The Challenges Associated with High Burnup and High Temperature for UO2 TRISO-Coated Particle Fuel

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ABSTRACT

The fuel service conditions for the DOE Next Generation Nuclear Plant (NGNP) will be challenging. All major fuel related design parameters (burnup, temperature, fast neutron fluence, power density, particle packing fraction) exceed the values that were qualified in the successful German UO$_2$ TRISO-coated particle fuel development program in the 1980s. While TRISO-coated particle fuel has been irradiated at NGNP relevant levels for two or three of the design parameters, no data exist for TRISO-coated particle fuel for all five parameters simultaneously. Of particular concern are the high burnup and high temperatures expected in the NGNP. In this paper, where possible, we evaluate the challenges associated with high burnup and high temperature quantitatively by examining the performance of the fuel in terms of different known failure mechanisms. Potential design solutions to ameliorate the negative effects of high burnup and high temperature are also discussed.

1. INTRODUCTION

The fuel service conditions for the DOE Next Generation Nuclear Plant (NGNP) will be challenging. Based on our knowledge to date, the highly successful German coated particle fuel program established an acceptable design envelope for the five key fuel-related parameters (burnup, temperature, fast fluence, particle packing fraction, power density). Table 1 and Figure 1 compare these parameters for the NGNP with those of other programs around the world. The results indicate that German fuel does not adequately envelope the conditions expected for the NGNP for any of these five key fuel-related parameters and neither does any other program around the world. Thus, additional fuel development will be required.

An assessment has been performed using modeling in the PARFUME code [Miller, 2004] to quantitatively evaluate the challenges associated with high temperature and high burnup with TRISO-coated particle fuel. There are a number of known fuel failure mechanisms that are temperature and burnup dependent. These include: thermomechanical response of PyC, fission gas release and CO production, amoeba effect, and Pd attack of the SiC. For each mechanism the effects of increasing burnup and/or temperature were evaluated and where possible the results normalized to results at 1100°C and 8% FIMA, the upper end of the German performance envelope. These numerical values then provide a metric to determine how the fuel performance will change as the temperature and burnup are increased.

Table 1. Comparison of fuel service conditions

	NGNP	Germany	Japan	South Africa	France	China
Burnup (% FIMA)	15-20	8	4	8-10	10-15	8
Temperature (°C)	1250	1100	1200	1100	1100-1200	1100
Fast Fluence (10^{25} n/m^2)	4	3.5	4	3.5	4	3.5
Packing Fraction (%)	< 35	10	30	10	10-15	10
Power Density (W/cc)	6	3	3-6	3	3-6	3
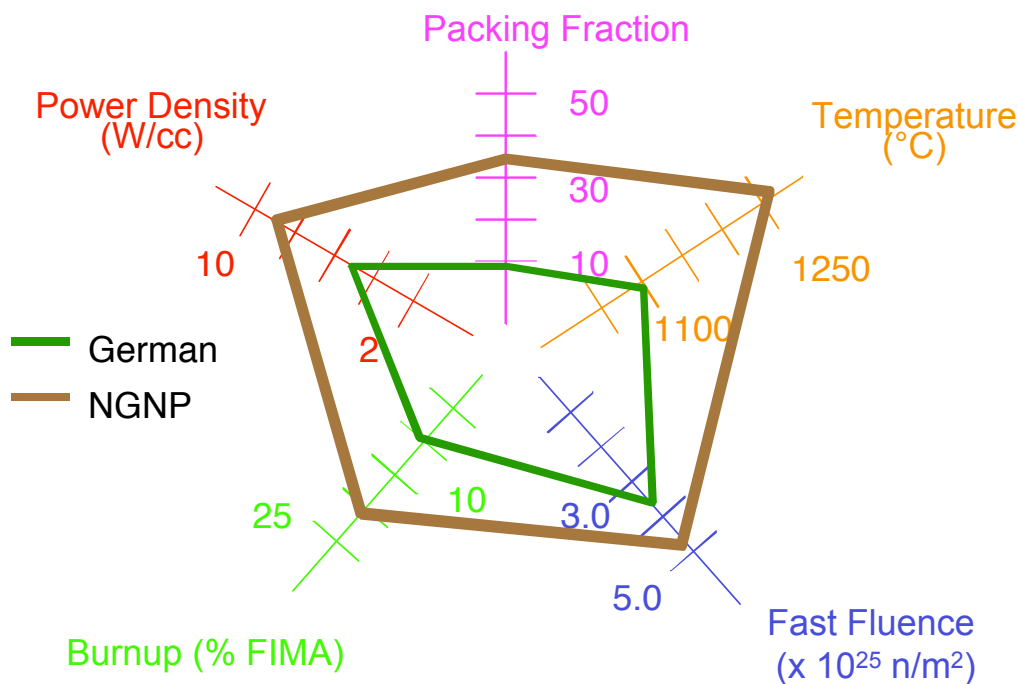
Figure 1. Comparison of NGNP and German fuel operating envelope.

2. CHALLENGES

2.1 Thermomechanical response of PyC

The shrinkage/swelling response of PyC is highly anisotropic and depends on the irradiation temperature and the isotropy of the PyC (as measured by the Bacon Anisotropy Factor (BAF)). As the irradiation temperature increases (see Figure 2), the shrinkage increases and the stress in the IPyC increases. Offsetting the shrinkage is irradiation-induced creep. Although the data is uncertain, the limited data available suggest that irradiation induced creep of PyC depends on the density of the PyC and the irradiation temperature. [Ho, 1993] (see Figure 3) The greater creep at higher temperature reduces stress in the IPyC layer of the particle. For the highly non-linear thermomechanical response of the coating system, creep dominates and the stress in the IPyC layer decreases as the irradiation temperature increases.
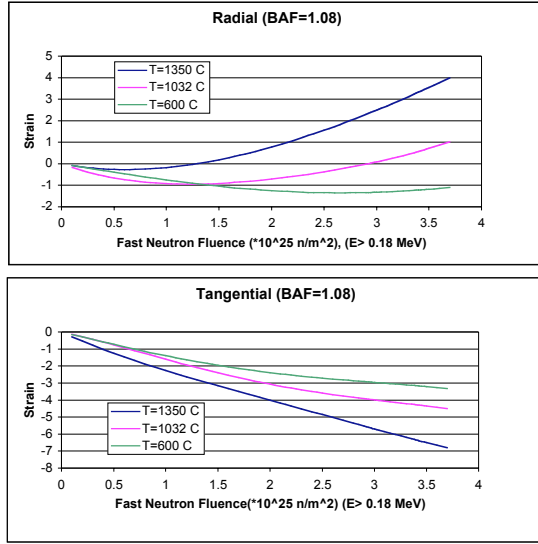
Figure 2. Radial and Tangential irradiation induced strains in PyC as a function of irradiation temperature and fast neutron fluence.
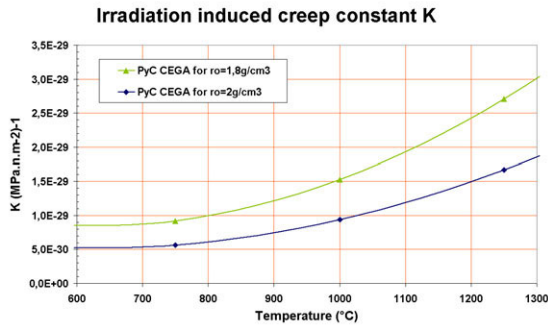

Figure 3. Irradiation induced creep of PyC

2.2 Fission Gas Pressure

Fission gases released during irradiation from the kernel of a coated particle depend on temperature, burnup and time. [Petti, 2004] Table 2 presents the normalized fission gas pressure that builds up in a 500 μm German UO_2 particle irradiated for three years at the indicated temperature and burnup. (The enrichment of the particle is assumed to scale with the burnup in this calculation.) The results indicate a factor of 8 increase in pressure as the burnup approaches 20% FIMA and the temperature approaches 1300°C.

2.3 CO Pressure

Oxygen is released during fission. In coated particle UO_2 fuels, there is net excess or "free" oxygen because the fission products that are produced do not consume all of the oxygen released. The excess oxygen reacts with the buffer to form CO gas. The amount of CO produced is a function of temperature and burnup. Depending on operating conditions and fuel design, the CO contribution to total internal pressure can be as high as four times the contribution from fission product gases. Table 3 presents the results of thermodynamic calculations of the CO pressure that builds up in a 500-μm German particle irradiated for three years at the indicated temperature and burnup. (The enrichment of the particle is assumed to scale with the burnup in this calculation.) The results indicate a factor of 4 increase in pressure as the burnup approaches 20% FIMA and the temperature approaches 1300°C. Under accident conditions, the pressure increase would be significantly higher.

2.4 Kernel Migration

Kernel migration is the tendency for the kernel to migrate up the temperature gradient. It has been observed in all UO_2 TRISO-coated fuel particles. The migration is a function of the kernel migration coefficient (KMC), temperature and temperature gradient. [Petti, 2004] (There is no burnup dependence.) An example of kernel migration is shown in Figure 4.
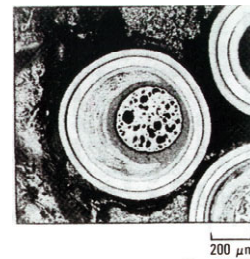

Figure 4. Migration of a UO_2 kernel.

The migration distance is given by the following relationship:

$$\bar{x}_{MIG} = -KMC \bullet \frac{\bar{\nabla} T}{T^2} d \bar{x}$$
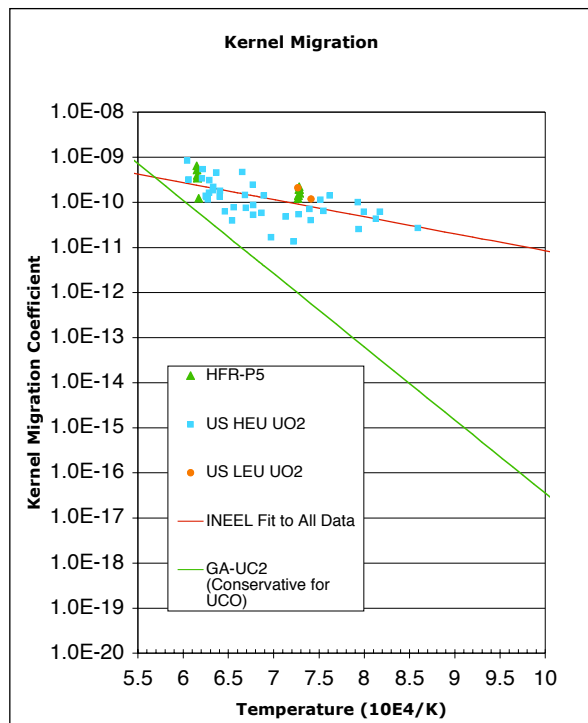
$$KMC = KMC_O \cdot \exp(-Q/RT)$$

Figure 5. Kernel migration coefficient for different fuel forms

composition of the kernel, although theoretically this could be important. Based on the international historical database the penetration rate of Pd into SiC is found to have an Arrenhius temperature dependence [Petti, 2004]. (see Figure 6). Table 5 below indicates that the rate of Pd penetration into the SiC is almost a factor of 3 greater at 1300°C than at 1100°C.
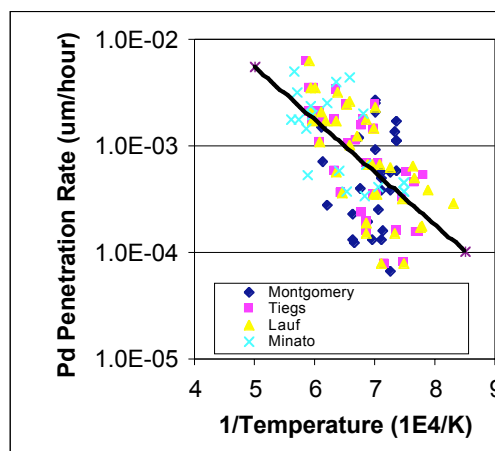

Figure 6. Pd penetration rate based on international data

For a given temperature gradient, the kernel migration distance will depend on the quantity (KMC/T^2). Figure 5 plots the migration coefficient versus inverse temperature. This quantity serves as a convenient metric for this phenomenon and is calculated as a function of temperature in Table 4. The results indicate that as the fuel temperature increases from 1100 to 1300°C the propensity for kernel migration in UO_2 TRISO-coated fuel particles increases by a factor of 1.7.

2.5 Pd Attack

Fission product palladium is known to attack SiC at localized reaction sites. These interactions have been the subject of extensive study. In high burnup LEU fuels, 25 to 50x more Pd is produced than in either high burnup HEU fuels or LEU low burnup fuels because of the large fraction of fissions from Pu that are expected at high burnup. As a result, the potential for Pd attack of the SiC could be higher in LEU high burnup fuels like that proposed for NGNP. A review of the international database shows no strong dependence on burnup or the

2.6 Cesium Release

The high temperature accident response of TRISO-coated particle fuel has been little studied, especially at the high burnups expected for the NGNP. German pebbles irradiated to burnups of 14% FIMA and fluences of 5-6 x 10^{25} (E > 0.1 MeV) have shown elevated releases of both cesium and noble gases compared to pebbles with burnups of < 10% FIMA heated to similar conditions (see Figure 7). [Gontard, 1990] The reasons for the increased release are not known with certainty.

A photomicrograph of the SiC in a coated particle from one such test is shown in Figure 8. [Gontard, 1990] The SiC layer from these particles does show some degradation. The Germans attributed the release to degradation of the SiC by fission products (cesium in particular) but no chemical analysis was performed to confirm that the degradation was due to fission products. Two hypotheses can be formulated concerning the degradation:
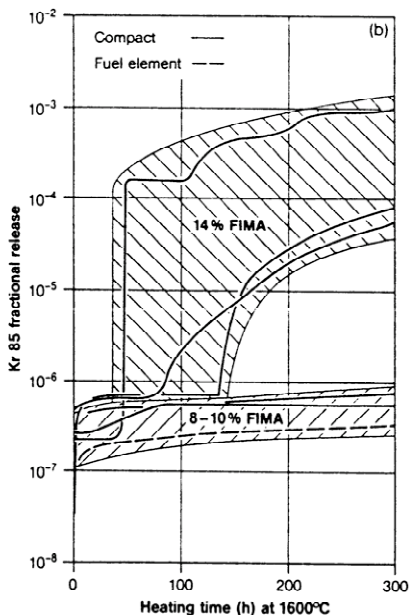
Figure 7. Kr-85 release from German UO$_2$ TRISO pebbles

(a) Cesium attack of the SiC. Experiments performed by Coen [Coen, 1972; Coen, 1973] in the 1970s demonstrate that cesium vapor can attack SiC at temperatures in excess of 1500°C. SiC samples exposed to cesium vapor indicate a pitting of the SiC layer indicative of an attack of the layer and not simple diffusion. The kinetics of the attack correlate reasonably well with the timing of cesium release from the German pebbles. Unfortunately no additional experiments were performed.

(b) CO attack of the SiC layer. At low partial pressures of CO, CO will react with SiC to form SiO, a gas. [Minato 1994] It is known that German pyrocarbon is somewhat permeable and that CO can be intercalated into graphitic structures [Minato, 1994]. The higher burnup of these particles may have produced enough CO that breakthrough of the PyC layer was achieved and a small amount of CO could attack the SiC layer and cause degradation.

There are not enough data to confirm or refute either of these two hypotheses.
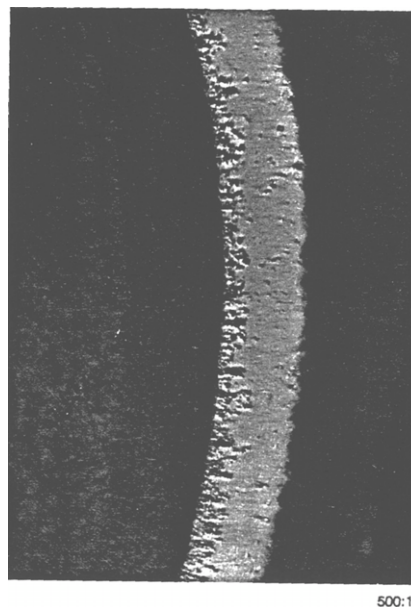

Figure 8. Photomicrograph of SiC showing degradation at SiC/IPyC interface
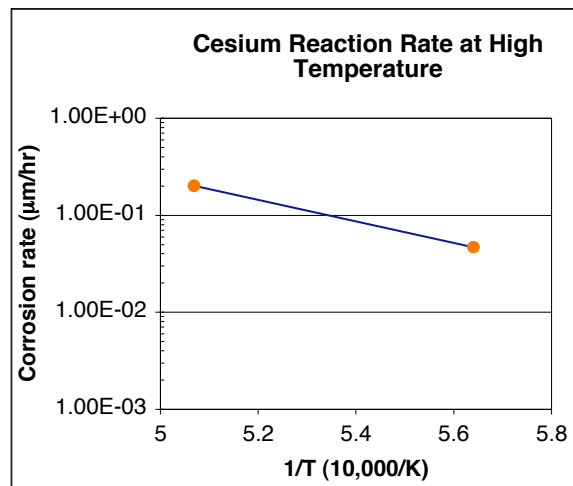

Figure 9. Cesium corrosion rate derived from Coen experiments

Table 2. Comparison of fission gas pressure in a German particle as the temperature and burnup are increased (normalized to 1.0 at 8% FIMA and 1100°C).

	Temperature (°C)				
Burnup	1100	1150	1200	1250	1300
8%	1.00	1.28	1.62	2.04	2.52
10%	1.33	1.69	2.14	2.68	3.28
15%	2.26	2.86	3.60	4.47	5.42
20%	3.32	4.21	5.28	6.53	7.89

Table 3. Comparison of CO pressure in a German particle as the temperature and burnup are increased (normalized to 1.0 at 8% FIMA and 1100°C).

	Temperature (*C)				
Burnup	1100	1150	1200	1250	1300
8%	1.00	1.15	1.28	1.38	1.44
10%	1.35	1.55	1.71	1.84	1.92
15%	2.16	2.46	2.72	2.92	3.06
20%	2.84	3.24	3.60	3.91	4.16

Table 4. Kernel migration metric as a function of temperature (normalized to 1.0 at 8% FIMA and 1100°C).

Temp (°C)	1100	1150	1200	1250	1300
KMC/T^2	1.00	1.16	1.33	1.52	1.70

Table 5. Penetration rate of Pd into SiC as a function of temperature(normalized to 1.0 at 8% FIMA and 1100°C).

Temp (°C)	1100	1150	1200	1250	1300
Pen. Rate	1.0	1.34	1.75	2.26	2.86

3. POTENTIAL DESIGN SOLUTIONS

There are potential design solutions to mitigate the deleterious effects as coated particles are taken to high temperature and higher burnup. These include: reducing the kernel size, changing the kernel to UCO, changing the kernel to UO_2* and replacing the SiC layer with ZrC.

3.1 Reduce Kernel Size

Reducing kernel size as enrichment/burnup goes up will reduce CO and fission gas pressures. It will also decrease total fission product content for a given burnup but leave the fission product concentration unchanged. However, as kernel size decreases, the diffusion length for a fission product to the SiC layer decreases and the flux of fission product atoms per unit surface area increases, both of which may exacerbate fission product attack mechanisms. Thus, reducing the kernel size helps with the gas pressure related mechanisms but hinders with respect to fission product attack of the SiC.

3.2 Change Kernel to UCO

The use of UCO will reduce the CO pressure and effectively reduces the potential for kernel migration because the uranium carbide content of the kernel prevents CO from being produced and greatly reduces the propensity of the kernel to migrate in a temperature gradient. As shown in Table 6, side by side US irradiations (HRB-14, HRB-15A, HRB-16 demonstrated no kernel migration in UCO but significant migration in UO_2 coated particles at high burnup and modest temperatures. [Young, 1980; Ketterer et al., 1984; Ketterer and Myers, 1985]

In the 1980s, the Germans irradiated 50,000 300-μm LEU UCO TRISO-coated fuel in irradiation experiment FRJ-P24. [Borschardt, 1982] No failures were observed after irradiation to 18-22% FIMA, 1.4-2.5 X 10^{25} n/m^2 (E >0.1 MeV) fast fluence and maximum fuel temperatures between 850 and 1350°C depending on the specific cell. Photomicrographs of a coated particle from this experiment are shown in Figure 10. [Bauer, 1983] Unfortunately, no postirradiation heating tests were performed. Based on these irradiation

results and the performance advantages associated with UCO at high burnup and high temperature, the US DOE Advanced Gas Reactor Fuel Development and Qualification Program has adopted UCO as its baseline fuel kernel. This selection confirms early work in the US concerning the selection of a fuel form for prismatic HTRs. [Scott, 1982]

Table 6. Kernel Migration Results from US Irradiations

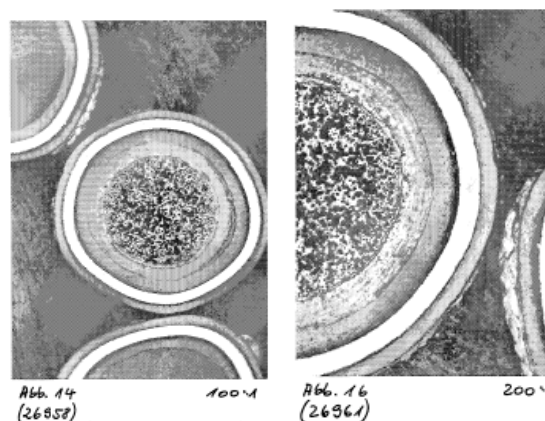| Capsule	HRB-14	HRB-15A	HRB-16
Max. Average Temperature	1070°C	1125°C	1150°C
UO_2 Peak Burnup (%FIMA)	29.5	28.5	27.8
Kernel Migration	16 μm	<30 μm in 22%	20-55 μm
Max. Average Temperature	1100°C	1100°C	1105°C
UCO Peak Burnup (%FIMA)	28.6	25	27
Kernel Migration	None	None	None

Figure 10. Photomicrograph of TRISO-coated UCO particles from irradiation FRJ-P24.

3.3 Change Kernel to UO_2*

Another alternative fuel kernel that may have promise is UO_2*. [Kendall, 2004] This fuel form is similar to a traditional TRISO-coated UO_2 particle except that a thin carbon seal coat and a

ZrC layer (~ 10 μm) are applied directly onto the kernel. Another form of UO$_2$* has ZrC, equivalent to the amount in a 10 μm hard layer, dispersed in the buffer. Irradiation of such particles in the HRB-15A and 15B to burnups of 26% FGIMA at time average temperatures of about 900°C [Bullock, 1983; Bullock, 1984] has shown very promising fuel performance. Figure 11 is a photomicrograph of a UO$_2$* particle. Very little kernel swelling was observed and kernel migration was greatly reduced. In addition, fission product retention in postirradiation annealing tests was much better than in conventional TRISO-coated particles. Finally, the ZrC in principle would be an excellent getter for any CO generated by irradiation of UO$_2$, which would reduce both internal gas pressures and kernel migration, especially at high burnups and temperature.
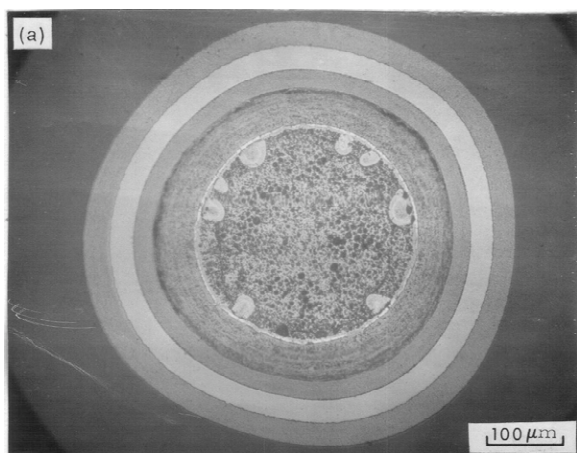

Figure 11. Photomicrograph of UO$_2$* irradiated to 22% FIMA at 900°C in HRB-15B.

3.4 Replace SiC with ZrC

ZrC has great potential as a coating for particle fuel. Testing to date suggests it may have higher performance capability than SiC. However, ZrC has a number of significant development issues that need to be addressed before it could be considered a reference coating for fuel particles.

There are no reference deposition processes or product specifications for ZrC. Significant additional fabrication development would be

required to develop the process and product specifications to make acceptable ZrC. ZrC$_x$ can be fabricated depending on conditions and performance can vary significantly depending on value of x. It is also unclear if ZrC can be made in an uninterrupted coating process, which is considered a key part of the successful German TRISO fuel development activity. Most importantly, because ZrC will oxidize in air, the leach-burn-leach test cannot be used with ZrC coated particle fuel. Thus, a new method needs to be developed and qualified to determine the quality of the ZrC layer in the same way that leach-burn-leach is used to determine the quality of the SiC layer in traditional TRISO fuel.

There is a lack of an optimized design for particles containing ZrC for an NGNP. Scoping irradiations would probably be needed to test different design configurations to establish the most promising candidates. Such design and testing is needed to establish a baseline for this fuel form. The current irradiation and accident-heating database, while promising, is inadequate from a fuel qualification perspective. The amount of ZrC fuel that has been irradiated is much less than SiC TRISO-coated particle fuel. Significant quantities of Zr-coated particle fuel would need to be irradiated and tested at accident conditions to demonstrate the requisite high burnup and high temperature performance capabilities. Furthermore, unexplained results have been found in the Japanese program. Irradiation and heating tests for ZrC particles found lower retention of Ru, Ce, and Eu than in SiC TRISO particles. No Pd/ZrC interaction was observed in the particles but at the same time no Pd could be found in the particles. These issues will need to be resolved.

All of these factors lead us to believe that ZrC still has a lot of promise but it also will take a long-term fuel development program to truly demonstrate the performance capability of this fuel. The long fuel development time is inconsistent with the current NGNP schedule.

4.0 SUMMARY

With the exception of the thermo-mechanical response of the particle, these calculations

indicate that high temperature and high burnup will erode existing fuel performance margins in the traditional UO_2 German TRISO-coated particle system. Additional fuel development will be required to demonstrate that UO_2 TRISO-coated particles will work under NGNP conditions. Irradiations and accident heating tests are proposed as part of the European gas reactor program to understand the limits of UO_2 at high burnup and high temperature.

Potential solutions exist to recover some of the performance margin that is expected to be lost in going to higher burnup and higher temperature. However, all of them require extensive testing and analysis. The solutions are at different stages of maturity. Some require scoping irradiations and heating tests to demonstrate satisfactory proof of performance while other options are more mature and only require the more extensive set of activities related to formal fuel qualification.

The Advanced Gas Reactor (AGR) Fuel Develop and Qualification Program has adopted UCO as a design solution given its satisfactory performance in German and US irradiations and its ability to prevent CO formation and kernel migration both of which are a concern at high burnup and high temperature. Thus, the AGR program is largely focused on irradiation testing (that extends the fuel operating envelope to NGNP conditions listed in Table 1) and subsequent accident heating testing required for fuel qualification.

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