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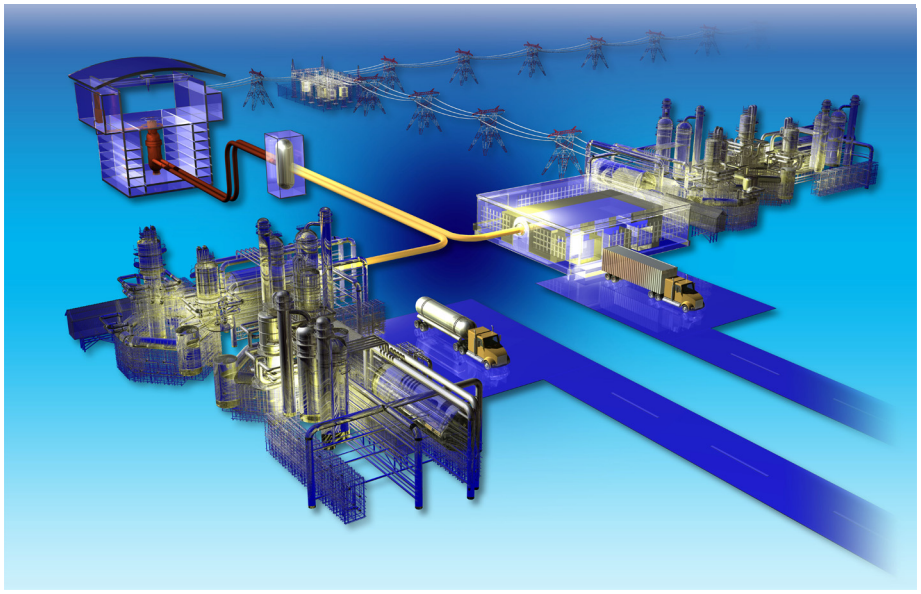
Technical Evaluation Study

Project No. 23841

Moisture Ingress from Direct Cycle Steam Generation—Effect on Fuel Performance and Fission Product Transport Technology Development



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MOISTURE INGRESS FROM DIRECT CYCLE STEAM GENERATION—EFFECT ON FUEL PERFORMANCE AND FISSION PRODUCT TRANSPORT TECHNOLOGY DEVELOPMENT	Identifier: TEV-583 Revision: 0 Effective Date: 07/06/09	Page: 1 of 32

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Signatures

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1. INTRODUCTION

The Next Generation Nuclear Plant (NGNP) Project is considering modifying the prismatic core reactor design from an indirect cycle gas turbine to a configuration with a steam generator in the primary loop feeding a steam turbine in a secondary loop. The revised configuration would substantially increase the likelihood and magnitude of moisture ingress events in the primary system. Since the fuel development and qualification program was developed based on the gas turbine concept, the scope of the program could be altered by the change in design configuration. This study evaluates the need for additional fuel development and qualification scope to address conditions associated with moisture ingress events.

The Modular High Temperature Gas-Cooled Reactor (MHTGR) development program conducted by the U.S. Department of Energy (DOE) in the 1980s and early 1990s originally focused on a design with a steam generator in the primary loop. Substantial design description, safety analysis, and licensing documentation were produced for this design, and serve as a basis for assessing the moisture ingress events and conditions that would likely result for a direct steam cycle NGNP. These events and conditions were reviewed and compared to the available data from testing conducted to address moisture ingress effects on fuel performance and fission product release.

Earlier testing had shown that the radionuclide retention characteristics of intact tri-isotropic (TRISO) particles are unaffected by moisture ingress, thus the focus of low-enriched uranium (LEU) TRISO testing was on the effect of moisture on the release characteristics of particles with exposed kernel defects. Fuel irradiation with simulated moisture ingress conditions were conducted on uranium oxycarbide (UCO) TRISO and ThO_2 fuel compacts containing simulated UCO defective particles in the High-Flux Isotope Reactor (HFIR) reactor at Oak Ridge National Laboratory and in the High-Flux Reactor (HFR) reactor at the Joint Research Centre – Petten, the Netherlands. Additional postirradiation moisture ingress testing was conducted on fuel irradiated in the HFR. In addition, moisture ingress testing was conducted on a German UO_2 TRISO fuel sphere having known particles with exposed kernel defects. While UCO response to moisture differs considerably from UO_2, the UCO becomes predominantly UO_2 as a result of irradiation and exposure to moisture, thus the German fuel testing is relevant to UCO fuel.

The effect of moisture ingress on metallic fission product release was not measured in the in-pile testing, but some data were obtained based on comparison of postirradiation metallic fission product distributions in fuel compacts and graphite fuel bodies with and without moisture injection during irradiation. The results indicated that release of metallic fission products from the fuel compact and surrounding graphite may be increased by moisture ingress.

The data produced by the moisture ingress testing at HFIR and HFR have been used to develop analytical models for predicting gaseous fission product release under moisture ingress conditions. These models are applicable to the NGNP fuel, subject to confirmation of similar behavior as observed in existing testing under selected conditions. However, application to the calculated range of conditions for the MHTGR, and expected for the NGNP has required extrapolation of the data to water partial pressures well above the range of existing data. Models have not been developed to predict the effect of moisture ingress on metallic fission product release.

The technical program plan for NGNP fuel development and qualification[i] was established based on a direct cycle gas turbine power conversion system design. The NGNP Project recently reviewed power conversion system options,[ii,iii] resulting in recommendations for a steam generator in the primary loop of the prismatic core plant design. This design change will significantly increase the likelihood and potential amount of moisture entering the primary system, thus the scope of the fuel program must be reviewed to determine whether additional testing is needed.

While the effects of increased moisture ingress associated with the design change must be taken into account, high temperature gas-cooled reactors (HTGRs) are generally tolerant of moisture as demonstrated by experience with the Fort St. Vrain Generating Station. A total of 29 moisture intrusion events were experienced during its operation from sources including thermal hydraulic moisture out-gassing of reactor internals components, tube leaks, moisture detection instrument failures and plugging of process lines.[iv] The majority of the intrusion events were produced by design features (e.g., PCRV liner cooling tube leaks, circulator bearing seal leaks) that are not applicable to the NGNP design. The Fort St. Vrain HTGR utilized carbide TRISO fuel with higher defect fractions than specified for the NGNP, thus it was more susceptible to the effects of moisture ingress on fuel performance. However, the gaseous and metallic fission product released from the fuel remained low considering the fuel quality, and generally well below predicted values.[v] While the Fort St. Vrain experience demonstrated that moisture ingress can be tolerated from a safety perspective, it also demonstrated that moisture ingress can substantially impact plant availability. Thus the plant should be designed so that water ingress events are infrequent enough to prevent a significant reduction of plant availability.

This paper addresses the following topics related to the need for testing to address moisture ingress:

- Primary system conditions resulting from moisture ingress events
- Key phenomena, analysis models, and design data needs
- Existing experimental data
- Additional testing required to address design data needs.

As will be discussed in Section 4, existing data show that intact coated particles are not significantly affected by exposure to moisture over the range of conditions which could credibly exist in the reactor fuel during normal operation and accident conditions. However, the data also show that release from failed particles with exposed kernels increases due to the interaction of water vapor and reaction products (from water-graphite reactions) with the kernel (hydrolysis), and that hydrolysis has a greater effect for UCO than for UO_2 kernels due to conversion of the carbide phase to oxides. This review addresses the effect of moisture ingress during normal operation and accident conditions on offsite dose calculations and the associated need for additional fuel testing.

2. EXPECTED RANGE AND CONDITIONS OF MOISTURE INGRESS EVENTS

The primary conditions of interest in establishing fuel response to moisture ingress are the fuel burnup, local temperature, and partial pressures of water and reaction products. The partial pressures within the fuel compacts where the fuel particles reside may be substantially less than in the coolant channels, particularly at higher temperatures when the water vapor is more rapidly consumed by reaction with the graphite in the reactor internals and fuel elements. The expected range and conditions of moisture ingress events for this paper are based primarily on a review of documentation produced for the 350 MWt MHTGR design developed in the 1980s, which incorporated a steam generator in the primary loop. The results of those analyses are briefly discussed below followed by a discussion of their applicability to the NGNP.

2.1 350 MWt MHTGR Analysis

Analyses of moisture ingress events are documented in the MHTGR Preliminary Safety Information Document[vi] (PSID), the Probabilistic Risk Assessment (PRA) of the Modular HTGR,[vii] and the Emergency Planning Basis Report,[viii] with Nuclear Regulatory Commission (NRC) review documented in the Preapplication Safety Evaluation Report (PSER).[ix,x] The majority of moisture ingress events are terminated by protection system actions without release into the reactor building, and thus do not contribute to offsite dose. This evaluation is limited to steam generator leaks that affect offsite dose. The events associated with moisture ingress from a steam generator that affect offsite dose are combinations of:

- Long term exposure of fuel to moisture and reaction products at impurity levels acceptable for normal operation.
- Steam generator leaks in conjunction with protection system failures that exceed the primary system overpressure protection set points resulting in release from the primary system through the primary system safety valve and associated discharge train.

- Steam generator leaks in conjunction with protection system failures that exceed the steam generator dump system overpressure protection set points resulting in release from the steam generator dump system through the dump system safety valve and associated discharge train.

- Steam generator leaks in conjunction with failures of the primary coolant pressure boundary resulting in release directly into the reactor building.

The PSID identifies two categories of events that result in offsite dose for moisture ingress: (1) Design Basis Events (DBEs) analyzed using conservative models assuming all systems function as designed, and (2) Safety Related Design Conditions (SRDCs) that assume only safety related systems function as designed. Among the moisture ingress DBEs, only DBE-7—moisture inleakage without shutdown cooling system cooling—results in offsite dose (0.035 rem thyroid, 95th percentile vs. 300 rem 10 CFR 100 limit). Among the moisture ingress SRDCs, SRDCs 6 and 7 (depressurized conduction cooldown with moderate moisture ingress), which are identical under the rules of SRDC analysis, result in an offsite dose of 3.8 rem thyroid (95th percentile). The analyses include models for release from failed particles, with a predicted iodine release fraction of 7.9% for DBE-7. Additional analyses were provided in response to NRC staff questions on the PSID, with one case (R 15-17 on the effect of relief time on SRDC-6 dose) resulting in a maximum thyroid dose of 25 rem and 100% hydrolysis of exposed kernels. The results documented in the PSID do not include water vapor partial pressure data, and the fuel temperatures vary with time and location within the reactor (normal operation ~300–1330°C, maximum accident condition ~1550°C).

The PRA results indicate that overall risk is dominated by higher frequency dry conduction cooldown events. The wet conduction cooldown category (WC) addresses small and moderate steam generator leaks with subsequent failures in the steam generator dump and isolation, and primary pressure relief systems. The largest thyroid dose at the exclusion area boundary is obtained for WC-1, with a mean of 9 rem, and median of 2.4 rem, whose low frequency ($2x10^{-7}$/yr) offsets the dose from a risk standpoint. The largest fractional release from failed particles due to hydrolysis is reported as 20% for WC-3, with other fractional releases ranging from 5.7 to 8%.

The emergency planning basis analysis results[viii] indicate that the highest plume exposure whole-body and thyroid dose are produced by EPBE-1 (Moisture inleakage with delayed steam generator isolation and without forced cooling). The margin to the plume exposure PAG limits at a 425 m exclusion area boundary radius were ~200× for whole body dose and ~5× for thyroid inhalation dose. Reference viii did not include ingestion pathway doses, which were addressed in a

later report.[xi] The ingestion pathway document was not available before the initial draft PSER,[ix] was not referenced in the later version of the NRC PSER.

In the later version of the PSER,[x] the NRC noted the higher thyroid dose that resulted from the DOE reply to PSID comment R 15-17, and that the worst meteorology was not assumed to occur during the depressurization, identifying this as a licensing issue. It also referred to statements regarding technology development in the first version of the PSER,[ix] which included the statement "it is essential that the final data offered in support of the MHTGR concept be obtained on the reference fuel design in test environments that include the conditions given in Section 15.2."

A paper presented by General Atomics at an International Atomic Energy Agency (IAEA) meeting[xii] stated that during MHTGR accident scenarios H_2O partial pressures could be as high as 355 kPa (3.5 atm) prior to depressurization. This is consistent with the mass ingress and primary system pressure response values reported in the MHTGR PSID.

2.2 Applicability to NGNP

The range of conditions experienced by the fuel during moisture ingress events is determined by reactor, primary coolant system and protection system design and operating conditions. The NGNP operating parameters recommended by the PCS alternative studies[ii,iii] are compared with the MHTGR in Table 1.

Table 1 Comparison of MHTGR and NGNP

	MHTGR	NGNP/GA	NGNP/AREVA
Reactor Inlet Temp, °C	259	322	350
Reactor Outlet Temp, °C	687	750	750
Reactor Power, MWt	350	600	600
Steam Pressure, MPa	17.3	17.2	16.7
Steam Temp, °C	541	540	566

The NGNP temperatures and steam pressures are sufficiently similar to the MHTGR that no substantial difference in the range of moisture ingress conditions for the fuel would be expected, assuming comparable protection systems (moisture detection, steam generator isolation and dump, overpressure protection) and accident condition temperature response. The higher power level would require a proportionately lower release fraction for the same offsite dose, but in general, the MHTGR results discussed above are applicable for the NGNP.

3. EXISTING EXPERIMENTAL DATA

As discussed above and confirmed by testing, the radionuclide retention characteristics of intact coated particles are unaffected by exposure to moisture within the range of conditions that may occur in the NGNP and previous HTGR designs. Thus experiments on the effect of moisture have generally been conducted using fuel specimens with known quantities of exposed kernels. The effect of moisture is more pronounced on uranium carbides (e.g., UC_2) than uranium oxides (e.g., UO_2), as the carbon atoms are replaced with oxygen atoms in the reaction process. For the UCO fuel (a mixture of UO_2, UC and UC_2), to be used in the prismatic NGNP, the carbide molecules are depleted and converted to oxide under irradiation and during hydrolysis, changing the kernel toward UO_2 as irradiation proceeds.

Existing data on the effects of moisture ingress on fuel performance include testing of LEU UCO fuel in the ORNL HFIR reactor (HRB-17/18) and the Petten HFR reactor (HFR-B1), and testing of modern, high-quality German LEU UO_2 fuel in HFR-K6. Qualification of the data for use in an NGNP licensing proceeding will need to be addressed with the NRC. The majority of the results are documented in reports referenced in the following discussion, which for the most part do not include raw primary data. This review was not able to confirm the existence of complete raw primary data such as irradiation data tapes, instrument calibration records, and operating procedures. A copy of the log of the HRB-17/18 irradiation, including selected data tables and strip chart recorder segments, was obtained,[xiii] along with a review of the HFR-B1 quality assurance measures.[xiv] Given the limited amount of existing primary data, it is likely that some replication of existing data will be necessary.

3.1 HRB-17/18

The HRB-17/18 irradiations were conducted in the HFIR reactor at ORNL to investigate the effect of moisture on gaseous radionuclide release from exposed UCO kernels distributed within a fuel compact.[xv] The exposed UCO kernels were produced by including five designed-to-fail (DTF) particles in each of six ~1.9 cm long by ~1.27 cm diameter compacts. The 347 μm diameter DTF LEU (~19.5% enriched) $UC_{0.4}O_{1.6}$ kernels included an inner region of UO_2 and UC_2 surrounded by a ~30 μm thick rind of UO_2 and a ~20 μm anisotropic pyrocarbon seal coat.[xxiii] In addition to the 5 DTF particles, each compact contained ~175 intact LEU UCO TRISO fissile particles and ~400 ThO_2 TRISO fertile particles. Figure 1 provides a sketch of the HRB-17/18 compacts and DTF particles and Figure 2 provides a microphotograph of representative unirradiated DTF particles.

In the HRB-17/18 experiments the purge gas, consisting of a mixture of helium and neon with water vapor added during the moisture injection tests, flowed directly around the stack of six compacts in each capsule. The purge gas pressure

was held constant at 15 psig. The irradiations and moisture injections were conducted for a total time of ~123 days with time averaged fuel temperatures in the range of ~700–840°C, with the exception of shorter periods in the range of

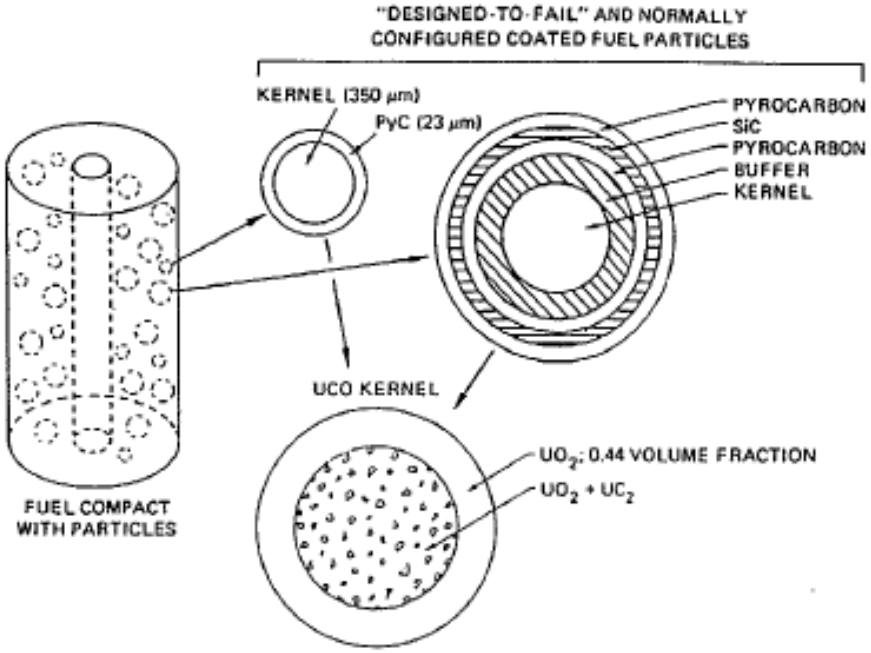

Figure 1. HRB-17/18 compacts and DTF particles.[xvi]

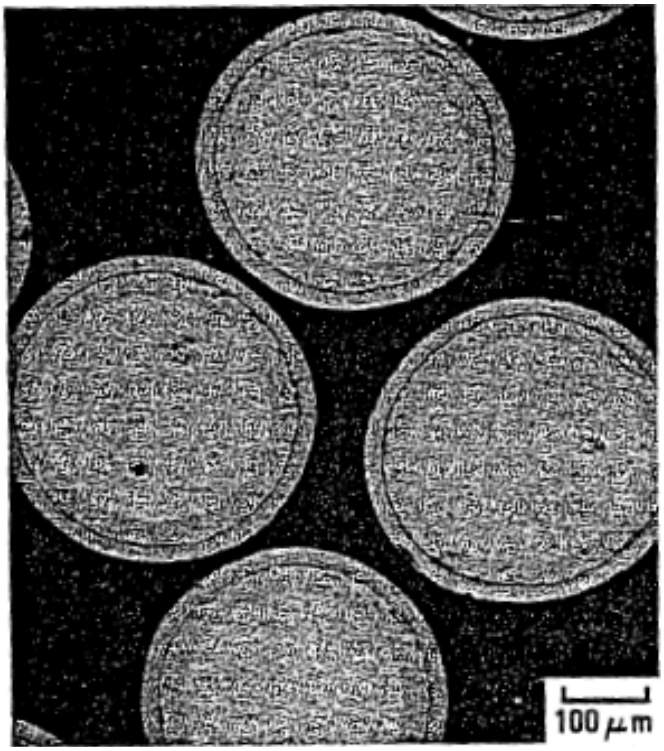

Figure 2 DTF particle photomicrograph.[xv]

~900–1040°C. Fissile particle burnup at the end of the irradiation was ~25% FIMA and fast fluence (E>29 fJ) was ~4.7×10^{25} n/m^2. Considerations unique to each of the capsules are addressed below.

3.1.1 HRB-17

Four moisture injections were performed with the capsule under irradiation. The injection start time (measured in days from the start of the irradiation), duration, and water vapor density are given in Table 2. As noted earlier, the total irradiation time was 123 days.

Table 2 HRB 17 water vapor injections.

Injection Number	1	2	3	4
Irradiation time at start, full power days	32	52	91	104
Injection duration, days	13.3	3.8	5.0	11.7
Water vapor concentration, ppmv	100	1,000	1,000	300
Water vapor partial pressure, psia	0.003	0.03	0.03	0.009
Water vapor partial pressure, kPa	0.02	0.2	0.2	0.06

The most significant data produced were noble gas release measurements from grab samples taken periodically during the irradiation. The response indicated a substantial release of gas stored in the exposed kernels at the beginning of moisture injection and changes in the steady state release characteristics following injection. These data were considered in the development of models to predict the effect of moisture ingress on gaseous release from failed LEU UCO particles.

3.1.2 HRB-18

Fuel in the HRB-18 capsule was subjected to moisture injection at a concentration of 10,000 ppmv at the end of a HFIR cycle at 104 full-power days. The injection was continued for 27.6 days with the capsule removed from the HFIR core and maintained at 200°C, with the objective of investigating the effect of a moisture ingress event during refueling. No gas release data were taken during the moisture injection. These data are relevant for moisture ingress during refueling.

3.2 HFR-B1

The HFR-B1 irradiation was conducted in the HFR reactor at the Joint Research Centre – Petten, the Netherlands. ORNL reports describe the irradiation[xvii] and postirradiation examination and testing,[xviii] and a recent General Atomics report[xix] provides an overall summary of the results. The irradiation rig contained three sealed capsules, with Capsule 3 subjected to moisture ingress during the irradiation and fuel compacts from Capsules 2 and 3, and fuel kernels from Capsule 3 exposed to moisture during postirradiation heating. The driver and DTF particles were taken from the same batches as those used for HRB-17/18 as described in Section 3.1. Each capsule contained 12 compacts 12.85 mm in diameter and 19 mm long, with 87 DTF particles in each compact (1,044 per capsule). The compacts were contained in a graphite fuel body, and during the moisture ingress during irradiation of Capsule 3, the moisture laden purge gas flowed around the fuel bodies. The results from Capsules 2 and 3 are briefly summarized below.

3.2.1 Capsule 2

The estimated fuel temperatures during irradiation in Capsule 2 ranged from 880 to 1,230°C with temperatures varying with time over a range of ~200°C to measure the effect of temperature on fission gas release. Fast neutron fluence at the end of the irradiation was 6.8×10^{25} n/m^2 (E > 0.1 MeV). The burnup for the compacts tested in postirradiation heating was reported as 21.7% FIMA.

Two compacts from Capsule 2 were subjected to postirradiation heating (800°C) and moisture ingress events (two moisture injections at a partial pressure of 1 kPa of durations 165 and 95 hours). A relatively mild increase in Kr^{85} release was observed in conjunction with the moisture injections.

3.2.2 Capsule 3

The estimated fuel temperatures during irradiation in Capsule 3 ranged from 820 to 1,050°C, with temperatures varying with time over a range of ~100°C to measure the effect of temperature on fission gas release. Fast neutron fluence at the end of the irradiation was $5.0x10^{25}$ n/m^2 (E > 0.1 MeV). The burnup for the compacts tested in postirradiation heating was reported as 19.7% FIMA. During the irradiation, Capsule 3 was subjected to a total of 16 moisture injections ranging from 0.018 to 1.06 kPa inlet partial pressure, and durations from 6 to 241 hours. The gaseous fission product R/B response is provided in both tabular and graphic form in the operating history report.[xvii]

Two compacts from Capsule 3 were also subjected to postirradiation heating (800°C) and nine moisture ingress events. The water partial pressures ranged from 1 to 50 kPa with durations from 24 to 48 hours. The initial ingress at 1 kPa and all the events at 14 to 50 kPa showed a marked increase in Kr^{85} release at the beginning of the ingress. As with the in-pile R/B, the Kr release is provided in both tabular and graphic form for both the compact tests and the loose particle tests discussed below.

Additional postirradiation heating and moisture ingress tests were conducted on two sets of five bare kernels produced by removing the coatings from loose particles irradiated in the same tray in Capsule 3. The first set of kernels was subjected to four water injections with the first at a partial pressure of 1 kPa and the following three at 50 kPa, with all injection durations of 10 hours. The second set of kernels was subjected to water injections of 1 kPa for 10 hours followed immediately by 2 kPa for 10 hours. The release characteristics differed significantly in that the first case indicated ~20% release of Kr^{85} inventory on heatup and ~70% release on the first water injection (1 kPa) while the second indicated ~70% release on the heatup and ~15% release on the first injection.

The effect of water vapor on metallic fission product release was addressed to a limited degree based on postirradiation gamma scanning of the compacts and of the graphite fuel bodies with the compacts removed for both Capsules 2 and 3. The report concludes that water vapor induces

an increase of release of cesium and cerium from exposed kernels and a reduction in retention (increase in diffusivity) of these fission products in the compact matrix. In the absence of a mass balance on these isotopes in the capsule, these conclusions do not appear to be unambiguously supported by the available data.

3.3 HFR-K6

The HFR-K6 irradiation was conducted in the HFR reactor at the Joint Research Centre – Petten. The test rig contained three cells, with the top and bottom cells each containing one sphere of German proof test fuel (UO_2 kernels), and the center cell containing two spheres. Based on the initial in-pile release data, the sphere in the bottom cell (sphere 4) was determined to have two particles with exposed kernel defects produced during manufacturing, thus this cell was the focus of a series of moisture ingress tests.[xx] The burnup and fast neutron fluence in Sphere 4 after 26 HFR irradiation cycles were 9.2% FIMA and 4.5×10^{25} m^{-2} ($E > 0.1$ MeV), with moisture injections conducted in Cycles 22 and 25. A total of eight injections were conducted with an average fuel sphere temperature of 680°C at water partial pressures of 0.2, 0.5, and 2 kPa, with two additional injections conducted at 930°C and partial pressures of 0.002 and 0.03 kPa. Enhanced release of gaseous fission products was observed in all the injections.

3.4 Applicability to NGNP Fuel

The NGNP UCO TRISO coated particle fuel is very similar to the UCO TRISO fuel irradiated and tested in HRB-17/18 and HFR-B1. In addition, as the fuel is consumed in a reactor, the carbon is displaced by excess oxygen released during fission, thus the composition becomes increasingly UO_2. Thus the results of the HFR-K6 testing of UO_2 TRISO are also relevant in determining the effect of moisture ingress on NGNP fuel.

However, the process used to produce the UCO kernels has been changed from an external gelation process used to produce the kernels irradiated in HRB-17/18 and HFR-B1, to an internal gelation process used for NGNP fuel. In addition the kernel heat treatment process for the NGNP fuel differs from the earlier fuel. While the kernel microstructure appears to be similar, there may be differences in the phase distributions within the kernels, which is relevant to response to moisture ingress, particularly at lower burnups when more carbides are present.

As noted in Section 2.1, the water partial pressure for moisture ingress events in the MHTGR was found to be as high as 355 kPa. This is well above the maximum water partial pressure in the tests described above of 50 kPa.

4. KEY PHENOMENA AND ANALYSIS MODELS, DESIGN DATA NEEDS

Key phenomena and the models for analyzing those phenomena are briefly described and design data needs that have been identified in this area are presented.

4.1 Key Phenomena

There are two principal phenomena that occur as the result of moisture ingress into the core: oxidation of graphitic core components and hydrolysis of exposed fuel kernels. These phenomena are coupled in that water vapor is depleted by oxidation of graphitic core components before reaching the fuel compacts, especially at high temperatures.

4.1.1 Graphite Oxidation

Moisture ingress results in access of water vapor to the cooling channels in the graphite blocks in the core. In order for water vapor to interact with the fuel compacts that are in adjacent holes in the graphite blocks, the water vapor must penetrate the graphite web (~5 mm thick) separating the coolant channels from the fuel compacts. At higher temperatures, water vapor is depleted by oxidation in the graphite web before reaching the fuel compacts and by oxidation with the compact matrix before reaching exposed fuel kernels.[xxi] Thus, oxidation of graphitic components affects the concentration of water vapor that eventually may interact with exposed fuel kernels. The principal reaction is $H_2O + C = CO + H_2$.

In addition, the water-gas-shift reaction, which will convert some CO to CO_2 is $H_2O + CO = CO_2 + H_2$.

A third reaction (Boudouard reaction), $C + CO_2 = 2CO$, is known[xxi] to be about an order of magnitude slower than the steam-graphite reaction.

4.1.2 Fuel Hydrolysis

Water vapor causes hydrolysis of the fuel only in particles with exposed kernels. Fuel particles with exposed kernels may be present as a result of fabrication defects or incremental failure from normal operation. Fuel with either UCO kernels (having a mixture of UC, UC_2 and UO_2 in the kernel) or UO_2 kernels is susceptible to hydrolysis. The UC and UC_2 content of UCO fuel (typically about 20% initially in the experiments performed to date) is diminished under normal irradiation conditions by reaction with oxygen released by the fission of UO_2 and PuO_2, and by fission of the UC and UC_2. During hydrolysis, UC and UC_2 are oxidized to UO_2, and UO_2 is oxidized to UO_{2+x}.

During irradiation, the sequential response of exposed UCO fuel kernels embedded in compacts to moisture ingress consists of three stages: (1) a rapid transient release of stored fission gas, (2), a period of constant steady-state release, and (3) a decline in the release rate to a steady-state post-hydrolysis value. This response is shown for UCO fuel containing both UC_2 and UO_2 phases in Figure 3 from experiment HRB-17.[xxii] The same general behavior results from the hydrolysis of compacts containing exposed UO_2 fuel kernels (or UCO kernels without any remaining UC_2 phase) except that the decline in R/B from the steady-state value in Stage 2 reaches a post-water injection steady-state value equivalent to the prehydrolysis value, as is shown in Figure 4.[xxii] The same general stages of fission gas release are seen in postirradiation moisture ingress tests of compacts containing exposed kernels as can be seen in Figure 5[xxiii] for UCO (no UC_2 remaining postirradiation) from the HFR-B1 test.

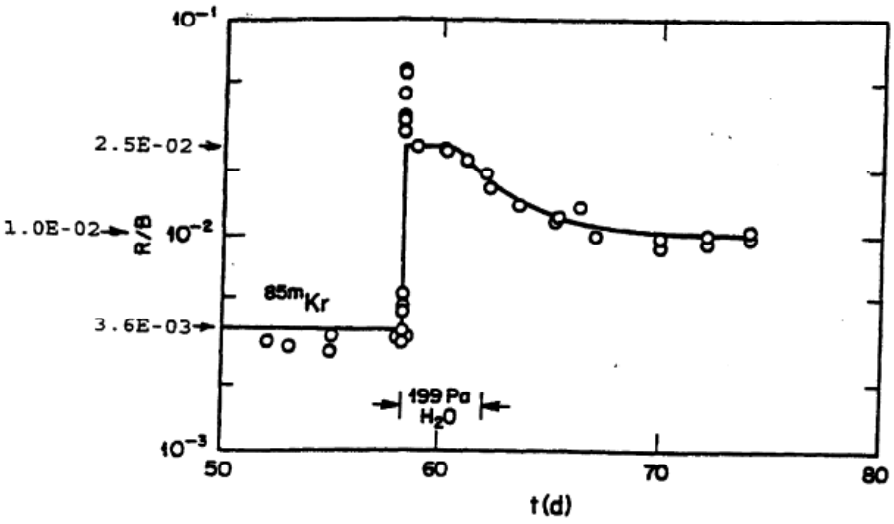

Figure 3. R/B time profile from in-pile experiment HRB-17 for Kr-85m before, during, and after water injection at 199 Pa water vapor partial pressure and 779°C for UCO fuel containing both UC_2 and UO_2 phases.

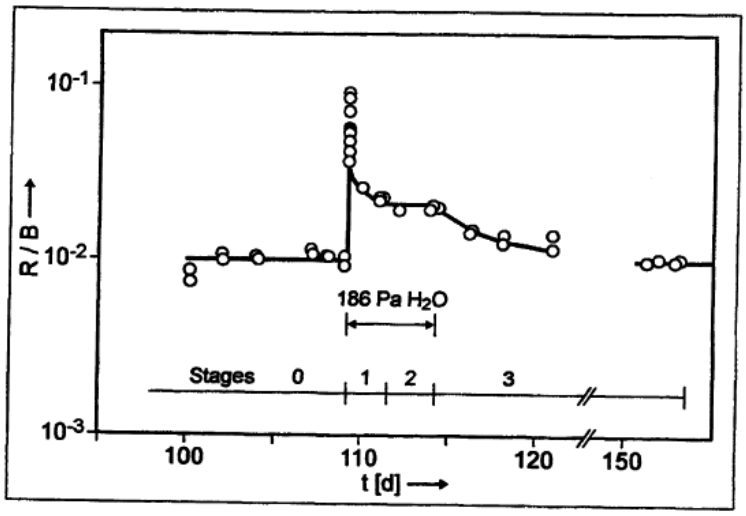

Figure 4 R/B time profile from in-pile experiment HRB-17 for Kr-85m before, during, and after water injection at 186 Pa water vapor partial pressure and 767°C for UCO fuel containing only the UO_2 phase.

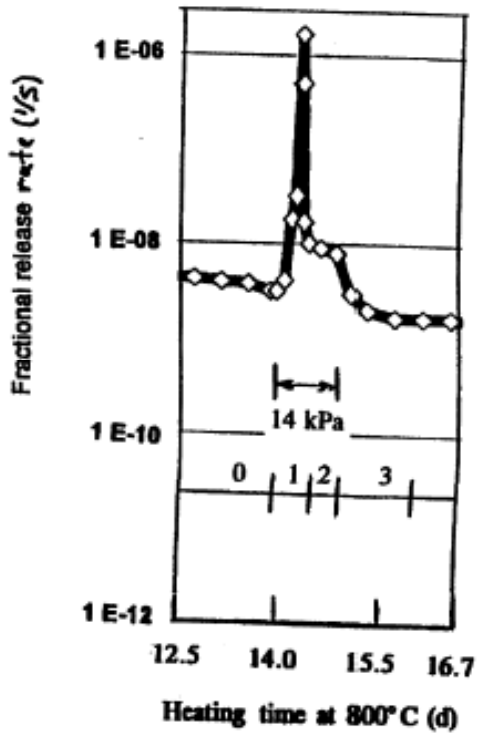

Figure 5. Fractional release rate for Kr-85m before, during, and after a postirradiation water vapor injection test conducted in KORA of

Compacts 3.2.1 and 3.2.2 from experiment HFR-B1. The fuel was UCO with no remaining UC_2 phase.

The greatest fractional release of fission gas occurs in the transient release phase, and the fractional release is independent of the chemical species of the gas (Kr or Xe) or the isotope, and also independent of the fuel species (UCO or UO_2). This release is thought to be a result of changes in fuel structure resulting from the oxidation of the UC_2 and UO_2 phases by hydrolysis that causes gases stored in bubbles to be released. At low partial pressures of water vapor, the fractional release of stored fission gas was found to be a simple function of the partial pressure of the water vapor. This result is shown in Figure 6[xxiv] from HRB-17, in which UC_2 has been consumed by irradiation and hydrolysis, leaving only UO_2 in two instances. Evidence is presented[xxiv] that carbide hydrolysis is much more rapid than oxide hydrolysis in exposed UCO kernels, but the carbide fraction in the UCO was generally 10% or less at the times of water injections, thus minimizing the contribution of the carbide phase to fission gas release relative to the oxide phase.

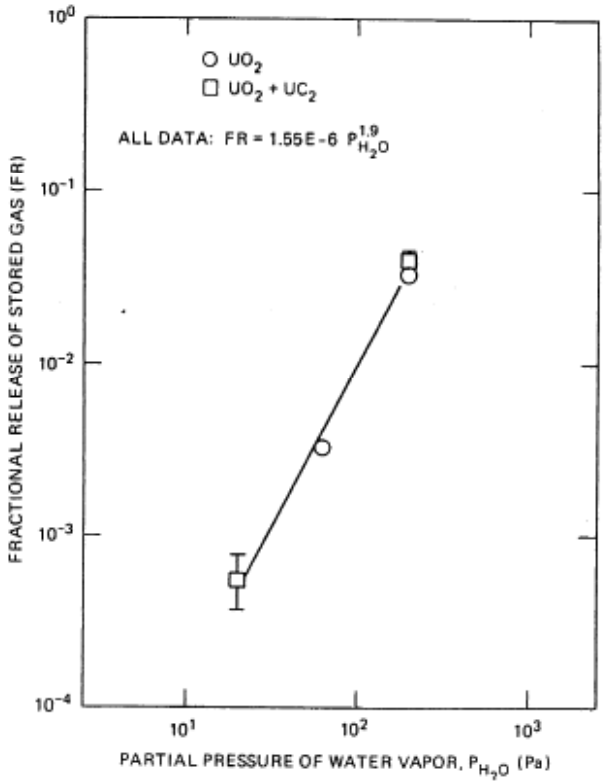

Figure 6. Dependence of the release of stored fission gas as a fraction of the inventory on the partial pressure of water vapor, experiment HRB-17.

4.2 Analysis Models

Existing analysis models for both graphite oxidation and fuel hydrolysis are discussed below.

4.2.1 Models for Graphite Oxidation

A model for the reaction of fuel element graphite with low concentrations of steam in the helium coolant (0.01-0.1 ppm, equivalent to 0.06 to 0.6 Pa in primary coolant He at a pressure of 6 MPa) to calculate graphite corrosion under normal operating conditions in an MHTGR has been presented by Richards.[xxv] It is stated that this model may readily be used to analyze abnormal occurrences when steam concentrations are much higher. In a subsequent publication,[xxvi] the REACT program based on the model described in Ref. xxv was used to analyze graphite corrosion in the HFR-B1 test. It was found that the model did well in calculating the graphite oxidation at high temperatures,

but underpredicted the oxidation at low temperatures by about 30%. The explanation given is that the oxidation of the compact matrix material and the UC_2 phase of the exposed UCO kernels was not modeled. The UC_2 contribution to the CO and CO_2 reaction product gases measured in the experiment would be very small, but the compact matrix material, which is an order of magnitude more reactive than graphite, could easily account for the additional gases. The REACT program calculates that water vapor readily penetrates the graphite at lower temperatures to reach the fuel compacts, but not at higher temperatures, as shown in Figure 7. This is a graphic example of the importance of coupling models for graphite oxidation and fuel hydrolysis.

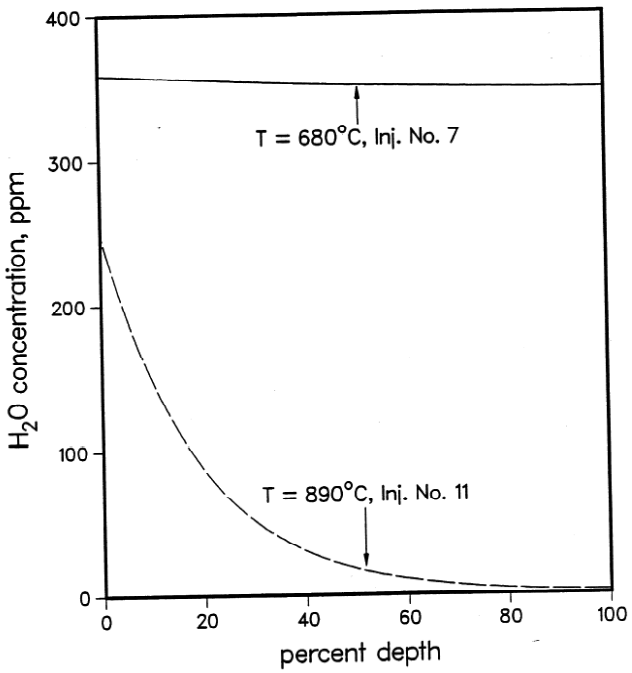

Figure 7. REACT calculations for water-vapor concentration as a function of depth from the graphite-gas interface in experiment HFR-B1.

4.2.2 Models for Fuel Hydrolysis

Models have been developed to fit experimental data for each of the three stages of fuel hydrolysis. For fractional releases of fission gases from bubbles during Stage 1 of fuel hydrolysis, the following equation has been developed[xxii] based on the in-pile experiments HRB-17 and HFR-B1:

$$F = 2.13E13\ p^{(-4.353 + 6503/T)}\ e^{(-4.7257E4/T)} \quad (3\text{-}1)$$

Where p is the partial pressure of water vapor in Pa and T is the temperature in K. Equation 3-1 is valid only over the range of water vapor pressures and temperatures of water injections in the HRB-17 and HFR-B1 experiments (20.6-199 Pa, 756–779°C) and (2.8–1050 Pa, 820–1,040°C), respectively, a full range of 2.8–1,050 Pa and 756–1040°C. The exponent of the water vapor pressure varies from 1.9 at 767°C, to 1.7 at 800°C. Equation 3-1 is plotted as a function of water vapor partial pressure at 770°C in Figure 8.[xxii] At 800°C, data from the hydrolysis of two exposed kernels in an irradiated German UO_2 fuel sphere (HFR-K6 experiment) are included along with data from HFR-B1 and Equation 3-1 in Figure 9.[xxii] This curve shows that at water vapor partial pressures above 1 kPa, the fractional release deviates from Equation 3-1, with the data point at 2 kPa falling about a factor of 4 below the line. Also at 800°C, Figure 10[xxiii] shows fractional release data from postirradiation moisture ingress tests at even higher partial pressures of water vapor, showing a very shallow slope at high pressures.

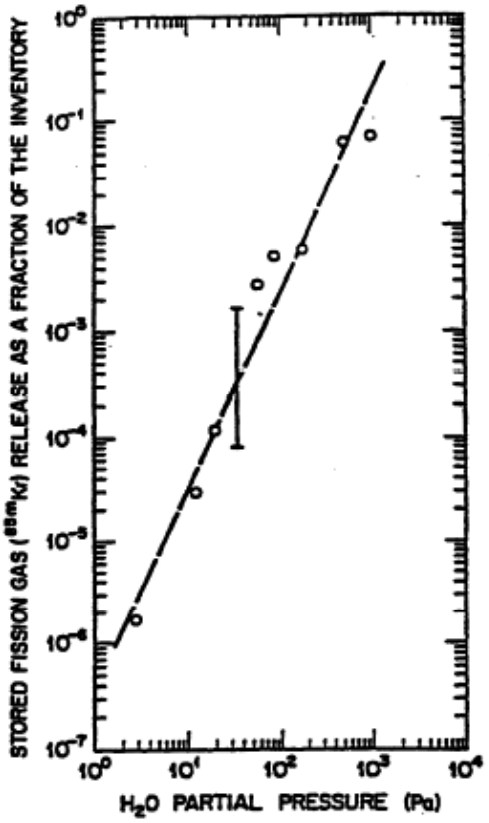

Figure 8. The dependence of the release of stored fission gas on the partial pressure of water vapor at 770°C derived from experiments HRB-17 and HFR-B1.

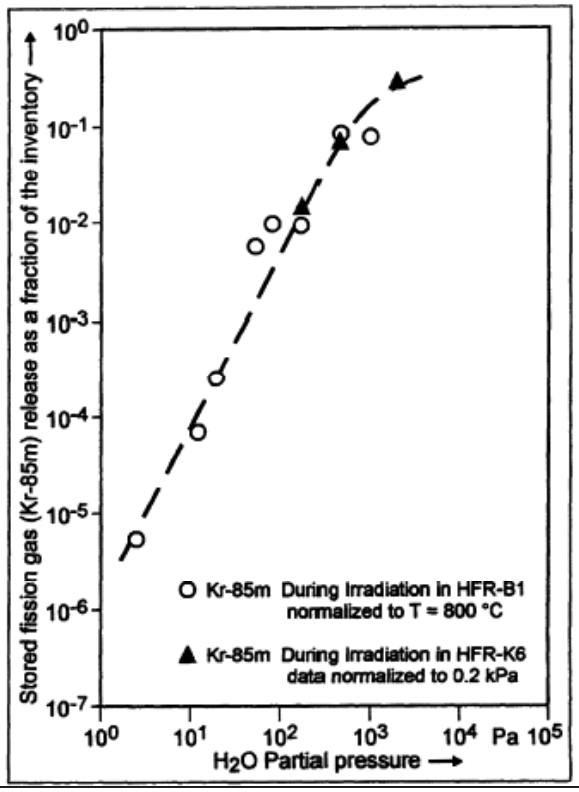

Figure 9. Fractional transient release of stored fission gas as a function of the partial pressure of water vapor at 800 C based on experiments HFR-B1 and HFR-K6.

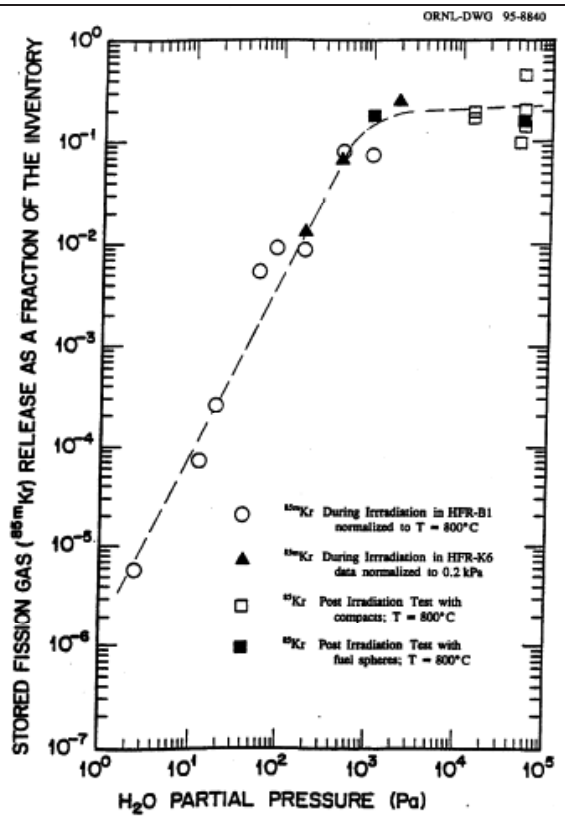

Figure 10 Fractional transient release of fission gas as a function of partial pressure of water vapor. Data from HFR-B1 and HFR-K6 during irradiation and post-irradiation testing of irradiated HFR-B1 compacts.

Correlations have been developed[xxii] for the ratio of R/B during Stage 2 steady-state release under water injection, $(R/B)_2$ to R/B prior to water injection, $(R/B)_0$ as

$$h_0 = (R/B)_2/(R/B)_0. \quad (3\text{-}2)$$

In Experiment HRB-17, h_0 for hydrolysis of UCO containing both UC_2 and UO_2 phases is 6.9 for krypton and 8.6 for xenon. For UCO fuel with no UC_2 remaining (only UO_2 present) the values of h_0 are 2.0 for krypton and 2.5 for xenon.

Correlations of the following form have been developed[xxii] that describe the decrease in R/B to a steady-state value, $(R/B)_p$ following cessation of water injection:

$$(R/B)_d = (R/B)_p\,[1 + (h_p - 1)\exp\{-\delta\,(t - t_T)\}] \quad (3\text{-}3)$$

where h_p is the ratio $(R/B)_2/(R/B)_p$ and t. is the time at which the water injection is terminated. In the case of UCO containing both the UC_2 and UO_2 phases, $h_p = 2.5$ (see Figure 3); whereas for fuel containing only the UO_2 phase, $(R/B)_p = (R/B)_0$ and $h_p = h_0 = 2.0$ (see Figure 4). The value of $\delta = 0.425$ was found to be independent of the isotope, element, water vapor pressure and fuel composition in HRB-17. The time at which the approach to $(R/B)_p$ is 95% complete is given by

$$\Delta t_{95} \approx 3/\delta \qquad (3\text{-}4)$$

which is approximately 7 days for experiment HRB-17.

These detailed correlations for h_0, h_p, and $(R/B)_d$ are interesting but probably not very important, given that the largest releases of noble gases and iodine are likely to occur during Stage 1, the transient release of stored gas.

In test HRB-18,[xxvii] water vapor was injected at a level of 2 kPa for 28 days, while the capsule was maintained at 200°C in a furnace in a pool following five cycles of irradiation at a fuel temperature of about 800°C in the HFIR. At the end of this period, the capsule was returned to the reactor and irradiation was resumed at a fuel temperature of 800°C. The Kr-85m R/B showed an increase by a factor of 1.8 times the prehydrolysis level, but R/B decreased to a steady-state level about 1.2× the prehydrolysis level within about a day. During the 28 days at 200°C and 2 kPa water vapor partial pressure, both the UC_2 and UO_2 portions of the exposed UCO kernels were calculated to be completely hydrolyzed.

4.3 Design Data Needs

Design data needs (DDNs) that include moisture ingress were developed for the steam cycle MHTGR[xxviii] and the gas turbine GT-MHR[xxix] and are collected in Table 3. DDNs addressing similar technical issues are indicated by a category letter designation, such as (a), (b), etc., following the DDN number.

Table 3 DDNs addressing effects of moisture ingress.

DDN No.	DDN Title	PHENOMENA Addressed
M.XX.ZZ	Commercial MHTGR [steam-cycle]	
M.07.01 (a)	Fission Gas Release from Core Materials	Effect of H_2O and impurities on fission gas release, including iodines, during normal operation and core heatup accidents
M.07.02 (a)	Fission Metal Effective Diffusivités in Fuel Kernels	Effect of H_2O and impurities on fission metal release from kernels during normal operation and core heatup accidents

DDN No.	DDN Title	PHENOMENA Addressed
M07.04 (d)	Fission Product Diffusivities/Sorptivities in Graphite	Effect of H_2O on fission metal transport in matrix and graphite during normal operation and core heatup accidents
M.07.06 (e)	Tritium Transport in Core Materials	Effect of H_2O on H-3 sorption on graphite
M.07.08 (h)	Radionuclide Removal Characteristics for Wet Depressurization [single-effects data]	Removal of plateout activity, especially iodines, by steam and liquid H_2O
M.07.12 (b)	Data for Validation of Fission Gas Release [integral validation data]	Effect of H_2O on fission gas release, including iodines, during normal operation and core heatup accidents
M.07.13 (b)	Data for Validation of Fission Metal Release [integral validation data]	Effect of H_2O on fission metal release from the core during normal operation and core heatup accidents
M.07.16 (i)	Integral Test Data For Radionuclide "Washoff"	Removal of plateout activity, especially iodines, by steam and liquid H_2O
M.07.26 (c)	Data for Validation of Fuel Performance under Normal Operating Conditions	Effect of trace H_2O (≤630 μatm) on normal operation coating integrity
M.07.27 (c)	Data for Validation of Fuel Performance under Core Conduction Cooldown Conditions	Effect of H_2O on coating intergrity during core heatup accidents
C.XX.YY.ZZ	Commercial GT-MHR [direct-cycle]	
C.07.02.05 (c)	Normal Operation Fuel Performance Validation Data	Effect of trace H_2O (≤700 μatm) on normal operation coating integrity
C.07.02.06 (c)	Accident Fuel Performance Validation Data	Effect of H_2O on coating intergrity during core heatup accidents
C.07.02.07 (c)	Fuel Proof Test Data	Same DDNs C.07.02.05 & C.07.02.06 but with proof test fuel
C.07.03.01 (a)	Fission Gas Release from Core Materials	Effect of H_2O and impurities on fission gas release, including iodines, during normal operation and core heatup accidents
C.07.03.02 (a)	Fission Metal Effective Diffusivities in Fuel Kernels	Effect of H_2O and impurities on fission metal release from kernels during normal operation and core heatup accidents
C.07.03.04 (d)	Fission Product Diffusivities/Sorptivities in Graphite	Effect of H_2O and impurities on fission metal transport in matrix and graphite during normal operation and core heatup accidents
C.07.03.06 (e)	Tritium Transport in Core Materials	Effect of H_2O on H-3 sorption on graphite
C.07.03.10 (h)	Radionuclide Removal Characteristics for Wet Depressurization [single-effects data]	Removal of plateout activity, especially iodines, by steam and liquid H_2O
C.07.03.12 (f)	Fission Product Transport in a Vented Low-Pressure Containment	Condensation, settling, and plateout on reactor building materials of construction

DDN No.	DDN Title	PHENOMENA Addressed
C.07.03.13 (g)	Decontamination Efficiency of Pressure Relief Train Filter	Effectiveness of candidate filter media for removal of radionuclides, especially iodines, during wet depressurization
C.07.03.14 (b)	Fission Gas Release Validation Data [integral validation data]	Effect of H_2O on fission gas release, including iodines, during normal operation and core heatup accidents
C.07.03.015 (b)	Fission Metal Release Validation Data	Effect of H_2O on fission metal release from the core during normal operation and core heatup accidents
C.07.03.18 (i)	Radionuclide "Washoff" Validation Data [integral validation data]	Removal of plateout activity, especially iodines, by steam and liquid H_2O

5. COVERAGE OF DESIGN DATA NEEDS

This section discusses coverage of DDNs by existing data, DDNs requiring additional data, and testing needed to provide additional data.

5.1 DDNs Addressed by Existing Data

Quantification of the effect of hydrolysis on fission gas release from exposed kernels during normal irradiation is provided by the results of the in-pile experiments HRB-17, HFR-B1, and HFR-K6 described in Section 4. These results address the moisture portion of impurities in the helium coolant, but not the CO and CO_2 impurities identified in the DDNs under category (a). The results of postirradiation moisture ingress experiments included in Figure 10 address the data needs identified for pressurized core conduction cooldown transients under category (a) for moisture ingress, but not the CO and CO_2 impurities.

5.2 DDNs Requiring Additional Data

New data are required to address the need, identified in the DDNs in category(a) for the effects of impurities beyond moisture, namely CO and CO_2 on fission product releases, both gases and metals, from exposed kernels under normal irradiation conditions. Moisture should also be included to fully address the question of the effect of low levels of impurities in helium during normal irradiation.

New data on the effects of moisture during pressurized CCCD will validate results from earlier experiments, such as those in Figure 10, as called out in DDNs in category (a). These same DDNs call for the effects of impurities CO and CO_2 on the releases of fission products under pressurized CCCD conditions.

It should be noted that none of the DDNs listed in Table 3 identify the need to conduct experiments with moisture under depressurized CCCD conditions.

Rather, the impurities CO and N_2 are identified under depressurized conditions. The other principal difference between pressurized and depressurized CCCD conditions is the temperature: 900–1,300°C for pressurized and 1200–1,800°C for depressurized. It should also be noted that previous experiments with large amounts of moisture under CCCD conditions (results in Figure 10) were carried out at a system pressure of 1 atm, not the >10 atm identified in the DDNs. Given the difficulty of performing high temperature tests in oxidizing atmospheres, it seems exceedingly difficult to design a furnace to also operate at very high pressures.

New data are required to address the remaining DDNs identified in Table 3 and the tests identified to satisfy these DDNs are described briefly in the Section 5.3.

5.3 Testing Needed to Provide Additional Data

It may be possible to supply one of the capsules in irradiation test AGR-3/4[i,xxx] with helium coolant containing impurities identified in the DDNs in category (a): 14 Pa H_2O, 35 Pa CO, and 14 Pa CO_2. This experiment, with DTF particles, will measure fission gas (on-line) and fission metal (PIE) releases from a known number of exposed kernels, and also measure the transport of fission metals in annuli of matrix graphite and fuel element graphite. The irradiation, PIE, and safety testing (pressurized and depressurized CCCD) would address the DDNs in categories (a) and (d). If, during the design of the AGR-3/4 experiment, it turns out not to be practical or desirable to dedicate a capsule to the effects of moisture and/or impurities, a separate irradiation experiment could be considered.

Adding helium coolant with impurities to one of the capsules in irradiation test AGR-8,[i] which is devoted to the validation of fission product releases from exposed kernels and in some of the safety tests, would satisfy the DDNs in category (b). The safety tests should be performed on irradiated compacts containing DTF particles, rather than individual exposed kernels, to maintain the appropriate oxygen potential of the test specimen. During irradiation and subsequent testing, the compact(s) should be contained in a cylindrical graphite fuel body of thickness simulating the diffusion characteristics between coolant and fuel channels in a prismatic fuel element. The postirradiation test facility should have the capability of measuring metallic as well as gaseous fission products. It is further recommended that each test be limited to a single water injection because multiple injections become difficult to interpret. If, during the design of test AGR-8, it is deemed impractical or undesirable to dedicate one capsule to the effects of impure helium, a separate irradiation test could be considered.

Including impurities in the helium coolant to one of the capsules in the AGR-7[i] irradiation and in some of the postirradiation safety tests, would satisfy the DDNs identified in category (c) in Table 3. A separate irradiation test could be considered as an alternative. Such a test could furnish in-pile data on fission gas release and irradiated sample materials for postirradiation examination and safety testing to address DDNs in categories (a), (b), (c), and (d). Note that the furnaces currently available for safety testing are not designed to operate in oxidizing atmospheres, so a new furnace will need to be designed and constructed for this purpose.[i]

The test plan proposed for characterizing tritium transport in a VHTR[xxxi] calls for measuring tritium transport in core graphites as a function of coolant chemistry (impurities in helium), which addresses DDNs in category (e).

Coolant chemistry (helium impurities) are included in a plan[i,xxxii] proposed to determine fission product behavior in the vented low-pressure containment under dry and wet core conduction cooldown conditions. This research addresses the DDNs under category (f).

Coolant chemistry (helium impurities) is included in the plan[i] to determine the decontamination efficiency of a pressure relief train filter. This research addresses the DDN under category (g).

A plan[xxxiii,xxxiv] has been proposed to address radionuclide removal from primary coolant surfaces under wet depressurization to satisfy the DDNs under category (h).

A plan[xxxv] has been proposed to address the validation of fission product washoff to satisfy the DDNs under category (i).

A systematic review needs to be undertaken of current computer codes and methods for modeling the effects of water ingress, and scoping calculations need to be performed to better define the conditions for tests that support the DDNs.

6. CONCLUSIONS

Additional testing is considered necessary to address the effect of moisture ingress on radionuclide release from the fuel for a prismatic NGNP with a steam generator in the primary loop. In-pile testing with impure helium, including moisture, is needed as is postirradiation safety testing. Additional testing with moisture is identified to address design data needs established for previous MHTGR designs that are applicable to the NGNP. The work includes the following:

- Confirmation that NGNP DTF UCO TRISO fuel performance is equivalent to DTF UCO TRISO fuel used in existing tests by repetition of selected test conditions
- In-pile testing under normal operating conditions with low levels of impurities in the helium coolant, including moisture
- Post-irradiation testing to moisture levels enveloping the maximum water partial pressure predicted for the NGNP
- Additional data on the effect of moisture ingress on metallic fission product release from fuel compacts and graphite
- A systematic review of current computer codes and methods for modeling the effects of water ingress and scoping calculations to better define the conditions for tests that support the DDNs.

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