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Journal of Nuclear Materials 371 (2007) 270–280
journal of nuclear materials
www.elsevier.com/locate/jnucmat

The challenges associated with high burnup, high temperature and accelerated irradiation for TRISO-coated particle fuel

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Abstract

Fuel service conditions proposed for the very high temperature reactor 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 coated particle fuel development program in the 1980s. Of particular concern are the high burnup and high temperatures expected in the very high temperature reactor. In this paper, the challenges associated with high burnup and high temperature are evaluated 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 discussed. Also of concern are the effects of accelerated irradiation on coated fuel that often occur during irradiation testing. These effects are evaluated in this paper and recommendations concerning allowable levels of accelerations are presented.

1. Introduction

The fuel service conditions proposed for the very high temperature reactor (VHTR) will be challenging [1]. 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 Fig. 1 compare these parameters as currently estimated for the VHTR (with a prismatic core which envelopes the parameters required for a pebble bed core) with those of other programs around the world. The results indicate that German fuel does not adequately envelope the conditions expected for the VHTR 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 [2] 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 and fission product release mechanisms that are temperature and burnup dependent. These include: thermomechanical response of PyC, fission gas release and CO production, amoeba effect, metallic fission

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doi:10.1016/j.jnucmat.2007.05.019

Table 1
Comparison of fuel service conditions

Parameter	US VHTR	Germany	Japan	South Africa	France	China
Burnup (% FIMA)	15–20	8	4	8–10	10–15	8
Peak temperature (°C)	1250	1100	1200	1100	1200	1100
Fast neutron fluence (10^{25} n/m^2, $E > 0.18$ MeV)	4	3.5	4	3.5	4	3.5
Packing fraction (%)	<35	10	30	10	10–15	10
Power density (MW/m^3)	6	3	3–6	3	3–6	3

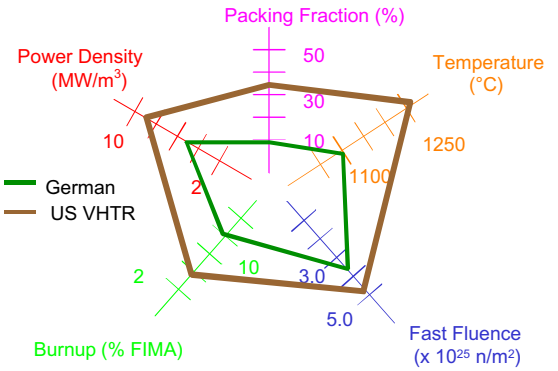


Fig. 1. Comparison of US VHTR and German fuel operating envelope.

product diffusion 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.

Fuel development and qualification programs usually irradiate fuel in test reactors. These high neutron flux irradiations accelerate (or reduce) the time required to reach full design burnup and/or neutron fluence. Accelerated irradiations also result in increased power per particle and increased temperature gradients in the TRISO-coated particles. Several fuel-related phenomena are affected by these variations in time and temperature.

The PARFUME code has been used to evaluate some of the most significant effects of accelerated irradiation that are dependent upon time at temperature. These evaluations were based on both German UO_2 and US UCO fuels. The UCO fuel is representative of the fuel being developed by the US Department of Energy (DOE) Advanced Gas Reactor (AGR) Fuel Development and Qualification Program which supports the Generation IV VHTR concept.

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 (as illustrated in Fig. 2 for representative US UCO fuel), the shrinkage increases and the stress in the IPyC increases.

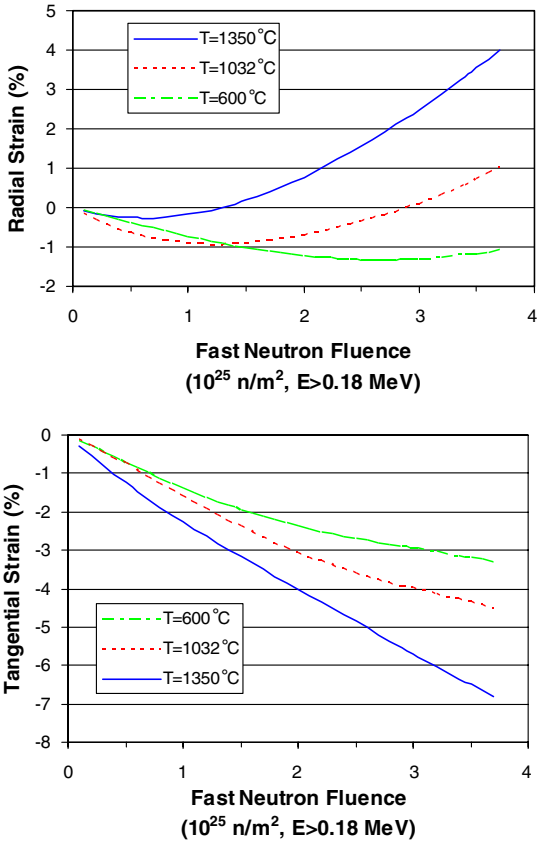


Fig. 2. Radial and tangential irradiation-induced strains in PyC (BAF = 1.08) as a function of irradiation temperature and fast neutron fluence.

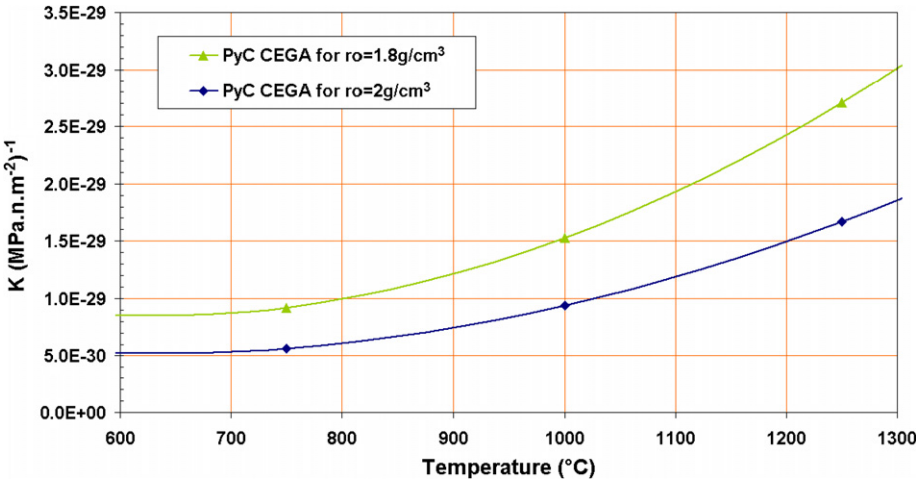


Fig. 3. Irradiation-induced creep constant for PyC.

Offsetting the shrinkage is irradiation-induced creep. Although the data are uncertain, the limited data available suggest that irradiation induced creep of PyC depends on the density of the PyC and the irradiation temperature [3] (see Fig. 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.

2.2. Fission gas pressure

Fission gases released during irradiation from the kernel of a coated particle depend on temperature, burnup and time [4]. Table 2 presents the normalized fission gas pressure that builds up in a 500 μm (kernel diameter) 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 eight increase in pressure as the burnup increases from 8% to 20% FIMA and the temperature increases from 1100 to 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 four increase in pressure as the burnup increases

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

from 8% to 20% FIMA and the temperature increases from 1100 to 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 [4] (there is no burnup dependence). An example of kernel migration is shown in Fig. 4.

The migration distance is given by the following relationship:

$$\vec{\delta}_{\mathrm{MIG}} = \int \mathrm{KMC} \cdot \frac{\vec{\nabla} T}{T^2} \mathrm{d}\tau, \tag{1}$$

$$\mathrm{KMC} = \mathrm{KMC_O} * \exp(-Q/RT). \tag{2}$$

For a given temperature gradient, the kernel migration distance will depend on the quantity (KMC/T^2). Fig. 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

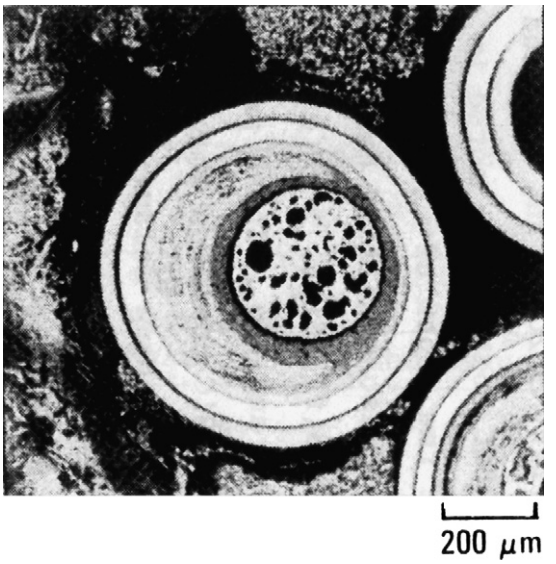


Fig. 4. Migration of a UO_2 kernel.

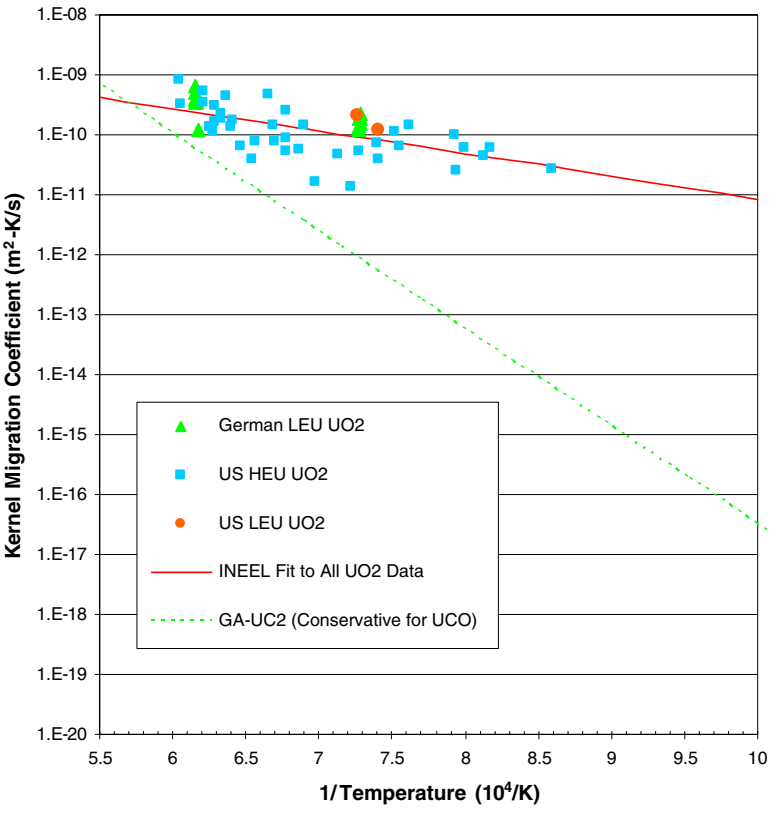


Fig. 5. Kernel migration coefficient for different fuel forms.

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

Temperature (°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)

Temperature (°C)	1100	1150	1200	1250	1300
Normalized penetration rate	1.0	1.34	1.75	2.26	2.86

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, 25–50 times 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 VHTR. A review of the international database shows no strong dependence on burnup or the 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 [4] (see Fig. 6). Table 5 indicates that the rate of Pd penetration into the SiC is almost a factor of three greater at 1300 °C than at 1100 °C.

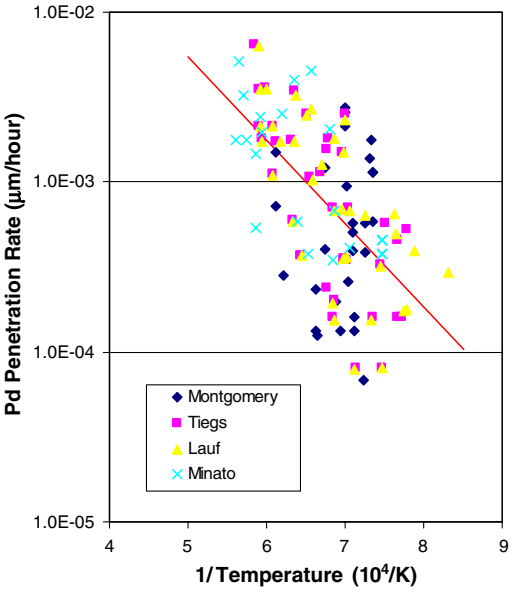


Fig. 6. Pd penetration rate based on international data.

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 VHTR. German pebbles irradiated to burnups of 14% FIMA and fluences of $5–6 \times 10^{25}$ n/m^2 ($E > 0.1$ MeV) have shown elevated releases of both cesium and noble gases compared to pebbles with burnups of less than 10% FIMA heated to similar conditions [5] (see Fig. 7). The reasons for the increased release are not known with certainty.

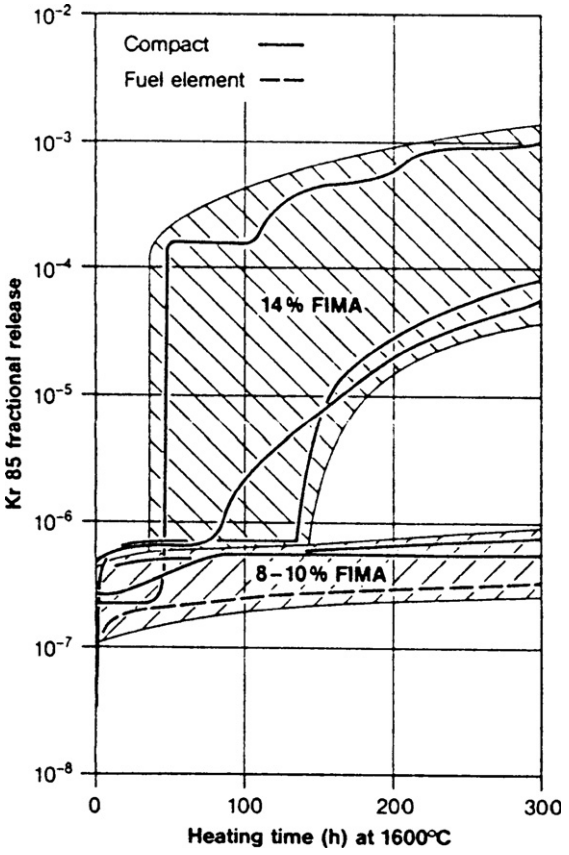


Fig. 7. Kr-85 release from German UO_2 TRISO pebbles.

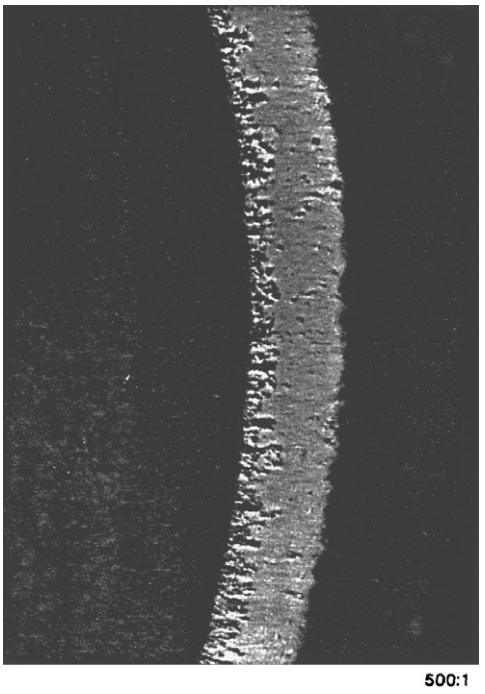


Fig. 8. Photomicrograph of SiC showing degradation at SiC/IPyC interface.

A photomicrograph of the SiC in a coated particle from one such test [5] is shown in Fig. 8. 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:

(a) Cesium attack of the SiC. Experiments performed by Coen et al. [6,7] 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 [8]. It is known that German pyrocarbon is somewhat permeable and that CO can be intercalated into graphitic structures [8]. 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.

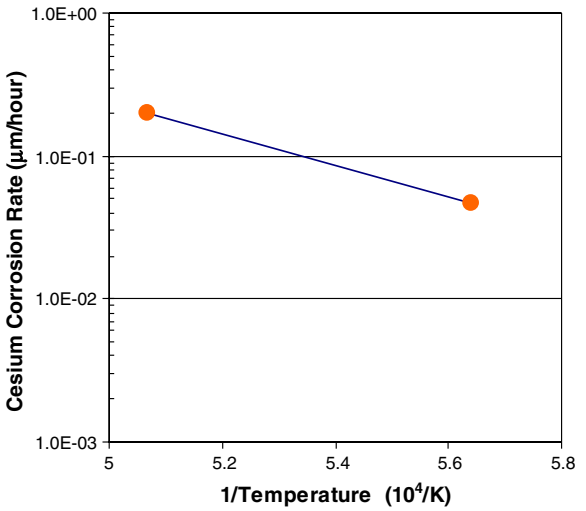


Fig. 9. Cesium corrosion rate derived from Coen experiments.

There are not enough data to confirm or refute either of these two hypotheses (see Fig. 9).

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 maximum average temperatures under 1150 °C [9–11].

In the 1980s, the Germans irradiated 50 000 LEU UCO TRISO-coated fuel particles (with 300 μm kernel diameters) in the FRJ-P24 irradiation experiment [12]. No failures were observed after irradiation to 18–22% FIMA, $1.4–2.5 \times 10^{25}$ n/m^2 ($E > 0.1$ MeV) fast neutron 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 Fig. 10 [13]. 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 [14].

Table 6
Kernel migration results from US irradiations

Parameter	Experiment		
	HRB-14	HRB-15A	HRB-16
Maximum average UO_2 temperature (°C)	1070	1125	1150
UO_2 peak burnup (% FIMA)	29.5	28.5	27.8
UO_2 kernel migration distance (μm)	16	<30 in 22%	20–55
Maximum average UCO temperature (°C)	1100	1100	1105
UCO peak burnup (% FIMA)	28.6	25	27
UCO kernel migration distance (μm)	None	None	None

3.3. Change kernel to UO_2^*

Another alternative fuel kernel that may have promise is UO_2^* [15]. 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 thick) 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, 15B and 16 experiments to burnups of 26% FIMA at time

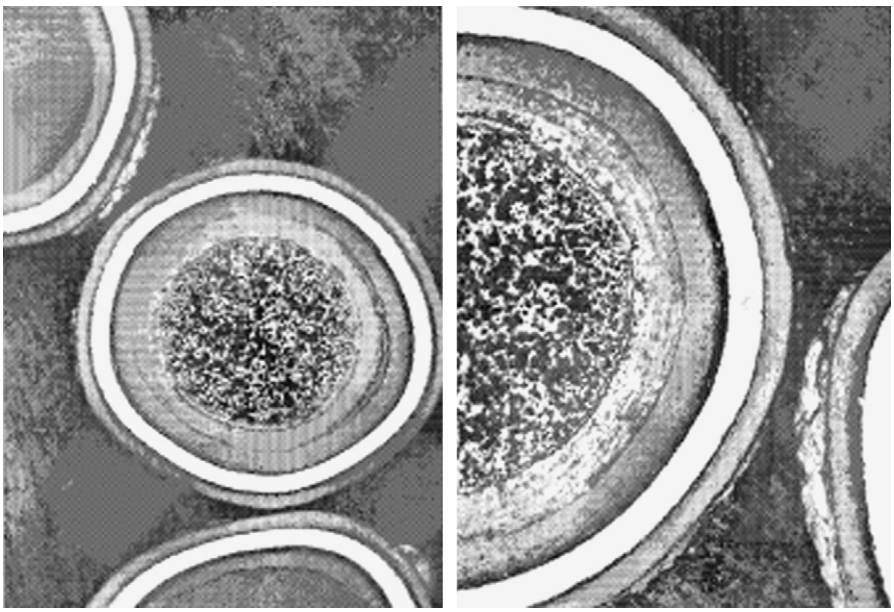


Fig. 10. Photomicrograph of TRISO-coated UCO particles from irradiation FRJ-P24.

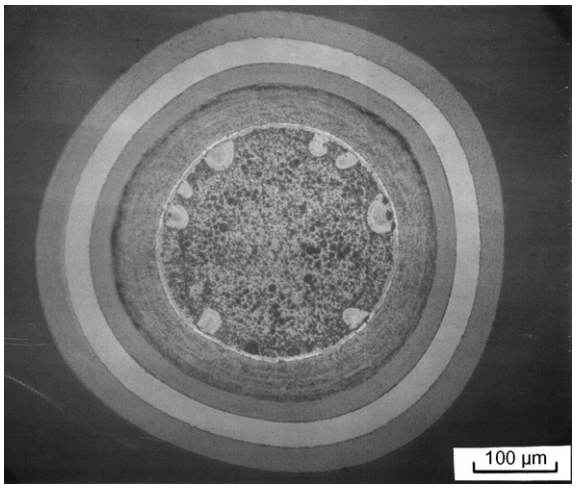


Fig. 11. Photomicrograph of UO_2^* irradiated to 22% FIMA at 900 °C in HRB-15B.

average temperatures ranging from 900 to 1200 °C [16,17] has shown very promising fuel performance. Fig. 11 is a photomicrograph of a UO_2^* particle. Very little kernel swelling was observed and kernel migration was not observed. 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.

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 a VHTR. 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 indicate that ZrC still has 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 VHTR schedule.

4. Accelerated irradiation

The PARFUME code was used to examine the effects of accelerated irradiation on coated particle fuel performance. Two fuel forms representing typical German particles and US AGR particles were used in the evaluations. German fuel consisted of coated 500 μm diameter UO_2 kernels and the US AGR fuel consisted of coated 350 μm UCO kernels. Particle performance was examined at power levels ranging between 50 and 500 mW/particle which corresponds to approximately real time irradiation up to 10 times acceleration to reach end of life (EOL) service conditions. For these calculations, end of life conditions were 10% FIMA for the German fuel and 20% FIMA for the US AGR fuel, with each fuel form experiencing an EOL fast neutron fluence of 4.0×10^{25} n/m^2 ($E > 0.18$ MeV). Both fuel burnup and fast neutron fluence were accelerated in this

analysis, since in many reactors fast and thermal neutron fluxes scale. To simplify the comparisons, these evaluations also assumed that all particles were at the given power throughout its entire life and were held at a thermal boundary condition of 1000 °C at the outer surface of the OPyC layer.

As power or acceleration increases, the time required to reach full burnup decreases and fuel temperatures increase. These expected results are illustrated in the Figs. 12 and 13. For a given power, the US AGR fuel temperature is higher than for the German fuel due to the higher power density associated with the smaller AGR kernels (at 500 mW/particle, the corresponding power density for AGR particles is 22.3 kW/cm^3 and for the German particles it is 7.6 kW/cm^3).

Accelerated irradiation increases total internal gas pressure in both fuel forms as shown in Fig. 14. This pressure increase is primarily due to the increased temperature of the fuel with increasing power. German fuel pressures are higher than AGR fuel pressures due to the formation of CO in the

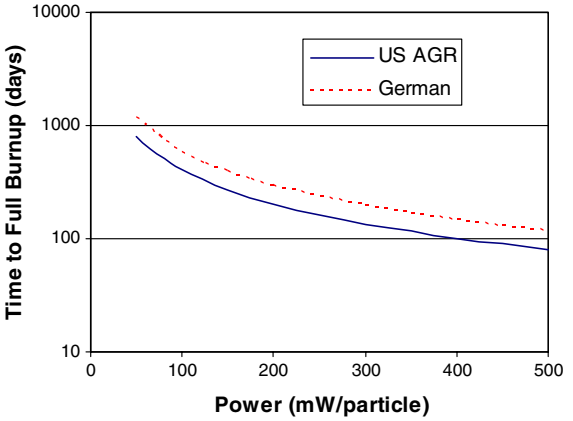


Fig. 12. Effect of particle power on time to full burnup.

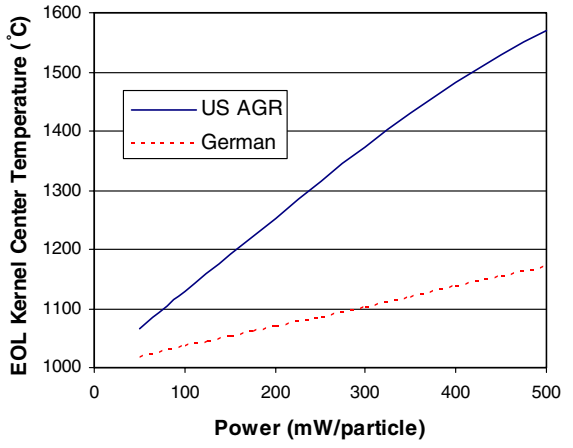


Fig. 13. Effect of power on particle center temperature.

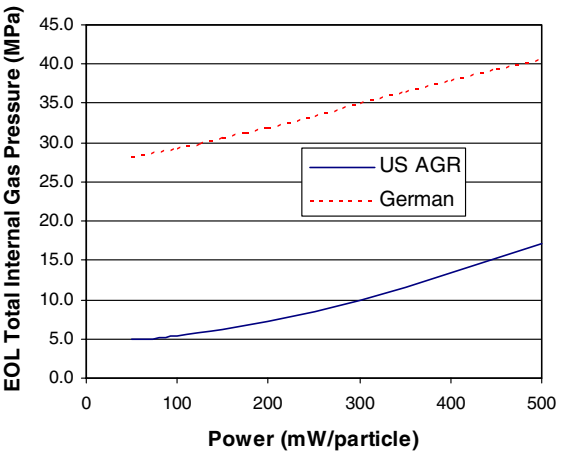
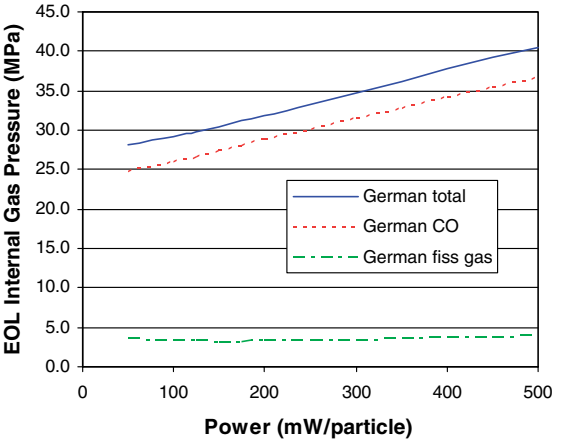


Fig. 14. Effect of particle power on internal gas pressure.



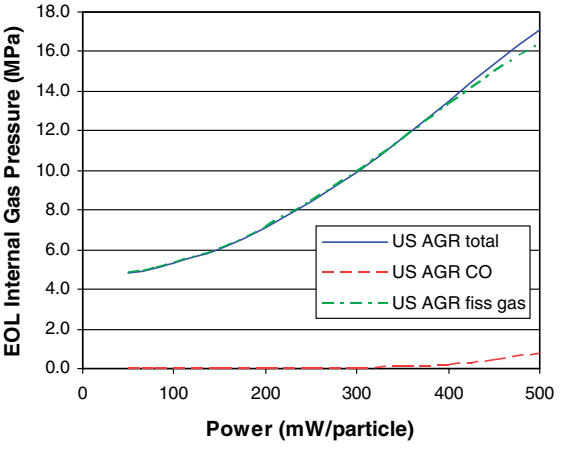


Fig. 15. Contributions to internal gas pressure.

UO_2 fuel which is negligible in UCO fuel. Internal pressures are also affected by the amount of fission product gases released to the void volume which is a complex function of time and temperature. The contributions of CO and fission product gas to the total gas pressure are displayed in Fig. 15.

Metallic fission product release is modeled by Fickian diffusion with Arrehnius diffusion coefficients [18]. The calculated results demonstrated that the behavior is a complex function of time and temperature as illustrated in Figs. 16 and 17 for cesium and silver release. Generally, as power increases, fractional release decreases due to less time available for diffusion. This trend continues until the diffusion rate increases sufficiently (due to increasing temperature and its impact on diffusion coefficients) to dominate over the irradiation time and release begins to increase with increasing power. For some fission products and irradiation conditions, this overall trend may not be displayed as illustrated by Pd penetration in SiC (which is rate limited by diffusive release from the kernel) for German fuel. As shown in Fig. 18, Pd penetration continuously decreases with increasing power for German fuel, while for US AGR fuel, Pd penetration initially decreases and then increases with increasing power.

Effects of acceleration on fuel performance metrics are complicated due to varying degrees of dependence on temperature, time, burnup and fast fluence. Therefore, fuel performance models are required to accurately predict these effects for a specific fuel form and irradiation history. However, this evaluation has shown the substantial increase in fuel temperatures associated with high levels of acceleration. This gives support to the historic German approach of limiting irradiations to less than three times acceleration.

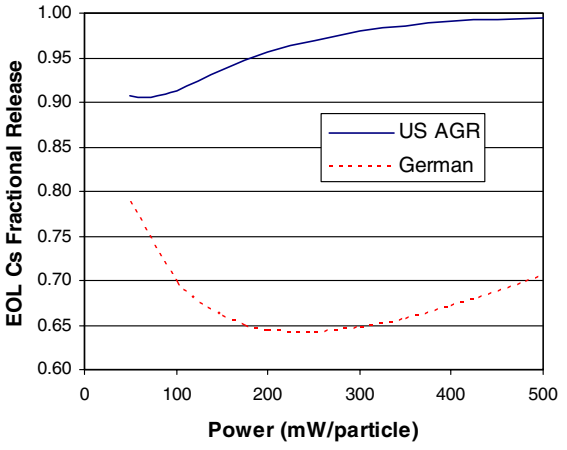


Fig. 16. Effect of particle power on cesium release.

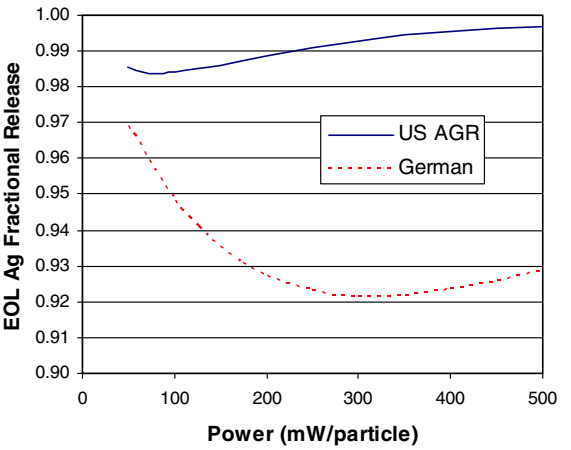


Fig. 17. Effect of particle power on silver release.

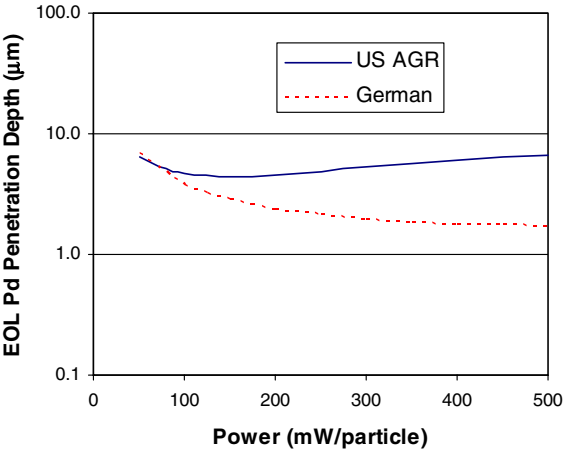


Fig. 18. Effect of particle power on Pd penetration into SiC.

5. Conclusions

With the exception of the thermomechanical 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 VHTR 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 DOE Advanced Gas Reactor Fuel Development and Qualification Program has adopted UCO as a design solution given its satisfactory performance in German 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 VHTR conditions listed in Table 1) and subsequent accident heating testing required for fuel qualification. Considering the deleterious effects of high temperature and high thermal gradients associated with high irradiation accelerations, these irradiation tests will also be conducted with limited accelerations (up to two times expected real time).

References

[1] US DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum, A technology roadmap for generation IV nuclear energy systems, GIF-002-00, December 2002.
[2] G.K. Miller et al., in: Proceedings of the High Temperature Reactor Conference HTR-2004, Beijing, China, September 2004.
[3] F. Ho, Material models of pyrocarbon and pyrolytic silicon carbide, CEGA-002820, July 1993.
[4] D.A. Petti (Ed.), Development of improved models and designs for coated-particle gas reactor fuels, INEEL/EXT-05-02615, December 2004.
[5] R. Gontard, H. Nabielek, Performance evaluation of modern HTR TRISO fuels, HTA-IB-05/90, July 1990.
[6] V. Coen et al., J. Nucl. Mater. 45 (1972/1973) 96.
[7] V. Coen et al., in: Proceedings of the BNES International Conference on Nuclear Fuel Performance, British Nuclear Energy Society, London, 1973, p. 19.1.
[8] K. Minato et al., J. Nucl. Mater. 208 (1994) 266.
[9] C.A. Young, Pre- and postirradiation evaluation of fuel capsule HRB-14, GA-A15969, UC-77, 1980.
[10] J. W. Ketterer et al., Capsule HRB-15A Postirradiation Examiantion Report, GA-A16758, UC-777, 1984.
[11] J. W. Ketterer, B.F. Myers, Capsule HRB-16 Postirradiation Examination Report, HTGR-85-053, 1985.
[12] G. Borschat, B. Hurttlen, G. Pott, Experiment FRJ2-P24 bestrahlungsbericht, KFA-ZBB-IB-19/82, August 1982.
[13] Ch. Bauer, H. Klocher, Elektrolytische desintegration an 3 compacts aus dem experiment, FRJ-P24IRW-IN-8/83, 1983.
[14] C.B. Scott, Summary Report on Selection of Reference LEU/Th Fuel System, GA Document 906330, Issue A, 12 April 1982.
[15] J. Kendall, R. Bullock, in: Proceedings of the Second International Topical Meeting on High Temperature Reactor Technology, HTR-2004, Bejing, China, September 2004.
[16] R.E. Bullock, J.L. Kaae, J. Nucl. Mater. 115 (1983) 69.
[17] R.E. Bullock, J. Nucl. Mater. 125 (1984) 304.
[18] International Atomic Energy Agency, Fuel performance and fission product behaviour in gas cooled reactors, IAEA-TECDOC-978, November 1997.