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Title: “Evaluation of Design Parameters for TRISO-coated Fuel Particles to Establish Manufacturing Critical Limits Using PARFUME”

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Abstract

The success of modular high temperature gas-cooled reactors is highly dependent on the performance of the tristructural-isotopic (TRISO) coated fuel particle and the quality to which it can be manufactured. During irradiation, TRISO-coated fuel particles act as a pressure vessel to contain fission gas and mitigate the diffusion of fission products to the coolant boundary. The fuel specifications place limits on key attributes to minimize fuel particle failure under irradiation and postulated accident conditions. PARFUME (an integrated mechanistic coated particle fuel performance code developed at the Idaho National Laboratory) was used to calculate fuel particle failure probabilities. By systematically varying key TRISO-coated particle attributes, failure probability functions were developed to understand how each attribute contributes to fuel particle failure. Critical manufacturing limits were calculated for the key attributes of a low enriched TRISO-coated nuclear fuel particle with a kernel diameter of 425 μm . These critical manufacturing limits identify ranges beyond where an increase in fuel particle failure probability is expected to occur.

1. Introduction

Critical to the success of modular high temperature gas-cooled reactors is the performance of the TRISO-coated fuel particle and the quality level it can be manufactured. The TRISO-coated fuel particle consists of a fuel kernel and buffer layer, inner pyrolytic carbon (IPyC) layer, silicon carbide (SiC) layer, and outer pyrolytic carbon (OPyC) layer (Fig. 1). The TRISO-coated fuel particles serve as a pressure vessel to contain fission gas and fission product migration during irradiation. The fuel specifications put limits on key attributes to minimize fuel particle failure under irradiation and postulated accident conditions. Fuel performance codes simulating these conditions can be used to develop fuel specifications and determine critical limits on material characteristics for use in a high temperature gas-cooled reactor. The Idaho National Laboratory has developed the fuel performance code PARFUME [1] to calculate the failure probability of a population of TRISO-coated fuel particles under irradiation conditions and determine such limits in the frame of the high temperature gas-cooled reactor program.

PARFUME is an integrated mechanistic computer code that evaluates the thermal, mechanical, and physicochemical behavior of coated fuel particles and the probability for fuel failure given the particle-to-particle statistical variations in physical dimensions and material properties that arise during the fuel fabrication process. The objective in developing PARFUME is to physically describe both the mechanical and physico-chemical behavior of the fuel particle under irradiation and postulated accident conditions, while capturing the statistical nature of the fuel. The PARFUME code has been developed to determine the failure probability of a population of fuel particles, accounting for all viable mechanisms that can lead to particle failure. In addition, PARFUME calculates fission product transport and accounts for these particle failures by determining the diffusion of fission products from the fuel through the particle coating layers and their subsequent release through the fuel matrix to the coolant boundary.

Coated particle fuel exhibits statistical variations in physical dimensions and material properties from particle to particle due to the nature of its fabrication process. Particle behavior is also inherently multidimensional, further complicating development of the model. The failure probability of a batch of fuel particles depends on statistical variations in the fuel design parameters as well as variation in the characteristic strengths of the coating layers in a batch. The calculation of fuel particle failures implemented in PARFUME samples the fuel design parameters from a Gaussian statistical distribution and the layer strengths are sampled from a Weibull statistical distribution [2,3]. PARFUME allows for statistical variations in the kernel diameter, the four layer thicknesses, the pyrocarbon densities, the degree of anisotropy of the pyrocarbons (as measured by the Bacon anisotropy factor [BAF]), the creep coefficient for the pyrocarbons, Poisson's ratio in creep for the pyrocarbons, bond strength between the IPyC and SiC layers, and particle asphericity (as measured by the aspect ratio).

The Department of Energy's (DOE) Advanced Gas Reactor (AGR) program is a nuclear fuel development and qualification campaign that supports licensing, deploying, and operation of a high temperature gas-cooled reactor [4]. The irradiation test AGR-5/6 is the fourth in a series of experiments that will use fuel manufactured from engineering-scale equipment to demonstrate the performance of fuels fabricated in a production environment. Previous AGR experiments utilized fuel manufactured

partially or completely in a lab-scale production environment. The AGR-5/6 fuel contains low-enriched uranium carbide/oxide (UCO) kernels with a TRISO coating and an overcoating pressed in a carbonaceous matrix to form a fuel compact [5]. After fabrication, the fuel compacts will be irradiated in the Advanced Test Reactor (ATR) at Idaho National Laboratory and subjected to high temperature heat-up tests after irradiation to simulate accident conditions. The results from the AGR-5/6 experiment will be used to qualify TRISO-coated fuel particles for use in a high temperature gas-cooled reactor.

The critical limits for various TRISO-coated fuel particle attributes were investigated with the fuel performance code PARFUME. These limits are used to establish particle attribute values that could result in increased fuel particle failure. Fuel particle failure probability was investigated by sampling each particle attribute considering the manufacturing variability for the sample population defined by its mean and corresponding standard deviation. Discrete PARFUME calculations were performed at each selected value within the attribute range while the other attributes were only varied about their means using their corresponding standard deviations. This analysis provides a failure probability curve within the range for each attribute that can then be used to determine the cumulative failure fraction curve to establish the critical limits for that attribute.

Fig. 1. Typical TRISO-coated particle geometry.

2. Material and Method

The irradiation conditions, fuel particle geometry, and nominal material properties used in this analysis are summarized in Table 1. These values were selected as a basis for typical operation and represent a standard particle for the DOE AGR program. Either a high- or low- irradiation temperature was used depending on which value resulted in higher failure probability for a given attribute. The high

temperature value was used for the buffer thickness and particle asphericity analysis where typical pressure vessel failures are of particular concern while the low temperature value was used for the remaining attributes which influence particle failure associated with IPyC cracking because of reduced irradiation-induced creep at lower temperatures. In both cases, the temperature was held constant during irradiation. The burnup and fast neutron fluence ramp linearly during the specified duration of the irradiation. Intrinsic in the PARFUME code are additional material properties that are consistent with the existing data that are available for TRISO-coated fuel particles [1,6].

The attributes considered in this analysis are listed in Table 2. Table 2 also provides the range over which the attribute was analyzed, the specific values at which each calculation was performed, and the irradiation temperature for the analysis. The fuel performance motivation for selecting these attributes is also summarized in Table 2, which were previously analyzed and found to warrant further investigation [7]. The range of values of each attribute cover a wide range of potential manufacturing variations to capture the potential tails of the distribution in establishing the critical limits. The specific values analyzed within the range were selected to cover the low and high ends of the distribution and to ensure there was sufficient breadth in the range so an increased failure probability would be captured within the range.

Table 1. Input parameters for failure calculations.

Attribute	Units	Value
Irradiation duration	Effective Full Power Days	400
End-of-life burnup	% FIMA	16
End-of-life fluence	10^{25} n/m ² [$E_n > 0.18$ MeV]	5
Irradiation temperature	K	1173 (Low) 1573 (High)
Ambient pressure	MPa	0.1
Kernel diameter	μm	425 ± 10
Buffer thickness	μm	100 ± 10
IPyC thickness	μm	40 ± 4
SiC thickness	μm	35 ± 3
OPyC thickness	μm	40 ± 4
Particle asphericity		1.04 ± 0.02
Kernel density	Mg/m ³	11.00
Buffer density	Mg/m ³	1.04
IPyC density	Mg/m ³	1.90 ± 0.02
OPyC density	Mg/m ³	1.90 ± 0.03
IPyC BAF		1.04 ± 0.005
OPyC BAF		1.03 ± 0.005
U-235 enrichment	Weight %	15.50
Oxygen-to-uranium	Atom Ratio	1.50
Carbon-to-uranium	Atom Ratio	0.39

Table 2. Attributes addressed to determine critical limits of AGR-5/6 fuel.

Attribute	Range of Values Analyzed	Specific Values	Temperature of Analysis (K)	Fuel Performance Concern
IPyC thickness (μm)	10 - 70	10, 20, 30, 40, 50, 60, 70	1173	Thickness influences stress in the IPyC layer and thus its propensity to fail.
IPyC density (Mg/m^3)	1.8 – 2.0	1.80, 1.85, 1.90, 1.95, 2.00	1173	Density in part determines dimensional change and thus determines IPyC failure probability.
IPyC BAF	1.0 – 1.16	1.00, 1.01, 1.02, 1.03, 1.04, 1.05, 1.06, 1.07, 1.08, 1.16	1173	Anisotropy is critical to determining the magnitude of the dimensional change and thus the IPyC failure probability.
OPyC density (Mg/m^3)	1.8 – 2.0	1.80, 1.85, 1.90, 1.95, 2.00	1173	Same reasoning as IPyC density.
OPyC BAF	1.0 – 1.16	1.00, 1.01, 1.02, 1.03, 1.04, 1.05, 1.06, 1.16	1173	Same reasoning as IPyC BAF.
SiC thickness (μm)	10 - 60	10, 20, 25, 30, 35, 40, 45, 50, 60	1173	Thickness of SiC layer influences overall particle failure probability.
Buffer thickness (μm)	10 - 150	10, 25, 40, 50, 60, 75, 100, 125, 150	1523	Determines available void volume for fission gases and subsequent particle pressure.
Particle asphericity (aspect ratio)	1.0 – 1.24	1.00, 1.02, 1.04, 1.06, 1.08, 1.10, 1.12, 1.15, 1.18, 1.21, 1.24	1523	Aspherical particles can generate tensile stresses in the SiC layer and cause failure.

3. Fuel Particle Failure Mechanisms

For the attributes considered at the low irradiation temperature condition, the primary failure mode of the SiC layer is caused by an irradiation-induced shrinkage crack in the IPyC layer. The crack creates a stress concentration in the SiC layer [8] that may or may not lead to total fuel particle failure. Since PARFUME is a one-dimensional fuel performance code and the effects of an IPyC crack on a TRISO fuel particle is multidimensional, the finite element analysis code Abaqus [9] was used to generate the required PARFUME input to estimate the stresses in the particle using the PARFUME statistical methodology [7]. The variations in the seven parameters listed in Table 2 (with the exception of particle asphericity) affect the stress levels in the SiC layer to some degree and require Abaqus calculations at each value analyzed with the other six attributes fixed at their mean values resulting in seven Abaqus calculations for each value listed [7]. The results from the Abaqus calculations are then used in PARFUME to correlate the stresses in a cracked particle to an intact particle at that attribute's specific

value. This methodology was performed for each specific value listed in Table 2 for each attribute requiring a total of 448 Abaqus calculations.

In addition, fuel particle failure due to typical pressure vessel failure was calculated with consideration of particle asphericity. Again, the multidimensional nature of particle asphericity required the use of Abaqus to generate the multidimensional input required by PARFUME to correlate the stresses of an aspherical particle to a perfectly spherical particle using the PARFUME statistical methodology [10]. The degree of asphericity is measured in terms of the aspect ratio, defined as the ratio between the major and minor outer diameters for the particle. For each specific value listed in Table 2, nine Abaqus calculations were required for a total of 576 Abaqus calculations.

IPyC/SiC debonding was not considered in this analysis. This eliminates any particle failures due to debonding. The current fuel manufacturing practices have greatly improved the IPyC/SiC bond strength (100 MPa) resulting in zero fuel particle failures due to debonding as calculated by PARFUME [10].

4. Fuel Particle Failure Probability Results

PARFUME was used to calculate the probability of SiC failure (particle failure) for each of the discrete values listed in Table 2. Each attribute was fixed at the specified value listed while the remaining attributes were statistically varied about their mean values using the standard deviation listed in Table 1. The failure probability for each attribute is presented in Figs. 2 – 9. A failure probability function was then fitted for each attribute throughout the range specified except for asphericity (Fig. 8) where the curve is discontinuous and had to be integrated numerically.

The following key findings are noted below:

- There was little change in the failure probability as the attributes for density (both IPyC and OPyC) and BAF were varied over the range under consideration. The historical concern in the program regarding anisotropic caused us to examine BAF but the lack of response is probably due to the uncertainties in the material properties, especially irradiation-induced creep.
- When varying the thickness of the SiC layer, the failure probability increased as the thickness decreased. This can be attributed to less structural material to retain the fission gas pressure and subsequent increase in tangential stress in the layer.
- Conversely, failure probability increased as the IPyC layer thickness increased. This is due to the increase in pyrocarbon material causing an increase in shrinkage early in irradiation. This results in a higher IPyC cracking probability causing localized stress concentrations in the SiC layer.
- As the buffer thickness increases, the volume available to store fission gas increases resulting in a lower pressure and hence less stress in the SiC layer.
- For increases in aspect ratio, the Abaqus model used in the analysis treats asphericity essentially as a flat plate on one side of the particle. Increasing the aspect ratio increases the surface area of the flat plate increasing the stress in the SiC layer due to pressure accumulation.

Fig. 2. Failure probability versus SiC thickness.

Fig. 3. Failure probability versus IPyC thickness.

Fig. 4. Failure probability versus IPyC BAF.

Fig.5. Failure probability versus IPyC density.

Fig. 6. Failure probability versus OPyC BAF.

Fig. 7. Failure probability versus OPyC density.

Fig. 8. Failure probability versus buffer thickness.

Fig. 9. Failure probability versus aspect ratio.

5. Determination of Critical Limits

The failure probability functions were then used to determine the critical limits for the eight attributes under consideration. The cumulative failure fraction (F) transforms the previous point calculations to a value that is representative of a population of particles and is calculated using Equation 1:

$$F = \frac{\int_A^B n(x)p(x)dx}{\int_A^B n(x)dx} \quad (1)$$

Where:

F = failure fraction

A = lower integration limit of the range considered for the attribute in question.

B = upper integration limit of the range considered for the attribute in question.

$n(x)$ = Gaussian distribution, $n(x) = e^{\frac{-(x-\mu)^2}{2\sigma^2}}$

μ = mean value for the attribute considered

σ = standard deviation for the attribute considered

$p(x)$ = discrete failure fraction function as shown in Figures 2 – 8.

With the exception of the aspect ratio, the remaining seven attributes failure probability curves were fitted to obtain a failure probability function ($p(x)$) as shown in Figs. 2 – 8. The aspect ratio failure probability curve ($p(x)$) required the numerical integration (Fig. 9) to obtain the cumulative failure fraction.

The mean value of μ was set either to the lower or upper limit of the mean specification range depending on whether a lower or upper critical limit was to be determined. For example, the mean specification for the SiC thickness is specified as $35 \pm 3 \mu\text{m}$ and since a lower critical limit is desired, a value $\mu = 32 \mu\text{m}$ was used.

Using Equation 1 and the curve fit functions in Figs. 2 – 8, the cumulative failure fraction was calculated as a function of standard deviation (σ). Theoretically, Equation 1 is integrated from zero to infinity, however, for this analysis, the integration was performed between the minimum and maximum values specified in the range for each attribute considered. This assumption results in little loss of precision due to the very small contributions to the integrand from the tails. The results of the cumulative failure fraction for the eight attributes under investigation are presented in Figs. 10 – 17. Using Figs. 10 – 17, a critical standard deviation (σ_L) was determined by selecting the point on the cumulative failure fraction curve where there was an inflection or change in slope in the function F . This point can be considered the threshold at which the attribute becomes susceptible to increased particle failure and becomes the point at which the critical limit (x_L) can be established using the following equation [11]:

$$x_L = \mu \pm 2.326\sigma_L \quad (2)$$

Depending on whether it is a lower or upper critical limit, the plus or minus in Equation 2 is used for that attribute. The coefficient 2.326 was determined based on allowing only 1% of the particle population to lie outside the critical limit (see Reference [11] for a detailed statistical methodology) when the standard deviation is equal to the critical standard deviation (σ_L).

The results of the cumulative failure fraction analysis are summarized in Table 3 for each of the eight attributes selected. Included in Table 3 is the critical region of interest (lower or upper), mean value used in the analysis, the point of inflection in the cumulative failure fraction curve (σ_L), and the calculated critical limit (x_L). Because the OPyC BAF and OPyC density cumulative failure fraction curves are essentially flat, there are no critical limits for these attributes.

Fig. 10. Cumulative failure fraction (F) for SiC thickness.

Fig. 11. Cumulative failure fraction (F) for IPyC thickness.

Fig. 12. Cumulative failure fraction (F) for IPyC BAF.

Fig. 13. Cumulative failure fraction (F) for IPyC density.

Fig.14. Cumulative failure fraction (F) for OPyC BAF.

Fig. 15. Cumulative failure fraction (F) for OPyC density.

Fig. 16. Cumulative failure fraction (F) for buffer thickness.

Fig. 17. Cumulative failure fraction (F) for aspect ratio.

Table 3. Critical Limits Determined for Each Attribute.

Attribute	Critical Region	Mean Value μ (at edge of range)	Standard Deviation σ_L (at point of inflection)	Critical Limit x_L (per Equation 2)
IPyC thickness (μm)	Upper	44	3.5	52
IPyC density (g/cm^3)	Upper	1.95	0.02	2
IPyC BAF	Upper	1.045	0.02	1.09
OPyC density (g/cm^3)	Upper	1.95	None	None
OPyC BAF	Upper	1.035	None	None
SiC thickness (μm)	Lower	32	5	20
Buffer thickness (μm)	Lower	85	15	50
Particle sphericity	Upper	1.014	0.02	1.06

6. Conclusion

Critical manufacturing limits have been confirmed for the key attributes of a TRISO-coated nuclear fuel particle to identify ranges above or below which an increase in fuel particle failure is expected to occur. These limits are used to verify that the mean values specified in the AGR-5/6 fuel specification will not result in an abnormal increase in fuel particle failure. The range of each attribute was selected to ensure that the tails of the distribution were considered when establishing the critical limits. The critical limits identified are at or beyond those specified for the AGR-5/6 fuel particles and confirm historical critical regions used in the development of the fuel specification. As a result, increased fuel particle failure probability is not expected to occur for the AGR-5/6 experiment. The results described above are only applicable to a low enriched UCO fuel particle with a kernel diameter of 425 μm with similar coating layer thicknesses. This analysis would have to be extended to cover additional kernel sizes and fuel composition (i.e. UO_2).

Further, the material properties and fuel attributes selected for this analysis are based on historical values that have been associated with the manufacturing of TRISO-coated fuel particles. In addition, PARFUME is not a safety-validated tool and is only being used to identify possible regions in which an increase (or decrease) in fuel particle failure can be expected and for this reason, a recommended optimized TRISO fuel particle is not provided. However, the attribute range and critical

limits provided for typical TRISO-coated particles can be used as a baseline comparison for fuel particle failure when one parameter is singularly varied from its historical value.

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