EVALUATION OF NEED FOR
INTEGRAL FUEL OXIDATION TESTS

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## EVALUATION OF NEED FOR INTEGRAL OXIDATION TESTS

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1. ABSTRACT

This document establishes the need for an integral fuel oxidation test which can give confidence to the predictions made by the OXIDE computer code for fuel and core damage during water ingress events in the Modular High Temperature Gas Cooled Reactor (MHTGR). This testing will provide clear engineering evidence to demonstrate that the core of the MHTGR can survive a moisture ingress incident with minimum investment risk and without danger to the reactor personnel or to the public. In particular, these tests will determine the degree of particle debonding and compact stack densification as a function of the fractional compact matrix burnoff.

Also included in the document is (1) a description of the proposed tests and, (2) a test matrix of the planned experiments.

2. BACKGROUND

The MHTGR is an inherently safe nuclear heat source designed to operate within a safe temperature regime to minimize investment risk and danger to the public even during a loss of forced circulation accident.

For licensing of the MHTGR, Design Basis Events (DBEs) and Safety-Related Design Conditions (SRDCs) were identified which, in the unlikely event of occurrence, could possibly result in significant offsite radioactive doses. Engineering studies utilizing complex computer codes were conducted primarily to study the effects that these DBEs and SRDCs would have on the safety of the public.

DBEs 6 to 9 (Ref. 1) result in a moisture ingress to the primary coolant, due to a steam generator tube leak. In these DBE scenarios the amount of moisture ingress is limited by assuming the plant protective
system operates properly to detect moisture and isolate the leaking steam generator or by assuming a small steam leak and a trip on high reactor pressure.

The most severe moisture ingress is considered in the SRDC-6 case. The basis for SRDC-6 is a steam generator tube leak allowing 12.5 lbm/s moisture ingress into the MHTGR primary circuit which results in a reactor trip and pressurized loss of forced cooling. The loss of forced circulation results in a slow heatup of the core. Natural circulation within the core redistributes heat and enhances the conduction and radiative heat transfer from the core, but the core temperature peaks at approximately 1500°C after 95 h, beyond which the core heat removal rate exceeds the afterheat generation and results in a slow cooldown.

Isolation of the leaking steam generator limits steam ingress for SRDC-6 to a maximum of 8900 lb. However, due to the increase in pressure the primary coolant pressure relief valve opens and much of the steam has been vented by the time the core temperature has increased to about 1250°C.

Leakage of moisture into the primary coolant system of the MHTGR raises the potential for a reactivity increase, fuel hydrolysis, graphite oxidation, and venting of primary coolant through the primary system pressure relief valve.

The moisture in the primary coolant is available for chemical attack. The carbonaceous matrix materials used as binders in the fuel compact, in the lumped burnable poison (LBP) compact, and the reserve shutdown control (RSC) pellets consists of finely divided graphite flakes bonded together with residual carbon from the carbonized pitch binder. The function of the matrix is to provide a stable, refractory bond between components such as fuel particles or poison materials. The matrix may be corroded by coolant impurities, principally H₂O. Under certain circumstances, these matrix materials may serve as getters and
protect the fuel particles and $\text{B}_4\text{C}$ control materials from corrosive agents. However, if the corrosion of the matrix is extensive, there could be deleterious effects. For example, oxidation could potentially lead to loss of structural integrity for the fuel compact with reduced thermal conductivity and associated higher temperatures. In the $\text{B}_4\text{C}$ containing compact, complete oxidation of the matrix could decrease the height of the $\text{B}_4\text{C}$ column in the core.

As part of the MHTGR design effort, analytical codes were used to determine the impact that compaction (slumping) of the fuel, $\text{LBP}$ and $\text{RSC}$ materials would have on the MHTGR core reactivity. Since laboratory data are not available, it was assumed that after approximately 30% of the matrix had oxidized, the stack collapsed in order to fill in the void left as the matrix oxidized. It was found that compaction of the fuel stacks result in a negative reactivity coefficient. However, if oxidation of the matrix decrease the height of the $\text{LBP}$ and $\text{RSC}$ columns, the worth of the poison is reduced significantly and a positive reactivity coefficient occurs.

The OXIDE computer code (Ref. 2) is used extensively to determine the localized oxidation by steam of the bulk moderator core graphite and the extent of the reaction of steam with the fuel rod matrix material.

Based on defined (input) primary system transients and oxidant ingress rates, OXIDE solves mass continuity and state equations for all gaseous species assuming perfect mixing in core inlet and outlet plenums. The core geometry is modeled by multiple regions with a variable number of columns and with axial segments that can extend beyond the active core to include reflector and core support blocks. Up to 20 individually characteristic regions can be treated with up to 10 variable-power axial segments in each region. In each segment a typical element of symmetry (triangular in shape) around HTGR element coolant channel is modeled with 17 nodes, which extend from the center of a fuel rod to the center of an adjacent coolant hole. The general
transient diffusion equation for heat or mass transport is solved implicitly with variable properties and a source or sink term. In-core diffusion of reacting gases, oxygen and steam, and product gases, carbon monoxide and hydrogen are simulated along with local chemical reaction in the graphite and fuel. Accumulative fission product release due to graphite oxidation and fuel hydrolysis is computed. Plant protective system actions of moisture detection and reactor trip, steam generator isolation and dump, and safety valve pressure relief to the reactor building can be modeled. Thermodynamic conditions in the reactor building are also treated.

OXIDE analyzes the reactions of steam with the hot core graphite based on the Langmuir-Hinshelwood equation for the reaction rate:

\[ R_{st} = \frac{K_1 P_{st} F_b F_c}{1 + K_2 P_{hyd} + K_3 P_{st}} \]  \hspace{1cm} (1)

where

- \( R_{st} \) = the local mass fraction or percent reacting, \%/h
- \( P_{st}, P_{hyd} \) = partial pressures of steam and hydrogen, atm
- \( F_b, F_c \) = linear modifiers for burnoff and catalyst effects

The temperature-dependent Arrhenius coefficients

\[ K_i = k_i \exp\left(-\frac{E_i}{RT}\right) \]  \hspace{1cm} (2)

are derived from experimental data, as is the exponential constant \( n \). Depending on the relative magnitude of the terms in the denominator, the reaction goes from a first-order reaction at low water pressures to a zero-order reaction at high water pressures (approximately 1 atm).

During a moisture ingress into the primary circuit of the MHTGR there are typically six flowing mass species. The major species is the helium primary coolant, while the others are contaminants. Steam, oxygen, and nitrogen are present in the system due to the leak, and
carbon monoxide and hydrogen occur as reaction products from the steam-graphite and oxygen-graphite reaction.

OXIDE solves the mass diffusion equation for flow of each contaminant species. Within the core graphite these gases are diffusing through helium-filled pores of varying length and pore diameter. This inhibits the rate of diffusion. To account for this, OXIDE uses an effective diffusion coefficient ($D_{\text{eff}}$) to calculate the rate of diffusion in each carbonaceous component. Typically, for core graphite $D_{\text{eff}}$ is $<1\%$ of the true gas phase diffusion coefficient. For rod matrix material, however, the increased porosity should result in a higher $D_{\text{eff}}$. Currently the magnitude of $D_{\text{eff}}$ in matrix is uncertain.

3. JUSTIFICATION FOR INTEGRAL FUEL ELEMENT OXIDATION TEST

The accident analysis in the Preliminary Safety Information Document (PSID) for the MHTGR (Ref. 1), utilized the most recent values for Arrhenius coefficients (Eq. 2) for oxidation of each of the carbon components of the MHTGR core. However, for the graphite components, these coefficients were obtained using a microbalance technique at temperatures only up to approximately 1000°C, and extrapolated up to the accident temperatures using Eq. 2. Determination of the rate factors using the microbalance technique is not possible at temperatures exceeding approximately 1050°C, because the high gas velocity needed to prevent a transport limiting situation causes unacceptable oscillation of the sample. Thus, integral tests are needed to verify that this extrapolation of the rate coefficients to higher temperatures is valid. In addition, the data base in support of the assumed matrix oxidation rate is extremely limited and the integral tests are needed to verify that the correct order of magnitude rates have been used in predicting oxidation.

The integral oxidation tests are designed so that a fuel rod is exposed to water vapor in a geometry which simulates that in the MHTGR.
The results of these tests would not only verify that extrapolation of oxidation rate to higher temperature is valid, but they would also lend additional support to the assumption that the complex mathematical chemical reaction and diffusion equations used in OXIDE are correct for MHTGR core geometry. Because of the similarity of fuel and poison compact matrix material the work with fuel compacts will be applicable to conclusions regarding poison material oxidation.

Because the test rod is surrounded by a rigid structure of graphite in the integral test, this test is an ideal experiment in which to measure the degree of particle debonding and subsequent compaction of the fuel (or burnable poison) columns as a function of the fractional matrix burnoff. This could not be determined in unconstrained tests as a consequence of particle spalling as the rod matrix oxidizes.

The integral oxidation tests would experimentally demonstrate that the carbonaceous material in the core can withstand these higher temperatures and high moisture conditions, thus maintaining fuel and poison geometry in the core.

There currently exist two Design Data Needs (DDN M.10.18.23 and M.10.18.24) which identify several aspects of the steam graphite oxidation reaction where additional intrinsic oxidation rate data is needed to substantiate predicted performance (Ref. 3). These DDNs are being modified to include these integral oxidation tests since conducting the integral oxidation test will help to clarify which material components and aspects of the steam graphite oxidation need more characterization.
In summary, there is need for a series of integral steam oxidation tests of MHTGR core components in order to lend support to predictions made by the OXIDE computer code. These tests are needed to:

1. Verify that the temperature extrapolation used for the oxidation of fuel block graphite is valid.

2. Experimentally demonstrate that fuel rod matrix material can continue to provide the required function of maintaining the fuel and poison geometry during the licensing basis events with water ingress.

3. Determine if additional testing is needed to characterize intrinsic oxidation properties of individual fuel components.

4. EXPERIMENTAL

4.1 Test Description

Integral oxidation tests are proposed to simulate MHTGR accident conditions by exposing a mockup of an H451 fuel block containing a fuel rod to flowing wet helium in a once through system. Figure 1 shows the integral test geometry in which fuel rods and graphite are oxidized together at 1000°C, 1100°C, and 1250°C.

Under these conditions, depletion of oxidant water vapor will occur resulting in an oxidation profile. The matrix material which binds fuel particles and PyC-coated B₄C into rods and RSC pellets is thought to be 10 to 20 times more reactive than graphite. Thus, some degree of debonding, particularly at the surface of the rods and pellets, could occur. Since the reaction rate of matrix should be independent of the type of particle in the rod, only fuel rod samples will be tested.
4.2 Test Matrix

Table 1 shows the proposed test matrix. Calculations show that above about 0.3 atm water vapor, the rate of H-451 oxidation, is independent of steam pressure. Since this partial pressure is easily obtained with a room temperature water saturator, it is proposed to conduct the initial tests with this concentration. In addition, hydrogen is added to the reactant gas in order to insure that the concentration of hydrogen is constant at the local level throughout the test specimen.

Several entries in Table 1 are To Be Determined (TBD) during the course of the integral experiments. The % matrix burnoff is not predicted in Table 1, because matrix oxidation has not been studied above about 950°C and because no water vapor dependence has been measured. Furthermore, no tests on the current matrix composition have been conducted.

The longest oxidation time at each temperature is also not specified in Table 1. These times will be chosen to give a matrix burnoff close to 100% and are thus based on the results of the other integral tests.
Figure 1. Schematic Drawing Showing Integral Test Geometry
TABLE 1
INTEGRAL TEST MATRIX

<table>
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<tr>
<th>Temp (°C)</th>
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(a) To be determined.

(b) Approximately 80 L/min He and 0.3 atm H₂O with 1% H₂.
5. REFERENCES

