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IRRADIATION TESTING IN THE DEVELOPMENT OF FUEL ELEMENTS FOR THE GAS-COOLED FAST BREEDER REACTOR*

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The fuel rods for the Gas-Cooled Fast Breeder Reactor (GCFR) are vented through a pressure-equalization system to a helium purification system; thus the pressure differential across the fuel-rod cladding is very small (~40 lbf/in.^{2}(psi)). With this feature, it is possible to make the design of the GCFR fuel rods similar to that of the Liquid Metal Fast Breeder Reactor (LMFBR) fuel rods, which are sealed rods that have a small pressure differential across the cladding at the beginning of life. As the fuel-rod materials and many of the operating conditions are also very similar, LMFBR technology can be used extensively for the GCFR fuel-rod development and only tests specific to the GCFR need be performed. Features related to venting of the fuel rods, such as the use of charcoal traps, and those related to the use of the helium coolant, such as roughening the cladding surface to improve heat transfer to the coolant, are being tested.

An irradiation program for the development of fuel elements for the GCFR is being conducted using the Oak Ridge Research Reactor (ORR) for thermal-flux irradiation of vented fuel rods and the Experimental Breeder Reactor (EBR-II) for fast-flux irradiation of sealed rods. The irradiation program is a cooperative effort among Gulf General Atomic (GGA), Argonne National Laboratory (ANL), and Oak Ridge National Laboratory (ORNL).

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THERMAL-FLUX IRRADIATIONS

Irradiation Capsule GB-9

The vented, sweep-gas, fuel-rod capsule GB-9 was irradiated in the ORR at 15 kW/ft and 685°C ±15°C cladding outside temperature to an exposure of 54,000 MWd/t.

The GB-9 capsule was designed to evaluate the overall performance and adequacy of the GCFR vented and pressure-equalized fuel rod. Test conditions were typical of a GCFR fuel rod, except for the fast-neutron exposure. Measurements were made of the fission-product release from the charcoal trap and blanket regions of the rod during the irradiation. The experimental results obtained are providing a basis for much of the GCFR vented fuel rod and pressure equalization system design. The capsule and fuel-rod design, planned operating conditions, and experimental results obtained early in the irradiation have been reported previously [1].

Details of the GB-9 fuel rod and test conditions are given in Table I. The fuel rod and its charcoal trap were normally operated under steady-state conditions. Sweep-gas flow was maintained normally across the top of the trap at all times when the fuel rod was at power. Sweep-gas samples were taken periodically and analyzed by gamma-ray spectrometry to determine isotopic fission-gas release rates. Fission-product release from the rod was monitored with the sweep gas flowing either across the top of the trap, which was the normal flow mode, or upward through the bottom of the trap.

The sweep flow rate, normally 150 to 250 cm^3 s.t.p./min during non-sampling periods to conserve helium, was increased to 1300 cm^3 s.t.p./min (~19 cm^3/min at 1000 lbf/in.^{2}-gauge) prior to sampling. At the sampling flow rate of 1300 cm^3 s.t.p./min, the sweep-gas transit time from the fuel rod to the sampling point was only 47 sec, thereby making analysis for short-lived fission gases possible.

Although the capsule was operated under steady-state design conditions most of the time, special tests were also performed to determine fission-gas release dependence on charcoal trap temperature, fuel region temperature, and sweep pressure. Tests to obtain information on fission-gas release during pressure cycling, fission-product decay heating in the charcoal trap, and iodine deposition in the trap were also made.

Steady-state Fission-gas Release versus Burnup. The results for fission-gas release measurements made under steady-state conditions during full-power operation are shown in Fig. 1 for sweep flow across the top of the rod. The fractional release values, i.e., release rate to birth rate (R/B), showed a general increase up to about 10,000 MWd/t of heavy metal burnup (~88 d) and then remained about constant. Calculated predictions showed that the charcoal trap would be less effective than the blanket region in delaying active fission gases, because the integrated mean free path for diffusion is greater in the rod trap. The effectiveness of the charcoal trap in reducing the steady-state fission-gas release rates was shown to be a function of the half-life of the isotopes, as expected. The trap reduction values were found to be reasonably close to those predicted.

Fission-gas Release during Slow Pressure Cycling. Several slow-pressure-cycling tests were conducted at a burnup level of about 7500 MWd/t.

Table I

TEST CONDITIONS FOR CAPSULE GB-9

Fuel rod number	GA-20
Cladding material	316 SS, annealed
Cladding OD	0.355 in.
Cladding thickness	0.024 in.
Cladding OD/ID	1.157
Bond gap fluid	Helium
Cladding OD surface temperature	700°C (+0°C, -30°C)
Fuel	$(U,Pu)O_2$
Smear density	~85% of theoretical
Linear power generation (max)	15 kW/ft
Burnup exposure	~50,000 MWd/t (15×10^{20} fissions/cm^3)
External pressure	975 lbf/$in.^2$ gauge
Initial internal trap pressure, hot	1000 lbf/$in.^2$ gauge
End-of-life internal trap pressure, hot	1000 lbf/$in.^2$ gauge
Trap temperature, surface	300°C ±50°C
Pressure differential, fuel-rod wall	25 lbf/$in.^2$ nominal, not to exceed 300 lbf/$in.^2$ whenever capsule is at power
Number of power (temperature) cycles	100 standard: 700°C → 300°C → 700°C 6 nonstandard: 700°C → x → 700°C
Number of pressure cycles	22 standard: 1000 lbf/$in.^2$ gauge → 100 lbf/$in.^2$ gauge → 1000 lbf/$in.^2$ gauge 4 nonstandard: 1000 lbf/$in.^2$ gauge → x → 1000 lbf/$in.^2$ gauge

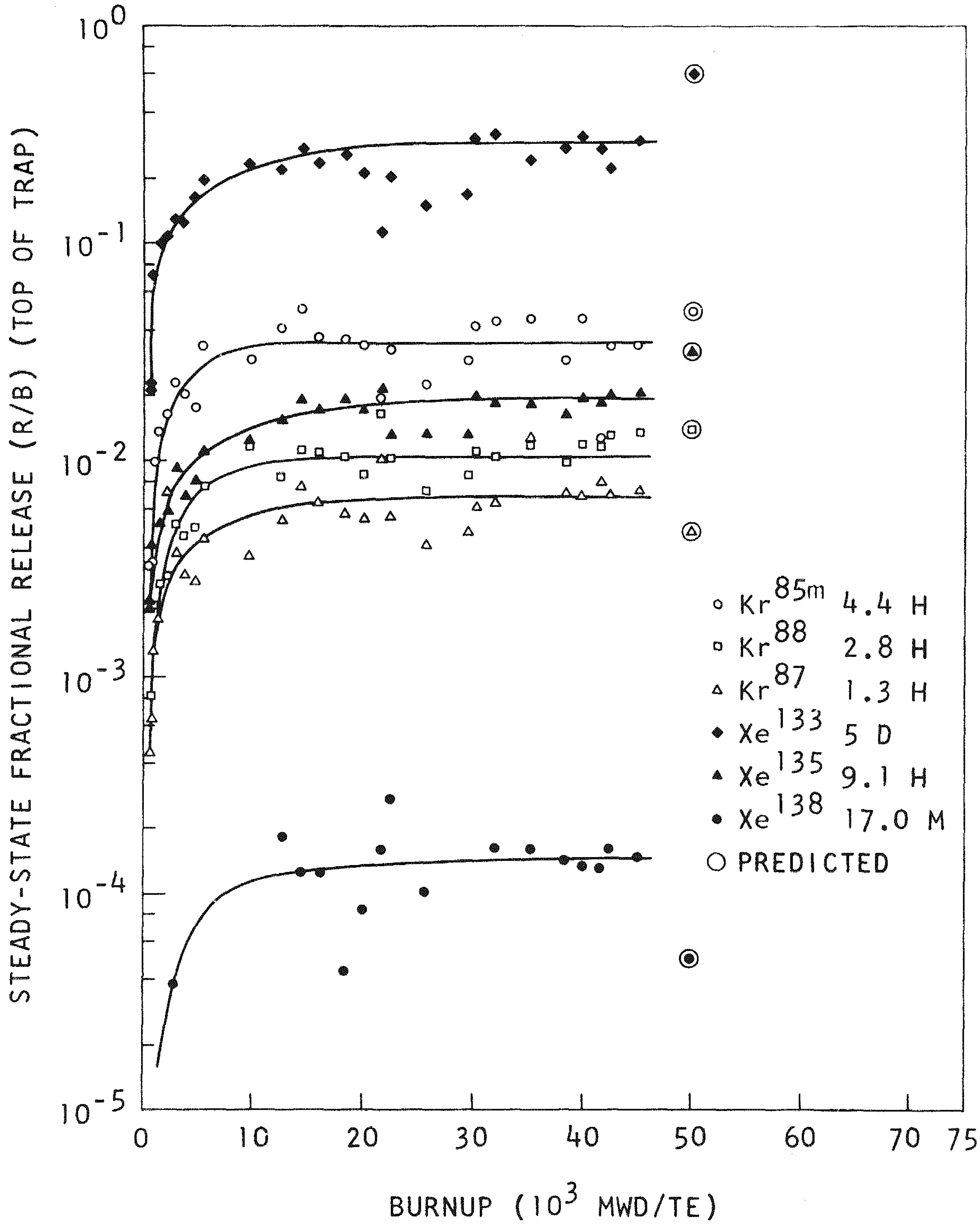

Fig. 1 Measured fission-product gas release from the vented fuel rod in capsule GB-9 as a function of burnup

Each of these tests was conducted with the sweep flow across the top of the trap starting from steady-state levels. During each depressurization, the volumetric flow rate of sweep gas past the radiation monitor was approximately constant at 19 cm^3/min as a consequence of the approximately exponential pressure decay. The response of the radiation monitor was interpreted as changes in the gross gamma activity release rate from the fuel rod, provided the deposition activity in the sweep line was small compared with the gaseous activity.

The release behavior during these tests is discussed in Ref. 1. Although all of the detailed variations in activity levels observed during the depressurizations are not fully understood, it appears that the observed increased activity was associated primarily with displacement of the concentrated gaseous activity in the trap at the start of depressurization by the mixture of the gas expanding from the lower inlet sweep line ($\sim$68 cm^3) and from the free volume in the fuel rod below the trap ($\sim$2.4 cm^3). In general, the release behavior during these slow pressure-cycling tests with the sweep gas flowing across the top of the trap agreed with expectations once the flow conditions within the rod were properly evaluated.

Fission-gas Release versus Charcoal-trap Temperature. A special test was conducted at a burnup level of $\sim$35,000 MWd/t to determine the effect of temperature level of the charcoal trap and upper blanket region on fission-gas release. In this test, steady-state fission-gas release rates were measured at charcoal trap temperatures of 200°, 300°, and 400°C while the fuel region peak power was held constant at 15 kW/ft. The effluent sweep-line activity levels indicated by the radiation monitors were reasonably consistent with the gas sample results; in going from 200° to 400°C trap temperature, the line activity increased by a factor of 2 when the sweep flow was across the top of the trap and by only 25% when the sweep flow was through the trap.

Fission-gas Release versus Fuel-rod Power and Temperature. Steady-state fission-gas release was measured as a function of fuel-rod power and temperature in special tests conducted at a burnup level of $\sim$36,000 MWd/t and at a burnup level of $\sim$43,000 MWd/t. These tests were conducted at peak cladding outside temperatures ranging from 550° to 685°C (fuel-rod power ranging from 12 to 15 kW/ft) while the charcoal trap temperature was held constant at 300°C. The gas-sample results and the indicated sweep-line activity levels were consistent in these tests; both showed an increase in fission-gas release of a factor of 10 in going from a peak cladding temperature of 550° to 685°C. The results of these tests, together with the results of the trap temperature test, show the fission-gas release from the rod to be much more sensitive to cladding temperature changes and temperature profile changes over the fuel region than to the temperature changes of the charcoal trap and blanket region of the rod.

Fission-gas Release versus Sweep Pressure. Attempts were made to separate the delay in release of fission products from the fuel matrix from the delay due to gas-phase diffusion by measuring the release at lower pressure (500 lbf/in.^{2} gauge). The scatter in the data was so large that determination of the solid-state release rates could not be made.

Postirradiation Examination. Upon termination of the GB-9 experiment, gamma scanning for fission-product distribution was performed [2]. The volatile fission products ^{137}Cs and ^{131}I moved somewhat from the fueled region in both the radial and axial directions. Peaking of volatile fission-product activity was observed at the fuel-cladding interface and at the fuel-blanket interfaces. Some volatile fission-product activity was found in the blanket region (most born *in situ*), but none was found in the charcoal trap. Typical volatile fission-product gamma-scan results are shown in Fig. 2. Dimensional measurements showed a maximum diametral strain of the fuel-rod cladding of 0.3%.

Postirradiation helium flow testing of the GB-9 fuel rod was performed to determine if irradiation had changed the resistance to flow through the rod passages. The resistance of the charcoal trap and upper blanket was essentially unchanged by irradiation and the flow through the fuel region was found to be sufficient to easily accommodate pressure equalization of the cladding.

Electron microprobe analysis of a transverse metallographic sample taken 4-3/16 in. above the bottom of the fuel column was made. Radial point-counting traverses across the fuel for zirconium, cesium, uranium, and plutonium showed distributions of these elements that substantiate the calculated radial power production. Slightly more cesium was found on the cold side than on the hot side, indicating that cesium migrated to the cooler side of the rod, whereas the zirconium and plutonium profiles indicated a higher burnup on the hot side. The most severe cladding attack in the mixed-oxide fueled region occurred also on the cold side of the rod at this axial position, which was 4-3/16 in. above the bottom of the fuel column.

Optical metallography was performed on five axial positions along the fuel region of the GB-9 rod. Attack of the cladding adjacent to the fuel was observed in each section except the one taken at the bottom of the rod. The maximum attack penetrated approximately 0.004 in. into the cladding in the fueled region. The attack was intergranular at its leading edge, but all of the grains were consumed by the reaction nearer the fuel-cladding interface. Examination of the longitudinal sections taken at the ends of the fuel column indicated that relatively large amounts of fission-product cesium were present adjacent to the cladding. At the bottom of the fuel column where the cladding temperatures were relatively low, less than 500°C, no attack occurred in that region of the rod. The maximum attack occurred in the region of the 14.9% enriched UO_2 half-pellet at the top of the mixed-oxide fuel column, as shown in Fig. 3. These enriched half-pellets are used to suppress power peaking at the ends of the fuel rods in the thermal flux. Figure 3 is a composite of photomicrographs of the top longitudinal section of the rod and shows that the central hole had closed with porous fuel material at the top of the 14.9% enriched UO_2 half-pellet. The fuel pellets (the 14.9% and the 8.3% enriched UO_2 pellets and the mixed-oxide pellets) were fabricated with a 0.060-in.-diam central hole.

Based on the microprobe analysis, closing of the central hole appears to have resulted from fuel vaporization in the hotter regions of the rod and condensation in the cooler regions at the ends of the fuel column. The central hole was enlarged and displaced toward the hot side of the fuel rod except at the bottom of the column of mixed-oxide fuel pellets. At the axial position where the constricting deposit occurred, cesium was found only in the outer ~1/3 of the fuel and not in the deposit. Cesium was also found in axial locations above the constrictions where temperatures during irradiation were lower than in the fueled region.

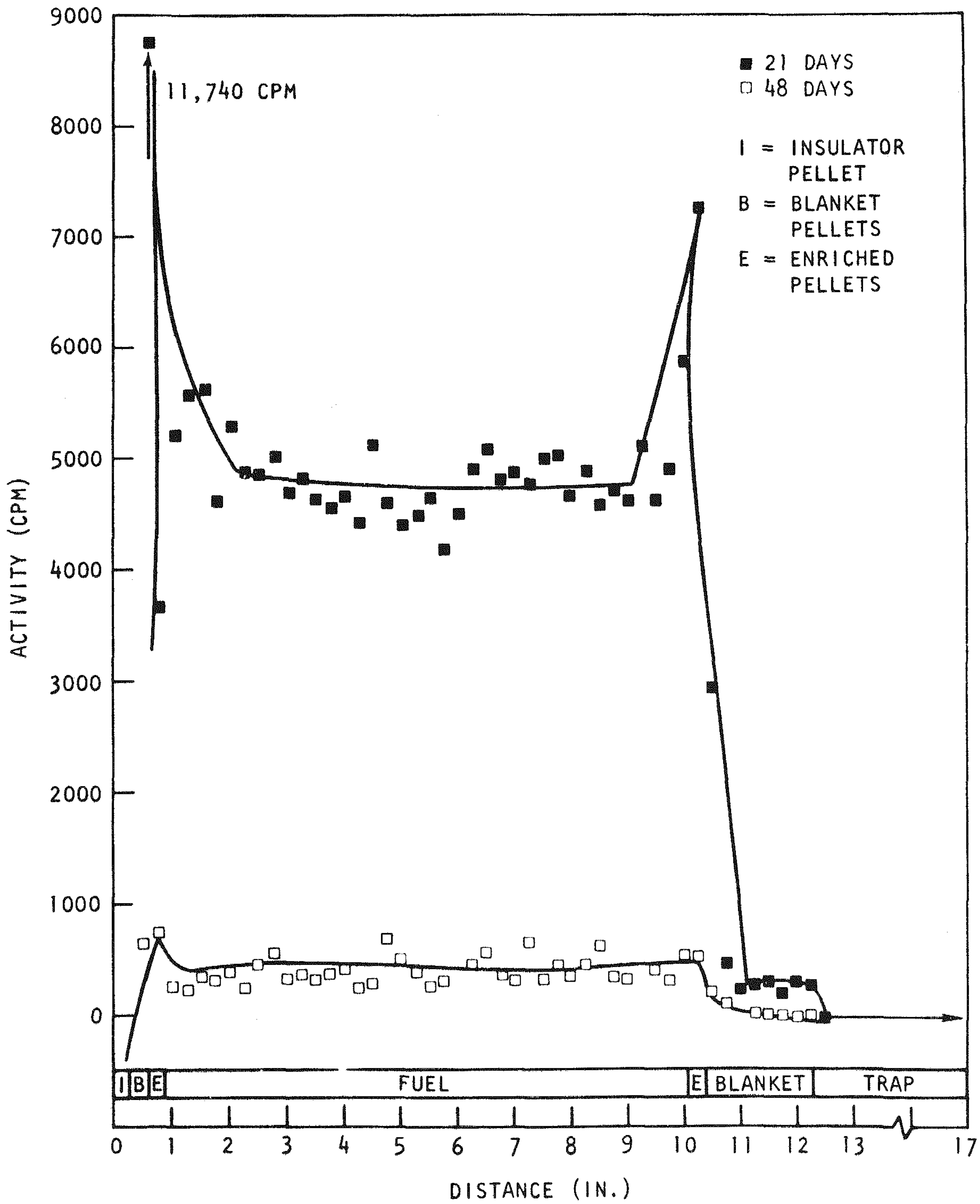

Fig. 2 Axial scan of capsule GB-9 for ^{131}I

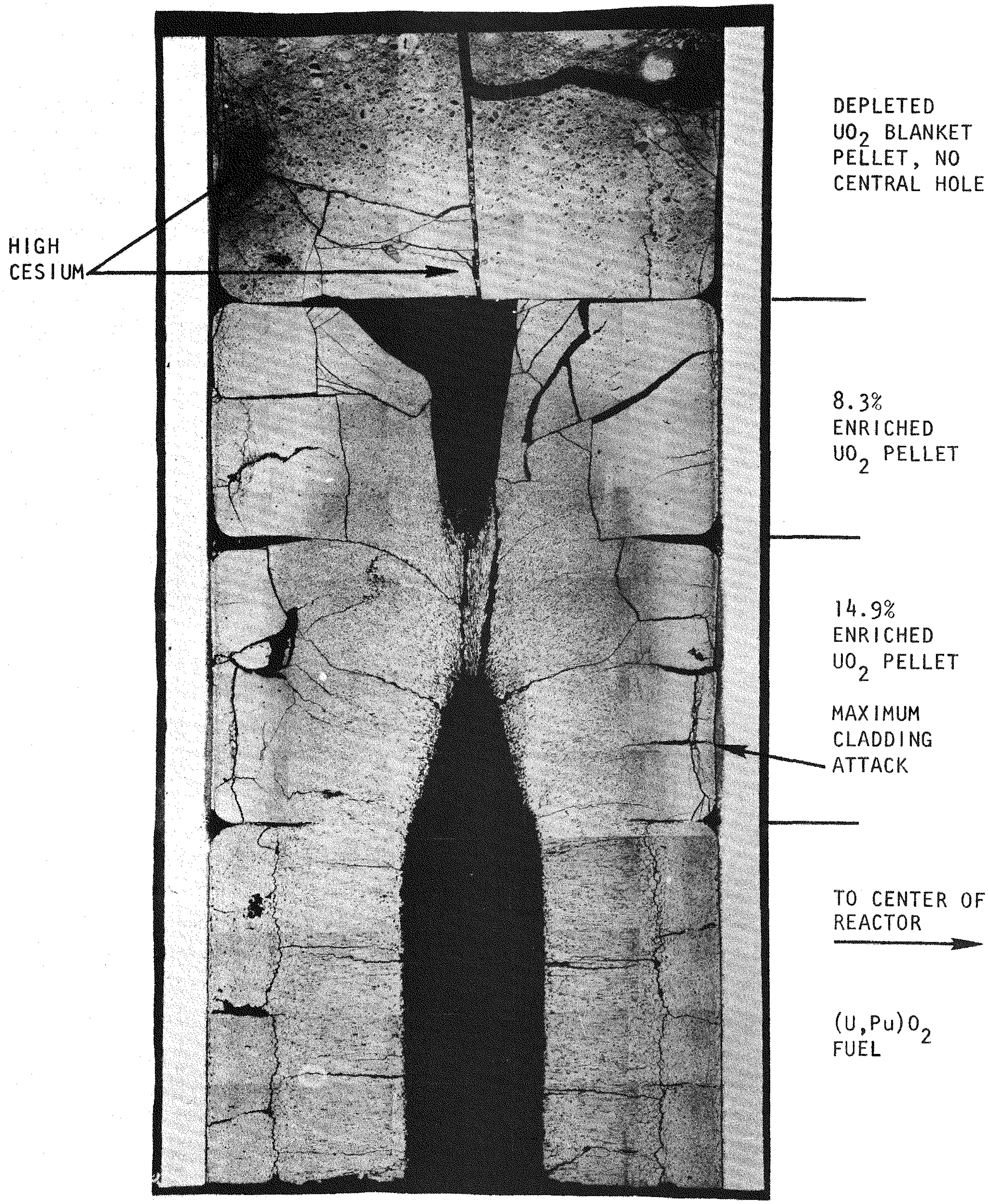

Fig. 3 Longitudinal section of the GB-9 fuel rod at the top of the fuel column

To determine the effect of the irradiation on the properties of the cladding, biaxial stress-rupture tests at 700°C were performed on a section of the Type 316 stainless steel cladding of the GB-9 fuel rod (taken from the fueled region) and on two unirradiated tube samples of the same cladding material. The amount of loss in rupture life and ductility agrees with values reported in the literature for SA 316 stainless steel at comparable fluence levels.

Irradiation Capsule GB-10

The GB-10 vented sweep-gas fuel-rod capsule has been irradiated to a burnup of 17,000 MWd/t. The rod is being operated initially at a linear heat-generation rating of 12 kW/ft until steady fission-gas release conditions are attained. After this initial period, the heat-generation rate will be increased, possibly in two steps, to 14.8 kW/ft, at which level it will continue for the remainder of its exposure to a burnup goal of 75,000 MWd/t.

Capsule GB-10 is similar to the capsule GB-9 experiment described above. From the experience gained in the design and operation of capsule GB-9, capsule GB-10 was designed with increased capability to measure fission-product release and transport and to allow simulation of a leaking vented fuel rod.

The design of the fuel rod in capsule GB-10 is similar to the GB-9 fuel rod, except that the outer surface of the cladding is roughened and solid $(U,Pu)O_2$ fuel pellets rather than annular pellets are used. The fuel stack height is slightly less than in the GB-9 rod, the upper blanket region of depleted UO_2 pellets is one pellet longer, and the charcoal trap is shorter (1 in. long instead of 3 in.). The charcoal trap was shortened to 1 in. to provide the same potential fission-product loading as the rod trap in full-length GCFR fuel rods, i.e., the GB-10 trap contains the same ratio of charcoal mass to power generated within the rod.

The design of the GB-10 capsule is similar to the GB-9 capsule except for the addition of sweep-gas lines at the bottom of the fuel and blanket regions. The sweep-gas lines and valving arrangement for capsule GB-10 are shown in Fig. 4. With these connections and valving, it is possible to measure fission-gas release from each of the three main regions of the rod (trap, blanket, and fuel) individually or in combinations. This increased capability for measuring fission-product release and transport permits studying release mechanisms and transport times in greater detail than was possible with capsule GB-9. By directing the sweep gas through the fuel region, for example, fission-gas release rates from the oxide fuel matrix can be determined. Sweep-gas flow through all three regions simulates a leak in the cladding of a GCFR rod and thus such effects as fission-product decay heating in the charcoal trap can be observed.

The data on the active fission-gas release from the GB-10 fuel rod during operation at the 12 kW/ft power level with several flow modes have been analyzed. A comparison between release fractions obtained in GB-9 and those obtained in GB-10 for the flow mode in which effluent was sampled from the top of the trap is shown in Fig. 5. It is apparent that the release fractions from capsule GB-10 operating at 12 kW/ft are significantly lower than those from capsule GB-9 operating at 14.8 kW/ft. It should also be noted that although the activity levels in GB-10 at 12 kW/ft have not reached the saturation value after 10,000 MWd/t burnup, steady values had essentially been reached in GB-9 at the same burnup level.

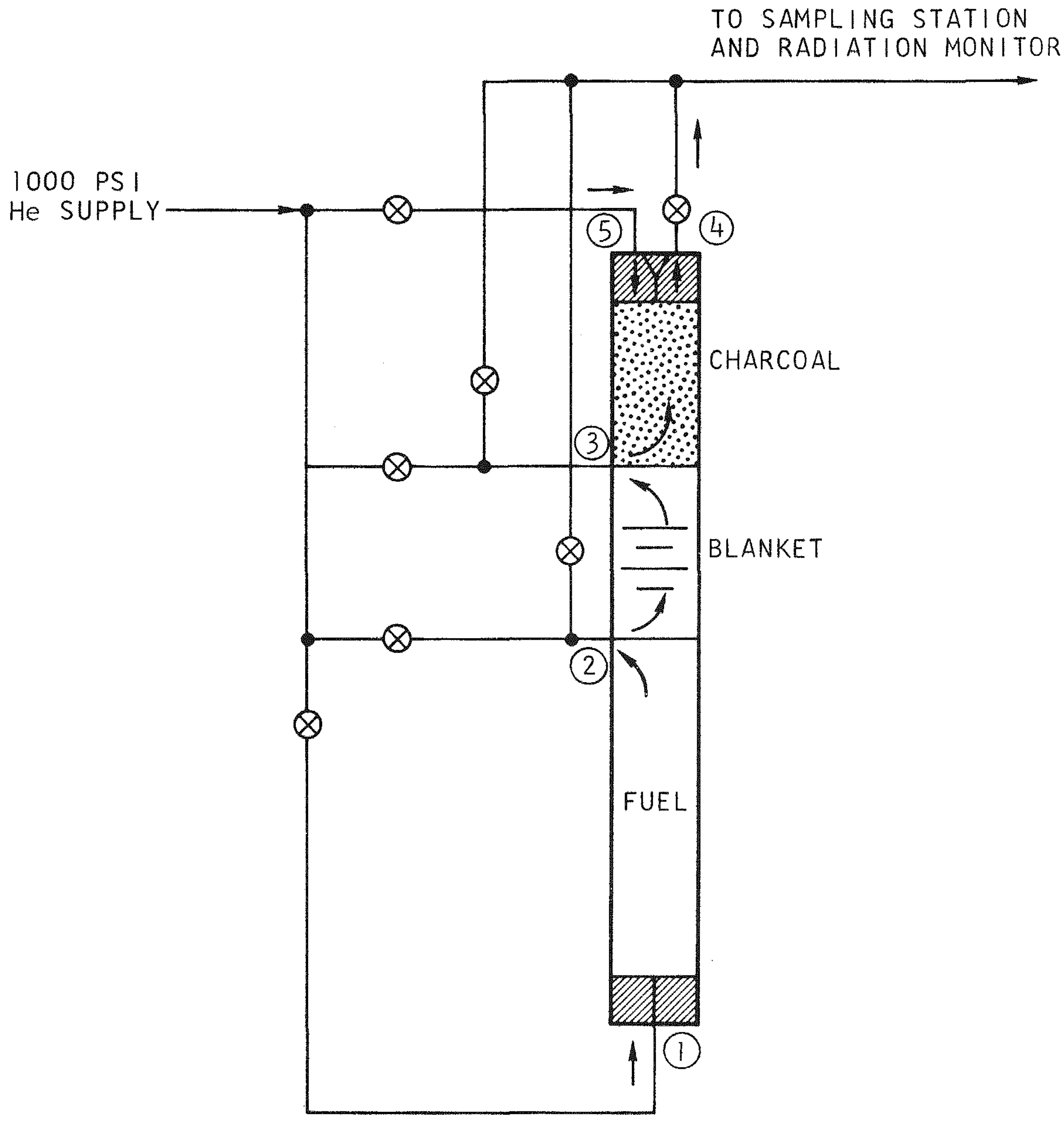

Fig. 4 Helium sweep- and monitoring-line arrangements for capsule GB-10

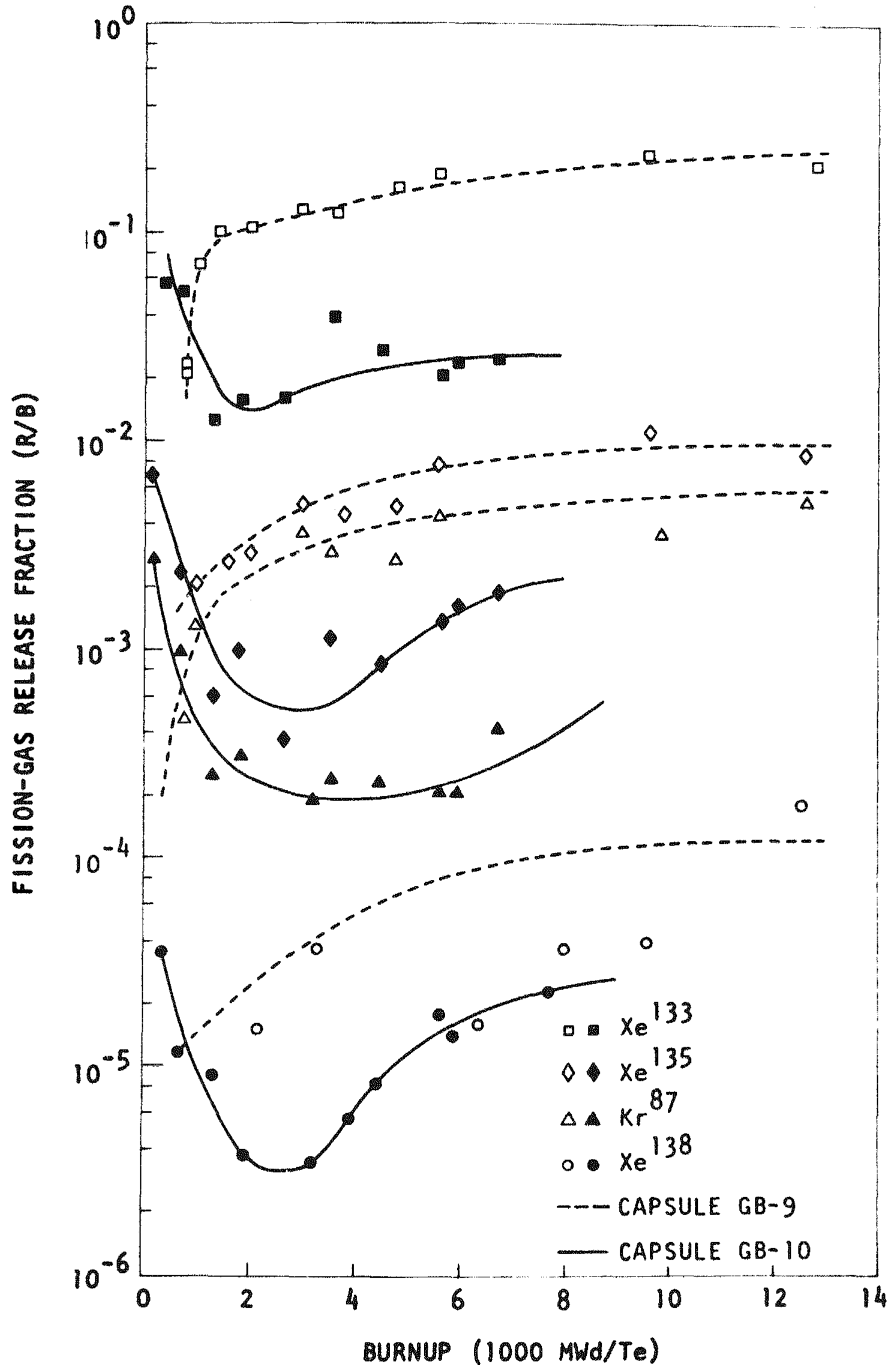

Fig. 5 Comparison of fission-gas release during early irradiation of fuel rods in capsules GB-9 and GB-10

The release of all of the isotopes for which measurements were obtained from capsule GB-10 decreased during the first 30 d of irradiation (~2,900 MWd/t) and then began to increase. During the initial irradiation period, most of the fissions occur in the outer periphery of the fuel and in the region where 50% of the volume of the fuel exists, which is beyond 70% of the fuel radius, because of self-shielding effects in the thermal flux in the ORR. The initial decrease in release is also thought to be due to the formation of a central hole in the fuel (solid pellets initially) and the closure of the fuel-cladding gap. It is hypothesized that the formation of the central hole and closure of the gap lowered the central and overall fuel temperatures and resulted in a lowered fractional release. The release fractions for the various isotopes in the sweep-gas flow mode through the top of the trap in comparison with those from the flow mode in through the bottom of the fuel and out through the bottom of the blanket are very different for the short-lived isotopes (e.g., ^{138}Xe shows a factor of 200 greater release in the latter mode), whereas the release fractions for the longer-lived isotopes (e.g., ^{133}Xe) are very nearly the same for both flow modes, which is in agreement with expectations.

FAST-FLUX IRRADIATION EXPERIMENTS

F-1 (X094) Irradiation Experiment

The general objective of the GCFR fast-flux experiment F-1 (X094) in the EBR-II is to study fuel-rod specimens over a wider range of fuel temperatures and to a higher range of cladding temperatures than heretofore carried out in the LMFBR program. Variables kept constant are the cladding material (Type 316 stainless steel, 20% cold-worked), the fuel (mixed $(U,Pu)O_2$), and the fuel fabrication technique. These fixed test conditions are summarized in Table II. The temperature range overlaps that already being studied under the LMFBR program to provide a point of commonality for comparison of results. The reference condition selected is the same as that used in the thermal irradiation of capsules GB-9 and GB-10 in the ORR so that a direct comparison can be made of a fast-flux versus a thermal-flux test environment.

The standard EBR-II B-7 core subassembly was chosen as the design basis because it provided space within each capsule for a thermal barrier to raise the cladding temperatures to the desired levels.

Table II

FIXED TEST CONDITIONS FOR THE F-1 EXPERIMENT

Cladding	316 SS, 20% cold-worked
Cladding OD	0.300 in.
Cladding OD/ID ratio	~1.15
Linear power generation (fission)	~15 kW/ft
Fuel pellets, sol-gel derived	85 wt-% UO_2, 15 wt-% PuO_2
Fuel smear density	85% of theoretical
External pressure	Ambient
Internal helium pressure, hot, beginning of life	~28 lbf/in.^{2} abs.

Active charcoal fission-product traps, which are in communication with the fuel through the vapor phase, will provide information on the ability of hot charcoal to remove condensable fission products and to delay short-lived gaseous fission products in the fast-flux environment. In addition to the active charcoal traps, two sealed containers of the same charcoal were inserted at the ends of each capsule to assess the effects of the fast neutron environment on the sorption properties of charcoal, apart from effects due to fission-product loading, by comparison of preirradiation and postirradiation measurements. Passive temperature and flux monitors are used to define the actual experimental environment.

The first seven encapsulated fuel rods in the F-1 (X094) subassembly started irradiation in EBR-II in March 1971. Interim nondestructive tests were performed at 27,000 MWd/t burnup exposure and at 52,000 MWd/t. One rod (G-3) was removed and replaced during the examination at 27,000 MWd/t. Destructive postirradiation examinations are being carried out to study the influence of the test variables on integral rod behavior and on individual components, including the activated charcoal fission-product traps. Five additional rods were removed when a nominal burnup of about 52,000 MWd/t was reached and were replaced with five new fuel-rod capsules for irradiation to the termination of the experiment.

Gamma spectrometric scanning of the fuel-rod plenums for ^{133}Xe during the interim examinations (i.e., at 27,000 MWd/t and 52,000 MWd/t) showed that the integrity of the cladding on all the rods was maintained. Detailed diametral and axial gamma scanning has been conducted at selected locations. Volatile fission-product peaks have been observed in the pellet-cladding gap at both fuel-blanket interfaces. The scanning results at 27,000 MWd/t, which were reported in Ref. 2, are similar to those observed in capsule GB-9, described above.

Postirradiation examination of the first rod removed is in progress. Dimensional measurements of rod G-3 showed a maximum of 0.3% total diameter change during irradiation to 27,000 MWd/t. Metallographic examination of rod G-3, which was operated at a maximum cladding outside temperature of 700°C, revealed no more than 0.0003 in. attack of the inside of the cladding by fuel and fission products.

F-3 Irradiation Experiment

The F-3 fast-flux irradiation experiment is to be conducted in EBR-II for the purpose of extending the plutonium-oxide, uranium-oxide fuel-rod irradiation technology being developed under the LMFBR program to conditions that are more specific to the GCFR. In particular, since the GCFR will employ a vented fuel rod, rather than a sealed fuel rod as does the LMFBR, the F-3 fuel rods are designed with large fission-gas plenums so that pressure buildup will result in a relatively small pressure differential across the cladding and thereby simulate the venting design as closely as is practical in EBR-II tests. In addition, the LMFBR tests to date have achieved maximum cladding fluences of $\sim 1 \times 10^{23}$ n/cm^2 at ~100,000 MWd/t burnup in EBR-II tests, whereas the F-3 experiment fuel-rod cladding will receive maximum fluences up to $\sim 1.5 \times 10^{23}$ n/cm^2 at the 100,000 burnup exposure. The F-3 fluences will be very near the maximum fluences to be experienced by fuel-rod claddings in the GCFR ($\sim 1.8 \times 10^{23}$ n/cm^2).

The GCFR F-3 experiment will share a J19A subassembly with an ANL

Group-08 LMFBR high-temperature chemistry experiment. The initial loading of the subassembly will contain ten F-3 fuel-rod capsules and nine Group-08 fuel-rod capsules.

The general objective of the F-3 experiment is to extend the study of fuel-rod behavior initiated in the F-1 (X094) experiment: (1) to higher fast fluences; (2) to lower oxygen-to-metal (O/M) ratio at higher temperatures than studied previously; (3) to observe the effect of variations in the O/M ratio on the release of volatile species, especially cesium, from the fuel pellets; (4) to observe the effect of the release of volatile species (or precursors thereof) from the fuel and their sorption in the activated carbon traps on the fuel—fission-product—cladding compatibility with cladding surface temperatures of 675° and 750°C; and (5) to observe the effect of varying fuel density. Variables that will be kept the same as in the F-1 (X094) irradiation experiment are: (1) the cladding material (Type 316 stainless steel, 20% cold-worked, except possibly with improved cladding), (2) the fuel (mixed $(U,Pu)O_2$), and (3) the fuel fabrication technique.

The F-3 experiment will continue to emphasize the performance of the GCFR fuel rods at cladding temperatures higher than those presently included in the LMFBR program while maintaining an overlap of temperatures to permit comparison with data generated in the LMFBR program. Irradiation of the F-3 fuel-rod capsules is scheduled to begin in early 1974.

CONCLUSIONS

From the results of the two GCFR vented fuel rods, GB-9 and GB-10, irradiated in the ORR, the following conclusions can be made:

1. Pressure equalization has been maintained over the range of pressures, temperatures, and rod power densities representative of GCFR operating conditions.

2. Volatile fission products have not been observed beyond the fuel end of the blanket region in the GB-9 rod, which operated with no simulation of a cladding leak. This means that for normal operation up to the maximum burnup maintained in the GB-9 rod, i.e., 54,000 MWd/t, the fuel-rod charcoal trap was not required to limit the transport of volatile fission products out of the fuel rod.

3. No significant dimensional change was observed for the fuel rod irradiated to a burnup of 54,000 MWd/t.

4. Attack of the cladding by fuel or fission products is in agreement with that observed in sealed rods tested under the LMFBR program.

5. The observed gaseous fission-product releases and transport rates under steady-state conditions have verified the models being employed for systems analysis.

6. The behavior of the gaseous fission products during transients of temperature, power, and pressure has been in agreement with analytical predictions.

7. Solid-state migration and gas-phase interdiffusion limit the rate of the release of gaseous fission products during normal operation. However, when a simulated leak is introduced, convective transport becomes dominant.

8. The delay of gaseous fission products by the rod charcoal traps was as predicted over a temperature range simulating both normal and off-design GCFR operation.

The fast-flux testing of the sealed rods is still in progress, and although in-process information is not available as with the vented rods, several conclusions may be reached from the data obtained thus far.

1. Preliminary results indicate that the six fuel rods irradiated with maximum cladding temperatures in the range of 600° to 800°C to burnup exposures up to 52,500 MWd/t have remained intact. This indicates that considerable margin may exist when the fuel-rod cladding peak hot-spot temperature is maintained at or below 700°C.

2. The rod that was irradiated at a temperature of 700°C (calculated) to a burnup of 27,000 MWd/t showed (a) a maximum diametral change of 0.3%, (b) a maximum cladding attack penetration of 0.0003 in., and (c) no volatile fission-product transport beyond the fuel end of the blanket region to the traps or plenums, which is in agreement with the results of the GB-9 vented rod irradiated in the ORR.

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