The Behavior of HTR Fuel Under Irradiation

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SUMMARY

In the FRG, three high-enriched fuel particle types were developed and qualified in irradiation tests from 1977 to 81. These are Variant 1: (Th,U)O₂ BISO²; Variant 2: (Th,U)O₂ TRISO³; and Variant 3 comprising UC₂ TRISO/ThO₂ TRISO particles. The goal of this program was to demonstrate acceptable performance for: HTR⁴ steam cycle applications with 700 - 750°C; gas turbine applications with 850°C; and process heat applications with 900 - 950°C He coolant outlet temperatures.

Recently, Germany switched to low-enriched uranium fuels for HTR, and the reference particle is now UO₂ TRISO.

As part of the conceptual core designs for future HTRs, fuel performance must be predicted in terms of: particle failure; fission product release from failed particles; and fission product release from intact particles. Based mainly on the FRG program to qualify high-enriched fuel, this paper presents the status of the structural analysis in modeling failure under normal operating and accident conditions.

In-pile failure is low, because particles have been designed by taking account of the pressure vessel performance model. Few data are available at the higher failure fractions where comparison with the theory is possible. Nevertheless, refined performance models are helpful in particle design and in making predictions under conditions outside the experimental range. Under accident conditions, good agreement between models and predictions can be observed. Further work is required to achieve a more consistent modeling of the transition from normal operating to accident conditions. Verification of the LEU⁵ irradiation program remains as the major outstanding issue.

a) The BISO coating consists of a porous buffer layer and a dense pyrocarbon (PyC) layer
b) The TRISO coating consists of buffer and dense PyC/dense silicon carbide (SiC)/dense PyC layers
c) High Temperature Reactor
d) Low Enriched Uranium
1. **INTRODUCTION**

The coated particle of ≤1 mm dia. is the basic structural unit of HTR cores. These miniature fuel elements consist of a central microsphere of heavy metal encapsulated by multiple coating layers. The series of coating layers serve a dual purpose: to provide structural integrity of the coated particle; and to remain retentive of fission products. Fuel failure is defined by the breach of all coating layers. The target of the qualification program has been to demonstrate $<6 \times 10^{-5}$ failure fraction from manufacture and $<2 \times 10^{-4}$ irradiation induced failures under normal operating conditions.

The buildup of gas pressure leads to stresses eventually exceeding coating layer strength. This is the most basic failure mechanism and has been studied extensively. [1-10] The diagram below shows how the calculation of gas pressure is combined with changing SiC strength to arrive at a failure prediction which will be compared to experimental observations.

Irradiation induced shrinkage of PyC effectively prestresses the SiC layer thereby delaying the change from compression into tension. These tedious calculations are not required for accident conditions where PyC thermal creep relaxes stresses. On the other hand, it may become necessary to introduce the effects of silicon carbide corrosion. The diagram below shows the combination of mechanisms at work between 1400 and 2000°C.
At the extreme temperatures 2000 - 2500°C characteristic of hypothetical accidents, the details of TRISO particle design, prior burnup, and fuel composition are negligible by comparison to the dominating mechanism - silicon carbide thermal decomposition. The scheme comparing model predictions to experiments is shown below.

Chapters 2, 3 and 4 deal with assumptions and laws used in the failure predictions. The details of modeling both the release of short-lived noble fission gases to derive in-pile failure fractions and the release of stable and long-lived fission gases are summarized in chapter 5.

2. STRUCTURAL MODELING OF PARTICLE FAILURE DURING IRRADIATION

The usual relationships to compute the stress distribution in multilayered coated particles [4-7,14] have been used

- equilibrium between the stress components,
- compatibility between elongation and displacement, and
- constitutive law relating elongation with stress (elastic), thermal expansion, shrinkage and creep.

Computations have been simplified by taking account of the observation that SiC does not shrink and creep[10]. A further simplification was to assume that elastic and inelastic constants do not change with irradiation (with the exception of PyC shrinkage). The "classical" TRISO particle failure criterion has been applied, whereby particle failure occurs when "the maximum hoop stress within SiC exceeds the SiC ultimate tensile strength (UTS)".

The statistical nature of coated particle manufacture and the distribution of SiC UTS values have been implemented in the calculations as has been proposed previously [8,9]. As is typical for brittle materials, the SiC strength distribution follows a Weibull statistic with

\[ F(S) = 1 - \exp \left[ -\ln \left( \frac{2}{(S/UTS)^m} \right) \right] \]  \hspace{1cm} (1)

representing the cumulative probability for strength values between 0 and S. UTS is the median strength, and m is the Weibull modulus. In the failure predictions reported here, only fuels have been used where the (UTS,m) data pairs have been characterized before irradiation.
ation with the brittle ring test [11]. Furthermore, rings from one particle batch have been irradiated to obtain the variation of $(UTS,m)$ with fast fluence and irradiation temperature. In the particle failure predictions, the following correlations have been used

$$UTS_{(irr)} = \max \left[ UTS_0 (1 - \Gamma / \Gamma_S), 200 \text{ MPa} \right] ; \log \Gamma_S = 0.556 + 0.065 \text{ E4}/T$$

(2)

$$m_{(irr)} = \max \left[ m_0 (1 - \Gamma / \Gamma_m), 2 \right] ; \log \Gamma_m = 0.394 + 0.065 \text{ E4}/T$$

(3)

where subscript 0 stands for the unirradiated state. Fast fluence $\Gamma$ is given in units $10^{25}$ m$^{-2}$. Equivalent Dido Nickel, temperature $T$ in K. Minimum values of 200 MPa and $m = 2$ are plausible values expected for poor material.

The number of gas atoms per heavy metal atom is given by $n_g / n_{HM} = (FY_F + O/f) F_{Bu}$. $FY$ is the fission yield for all xenon and krypton isotopes, $F_F$ is the fractional gas release. $O/f$ is the number of oxygen atoms released per fission, and $F_{Bu}$ is the fractional heavy metal burnup (FIMA). The gas release model (Model Ia) is detailed in chapter 5. Oxygen release has been assumed as follows

$$O/f = 0 \text{ for carbide and oxy carbide fuels;}$$

(4)

$$\log (O/f) = -1.08 - 0.85 \text{ E4}/T + 2 \log t \text{ for UO}_2 \text{ fuels \cite{12}};$$

(5)

$$\log (O/f) = 1.24 - 0.478 \text{ E4}/T + 0.5 \log N_F \text{ for (Th,U)O}_2 \text{ fuels.}$$

(6)

$N$ is the thorium/U 235-atom ratio, $F_{Bu}$ fractional FIMA, $T$ (K) irradiation temperature, and $t$ (s) irradiation time.

The Redlich-Kwong equation of state \cite{13} is used to compute the gas pressure in the void volume in the buffer layer and within the kernel porosity. The differences in kernel swelling of oxide fuels (1 volume % per % FIMA) and carbide fuels (0.2 % / % FIMA) have been accounted for.

Observed failure levels shown in fig. 1 have been derived from in-pile gas release measurements of short-lived fission gases (Model II; see chapter 5). Failure fraction predictions have been performed both without and with additionally modeling SiC corrosion, i.e., SiC thinning due to fission product-silicon carbide interaction. Bongartz' corrosion model \cite{15} has been used. Computational results shown in Table 1 and Fig. 1 represent a first attempt to reconcile a large number of independent measurements and models with overall irradiation results. It is clearly visible that the fuel development program is geared to investigate in-pile particle failure behavior, rather than providing systematic data for performance modeling. Invariably, experiments with observed failure fractions $\Phi_{exp} > 10^{-3}$ are older tests where the TRISO particle design was not optimized and/or testing conditions were extreme.

The described model is satisfactory because the predicted failures higher than $10^{-4}$

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a) Particles irradiated in the tests 8R2-P25, R2-K12, and FRJ2-P23 (see table 1)
are more conservative than the experimentally observed ones and for predicted failures lower than $10^{-5}$ no broken particles have been observed. However only test BR2-P23 failure level $7 \times 10^{-4}$ deviates significantly from the model prediction. One hypothesis is that in this test moisture led to hydrolysis which increases gas release from failed $U_{2}O_{2}$ particles by a factor 20. This would give a BR2-P23 failure fraction of $3.5 \times 10^{-5}$ well within the specified limit for failure from manufacture.

Structural model predictions including corrosive effects for test BR2-P21 have been reported previously [15] without using the decrease in SiC strength. With both mechanisms at work, failure is overpredicted which, however, is satisfying given the extreme irradiation conditions of this experiment.

3. MODELING PARTICLE FAILURE AT ELEVATED TEMPERATURES 1400 - 2000°C

At temperatures in excess of 1400°C, the calculation of stress distributions are simplified by virtue of PyC thermal creep relaxing the SiC prestressing. However, experimental observations have shown irradiation induced SiC embrittlement [16]. This will essentially increase the number of flaws that are present, e.g., due to widening of the grain boundaries or production of the internal cavities of significant size, so that SiC will tend to be weaker. If we assume that grain boundary corrosion results primarily in an increase in the range of flaw sizes in the SiC layer, failure will then occur over a wider range of tangential stresses. Mathematically, this has been simulated by

\[
m(\text{ann}) = m_0 \left[ 0.44 + 0.56 \exp \left( - \frac{t}{\tau_m} \right) \right]
\]

(7a)

where $m_0$ denotes the modulus before annealing, $t(s)$ annealing time. $\tau_m (s)$ is given by the correlation

\[
\log \tau_m = 0.248 + 0.979 \frac{E4}{T},
\]

(7b)

where $T$ is the annealing temperature simulating an accident. Model predictions at 1400/1600/1800/2000°C are shown in Fig. 2 in comparison with experimental results obtained by Schenk [17].

4. MODELING PARTICLE FAILURE AT EXTREME TEMPERATURES

Experiments in the temperature range 2000-2500°C are performed to obtain data for the risk analysis of hypothetical accidents. Modeling is greatly simplified because the underlying mechanism is simply SiC thermal decomposition. Failure is described by a Reliability Analysis approach where the failure rate - $\dot{R}/R = \Phi/(1-\Phi)$ is set proportional to a scaled time variable to a positive exponent. Here, $R = 1 - \Phi$ is the survival curve (reliability function), and $\Phi$ is the failure fraction. Choosing appropriate constants leads to

\[
\Phi = 1 - \exp \left\{ -(kt)^{m} \right\}.
\]

(8a)
A value of parameter \( m \) in excess of 1 describes wear-out of a component, while \( m = 1 \) stands for random failure, and \( m < 1 \) represents infant mortality. Particle failure increases steeply with annealing time [18] at temperatures towards 2500°C, thereby requiring a \( m \)-value significantly larger than one. However, there is little difference between various fuel types and between irradiated and unirradiated particles [19].

As for any thermally activated process, one would expect an Arrhenius-type correlation for the scaling parameter \( k \). Based on a wide range of experiments, linear correlations as shown in Fig. 3 have been obtained by the transformation

\[
\log \left( \frac{1}{\Phi} \right) = m(8.80 - 3.50 \, \text{E4}/T) + m \cdot \log t; \quad m = 4; \tag{8b}
\]

where \( \Phi \) is the failure fraction, \( t(s) \) annealing time, \( T(\text{K}) \) annealing temperature.

5. GAS RELEASE MODELING

Fission Product Transport and Release in Fuel

The Equivalent Sphere model [20] for fuel release from UO₂ or ThO₂ leads to an easy diffusional model via simulating the fuel grains by a sphere of radius \( a \). The diffusion equation is derived by combining the mass balance equation

\[
\int (p - D \frac{\partial c}{\partial t}) \, dV = \frac{\partial}{\partial t} \int (D \frac{\partial c}{\partial s}) \, dS
\]

with Fick's law \( \nabla = -D \text{grad} c \) to give \( \frac{\partial c}{\partial t} = \text{div} (D \text{grad} c) \cdot \frac{\partial c}{\partial t} + p \),

where \( p \) and \( c \) are volume specific source term and concentration, \( j \) the diffusive flux, \( V \) and \( S \) are volume and surface of a given element. The kinetic constants \( \lambda \) and \( D \) are radioactive decay constant and diffusion coefficient. Release from fuel grains is usually modeled with infinitely fast evaporation from the grain surface: \( -D \text{grad} c \, (r = a) = \alpha c; \lambda \to \infty \). This is equivalent to \( c(r = a) = 0 \). Solutions in the Equivalent Sphere model have \( D' = D/a^2 \) as a parameter, denoted Reduced Diffusion Coefficient.

Shortlived Fission Gases (Model II)

Both for reactor applications and for fuel test monitoring, the isotopes Kr 85m, Kr 88, Kr 87, Xe 133, and Xe 135 are important. In the temperature range 800 – 1200°C, typical of HTR fuel, radioactive decay is faster than diffusion, i.e. \( D' < \lambda \).

The steady state solution of the diffusion equation gives

\[
\frac{R/B}{\lambda} = 3 \sqrt{D'\lambda} \left( \coth \sqrt{\frac{4}{D' \lambda}} - \frac{1}{\sqrt{D' \lambda}} \right), \tag{9}
\]

where \( R/B \) is the ratio of release/birthrate.

Gas release from coated particle HTR fuel indicates failure of the complete coating. However, fission products do not only recoil into fuel grains, but also into pores and into the surrounding graphite buffer layer. Also, a minor amount of heavy metal contamination in
the buffer layer may contribute to the graphite grain/pore source term. While release from pores is assumed to be instantaneous, some diffusional delay is assumed from graphite. Consequently, gas release from defect particles is modeled by

\[ \frac{R/B}{\text{def}} = g_F \left( R/B \right)_F + g_C \left( R/B \right)_C + g_P, \]

(10)

where the subscript F stands for fuel, C for carbon and P for pores. The parameters \( g_F, g_C \) and \( g_P \) are derived from fuel characterization data, \( D'_F \) and \( D'_C \) have been obtained from an irradiation experiment where defects have been simulated by laser holes through the coating \([21]\). Typical values for krypton release from \((\text{Th}_2\text{U})_2\text{O}_2\) fuels are \( \log D'_F = 1.80 - 1.64 \text{E}4/\text{T}, \log D'_C = -5.60 - 0.284 \text{E}4/\text{T} \), where \( D' \text{ (s}^{-1}) \) are reduced diffusion coefficients, and \( T(\text{K}) \) is the irradiation temperature. In a large assembly of coated particles, the failure fraction can be obtained by dividing measured \( R/B \)s by the model prediction \( R/B \) per defect particle. The majority of failure levels in Fig. 1 have been obtained this way from in-pile gas release data.

Release of Long-Lived Fission Products from Fuel (Model I)

The release of stable and long-lived fission products is modeled by setting \( \lambda = 0 \) giving \( \partial c/\partial t = D \text{ div (grad } c) + p \), and by assuming zero concentration at grain surfaces. The fraction released after irradiation for \( t(s) \) is given by (Model Ia)

\[ F_i = 1 - \left(6/D'_i t_i\right) \sum_{n=1}^{\infty} \left[1 - \exp\left(-n^2\pi^2 D'_i t_i\right)\right]/[n^4 \pi^4]. \]

(11)

After cold irradiation with negligible kernel release, the fraction of fission products released in an annealing experiment is given by (Model Ib)

\[ F_a = 1 - \left(6 \sum_{n=1}^{\infty} [\exp\left(-n^2\pi^2 D'_i t_i\right)]/[n^2 \pi^2]\right). \]

(12)

If an irradiation for time \( t_i \) is followed by an anneal for \( t_a \), the fraction released is given by

\[ F_{i+a} = 1 - \left(6/D'_i t_i\right) \sum_{n=1}^{\infty} \left[1 - \exp\left(-n^2\pi^2 D'_i t_i\right)\right] \times \left[\exp\left(-n^2\pi^2 D'_a t_a\right)\right]/[n^4 \pi^4]. \]

(13)

6. CONCLUSIONS

In the evolutionary process establishing fuel performance models for normal operation and accidents, practically all sub-models have been validated and provide a realistic base for prediction. In the process, coated particle design and manufacturing conditions/quality assurance have been optimized so that the classical pressure vessel failure is extremely low. Therefore, few data exist to verify the model predictions which combine all sub-models. Nevertheless, the presently available refined stress model is helpful in particle design, should new requirements arrive, and in making predictions under conditions outside the experimental range.
Under accident conditions, good agreement between models and predictions can be observed. While there is some uncertainty in the input data to the models, predictions can be used for the layout of safety experiments. Further work is required for a more consistent modeling of the transition from normal operations to accident conditions.

7. ACKNOWLEDGMENTS

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8. REFERENCES


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### Tab. 1: Fuel irradiation test data; calculated results on gas release, SiC strength and SiC corrosion; and coated particle failure predictions

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Fuel</th>
<th>Burnup (% FIMA)</th>
<th>Irr.Time (d)</th>
<th>Temp (°C)</th>
<th>Fluence (10²⁵ m⁻²)</th>
<th>Gas atoms per Fission</th>
<th>SiC-UTS (MPa)</th>
<th>m</th>
<th>SiC-Thick. (μm)</th>
<th>Predicted Failure Fraction</th>
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a) Lower SiC thickness represents the effect of fission product - SiC interaction (corrosion) leading to a thinner SiC layer
Fig. 3: Percentage particle failure predicted in extreme temperature accidents. Comparison to results of annealing experiments with irradiated and unirradiated fuel.