reduced construction costs. Although the investigation is

only in the conceptual phase, we envision that the evaluation will include (1) identification of a suitable fibrous concrete mix, (2) development of fibrous concrete material properties, (3) selection of a structural model for evaluation, and (4) conduction of an analytical evaluation of the model to determine improved performance provided by the use of the fibrous concrete material system. Once improved performance is demonstrated through analytical studies, model tests will be conducted to verify results, and other potential areas in which fibrous concrete may be utilized to effect reduced PCRV costs will be identified and evaluated.

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4. STRUCTURAL MATERIALS

P. L. Rittenhouse

4.1 INTRODUCTION

The ORNL program on Gas-Cooled Reactor (GCR) structural materials provides assistance in the development of materials, materials technology, and materials data needed for the design, licensing, construction, and operation of advanced GCR systems. The program is interfaced with, and complements, the GCR materials efforts at General Atomic Company (GA) and General Electric Company (GE) and, together with these efforts, forms the national structural materials program in support of GCR technology.

The four main tasks covered in the ORNL program are

- mechanical properties of GCR alloys, weldments, and ceramics,
- corrosion of GCR alloys,
- joining technology, and
- structural ceramics.

The mechanical properties task involves the determination of tensile, creep, fatigue, creep-fatigue, and crack-growth properties of selected candidate alloys and their weldments as well as various mechanical tests on structural ceramic materials. Many of the tests are conducted in a simulated GCR helium environment to determine any effects of reaction between the materials and oxidizing and carburizing components of the environment. Thermal stability and its effect on properties are evaluated by aging and postexposure testing.

Work on corrosion is divided into two distinct subtasks: steam corrosion and oxidation/carburization behavior in GCR helium. The steam corrosion work is relevant to materials for steam generators (electric and process heat plants) and recuperators (direct-cycle gas turbine system). Determination of the kinetics of carburization of selected alloys is being emphasized at present in the oxidation/carburization subtask. The effect of carbon concentration gradients on dimensional stability is also being studied.

In the joining technology task, materials with potential application for advanced GCRs are being studied to determine their suitability for joining by welding. Various welding processes and usually more than one filler metal are employed for each alloy. The weldments produced are evaluated for hot cracking susceptibility, general weldability, and mechanical properties.

In our newest area of work, structural ceramics, both fibrous insulations and dense ceramics are being characterized, and tests of their compatibility/stability in the GCR helium environment are being initiated. Studies to determine the effects of rapid depressurization on integrity are also being planned.

During the past year the emphasis of our studies was shifted from materials for steam-cycle application toward materials for higher temperature, advanced GCR systems. For example, all of our work on the mechanical properties of ferritic steels was phased out, and the facilities used for this work were dedicated to testing advanced alloys and structural ceramics. These shifts in program direction did not significantly perturb our work because most of the ongoing efforts were, and continue to be, generic to the technology.

4.2 MECHANICAL PROPERTIES OF GCR ALLOYS, WELDMENTS, AND CERAMICS — C. R. Brinkman

4.2.1 Creep and Tensile Testing — H. E. McCoy

The materials being evaluated in this program are described in Tables 4.1 and 4.2. Tensile tests have been run on most of these materials and the results have been reported.¹⁻⁴ Comparative tensile properties of several alloys are shown in Fig. 4.1. The yield and tensile strength of Inconel 617, Hastelloy X, and Incoloy 802 fall into a rather narrow band, but HD 556 is significantly stronger over the entire test temperature range.

All-weld-metal specimens of Hastelloy X and Inconel 617 were tensile tested and the properties compared with those of the base metal. The welds were prepared by the gas tungsten-arc (GTA) process, and the filler metals used had the same nominal compositions as the base metals.

Table 4.1. Alloy Characterizations

Material	Heat	Source	Product form	Grain size (μm)	Heat treatment
Alloy 617	XX01A3US	INCO	12.7-mm (1/2-in.) plate		Solution annealed by vendor
Hastelloy X	2600-3-4936	Cabot	12.7-mm (1/2-in.) plate	80	Solution annealed by vendor
	2600-3-2792	Cabot	31.8-mm (1 1/4-in.) bar	55	Solution annealed by vendor
Alloy 800H	HH6993	INCO	19-mm (3/4-in.) bar		
2 1/4 Cr-1 Mo	X-6216	B&W	51-mm-OD (2.0-in.) tube	27	Isothermally annealed ^a
	36202	B&W	51-mm-OD (2.0-in.) tube	20-27	Isothermally annealed ^a
	72768	B&W	51-mm-OD (2.0-in.) tube	27	Isothermally annealed ^a
9 Cr-1 Mo	316381				
Hastelloy S	8635-3-8365T	Cabot	12.7-mm (1/2-in.) plate		
HD 556	8556-5-7305	Cabot	6.4-mm (1/4-in.) plate		Unknown
Inconel 618	Y09B9	INCO	12.7-mm (1/2-in.) plate		Unknown
Incoloy 802	HH6863B	INCO	7.9-mm (0.312-in.) plate		Solution annealed by vendor

^a927 $\pm$ 14°C for 30 min, cooled to 704 $\pm$ 14°C at a maximum rate of 83°C/h, held at 704 $\pm$ 14°C for 2 h, cooled to room temperature at a maximum rate of 6°C/min.

Table 4.2. Alloy Compositions

Material	Content (%)													
	Ni	Cr	Co	Mo	Fe	Al	Si	C	Mn	S	W	Ti	Cu	Others
Alloy 617	57.35	20.30	11.72	8.58	1.01	0.76	0.16	0.07	0.05	0.004				
Hastelloy X 4936	Bal.	21.82	1.68	9.42	19.09		0.44	0.07	0.58	<0.005	0.63			
2792	Bal.	21.25	1.94	8.99	18.96		0.41	0.10	0.57	<0.005	0.56			
Alloy 800H 6993 ^a	33.43	20.36			43.60	0.43	0.39	0.07	0.77	0.004		0.46	0.49	
2 1/4 Cr-1 Mo X-6216		2.19		1.00	Bal.		0.26	0.12	0.44	0.016				
36202		2.24		0.96	Bal.		0.34	0.12	0.43	0.023				
72768		2.16		0.94	Bal.		0.38	0.10	0.41	0.012				
9 Cr-1 Mo	0.4	8.83		1.0	Bal.		0.6	0.0803	0.5	0.0208				0.02 V
Hastelloy S 8365T	Bal.	15.3	0.15	15.3	0.5	0.40	0.40	0.0129	0.5			0.01	0.04	0.02 La
HD 556	19.44	21.39	19.08	2.72	Bal.	0.26	0.49	0.11	1.20	<0.005	2.08			0.01 La, 0.76 Cb + Ta
Inconel 618	56.49	22.5			14.79	0.01	0.021	0.05	0.09	0.005	5.85		0.04	
Incoloy 802	33.45	19.74			44.57		0.37	0.30	1.11	0.001			0.46	

^aChemical composition not available for this heat; analysis shown is for HH13-6979.

The yield and tensile strengths of the all-weld-metal samples were higher than those of the base metal. The yield and tensile strengths of the Hastelloy X weld metal were 50 and 10%, respectively, higher than those of the base metal, and the yield and tensile strengths of Inconel 617 weld metal were 75 and 10%, respectively, higher than those of the base metal. The fracture strains of all-weld-metal specimens were in the range of 20 to 40%.

Tensile tests were run on all-weld-metal samples of Incoweld A, a filler metal for making welds in nickel- or iron-base alloys by the shielded metal arc weld (SMAW) process. Incoweld A weld deposits were stronger than those for Incoloy 800H at all test temperatures and compared favorably with Hastelloy X in short-term tensile properties.

Fig. 4.1. Tensile Properties of Several Alloys (Strain Rate = 0.05 min^{-1}).

The yield strength of Incoweld A was 20% higher than that of Hastelloy X, and the ultimate tensile strength of Incoweld A was 20% lower than that of Hastelloy X. The fracture strains were in the range of 40 to 60%.

Inconel 617 was investigated as a filler metal for Incoloy 802. A sound test weld was made by the GTA process, and test specimens were made for evaluation (Sect. 4.4.1). In tensile tests at 25 to 871°C , failure consistently occurred in the Incoloy 802 base metal. Thus, Inconel 617 makes a suitable filler alloy for Incoloy 802. Alloy HD 556 was joined with HD 556 filler metal. The transverse weld specimens had yield strengths about 10% higher than the base material and ultimate tensile strengths about 20% lower than the base material. Failure occurred in the weld metal, and the fracture strains were about one-half those of the base metal.

All the materials described in Tables 4.1 and 4.2 have been creep tested in metal chambers. The test environment was premixed helium of the composition noted in Table 4.3. The testing procedures were described previously.^{4,5} Currently, 32 creep machines are operating

Table 4.3. Composition Limits of Test Gases^a

Impurity	Level noted (ppm)
H ₂	170-180
O ₂	10 ⁻²² atm ^b
N ₂	<1
CH ₄	15-18
H ₂ O	1-2 ^c
CO	9-12

^aTest chambers operate at 184 kPa (12 psig, 26.7 psia). Multiply ppm $\times$ 1.82 to obtain μ atm, by 0.184 to obtain Pa.

^bGas passed through furnace at 500°C to react O₂ with H₂. Estimate based on equilibrium.

^cConcentration range thought to be correct, but measuring techniques being reevaluated.

in a helium environment and 16 machines are operating in air. The materials described in Tables 4.1 and 4.2 are evaluated in the creep program, and some results have been reported.⁴⁻⁶

Testing has been in progress for several years on 2 1/4 Cr-1 Mo steel and Hastelloy X, and some tests have run over 25,000 h. The most important finding is that Hastelloy X is carburized by exposure to HTGR helium and that 2 1/4 Cr-1 Mo steel is decarburized at test temperatures of 538°C and higher. The decarburization of the steel leads to a decrease in creep strength but not to degradation of ductility. The carburization of Hastelloy X has not caused a detectable change in creep strength but does seem to reduce creep fracture strain. The most important effect is the reduction in low-temperature ductility because of the carburization.

Testing on a number of other materials has begun, and some results are shown in Fig. 4.2. These results help to scope the strengths of the materials relative to those of Hastelloy X. The stress-rupture properties of Inconel 618 and Hastelloy S are lower than those of Hastelloy X, whereas those of Incoloy 802, Inconel 617, and HD 556

Fig. 4.2. Stress-Rupture Properties of Several Alloys.

are higher. Incoloy 802 has fracture strains of 2 to 4%, but the other alloys have fracture strains greater than 10%.

Creep tests were run on a number of weldments. Incoloy 802 base metal joined with Inconel 617 filler metal was found to fail consistently in the Incoloy 802 so that creep strength was controlled by the Incoloy 802. Hastelloy X joined with Hastelloy X filler consistently failed in the weld metal, but the rupture time was within a factor of 2 of that for the base metal. Alloy HD 556 was joined with HD 556 filler metal, and the creep strength of the transverse weld is approximately equivalent to that of the base metal.

All-weld-metal specimens of Hastelloy X and Inconel 617 had rupture lives quite close to those of the base metal. Two samples of Inconel 617 weld metal tested at 649°C had fracture strains of only 2%, but all other specimens had acceptable ductility. Several all-weld-metal samples of Incoweld A had stress limits about 10% higher than those of Incoloy 800 H and 50% lower than those of Hastelloy X. The fracture strains on Incoweld A weld deposits were a minimum of 8%.

The carburization resistance of Ti-modified Hastelloy N was compared with that of Hastelloy X. The Ti-modified Hastelloy N has a nominal composition of Ni-12% Mo-7% Cr-2% Ti-0.05% C. Small coils of 0.28-mm-diam wire of Hastelloy X and Ti-modified Hastelloy N were exposed to HTGR helium for 1028 h at 871°C. The average carbon content of the Hastelloy X increased from 0.10 to 0.44%, and the carbon content of the Ti-modified Hastelloy N changed from 0.021 to 0.017%. Limited creep testing on Ti-modified Hastelloy N at 760°C has shown the creep strength to be slightly less than that of Hastelloy X.

4.2.2 Aging Studies — H. E. McCoy

Many of the alloys being considered change properties as a function of time at temperature, and the problem is likely worsened by carburization in HTGR helium. The programs in progress to study these effects are summarized in Table 4.4. The aged specimens are being evaluated by both tensile and creep tests, which are still in progress. Many of the samples are still in the furnaces undergoing aging.

Table 4.4. Summary of ORNL Aging Programs

Material	Aging conditions			Status
	Temp. (°C)	Time (h)	Environment	
Base metals				
Hastelloy X	538	2.5, 10, 20	Inert	Complete
Hastelloy X	704	2.5, 10, 20	Inert	Complete
Hastelloy X	871	2.5, 10, 20	Inert	Complete
Incoloy 802	593, 704, 871	10	HTGR He	5,000 h
Inconel 617	538	2.5, 10, 20	Inert	Complete
Inconel 617	704	2.5, 10, 20	Inert	Complete
Inconel 617	871	2.5, 10, 20	Inert	Complete
Hastelloy X	593, 704, 871	10	HTGR He	Complete
Hastelloy X	593, 704, 871	20, 50	HTGR He	15,000 h
Inconel 617	593, 704, 871	10	HTGR He	Complete
Inconel 617	593, 704, 871	20, 50	HTGR He	15,000 h
Welds ^a				
X/X/X, GTA	538, 704, 871	2	Inert	Complete
X/82/Incoloy 800 H, GTA	427, 538, 649	2	Inert	Complete
X/X/X, GTA	593, 649, 760,871	2, 10	Inert	Complete
X/X/X, SMAW	593, 649, 760,871	2, 10	Inert	Complete
X/S/X/, GTA	593, 649, 760,871	2, 10	Inert	Complete
X/182/800 H, GTA	482, 538, 649	2, 10	Inert	Complete
X/A/800 H, SMAW	482, 538, 649	2, 10	Inert	Complete
X/X/X, GTA	593, 704, 871	10	HTGR He	Complete
X/X/X, GTA	593, 704, 871	20, 50	HTGR He	15,000 h
617/617/617, GTA	593, 704, 871	10	HTGR He	Complete
617/617/617, GTA	593, 704, 871	20, 50	HTGR He	15,000 h
802/617/802, GTA	593, 704, 871	10	HTGR He	5,000 h

^aThe following designation is used for welds: Base metal 1/filler metal/base metal 2; GTA = gas tungsten-arc process; and SMAW = shielded metal arc weld.

The main property change of concern is the loss of low-temperature ductility. This effect is shown in Table 4.5 for Hastelloy X and Inconel 617 base metal. Samples of both alloys were aged for up to 20,000 h in an inert environment. The lowest ductilities, about 10%, were found for Hastelloy X after aging at 704°C. Generally, the ductilities were higher for Inconel 617 than for Hastelloy X, but very similar values of about 20% were obtained after aging for 20,000 h at 871°C. Similar specimens are being aged in HTGR helium, and the results of some of these tests are also shown in Table 4.5. The two sets of samples have different gage geometries, and this factor may influence the results slightly. However, the data in Table 4.5 indicate that aging at 704 and 871°C in HTGR helium resulted in lower fracture strains than aging in an inert environment for both alloys.

Table 4.5. Tensile Fracture Strains of Samples Tested at Room Temperature

Material	Aging conditions		Fracture elongation (%)	
	Time (h)	Temp. (°C)	Aged in inert gas ^a	Aged in HTGR He ^b
Hastelloy X	0	0	52	
	2,500	538	55	
	10,000	538	43	43 ^c
	20,000	538	47	
	2,500	704	9	
	10,000	704	18	11
	20,000	704	10	
	2,500	871	15	
	10,000	871	15	10
	20,000	871	18	
Inconel 617	0	0	58	
	2,500	538	69	
	10,000	538	53	60 ^c
	20,000	538	66	
	2,500	704	42	
	10,000	704	30	14
	20,000	704	21	
	2,500	871	32	
	10,000	871	28	11
	20,000	871	20	

^aTest section 6.35-mm diam by 31.8 mm (1/4-in. diam by 1.25 in.) long.

^bTest section 3.18-mm diam by 25.4 mm (1/8-in. diam by 1.00 in.) long.

^cAged at 593°C.

Impact specimens were made of the two alloys; the test results are given in Table 4.6. The lowest value, 5 J (4 ft-lb), was obtained for Hastelloy X after aging for 20,000 h at 704°C in an inert environment. The lowest value for Inconel 617 was 20 J (15 ft-lb) after aging 20,000 h at 704 and 871°C. Data are insufficient to draw a conclusion about whether or not carburization reduces the impact energy further.

Table 4.6. Impact Properties of Hastelloy X and Inconel 617

Treatment	Impact energy at 25°C			
	Hastelloy X		Inconel 617	
	J	(ft-lb)	J	(ft-lb)
As received	136,137	(100,101)	140,211	(103,155)
100 h at 871°C in inert atmosphere			97	(71.5)
100 h at 871°C in HTGR He	14	(10.5)	91	(67)
1,000 h at 871°C in HTGR He	15	(11)	53	(39)
10,000 h at 538°C in inert atmosphere	39	(29)	80.5	(59.5)
10,000 h at 704°C in inert atmosphere	15	(11)	31	(23)
10,000 h at 871°C in inert atmosphere	7.5	(5.5)	2.5	(18.5)
20,000 h at 538°C in inert atmosphere	40	(29.5)	103	(76)
20,000 h at 704°C in inert atmosphere	5	(4)	20	(15)
20,000 h at 871°C in inert atmosphere	11	(8)	20	(15)

Transverse weld specimens of Hastelloy X joined by three combinations of filler metal and welding process were aged 2000 and 10,000 h in inert gas at 593, 649, 760, and 871°C. Tensile tests have been completed on these specimens, and creep tests are in progress. Analysis of the data on specimens aged and tested at room temperature revealed that aging reduced the fracture strain drastically. The most severe embrittlement occurred after aging at 760°C. The fracture strain was lower for Hastelloy X filler metal deposited by the SMAW process than for Hastelloy X deposited by the GTA process. Hastelloy S filler deposited by the GTA process exhibited the least embrittlement. Aging of Hastelloy X weldments with Hastelloy X filler at 650°C resulted in the maximum strengthening (67% in yield strength). Aging at 593°C had very little

effect on strength, and aging at 871°C resulted in a decrease in strength of about 25%.

Transverse welds of Incoloy 800H and Hastelloy X joined with INCO 82 and Incoweld A were aged for 2000 and 10,000 h at 482, 538, and 649°C. The changes in measured ductility and strength were small. However, all fractures occurred in the Incoloy 800H base metal so that the measured properties do not reflect changes that might have occurred in the weld deposit.

The creep tests on the weld series described in the previous paragraphs are still in progress, but sufficient results are available to reflect trends. The minimum fracture strain of the Hastelloy X weldments prepared by the GTA process was about 6.5%, which was noted in the as-welded condition, and aging generally resulted in improved ductility and reduced rupture life. Similar trends were noted for the Hastelloy X weldment prepared by the SMAW process, but the minimum strain levels were about 3%. In the Hastelloy X weldment involving Hastelloy S filler metal, the minimum fracture strain was about 10% in the as-welded condition and improved with aging. In the two weldments with Hastelloy X filler metal, fracture consistently occurred in the weld deposit; with Hastelloy S filler metal, the fracture location varied with test conditions. The two weldments of Hastelloy X to Incoloy 800H consistently fractured in the Incoloy 800H at fracture strains of 3 to 5%. Aging had little effect on the fracture strain or the strength.

Hastelloy X and Inconel 617 weldments prepared by the GTA process were aged for 10,000 h in HTGR helium. The elongation at rupture in a room-temperature tensile test is shown in Table 4.7, along with values for Hastelloy X specimens aged in an inert environment for comparison. At the higher aging temperatures the fracture strain is slightly lower for samples aged in HTGR helium than for those aged in an inert environment.

Some creep tests have been started on the Hastelloy X and Inconel 617 transverse weld specimens aged in HTGR helium for 10,000 h. The changes caused by aging currently appear to be very similar to those noted for the Hastelloy X specimens aged in an inert environment (i.e.,

Table 4.7. Room-Temperature Tensile Fracture Strains for Transverse Welds Aged for 10,000 h

Alloy	Aging temp. (°C)	Elongation at rupture (%)	
		Inert environment	HTGR He
Hastelloy X	593	18.5	30.7
	650	6.1	
	704		4.1
	760	6.5	
	871	10.6	4.6
Inconel 617	593		26.3
	704		14.4
	871		0.7

aging increases the fracture strain and slightly decreases the rupture life).

4.2.3 Crack Propagation

4.2.3.1 Fatigue Crack Growth — C. R. Brinkman and J. P. Strizak

This task involves conducting fatigue crack propagation studies on a number of candidate structural alloys and their weldments. Materials being investigated include Hastelloy X, Inconel 617, and type 316 stainless steel. Testing parameters include temperature, environment, frequency, wave form, and metallurgical state. Materials (blanks or specimens) that have been thermally aged and are waiting to be tested are shown in Table 4.8. In addition to the above material, the following

Table 4.8. Thermally Aged Materials for Crack Propagation Testing

Alloy	538°C			704°C			871°C		
	2,500 h	10,000 h	20,000 h	2,500 h	10,000 h	20,000 h	2,500 h	10,000 h	20,000 h
Hastelloy X ^a	2 ^b	2 ^b	2 ^c	2 ^b	2 ^b	2 ^c	2 ^b	2 ^b	2 ^c
Inconel 617 ^d	2 ^b	2 ^b	2 ^c	2 ^b	2 ^b	2 ^c	2 ^b	2 ^b	2 ^c

^aHeat 2600-3-4936.

^bBlank material in postaged condition.

^cSpecimens already fabricated.

^dHeat XX01A3U5.

blanks or specimens are available for material in the solution annealed condition:

Hastelloy X — base material: 2 specimens; weld metal: 4 specimens

Inconel 617 — base material: 9 blanks

We plan to complete scoping tests in the near future for comparing the crack propagation behavior of aged (20,000 h) and unaged Hastelloy X and Inconel 617. In addition, a single test on a type 316 stainless steel specimen that has seen elevated-temperature service as a component for 50,000 h at elevated temperature is planned.

Installation of an additional environmental fatigue test facility was completed recently. The unit, shown in Figs. 4.3 and 4.4, permits

PHOTO Y161800

Fig. 4.3. Environmental Fatigue Crack Propagation Test Facility Capable of Either High Vacuum [1.33×10^{-7} Pa (10^{-9} torr)] or Controlled Impurity Helium Environments.

Fig. 4.4. Close-Up View of Environmental Fatigue Test Chamber.

conducting elevated-temperature crack propagation tests under high vacuum [1.33×10^{-7} Pa (10^{-9} torr)] or under controlled impurity helium conditions. The specimen under test will be heated by induction, and rates of crack propagation will be determined by optical and by crack opening displacement (COD) measurement techniques.

During the past year T. Weerasooriya completed three ORNL reports. These reports have been or are being published. Abstracts of these reports are as follows:

*Fatigue Crack Propagation in the Heat-Affected Zone of
2 1/4 Cr-1 Mo Steel and ERNiCr-3 Weldments - Interim Report,
ORNL/TM-6971 (October 1979).*

We examined the fatigue crack growth rate at 510°C in the heat-affected zone (HAZ) of a 2 1/4 Cr-1 Mo steel weldment as a function of frequency and stress intensity factor. The weldment was made of isothermally annealed 2 1/4 Cr-1 Mo steel base metal and ERNiCr-3 weld metal. Fatigue crack-growth rate data for thermally aged HAZ material lies between the two scatter bands of data for the base metal and ERNiCr-3 filler metal at 510°C. Prior thermal aging up to 2000 h at 566°C does not substantially change the fatigue crack-growth rate of HAZ material at 510°C.

Fatigue Crack Propagation in Hastelloy X Weld Metal,
ORNL/TM-6999 (November 1979).

The fatigue crack growth rate of Hastelloy X weld metal increased with stress intensity, temperature, and inverse frequency. The results were correlated with the equation

$$da/dN = A(\Delta K)^n,$$

for a constant frequency or a constant temperature. The values of A and n were computed with a linear regression algorithm. With decreasing frequency at a constant ΔK and constant temperature (538°C) fatigue crack-growth rates approach an upper limit. Fatigue crack-growth rate of the weld metal was lower than that reported for base metal at 538°C and lower at 649°C for a frequency of 1 Hz.

Crack Propagation in Hastelloy X, to be published.

The fatigue and creep crack-growth rates of Hastelloy X were examined in air and impure helium environments. Creep crack-growth rate in air is higher than is that in impure helium at 650°C. Initial creep crack-growth from the original sharp fatigue crack is by an intergranular mode of fracture. As the crack accelerates at higher ΔK levels, growth is by a mixed mode of both intergranular and transgranular fracture. Fatigue crack-growth rate increases with increasing temperature and decreasing frequency for the range of stress intensities reported in the literature. Fatigue crack-growth rate in impure helium is lower than is that in air.

4.2.3.2 Creep Crack Growth in Hastelloy X — R. W. Swindeman and B. C. Williams

Crack growth in Hastelloy X sheet specimens has been examined by Weerasooriya^{7,8} for both fatigue and creep conditions. A side edge notch geometry specimen was utilized for the creep crack-growth work at 650°C. Experimental data were examined to determine whether stress

intensity or mean stress across the uncracked ligament controlled the crack growth rate. No conclusion could be reached because the specimen geometry was severely distorted by large bending strains. Further, the measurement of creep crack growth was complicated by the nature of the secondary cracks that appeared in advance of the primary crack tip. Finally, it was difficult to get many measurements because of the relatively rapid crack growth and the need to remove the specimen from the furnace to make each measurement.

We decided to examine the usefulness of the center cracked panel specimen to study creep crack growth. Initial experiments will be performed in the lever arm creep machines used by Weerasooriya. We hope that the specimen distortion will be decreased by the more symmetric stress profile across the specimen width.

We also plan to install capability to control deflection rate rather than load, and a machine is being purchased that will accomplish this. The center cracked panel will be gradually stretched, and the COD will be increased at a constant rate. The resulting data will be interpreted by the method developed by Landis and Begley to determine C^* .⁹

4.2.4 Fatigue — J. P. Strizak and C. R. Brinkman

An investigation of the effect of a simulated HTGR helium environment on the fatigue behavior of 2 1/4 Cr-1 Mo steel was completed. The abstract from a report that is being published follows.

The Influence of Environment, Metallurgical Variables, and Prolonged Test Times on the Fatigue and Creep-Fatigue Behavior of Annealed 2 1/4 Cr-1 Mo Steel, C. R. Brinkman, J. P. Strizak, and M. K. Booker.

Results are reported from strain-controlled fatigue tests conducted in air and impure helium on annealed 2 1/4 Cr-1 Mo steel. Tests were conducted in the temperature range of 482 to 593°C, with and without hold periods at peak strain and near zero stress values. The intent of these tests was to determine the influence of variations in melting practice, postweld heat treatment (PWHT), environment, cyclic wave form, and thermal aging experience on fatigue life. It was concluded from this study that, at low strain ranges, while a tensile hold period introduced in each cycle was more damaging than an identical compression hold period in an impure helium environment, the reverse was true for an air environment. It was further concluded that hold periods introduced both in the

tensile and compression side of the hysteresis loop at peak strain value in both air and helium were a more damaging situation in terms of cycle life than either of the above cases. A single test conducted with a hold period on the tension-going side of the hysteresis loop near zero stress showed a marked reduction in fatigue life in comparison with the cyclic life of an identical specimen subjected to continuous cycling at the same temperature and strain range. It was demonstrated that much of the time-dependent fatigue behavior at temperature within the creep regime can be attributed to wave form-environment interaction. Considering the scatter in the data, metallurgical differences brought about by variations in melting practice, heat-to-heat variations, thermal aging, and PWHT for material in the annealed condition result in little difference in the magnitude of the reduction in cyclic lives induced by a hold period of given length and form.

Several methods for estimating long-term, time-dependent fatigue behavior were used to compare predictions versus actual cyclic lives of a number of specimens. When linear damage summation was employed, it was concluded that available uniaxial time-dependent fatigue data do not support use of a single D value such as 1.0 for making long-term extrapolations. Available data do show that the method of strain range partitioning is superior to that of linear damage summation for estimating strain-controlled fatigue behavior. However, it was shown that there is a need to conduct more complex and longer-term tests in order to more fully identify and clarify various aspects of material behavior as well as to identify any shortcomings in the ability of a given method to capture basic behavior. A number of these tests are identified and discussed in some detail.

A number of continuous cycling fatigue tests have been conducted on aged Hastelloy X. Interim data on material aged at 538 and 871°C for 10,000 h and tested at the respective aging temperature are given in Table 4.9. Comparison of the interim results on aged material with available data on solution-annealed material from the same heat reveals that thermal aging reduces cyclic life.

Scoping tests to compare the fatigue behavior of aged and unaged Hastelloy X and Inconel 617 will continue. The materials shown in Table 4.10 have been thermally aged, and specimens are waiting to be tested.

In addition to the above material, the specimens shown in Table 4.11 are available for material aged in HTGR helium.

Table 4.9. Comparison of Interim Results of Continuous Cycling Fatigue Tests on Aged Hastelloy X with Data on Solution-Annealed Material from Heat 2600-3-4936

Specimen No.	Aged 10,000 h		Solution annealed	
	Total strain range (%)	Cycles to failure	Total strain range (%)	Cycles to failure
	Aged and tested at 538°C		Tested at 538°C	
X1203	3.59	116	4.06	177
X1209	1.96	753	3.02	418
X1206	1.18	1,632	1.56	1,181
X1207	0.84	4,150	0.99	5,182
X1208	0.77	5,226	0.79	7,596
X1210	0.50	11,713	0.71	14,240
X1204	0.5	9,298	0.65	13,675
			0.60	225,000
	Aged and tested at 871°C		Tested at 871°C	
X3207	2.38	170	4.00	96
X3203	1.78	255	3.01	138
X3292	1.19	624	2.01	312
X3205	0.71	1,950	1.50	466
X3201	0.59	2,991	1.00	1,156
X3206	0.42	11,570	0.80	2,002
			0.59	3,433
			0.5	13,234

Table 4.10. Aged Specimens for Fatigue Testing^a

Alloy	538°C			704°C			871°C		
	2,500 h	10,000 h	20,000 h	2,500 h	10,000 h	20,000 h	2,500 h	10,000 h	20,000 h
Hastelloy X ^b	10	10	10	10	10	10	10	10	10
Inconel 617 ^c	10	10	10	10	10	10	10	10	10

^a Blanks for specimen fabrication were encapsulated in stainless steel containers having an argon environment.

^b Heat 2600-3-4936.

^c Heat XX01A3U5.

Table 4.11. Fatigue Test Specimens Aged in HTGR Helium^a

Alloy	Aging temperature (°C)	
	593	871
Hastelloy X		
Base metal	2	2
Weld metal	2	2
Inconel 617		
Base metal	2	2
Weld metal	2	2

^a Aging period was 10,000 h.

4.2.5 Testing of Ceramics — J. E. Selle and H. E. McCoy

4.2.5.1 Creep and Stress Relaxation Correlation — J. E. Selle

An assessment was made of published work on creep and stress relaxation phenomena in structural ceramics of interest to the HTGR program. These materials include fused silica, alumina, silicon nitride, and silicon carbide. The ability of the core support structure to function for the lifetime of the reactor is directly related to the allowable creep strain and to the ability of this component to withstand thermal transients. The thermal-mechanical response of the core support pads to steady-state stress and potential thermal transients is dependent, in part, on the ability of the ceramic to undergo some stress relaxation in relatively short times.

Of the materials listed above, alumina has been investigated in creep more than have the others. Activation energies reported vary between 482 and 837 kJ/mol (115 to 200 kcal/mol). Consequently, variations in the assigned mechanisms were noted. Nabarro-Herring creep has been designated by most investigators as the primary creep mechanism, although no definite grain size dependence has been identified. Reported activation energies for silicon nitride (586 to 649 kJ/mol) are in reasonable agreement with the Nabarro-Herring creep mechanism. No creep rate data were found for fused silica or silicon carbide. No stress relaxation data were found for any of the candidate materials.

Although creep and stress relaxation are similar and it is theoretically possible to derive the value of one property when the other is known, no explicit demonstrated relationship exists between the two. We therefore conclude that it is necessary to determine experimentally both properties to obtain information necessary for use in high-temperature design and safety analyses.

4.2.5.2 Mechanical Testing of Ceramics — H. E. McCoy

Test equipment was installed to run compressive creep tests on ceramic materials in HTGR helium. The test chamber consists of an alumina tube with cooled aluminum ends. All the internal parts are constructed of Mo-TZM. Four chambers of this type have been installed and are operational.

The first material to be tested is SiO_2 , type Corning 7941. The test specimens, 11.1 mm diam (7/16 in.) by 31.25 mm (1-1/4 in.) long, were provided by GA. The initial tests will be run at 816, 871, 927, and 982°C at a stress level of 13.8 MPa (2000 psi).

Two test chambers have been partially assembled for running four-point bend creep tests on small ceramic beams in HTGR helium. These chambers utilize a ceramic tube and Mo-TZM internal parts.

Equipment for running stress-relaxation tests on ceramics has been designed. Some of the parts have been procured, and the remaining parts will be fabricated when funds become available.

4.3 CORROSION OF GCR ALLOYS — J. H. DeVan

4.3.1 Oxidation/Carburization in GCR Helium — H. Inouye

The objective of this task is to provide a basic understanding of the corrosion behavior of structural alloys in GCR helium. Available published data are initially reviewed, and specific experiments are conducted to provide additional data deemed necessary to characterize adequately the corrosion resistance of candidate alloys. During 1979 one phase of the work included several runs to define better the carburization kinetics of Hastelloy X and Incoloy 800H, particularly with respect to the effects of exposure time and temperature on the carburization rate and depth of carbon penetration. A second phase of the work concerned providing an explanation for the creep rates of certain alloys that were higher in GCR helium than in air. Experiments conducted on this phase of the work were based on the assumption that the measured creep strains of alloys in GCR helium consisted of an alloy dilation (dimensional instabilities associated with carburization) component superimposed on that produced by the applied creep stress.

As described in detail in the 1978 annual report,¹⁰ the test gas composition was controlled by the addition of H_2 , CH_4 , and CO to pure helium assaying 99.998% He and circulated with a metal bellows pump in a stainless steel loop at a flow rate of 1 liter/min. The pumped test gas was preheated in the annulus of a double-walled porcelain retort and reacted with 6.35-mm-diam by 22.2-cm-long specimens supported on

two 3.2-mm-diam nickel rods in the inner tube. A five-zone furnace (91 cm long) was used to obtain a temperature gradient between 750 and 1000°C along about 46 cm of the retort. The specimens were located at those points where a traveling Pt/Pt-10% Rh thermocouple indicated temperatures of 750, 800, 850, 900, 950, and 1000°C. Based on the retort dimensions, the velocity of the test gas over the specimens was 23 mm/s, and its residence time above 750°C was 40 s (including preheat time). To expand on prior kinetic data and to correlate the creep results of the Central Institute for Industrial Research, Oslo, Norway (CIIR) and GA with our dilation experiments, all experiments were conducted in phase IV helium [500 H₂/50 CH₄/33 CO (μatm)].

4.3.1.1 Carburization Kinetics

Carbon increases for Hastelloy X and Incoloy 800H resulting from exposures to 4000 h are plotted in Figs. 4.5 and 4.6, respectively. The data points for both alloys suggest that carburization initially follows a parabolic rate law but then becomes accelerated. This result is a characteristic not reported previously. This "breakaway" carburization requires an incubation period that lengthens as the exposure temperature decreases. Because both alloys show this behavior, the transition is not significantly influenced by the alloy composition.

Fig. 4.5. Carburization of Hastelloy X as a Function of Exposure Time to Phase IV Helium.

Fig. 4.6. Carburization of Incoloy 800H as a Function of Exposure Time to Phase IV Helium.

In contrast, Lupton's¹¹ analysis of Hastelloy X data from several sources suggests that carburization can be described by a long transient period followed by "parabolic-linear" kinetics.

Figure 4.7 shows plots of the carbon increase versus T^{-1} for Hastelloy X after 1200-* and 4000-h[†] exposures. The curves for both exposure times appear to consist of two straight lines intersecting at about 850°C. The inflections in these plots are interpreted as a change in the character or in the type of reaction products on or near the alloy surface that probably occurred when the carbon increase versus time plots sharply increased (Figs. 4.5 and 4.6). The decrease in the H₂O level*[†] from an average of 0.3 to <0.1 μatm may account for the large differences in the slopes of the two curves. The ratio of $\Delta C/\Delta W$ (Table 4.12) for Hastelloy X remains constant for the 4000-h exposure between 750 and 1000°C, indicating that the inflection in the ΔC versus T^{-1} plot is caused by a morphology change in the reaction

*Time-compensated average gas composition is 504 H₂/49 CH₄/38 CO/0.036 CO₂/0.3 H₂O (μatm).

†Time-compensated average gas consumption is 502 H₂/50 CH₄/36 CO/0.042 CO₂/ <0.1 H₂O (μatm).

Fig. 4.7. Carburization of Hastelloy X as a Function of Exposure Temperature to Phase IV Helium.

Table 4.12. Comparison of 4000-h Carburization of Hastelloy X and Incoloy 800H with Total Weight Gain in Phase IV Helium^a

Test temp. (°C)	Weight increase, ΔW (μg/cm ²)	Carbon increase, ΔC (μg/cm ²)	ΔC/ΔW × 100 (%)
<u>Hastelloy X</u>			
750	600	420	70
800	710	560	79
850	850	730	86
900	2,700	2,100	78
950	8,300	6,300	76
1,000	19,000	14,800	78
<u>Incoloy 800H</u>			
750	590	330	56
800	780	220	28
850	1,000	160	16
900	2,000	800	40
950	5,200	3,700	71
1,000	11,000	15,000	136

^a502 H₂/50 CH₄/36 CO/0.042 CO₂/0.1 H₂O (μatm).

product (e.g., surface cracking). In contrast, $\Delta C/\Delta W$ for Incoloy 800H goes through a minimum at 850°C, possibly caused by a change in the type of reaction products at the surface (e.g., oxide + carbide to principally carbide). Like Hastelloy X, an inflection in the ΔC versus T^{-1} plot near 850°C is also observed for Incoloy 800H.

The ΔC values in Table 4.12 correspond to carbon increases in excess of 1% at 1000°C. Dean¹² also reported carburization levels between 1 and 2% in phase IV helium for several alloys tested in the 1975 to 1977 period, but his later tests (1977-1978) on the same alloys showed carburization levels of about 0.1 to 0.2%. The cause(s) of the large difference in carburization observed between his two test periods was not determined, but our results support his suggestion that carburization in phase IV helium is sensitive to small differences in the test conditions. Moreover, because both Hastelloy X and Incoloy 800H show the same high carburization levels, carburization appears to be only moderately dependent on the alloy composition. Two factors may account for Dean's observed differences in carburization. First, his initial results were duplicated by our tests; thus, the H_2O level in those tests may have been about 0.2 μatm but were possibly higher in his later tests. Second, the apparently stronger carburization tendency of our test gas could be caused by a lower level of CH_4 pyrolysis as a consequence of the retort design. The rate constant for the thermal decomposition of CH_4 in a clean system is given by $\dot{r} = (-103 \text{ kcal}/RT)/s$ and increases further when carbon nuclei deposit on hot surfaces.^{13,14} At 1000°C the above expression gives a pyrolysis rate of 0.14%/s and values of 0.026 and 0.004%/s at 950 and 900°C, respectively. Based on the temperature gradient along the retort, its internal dimensions, and the flow rate,* the residence time of CH_4 at each of the above temperatures is estimated to be 8 s, or about 1.4% of the CH_4 is pyrolyzed as it passes through the retort. The residence time of CH_4 at a flow rate

* Retort length at 1000, 950, or 900°C around 9.2 cm each, cross-sectional area of annulus and specimen tube 4.2 and 7.2 cm^2 , respectively. Helium flow rate 17 cm^3/s .

of 1 liter/min in Dean's retort is estimated to be about 173 s (assumes CIIR retort dimensions) or a pyrolysis rate of about 30% as the test gas passes through the retort. Methane depletion, through its reaction with the test specimens and their supports, was not included in the above calculations.

4.3.1.2 Carburization Depth

Plots of the carbon increase ΔC versus distance from the surface X were constructed by calculating the difference in the carbon content of specimens machined to successfully smaller diameters. Their extrapolation to $X = 0$ provided an estimate of the surface concentration C_S , listed in Table 4.13. For exposure of 1200 h, C_S goes through a maximum at 850°C, reaching concentrations between 3000 and 4000 ppm, before decreasing at the higher temperatures. This result suggests that carburization is controlled by surface reactions, not by the diffusion rate of carbon. This interpretation is further supported by the observation that C_S is not constant but increases with the exposure time.

Table 4.13. Effect of Phase IV Helium^a on Surface Carbon Concentration of Hastelloy X and Incoloy 800H

Exposure temp. (°C)	Surface carbon concentration, C_S^b (ppm)	
	1200 h	4000 h
<u>Hastelloy X</u>		
750	1,800	4,940
800	2,100	9,600
850	4,300	5,000
900	3,000	6,800
950	2,400	50,400
1,000	No test	0
<u>Incoloy 800H</u>		
750	2,400	15,700
800	2,900	7,500
850	3,300	700
900	2,000	4,900
950	1,200	132,000
1,000	1,100	268,000

^a500 H₂/50 CH₄/36-38 CO/0.04 CO₂/ $<0.1-0.3$ H₂O (μ atm).

^bValues extrapolated.

The exceptionally high C_S values of 13.2 and 26.8% for Incoloy 800H after 4000 h at 950 and 1000°C indicate that free carbon may have deposited on the surface.

The carburization depth, arbitrarily taken where X has a ΔC value of 200 ppm, listed in Table 4.14 shows the expected effects of temperature and time. Carburization depth in excess of the radius of the specimens is obtained in 4000 h at 1000°C.

Table 4.14. Carburization Depth of Hastelloy X and Incoloy 800H Exposed to Phase IV Helium^a

Exposure temp. (°C)	Carburization depth, X^b (mm)	
	1200 h	4000 h
<u>Hastelloy X</u>		
750	0.30	0.25
800	0.58	0.34
850	0.35	0.50
900	0.93	1.42
950	1.20	>3.17
1000	No test	>3.17
<u>Incoloy 800H</u>		
750	0.24	0.27
800	0.38	0.75
850	0.64	1.25
900	0.92	1.80
950	1.26	2.45
1000	1.67	3.45 ^c

^a502 H₂/50 CH₄/36 CO/0.042 CO₂/0.1 H₂O (μatm).

^bDepth below surface where $\Delta C = 200$ ppm.

^cEstimate. Specimen radius is 3.17 mm.

Long-term and/or high-temperature exposure produced unusual carbon profiles such as shown in Figs. 4.8 and 4.9. Between exposure times of 1200 and 4000 h at 950°C, calculations indicate that a large subsurface carbon peak formed in Hastelloy X. At 1000°C the data indicate surfaces with virtually no carbon in addition to still higher subsurface carbon peaks. Evaporation or spalling of surface reaction products could explain C_S values near zero at 1000°C, and countercurrent diffusion of carbide formers toward the surface could explain the subsurface carbon peaks. Data for Incoloy 800H show similar features.

Fig. 4.8. Carbon Gradient Resulting from the Exposure of Hastelloy X to Phase IV Helium at 950°C.

4.3.1.3 Carburization-Induced Dilation

The basis for this phase of study is Table 4.15, compiled from data reported by GA,¹⁵ which shows that the creep rate of several alloys is greater in GCR helium than in air. To account for this creep behavior, it is postulated that the carbon gradients will generate an internal stress whose magnitude is proportional to the carbon gradient dc/dx . The internal stress, when superimposed on a creep stress, will accelerate the creep rate by the factor $(\sigma_\alpha + \sigma_i)n/\sigma_\alpha$, where σ_α is the creep stress, σ_i is the internal stress, and n is the stress exponent. The environmental effect will therefore have the greatest effect when σ_α is low

Fig. 4.9. Carbon Resulting from the Exposure of Hastelloy X to Phase IV Helium at 1000°C.

Table 4.15. Correlation Between Carburization and Creep Acceleration in Phase IV Helium^a

Alloy	Carbon increase (%)	<u>Creep strain in He</u> <u>creep strain in air</u> (%)
IN-519	0.20	13
M-21	0.17	2.4
Hastelloy X	0.06	4
IN-713	0.07	1.5
IN-100	0.01	1.3

^a900°C.

Source: Compiled from S. N. Rosenwasser and W. R. Johnson, "Gas Turbine and Advanced HTGR Materials Screening Test Program," *Semiannual Progress Report for Period Ending March 31, 1977*, GA-144077, General Atomic Company, San Diego, Calif. (July 1977).

and dc/dx is a maximum and, conversely, will be masked at high values of σ_α and low gradients. Chemical (carbon) gradient creep is thus a distinct possibility and is supported by the data listed in Table 4.16. Both Hastelloy X and Incoloy 800H dilate (i.e., creep as a result of dc/dx -induced σ_z) and reach a maximum value of about 0.8% strain in 2000 h at the highest temperature. The data further show that dilation is not strictly proportional to ΔC .

Table 4.16. Dimensional Stability of Alloys Exposed to Phase IV Helium

Exposure temp. (°C)	Hastelloy X		Incoloy 800H	
	Length, Δ^a (%)	Carbon, Δ ($\mu\text{g}/\text{cm}^2$)	Length, Δ^a (%)	Carbon, Δ ($\mu\text{g}/\text{cm}^2$)
2000 h				
850	0.35	340	0.24	100
900	0.54	430	0.29	400
1,000	0.93	4,500	0.74	4,600
4000 h				
750	-0.07	420	0.02	520
800	0	560	0.28	230
850	0.16	740	0.04	160
900	0.23	2,200	-0.06	800
950	0.47	6,800	0.12	3,600
1,000	0.60	16,000	0.48	16,500

^a6.35-mm-diam by 22.2-mm-long bar specimens machined from 12.7-mm-thick plate previously annealed at 1177°C for 15 min and water quenched.

4.3.2 Steam Corrosion — J. C. Griess, J. H. DeVan, and W. A. Maxwell

This task is supported jointly by the GCR and the Liquid-Metal Fast-Breeder Reactor (LMFBR) programs. Its purpose is to determine the corrosion behavior of selected materials in superheated steam under both isothermal and heat-transfer conditions. All testing is performed by the NUS Corporation, under subcontract to ONRL, at the Bartow Plant of the Florida Power Corporation in St. Petersburg, Florida. The steam for these tests comes directly from the superheater circuit of that plant. The isothermal tests are conducted in a nonrecirculating loop

made of Inconel 617, and the heat-transfer tests are conducted in a type 316 stainless steel loop, which also uses once-through steam.

4.3.2.1 Inconel 617 Loop

This loop contains two chambers for exposure of test specimens; one is maintained at 482°C and the other at 538°C. The steam pressure is 10.5 MPa. The loop has been in operation for 28,000 h, and a report describing the results for the first 25,000 h has been prepared.¹⁶ An abstract of that report follows.

We are studying the corrosion of several ferritic and austenitic materials in flowing superheated steam at 482 and 538°C (900 and 1000°F). This report summarizes the results obtained during the first 25,000 h. The test specimens are mounted in a nonrecirculating loop that receives steam from the superheater circuit of a fossil-fired power plant.

At both temperatures all materials exhibited parabolic oxidation kinetics during the first year and subsequently have oxidized at low constant rates. The ferritic steels containing 2 1/4 and 9% Cr plus 1% Mo have oxidized at about the same rates, averaging 4.2 and 8.6 µm/year (0.17 and 0.34 mils/year) at 482 and 538°C, respectively, after the first year. Sandvik HT-9 (11.4% Cr) has corroded at slightly lower rates. Annealed and cold-worked surfaces of these alloys have exhibited identical behavior. At 482°C all materials have retained their corrosion products completely, but at 538°C some began experiencing exfoliation after 12,000 h. The data suggest that a high silicon content in the alloy minimizes exfoliation.

Cold-worked surfaces of alloy 800 are corroding at lower rates than annealed and pickled ones, but in both cases the rates are very low. Alloy 800 specimens that had been intergranularly corroded before exposure to steam are oxidizing at much higher rates, but intergranular penetration has not progressed. Type 304 stainless steel is corroding uniformly, but the attack rates are low at both temperatures. Alloy 617 is corroding at the lowest rate of any material in the loop; even after 25,000 h surface films are thin enough to show interference colors.

4.3.2.2 Heat-Transfer Loop

During the past year this loop has been used to investigate the effect of a high heat flux on the corrosion of 2 1/4 Cr-1 Mo steel in

superheated steam. For these tests the heat flux has been 189 kW/m^2 compared with 125 kW/m^2 in previous similar tests.¹⁷ In the first of this year's tests the surface temperature of the tubular test specimen was 484°C at the steam inlet end, increasing to 537°C at the end of the heated section. This range of surface temperatures was the same as that for the series of tests conducted previously at 125 kW/m^2 . The first specimen was removed from test after 988 h and the second after 1322 h, at which time the test was terminated because loss of coolant to a seal around a heated specimen resulted in momentary overheating of the specimen. Weight losses on internally heated specimens (outside or convex surfaces exposed to steam) were obtained after chemically descaling the specimens. Calculated average penetrations were 20.1 and $26.4 \text{ }\mu\text{m}$ for the 988- and 1322-h exposures. These values are approximately 35% higher than comparable values obtained at the same average surface temperatures but at the lower heat flux, indicating that heat flux has a significant effect on corrosion. Externally heated specimens (inside or concave surface exposed to steam) exposed under the same conditions (189 kW/m^2) could not be weighed, but visually the oxide appeared to be similar to that formed on the internally heated specimens.

Currently, 2 1/4 Cr-1 Mo steel is being tested under the same conditions as above, but the steam temperature has been lowered so that the surface temperatures over the length of the specimen range from 426 to 482°C instead of from 484 to 537°C . The test has been in operation for 2900 h. One of the internally heated specimens was recently visually examined; no evidence of oxide exfoliation could be found. The test is continuing.

4.3.2.3 Future Plans

The Florida Power Corporation has recently announced that the Bartow Plant will be used as a peaking rather than as a base-load plant; that is, operation will be confined to periods of high power demand. For the winter and summer months the plant is expected to operate an appreciable part of the time, depending on the temperature, but for the rest of the year operation will be very sporadic. In view

of this development, we must locate a base-load power plant to supply steam to our test loops if these tests are to continue. We are attempting to find such a plant.

4.4 JOINING TECHNOLOGY — G. M. Goodwin

Materials with potential application for advanced GCRs were evaluated to determine their suitability for joining by welding. Candidate materials were obtained and tested for hot cracking susceptibility, general weldability, and mechanical properties. Weldments in alloys that have been or are being evaluated include Hastelloy X, Inconel 617, Incoloy 802, HD 556, and Inconel 618E. The latter three were studied during this reporting period.

4.4.1 Incoloy 802 — J. F. King

Incoloy 802 is a high-carbon modification of alloy 800, which is strengthened by a carbide dispersion.¹⁸ This alloy was claimed to provide superior strength and corrosion resistance at elevated temperatures and was therefore considered for possible application in GCRs.

An 8-mm-thick plate of Incoloy 802 was obtained with the following chemical composition: 0.3% C, 1.11% Mn, 44.6% Fe, 0.001% S, 0.37% Si, 0.46% Cu, 33.5% Ni, and 19.7% Cr. Because no matching composition filler metal is commercially available for this alloy, it was necessary to evaluate possible candidates. The manufacturer's recommendation was to try Inconel 625 filler metal for gas tungsten-arc welding and Inconel 112 electrodes for the shielded metal-arc process. The Inconel 625 and 112 produce weld deposits with essentially the same composition (although different from that of Incoloy 802) and with tensile strengths compatible with the Incoloy 802.

Weldments were made in plates by using the Inconel 112 and Inconel 625 filler metals and normal welding practices for this class of materials. The welds were examined visually and by liquid penetrant and radiography. No weld defects were revealed by this inspection or by guided bend tests of welded specimens. Tensile testing at room temperature on specimens taken transverse to the weld produced ruptures in

the base metal outside of the heat-affected zone (HAZ), with an ultimate tensile strength averaging 666 MPa.

An additional filler metal that had shown good potential for high-temperature applications was used to join Incoloy 802. This metal was Inconel 617, deposited by the gas tungsten-arc welding process. Again, defect-free welds were made by using this alloy. The alloy has the added advantages of not age hardening and of maintaining better ductility than the Inconel 625 alloy. Figure 4.10 is a photograph of guided bend test specimens made from plate welds of Incoloy 802 joined with Inconel 617 filler metal. These bend specimens show the ductile nature of the weld and the absence of cracking or defects.

Hot cracking evaluations were made on the Incoloy 802 base material by the spot-Varestraint testing procedure. Incoloy 802 and other alloys of interest to this program were compared by this technique to obtain a relative ranking of their susceptibility to hot cracking during welding. In this test,^{19,20} the specimen is subjected to a gas tungsten-arc spot weld thermal cycle for sufficient time to establish steady-state thermal conditions. As the arc is interrupted, an augmented strain is applied to the specimen by bending. Cracks are produced in the HAZ

Fig. 4.10. Guided-Bend Test Specimens of Incoloy 802 Joined with Inconel 617 Filler Metal.

surrounding the fusion zone, and the quantity of these cracks produces a measure of the hot cracking susceptibility of the alloy.

The total crack length measured on specimens of Incoloy 802 tested at various strains (ranging from 1.38 to 4.79%) was compared with crack measurements of other alloys as presented in Fig. 4.11. The alloys used for comparison include Hastelloy X, Inconel 618E, Inconel 617, and Haynes Development alloy 556. The results of this testing showed Incoloy 802 to be the least sensitive to hot cracking of the group tested, but all alloys were grouped close together. Previous work on Hastelloy X (ref. 21) and Inconel 617 indicated that these alloys do not present great hot cracking problems during welding, so one can conclude that all other alloys in this group should be similar.

Welds were prepared in alloy 802 by using the three filler metals previously described in this section. They have been used for aging studies and mechanical property testing. Results of this work are reported in Sects. 4.2.1 and 4.2.2.

Fig. 4.11. Spot-Varestraint Test Results for Potential GCR Materials.

4.4.2 Inconel 618E — J. F. King

Experimental alloy Inconel 618E was obtained for evaluation of its mechanical properties and weldability. Plate and wire product forms were received with the following chemical analysis: 22.5% Cr, 14.8% Fe, 56.5% Ni, 5.85% W, 0.05% C, and minor amounts of Al, Mn, S, Si, and Cu. This alloy was developed to be similar to Inconel 617 but without cobalt.

Hot cracking tests with the Inconel 618E alloy were conducted as described in Sect. 4.4.1; the results are presented in Fig. 4.11. These results fall within the group of five alloys, and thus Inconel 618E would be expected to have the same general hot cracking susceptibility as Hastelloy X but better than Inconel 617.

Welds were made in 20-mm-thick plates of this material by using the gas tungsten-arc welding process and matching composition Inconel 618E filler metal. Although some minor defects were found in these weldments by the various examination methods, the defects were determined to be related to welding technique rather than to hot cracking.

Transverse tensile specimens were tested from an Inconel 618E weldment. In room-temperature tests, the specimens containing weld metal, base metal, and HAZ in the gage length failed in the weld metal, with a yield strength of 312 MPa and an ultimate strength of 685 MPa.

Preliminary creep data on this alloy indicate that it has lower creep strength than Inconel 617 until above 980°C. This offers no great benefit to the program at this time so that work on the alloy has been discontinued.

4.4.3 Haynes Developmental Alloy 556 — J. F. King

Alloy HD 556 is a solid-solution-strengthened, iron-base alloy containing 20% Ni, 20% Co, 22% Cr, and several other alloying additions, including lanthanum and zirconium. This alloy offers improved strength and oxidation resistance over that of Hastelloy X and was therefore included in the program.

The spot-Varestraint test results (Fig. 4.11) show HD 556 to have very low susceptibility to hot cracking. General weldability is expected

to be excellent for this alloy. Plate weldments produced with this alloy by the gas tungsten-arc welding process and matching composition filler metal produced sound, defect-free welds. Four specimens were tested in a guided-bend wrap-around test jig in accordance with Section IX, Weld and Brazing Qualifications, of the *ASME Code*. Two-face and two-root side specimens were tested. No open defects or cracks were found in any of these specimens after bending, which indicates sound weldments and excellent ductility. Additional weldments were prepared for aging and mechanical property testing.

4.5 STRUCTURAL CERAMICS — V. J. Tennery and R. L. Beatty

4.5.1 Characterization of Materials

4.5.1.1 Fibrous Insulations — J. E. Selle

Characterization of two fibrous insulation boards was initiated. These insulation boards, a product of Babcock and Wilcox Company, are formed from a slurry of Kaowool ceramic fiber, Saffil alumina fiber, and organic binders. The trade names for these products are Kaowool Unifelt 2600 and Kaowool Unifelt 3000. Kaowool Unifelt 2600 is useful to 1427°C (2600°F) and Kaowool Unifelt 3000 is useful to 1649°C (3000°F) in air. As-received densities of 150.4 kg/m³ (9.4 lb/ft³) and 124.15 kg/m³ (7.8 lb/ft³), respectively, were measured.

X-ray diffractometry showed that the Kaowool Unifelt 2600 contains a considerable amount of glassy phase with α -Al₂O₃ (corundum), δ -Al₂O₃, SiO₂, Al₂SiO₅, Na₆Al₄Si₄O₁₇, and LiOH. Other peaks were observed but could not be identified. Kaowool Unifelt 3000 contained a small amount of glassy phase along with α -Al₂O₃ (corundum), δ -Al₂O₃, Al₂SiO₅, Na₆Al₄Si₄O₁₇, and MoSi₂. A few other indeterminate peaks were also found.

Work was begun on the development of tests for determining total deformation as a function of load, stress relaxation, and deformation recovery. The effects of aging on these properties at elevated temperatures in a simulated GCR atmosphere will be studied.

4.5.1.2 Dense Ceramics — R. J. Lauf and J. E. Selle

Two types of silicon nitride are being studied by using a variety of characterization techniques. NC-132 is composed of Si_3N_4 with the addition of MgO ; NCX-34 is Si_3N_4 plus Y_2O_3 . Specimens of each have been studied by x-ray diffraction, optical metallography, electron microprobe, and transmission electron microscopy.

X-ray diffraction for NC-132 detected primarily $\beta\text{-Si}_3\text{N}_4$ with small amounts of $\alpha\text{-Si}_3\text{N}_4$ and MgO . NCX-34 is predominantly $\beta\text{-Si}_3\text{N}_4$, with no $\alpha\text{-Si}_3\text{N}_4$ detected. Each material contains several minor phases that have not yet been identified, and analysis of their diffraction patterns is continuing. Comparison of NCX-34 and NC-132 patterns (taken from solid bars of each) showed substantial broadening of the $\beta\text{-Si}_3\text{N}_4$ peaks in NCX-34. This finding could indicate the presence of residual stresses or possibly a difference in crystallite size. This matter will be clarified by comparing the diffraction patterns of the solid bars with patterns taken from crushed samples.

Optical micrographs of NC-132 and NCX-34 were taken in the as-polished condition and also after a molten salt etch. Both materials exhibit a very fine grain structure; the individual Si_3N_4 grains are generally acicular in shape, the largest ones being about 2 μm thick and 10 μm long. Very small ($\sim 1\text{-}\mu\text{m}$) particles of a second phase are present. NCX-34 contains a greater number of these particles per unit volume than does NC-132. Electron microprobe analysis has indicated that these particles are rich in tungsten, iron, and cobalt. It is therefore likely that they were introduced during ball milling of the original silicon nitride powders, which probably involved tungsten carbide balls.

Characterization of NC-132 and NCX-34 by transmission and scanning transmission electron microscopy (TEM) is nearly complete. Detailed micrographs have been taken of the general microstructure at about 100,000 $\times$. Dislocations and other defects are being examined as well as grain boundary films, second phases, and microporosity. Crystallographic information is being obtained from selected-area electron diffraction patterns. We expect that comparison of TEM photographs before

and after aging or high-temperature mechanical testing will help to identify deformation modes and critical microstructural properties.

4.5.2 Environmental Compatibilities — R. J. Lauf and J. E. Selle

A structural ceramic atmospheric test system (SCATS) was designed and built. This system will be used to expose candidate structural ceramics to a simulated GCR helium atmosphere at appropriate temperatures for studying degradation effects. The system includes gas analysis equipment, a means of adjusting the gas composition, and aging furnaces.

4.5.3 Depressurization Studies — J. E. Selle

Work has been initiated on the design and assembly of a system to study the effects of rapid depressurization on the integrity of structural ceramics and fibrous insulations. Tests will be conducted at room temperature and at elevated temperatures. An autoclave containing a high-temperature furnace, designed for use in a different type experiment, has been located, and calculations indicate that only minor modifications are necessary to convert this autoclave to a depressurization apparatus. These modifications are under way.

The system will be capable of depressurizing the autoclave from 7.69 N/mm^2 (1100 psig) to 0.1 N/mm^2 (0 psig) in less than 1 s, at a rate of at least $27.6 \text{ N/mm}^2 \text{ s}^{-1}$ (4000 psi/s). The depressurization time of the system can be varied to obtain faster and slower depressurization rates if desired. Temperatures to 850°C for short periods should be possible.

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5. HTGR GRAPHITE STUDIES

W. P. Eatherly and J. A. Conlin

5.1 CAPSULE IRRADIATIONS — R. L. Senn

Five capsules were inserted into or removed from the High-Flux Isotope Reactor (HFIR) during 1979. In addition, creep capsule OC-3 was inserted in the ORR on February 28 and removed on July 8. German graphite irradiations HTK-2 (removed June 28) and HTK-3 (in May 30, removed August 30) were completed. A flux calibration experiment (HTC-1) was performed in HFIR from April 6 to 27, and a special irradiation of the N-reactor graphite was initiated on August 9 and will be removed from HFIR in March 1980. This last irradiation is being performed for United Nuclear Corporation.

Graphite creep experiment OC-3 was the second irradiation at 900°C. During operation with an adjusted helium-neon gas mixture and auxiliary electrical heaters, the ORRDACS computer maintained temperatures to within $\pm 5^\circ\text{C}$. Note that the sample temperatures are measured by two traversing thermocouples moving in the two columns of unloaded samples. This system has also been automated; the computer automatically scans and records all temperatures at the various sample locations.

A number of minor difficulties were encountered during the irradiation, all of which were circumventable and several repairable. A monthly account of misadventures follows:

March. One of the control thermocouples in zone 3 operated only intermittently, and control was shifted to its companion. (Each of the ten heater zones has at least one backup thermocouple.) One of the two movable thermocouples showed an intermittent shunting effect when used in the automatic mode after several weeks of successful operation. Operation was successful in the manual mode.

April. An extended reactor shutdown occurred to accommodate a very-low-flux experiment. During this period, OC-2 was removed from the core to an adjacent pool position and examined. The fault in the movable thermocouple was traced to a faulty insulator, which was replaced.

The error in the third-zone thermocouple was found to be in the interior of the capsule, presumably a faulty weld at the junction. No repair was possible. After the experiment was resumed, a control thermocouple in zone 5 failed, and control was switched to its backup.

May. The same movable thermocouple again faulted, but this time as a ground. Control thermocouples in zones 2 and 10 failed and were switched to their backups.

June. A control thermocouple in a guard zone failed and was switched to its backup. A small leak in a bellows seal was detected but did not interfere with capsule operation.

A noncapsule-related failure of some interest occurred involving the computer. The actual control circuitry for the capsule has "memory" for the last information sent it from the computer, and, in case of computer failure, the memory takes over and continues to operate the experiment. The computer has a "watchdog" routine to check its own programming. In May the computer alarmed and switched control to the memory units in the control circuitry. The flaw was quickly traced to a defect in the operating disk. A duplicate disk was inserted, and the computer resumed control without interruption to the experiment.

The above history of "failures" has increased our confidence in the entire system. We have operated experiments for 55 and 110 d and will now plan for 165 d.

The German graphite irradiations have been uneventful, except for a worrisome discrepancy in data. A comparison of dimensional distortion results indicated complete agreement between the experiments in HFIR and those at Petten, but dimensional data on low fluences are not sensitive to temperature. A comparison of Young's modulus data for the two reactors indicated that the temperatures calculated for the HFIR irradiations could be in error. Silicon carbide monitors implied such, but these are not reliable at the high fluences being attained in the HFIR capsules. In view of the planned Hanford reactor irradiation, a 575°C test, we decided to do a short (one-cycle) HFIR test to establish temperatures unambiguously. This experiment indicated that the German irradiations were actually performed at 600 to 620°C near the centerline

rather than at the planned 500°C. Although gamma and neutron fluxes are well established in the HFIR reflector, only the fluxes are well known in the target region. Hence, presumably the extrapolated and calculated gamma fluxes are in error.

As a result of these findings, the Hanford experiment was redesigned to increase the temperatures at the ends of the capsule by 50°C and to decrease those near the midplane by 40°C. This change should provide a more uniform temperature of 575°C throughout the Hanford capsule.

5.2 FLUX ANALYSIS FOR CREEP EXPERIMENT OC-2 — W. E. Thomas

The analytical results desired for this experiment were (1) time-averaged total neutron fluxes, (2) time-averaged fast-neutron fluxes ($E > 29.3$ fJ), and (3) graphite damage cross sections.

The experiment contained 20 dosimeters located at five axial and four azimuthal positions. Two of the dosimeters, at positions NE-4 and SW-5, were not recovered during disassembly of the experiment. Each dosimeter contained Fe, Ti, V-Co, and V-Fe wires enclosed in stainless steel tubes. The dosimeters were opened in the hot cell to recover the wires for counting. Activities resulting from the following nuclear reactions were used to compute flux magnitudes:

$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ — total flux,

$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ — total flux,

$^{46}\text{Ti}(n,p)^{46}\text{Sc}$ — fast flux,

$^{54}\text{Fe}(n,p)^{54}\text{Mn}$ — fast flux.

The experiment was in Oak Ridge Research Reactor (ORR) core position E-5 from 3:10 PM, June 30, 1978, to 12:00 PM, October 15, 1978. The flux monitor wires were removed from the stainless steel tubes on January 16, 1979, and gamma-ray counting was completed on March 28, 1979. During the time that the experiment was in the reactor, the maximum reactor power level (for any extended length of time) varied from about 28 MW(t), in July, to about 25.5 MW(t) thereafter. There were six reactor shutdowns ranging from about 7 h to about 214 h each. Total

reactor downtime was 330.3 h during the test period. In addition to the above outages, the ORR was either brought to power or was significantly reduced in power level (briefly) and then returned to the operating power level about 21 times while OC-2 was in the reactor. To consider both downtime and power fluctuations, the irradiation period had to be subdivided into 74 time increments for use in computing flux magnitudes. The ORR accumulated 209.31 TJ (thermal) during the test period. Based on the ORR rated power level [30 MW(t)], the "capacity factor" was 75.2%. From startups to shutdowns (excluding downtime), the time-averaged power level was 25.88 MW(t). Hence, for calculating neutron fluence, a value of 8.088 Ms should be used with the time-averaged fluxes.

Flux magnitudes are shown in Figs. 5.1 to 5.5. Only a qualitative comparison between the curves labeled reference data¹ and the OC-2 data should be made. The reference fluxes reflect a 30-MW(t) power level versus a 25.88-MW(t) average power level during the time OC-2 was in the core; these fluxes were also characteristic of the center of the E-5 position in a mock-up experiment with a somewhat different ORR core configuration from that for the irradiation of OC-2.

Fig. 5.1. OC-2 Flux Magnitude in the NE Dosimeter Column; Neutron Flux Versus Axial Position.

Fig. 5.2. OC-2 Flux Magnitudes in the SE Dosimeter Column; Neutron Flux Versus Axial Position.

Fig. 5.3. OC-2 Flux Magnitude in the SW Dosimeter Column; Neutron Flux Versus Axial Position.

Fig. 5.4. OC-2 Flux Magnitudes in the NW Dosimeter Column; Neutron Flux Versus Axial Position.

Fig. 5.5. Average Flux Magnitudes for Experiment OC-2; Neutron Flux Versus Axial Position.

The average flux magnitude (Fig. 5.5) is the arithmetic average of the flux magnitudes at each axial location. Because dosimeter capsule SW-5 (in the top of the experiment) had been lost, it was necessary to use the average flux magnitudes at that axial position to generate the curves in Fig. 5.3. An attempt to fit the curves through only four axial positions resulted in unrealistic (negative) extrapolated fluxes at the top of the experiment.

Both the total flux and the fast flux for OC-2 can also be evaluated from an equation of the form:

$$\phi(z) = \alpha(1.0 + \beta_1 z + \beta_2 z^2 + \beta_3 z^3) .$$

Values for α , β_1 , β_2 , and β_3 are given in Table 5.1. The axial distance z for use in the above equation should be expressed in centimeters and referenced from the horizontal midplane of the core. Positive z values are toward the top of the experiment.

Graphite damage cross sections are 807 and $261 \times 10^{-28} \text{ m}^2$ (807 and 261 barns) for use with the fast- and total-flux magnitudes, respectively.

Table 5.1. Experiment OC-2 — Coefficients for the Neutron Flux Equation

Azimuth ^a	α	β_1	β_2	β_3
<u>Total flux</u>				
NE	7.7460E 14	-1.0176E-02	-7.0483E-04	3.6987E-06
SE	8.1032E 14	-1.5638E-02	-6.2609E-04	7.8564E-06
SW	8.1523E 14	-2.2223E-02	-6.9532E-04	2.0498E-05
NW	7.9614E 14	-1.8520E-02	-6.1588E-04	1.1480E-05
Av	7.9547E 14	-1.7421E-02	-6.5617E-04	1.1996E-05
<u>Fast flux ($E > 29.3 \text{ eV}$)</u>				
NE	2.0943E 14	-1.6413E-02	-6.1879E-04	1.1438E-05
SE	2.1995E 14	-1.3345E-02	-5.7168E-04	4.1962E-06
SW	2.2284E 14	-1.9820E-02	-6.5486E-04	1.6782E-05
NW	2.1256E 14	-1.1267E-02	-5.5402E-04	-7.3518E-08
Av	2.2651E 14	-1.1782E-02	-8.0705E-04	1.0032E-05

^aAv = arithmetic mean of the fluxes at the four azimuthal positions.

5.3 RESULTS OF THE THIRD COMPRESSIVE CREEP TEST — C. R. Kennedy

The compressive creep experiments continue in the ORR with the irradiation of a second capsule (OC-3) at 900°C. This experiment is of particular interest for several reasons. The total fluence at 900°C is now extended to 3.2×10^{25} neutrons/m² (ϕ_G), and several samples from the first 900°C experiment were replaced in unstressed positions to allow recovery of the primary creep. Total strains of up to 3.2% have now been observed. The current results are generally in agreement with the results of the initial experiments at 900°C (OC-1) and 600°C (OC-2). Several marked differences have also been observed and are discussed here.

The portion of the strain due to compressive creep of H451 graphite is shown in Fig. 5.6. There appears to be a tendency for the creep rate to be decreasing at the higher fluences. The total densification of the H451 graphite for both the stressed and unstressed samples is given in Fig. 5.7. It is of interest to compare these figures in detail. Note first that the densification rates at fluences beyond about 1.5×10^{25} are identical, particularly between the stressed and unstressed samples. This fact simply means that the creep is now taking

Fig. 5.6. Compressive Creep Curves for H451 at 900°C.

Fig. 5.7. Change in Bulk Density with Irradiation and Creep.

place at constant volume. The implication is that only the primary creep occurs at nonconstant volume.

This implication is further substantiated by the observations on the relaxed specimens. Five samples stressed in experiment OC-1 were reirradiated in OC-3 under no stress. Their original creep strains on removal from OC-1 ranged from 0.6 to 2.7%. Not only was the primary creep recovered, but the densities returned to that of the control samples irradiated to the same fluence. Clearly, for these samples the secondary creep also occurred at constant volume. Linear dimensional strains were not, of course, identical for the relaxed specimens and the controls, reflecting the effects of unstressed dimensional changes superimposed on the secondary creep strains.

Data analysis is not complete. However, the electrical conductivities continue to be unaffected by creep. Young's and shear moduli changes are still less for the creep samples, but the difference is not increasing with the additional creep deformation. Sonic attenuation remains slightly higher for the creep specimens. Thermal expansion measurements are in progress.

5.4 RESULTS OF IRRADIATIONS OF KERNFORSCHUNGSANLAGE GRAPHITES IN HFIR — C. R. Kennedy

The development of the reflector and support structure graphites in the Federal Republic of Germany (FRG) has been expanded to include the more demanding requirements of systems for process heat as well as for electricity generation. The previously established reference-grade ASR-2E is considered too anisotropic to withstand the more severe neutron irradiation exposure without excessive dimensional instability. Therefore, new isotropic graphites with greater dimensional stability were needed for use in those regions of more severe exposure in the reactor. These graphites are primarily based on domestically (German) available fillers derived from coal pitch coke, in contrast to the petroleum-derived filler cokes developed in the United States. The graphites to be evaluated are listed in Table 5.2 and the properties are given by Haag et al.²

Table 5.2. Graphites Irradiated in HFIR

Grade	Filler	Fabrication
V483	Pitch coke	Isostatic molding
V356	Petroleum coke	Isostatic molding
ATR-2R	Semi-isotropic pitch coke	Vibrational molding
ATR-2E	Semi-isotropic pitch coke	Extrusion
ASR-1R	Pitch coke	Vibrational molding
ASR-2R	Pitch coke	Vibrational molding
UK-11 ^a	Pitch coke	Extrusion

^aAn experimental United Kingdom Atomic Energy Authority grade run for comparison.

This development has advanced to the stage that a number of candidate materials have been fabricated on full-scale production runs. These grades have been included in the irradiation program experiments in the High-Flux Reactor (HFR), Petten, for engineering data and in the higher-flux HFIR, Oak Ridge, for results allowing the selection of

reference grades. The graphites irradiated in HFIR have now been evaluated up to a fluence of 2.0×10^{22} EDN (equivalent Dido nickel). Dimensional changes, electrical resistivity by eddy-current measurement, elastic constants by sonic measurements, fracture strength, and the 500°C coefficient of thermal expansion (CTE) have been measured. The irradiations were performed at 600 to 620°C as indicated by silicon carbide thermal monitors.

The dimensional changes of each of these fairly isotropic grades are primarily a result of densification rather than of anisotropic growth. The maximum density is reached between 0.9 and 1.0×10^{22} EDN for all the German grades and for a United Kingdom Atomic Energy Authority (UKAEA) reference grade, as seen in Fig. 5.8. The maximum densification and the rate of densification appear to be related to the initial bulk density and the initial CTE. Obviously, graphites with lower bulk densities would be expected to densify the most. However, the same defect morphology of each structure that reduces the CTE will also influence the eventual closing of the available porosity. Therefore, a

Fig. 5.8. Volume Change of Two German Grades Compared with Reference-Grade UKAEA-11.

measure of the potential for densification should also be proportional to the thermal densification or to the difference between the theoretical and actual volumetric CTEs of each grade. We found that the maximum densification was linearly proportional to the factor $(27 - \text{CTE}_v)(2 - d)$, where CTE_v is the initial volumetric CTE in $10^{-6}/^\circ\text{C}$ and d is the initial density in Mg/m^3 . This correlation is shown in Fig. 5.9.

The effect of irradiation on the 500°C CTE was found to depend on the initial volumetric CTE. Those grades having a very low CTE experienced only a slight change, but those with high initial CTEs showed a significant decrease with fluence. These results are summarized in Fig. 5.10.

The moduli of elasticity increased in two stages before the maximum densities of the graphites were achieved (Fig. 5.11). As the graphites achieve maximum density and begin to expand, the structure appears to degrade, as indicated by a decrease in both Young's modulus and Poisson's

Fig. 5.9. Maximum Densification as Related to the Original Density d and the Volume Coefficient of Thermal Expansion CTE_v .

Fig. 5.10. Coefficient of Thermal Expansion Decreases with Irradiation as Correlated with the Initial Coefficient of Volumetric Thermal Expansion.

Fig. 5.11. Young's and Shear Moduli Changes Induced by Irradiation.

ratio. This loss in structural integrity is also observed (Fig. 5.12) by an additional increase in the electrical resistivity as the volume begins to increase.

The specimens that did not require further irradiation have all been broken in brittle-ring testing. In all, over 150 irradiated specimens have been broken, yielding almost 300 values for fracture strength.

Fig. 5.12. Effect of Irradiation on Electrical Resistivity.

The results have been correlated by using the Griffith-Irwin expression for fracture:

$$\sigma_f = (G_{Ic} E / \pi c)^{1/2} f(g) ,$$

where

σ_f = fracture stress,

G_{Ic} = energy for fracture,

E = Young's modulus,

c = effective radius of defect,

$f(g)$ = geometric factor.

Past studies have assumed that both G_{Ic} and c were unaltered by irradiation and the associated volume changes, and that, without an independent method to measure either G_{Ic} or c , the above equation could not be confirmed. We have found that measuring the sonic attenuation does, indeed, yield an estimate of defect size as it is affected by irradiation damage. Thus, we have further found that the decrease in defect size (or attenuation) with densification was as expected from independent studies on changes in porosity by fabrication or oxidation. However, as the graphite begins to expand after maximum densification due to irradiation, the increase in defect size is greater than would be expected from the simple increase in porosity, which is illustrated in Fig. 5.13.

Fig. 5.13. Sonic Attenuation Versus Fluence. Notice that the attenuation degrades faster than predicted from the observed porosity changes.

If we assume that G_{Ic} is constant and unaffected by irradiation, then the fracture stress σ_f would be proportional to $(E/\alpha)^{1/2}$. This finding is shown in Fig. 5.14 for grade ATR-2EL over a strength variation of a factor of 2. The data clearly confirm this relationship and also that G_{Ic} does not vary with irradiation, which also implies that the stress intensity factor K_{Ic} can be calculated from the modulus data, because

$$K_{Ic} = (G_{Ic} E)^{1/2} .$$

The value of G_{Ic} is constant; therefore, the value obtained from unirradiated data can be applied throughout.

The lifetime of the graphite has generally been defined as the fluence at which the irradiation-induced volume charge returns to zero. These values for the German graphites are given in Table 5.3. The values tend to suggest that the method of fabrication is relevant in controlling lifetime expectancy. It should also be emphasized that the measured physical properties given in Figs. 5.9 through 5.14 clearly demonstrate that graphite does retain its structural integrity within this definition of lifetime.

Table 5.3. Graphite Lifetime as a Function of Fabrication

Grade	Life (Fluence at $V/V_0 = 0$)	Fabrication
ASR-2R	1.35×10^{22} EDN	Vibrational molding
ATR-2R	1.4	Vibrational molding
ASR-1R	1.45	Vibrational molding
V483	1.55	Isostatic molding
V356	1.6	Isostatic molding
ATR-2E	1.7	Extrusion
UKAEA-11	1.7	Extrusion

Fig. 5.14. Correlation Between the Strength of Graphite and the Griffith-Irwin Relationship.

5.5 BENCH TEST OF THE 1200°C CREEP CAPSULE -- H. C. Roland and B. F. Johnsen

All tests on the 1200°C capsule mock-up have been finished. A 50-d run at 1200°C was completed, with no indication of change in emissivity of the tungsten gamma heater, which also doubles as an infrared reflector. After the mock-up was dismantled, the tungsten surface was unchanged other than for a minor deposit of a few carbon particles. Attention has now shifted to detailed design of the actual irradiation capsule.

5.6 REFERENCES

1. E. J. Allen and H. T. Kerr, *A Summary of Neutronics Information for Irradiation Experiments in the Oak Ridge Research Reactor*, ORNL/TM-5271 (May 1976).
2. G. Haag, W. Hammer, and M. F. O'Connor, "Preliminary Results from the FRG Graphite Programme for the Process Heat HTR," pp. 558-63 in *Proceedings of the Fifth London International Carbon and Graphite Conference, London, 1978*, The Society of Chemical Industry, London, 1978.

6. EVALUATION OF THE PEBBLE-BED HTR

6.1 INTRODUCTION — D. E. Bartine

The purpose of this effort is to examine the design and operation of the pebble-bed high-temperature reactor (PBR) concept. The work performed supports two major efforts: (1) analysis of the PBR under a technical information exchange umbrella agreement with the Federal Republic of Germany (FRG) and (2) comparison of the PBR and the prismatic High-Temperature Gas-Cooled Reactor (HTGR).

Previous ORNL analyses of PBR performance and its potential for improved ore utilization were presented in last year's annual report.¹ During this past year, ORNL results on the projected ore requirements, fuel cycle costs, and flexibility in application of the PBR were compared with prior analyses at Kernforschungsanlage (KFA). The results of the comparison were published in a joint U.S./FRG report.² Cooperation with KFA has continued this year through a joint analysis of a well-defined benchmark problem for comparison of methods and data.

Basic evaluation of the PBR has been directed toward more detailed studies of reactor operation, particularly reactor control and stability.

Development of analysis methods continues under this program in the areas of cross-section processing for double heterogeneous systems, temperature feedback effects on cross sections, stability analysis, thermal-hydraulic analysis capability, and fuel management with selective recycle.

The second major effort of this program, a comparison of the PBR and the prismatic HTGR, was initiated during the past year and is new to the program. A preliminary assessment of the relative advantages and disadvantages of the PBR compared with the HTGR is to be provided to the U.S. Department of Energy (DOE) by March 31, 1980. The ORNL work consists of an overall assessment as well as technical input in the areas of reactor control and stability, ore requirements, and fuel cycle costs; prestressed concrete reactor vessel (PCR) costs; fission product release; fabrication and reprocessing unit costs; material and

fuel requirements; graphite reflector damage; instrumentation requirements; seismic effects safety; estimated reactor availability; and overall capital costs. The ORNL work is supported by technical input from General Atomic (GA), General Electric (GE), and Gas Cooled Reactor Associates (GCRA).

6.2 METHODS DEVELOPMENT — M. A. Bjerke, R. M. Westfall, and D. R. Vondy

6.2.1 Cross Sections — R. M. Westfall and M. A. Bjerke

The analytical model implemented into the ROLAIDS resonance processor to treat the PBR fuel geometry was tested by a series of calculations. Results from these analyses of single- and double-pebble systems indicate the importance of resonance shielding as a function of fuel kernel diameter, fuel loading in each pebble, and the presence of more than one type of pebble in the system.

The one-pebble analyses indicate that the consideration of separate fissile and fertile grains is important for treating the fissile nuclide but unimportant for treating the fertile nuclide. This result is due to the small amount of fissile material in the system. However, for a fixed fuel loading and pebble shell geometry, the variation of the grain diameter has a pronounced effect on the thorium capture integral. Furthermore, the two-pebble analyses, in which fissile and fertile pebbles are treated simultaneously, demonstrated the importance of neutronic coupling between pebbles. These results, as well as a description of the analytical model, have been documented in a paper³ presented at the recent American Nuclear Society (ANS) topical meeting on computational methods in nuclear engineering.

The ROLAIDS code, as modified to treat doubly heterogeneous (grains and pebbles) problems, was used to generate 123-group cross-section sets for several PBR fuel designs. The capture resonance integral was found to be an increasing function of the carbon/heavy-metal ratio (C/HM) and a decreasing function of the kernel diameter. Figure 6.1 illustrates these effects for the reference fuel temperature of 1273 K.

Fig. 6.1. Thorium-232 Capture Resonance Integral.

An effort to extend the ROLAIDS geometry model to include a simultaneous treatment of the spherical grain geometry and the cylindrical fuel matrix geometry of the HTGR is proceeding. The calculation of escape and transmission probabilities for the cylindrical and spherical geometries has been reprogrammed to follow the recently published formalisms of Hwang and Toppel at Argonne National Laboratory (ANL). The numerical implementation of these formalisms has been documented in a paper⁴ submitted for presentation at the 1980 annual meeting of the ANS.

6.2.2 Neutronics — D. R. Vondy

A study was made of the requirements for accurate core modeling and potential of the Monte Carlo method. Of special interest is its application to the control rod worth assessment, which cannot be done

reliably with available deterministic methods. The problems are three dimensional, and simple diffusion theory would not be expected to model accurately the gas space above the core, the reflector response, and the partial control rod insertion. Use of the Monte Carlo method appears feasible to yield differential rod worth information, treating discrete positions that cause sufficient reactivity changes, and to allow resolution within the uncertainty associated with the statistical results. Perturbation schemes show promise of producing additional information at a modest cost. The inability to effect resolution of secondary information such as power density peaking, however, is a disadvantage of the method. It may be possible to account for the effect of voids (increased neutron transport) in a statistical sense by methods development.

Some effort went into the implementation of a coarse mesh method. By applying a polynomial fit to the flux within a discrete volumetric element, the node coupling coefficients may be recalculated. This may reduce the error associated with coarse finite-difference representation. The three-dimensional neutronics problem is a special challenge, and accurate solutions must be produced at a lower cost than is possible with conventional methods (finite-difference, finite-element) before time-dependent problems (such as control of xenon-driven oscillation) may be accurately modeled at an acceptable cost.

The VENTURE diffusion theory code has been enhanced so that the effects of local temperatures on microscopic cross sections can be accounted for. Thus, the steady-state, continuous-fueling problem has been solved by treating only the axial dimension with thermal-hydraulics feedback; the two-dimensional problem can be treated when the thermal-hydraulics capability becomes available. This capability will allow examination of reactivity changes caused by power level and coolant flow changes.

Additions were also made to the VENTURE code to support stability and sensitivity analyses. For the azimuthal first harmonic, a D/r^2 loss term is included that allows solution of the three-dimensional problem by treating only two dimensions (RZ) when the core has azimuthal symmetry and the first harmonic is simply a cosine. Procedures have

also been incorporated for solving the general dominant harmonic neutronics problem by extracting contributions from the fundamental during iteration given regular and adjoint solutions, treating up to three space dimensions. Methods testing has been done on a bare reactor problem that we could understand in detail. Figure 6.2 shows the dependence of the separation of the harmonic and fundamental eigenvalues on the height of a large core. Note that stability increases as this eigenvalue separation increases and that this separation is the same at one point for the azimuthal and axial first harmonics. Unfortunately, in treating multiple dimensions we find that effecting an accurate solution is easy when the harmonic eigenvalues have a large separation but not as the separation goes to zero. A dominant harmonic can be missed, and problem initialization has frustratingly been found to be crucial. This finding casts some uncertainty on the ability to treat the three-dimensional prismatic core problem.

Fig. 6.2. Bare Reactor Results for the Pebble Bed Reactor Concept.

6.2.3 Thermal Hydraulics — D. R. Vondy

One-dimensional, steady-state capability has been developed and is now being used with the VENTURE and related codes. The Los Alamos Scientific Laboratory (LASL) PEBBLE code was enhanced and is being interfaced with our local computation system. It was necessary to correlate the dependence of the graphite thermal conductivity on temperature and fast-neutron flux exposure to produce accurate temperatures.

6.2.4 Fuel Management — D. R. Vondy

The fuel management capability has been developed and is being used in our local computation system for treating the throwaway cycle. The programming has been done for flexible, selective, delayed recycle for general application to reactor problems. Proofing of this capability is still in progress. For completeness, the reporting of heavy-metal mass balances must be added and fuel cost calculations programmed.

6.3 PROJECT WORK STATEMENT ACTIVITIES UNDER U.S./FRG UMBRELLA AGREEMENT — D. R. Vondy and B. A. Worley

6.3.1 Benchmark Analysis — B. A. Worley

Neutronic analysis for the benchmark problem chosen by KFA continued this quarter. Two-dimensional calculations were completed, and comparison with the KFA calculations will be made upon receipt of their results; the ORNL results were sent to KFA in late November.

The benchmark problem is a two-dimensional description of a 1640-MW(t) reactor core using highly enriched fuel and operating on a once-through, throwaway/stowaway cycle. The reactor inlet is modeled by smearing the gas gap, upper reflector, and absorbers into one effective absorbing region. Eight feed paths (radially) are modeled. The fuel for the inner five paths has the same $^{235}\text{U}/\text{HM}$ ratio, and that for the three outer paths has a larger ratio to obtain power flattening across the core. Data and results are listed in Tables 6.1 through 6.3, and fast and thermal flux profiles are shown in Figs. 6.3 and 6.4.

Table 6.1. Data for 1640-MW(t) Benchmark Problem

<u>Reactor core</u>			
Power, MW(t)		1640	
Power density, W/cm ³		5.511	
Active core height, m		5.50	
Active core radius, m		4.15	
Volume fraction of pebbles		0.61	
<u>Reflectors</u>	<u>Top</u>	<u>Bottom</u>	<u>Radial</u>
Thickness, m	2.0	1.0	1.0
Graphite density, g/cm ³	0.32	1.40	1.60
Homogenized control poison, Σ_a/cm	0.0005		
<u>Pebble design</u>			
Diameter, cm		6.0	
Graphite shell thickness, cm		0.5	
Graphite density, g/cm ³		1.70	
<u>Coated particle design</u>			
Kernel material		U/ThO ₂	
Kernel diameter, μm		400	
Kernel density, g/cm ³		9.50	
Uranium enrichment		0.93	
Graphite coating thickness, μm		85/30/80 ^a	
Graphite coating density, g/cm ³		1.0/1.6/1.85 ^a	
<u>Pebble-averaged nuclide density, atoms/b-cm</u>			
Inner zone paths (1-5)			
²³⁵ U		1.94848E-5	
²³⁸ U		1.44810E-6	
²³² Th		2.39810E-4	
¹² C		8.37960E-2	
¹⁶ O		5.14020E-4	
Outer zone paths (6-8)			
²³⁵ U		2.27848E-5	
²³⁸ U		1.69335E-6	
²³² Th		2.39810E-4	
¹² C		8.37960E-2	
¹⁶ O		5.14020E-4	

^aBuffer layer/sealer layer/pyrolytic carbon layer.

Table 6.2. Key Results for 1640-MW(t) Benchmark Problem

Geometrical representation	2-D
<u>Fuel loadings — average over core</u>	
²³⁵ U/heavy metal (mass ratio)	0.0806
Grams of heavy metal/pebble	11.44
Carbon/heavy metal (atom ratio)	320
<u>Physical characteristics</u>	
$\bar{\eta}$ (fissile U, Pu)	2.088
$\bar{\alpha}$ (fissile U, Pu)	0.180
<u>Global characteristics</u>	
Residence time, years	3.15
Burnup, MWD/kg heavy metal	103
Conversion ratio	0.575
Leakage loss fraction	0.0407
Fission product loss fraction	0.1010
Peak kW/pebble	2.98
Fissile inventory, kg	657
<u>Mass flow rates, kg/d</u>	
<u>Feed</u>	
²³⁵ U	1.2866
²³⁸ U	0.0968
²³² Th	14.5852
<u>Discharge</u>	
²³⁵ U	0.0737
²³⁸ U	0.0778
²³² Th	13.4405
²³³ U	0.3055
²³³ Pa	0.0052
²³⁴ U	0.0933
²³⁶ U	0.1901
²³⁹ Pu	0.0008
²⁴⁰ Pu	0.0005
²⁴¹ Pu	0.0005
²⁴² Pu	0.0011

Table 6.3. Fractional Neutron Losses for 1640 MW(t)
Benchmark Problem

Leakage	0.04069
<u>Absorption in fission products</u>	
¹³⁵ Xe	0.02247
¹⁴³ Nd	0.01348
Lumped, saturating	0.00838
¹⁴⁹ Sm	0.00726
¹⁰³ Rh	0.00649
¹³¹ Xe	0.00555
¹⁴⁷ Pm	0.00550
¹³³ Cs	0.00507
¹⁴⁵ Nd	0.00381
¹⁵¹ Sm	0.00356
Lumped, nonsaturating	0.00323
¹⁵² Sm	0.00284
^{148m} Pm	0.00236
¹⁵⁰ Sm	0.00201
¹⁵³ Eu	0.00190
¹⁵⁵ Eu	0.00157
¹⁵⁴ Eu	0.00151
⁸³ Kr	0.00139
¹³⁴ Cs	0.00097
¹⁴⁸ Pm	0.00089
⁸² Kr	0.00030
¹³³ Xe	0.00024
¹⁰³ Ag	0.00010
¹³⁵ Cs	0.00010
¹⁰⁵ Rh	0.00007
¹⁴⁷ Nd	
¹⁴⁹ Pm	
¹³⁵ I	
Total	0.10096
<u>Absorption in actinides</u>	
²³⁵ U	0.29126
²³² Th	0.26721
²³³ U	0.18138
²³³ Pa	0.01251
²³⁴ U	0.00969
²³⁶ U	0.00840
²³⁸ U	0.00433
²³⁹ Pu	0.00409
²⁴⁰ Pu	0.00145
²⁴¹ Pu	0.00132
²⁴² Pu	0.00010
²³⁴ Am	0.00004
²³⁹ Np	0.00001
Total	0.78185
<u>Absorption in other materials</u>	
¹² C	0.04284
Absorber in top reflector	0.03350
¹⁶ O	0.00008
He	0.00007
Total	0.07650
Total fraction losses	1.00000

In addition to comparison with KFA results, work is now planned for comparison with a two-dimensional model that explicitly treats the gas space above the core. Discrete ordinates transport theory and diffusion theory calculations will be performed.

6.3.2 Control and Stability Assessment — D. R. Vondy

The assessment of control and stability of the PBR core concept progressed this year. Calculations of control rod worth were done with

Fig. 6.3. Fast Flux Profile for 1640 MW(t) Benchmark Problem.

Fig. 6.4. Thermal Flux Profile for 1640-MW(t) Benchmark Problem.

the newly developed ORNL triangular mesh diffusion theory neutronics code VALE for a 1640-MW(t) core without power density flattening. With rods on a 102-cm triangular pitch within the core, 1.11 rods per square meter (except for a full ring near the reflector) and a ring within the reflector, the following results were obtained:

<u>Rods inserted</u>	<u>Reactivity worth per rod</u>
Central rod (1)	-0.00684
Outer ring (24)	-0.00055
Reflector rods (24)	-0.00013
All rods in core (61)	-0.00519
Reflector rods, core rods in (24)	-0.00023
All rods (85)	-0.00379

Thus, reactivity shutdown capability can be provided, but rods toward the outside of a large core have low worth, and rods in the reflector have decreasing worth as the core size is increased. A large perturbation of the neutron flux is associated with selective insertion, causing decrease in rod worth. Preferred rod arrangements may cause discrete core diameter changes to be desirable, especially in small core sizes.

Effort has gone into analysis of stability against xenon-driven oscillation. In a generic study, the emphasis is on identifying the core size above which oscillation is not self-dampening and, hence, requires control; the effect of the variables on this size; and the control requirements for larger cores. For the low height-to-diameter reference design, the core seems to have the very desirable characteristic of axial stability, the dominant harmonic being azimuthal above some small size. (Note the bare reactor results in Fig. 6.2.) Once-through pebble management and associated peaking of the flux near the top of the bed tend to increase axial stability but may decrease azimuthal stability. Calculations indicate that the stability boundary is sensitive to the core conditions and the neutron flux distribution. It is also influenced by the details above the core, including control rod positions, that affect the neutron flux at the top of the core (where the power density is high); and by the modeling of these details.

Subject to further analysis and certain limitations, the pebble-bed core appears to have a bound for stability at about 3000 MW(t) at a power density of 5 W(t)/cm^3 , but this bound can shift up or down appreciably. Increase in stability is associated with a decrease in the C/HM ratio, an increase in power density (smaller core), and ^{233}U buildup. A negative temperature coefficient of reactivity contributes to this stability, but it is difficult to evaluate temperature feedback effects, especially those caused by a positive contribution from the ^{135}Xe concentration and its cross section, which falls off rapidly with increased neutron energy in the thermal range. This positive contribution from the xenon increases as the temperature increases and as the ^{135}Xe concentration increases — with increase in the specific power density per fissile atom (increase in the C/HM ratio, for example). Stability requirements can place severe constraints on both early operation and startup with xenon override.

6.4 PRISMATIC/PEBBLE-BED REACTOR DESIGN COMPARISON — OVERALL ASSESSMENT — T. J. Burns

The basic methodology being used for the high-temperature reactor (HTR) core assessment consists of a cost-benefit analysis of each system under conditions of uncertainty about the basic data. Once the objective function is defined (in this case, cost), sensitivity calculations are performed to find the primary variables that determine the value of the objective function. The uncertainty in these variables is then assessed, followed by a probabilistic calculation to determine the uncertainty in the objective function. This methodology permits comparison of both HTRs at the same level of confidence in the objective function. The preliminary assessment effort consists of five tasks:

1. decision definition and generation,
2. structuring of decision logic,
3. deterministic sensitivity calculations,
4. probabilistic sensitivity calculation, and
5. documentation of results.

Tasks 1 and 2 above have been completed, task 3 is under way, and tasks 4 and 5 are scheduled for the first quarter of 1980.

6.5 PRISMATIC/PEBBLE-BED REACTOR DESIGN COMPARISON -- TECHNICAL ASSESSMENTS -- B. A. Worley, J. P. Renier, R. P. Wichner, M. F. Osborne, A. R. Olsen, P. L. Rittenhouse, W. P. Eatherly, S. J. Ditto, W. C. Stoddart, J. G. Delene, M. L. Myers, and D. J. Naus

6.5.1 Core Neutronic Analysis -- B. A. Worley and J. P. Renier

Calculations to obtain HTGR mass flows were performed in one-dimensional geometry. The primary emphasis was to understand and become familiar with calculational methods for HTGR fixed-fuel reactors. This experience is needed to produce consistent neutronic results for the pebble-bed and prismatic core designs.

Preliminary neutronics calculations for HTGR throwaway cycles were performed for different C/HM ratios, cycle lengths, fuel holes, diameters, etc. Both conventional loading and axial push-through cycles were analyzed for highly enriched uranium (HEU)/Th, 3360 MW(t), 7.1 W/cm^3 average power density cores. The neutronics calculations were performed in *R* geometry.

For benchmarking with GA's results, the following calculations were performed: HTGR HEU/Th, 3360 MW(t) throwaway cycle and HTGR medium-enriched uranium (MEU)/Th (25%), 3360-MW(t) throwaway cycle. Benchmark calculations in *R-Z* and hexagonal geometry are in progress.

6.5.2 PCRVR Comparative Costs -- D. J. Naus

Cost estimates have been prepared for both PCRVRs and secondary containments for five reactor systems: HTGR-SC, DCGCR, HTGR-GT, PBR demo, and PBR.

6.5.2.1 PCRVR Cost Estimates

Information for use in the development of PCRVR cost estimates was quite limited. The HTGR-SC and DCGCR data were obtained from ref. 5, the HTGR-GT information was obtained by using data in ref. 6 in conjunction with an extrapolation of data obtained from ref. 5, and HHT demo

plant data were supplied from refs. 7 and 8. The PBR data were then obtained by extrapolating the HHT data and using scaling factors to account for the larger output [3000 MW(t) vs 1600 MW(t)]. Design parameters used in preparing the cost estimates for the PCRVs for each of these systems are presented in Table 6.4.

In preparing PCRV cost estimates, several additional assumptions, especially for the PBRs, had to be made; for example, the PBR systems were assumed to use a cold liner rather than the warm liner currently proposed. Estimates based on the use of equivalent process equipment (gas turbines, number of loops, etc.) were made for the 3000-MW(t) PBR. Other assumptions, as well as calculations used to develop material quantity takeoffs, are presented in ref. 9. Table 6.5 presents unit cost factors, quantity takeoffs, and estimated PCRV costs for the two gas-turbine systems considered.

Table 6.4. Design Parameters

Item	HTGR-GT		PBR	
Output, MW(e)	1200		1200	
Output, MW(t)	3000		3000	
Coolant inlet, °C (°F)	453	(847)	453	(847)
Coolant outlet, °C (°F)	850	(1562)	850	(1562)
Coolant pressure, Pa (psia)	0.33	(1129)	0.33	(1129)
Loops, number	6		6	
Core cavity diameter, m (ft)	13.3	(43.83)	17	(56)
Core cavity height, m (ft)	16.1	(53.08)	14.3	(47)
PCRv diameter, m (ft)	39.3	(129.04)	48.1	(158)
PCRv height, m (ft)	35.4	(116.14)	41.1	(135)

Table 6.5. Prestressed Concrete Reactor Vessel Cost Factors

Material	Unit cost \$	HTGR-GT		PBR	
		Quantity	Cost (\$ × 10 ³)	Quantity	Cost (\$ × 10 ³)
Concrete	300/yd ³	48,832	14,650	89,290	26,787
Liners and penetrations	12,000/ton	3,493	41,916	3,522	42,264
Cooling tubes	30/ft	651,798	19,554	657,534	19,726
Tendons	10,000 ea	1,260	12,600	2,155	21,550
Circumferential steel	5,000/ton	3,757	18,785	6,819	34,095
Insulation	50/ft ²	325,899	16,295	328,867	16,443
Reinforcing steel	2,500/ton	2,589	6,473	4,734	11,835
Total cost			130,273		172,700

6.5.2.2 Secondary Containment Cost Estimates

Cost estimates were also prepared for the secondary containments for each of the five reactor systems. The HTGR-SC and DCGCR information was obtained from ref. 5. As noted in Table 6.4, the PCRV sizes for the DCGCR and HTGR-GT are almost identical; therefore the same containment data were used for both. The only containment information located for the pebble-bed systems, presented in ref. 10, consisted of clearances between the containment and the PCRV for a steam cycle plant. Table 6.6 presents the containment design data used for development of the initial cost estimates.

Table 6.6. Containment Geometries^a

Reactor system	Internal diameter [m (ft)]	Internal height [m (ft)]
HTGR-SC	42.7 (140)	68.0 (223)
DCGCR	48.5 (159)	62.5 (205)
HTGR-GT	48.5 (159)	62.5 (205)
PBR Demo	55.3 (181.4)	94.4 (309.7)
PBR plant	66.8 (219.3)	105.9 (347.6)

^aContainments are cylindrical with oblate spheroidal dome caps. Wall thicknesses are 1.07 m (3.5 ft) for all systems. The HTGR-SC containment utilizes reinforced concrete, whereas the others utilize a combination of reinforced and prestressed concrete.

As with the PCRV cost estimates, several assumptions had to be made for the pebble-bed containments; for example, clearances between PCRVs and containment internal height were the same as for the pebble-bed steam cycle plant, and the clearance between the PCRV and the containment internal diameter was maintained the same as that required for the prismatic plant. Other assumptions, as well as calculations used to develop the containment cost estimates, are presented in ref. 11. Table 6.7 presents unit cost factors, quantity takeoffs, and estimated costs of containments for the two equivalent GT systems considered in Table 6.5.

Table 6.7. Containment Cost Factors

Material	Unit cost \$	HTGR-GT ^a		PBR ^b	
		Quantity	Cost (\$ × 10 ³)	Quantity	Cost (\$ × 10 ³)
Concrete	300/yd ³	74,700	22,410	90,114	27,034
Liner steel	12,000/ton	1,700	20,400	2,266	27,192
Reinforcing steel	2,500/ton	6,400	16,000	7,721	19,303
Structural steel	1,200/ton	580	696	688	826
Tendons	5,000/ton	1,400	7,000	1,968	9,840
Total cost			66,506		84,195

^a48.4 m (159 ft) ID by 62.4 m (205 ft).

^b57.4 m (188.5 ft) ID by 89.5 m (293.8 ft).

6.5.3 Fission Product Release — R. P. Wichner

The following work scope for this task area has been prepared for review and comment from program managers.

6.5.3.1 Introduction

Each of the 12 reactor pairs considered in the comparative evaluation has a different amount of fission product activity outside the fuel and a different distribution of this activity around the primary loop. There are two primary reasons for varying the degrees of ex-core activity: (1) different fuel temperatures, particularly different temperature histories for the hottest approximately 10% of the fuel, and (2) the type of fissile mixture (i.e., HEU vs MEU). Fissile yields differ, and ^{110m}Ag becomes a particularly significant isotope for maintenance analyses, using MEU fuel.

The ex-core fission product activity affects the comparative evaluation in three ways: (1) the cost of operation and maintenance; (2) costs that may be ascribed to activity release under normal operating conditions, and (3) costs that may be ascribed to accident-caused activity release.

The above items appear to impact the evaluation in the order listed, especially in view of the impending lowering of the allowable occupational dose. Operation and maintenance (O&M) costs may be classified as scheduled (and hence designed for) and unscheduled. Expected routine

maintenance involves inspection and repair of all rotating equipment, control valves, heat exchangers, etc. The largest single maintenance task is expected to be the periodic examination and repair of the main turbine, and its costs may dominate the scheduled maintenance costs. Similarly, the refueling operation probably dominates the operating cost picture. Unscheduled maintenance is by definition unexpected and hence not planned. It is quite difficult, therefore, to gain estimates of unscheduled maintenance costs as a function of ambient activity level. Both labor (man-rem) costs and the cost of the reactor unavailability must be factored in.

Thus, some of the ways in which ex-core activity levels affect O&M costs are (1) the cost of labor (man-rem costs), (2) downtime costs (e.g., the cost of replacement power for nonscheduled maintenance), (3) health physics costs, and (4) cost of remotely operated equipment, if necessary.

The maximum allowable routine activity releases are specified in Appendix I of 10 CFR 50. Past calculations at GA have indicated that anticipated releases for older concepts are only a factor of 5 to 10 less than the designated maximum allowable airborne value. Noble gases and tritium would be the major contributors to routine airborne releases. Noble gas release is essentially a function of the ex-core, fissile contamination level and the failed fuel particle fraction (hence, mainly the temperature history of the hottest ~10% of the fuel). The variation in released activity with reactor type must subsequently be translated into an effective cost for use in the evaluation study.

Similarly, variations in accident-caused activity release as a function of reactor concept would have to be translated into a cost for application to the evaluations study. High-consequence, low-probability accidents, which generally involve core heatup and, consequently, fuel failures, are expected to show no variation in consequence as a result of variation in ex-core activity, because in all cases activity release from the fuel dominates the expected ex-core activity under normal operation. However, the consequence of higher-probability accidents, such as the design-basis disruptive accident (DBDA) or any event that breaches the PCRV with no core heatup, will be, to a degree, dependent

on ex-core activity. However, the safety margins for these are quite high (in the thousands) so that associated costs, however estimated, will probably be low in comparison with costs of routine releases where the margins are from 5 to 10.

6.5.3.2 General Work Scope

The scope of the work is to ensure development and review of fission product activity level estimates needed to evaluate (1) differences in O&M costs, (2) differences in routine releases, and (3) differences in the consequence of ex-core, activity-dependent accidents in the considered reactor concepts. However, it is recognized that this cannot be done in the required time scale for all the reactor pairs under consideration, and methods must therefore be found to acquire reasonable estimates, perhaps based on extrapolation of past calculations at GA.

In addition, an attempt should be made to assess possible differences in the probability-consequence curve at the high consequence end of the accident spectrum. For example, does the difference in core configuration affect the seriousness of unrestricted heatup accidents? If so, to what extent will the probability-consequence curve be affected? At present, there is no apparent reason for any significant difference between the concepts on this issue, but it is important to verify this impression with some care.

6.5.3.3 Specific Work Scope

Therefore, the work scope planned by Barsell and Henson will be reviewed with Winkler by mid-January. No activity distribution estimates are currently planned, as would be required for the determination of relative O&M costs. Some means for obtaining reasonable activity distribution estimates will be formulated with Hanson for all reactors under consideration. Total ex-core activity estimates are planned for only five concepts, four of which are for MEU fuel. These estimates will be extended to all the 24 reactor cases involved in the study. The consequence of the high-probability accidents will be estimated as a function of the degree of ex-core activity. This cannot be done in the

usual fashion (i.e., the way it was done in the AIPA work), but perhaps estimates can be made that are based on past calculations. The effect of the different core concept in each reactor pair on the high-consequence, low-probability end of the accident spectrum will also be examined.

6.5.4 Fuel Fabrication Technology Assessment and Comparative Fabrication Cost Estimates — A. R. Olsen

The objectives of this task are (1) to develop and document consistent cost estimates for fabrication of both prismatic and pebble-type fuel elements and (2) to identify the required research and development (R&D) needed to bring each process to a level suitable for commercial application together with estimates of the resource requirements for this R&D effort.

All fuel fabrication cost estimates should be specific to a given fuel element design and cycle description. Because the overall HTR core evaluation studies will address a variety of reactor cycles and specific details on cycle descriptions and fuel element descriptions are not currently available, the initial phase of this task is to develop and document fuel fabrication cost estimates for selected base cases. This approach is reasonable, because the fabrication processes and equipment requirements are similar for a given type of fuel element. Thus, consistent realistic estimates can be developed by compiling and arranging major process equipment items to fit a given cycle.

During this quarter four base case cycles were selected from existing published fuel cycle analyses. These analyses provided the necessary fuel element descriptions and reactor mass flow data to define fabrication process requirements and to derive commercial-scale plant production capacity requirements. The chosen cycles, a once-through and a recycle case for each reactor fuel element type, cover the fabrication of low-enriched uranium (LEU) ($<20\%$ ^{235}U)-Th fuel for once-through cycles and both fabrication and refabrication of HEU-Th fuel for the recycle cases. The parameter details obtained from available data and publications for these cycles are summarized in Tables 6.8

Table 6.8. Preliminary Pebble-Bed-Prismatic Comparison Cycle Reactor Characteristics

	Once-through cycles		Recycle	
	Pebble bed, ^a MO20	Prismatic, ^b MEU(5)-Th(OT)	Pebble bed, ^c Th/U	Prismatic, ^b HEU(5)/HEU(3)-Th recycle
Design MW(t) (Q_{Th})	3000	3360	3000	3360
Design MW(e) (Q_e)	1200	1332	1200	1332
Plant efficiency, Q_e/Q_{Th}	0.40	0.396	0.4	0.396
Load factor	0.80	0.75	0.8	0.75
Annual electricity generation MW(e)-years	960	999	960	999
Fuel elements per core	3,233,370	5288	3,233,370	5288
Fuel elements replaced per year	1,111,720	1322	1,111,720	1322
Core power density, W/cm ³	5.0	7.1	5.0	7.1
Average burnup, MWd/kg HM	100	133	36.5	59.5
Average fuel residence time, d	872	1460		
Conversion ratio (av during equilibrium)	0.575	0.54	0.824	0.75
N_C/N_{HM}	458	395	220	169

^aE. Teuchert, *Once Through Cycles in the Pebble Bed HTR*, Jul-1470 (December 1977), Case MO20.

^bGeneral Atomic Company, *INFCE Working Group 8 - Advanced Fuel Cycle and Reactor Concepts - Subgroup A: Once-Through Fuel Cycles*, Draft (July 1978).

^cE. Teuchert et al., *Closed Thorium Cycles in the Pebble Bed HTR*, Jul-1569 (January 1979).

and 6.9. Information derived from assessment of fabrication requirements for these cycles and the cost estimates of commercial plants, each to support 20 reactors of each type, will provide the basic process concepts and equipment costs to be used in assessing any other fuel element designs or cycle definitions that may be defined as the HTR core evaluation study proceeds.

In addition to the selection of base cases for phase 1 analysis, the methodology to be used in the fuel fabrication process assessment and cost estimation has been outlined, and preliminary study ground rules and assumptions have been defined. These have been incorporated in a preliminary partial draft report of the base case assessments and transmitted to both HOBEG in FRG and GA for review and comment. This draft includes preliminary functional flow diagrams and brief process descriptions for both the pebble and prismatic fuel element fabrication processes.

Visits were planned to both HOBEG and GA to review the process descriptions, to obtain additional functional flowsheet process informa-

Table 6.9. Pebble-Bed-Prismatic Comparison Cycles, Fuel Element Descriptions

	Once-through			Recycle					
	Pebble bed		Prismatic HTGR MEU(5), Th-OT cycle makeup	Pebble bed Th/U-total recycle		Prismatic R HEU(5)/HEU(3)-Th recycle			
	MO20 makeup			Fabrication F1	Refabrication R1	Fabrication T1	Fabrication Makeup	Refabrication	
	Inner	Outer						²³³ U recycle (23R)	²³⁵ U recycle (25R)
Fuel elements									
Elements per year	889,806	221,912	1,322	171,000	609,000	716,000	500	733	80
Heavy element loading (g/element)									
Total uranium	0.76	4.60	2,920	1.24	1.86		740	650	1.24
²³⁵ U	0.55	0.92	0,580	1.15	0.90		690	30	0.53
²³³ U					0.20			490	
Thorium	5.31	3.47	2,490	15.22	14.60	16.45	11,240	11,240	11.24
Total heavy metal	8.07	8.07	5,410	16.46	16.46	16.45	11,980	11,890	12.48
	1:92:1	1:1:33							
Chemical composition									
Fissile kernels	(U,Th)O ₂	(U,Th)O ₂ (20% U5)	UCO (20% U5)	(U,Th)O ₂ (93% U5)	(U,Th)O ₂ (59% fissile)		UCO (93% U5)	UCO (80% U3-U5)	UCO (43% U5)
Fertile kernels						ThO ₂	ThO ₂	ThO ₂	ThO ₂
Carbon ratios									
C/Th			850				180	180	180
C/U			740				2,730	3,110	1,630
C/heavy metal	458	458	395	220	220	220	169	170	162
Shipping requirements	Unshielded		Unshielded	Unshielded	Unshielded	Unshielded	Unshielded	Shielded	Shielded
Particle description									
Fissile particle									
Kernel diameter, μm	400		350	400	400		195	300	300
Kernel density, g/cm ³	0.5		10.8	9.5		10.8	10.8	10.8	
Buffer thickness, μm	85		105	85	85		110	105	105
ILTI thickness, μm	30		35	30	30		35	35	35
SiC thickness, μm			35				35	35	35
OLTI thickness, μm	80		45	80	80		40	40	40
Average particle diameter, μm	790		790	790	660		635	730	730
Fertile particle									
Kernel diameter, μm			500			"Assumed" 500	500	500	500
Kernel density, g/cm ³			9.9			9.9	9.9	9.9	9.9
Buffer thickness, μm			85			95	95	95	95

Table 6.9 (continued)

Once-through			Recycle					
Pebble bed MO20 makeup		Prismatic HTGR MEU(5), Th-OT cycle makeup	Pebble bed Th/U-total recycle			Prismatic R HEU(5)/HEU(3)-Th recycle		
Inner	Outer		Fabrication F1	Refabrication R1	Fabrication T1	Fabrication Makeup	Refabrication	
							²³³ U recycle (23R)	²³⁵ U recycle (25R)
ILTI thickness, μm								
SiC thickness, μm								
OLTI thickness, μm					80	80	80	80
Average particle diameter, μm					850	850	850	850
Prismatic								
Fuel rod description								
Rod diameter, cm						1.58	1.58	1.58
U content, g						0.40	0.75	0.75
Th content, g						6.79	6.79	6.79
Fuel element description								
No. of fueled holes ^a						138/76	138/76	138/76
Diameter of fueled holes, cm						1.61	1.61	1.61
No. of fuel rods/ element ^a						1,656/851	1,656/851	1,656/851
No. of cooling holes ^a						72/43	72/43	72/43
Diameter of cooling holes, cm						2.10	2.10	2.10
Poison content, kg						0.01	0.01	0.01
Graphite, kg						94	94	94
Pebble								
Ball diameter, cm			6	6	6			
Graphite shell thickness, cm			0.5	0.5	0.5			
Graphite density, g/cm ³			1.7	1.7	1.7			
Graphite, g								

^aStandard/control fuel elements.

tion, and to obtain all available information on process status, equipment designs, and operating experience before proceeding with the commercial-scale plant cost estimation. This type of consultation is particularly needed for a realistic evaluation of the pebble fuel element fabrication, processing, and costs because of the limited amount of detailed information on the processes and the equipment requirements available in the literature. Because the HOBEG visit could not be arranged in this quarter, it will be necessary to depend on written comments on the draft material forwarded in early December 1979 in order to develop cost estimates for the Preliminary Assessment Report in March 1980. However, this restriction will not apply to the prismatic process assessment and estimates because a large amount of detail from several recent U.S. studies is available.

6.5.5 High-Temperature Materials Assessment — P. L. Rittenhouse

An assessment of the structural integrity of the control rods for the pebble-bed core was initiated during this reporting period. This work is summarized below.

6.5.5.1 Mechanical Loading

Two types of control rods are being considered for use in the pebble-bed core. The first of these (the push-type rods) are forced directly through the bed of fuel spheres, and the second (rotating-type rods) are rotated as they penetrate the fuel bed. In both cases the rate of insertion is to be limited to 2 cm/s to minimize temperature increases in the control rod cladding. The force F required for full insertion (4.5 m) of a push-type rod is about 13,000 kg. However, this force can be lowered to about 3300 kg if ammonia is injected immediately before the start of rod insertion. The forces required for insertion are reduced significantly when rotating-type rods are employed. Even without ammonia only 1000 to 2000 kg is needed. (It must be noted that F values obtained from various sources were not entirely consistent. Those quoted above appear to be reasonable but will be checked further.)

The following assumptions have been made for both types of rods in the analysis to follow:

- Insertion rate of the control rods is 2 cm/s.
- Maximum temperature of the rods during insertion is 750°C.
- The column length L of the rods is 6.5 m.
- The outer cladding tube is 130 mm OD by 110 mm ID, the inner cladding tube is 90 mm OD by 80 mm ID, and cladding cross-sectional area A is 5100 mm².
- There is no support from the B₄C absorber material.
- No irradiation damage has occurred.

Failure of the control rods during their insertion into the core could occur by plastic yielding (i.e., when $F/A >$ yield strength of the cladding) or by buckling (i.e., when $F > F_{cr} = \pi^2 EAr^2/4L^2$). The term F_{cr} is the Euler critical load (buckling load), E is the elastic modulus of the cladding material, and r is the least radius of gyration of the concentric cladding tubes. Safety factors (i.e., force to cause failure/applied force) have been calculated for both failure modes over a range of forces encompassing those discussed earlier for push- and rotating-type rods. These are given in Table 6.10 for three cladding alloys that have been proposed. First, notice that in all cases the plastic yielding safety factor (proportional to F^{-1} , A , and yield strength) is >5 and, therefore, that failure of the control rods by this mode should be impossible even under the most severe conditions

Table 6.10. Calculated Control Rod Safety Factors for Normal Insertion Rates and Temperatures

Control rod insertion force (kg)	Failure mode					
	Plastic yielding			Column buckling		
	Alloy 800H	Hastelloy X	Inconel 625	Alloy 800H	Hastelloy X	Inconel 625
1,000	78	135	216	16	16	18
2,000	39	67	108	8.0	8.0	8.8
5,000	16	27	43	3.2	3.2	3.5
10,000	7.8	14	22	1.6	1.6	1.8
15,000	5.2	9.0	14	1.1	1.1	1.2

(i.e., push-type rods without ammonia, $F = 13,000$ kg). The significant difference in safety factors for the three alloys is a direct result of their yield strengths.

Column buckling safety factors (proportional to F^{-1} , L^{-2} , r^2 , A , and E) are much lower than those for plastic yielding but still seem sufficient for all cases except the push-type rods without ammonia. Values of E for the three alloys are essentially identical, and this results in the low variation in the safety factor observed between alloys.

Should, for some reason, the temperature* of the cladding reach 950°C during control rod insertion, safety factors would be reduced to those shown in Table 6.11. The plastic yielding safety factors are reduced by 50% or more, depending on alloy, because yield strength falls off rapidly from 750 to 950°C ; those for buckling are relatively unaffected because E is almost constant over this temperature range. However, even with these lower safety factor values, the conclusions on control rod integrity are unchanged for the reference (750°C) case. Even more-severe temperature conditions were considered by calculation of yielding safety factors for Hastelloy X and Inconel 625 at 1100°C and $10,000$ kg_f and of the buckling safety factor for Alloy 800H under the identical temperature and stress. The yielding safety factors calculated were 2.6 and 4.0, respectively, for Hastelloy X and Inconel 625, a reduction of approximately 50% from the 950°C case; that for buckling of Alloy 800H was 1.3 versus 1.4 for 950°C and 1.6 for 750°C .

* A temperature of 950°C would be achieved if the insertion rate were increased to 10 cm/s. Under such conditions, F might also be higher.

Table 6.11. Calculated Control Rod Safety Factors at 950°C

Control rod insertion force (kg)	Failure mode					
	Plastic yielding			Column buckling		
	Alloy 800H	Hastelloy X	Inconel 625	Alloy 800H	Hastelloy X	Inconel 625
1,000	39	52	91	14	14	15
2,000	19	26	45	7.0	7.0	7.5
5,000	7.8	10	18	2.8	2.8	3.0
10,000	3.9	5.2	9.1	1.4	1.4	1.5
15,000	2.6	3.5	6.1	0.94	0.94	1.0

6.5.5.2 Thermal Aging and Irradiation Damage

During the major portion of the service life (approximately four years) of the control rods, the cladding will be at temperatures in the range of 575 to 650°C; hardening of the cladding materials, with an accompanying reduction in ductility, will occur by thermal aging. Neutron doses of about 10^{22} neutrons/cm² (thermal) will be experienced and will further add to hardening and ductility reduction. Effects on such properties will begin to be seen at about 10^{16} neutrons/cm².

Evaluation of the performance of the control rods (as in the section above) as a function of service time will require tensile properties (yield strengths and elastic modulus values) as a function of neutron dose at a nominal temperature of 600°C. Most probably, both yield strength and elastic modulus will, at least initially, increase with exposure and result in calculated safety factors that will be enhanced, or at least not reduced, by service exposure.

More realistically, one must worry about the ductility and toughness characteristics of the cladding material (both will probably be degraded significantly by the service exposure) because of thermal shock, possible impact loads (e.g., the insertion forces generally occur as "spikes" rather than as a continuous loading), seismic events, etc. There is probably no method for accurately determining what levels of toughness and ductility are necessary for assurance of control rod integrity. (These properties are not involved in the safety factor calculations described earlier.) Therefore, it is not improbable that, for example, ductility minimums for the control rod cladding at end of life will be specified on an arbitrary but conservative basis. Input relative to the licensing experience of U.S. reactor vendors on control rods would be valuable in further analysis.

6.5.5.3 Cladding-Environment Interactions

The alloys being considered for control rod cladding are known to react with the impurities present in gas-cooled reactor (GCR) helium and thereby to undergo changes in mechanical properties, usually in a

detrimental fashion. However, for temperatures up to about 750°C the degree of such reactions should be inconsequential; therefore, there is no reason to expect environmental degradation of the cladding alloys during their approximately 30,000-h life at 650°C. Under normal full-insertion conditions the temperature is only slightly higher, and no damage should occur during the short periods in question. Even a few hours at temperatures as high as 900 to 950°C should cause no problems. Additional compatibility questions that need to be addressed are the possibilities of reaction between the B₄C absorber material and the cladding and between the fuel spheres and the cladding. Both could affect the properties of the cladding and its subsequent performance.

6.5.6 Graphite Reflector Damage — W. P. Eatherly

The study to evaluate graphite reflector design is proceeding along the lines of developing solutions of the constitutive equations in closed form for checking more exact solutions derived from finite-element analyses. Such simplified solutions will err in the direction of overconservatism and, hence, set upper bounds on the evolved internal graphite stresses. An operator formalism is being used, and this has been completed in first order for cylindrical geometries. This geometry is more constrictive than slab geometries and, hence, represents a better model for testing the power of the calculational procedure. The constraints on the forcing function (radiation- and temperature-induced dimensional changes) are being evaluated. Upon completion of the latter, the method will be extended to slab geometries, and the effect of boundary conditions will be considered.

6.5.7 Control System Assessment — S. J. Ditto

During the reporting period a study of the German PBR concepts was begun, based on GE documentation.¹⁰ Concurrently we began a study of the ERDA report⁵ on the direct-cycle gas-cooled reactor plant as a basis for a comparative assessment of the pebble and prismatic cores.

On November 12, E. P. Epler and S. J. Ditto met with several GE engineers at Sunnyvale for a briefing by GE. Previous gas-cooled reactor work by GE was discussed as well as future plans for the core evaluation studies. Preliminary information should be available early next quarter regarding the definition of the PBR control system design and mechanisms.

6.5.8 Seismic Effects — W. C. Stoddart

The choice of a prismatic or a pebble-bed reactor core leads to significantly different designs for the core, core support structures, and associated equipment such as fuel-handling machines. In the United States, seismic response is of particular importance in safety and licensing areas for nuclear reactors. The purpose of the seismic study is to assess the state of knowledge of the design basis for both core concepts. This assessment will then lead to a classification of those design areas for which knowledge is adequate or inadequate. The areas of knowledge that are judged to be inadequate will require further investigation if that particular core concept is to be implemented. The goal of the study is to identify and document this state of knowledge and to estimate the cost to arrive at an adequate basis for design.

During the first quarter of the study, a work scope was prepared, the literature available at ORNL was reviewed, visits were made to GA and GE, and a list of significant design features was started.

During the next quarter, a tentative report of the study findings will be prepared, based on the information available for review. Current activities at GA and GE are not expected to allow final conclusions to be reached in time for the March 1980 report to DOE.

6.5.9 Reactor Availability — W. C. Stoddart

The economic attractiveness of an HTR for thermal or electrical supply is strongly impacted by the operational characteristics of the plant. It would generally be preferred for the plant to produce power

on an almost uninterrupted basis. This high availability goal is affected by the number of scheduled and unscheduled outages. The prismatic and pebble-bed approaches to HTR core design result in different refueling and maintenance approaches. The purpose of this study is to assess the two design concepts to determine if there are significant differences in expected availability. If such areas are identified, the impact on economics may be determined.

During the first quarter of this study, a work scope was prepared, the literature available at ORNL was reviewed, and visits were made to GA and GE. The next quarter will concentrate on scheduled outages. The relative advantages and disadvantages of on-line refueling will be examined, and a tentative report on expected availability for the two concepts will be prepared. The question of unscheduled outages is not expected to be addressed in sufficient depth for conclusions to be included in the March 1980 report to DOE.

6.5.10 Overall Capital Costs — J. G. Delene and M. L. Myers

Estimates were made of the capital costs by cost category for the steam cycle, direct cycle, and process heat HTGR. These costs included both best estimates and expected cost ranges. All costs were in January 1979 dollars for 3000-MW(t) systems.

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