

## **2. TRISO-COATED FUEL PARTICLE PERFORMANCE**

### **2.1 General Description Of Coated Particle Fuel And Fuel Element**

#### **2.1.1 Operational Requirements**

The contemporary goal of coated particle fuel and the associated fuel form is to allow high temperature reactor operation with very high fractional fuel particle integrity during normal operation and accidents and very low fission product release during normal operation and under accident conditions [2-1 to 2-7]. A secondary goal is high fuel burnup (>10% FIMA for pebble bed and >20% FIMA for prismatic core) to allow economic operation of the reactor system and good fissile material utilization. The core average exit temperatures currently in the range of 800°C to 950°C with peak fuel temperatures of 1200°C to 1250°C, which is higher than for LWRs. Long-term plans are to go to even higher core average exit temperature with burnups in the range of 20-25% FIMA, which is also higher than for LWRs.

Achieving these goals requires the use of different materials than commonly used in LWR fuel and a different core coolant environment. Unlike LWR fuels, the use of metallic materials is minimized and the coolant is an inert, single-phase gas (helium). Reliance instead is placed on ceramic materials, primarily carbon based materials such as graphite, pyrocarbon, and silicon carbide [2-8]. The properties of these materials also have shifted the fuel design away from an array of rods to graphite blocks with fuel compacts or fuel spheres. Also in contrast to LWRs, the coolant and the moderator are separate.

The philosophy of HTGR coated particle fuel is somewhat different than that of LWR fuel pellets and cladding. The fuel in an HTGR core is contained in billions of coated particles, each of which acts as its own containment. The small kernels of fuel are each coated with layers of carbon and silicon carbide. The resulting particle is designed to withstand the pressure of the generated fission gases and to form an essentially leak tight barrier to fission product release. While LWR fuel cladding performs this function on a larger scale during normal operating conditions, coated particle fuel also requires this high level of integrity under accident conditions. Thus, the fuel particle is required to stay intact with high reliability during both normal operation and accident conditions.

#### **2.1.2 Basic Fuel Element**

The fundamental component of the HTGR fuel element is the coated fuel particle. The particle is composed of a kernel of fuel and several coating layers, each of which has a specific function. For the purposes of this report, we will consider the traditional four-layer particle as illustrated in Fig. 2-1.

The HTGR kernel is either  $\text{UO}_2$  or a two-phase mixture of  $\text{UO}_2$  and  $\text{UC}_2$  known as UCO. The diameter is about 500 microns for the pebble fuel kernels and about 350 and 500 microns for fissile and fertile kernels, respectively, in prismatic block fuel. Particles made from thorium and plutonium are also possible, but  $\text{UO}_2$  and UCO are the materials of current interest for commercial power reactors. The  $\text{UO}_2$  kernel has the most extensive civilian experience base, but the UCO kernel offers the ability to better control carbon

monoxide production and thus particle internal pressure buildup. This factor is important as the fuel is pushed to higher burnup and higher operating temperatures.

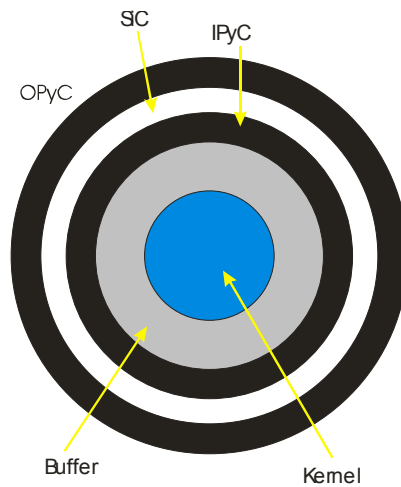


Figure 2-1 Schematic of the four-layer particle design

The HTGR kernel is either  $\text{UO}_2$  or a two-phase mixture of  $\text{UO}_2$  and  $\text{UC}_2$  known as UCO. The diameter is about 500 microns for the pebble fuel kernels and about 350 and 500 microns for fissile and fertile kernels, respectively, in prismatic block fuel. Particles made from thorium and plutonium are also possible, but  $\text{UO}_2$  and UCO are the materials of current interest for commercial power reactors. The  $\text{UO}_2$  kernel has the most extensive experience base, but the UCO kernel offers the ability to better control carbon monoxide production and thus particle pressure. This factor is important as the fuel is pushed to higher burnup and higher operating temperatures.

The buffer layer is a low-density porous carbon layer that acts as an expansion space to collect the released gases. The gases are generated as a result of (1) fission and (2) chemical reactions between the carbon buffer layer and oxygen liberated from the  $\text{UO}_2$ . The inner pyrocarbon (IPyC) layer provides a smooth surface for the silicon carbide layer to be deposited and also shields the kernel from chlorine released during fabrication. It also plays a role in the mechanical stress distribution within the particle. The silicon carbide (SiC) layer is the major fission product barrier and plays an important structural function. Finally, the outer pyrocarbon (OPyC) layer isolates the SiC layer from the matrix that binds the particles together and provides a compressive force on the SiC. These layer functions will be described in more detail in the following sections. As described in Chapter 1.0, the particles are aggregated in a matrix material to form a fuel element for the reactor core.

The reader may wonder why suspending and binding the particles in a fuel form is necessary and why it wouldn't be acceptable to pour them into a tube or a hole in a block. Two reasons preclude this approach. – particle heat transfer and mismatches between their thermal expansions.

Close packed particles would have limited contact area and a considerable amount of void space, thus the contact heat transfer would be modest. The flow impedance of very small closely packed particles would be too high, thus limiting the heat that can be removed by convection. These two effects would limit the effective heat transfer from a loose bed of small particles.

The second reason for combining particles in a fixed matrix material, differences in the thermal expansion of the particles and holder, is more limiting. Loosely packed particles would have a tendency to settle under vibration or temperature cycling. Settling would generally be irreversible and the application of significant forces would not reverse it. As the particles heat up and are subject to normal reactor system flow-induced vibrations, the particles would settle into a new, more closely packed configuration. This might be acceptable if the particles and their holder had the same thermal expansion and only modest temperature gradients, but this is generally not the case. Temperature cycling after the particles settle would result in differential expansion between the particles and the holder. Since the settling cannot be reversed by force, large compression forces would build up in the particles that could damage the particles or rupture the holder.

A practical approach to this problem is to suspend and bind the particles with a thermal-mechanically compatible medium [2-1 through 2-4, 2-9]. Figure 2-2 shows the basic concept. The fuel particles are fixed within a matrix of graphite powder and binder and may be encased within a non-fuelled layer depending on the application. In practice two basic fuel element forms have evolved.

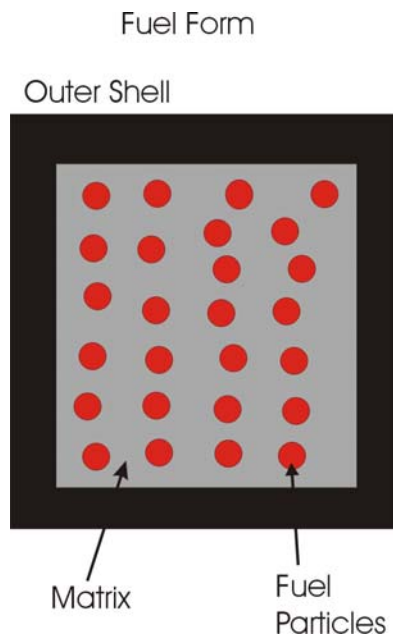


Figure 2-2 Fuel element components

The first involves fuel particles suspended in a matrix in the shape of a sphere surrounded by an outer unfueled layer of matrix material (i.e. a fuel “pebble”). This fuel form is used by the pebble bed type reactor designs. It must be sufficiently tough so that it can be

dropped repeatedly several meters onto a bed of pebbles without breaking. The fuel pebbles are typically 6 cm in diameter and contain about 15,000 fuel particles. The second design is a cylindrical fuel compact design that can be stacked and inserted into a graphite block. This form may be solid or annular in configuration. Each compact contains about 10,000 particles. See Fig. 2-3 for the designs. Over the years other designs such as a complex fuel block molded from particles have been considered, but these are currently not of interest [2-9]. The details of fuel element fabrication will be covered in later sections of this chapter.

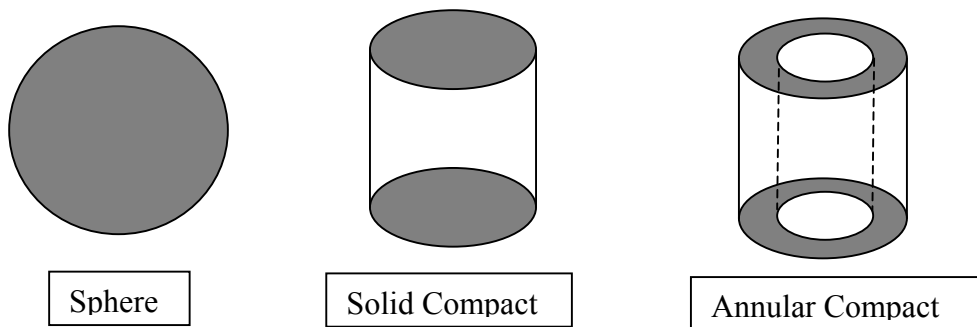


Figure 2-3 Three of many possible designs for a fuel element. The compact design can also be used as part of a pin in block design

## 2.2 Design Function of Each Component

### 2.2.1 General

Each component of the fuel particle and fuel element has specific functions, specific material needs and specific challenges. A description of each component and its design considerations is followed by its relevant phenomena in the PIRT Table.

### 2.2.2 Kernel

The kernel contains the nuclear fuel and its composition controls the basic chemistry of the particle environment. Contemporary design focuses on controlling the oxygen potential of the particles either by limiting burnup or by tailoring the kernel composition [2-8]. There are two reasons for this. The first is to tie up the oxygen liberated by fissioning the U in  $\text{UO}_2$  so that it cannot react with the carbon buffer layer and form CO. CO production can greatly increase the particle pressure and increase its failure likelihood [2-6, 2-8, 2-10, 2-11]. In addition, in the presence of a thermal gradient, CO can result in migration of the kernel away from its centered position [2-8]. The second is to make ensure that the rare earth elements are oxidized and thus immobilized so they do not migrate to the SiC and react with it.

Summarizing, the basic functions of the kernel are to:

- 1) Control particle internal pressure and migration potential by holding down CO production.
- 2) Tie up rare earths as oxides to limit their migration to the coating.
- 3) Produce the desired power.

The fission gases and volatile fission products are largely contained by the coatings. However, fission gas retention by the kernel is important at the low to moderate burnup levels (less than about 20%) [2-22]. Up to this burnup and at normal operating temperatures, the kernel provides significant holdup (~50%, see Chapter 3) of the fission gases krypton and xenon as well as volatile species such as iodine and cesium. This retention aids in controlling particle pressure and is important for exposed kernels, as it greatly reduces the amount of fission products that are released to reactor internal components. Significant retention of some isotopes can even occur as accident temperatures are approached.

As the kernel burnup increases, its ability to retain fission gases and volatile fission products can decrease, especially at the higher temperatures. Designers often assume high release levels (up to 100%) of fission gases (very high burnup, ~70% FIMA) at end of life or for accident conditions. This is different from LWR fuel (normal operating temperatures) where fuel fission gas retention is considered important (retention levels ~ 95%) and secondary gases such as CO are not important.

An important design consideration for the kernel is the oxygen potential [2-8]. Its importance comes about from the fact that the fission products from a fissioned uranium atom have an oxygen combining ability less than that of the original U, thus oxygen is available in the system to combine with other elements. Table 2-1 illustrates the available oxygen ratio for several actinides assuming the metal oxides do not become super stoichiometric. Note that the oxygen to metal ratio is not the same as oxygen potential. The ratio simply looks at the number of oxygen and metal atoms in the system and allows one to determine whether or not oxygen is likely to be available for chemical reactions. The oxygen potential is determined by the amount of oxygen in the system and the affinity of particular elements for it. The oxygen potential determines which elements are successful in competing for the oxygen and which are not.

The fission products, carbon, and uranium all compete for oxygen in this closed system and the system oxygen potential determines which elements are oxidized and which are not for a limited amount of oxygen. The oxygen potential,  $\mu_{O_2}$  is defined as:

$$\mu_{O_2} [kcal / mol] = RT \ln(P_{O_2} [atm])$$

where  $R$  is the gas constant,  $T$  is the absolute temperature, and  $P_{O_2}$  is partial pressure of oxygen. In pure  $UO_2$  fuel (our reference case), the oxygen potential increases as a function of burnup and results in the production of CO. This CO increases the pressure in the particle. Figure 2-4 illustrates how particle pressure can be affected as burnup increases.

**Table 2-1 Oxygen Excess per Fission**

Isotope	O/F
U-233	0.092
U-235	0.132
Pu-239	0.623
Pu-241	0.599

$2C + (O_2) = 2(CO)$	UO <sub>2</sub> Burnup	T (K)	P <sub>co</sub> (atm)
		Low	1300
	( $\mu_{O_2} = -100$ kcal/mol)	1600	5
	High	1300	580
	( $\mu_{O_2} = -75$ kcal/mol)	1600	1300

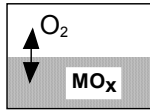


Fig. 2-4 Increase in particle pressure as a function of burnup for a UO<sub>2</sub> kernel (representative order of magnitude calculation)

Particle pressure translates directly into coating stresses [2-13 to 2-18]. This can be illustrated with a simple model. If it is assumed that the SiC layer carries the entire internal pressure load, the stress,  $\sigma$ , in the layer is approximately:

$$\sigma = \frac{rP}{2t}$$

where  $r$  is the radius,  $P$  is the pressure, and  $t$  is the layer thickness. If the stress in the SiC layer is greater than its ultimate tensile strength (UTS) the layer will fail. The total gas pressure in the particle is the sum of the CO gas pressure and the released fission gases:

$$P_{Total} = P_{Kr} + P_{Xe} + P_{CO}$$

The stress equation can be solved for  $P$  and, with an UTS of SiC of 350MPa, an SiC thickness of 35 $\mu$ m, and an SiC layer radius of 310  $\mu$ m, one gets:

$$P_{Max} = \frac{UTS_{SiC} 2t}{r} = 790 atm$$

This scoping calculation tells the designer that the particle internal pressure needs to be limited to a few hundred atmospheres to prevent overpressure failure of the SiC. Factors

such as uneven coating thickness, thin coatings, non-spherical shapes, and less than expected material properties would reduce this pressure considerably. A more sophisticated analysis for both oxygen potential and layer stress would be required for actual particle design work. To summarize, for  $\text{UO}_2$  fuel:

- 1) The krypton and xenon pressure depend on burnup, kernel gas retention, and free volume (buffer). The kernel gas retention diminishes with burnup and is often assumed to be nil for accidents. For the 10% FIMA burnup fuel kernel gas retention is fairly high at normal temperatures, likely in the 50% FIMA or more range for fission gases.
- 2) CO pressure depends only on oxygen potential and temperature. Oxygen comes from fissioning the U in  $\text{UO}_2$  and carbon comes from the buffer.
- 3) The oxygen potential depends on  $\text{UO}_2$  burnup.

Figure 2-5 shows a SiC layer overpressure failure (known in the literature as *pressure vessel failure*). For particles designed and operated within specifications, this failure is not commonly seen.

Carbon monoxide is involved in another particle damage phenomena known as the *Amoeba Effect* [2-8]. This phenomenon involves in the transfer of carbon from one side of the kernel to another in the presence of a temperature gradient and results from the differing equilibrium between CO and  $\text{CO}_2$  at different temperatures.

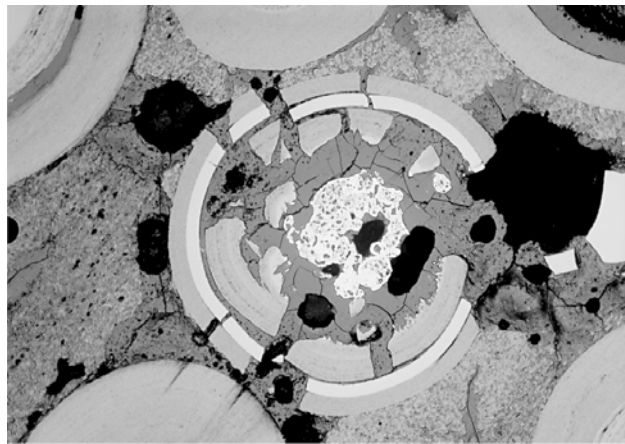


Figure 2-5 Pressure vessel failure in HRB-8, Specimen 5 ( $\text{UO}_2$ )

This effect can also occur in  $\text{UC}_2$ , but is due to solid-state carbon transport rather than CO/ $\text{CO}_2$  [2-12]. The greatest effect is with  $\text{UO}_2$ .

The net effect of this carbon transport is to gradually push the kernel in the direction of increasing temperature, across the kernel so that the kernel moves toward the SiC layer and damage the layer. This is clearly undesirable. Figure 2-6 illustrates the action.

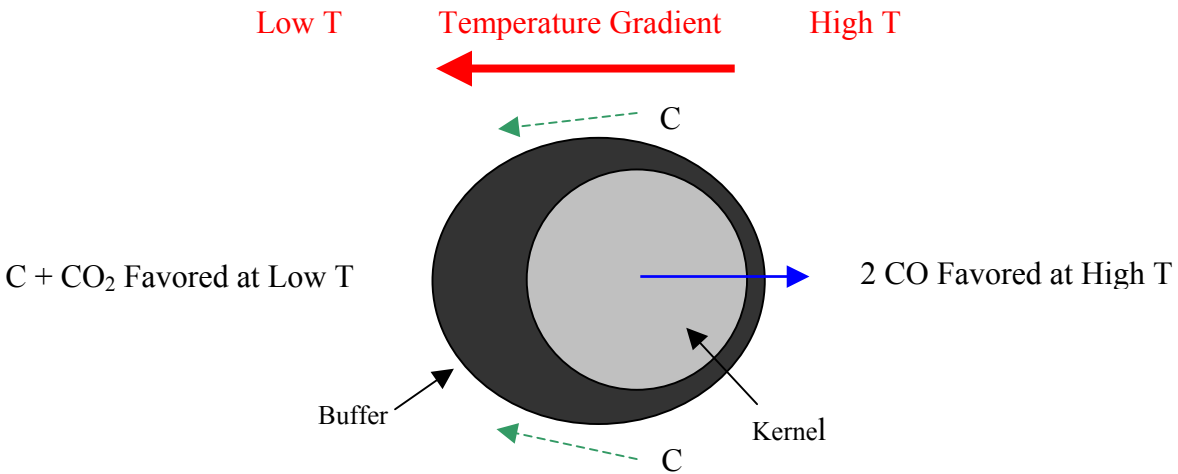


Figure 2-6 Illustration of the Amoeba Effect. Carbon is transported from the hot side to the cool side

An expression for the movement of the kernel is:

$$\Delta x = KMC \frac{1}{T^2} \frac{dT}{dx} t$$

where  $\Delta x$  is the kernel movement in meters,  $t$  is the time in seconds,  $T$  is the temperature ( $^{\circ}\text{K}$ ), and  $KMC$  is the migration factor. Typical migration factors are shown in Table 2-2.

**Table 2-2 Migration Factors**

Typical KMC at 1300 $^{\circ}\text{K}$ ( $\text{m}^2\text{K}/\text{s}$ )	
LEU UCO	$9 \times 10^{-13}$
LEU UO <sub>2</sub>	$6 \times 10^{-11}$
UC <sub>2</sub> (Solid state diffusion)	$9 \times 10^{-13}$

Particle failure is assumed to occur when the kernel touches the SiC layer. Examples of the Amoeba Effect are shown in Fig. 2-7.

The amoeba effect was a concern for the large (1000 MWe) HTGR designs using UC<sub>2</sub> and UO<sub>2</sub> kernels. Kernels with UCO significantly reduce this effect because CO production is minimized (see below) and solid-state carbon transport through the UCO is very low. Even at higher fuel operation temperatures, the modular prismatic designs that utilize UCO fuel are not expected to be significantly impacted by amoeba effect failures. Pebble bed reactors are also not expected to be significantly impacted by the amoeba effect because these reactors have small temperature gradients.

Thus, UO<sub>2</sub> fuel has been proposed as the fuel of choice in reactor systems with low temperature gradients and burnups in the range of 10%. This kernel type has had extensive testing under the conditions of interest for the modular steam cycle reactors and it is currently planned for direct cycle gas turbine systems (pebble bed).



Relative Thermal stability of HTGR Candidate Recycle Oxide Fuel Kernels Irradiated in HRB-7. Time-average temperature, 1200-1220EC; thermal gradient, 1000-1030EC/cm; fast fluence,  $6 \times 10^{21} \text{ n/cm}^2$ .

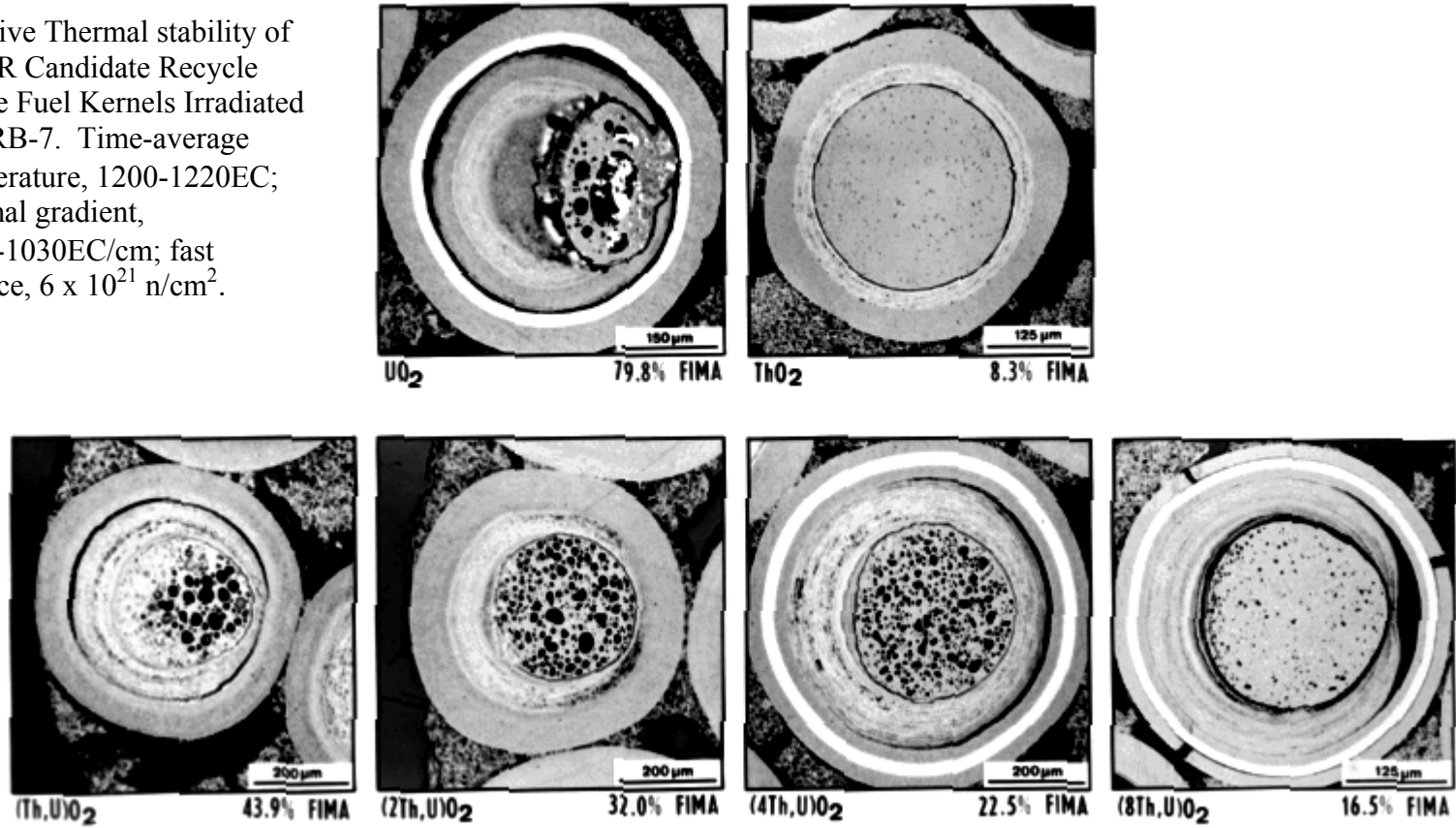
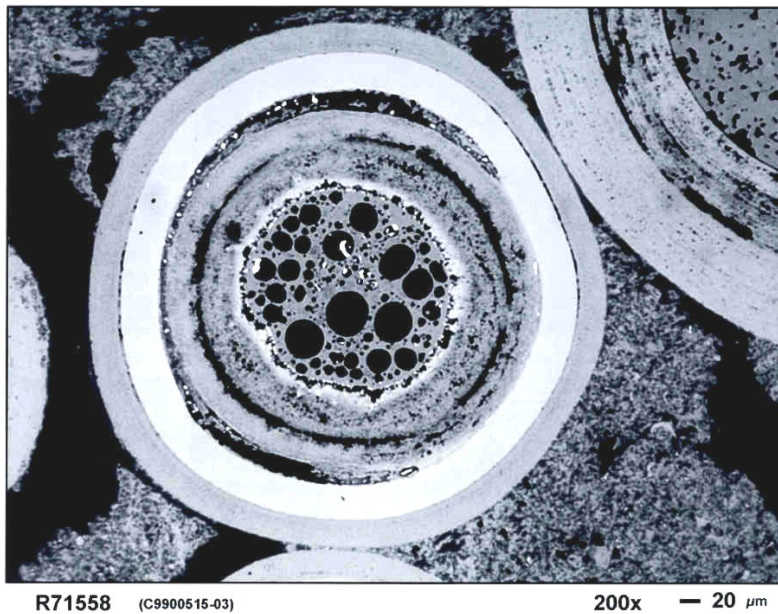


Figure 2-7 Oxide kernel fuel exhibiting the amoeba effect

Some reactor concepts have higher temperature gradients and greater fuel burnup needs and these reactors may not choose to use the  $\text{UO}_2$  based fuel. The prismatic core GT-MHR is likely to have fuel burnups in the range of  $\sim 20\%$  FIMA. Thus, there is interest in fuels that can achieve higher burnups without changing kernel migration or elevated internal pressures associated with CO.

Unlike lower burnup LWR fuel, the particle fuel designer cannot depend on the crystal structure of the kernel to contain a high percentage of fission gases and volatile fission products in high burnup fuel ( $>50\%$ ), especially under accident conditions. The relatively uniform kernel structure deteriorates with burnup and the kernel coatings must be relied upon for containing the mobile fission products. However, the kernel retains the refractory non-mobile compounds. A high burnup kernel is shown in Fig. 2-8. The figure shows a highly sub-stoichiometric plutonium dioxide kernel that limits CO problems by reducing the amount of oxygen initially available (note: it is not possible to fabricate highly sub-stoichiometric  $\text{UO}_2$ ). At high burnup the kernel structure becomes highly voided.



FTE-13,  $\text{PuO}_{1.68}$  Kernel,  $1150^\circ\text{C}$  ave,  $2.2 \times 10^{25}$  fluence, 70% BU

Figure 2-8 High burnup kernel showing the loss of crystal structure and the development of large voids

To avoid the problems associated with CO production, three approaches are possible. The first is to make a sub-stoichiometric kernel and thus limit the amount of oxygen available for CO production. This is possible with plutonium, but not with uranium. A second approach is to include a “getter” material in or near the kernel to absorb the released oxygen and make it unavailable for CO production. This approach has been

used, but involves greater complexity in particle production [2-40]. The third approach is to make a two-phase kernel consisting of both carbide and oxide phases.

The two-phase approach, known as UCO, allows the released oxygen from the oxide phase to oxidize the carbide phase at the expense of CO production [2-8]. This approach works because the oxidation energy of uranium is much lower than that of carbon. However, sufficient oxygen must be available to oxidize the rare earth elements, as the carbide forms of some rare earth elements are mobile and can migrate to the SiC and damage it. Figure 2-9 is a diagram of the process.

The two phases,  $UC_2$  and  $UO_2$ , interact in the following way. As oxygen is liberated, it first oxidizes the  $UC_2$  and rare earth elements because they have the greatest affinity for oxygen. Once they have been oxidized, oxygen is available for some of the elements with less affinity, such as Sr, Eu, Zr, and Ba, which were limited to carbide form earlier. Finally, only at the end of life is there enough oxygen for CO production. The goal is to balance this final CO production point with the need to oxidize the rare earths.

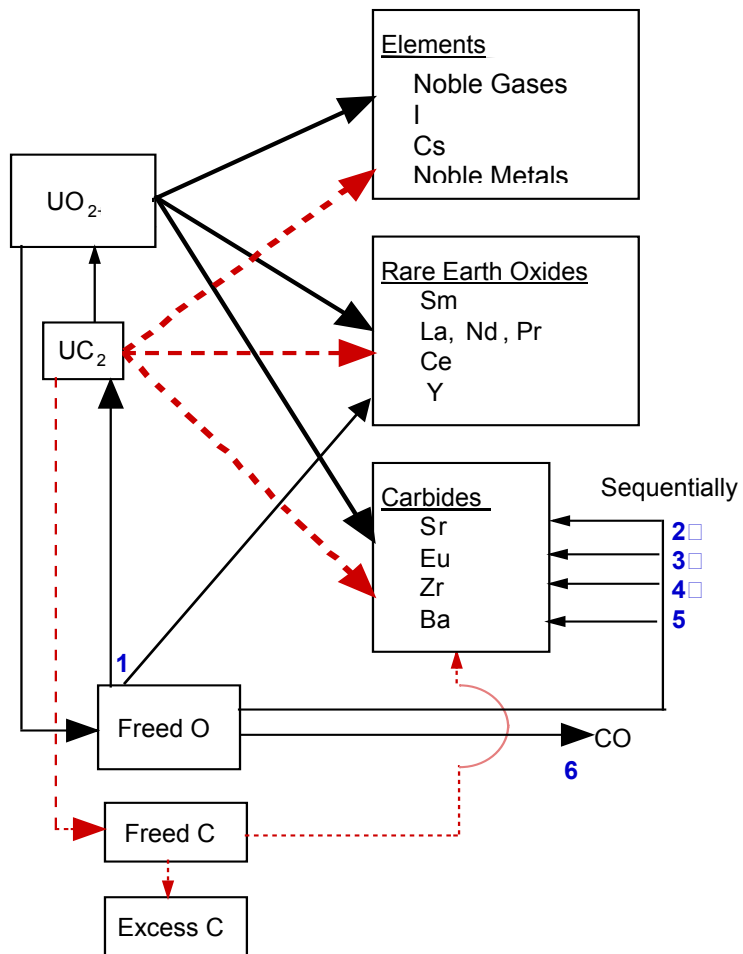


Figure 2-9 Oxygen distribution in a UCO kernel

The additional challenge of the UCO kernel is that it involves a more complex fabrication process and cesium, which can attack the SiC, may be more mobile (based on thermochemical calculations). Table 2-3 shows estimates for CO pressure. As seen from the calculations, UCO results in lower CO pressures for the higher burnups (>10%).

**Table 2-3 Order of Magnitude Estimates for CO Pressure**

T(°K)	Fuel	CO Pressure (atm)
1300	UCO	~0
	Low BU UO <sub>2</sub>	1
	High BU UO <sub>2</sub>	580
1600	UCO	~0
	Low BU UO <sub>2</sub>	5
	High BU UO <sub>2</sub>	1270

UCO kernels have had much less irradiation testing than UO<sub>2</sub> kernels and an important research objective is to further explore UCO behavior and performance limits.

To summarize, for higher burnup applications (>10% FIMA) it is important to control CO production (pressure) while still keeping the rare earth elements oxidized to limit their migration out of the kernel. The two-phase UCO kernel design is intended to provide these attributes without kernel migration so CO pressure problems can be avoided.

Kernel shape and density also have an impact on particle behavior. A manufacturing objective is to have a fairly round kernel so that the resulting coated particle departs little from sphericity [2-19]. The degree of sphericity effects the stress distribution in the coating. Significant departures from sphericity can result in high local stress in the coating layers. The density affects the amount of fissile material available in the particle, but it may also affect the degree to which the kernel can retain fission products (at least at lower burnup) and its reactivity with chlorine during the coating process (discussed later).

Unlike LWR fuel pellets, the kernel is at essentially a constant temperature and its behavior is more like that of “single crystal” release at low burnup; at high burnup the characteristic UO<sub>2</sub> structure is destroyed. In general, oxygen and carbon are used to form refractory compounds and the coatings are relied upon to hold the more volatile compounds.

Specifications for kernels are somewhat different than for LWR fuel pellets. The reader may note that there is less concern with grain structure or size and little mention of pores or complex structure. Table 2-4 lists a past kernel specification for typical German (pebble) fuel [2-2].

**Table 2-4 Typical German Kernel Specification**

Material	UO <sub>2</sub>
Enrichment, wt%	9.82
Sphericity	< 1.058
Diameter, μm	509 ± 9.7
Density, g/cm <sup>3</sup>	10.81 ± 0.048

**2.2.2.1 Kernel Design Factors Identified By The PIRT Panel**

Table 2-5 lists the important kernel design factors and their rationales. Both items discussed in the previous section and items of general interest are included.

**Table 2-5 Kernel Design Factors Identified By The PIRT Panel**

<b>Kernel Design Factor</b>	<b>Rationale</b>
Diameter	Power generation and fission product production
Density	Power generation and fission products, fabrication reactivity with chlorine (perhaps), and may impact fission product retention at low burnups
Sphericity	Effects the coating uniformity (stress distribution in coating layers)
Stoichiometry: Uranium to oxygen (UO <sub>2</sub> kernel)	CO production
Stoichiometry: Uranium to carbon and uranium to oxygen (UCO kernel)	CO production and oxidation of rare earths
Purity	General chemical and nuclear behavior (poisons)
Enrichment	General nuclear behavior and power production

**2.2.3 Buffer Layer**

The buffer layer surrounds the kernel and performs three main functions [2-6, 2-13 to 2-18, 2-25 to 2-27]:

- 1) Fission Product Recoil Attenuation. When uranium fissions, the resulting fission products are ejected at high velocity and are slowed down and stopped by the nearby material. Dense materials such as UO<sub>2</sub> and LWR fuel cladding limit the range of these recoils to roughly 10 microns. However, in low-density materials like carbon, the range of these recoils can be longer and they can cause significant local damage to the area they impact. The thickness of the dense outer layers in coated particle fuel is comparable to the recoil range. The buffer layer captures fission-produced recoils originating on the surface of the kernel and shields the IPyC from recoil damage.
- 2) Void Volume. The porous buffer layer provides the free volume for gas generation and expansion necessary to control the particle pressure.
- 3) Sacrificial layer. The buffer layer can distort to accommodate kernel swelling.

The thickness of the buffer layer affects the particle internal pressure. Too thin a buffer or a missing buffer layer will cause increased internal pressure, which can cause the particle to fail before the design burnup is reached.

The thermal conductivity of the buffer is not as high as the other coatings and too thick a buffer can raise kernel temperatures (somewhat) and thereby limit fuel core power density. Thus, the buffer thickness (already the thickest layer) is limited by pressure and heat transfer. The buffer layer is not required for particle strength, but it must be able to hold the kernel away from the IPyC.

Fast flux and recoils can cause shrinkage and cracking of the buffer layer. While not desirable, a certain amount of shrinkage and cracking is acceptable. However, a line of sight path from the kernel to the IPyC may expose the IPyC to serious recoil damage [2-28]. Figure 2-10 shows a particle with a distorted buffer layer. This particle did not perform well for other reasons (note the cracked OPyC and IPyC).



Figure 2-10 An example of a distorted buffer layer (HRB-21). (DOE-HTGR-100229)

It should be noted that the reference German pebble fuel did not exhibit buffer layer distortion under irradiation conditions. This may be because of different material properties and less challenging irradiation conditions [2-27]. The properties of coatings are currently a research topic.

The specifications for the buffer layer are shown in Table 2-6 (German fuel) [2-2]. These specifications are used in conjunction with process specifications; the layer generally has both process and product specifications.

**Table 2-6 Typical German Buffer Layer Specification**

Material	Carbon
Thickness, $\mu\text{m}$	$100 \pm 12.4$
Density, $\text{g/cm}^3$	$1.02 \pm 0.01$

### 2.2.3.1 Buffer Layer Design Factors Identified By The PIRT Panel

Table 2-7 details important buffer layer design factors and their rationale.

**Table 2-7 Buffer Layer Design Factors Identified By The PIRT Panel**

<b>Buffer Design Factor</b>	<b>Rationale</b>
Thickness	Void volume for gases, recoil attenuation, and distortion to accommodate kernel swelling. Also, the buffer affects the thermal impedance.
Density	

### 2.2.4 Inner Pyrocarbon Layer

The inner pyrocarbon layer (IPyC) is a higher density carbon layer deposited on the buffer layer [2-6, 2-13 to 2-18, 2-25 to 2-27]. It serves several functions:

- 1) It protects the kernel from chlorine (in the form of HCl) liberated during SiC deposition. Without the IPyC layer, chlorine would easily migrate through the buffer layer to the kernel, react with the uranium and produce volatile chlorides. These chlorides would then transport the uranium out of the kernel and contaminate the coatings. During operation, fissioning of this uranium contamination would then damage the layers. Fissioning outside of the kernel would also lead to increased fission product transport and releases from the particle.
- 2) It provides a smooth surface for SiC deposition (the buffer layer is too porous).
- 3) It delays transport of fission products to the SiC layer. The IPyC layer retains gases well and effectively isolates the SiC from CO, which can attack the SiC at higher temperatures. The layer does not effectively retain metals.
- 4) It can help maintain the SiC layer in compression. Depending on the IPyC/SiC layer bonding, the IPyC can place compressive forces on the SiC due to irradiation-induced shrinkage of the IPyC.

Good irradiation behavior requires that the pyrocarbon layer exhibit similar dimensional changes in the longitudinal and latitudinal directions for the fast fluence of interest [2-6, 2-8, 2-13, 2-25 to 2-27]. That is, it is desired that the carbon layer material and physical properties be anisotropic. This can be achieved by ensuring that the deposited carbon has a random rather than a preferred macroscopic crystal orientation. A measurement of anisotropy is known as the Bacon Anisotropy Factor (BAF). A BAF of 1 is completely isotropic, greater than 1 implies increasing crystal orientation. In practice, measurement techniques used to determine BAF have in some cases been inaccurate. In such cases the measured BAF has not correlated with irradiation performance as well as expected. This is an area of current research.

For the IPyC, there are six material and physical properties of interest to the designer of coated fuel particles:

- 1) Density
- 2) Permeability
- 3) Anisotropy
- 4) Creep
- 5) Shrinkage
- 6) Strength

These properties cannot be independently specified and two sets of conflicts are of particular designer interest. The first set is anisotropy and permeability. Some believe that a BAF of less than about 1.05 is necessary for good irradiation performance, with a BAF in the range of 1.02-1.05 as the desired target. However, permeability (in our case the transport of HCl to the kernel during SiC layer coating) tends to be less with a higher BAF (at least for past US fabrication experience; past German fabrication experience may have been somewhat better in this area). Thus, the designer has to balance irradiation stability and coating contamination. The situation can be made better or worst by the chlorine reactivity of the kernel.

Increased IPyC thickness would not resolve this problem as thicker IPyC can result in higher irradiation induced stresses in the IPyC and greater failure probabilities for the particle. Figure 2-10, previously shown, is an example of poor pyrocarbon performance. Both the inner and outer pyrocarbon layers failed by shrinkage-induced cracks. Figure 2-11 shows another pyrocarbon failure. This particle is an earlier design that lacked a SiC layer (buffer and OPyC only).

The other potential tradeoff is between IPyC shrinkage and IPyC creep. The fast flux causes shrinkage of the IPyC (influenced also by BAF), which is relieved by IPyC creep. The latter is a function of temperature. The designer has to minimize the rate of SiC stress increase caused by both particle pressure and pyrocarbon shrinkage due to fast flux with stress reduction by creep [2-13].

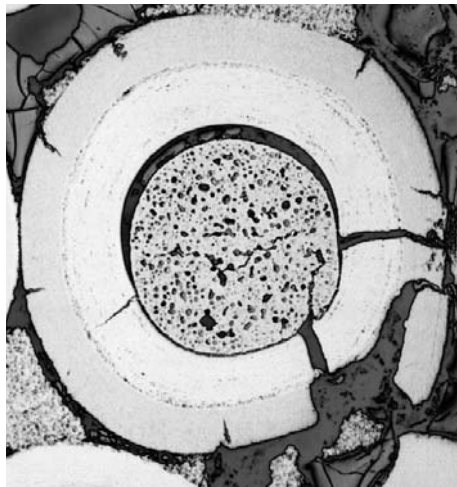


Figure 2-11 Example of pyrocarbon layer failure (BISO fuel)



Detailed modeling with accurate and manufacturing specific material properties is necessary to understand and optimize the tradeoffs in the particle design. The historical TRISO-coated particle design and fabrication process are an empirical solution to this problem for the design core environmental conditions.

The thermal properties of the (relatively thin and dense) IPyC are generally not as important as those of the buffer layer or the kernel. A more significant performance concern is IPyC radial cracking. Cracks (or debonding) in the IPyC can act as stress risers and induce locally high tensile stresses in the SiC layer if the between layer bond is strong [2-27, 2-29, 2-30]. Such cracks can also allow CO exposure to oxidize the SiC at high temperatures [2-31]. Figure 2-12 is an example of an IPyC crack that allowed CO to attack the SiC layer. Such attack occurs at elevated temperatures. Thus, this particle failure mode could be important in regions of power peaking.



Figure 2-12 CO oxidation of SiC (WAR UO<sub>2</sub> kernel at ~1500°C in HRB-10)

The past German product specifications for the IPyC layer are shown in Table 2-8 [2-2]. These product specifications are used in conjunction with manufacturing process specifications. The process specifications are used to control the other properties such as IPyC crystalline structure. PyC quality assurance will continue to require a process specification until such time that additional measured product parameters can be developed, measured, and controlled to ensure good irradiation performance.

**Table 2-8 Typical German IPyC Layer Specifications**

Material	Carbon
Thickness, $\mu\text{m}$	$39 \pm 3.9$
Density, $\text{g/cm}^3$	$1.92 \pm 0.007$
BAF	1.043

In summary, the IPyC plays three major roles. It protects the kernel from Cl attack during SiC layer processing, provides structural stability, and retain gases. Dimensional stability is important as cracking can lead to particle failure.

#### 2.2.4.1 IPyC Layer Design Factors Identified By The PIRT Panel

Table 2-9 details important IPyC layer design factors and their rationale.

**Table 2-9 IPyC Layer Design Factors Identified By The PIRT Panel**

IPyC Design Factor	Rationale
Thickness	Structural properties of the IPyC, gas retention, and control of possible HCl attack of kernel during fabrication
Density	Indirectly determines material properties. In particular, one is interested in radiation stability. Note that process specifications may also be needed to characterize this layer.
Anisotropy	The crystalline structure determines the irradiation stability of the layer.

#### 2.2.5 SiC Layer

The SiC layer is the primary fission product barrier in the coated particle. It was the major innovation that took the pyrocarbon-coated particle to its high level of fission product retention performance. The SiC layer has two major functions [2-6, 2-13 to 2-18, 2-25 to 2-27]:

- 1) Provides structural support to accommodate internal gas pressure.
- 2) It is the primary fission product barrier. It retains gases and metallics (except silver), but is subject to attack from palladium and rare earth elements.

For design purposes, the strength of the SiC is important as well as the strength distribution. For very low particle failure rates, the tails of the SiC strength distribution become important. In addition, SiC density, grain size and grain orientation as well as the trace amounts of free silicon in the layer are thought to be important. However, conclusive evidence connecting these properties to particle irradiation performance is lacking [2-8, 2-32 to 2-36].

The interaction of strains between the pyrocarbon layers and the SiC layer are important to SiC layer failure [2-13, 2-27, 2-29, 2-30]. Figure 2-13 shows the qualitative model stresses in a particle. Stress in the PyC layers is driven by gas pressure and irradiation induced shrinkage; it is relieved by irradiation-induced creep [2-13]. Both shrinkage and creep are temperature dependant. Stress in the SiC layer is driven by particle pressure and the relative stress distributions between the layers, which depend on material properties and layer bonding strengths. Two important points are made in this figure:

- 1) The particle should be designed so that the pyrocarbons keep the SiC in compression for as long as possible.
- 2) Failure of a pyrocarbon layer will change the stress distributions and will change the SiC stress from compression to tension at a lower burnup. Since the SiC is a brittle material, the particle designers seek to keep the pyrocarbon layers intact over the design burnup.

For design purposes, intact SiC is assumed to retain all fission products at normal operating temperatures except for silver, which has a high release rate above 1100°C.

As the operating temperatures increase (>1250°C) fission product attack of the SiC becomes more likely. The major concerns are the lanthanides (even at lower temperatures) and palladium. Design of the kernel can retain the lanthanides as oxides, but palladium (noble metal) cannot be tied up and migrates (diffuses) to the SiC at the higher temperatures where it attacks the layer. This behavior effectively limits the normal operating temperatures (below ~1300°C) [2-8, 2-35 to 2-36].

At accident temperatures, above ~1600-1800°C, fission product release quickly increases. Above about 2000°C, thermal decomposition of SiC is a dominant failure mechanism.

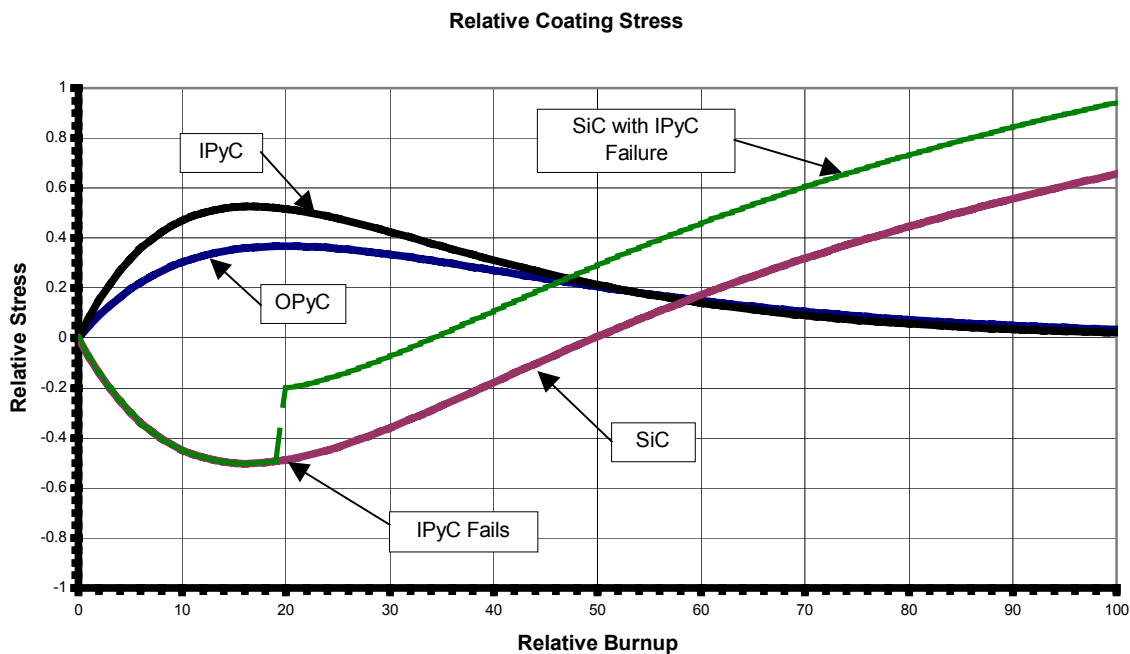


Figure 2-13 Qualitative stresses in coated particle layers for a model particle. (Assumes weak bonding between the IPyC and SiC – strong bonding may create local stress risers in the SiC)

However, above about 1600°C decomposition effects in the SiC such as the development of porosity are noted, implying that thermal decomposition mechanisms are active. At the assumed particle temperature limit, ~1600°C, diffusion of fission products begins to

increase (over normal operating values) and limits the time at temperature [2-6, 2-21 to 2-24].

To summarize, the designer tries to keep the SiC layer in compression during most of the irradiation and limits the operating and accident temperature to control SiC layer corrosion and decomposition.

The past German specifications for the SiC layer are shown in Table 2-10 [2-2]. These product specifications were used in conjunction with manufacturing process specifications. The process specifications are used to envelope the other properties such as SiC microstructure. The specifications for the SiC layer for contemporary HTGR particle fuel may include grain size and orientation as well as strength and strength distribution. This is an area of current research and the relevance of these items is being studied.

**Table 2-10 Typical German SiC Layer Specifications**

Material	SiC
Thickness, $\mu\text{m}$	$35 \pm 1.9$
Density, $\text{g/cm}^3$	$3.20 \pm 0.007$
Fraction Defective, (mean value)	$7.7 \times 10^{-6}$

### 2.2.5.1 SiC Layer Design Factors Identified By The PIRT Panel

Table 2-11 details important SiC layer design factors and their rationale.

**Table 2-11 SiC Layer Design Factors Identified By The PIRT Panel**

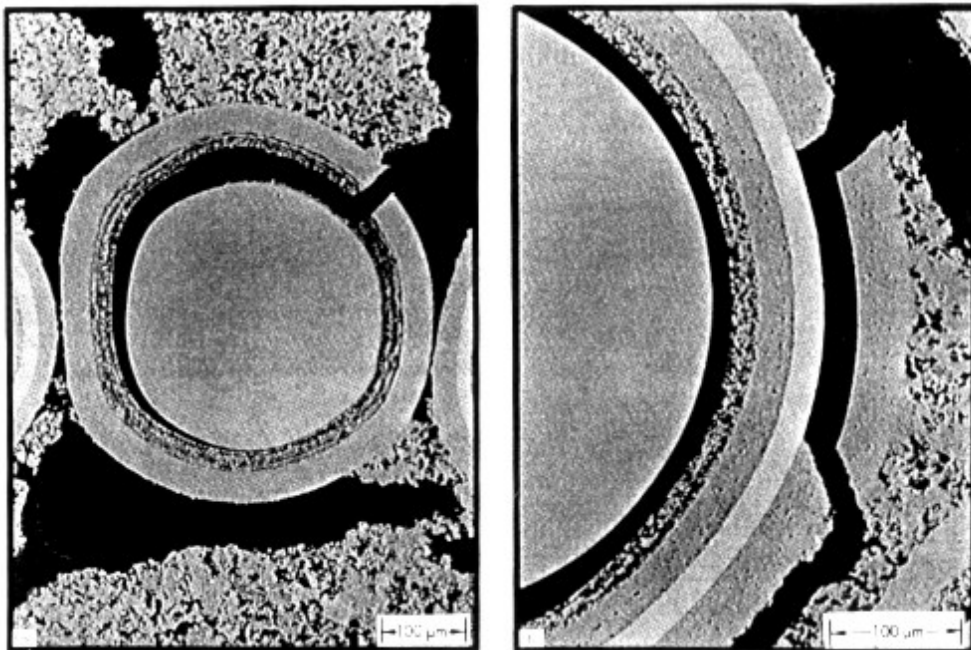
<b>SiC Layer Design Factor</b>	<b>Rationale</b>
Thickness	Determines the strength of the layer. Distribution of thickness is also important.
Density	Indirectly determines material properties. Desire high density and small grain size.
Fraction with defective SiC layers	The number of initially defective particles is an upper limit on fuel performance and impacts fabrication effort/cost.

### 2.2.6 Outer Pyrocarbon Layer

The outer pyrocarbon layer (OPyC) is the final layer on the coated particle and is the layer that binds the particle to the fuel form [2-6, 2-8, 2-9, 2-13 to 2-18, 2-25 to 2-27]. Many of the performance factors associated with the OPyC are similar to those of the IPyC, especially irradiation stability, but there are differences. The OPyC:

- 1) Protects the SiC during fuel particle handling prior to fuel element fabrication.
- 2) Provides a bonding surface for the matrix material.
- 3) Compresses the SiC during irradiation.
- 4) Acts as a final barrier to gaseous fission product release.
- 5) Provides some isolation of the SiC from external chemical reactions.

The six material properties of importance to the IPyC are important to the OPyC. Permeability of the OPyC is also important and is related to the intrusion of matrix material into the pores of the coating. If the OPyC were too permeable, it could result in too strong a bond between the OPyC and the fuel element matrix material. This could fail a coating as the materials differentially shrink from irradiation exposure. This is illustrated in Fig. 2-14.



This

strong bonding was a cause of fuel particle failures for past US made fuel and lead to the Figure 2-14 Particles Broken During Irradiation Due to Matrix-Particle Interaction.

Carbonized in graphite tube. Irradiated to  $3.6 \times 10^{21}$  n/cm<sup>2</sup> ( $E > 0.18$  MeV) at 940-1145°C. Left - BISO particle / Right TRISO particle

introduction of additives to the matrix binder to control the bonding strength. The contemporary US approach is to replace the fuel element injection fabrication approach (requires the thinner resin) with a new process that does not require the thinner resins and their resultant bonding concerns. The former and later fuel element fabrication methods are detailed in later sections.

The irradiation shrinkage and creep of the OPyC play similar roles as were outlined for the IPyC. The properties of interest to the OPyC are similar to those of the IPyC with the exception of permeation, which is important to matrix bonding rather than chlorine transport to the kernel.

The past German specifications for the OPyC layer are shown in Table 2-12 [2-2]. These product specifications were used in conjunction with process specifications. The process specifications are used to envelope the other important properties. OPyC quality assurance will continue to require a process specification until such time that additional measured parameters can be developed, measured, and controlled to guarantee good irradiation performance.

**Table 2-12 Typical German OPyC Layer Specifications**

Material	Carbon
Thickness, $\mu\text{m}$	$39 \pm 3.6$
Density, $\text{g/cm}^3$	$1.92 \pm 0.02$
BAF	1.028

#### 2.2.6.1 OPyC Layer Design Factors Identified By The PIRT Panel

Table 2-13 details important OPyC layer design factors and their rationale. They are similar to those identified for the IPyC.

**Table 2-13 OPyC Design Factors Identified By The PIRT Panel**

<b>OPyC Layer Design Factor</b>	<b>Rationale</b>
Thickness	Strength of the OPyC and gas retention if the SiC fails.
Density	Indirectly determines material properties. In particular, one is interested in radiation stability. Note that process specifications may be needed to characterize this layer. Also, bonding between the layer and the matrix must be controlled.
Anisotropy	The crystalline structure determines the irradiation stability of the layer.

### 2.2.7 Fuel Element

The fuel element provides a fixed uniformly random distribution of fuel particles and matrix material with good irradiation and thermal properties [2-2 to 2-4, 2-6, 2-9]. It satisfies several needs:

- 1) Allows the fuel to be handled and placed in the reactor without damage to the particles.
- 2) Stabilizes the particles so they do not shift or move relative to the matrix.
- 3) Provides a good heat transfer medium from the particles to the reactor environment.
- 4) In the cases of pebbles, isolates the particles from impacts.

Generally, two fuel forms are considered:

- 1) Fuel compacts in graphite prismatic blocks. The cylindrical fuel compacts are approximately 2" long by ½" diameter (US design).
- 2) Fuel pebbles. Spheres 6 cm in diameter (Germany, China, South Africa).

Pebble fuel is the reference case for this PIRT.

The fuel form is selected on the basis of reactor fuel cycle, thermal considerations, particle loading requirements, and whether or not it must be removed from a "block" at end of life. Also, the number of particles broken during fuel element fabrication can be a consideration.

The number of broken particles along with the tramp uranium in the matrix material determines the quality of the fuel as both release fission products into the primary system.

The power produced by a fuel element depends in part on the number of particles; however, more particles per element (particle loading) can lead to increased particle damage during fabrication. This is a fundamental trade-off that can greatly influence the integrity of the element by limiting fabrication choices (described later). Power distribution can be skewed by an inhomogeneous distribution of particles within the element. This distribution must be controlled within designer limits.

The fuel element can be made by placing particles (and shim) in a mold and injecting matrix material to form a unit or by coating the particles with a thin soft layer of matrix material and forming the overcoated particles into shape under pressure. For pebble fuel an unfueled layer of matrix material is added to the outside of the fueled zone to protect the fuel particles from fuel element impacts associated with on line refueling and from pebble contact with other pebbles or walls during pebble flow through the core. This will be described in greater detail in the fabrication section. At this point, the designer is interested in the shape, heat transfer properties, damaged particle fraction, and any impurities in the matrix that may cause problems.

Table 2-14 lists some key German design specifications for their past pebble fuel elements [2-2].

**Table 2-14 Historic Reference German Fuel Element Specifications**

Outer Diameter, mm	60.0
Fuel-free shell thickness, mm	5.0
Uranium loading, grams per element	7.0 to 10
<sup>235</sup> U enrichment, %	8 to 9.7
Free uranium fraction	$6 \times 10^{-5}$

### 2.2.7.1 Fuel Element Design Factors Identified By The PIRT Panel

Table 2-15 details the important fuel element design factors and their rationale.

**Table 2-15 Fuel Element Design Factors Identified By The PIRT Panel**

<b>Fuel Element Design Factor</b>	<b>Rationale</b>
Matrix material specification	The matrix material binds the fuel particles in a uniformly random distribution and contributes to the fuel element properties.
Particle packing fraction	This parameter determines in part the nuclear and thermal power properties of the fuel element. Particle damage during manufacture is more likely at higher packing fractions.
Unconfined heavy metal outside SiC layer	Unconfined heavy metal results in fission products in the primary circuit and the potential for releases during off-normal conditions.
Particle distribution in fuel element	Inhomogeneous particle distribution within fuel elements can result in hot spots.
Particle overcoat	This layer protects the particle during fuel element fabrication by deforming, providing a particle-to-particle spacing function, and integrating the particle into the matrix.
Fuel free zone (Pebble)	The fuel pebble requires a fairly strong outer layer to protect the inner-fueled region from damage as the pebble must be repeatedly dropped several meters into the pebble bed core.

## 2.3 Manufacturing

### 2.3.1 General

The kernel coating layers are added in a fluidized bed coater. The coating process is statistical and results in a distribution of attributes. The goal is to control the process so that the distribution meets the demanding manufacturing specifications and the process is predictable. A simplified diagram of a coater is shown in Fig. 2-15.



The coating chamber consists of a graphite tube that is maintained at the desired temperature by electrical heating. A fluidizing gas is introduced to the chamber by a gas distribution nozzle at the bottom. The flow rate of the fluidizing gas is enough to levitate and randomly circulate the bed of particles, but not so much as to eject particles out of the top of the coater chamber, which could result in coating defects. Separate coating gases are introduced and gas ratios automatically changed and controlled to produce the desired coating [2-8, 2-25 to 2-27, 2-37]. Hydrocarbon gases are pyrolytically decomposed into carbon and hydrogen for the application of the buffer, IPyC, and OPyC layers.

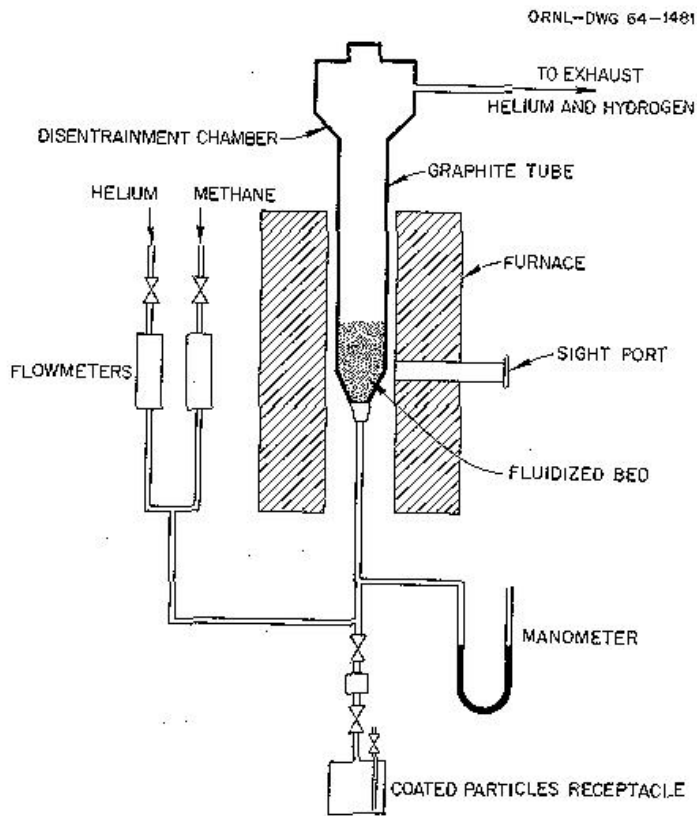


Figure 2-15 Diagram of a coater (from ORNL-4324)

During the coating process the bed of particles is agitated continuously by the levitation (fluidizing) gas and each particle is coated over time with about the same amount of material. Since heat transfer rates are very high in a fluidized bed, thermal gradients are generally very low within the bed. Temperatures can be monitored with optical pyrometers or thermocouples through access ports. When the desired coating run has been completed, the flow is reverted to pure fluidizing gas and reduced until the particles drop to the bottom.

Important variables in the design of the coater are the shape of the inlet nozzle and cone or “frit” at the inlet to the reaction chamber. The gas flow distribution angle affects the refluxing action of the fluidized bed, the tendency of soot plugging, and the gas flow distribution. The exact influence of all these factors combined with the operating parameters is not well understood and some experimentation is necessary with each coater design to make particles within specifications. The temperatures, sources gases, and coating deposition rates are particularly important and determine the coating properties. Also, during manufacture of a coated particle lot the diameter and mass of the particles increase, but compensating the flow rate for this fact has not always been done, although it appears to be necessary to assure uniformity of coating properties.

Coaters have been operated in either of two ways: continuous or interrupted. In a continuous coating process the coating layers are put on one right after another by changing the feed gases, flow rates, and temperatures as a function of time. That is, the particles are not removed from the coater after each layer is completed. This method has the advantage that there are no interruptions to introduce defects into the coatings or the way the coatings adhere to each other. The disadvantage is that defective particles cannot be removed or problems identified until the process is complete. However, particles can be siphoned off the bed for sampling after a layer is added. Interrupted coating empties the coater after a layer has been applied and allows a layer-by-layer inspection of a batch, but the extra handling can introduce problems of its own such as impurities and coating damage.

The typical feed gases are argon, hydrogen, acetylene, propylene, and methyltrichlorosilane (for the SiC). The coating rate effects PyC anisotropy, with low rates, 1-4 micron/min, favoring high BAF, and high rates, 4-10 micron/min, favoring low BAF [2-8, 2-25, 2-27]. A tradeoff between porosity and density occurs in PyC coating and optimizing the tradeoff in these properties to some degree may be necessary. To date, process knowledge generally has been necessary to characterize a coating, as measured product characteristics alone were not found to be sufficient to assure good irradiation performance.

### **2.3.1.1 Manufacturing Process Phenomenon Identified By The PIRT Panel**

Table 2-16 lists factors identified by the PIRT panel for coated particle manufacturing.

### **2.3.2 Kernel**

The kernel is made by an ammonia-based gel-precipitation process, referred to as either “internal” or “external” gelation. Briefly, for internal gelation, uranium is dissolved in nitric acid and mixed with urea (and carbon for a UCO kernel). The mixture is then chilled and mixed with hexamethylene (HMTA) to form a broth. This broth is only stable at low temperatures (~0°C) and is kept chilled. The broth is then pulsed through needle orifices to form droplets that fall into a heated column of immiscible liquid. The rise in temperature causes internal ammonia production and the droplet to gel. The resulting spheres sink to the bottom of the column, are removed, and are then washed in ammonium hydroxide to remove ammonium nitrate and dried.

**Table 2-16 Manufacturing Process Phenomena Identified By The PIRT Panel**

<b>Manufacturing Factor</b>	<b>Rationale</b>
Layer coating process specifications: Gases (levitation gas and coating gas)	The gases used in the coater directly influence the quality of the layer and the operation of the coater.
Layer coating process specifications: Ratio of gases	The gas mixtures affect the layer properties and production rate.
Layer coating process specifications: Temperature	The properties of the coating layer are dependant on the coater temperature.
Layer coating process specifications: Coating rate	The microstructure of the coating layer is influenced by the coating rate.
Layer coating process specifications: Pressure	Pressure affects reaction rates. (The coaters are generally operated at atmospheric pressure.)
Layer coating process specifications: Coater size	Coater size effects the distribution of layer properties.
Layer coating process	Continuous versus interrupted coating may affect coating layer interface properties.
Process control	Controlling the process is important. Coating product measurements may not be sufficient to guarantee good irradiation performance.
Product control	Coatings must meet designer specifications.

An external gelation process can also be used. This process also induces gelation with ammonia, but the source of the ammonia is external to the droplet. A somewhat different broth is prepared and pulsed through needle orifices, but this time the droplets fall through an ammonia vapor phase and then into an ammonium hydroxide containing aqueous column to induce the gelling. They are then washed and dried. The external process is used for the reference Pebble fuel [2-1 to 2-3, 2-25].

The dried spheres are calcined and sintered in a hydrogen atmosphere to remove the excess oxygen. If they are UCO kernels, they are next sintered in an argon or argon/CO atmosphere to adjust the O/C ratio for a UCO kernel.

The kernels are then screened for size and tumbled to eliminate the non-round and odd shapes. They are inspected for size, size distribution, density, and stoichiometry. Figure 2-16 is a flow diagram of the external gelation process (reference for this PIRT).

Table 2-17 outlines important properties and likely quality control methods [2-1 to 2-4, 2-38, 2-39].

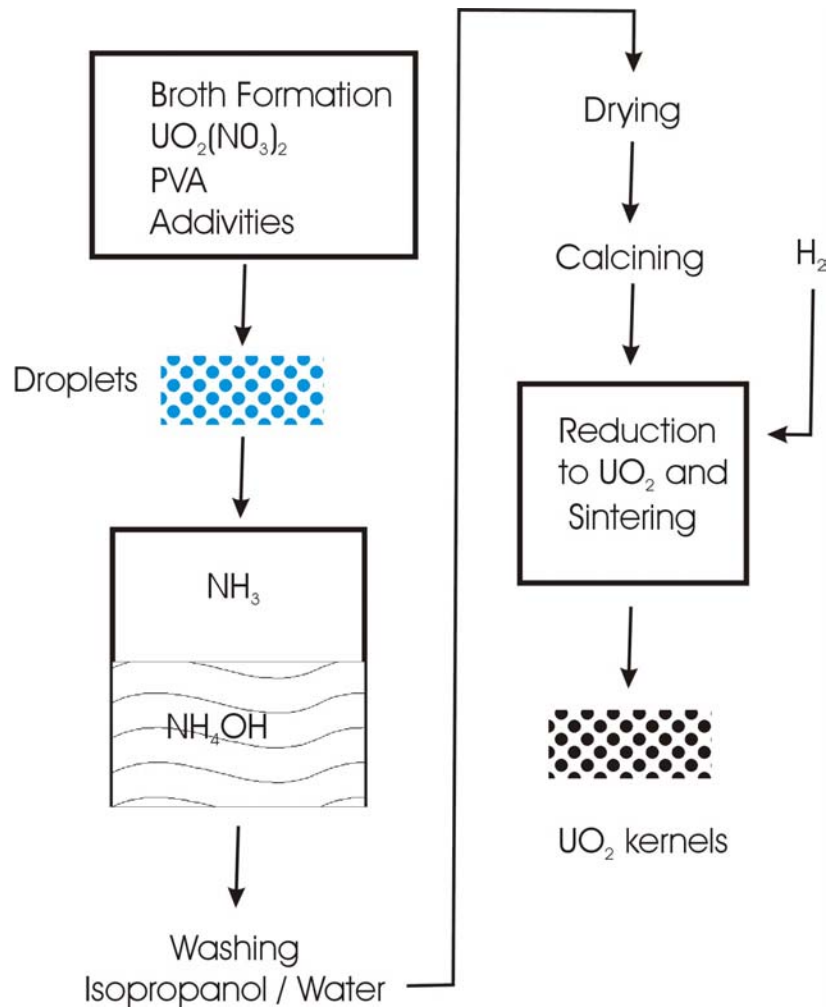


Figure 2-16 Flow diagram of external gelation kernel fabrication process

**Table 2-17 Kernel Properties and QC Methods**

**Table 2-17 Kernel Properties and QC Methods**

Kernel Attribute	QC Method
Density	Pycnometry, density column
Diameter	Particle size analyzer, imaging (sphericity)
C/U and O/U	Combustion and wet chemistry, metallography/image analysis (phases)
Impurities	Spectrographic (mass) methods and wet chemistry

### 2.3.2.1 Kernel Manufacturing Phenomenon Identified By The PIRT Panel

Table 2-18 lists the kernel manufacturing factors identified by the PIRT panel.

**Table 2-18 Kernel Manufacturing Factors Identified By The PIRT Panel**

<b>Kernel Manufacturing Factor</b>	<b>Rationale</b>
Density	The density of the kernel determines the fissile material present and thus power. It may also affect the HCl reactivity during coating.
Microstructure – UO <sub>2</sub>	Microstructure is known to affect performance in other types of reactor fuel.

### 2.3.3 Coated Particles

The coating layers are deposited on the kernel in a fluidized bed by the thermal cracking of the appropriate gas in a fluidizing gas such as argon [2-8, 2-18, 2-25 to 2-27, 2-37 to 2-38]. Hydrocarbon gases such as acetylene and propylene are used for the carbon layers. MTS is used for the SiC layer and it is reduced by hydrogen. Temperatures are in the range of 1200 to 1500°C and the flow rates of the gases are adjusted to achieve the desired deposition rate.

Layer properties are controlled by temperature, coating rate, coating gas composition, bed loading, and particle size. In general, each layer has its own optimal combination of parameters that are determined experimentally for a particular coater. A flow diagram of the process is shown in Fig. 2-17. Note that the process may be continuous or interrupted. In the continuous process, the particles remain in the coater and the composition of the gases and furnace temperature are changed so the coatings can be put on one after another. In the interrupted process, the coater is unloaded after each coating and the particles can be checked and sorted for gross defects such as out-of-roundness. Sampling can be used for destructive investigation. The bad particles (or a bad batch) are discarded before the next layer is applied.

At the present time, the continuous coating method has been demonstrated to give acceptable results, but this conclusion is still tentative. The current trend is toward continuous coating and the highest quality fuel (reference material) has been produced by this method.

An item of interest for the PIRT review is that the specification of layer product properties is not sufficient to ensure satisfactory irradiation performance. At the present state of the art, modest changes in the operation of the coater (such as design issues, coater size, and exactly where the bed temperature is measured) can lead to coating property changes that can result in substandard irradiation performance. These changes either cannot be observed by the present QC methods or the changes in material properties are not currently measured.

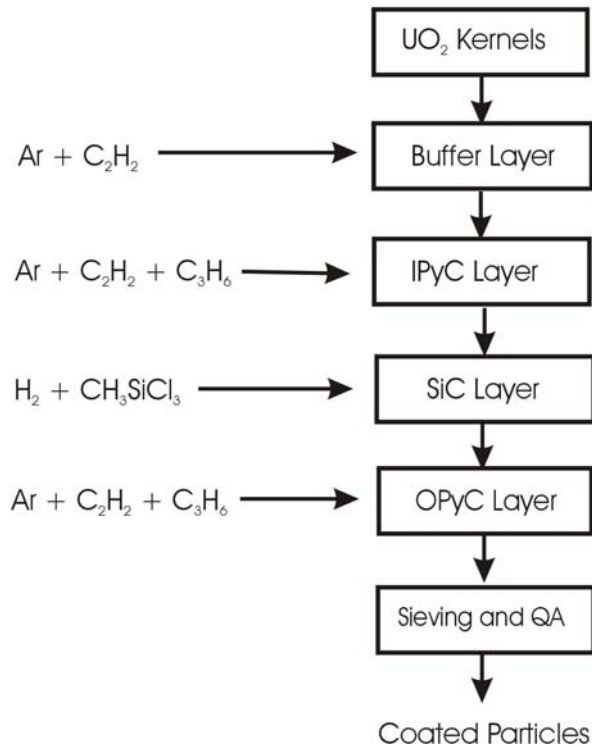


Figure 2-17. Flow diagram of the continuous coating process

This is addressed by having both layer fabrication process and layer product specification. Thus, both process knowledge and product measurements are required to determine if the fuel has been properly fabricated. This issue appears to be particularly important for pyrocarbon layers as was discussed in the design section. The BAF measurement technique is also important. The SiC layer is very important for the control of fission product transport and it is sensitive to the details of coater operation.

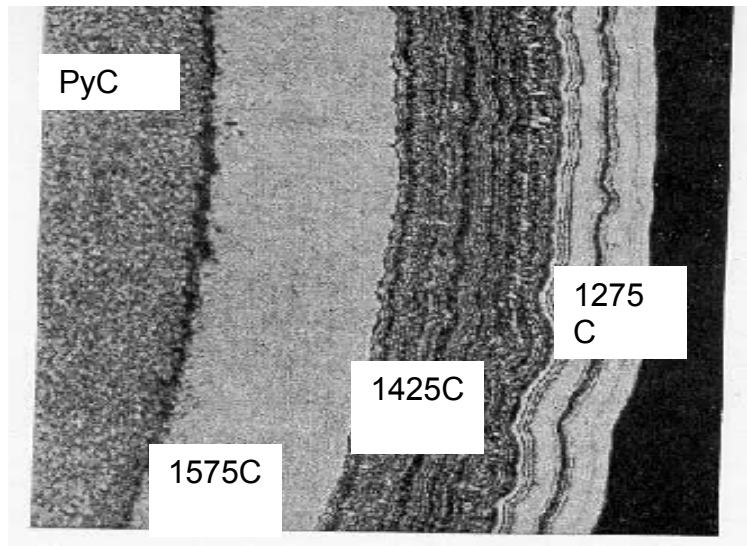
Figure 2-18 shows how the nature of deposited SiC can change with temperature. The fabricator would like to control free silicon, grain size and grain orientation. SiC has shown good irradiation properties, but like pyrocarbon, a clear one to one correlation between measured properties and irradiation behavior is not available at present.

Table 2-19 shows the coating layer product properties measured during fabrication and the measurement methods that are typically employed. The reader is cautioned that measurements alone do not provide a complete picture of the fabrication parameters and must be used in conjunction with process knowledge [2-27, 2-32, 2-33, 2-37, 2-39].

### 2.3.3.1 Coated Particles Manufacturing Factors Identified By The PIRT Panel

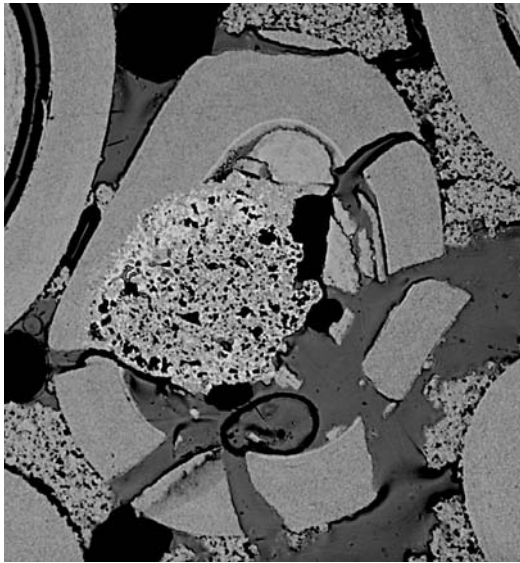
Table 2-20 lists the manufacturing factors identified by the PIRT panel.

Finally Fig. 2-19 shows that particles don't have to be perfect; some out of "roundness" can be tolerated. Severely malformed particles, however, are to be removed from the lot.

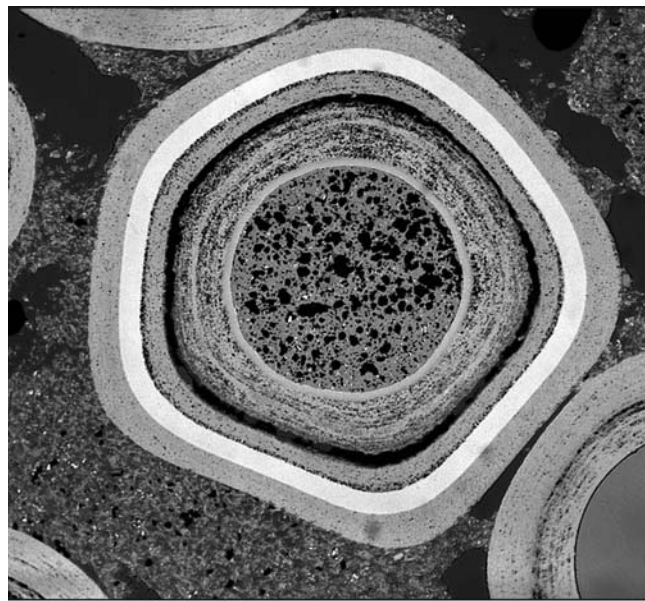


Etched SiC  
ORNL/TM-5152

Figure 2-18 SiC structure as a function of coater temperature



Malformed Particle



R71610 (C9900614-23)

200x

Particles with facets  
have performed well  
(FTE-13 Pu,Th)

Figure 2-19 Performance of non-spherical particles



**Table 2-19 Coating Layer Product Factors and Typical QC Methods**

<b>Layer Attribute</b>	<b>QC Method</b>
<b>Buffer Layer</b>	
Thickness	Radiography, metallography
Density	Mercury pycnometry and carbon content analysis (LECO)
Missing or thin layer (a failure mechanism)	Radiography
<b>IPyC Layer</b>	
Thickness	Radiography, metallography
Density	Liquid gradient column
Anisotropy	BAF (other methods under study)
Microstructure	Coating rate and process conditions (temperature, coating gases, time) [Process Knowledge]
Permeability (The heavy metal dispersion will signal a missing layer)	Heavy metal dispersion into layers (Radiography, chemical analysis)
<b>SiC Layer</b>	
Thickness	Radiography, metallography
Density	Liquid gradient column
Microstructure	Coating rate and process conditions (temperature, coating gases, time) [Process Knowledge], metallography
Spatial defects or missing layer	Burn/leach
Strength	Crush tests, brittle ring tests
<b>OPyC Layer</b>	
Thickness	Radiography, metallography
Density	Coating weight and pycnometry
Anisotropy	BAF
Microstructure	Coating rate and process conditions (temperature, coating gases, time) [Process Knowledge]
Missing or defective layer	Optical microscopy
Surface connected porosity	Mercury porosimetry

**Table 2-20 Manufacturing Layer Product Factors Identified by the PIRT Panel**

<b>Coating Layer Manufacturing Factor</b>	<b>Rationale</b>
<b>Buffer Layer</b>	
Thin (less than specified)	A thin buffer layer can lead to particle failure due to overpressure.
Density and open porosity	The buffer layer is the void volume to accumulate the released fission gases and any generated CO. It determines the internal pressure.
<b>IPyC Layer</b>	
Porosity	High porosity can allow HCl liberated during SiC deposition to attack the kernel and spread fissile material to the other layers.
Bonding strength to SiC	The bonding strength determines how forces are transmitted from one layer to another. May be most important for IPyC.
Anisotropy	The anisotropy is important as it determines the dimensional stability of the layer under irradiation.
<b>SiC Layer</b>	
Defects	Defects in the SiC can allow fission products to diffusion out of the particle.
Heavy metal dispersion	Heavy metal in the SiC layer will fission and damage it.
Stoichiometry	High quality SiC is important. Free silicon may be detrimental.
Bonding strength to OPyC	The bonding strength determines how forces are transmitted from one layer to another.
Density	Density can indirectly determine retention properties and strength.
Fracture strength	The strength of the layer determines the integrity of a particle. The tails of the distribution determine the particles with marginal strength
Grain size and microstructure	The microstructure determines the layer diffusion properties.
<b>OPyC Layer</b>	
Porosity	The porosity of the OPyC can determine the strength of the OPyC to matrix bond.
Anisotropy	The anisotropy is important as it determines the dimensional stability of the layer under irradiation.

### 2.3.4 Fuel Element Manufacture

Once the particles have been coated and inspected, the next step is to form the fuel element. Over the years, several fuel element forms have been considered, but two are now of contemporary interest to US HTGR licensing. They are: fuel compacts in prismatic graphite blocks (US) and fuel pebbles (Germany, reference case). The fuel form and particle loading depends on the power required from the element, the required fissile loading, and the temperature and temperature gradients. In addition, the tolerable particle breakage depends on the fuel element loading as higher particle loading generally increases the risk of particle damage [2-9].

An important goal of fuel element fabrication is to minimize the amount of uranium outside the particles by limiting the number of defective particles from the manufacturing process, minimizing the damage done to particles during the element forming process, and minimizing the uranium impurities in the fuel element feedstock materials. With particle defects at the  $10^{-5}$  level, limited particle damage during handling, and high purity materials, the exposed uranium in a fuel element is expected to be equivalent to no more than a particle defect fraction of  $\sim 10^{-5}$ .

The fuel element (compact or pebble) is formed from the fuel particles, which may or may not be overcoated with a matrix material, and the fuel element matrix material. The matrix material is a mixture of binder (resin or pitch), graphite flour, additives, and graphite shim (if required). The binder may be a thermosetting type (resin) that becomes rigid with exposure to elevated temperatures or a thermoplastic type (often a pitch) that always softens and flows with exposure to elevated temperatures.

Fuel element fabrication may be cast into two broad categories [2-1 to 2-4, 2-9]. The first is the so called "Admix" process where the particles, matrix, and any shim material are first mixed together and then molded to shape at temperature (used for reference fuel). With this process, the particles are often first overcoated with matrix material. The matrix material used is a highly viscous mixture of binder and graphite powder that does not flow. This process is generally limited to lower particle packing fractions of no more than roughly 30-40%. Somewhat higher packing fractions are possible, but the probability of particle breakage increases with higher packing fractions. Figure 2-20 illustrates the general process.

The particles are usually first overcoated with matrix material. The overcoated particles are then mixed with additional matrix material (as necessary) to create a uniformly random distribution of particles and matrix material of a specific volume, which is then placed into a mold. If the particles are not overcoated first, the matrix material may be introduced as a powder or flakes and melted during processing; it need not be "wet" to start with. Pressure and heat are applied to form the fuel element to shape. To add the unfueled layer of a pebble, the fueled center is fabricated as described above, then surrounded by more unfueled matrix material, placed in a larger spherical mold, and formed as before. The result is a "green" fuel element ready for carbonizing.

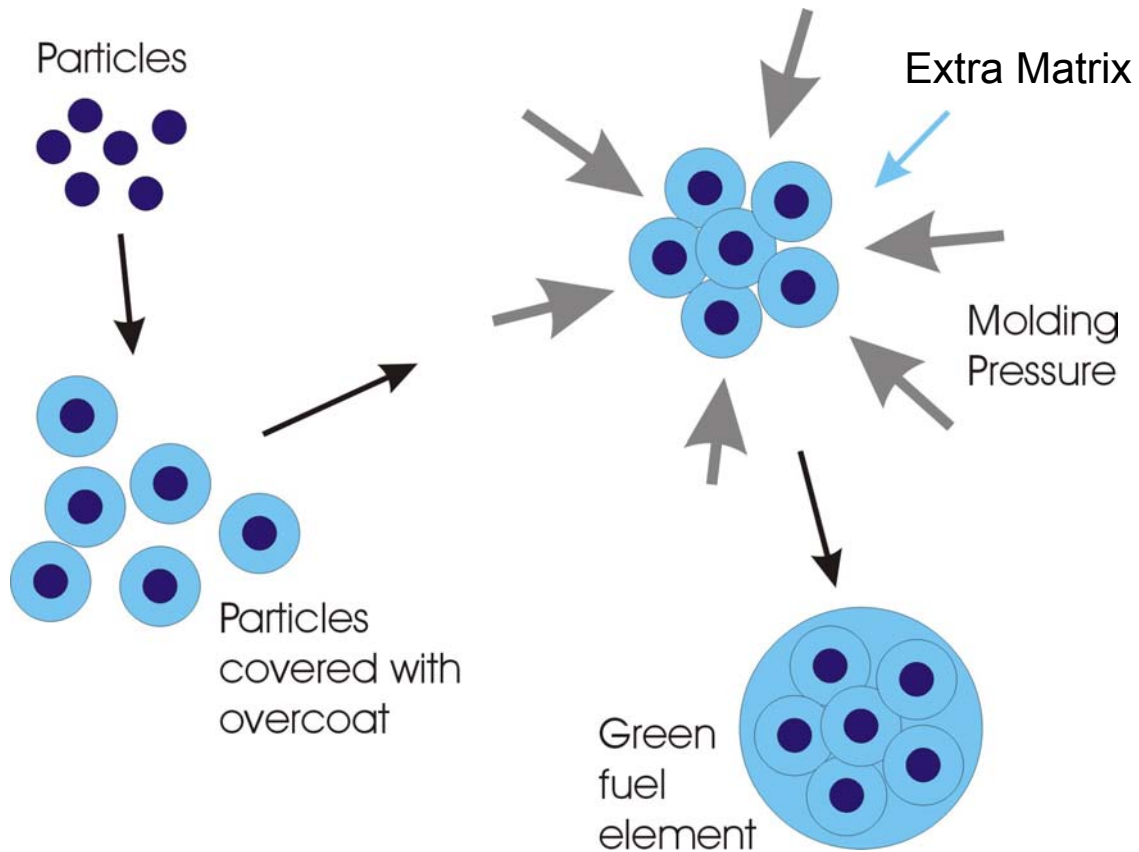


Figure 2-20 Illustration of the Admix fuel element forming process

Either thermosetting or thermoplastic binder may be used, but thermosetting resin is often chosen, as the “green” element will not slump upon further heating.

The second process is the injection method. With this process, the particles and any shim are first put into a mold and compressed by a piston. Next a flowable mixture of binder (usually pitch) and graphite flour is injected into the mold and allowed to harden by cooling the mold for thermoplastic binder or elevating the temperature to the set point for thermosetting binder. After the element has hardened, it is ejected from the mold. See Fig. 2-21. This method has often used thermoplastic pitch to get the desired matrix and fabrication properties.

Because this method needs a flowable mixture, less filler material can be used in the matrix, making it weaker. However, much higher particle packing fractions can be accommodated ~50-60%.

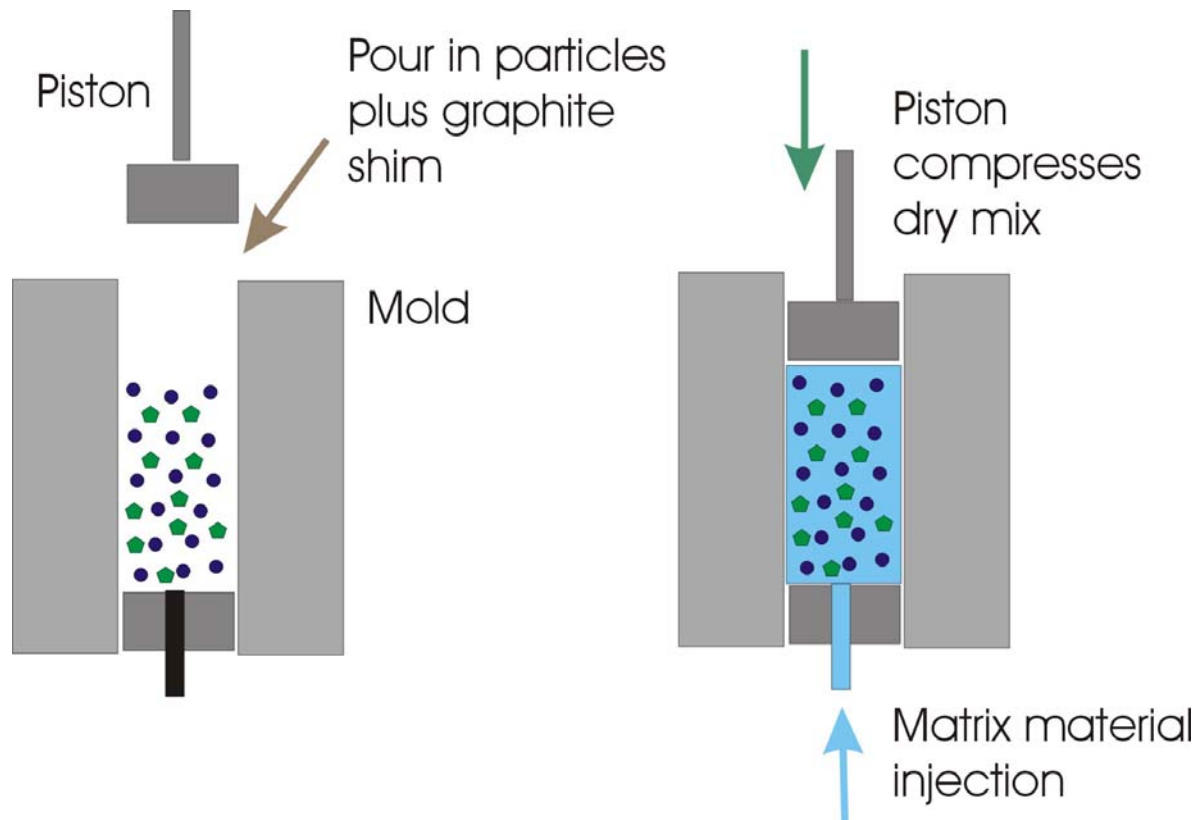


Figure 2-21 Illustration of the injection method

Both methods can damage particles by crushing and care needs to be taken to control forces and packing fraction. The current trend is toward the Admix process with thermosetting resin and the best performing fuel has used this method. It is likely that all future HTGR fuel element fabrication will be based on the Admix rather than the Injection process unless high particle packing fractions are required.

A common QC method to check for broken particles in a fuel element is the burn-leach method. A fuel element is slowly oxidized in air in a furnace at approximately 800°C to remove the matrix material and particle outer pyrocarbon layer. Flaws in the SiC larger than roughly 1 micron that expose the inner pyrocarbon will also allow the IPyC layer and the buffer layer to be oxidized during the burn [2-33, 2-38, 2-39]. The resulting burned back particles are then leached in hot nitric acid to dissolve any exposed uranium. The leachant is then analyzed for uranium and an estimate for the damaged particle fraction determined.

Another variable is the matrix mix. The resin is the glue (binder) that holds the mixture together, the graphite flour or flakes is the filler material that forms the foundation of the element, and the additives make the mixture free flowing (if necessary) and limit the adhesion of the resin to the OPyC.

Another factor of the element fabrication process is the adhesion between the matrix material and the OPyC. This was discussed previously (see Fig. 2-14). If the adhesion is too strong, the OPyC will be damaged as the matrix material shrinks during irradiation. This is less important with the Admix process because of the low percentage of binder and high filler content, but is more important for the injection process because of its high binder content and lower viscosity, which gives it better penetration ability. To limit the binder penetration into the OPyC, additives are added to the matrix material. These additives vaporize during the carbonizing process.

To summarize, the Admix process:

- 1) Uses a low percentage of binder and a high percentage of filler
- 2) Doesn't flow – can't be used for injection molding
- 3) Produces a relatively strong dense matrix
- 4) Few problems with OPyC penetration
- 5) Limits particle packing fractions to 30-40% (perhaps even up to 50% in some cases)

The Injection process:

- 1) Has a high binder content and a low filler content
- 2) Flows well – designed for injection molding
- 3) Produces a less dense, weaker matrix
- 4) Additives must be used to limit penetration into the OPyC
- 5) Supports higher packing fractions of 50-60%

The required strength of the fuel element depends on its application. A pebble, which is repeatedly dropped several meters, requires greater strength than a fuel compact that is surrounded by a fixed graphite block. Thus, element strength is a parameter that can be part of a design trade off – say, for higher packing fraction.

The next step after the green element has been made is carbonizing. The green fuel elements are baked in an inert atmosphere furnace at approximately 800°C to carbonize the binder material and vaporize and remove any process additives. Fuel elements made with thermosetting resin are carbonized free standing, as the resin will not resoften. Elements made with thermoplastic resin (pitch) are packed in beds of aluminum oxide to support them as they will “slump” (i.e. deform) as the resin softens with the temperature increase.

During the carbonizing process, care must be taken not to introduce impurities either from the furnace atmosphere or bakeout bed. Metals like iron and chrome can diffuse through the carbon layers and attack the SiC layer. To limit problems with impurities, the fuel elements can be purged with HCl after carbonizing. The HCl converts many impurities to volatile chlorides that leave the fuel element at processing temperatures. The HCl process was used by the US in a few cases. Other countries did not use this step.

Finally, the fuel elements are baked at 1650 to 1950°C to their final form. The purpose of this high temperature firing is to further the carbonization, improve the crystallinity of the

matrix binder, and to remove any residual volatile impurities. Short times, ~1 hour, at these temperatures do not appear to effect the SiC (slight grain growth). Impurity control during high firing is important because at these high temperatures impurities can quickly diffuse through the matrix and pyrocarbons to the SiC and damage it. Impurities can come from the initial matrix mix, the carbonizing bed, the firing furnace, and from handling equipment. Figure 2-22 illustrates the fuel element baking steps.

The final fuel element parameters of interest are:

- 1) Thermal conductivity and its changes with irradiation
- 2) Strength for general handling, and for pebbles, dropping toughness during normal operating conditions
- 3) Dimensional stability under irradiation and both cracking and microcracking
- 4) Corrosion behavior under air and water ingress conditions
- 5) Ability to sorb fission product metals

Some typical QC methods used for fuel element examination are detailed in Table 2-21 [2-38].

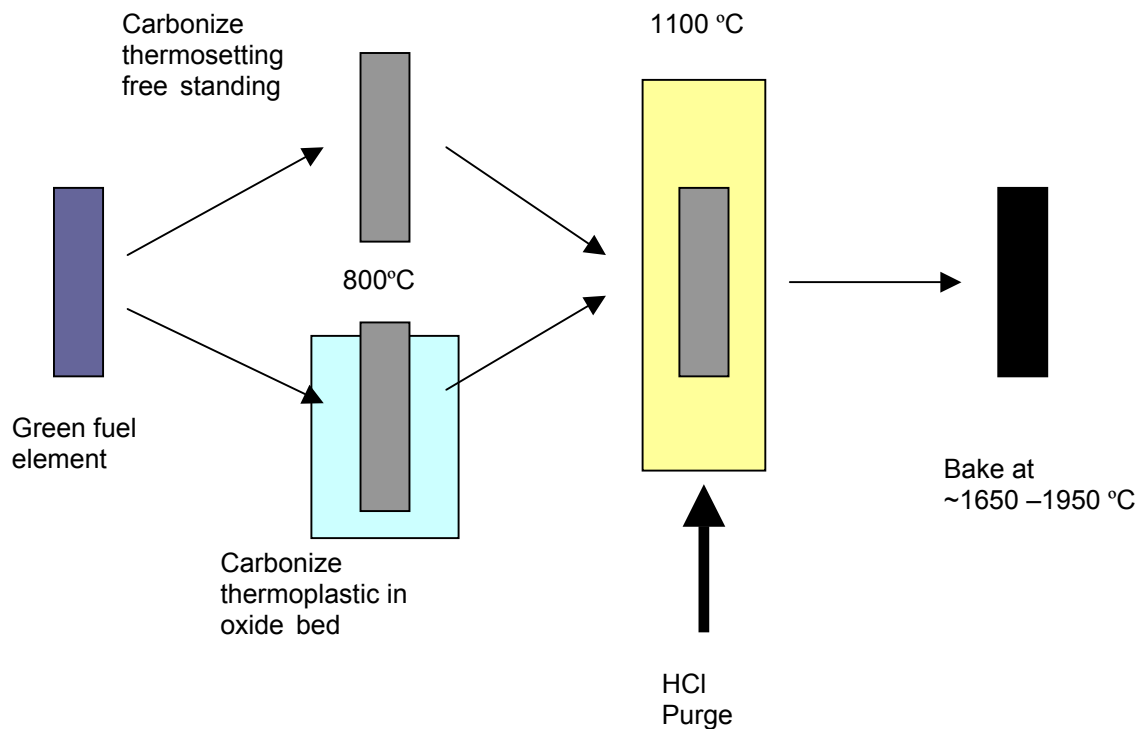


Figure 2-22 Green fuel element bakeout process

**Table 2-21 Typical Fuel Element QC Methods**

<b>Fuel Element Attribute</b>	<b>QC Method</b>
General integrity	Visual inspection
Dimensions	Gauging, contact and non-contact
U Content (total)	Wet chemistry
Particle distribution (homogeneity)	Gamma spectroscopy, radiography, metallography
Defective SiC	Burn leach
U contamination	HCl leaching
Impurities	Spectrographic methods and wet chemistry
Strength and toughness	Crush, drop
Thermal	General thermal analysis methods

**2.3.4.1 Fuel Element Manufacturing Factors Identified By The PIRT Panel**

The fuel element manufacturing factors identified by the PIRT panel are listed in Table 2-22.

**Table 2-22 Fuel Element Manufacturing Factors Identified by the PIRT Panel**

<b>Fuel Element Manufacturing Factor</b>	<b>Rationale</b>
Particle overcoating	The overcoating layer helps protect the particle during fuel element fabrication by slightly deforming, providing a spacing function, and integrates the particle into the matrix.
Matrix and Binder	The “glue” that holds the matrix together. May be thermosetting or thermoplastic.
Bonding strength between the OPyC and the matrix	If the bonding strength of the matrix to the fuel particle is too high, the OPyC may be torn away as the matrix undergoes irradiation-induced shrinkage.
Compacting (molding and pressing)	This process can result in broken or damaged particles.
Carbonization	This is the process of driving off the volatiles and converting the resin to carbon.
Heat treatment	The high temperature process that completes carbonization, improves the crystallinity of the matrix, and degasses the element.
Impurity control	Impurities can come from many sources and the metals can damage the SiC at high temperatures.
Tramp uranium	This is uranium contained in the raw materials used in the manufacturing process.
Strength	The fuel element must be strong enough for its application.
Initial particle defect fraction due to manufacture	Exposed kernels at the completion of fuel element fabrication increases fission product releases.



## 2.4 Normal Operation

### 2.4.1 General

During normal operation the fuel failure rate is required to be very low. The cause of fuel failures during operation is a function of its design, manufacture and operating environment. For example, in service failures of some past fuels were believed to come from the very small fraction of particles with thin or missing buffer layers [2-7, 2-20 to 2-24, 2-34]. Less than optimal pyrocarbon layers that crack or debond under irradiation can also contribute to failure [2-27, 2-30]. Failures due to misshaped particles are expected to be small with current QC methods. Adverse stress conditions due to statistical variations in fuel particle layer characteristics can also result in particle failures. Another potential source of failures are fuel particles that are in the “hot spots” of the core. Extended operation at temperatures near and above 1300°C can result in weakening or failure of particles due to fission product corrosion [2-35, 2-36]. Proper core design should eliminate these ‘hot spots’.

Fission product releases during normal operation come from at least four sources:

- 1) Fabrication-induced particle defects and tramp uranium. Release from defective (e.g. flawed SiC) particles, particles broken during fuel element fabrication, and uranium impurities in the fuel element fabrication materials will be present from the start of operation. All three of these sources have uranium outside of the SiC coating, the pyrocarbon, or both.
- 2) Releases from particles that fail during normal operation. For a properly designed core, one source of release may be from particles that fail due to missing or thin buffer layers (or perhaps, poor quality pyrocarbon that cracks). These particles will result in excessive internal pressure and break sometime during the core life. If the particles are fabricated correctly and operated within limits, failures due to statistical variations or coating faults such as pyrocarbon, overpressure, or SiC problems should be very small.
- 3) If there are hot spots in the core, the probability of failure from other sources such as amoeba (UO<sub>2</sub>) or fission product corrosion of the SiC could be significant for a limited number of particles.
- 4) Silver releases from particles at temperatures above 1100°C. SiC does not retain silver well above approximately 1100°C and silver is likely to be released. Silver-110m is considered to be an occupational dose issue rather than a public health and safety issue for present designs.

Released fission gases will quickly migrate to the coolant and can be removed by the coolant gas cleanup system. Metals, however, will remain on the reactor internal components and perhaps on dust. There appears to be no practical way to remove metallic releases from an operating reactor.

To meet the proposed HTGR licensing-basis offsite dose criteria, safety analysis assumptions, the initial releases would have to be equivalent to a particle failure fraction of about  $1-5 \times 10^{-5}$ . Failures during normal operation would also have to increase by no more than a factor of about 2 to 5. An illustration of selected operational failure mechanisms attributed to US fuel manufactured in the past is shown in Fig. 2-23. The

detailed analysis of acceptable fuel failure fractions and mechanisms was outside the scope of this PIRT. Other fission product transport means such as dust were deferred to future PIRT exercises.

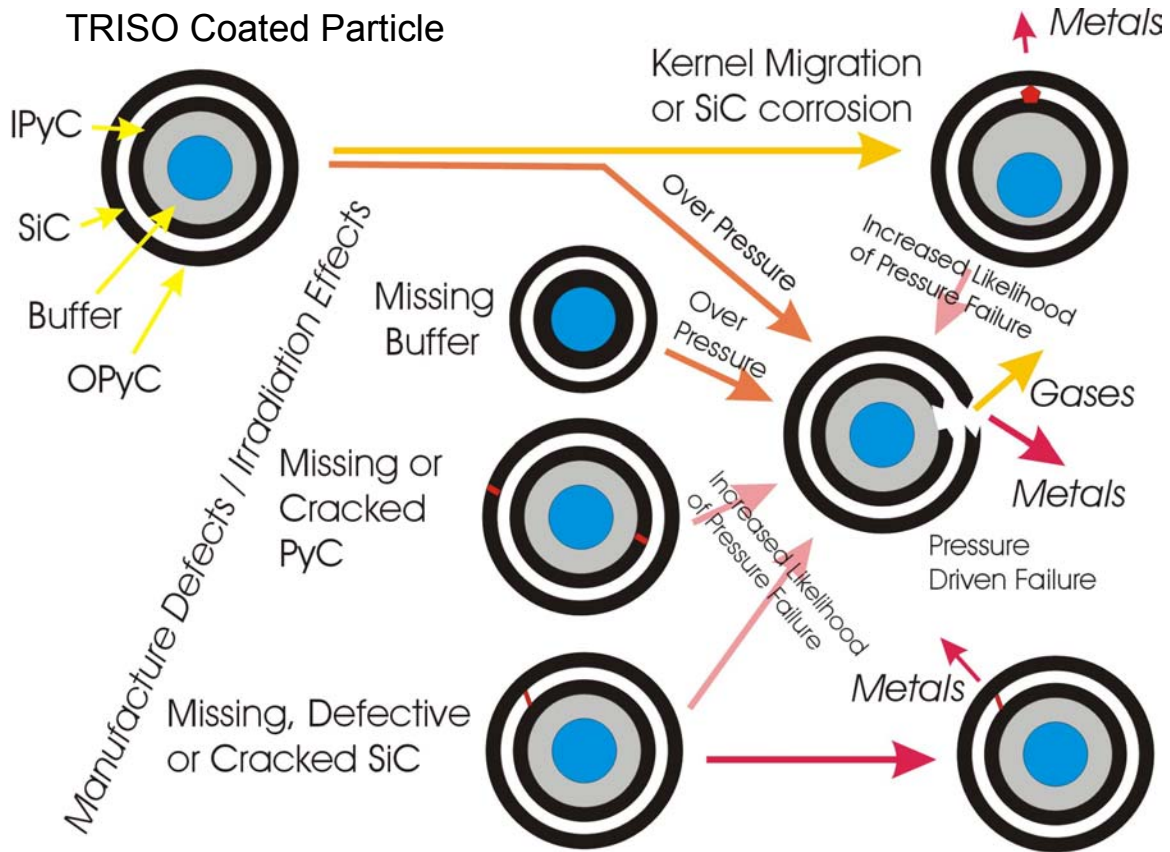


Figure 2-23 Illustration of selected particle failure mechanisms during normal operations

Figure 2-24 illustrates fractional releases (fission products outside fuel sphere divided by the total integrated fission products produced by fission) that have been calculated for pebble fuel during its life in the core of a small (170MW thermal) pebble bed reactor. Note the temperature cycling as it traverses the core. Also, note that the cesium release fraction is largely determined by the defective particles. These model calculations were done using the FRESKO-II code (German) [2-6].

Many of the particle failure mechanisms and phenomena associated with normal operation were covered in the design section. This section will not repeat these mechanisms and phenomena, but will focus on the general goals of operation.

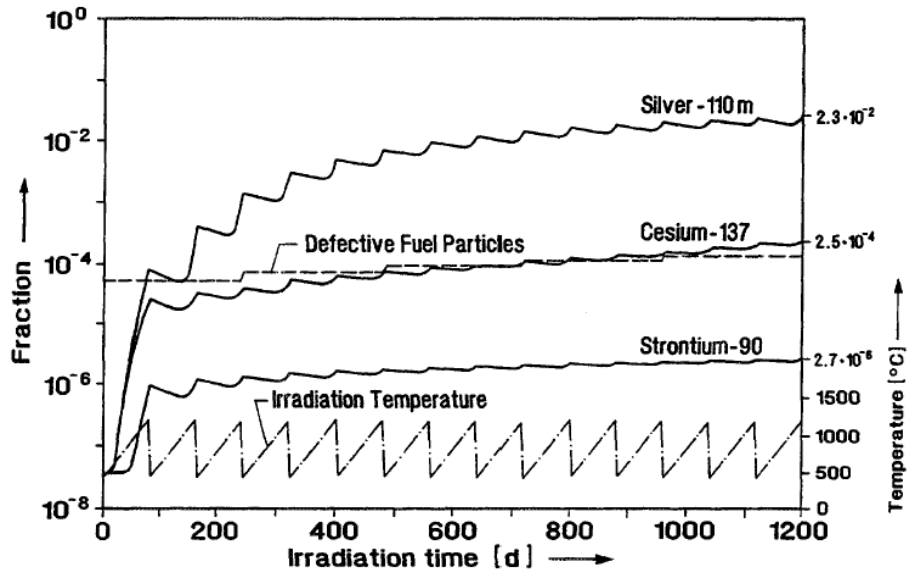


Figure 2-24 Model calculation-pebble fractional release over its lifetime (IAEA TECDOC-978)

### 2.4.2 Kernel

During normal operation, the kernel is expected to perform the functions outlined in the Section 2.2.2. Two design functional objectives are to control the oxygen potential to limit kernel migration and particle CO pressure and to retain the rare earths. Figure 2-25 shows two extremes of this situation. In the first case (left) a  $\text{UO}_2$  kernel has moved in the direction of increasing temperature (amoeba effect). The time averaged temperature was  $1180^\circ\text{C}$ , the temperature gradient was  $990^\circ\text{C}/\text{cm}$ , and the burnup was 80% FIMA. In the second case (right), rare earths from a  $\text{UC}_2$  kernel have migrated in the direction of decreasing temperature to the SiC layer. For this case, the time averaged temperature was  $1130^\circ\text{C}$ , the thermal gradient was  $990^\circ\text{C}/\text{cm}$ , and the burnup was also 80% FIMA. Both situations can cause SiC failure. In both cases the test conditions exceeded the design conditions that would be present in a power reactor environment.

While the kernel will also contain gases to some extent, the change in microstructure due to burnup may prevent high gas retention in the kernel, especially for accident temperatures. High burnup fuel contains many voids and the change in lattice structure with burnup increases the diffusion of fission products. Lower burnup fuel,  $<10\%$  FIMA, will have less kernel microstructure damage and may better retain fission gases. This is different than the LWR case where high levels of fission gas retention ( $>95\%$ ) are common. Thus, the kernel is not expected to be retentive of all fission products, but rather to focus on particular ones and assume a new role of controlling oxygen potential.

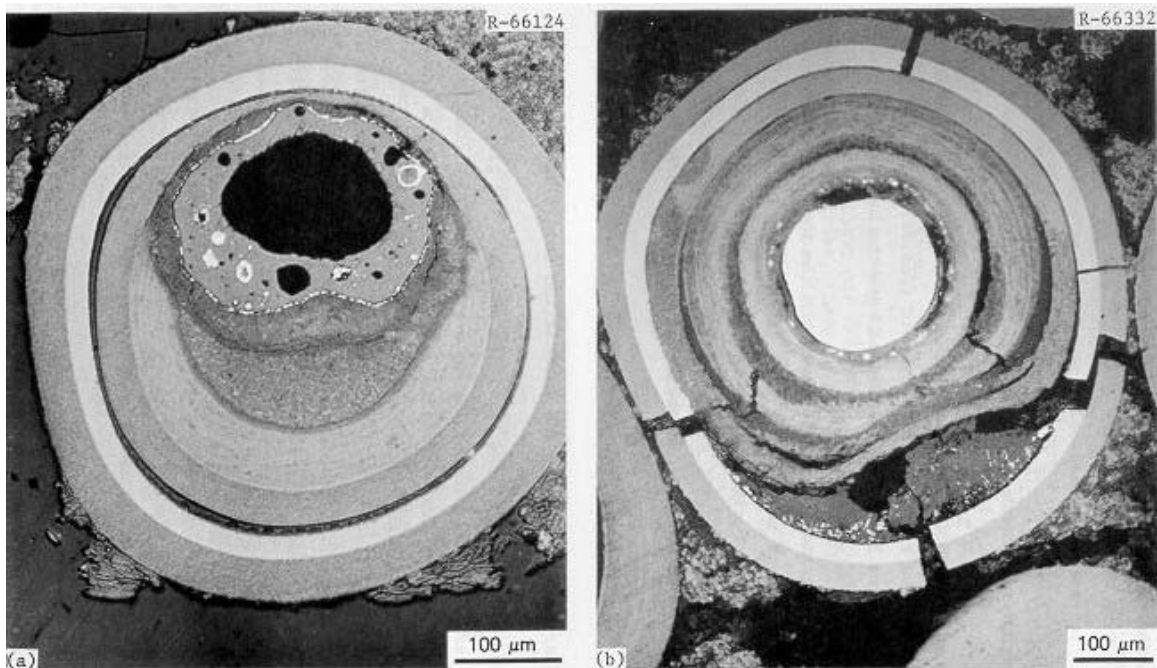


Figure 2-25 Kernel performance issues. The kernel on the left is a  $\text{UO}_2$  kernel that is moving up the temperature gradient (amoeba effect). The particle on the right is a  $\text{UC}_2$  kernel showing the accumulation of rare earth fission products on the cold side of the particle. The hot side is at the top of the photo

#### 2.4.2.1 Kernel Operating Factors Identified By The PIRT Panel

Table 2-23 lists the kernel operating factors identified by the PIRT panel. This table also includes some general items that apply.

#### 2.4.3 Buffer

The main design functional objective of the buffer is to provide a free space for the released fission gases and any generated CO. Service failures of fuel particles can be caused by missing buffers. Lacking sufficient volume to accommodate the volume of generated gas, the particle can fail by overpressure. Figure 2-26 shows a failure (in this case a BISO particle with no SiC layer) due to a missing buffer layer. The design section details other factors associated with the buffer.

An excessively thick (out of specification) buffer could increase thermal impedance. However, the temperature gradient across the particle due to its own heat generation is small ( $<100^\circ\text{C}$ ) and this extra impedance does not appear to adversely affect particle performance, at least with the current level of understanding.

However, some recent calculations indicate that large thermal gradients can drive fission product diffusion; this is an area of current study.

**Table 2-23 Kernel Operating Factors Identified By The PIRT Panel**

<b>Kernel Operations Factor</b>	<b>Rationale</b>
CO production	CO production influences particle pressure and kernel migration.
Burnup	The burnup state determines the chemical environment in the kernel and its ability to retain fission products.
Swelling	The buffer is designed to accommodate the kernel distortions.
Microstructure changes	The crystal structure of the kernel can influence the hold up of fission products
Fission product chemical form	The chemical form of the fission products determines their mobility within the kernel and affects the CO pressure.
Buffer interaction	The periphery of the kernel may react with the buffer layer carbon.
Kernel migration (fuel dependent)	Temperature gradients can drive carbon transfer and result in the movement of the kernel toward the coatings.
Fission product generation	Different fissile isotopes will give different mixes of fission products. Since the noble metals attack the SiC, isotopes that generate more noble metals may degrade performance.
Temperature gradient	The macro temperature gradient across the kernel drives the amoeba effect and rare earth migration.
Isotope half life	Determines which fission products will survive the diffusion to and through the coatings.

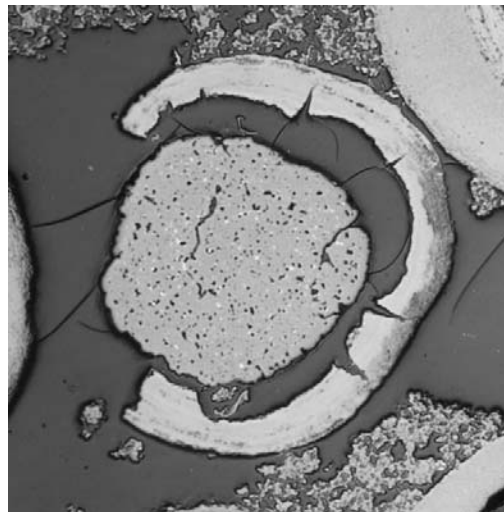


Figure 2-26 Failure in a BISO particle due to a missing Buffer layer

### 2.4.3.1 Buffer Operating Factors Identified By The PIRT Panel

The operating factors identified by the PIRT panel for the buffer layer are listed in Table 2-24. This table also carried forward the items discussed in previous sections that are relevant.

**Table 2-24 Buffer Operating Factors Identified By The PIRT Panel**

<b>Buffer Operations Factor</b>	<b>Rationale</b>
Pressure	The buffer function is to provide void volume to control particle pressure.
Shrinkage	Controlling buffer shrinkage is desirable to control cracks, and minimize kernel extrusion.
Cracking	See Shrinkage. May also allow recoil path to IPyC.
Carbonyl vapor species	No past examination of this; new issue identified by panel.
Temperature gradient	An excessive gradient can lead to higher kernel temperatures.
Condensed phase diffusion	The buffer layer is the medium connecting the kernel to the IPyC layer and first sees the fission products released from the kernel.
Gas phase diffusion	
Recoil effects	The buffer layer protects the IPyC layer by attenuating the fission product recoils.

### 2.4.4 IPyC Layer

The design functional objective of the IPyC is to remain dimensionally stable and intact during the fuel lifetime. Cracks (or debonding) in the IPyC can place high local tensile stresses on the SiC that can crack it if the bonding between layers is strong enough. Cracks can also expose the SiC to CO, which can corrode the SiC at high temperatures. The same issues that were covered in the design section are important during normal operation. In particular, the shrinkage of the IPyC is controlled and accommodated to some extent by IPyC creep to keep the stresses in IPyC the layer within acceptable limits.

The IPyC also limits transport of fission products to the SiC; it is highly impermeable to fission gases, but will allow the diffusion of metallic fission products.

#### 2.4.4.1 IPyC Operating Factors Identified by the PIRT Panel

Table 2-25 lists the operating factors identified by the PIRT panel for the IPyC layer. It also includes the relevant factors identified in the previous sections.

### 2.4.5 SiC Layer

The SiC layer is the principle barrier to the release of fission products from the fuel particle. During normal operation, the SiC diffusion coefficients and temperatures are low enough that releases to the coolant are expected to be dominated by the initial defective fuel fraction

**Table 2-25 IPyC Operating Factors Identified By The PIRT Panel**

<b>IPyC Operations Factor</b>	<b>Rationale</b>
Radiation induced creep	IPyC creep relieves some of the stress in the IPyC caused by irradiation-induced shrinkage and pressure.
Fast fluence	The fluence dose has an impact on the shrinkage, creep, and layer stresses.
Dimensional change	The stresses caused by irradiation can result distortion and debonding of the IPyC from the SiC.
Anisotropy	The anisotropy is considered to be any important predictor of the IPyC dimensional stability.
Cracking	Cracking can result from the shrinkage and debonding from the SiC.
Debonding	Shrinkage and dimensional instability can result in the IPyC pulling away from the SiC and increasing peak local tensile stresses on the SiC layer.
Condensed phase diffusion	The IPyC layer is the first reasonably retentive layer.
Gas phase diffusion	The IPyC layer is the first reasonably retentive layer. The layer retains fission gases well.

and failures caused by other phenomena. The exception is silver. Extended fuel operation above about 1100°C will allow diffusion of silver through the SiC and into the coolant. The implications of this diffusion depend on the reactor design.

Extended high temperature operation of the fuel, above approximately 1300°C, can lead to corrosion of the SiC by noble metals and CO, especially if the IPyC is cracked. The noble metal of greatest concern is palladium as it easily migrates (diffuses) to the SiC and attacks it.

Figure 2-27 shows migration of Pd to the SiC layer for a plutonium kernel. The kernel type can be important as different fissile materials can have much higher yields of noble metals.

If the kernel does not retain the rare earth elements as oxides, they too can attack the SiC as was noted in the section dealing with the kernel.

If heavy metal contamination of the SiC layer occurs during fabrication, fissioning of this material will damage the SiC and lead to its early failure. If the PyC layers are intact, A SiC layer failure will not be detected by an increase in the circulating fission gas inventory. Fission product metals will be released, however. The same behavior will occur if the SiC is cracked, perhaps by handling (again, good PyC).

In general, an intact PyC layer or SiC layer will retain the fission gases making individual layer failure difficult to detect during normal operation. During normal operation, migration of the metals is limited and collecting and analyzing them is difficult in the absence of a special collection device.

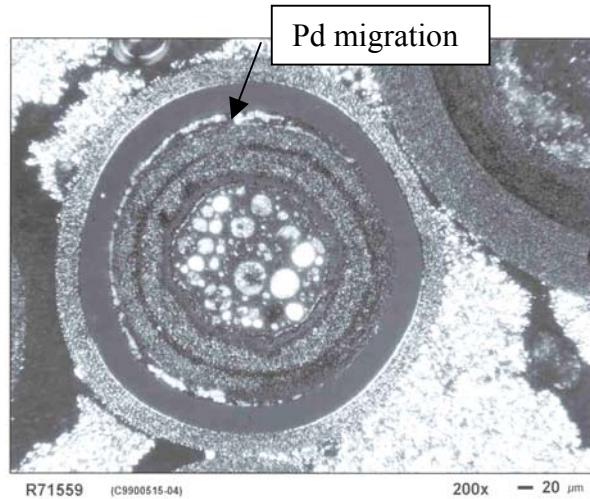


Figure 2-27 Pd migration: 70% burnup Pu kernel. FTE-13, PuO<sub>1.68</sub>, 1150°C, polarized light

#### 2.4.5.1 SiC Operating Factors Identified By The PIRT Panel

Table 2-26 lists the SiC layer operating factors identified by the PIRT panel.

**Table 2-26 SiC Operating Factors Identified By The PIRT Panel**

SiC Operations Factor	Rationale
Kernel interaction with SiC layer	See the previous comments on kernel migration.
Fission product corrosion	Noble metals present a corrosion hazard at the higher temperatures. CO can attack the SiC with a cracked IPyC at high temperatures.
Heavy metal attack	If fissile material is present in the SiC from fabrication, fissions in the SiC will damage it.
Cracking	Cracking during normal operation will cause the particle to release metallics, but not gases if one of the PyCs remains intact.
Condensed phase diffusion Gas phase diffusion	SiC is the primary diffusion barrier and it retains both metallic and gaseous fission products well, with the exception of silver.

#### 2.4.6 OPyC Layer

The design functional objective of the OPyC is to remain dimensionally stable and intact over the fuel lifetime. It is the interface between the SiC and the fuel element matrix. Failure of the OPyC could increase the failure probability of the SiC. The same phenomena that were covered in the design section are important during normal operation. In particular, the shrinkage of the OPyC is controlled and accommodated to some extent by OPyC creep to keep the stresses in the layer within acceptable limits.



The OPyC limits the transport of fission gases, but will allow the migration of metals. One also wants to control the OPyC-matrix interactions so that the OPyC is not damaged by matrix shrinkage or micro cracks that occur in the matrix material.

#### 2.4.6.1 OPyC Operating Factors Identified by The PIRT Panel

Table 2-27 lists the OPyC layer operating factors identified by the PIRT panel.

**Table 2-27 OPyC Operating Factors Identified by The PIRT Panel**

<b>OPyC Operations Factor</b>	<b>Rationale</b>
Radiation induced creep	This creep relieves some of the stress caused by shrinkage and pressure.
Dimensional change	The stresses caused by irradiation can result distortion and perhaps breakage of the OPyC layer.
Anisotropy	The anisotropy is considered to be an important predictor of the OPyC dimensional stability.
Condensed phase diffusion	The OPyC is not strongly retentive of metallics.
Gas phase diffusion	The OPyC layer functions as a barrier in the event of SiC failure.
Cracking	The OPyC layer functions as a barrier in the event of SiC failure.

#### 2.4.7 Fuel Element

The major design functional objective of the fuel element during normal operation is to

- 1) Maintain dimensional stability
- 2) Not transmit undue stresses to the fuel particles
- 3) Withstand being dropped for pebble fuel
- 4) Maintain good thermal properties
- 5) Act as a sink for any released metallic fission products

Stability is a function of the fabrication of the fuel element; elements made with a high filler content tend to be more stable than those with low filler content. Shrinkage of the matrix material may result in microcracks and voids in the fuel element. As long as particles are not damaged and the thermal properties are within design limits, minor cracking is tolerable.

The tolerance for large cracks depends on the application. For fuel compacts inserted into a prismatic graphite block, a through crack may not be desirable, but it may not seriously affect the performance of the compact. However, the situation is different for a pebble that must be capable of being dropped from a height of several meters and traversing the reactor core without breakage. A through crack would increase the probability of pebble breakage and the fragments would have to be removed.

Corrosion by coolant impurities can affect fuel elements and result in small regions (<0.2mm deep) of “peeling” or “spalling” on the surface of pebbles. Corrosion resistance

to coolant impurities can be greatly increased by high temperature baking of the pebbles at 1950°C during manufacture [2-9].

It should be noted that the fuel element matrix is not solid graphite and the irradiation performance is not the same as graphite and will vary with the actual composition of the matrix. Different sources and mixes of binder, graphite, shim materials, and processing conditions can lead to different results, both in initial properties and during irradiation.

As the matrix material accumulates irradiation damage, its mechanical and thermophysical properties are affected [2-9]. For the reference fuel pebbles, matrix irradiation performance was very good for the operating temperatures and fast fluences of interest (<1400°C and <9 x 10<sup>25</sup> n/m<sup>2</sup>). Under irradiation the pebbles shrank less than 2% in diameter. Shrinkage was about the same in the radial and tangential directions up to a fast fluence of 5 x 10<sup>25</sup> n/m<sup>2</sup> and then deviated by about 30% as the fast fluence increased. Strength increased about 10% at moderate fast fluences and the elastic modulus increased with irradiation, up to 70% (700°C), and then only slightly decreased at the higher fast fluences (the increase was much less and the decrease greater at higher temperatures). Crush strength almost doubled.

Thermal conductivity decreased about 60% and the coefficient of thermal expansion increased by about 40% with irradiation. Both of these properties decreased initially and then remained fairly constant with increasing fluence. Irradiation temperature and accumulated fast fluence strongly influences property changes, but the pebbles maintained integrity under the irradiation conditions of interest.

The extent to which of the fuel element matrix retains metallic fission products depends on many factors, but in practice the matrix material appears to either sorb metallic fission products or delay their migration into the coolant at normal operating temperatures. Gases readily migrate through the matrix material.

#### 2.4.7.1 Fuel Element Operating Factors Identified By The PIRT Panel

Table 2-28 lists the Fuel Element Operating Factors Identified by The PIRT Panel.

**Table 2-28 Fuel Element Operating Factors Identified By The PIRT Panel**

<b>Fuel Element Operations Factor</b>	<b>Rationale</b>
Temperature	Matrix behavior and stability is a function of temperature/fast fluence.
Fast fluence	Matrix behavior is related to the fast fluence. In particular shrinkage and cracks.
Power density	Power affects the operating temperature and gradients.
Temperature difference	The fuel particle behavior can be influenced by the macro temperature gradient.
Temperature time histories	Important for determining corrosion and diffusion.
Condensed phase diffusion	The matrix material can sorb/trap significant amounts of the less volatile fission products.
Gas phase diffusion	The element doesn't provide significant retention of gases.
Corrosion by coolant impurities	PPM levels of coolant impurities can corrode or damage the fuel element matrix material

## 2.5 Accident Conditions

For the purposes of identifying phenomena that are considered important to the performance of coated particle fuel, four classes of accident conditions were considered by the PIRT panel:

- 1) *Core heatup*: Loss of helium pressure and flow, core heatup and subsequent cooldown, with or without a SCRAM. This accident results in the reactor achieving a peak fuel temperature of roughly 1600°C; however, 1800°C was examined to envelope phenomena of interest. The core temperature rises from normal operating temperatures to a maximum over a period of 20 to 30 hours and slowly cools down over a period of several hundred hours in an assumed helium environment. Loss of coolant pressure is more challenging than simply a loss of flow, because natural convection can reduce the peak fuel temperature to some extent in the normal pressure case.
- 2) *Water ingress*: Failure of a heat exchanger tube with reactor depressurization. Since the reactor designs currently under study do not have steam generators and the water coolant in the heat exchangers is lower than core pressure, significant water entry into the core is not expected; however, 1% water vapor was assumed to be present in the core during the postulated accident (balance helium). The failure of the pressure boundary and depressuring the core with a heat exchanger failure would allow water entry under low driving pressure.
- 3) *Air ingress*: Complete severing of the horizontal vessel between the reactor vessel and the power conversion system vessel followed by depressurization and air diffusion into the core. Both high temperatures and chemical attack are considered. The PIRT panel considered oxidation of the core structure, but not structural collapse or major relocation of fissile material.
- 4) *Reactivity insertion*: A sudden reactivity addition could result in locally high fuel temperatures and/or fuel damage. Pulse lengths are on the order of seconds and a fraction of the fuel is assumed to fail by cracking of the PyC and SiC, but the kernel and buffer remain intact. A postulated rod ejection accident was considered to be the basis for the accident conditions. The on-line refueling of the pebble bed reactor results in small excess reactivity and the use of burnable poisons in the prismatic reactor also limits excess reactivity.

The PIRT panel considered the aforementioned postulated events as a basis for establishing an “envelope of accident conditions” as fuel environmental conditions from which to identify important fuel phenomena. The conditions were intended to be a “realistically conservative” upper bound for each postulated class of accidents that might be included in the licensing basis.

Unlike an LWR, an HTGR does not have core internals that would melt and rapidly lose core configuration under accident conditions, although an earthquake could cause some damage. In addition, the chemical reactions that result in the degradation of core internals proceed at a much slower rate. Thus, the dynamics of these types of accidents are much slower than LWR accidents. Reactivity insertion accidents are expected to be less severe in an HTGR, both in intensity and because the large thermal inertia of the

graphite provides a heat sink. However, this accident needs more study for the particular design of interest.

### 2.5.1 Heatup Accident

The most significant condition associated with this accident is the elevated temperature over time. The environment remains essentially inert (helium). Thermal power is at a low level, either from afterheat or from the low nuclear power level achieved by the negative reactivity with the failure to SCRAM. Heat is transferred from the core through the reactor vessel to the cavity cooling equipment and structures.

Figure 2-28 shows the calculated temperature versus time curve for a depressurized heatup accident, the temperatures used for experimental fuel testing (horizontal lines), and a conservative heatup accident curve for a small modular type reactor.

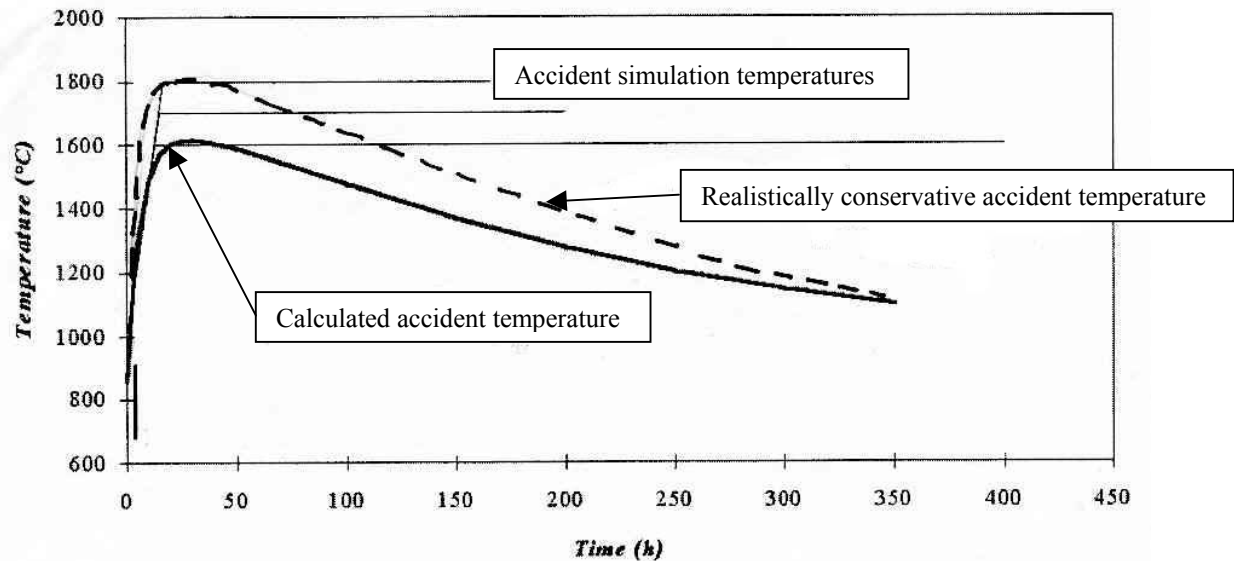


Figure 2-28 Time versus temperature heatup accident curve considered by the PIRT

In general, fuel element releases tend to increase with time at temperature, burnup, and temperatures above  $\sim 1600^{\circ}\text{C}$  [2-6, 2-7, 2-20 to 2-24]. Figure 2-29 shows  $^{85}\text{Kr}$  releases for German program test compacts illustrating the burnup and temperature observation for about a 300-hour heating time [2-6].

#### 2.5.1.1 Kernel

High temperatures during a Heatup accident will increase the diffusion of fission products from the kernel. The SiC is the primary barrier to release, but diffusion through the kernel can have a minor effect. Of special interest is the more rapid diffusion of fission products (mostly noble

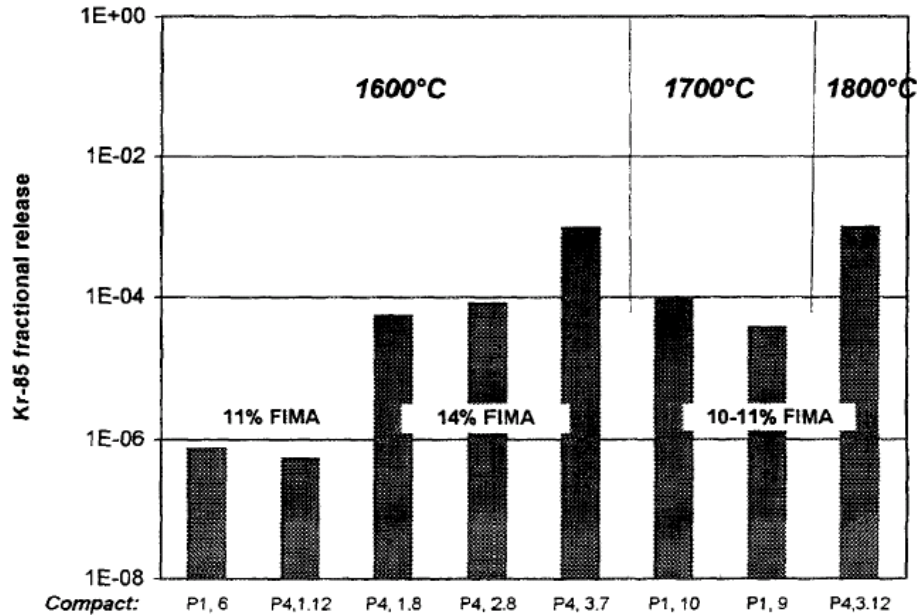


Figure 2-29  $^{85}\text{Kr}$  releases as a function of burnup and test temperature. (IAEA TECDOC-978)

metals) that are known to attack the SiC layer. If the SiC is already weak due to corrosion during normal operation, the additional attack due to newly diffusing material, higher gas pressures, and the higher temperature may fail the SiC layer if the time at temperature is long enough. Cracks in the IPyC may allow CO to attack the SiC as well as increasing the stress loading of the layers. These effects are considered a function of burnup.

The temperatures in the fuel are expected to be below the melting point of the kernel material. The vapor pressure of the volatile species could increase however. As the temperature increases, the oxygen potential of the kernel may change and this could shift the equilibrium somewhat, but the fuel design should anticipate this situation [2-8, 2-43].

Temperature gradients in the fuel are expected to decrease under accident conditions compared to normal operation.

#### 2.5.1.1.1 Kernel Heatup Accident Factors Identified By The PIRT Panel

Table 2-29 lists the phenomena identified by the PIRT panel for the kernel under heatup accident conditions.

**Table 2-29 Kernel Heatup Accident Factors Identified By The PIRT Panel**

<b>Kernel Heatup Accident Factor</b>	<b>Rationale</b>
Maximum fuel temperature	Temperature affects the rate at which material diffuses out of the kernel.
Temperature vs time transient conditions	The time-dependent variation of the fuel with time will determine diffusive release.
Energy Transport: conduction within kernel	The gradient within a kernel is due to the transport of decay heat
Thermodynamic state of fission products	The chemical state of the fission products determines how they migrate and vapor pressures. A goal is to tie the rare earths and limit CO production (same as normal operation).
Condensed phase diffusion	Higher temperature increases the diffusion rate.
Gas phase diffusion	Higher temperature increases the diffusion rate.
Oxygen flux	Diffusion would increase, but not known in detail.
Grain growth	Grain growth may increase fission product transport, but it has not been explored.
Buffer carbon-kernel interaction	Some interaction between the kernel and the buffer carbon may be possible.

### **2.5.1.2 Buffer Layer**

The design performance objective of the buffer layer during a heatup accident is to control the gas pressure by providing a void volume as it is for normal conditions. It provides little retention of fission products. Some distortion of the buffer is expected during irradiation and is not expected to affect accident performance.

#### **2.5.1.2.1 Buffer Layer Heatup Accident Factors Identified By The PIRT Panel**

Table 2-30 lists the buffer layer heatup accident factors identified by the PIRT panel under heatup accident conditions. Many of these issues are the same as noted for normal operation.

### **2.5.1.3 IPyC Layer**

Depending on the bonding between the IPyC and the SiC, the IPyC can help keep the SiC in compression; breaking or cracking of the IPyC can result in higher localized stresses in the SiC layer. A crack can also expose the SiC to CO that can cause corrosion at the higher accident temperatures. Complete debonding of the IPyC may cause a shift in the pressure loading; a structural model is necessary to determine the effect. This is an area of active research.

**Table 2-30 Buffer Layer Heatup Accident Factors Identified By The PIRT Panel**

<b>Buffer Heatup Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	The fission product transport through the buffer layer is expected to be high at accident temperatures.
Condensed phase diffusion	
Response to kernel swelling	The buffer layer is designed to be weak enough that it will deform to accommodate the kernel.
Maximum fuel gaseous fission product uptake	The buffer layer is designed to have sufficient free volume to accommodate the fission products at an acceptable pressure.
Layer oxidation	Any oxygen released from the kernel will oxidize a small portion of the buffer. This may be of little consequence for the buffer, but may increase the particle pressure.
Thermal gradient	During heatup conditions, the gradient across the buffer is much less than normal operation due to the much lower heat generation rate.
Irradiation and thermal shrinkage	The buffer is designed to isolate the kernel from the IPyC, but small cracks could locally concentrate fission products on the IPyC.

Decomposition or chemical reactions of the IPyC layer are not an important phenomena for a heatup accident; dimensional stability is the important concern as it is during normal operation. Metallic fission products will diffuse through the layer at a greater rate, but the gaseous diffusion should still be small for an intact layer.

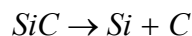
#### **2.5.1.3.1 IPyC Layer Heatup Accident Factors Identified By The PIRT Panel**

Table 2-31 lists the IPyC layer heatup accident factors identified by the PIRT panel. The relevant general issues identified before are also included.

#### **2.5.1.4 SiC Layer**

An intact SiC layer is the primary barrier to the release of metallic fission products at accident temperatures. It also effectively contains gaseous fission products. The primary challenges to layer integrity are pressure, corrosion, and decomposition. The pressure can be controlled by the design of the particle, corrosion by the tie up of most fission products and limiting the operating temperature to control Pd attack (and CO attack if the IPyC cracks), and decomposition by limiting the maximum accident temperature.

Decomposition results in the disassociation of the SiC into silicon and carbon at temperatures above about 1600-1800°C [2-6, 2-20 to 2-24, 2-34]:



**Table 2-31 IPyC Layer Heatup Accident Factors Identified By The PIRT Panel**

<b>IPyC Heatup Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	Gaseous fission products are generally retained well by the IPyC layer even at higher temperatures.
Condensed phase diffusion	Metallic fission products are not retained well.
Pressure loading (fission products)	The IPyC can help keep the SiC in tension and prevent it from failing if the bonding between layers is strong (same as normal operation).
Pressure loading (CO)	The same CO issues apply here. High CO production will result in high pressures (same as normal operation).
Layer oxidation	In some cases internal oxidation of the layer could be significant. IPyC cracks can allow CO to the SiC and corrosion may result.
Stress state (compression/tension)	If this layer breaks or debonds, the changes in stress distribution of the particle could cause it to fail.
Cracking	Cracks could change the stress distribution; same as normal operation.
Intercalation	The IPyC can accumulate fission products during normal operation that could be released during a heatup accident.
Trapping	

At about 2000°C, thermal decomposition of SiC is a dominate failure mechanism. However, above about 1600°C decomposition affects in the SiC such as the development of porosity are noted, implying that thermal decomposition mechanisms are active. Above the assumed particle temperature limit, ~1600°C, diffusion of fission products begins to increase (over normal operating values) and limits the time at temperature [2-6, 2-21 to 2-24].

Figure 2-30 shows the relative effective SiC thinning rates due to corrosion and decomposition predicted for past US fuel at 16% FIMA, a fast flux of  $4.0 \times 10^{15}$  n/m<sup>2</sup>/s, and an irradiation temperature of 1000°C. Decomposition is considered a significant factor above 1600°C. Corrosion can take place during normal operation and weaken the SiC layer, which then might fail under the greater pressure at accident temperatures.

SiC is stable to oxidation over the temperature region of interest. Figure 2-31 shows an oxidation diagram for converting selected carbides to oxides.



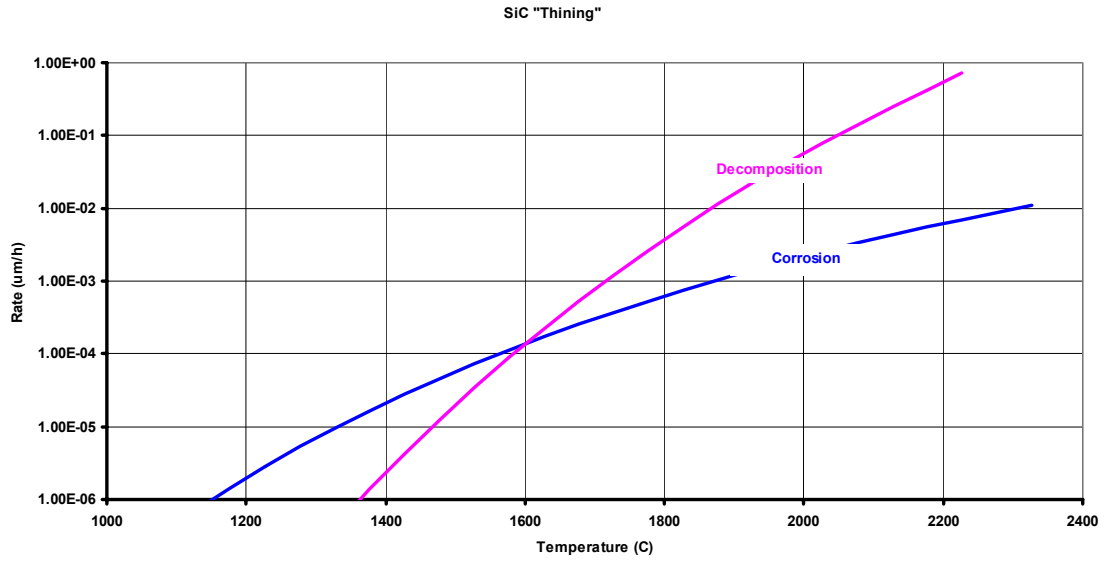


Figure 2-30 Effective SiC thinning due to corrosion and decomposition. Comparison of frequency factors for failure (data from Goodin, 1989)

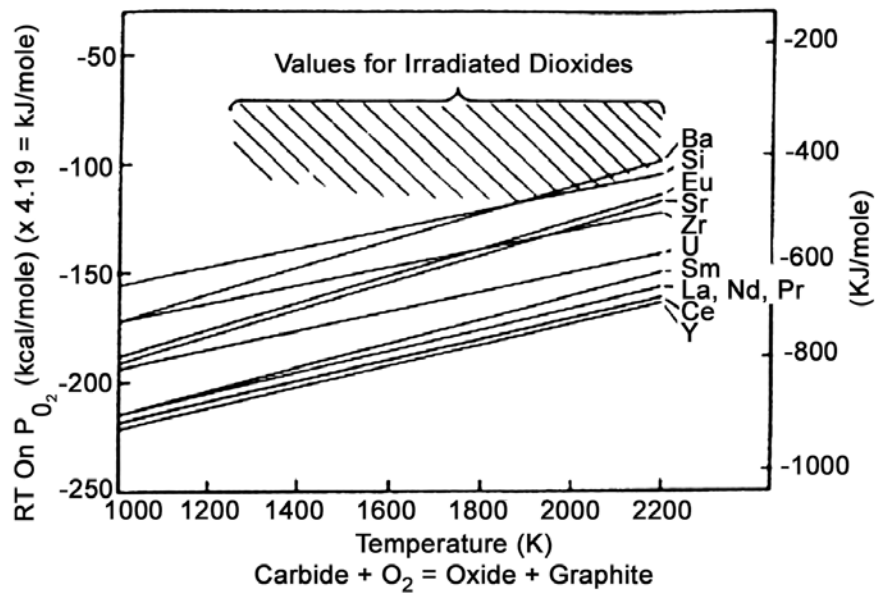


Figure 2-31 Oxidation diagram for carbides of interest

As the temperatures increase and the time at temperature becomes significant, diffusion of silver and radiological important fission products through the SiC can occur. Figure 2-32 shows the fractional releases from a German pebble at 1600°C. Note that silver is not effectively retained by the SiC layer and a significant portion of the relatively small amounts of the other released fission products are sorbed on the matrix graphite. In particular, <sup>90</sup>Sr levels approaching 1% (after 500h at 1600°C) are seen in the matrix, but the fractional release from the sphere is only  $\sim 10^{-7}$  [2-6].

Accident releases tend to increase with peak accident temperature. Figure 2-33 shows the fractional <sup>85</sup>Kr releases as a function of temperature. The general rule is that larger releases are seen above 1600°C.

#### 2.5.1.4.1 SiC Layer Heatup Accident Factors Identified By The PIRT Panel

Table 2-32 lists the SiC layer heatup accident factors identified by the PIRT panel. Included are the relevant general items discussed in previous sections.

**Table 2-32 SiC Layer Heatup Accident Factors Identified By The PIRT Panel**

<b>SiC Heatup Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	Except for silver, fission product diffusion is low through the SiC even at accident temperatures.
Condensed phase diffusion	
Thermal deterioration or decomposition	Above about 1600-1800°C the SiC begins to decompose and its ability to retain fission products is greatly reduced.
Fission product corrosion	Some fission products may migrate to the SiC and corrode it. This corrosion process is a function of temperature and can begin during normal operation. Pd is believed to be the main concern, but others are possible as well.
Heavy metal diffusion	The diffusion of heavy metal through SiC could relocate fissile material (not noted to date).
Layer oxidation	Local attack of the SiC layer by CO due to a cracked IPyC at high temperatures may be possible.
Fission gas release through undetected defects	In order to release gases, both PyCs must be cracked in addition to a failed SiC. The SiC can be attacked by fission products and fabrication impurities (see manufacturing).
Fission product release through failures (e.g. cracking)	
Thermodynamics of the SiC fission product system	At the higher temperatures of interest, SiC is just stable to oxidation in its intact particle environment. See corrosion issues.
Sintering	Accident temperatures could change the crystal or grain structure of the SiC layer (not noted to date).

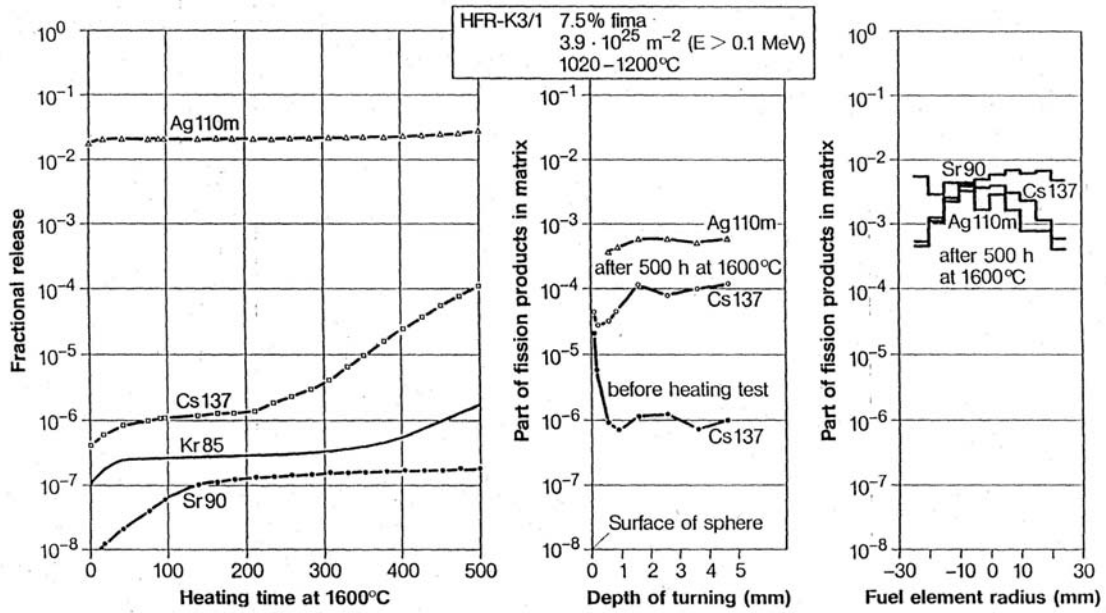


Figure 2-32 Fission product release from a fuel sphere at 1600°C. (IAEA TECDOC-978)

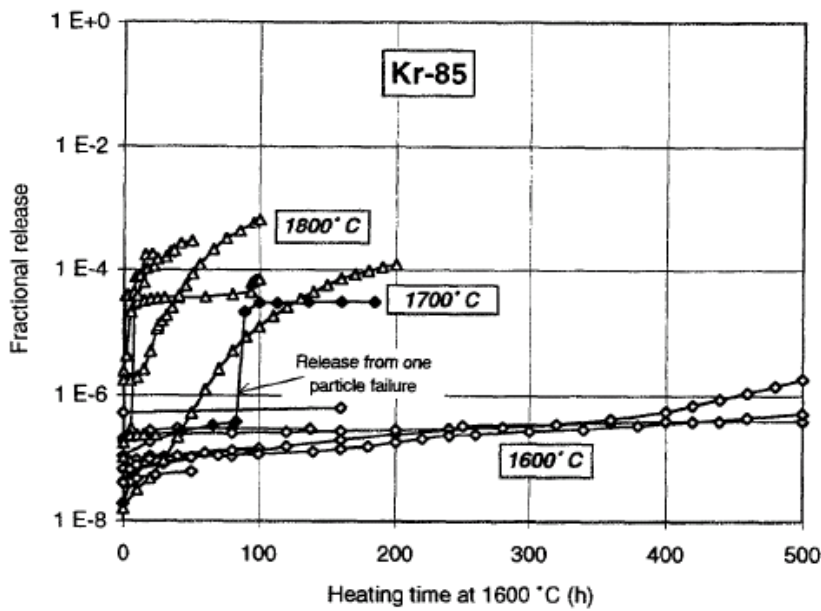


Figure 2-33  $^{85}\text{Kr}$  release as a function of heating temperature. (IAEA TECDOC-978)

### 2.5.1.5 OPyC Layer

The OPyC can help keep the SiC layer in compression; breaking or cracking of the OPyC can result in higher local stresses in the SiC and increase its likelihood of failure. It also protects (delays) the SiC layer from chemical attack

Decomposition or chemical reactions of the OPyC layer are not an issue for a heatup accident; dimensional stability is still the important concern. Metallics will diffuse through the layer at a greater rate, but the gaseous diffusion should still be small for an intact layer. It is possible that fission products trapped within the layer (intercalation) could be released at the higher temperatures. No new issues with matrix interactions are expected.

#### 2.5.1.5.1 OPyC Heatup Accident Factors Identified By The PIRT Panel

Table 2-33 lists the OPyC heatup accident factors identified by the PIRT panel.

**Table 2-33 Identified OPyC Heatup Accident Factors Identified By The PIRT Panel**

<b>OPyC Heatup Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	Gaseous diffusion is low, while metallic diffusion is high.
Condensed phase diffusion	
Layer oxidation	Cracked layers may allow CO to the OPyC or coolant impurities may attack it.
Stress state (compression/tension)	See general discussion on the stress of the particle. OPyC loss is generally not as bad as IPyC loss.
Intercalation	Because the fission product inventory is low in this layer, these two items may be important.
Trapping	
Cracking	Cracks can lead to a higher probability of SiC failure. Gases will be released if the other layers have failed.

### 2.5.1.6 Fuel Element

During a heatup accident, the fuel element performs three main functions that are essentially the same required during normal operation. The first is dimensional stability; by remaining stable and intact no undue mechanical stresses will be placed on the fuel particles. Modest element cracking can often be tolerated as long as the particles are not affected. The second function is heat transfer. Adequate thermal conductivity is necessary to couple the particles to the core and coolant. Finally, the third function is to sorb released metallic fission products. The matrix material of some fuel element types may be capable of retaining a significant portion of the released metallic fission products. Gases generally are not retained. Figure 2-32 shows that some of the less volatile fission products are retained in the element matrix material even if they are released from the

particles [2-6]. Some of these fission products migrate from the fuel element to the coolant and other reactor components [2-7, 2-20 to 2-24]

### 2.5.1.6.1 Fuel Element Heatup Accident Factors Identified By The PIRT Panel

Table 2-34 lists the Fuel Element heatup accident factors identified by the PIRT panel.

**Table 2-34 Fuel Element Heatup Accident Factors Identified By The PIRT Panel**

Fuel Element Heatup Accident Factor	Rationale
Irradiation history	Matrix behavior (shrinkage) is related to time temperature history, particularly temperatures and fast fluence.
Condensed phase diffusion Gas phase diffusion	In the matrix gas diffusion is high, while metallic diffusion is variable. The less volatile metals can be sorbed in the matrix material to a high degree.
Transport of metallic FPs through fuel element – chemical form	The chemical form of the fission product determines its transport behavior. The chemical environment outside the fuel kernel may be different than inside. Generally, significant sorbing of the metallics is seen.

### 2.5.2 Air and/or Water Intrusion Accident

Unlike the heatup accident detailed in the previous section, an accident with subsequent air and/or water intrusion can result in the additional effect of chemical attack of the fuel elements [2-6, 2-20 2-24, 2-42 to 2-44]. The temperature of the fuel depends on whether or not the reactor remains pressurized, if the control and shutdown rods insert, if significant heat is added by chemical reactions, or if heat transfer conditions change significantly during the accident.

A major difference between chemical intrusion accidents and a heatup accident is that the fuel element matrix material and the particle coating layers can be subject to a chemical reaction and transport of fission products can be by a means other than diffusion. Also, unlike the simple heatup case, the structural integrity of the fuel element may be degraded and particle coatings may even be damaged or fail. Some core designs involve changes in core geometry if the chemical reactions were sufficiently extensive. However, for purposes of the PIRT assessment, core support structural damage was assumed to be limited and slow.

The generally low chemical reactivity of the nuclear grade graphite fuel blocks in a prismatic core reactor can protect the fuel compacts and coated fuel particles to some extent. In a pebble bed reactor core, the fuel zone is surrounded by a relatively thin layer of matrix material that is directly exposed to the coolant. The matrix material is more

reactive than nuclear grade graphite so fuel element oxidation in air or water attack could be more rapid.

Current reactor designs are based on a direct cycle gas turbine design that does not have a source of high pressure, high volume water such as a steam generator, so extensive water ingress due to equipment failure is not possible as it was with the past designs. However, design specifics will need to be examined.

Exposed kernels are most affected by water ingress. Intact particles are much less affected even for extended periods of exposure to water [2-6, 2-43].

For air ingress, controlling or limiting the oxidation rate and exposure duration is a primary objective. This can be achieved by delaying the entry of air into the core until the core temperatures have been reduced and by limiting the air flow rate through the core. The loss of protective matrix material can expose fuel particles and release sorbed fission products into the coolant. Chemical attack of the fuel particles could follow. As long as the fuel temperatures and flow rate of reactants are limited, fuel particle damage can be avoided. The establishment of an air flow path that would allow rapid core matrix material loss to occur at temperatures above 1600°C could result in significant particle damage rates.

This flow path can principally be established in two ways. The first is the development of a "chimney" by having the reactor pressure boundary fail in both the top and bottom of the vessel. Natural convection via buoyancy forces then drives a flow. The second way involves a breach in a single location. In this case, gaseous diffusion allows the entry of air from outside the break into the core volume and the establishment of convective flow on a longer time scale [2-45].

Contemporary designs limit the water available by using heat exchanger water side pressures below core pressure and limit the natural convection with high core flow impedance. However, these are design specific issues and need to be assessed on a case-by-case basis. Figure 2-34 illustrates how chemical attack of the fuel might occur. Air attack of the carbon materials is the most aggressive [2-6].

Figure 2-35 shows the results of a fuel sphere exposed to water vapor at a temperature of 800°C. The water vapor reacts with exposed kernels (fabrication defects) and releases fission products [2-6, 2-43].

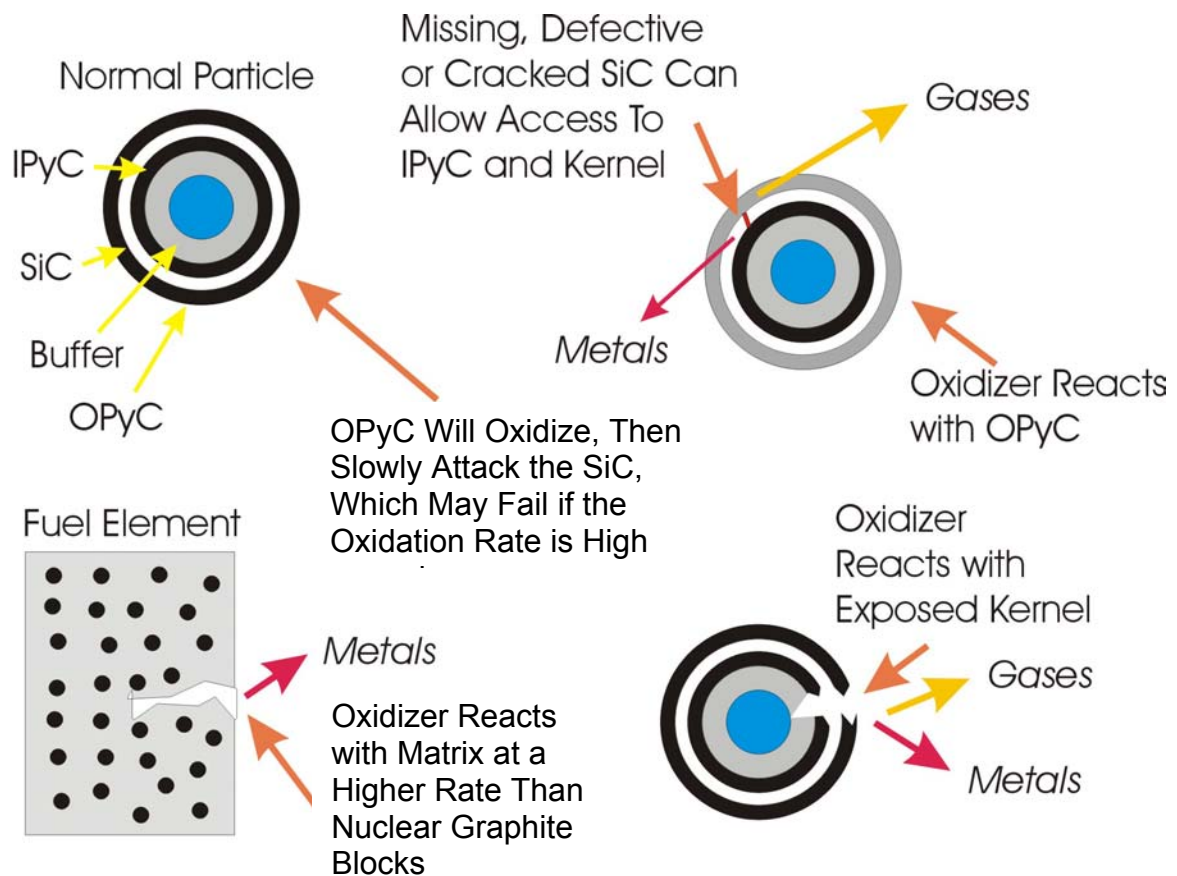


Figure 2-34 An illustration of the chemical attack of a fuel element and fuel particles

Air ingress is a complex thermochemical phenomena, as the reaction can be non-self sustaining or, in extreme situations, self sustaining. The balance between heat production, heat removal, and gas flows all influence the reaction. In addition, the fuel particles are usually shielded from the reactor coolant by several millimeters of highly graphitized material such as the webs of a prismatic fuel block or the outer few millimeters of relatively ungraphitized carbon matrix material of the outer shell of a pebble fuel element. The oxidizer must first penetrate this material before the fuel particles are exposed. Thus, there is a varying amount of “sacrificial” material to limit the damage that might be caused by a modest amount of air.

Sustained air ingress with high carbon reaction rates is possible only if a self-sustaining flow of air is established while the fuel is still at a relatively high temperature. If this should happen, the behavior of  $\text{SiO}_2$  becomes important. In such as case,  $\text{SiO}_2$  would be produced as SiC is oxidized and it forms a layer on the particle that impedes further reaction. However, in an oxygen-limited atmosphere above about  $1300^\circ\text{C}$ ,  $\text{SiO}_2$  may be

converted to SiO, which is volatile and will allow the SiC to SiO reaction to continue unabated.

Figure 2-36 shows the particle failure fraction from irradiated fuel spheres (about 9% burnup) at 1300°C and 1400°C as well as a set of 10 particles at 1500°C while heating in air [2-6]. In an unlimited air supply, failure rates can be high, much greater than in a heatup without air ingress. This event is more aggressive than water ingress, which is primarily a factor with exposed kernels.

### **2.5.3 Water Ingress Accident Phenomena**

The following sections detail the PIRT panel selected coated particle fuel factors, characteristics, and phenomena for water ingress.

#### **2.5.3.1 Kernel**

If the fuel particle is intact during a water ingress accident, the kernel behavior is much the same as during a heatup event (described previously). If the kernel is exposed, it can be oxidized by water [2-6, 2-22, 2-24, 2-43]. During the process of oxidation, the structure of the kernel changes and it releases much of its stored fission product inventory relatively quickly. This effect appears to be burnup dependent. After the kernel completes the process, the kernel release rates return to approximately the pre-oxidation level.

Figure 2-37 shows this effect after exposure to water vapor.

Exposed kernels are most susceptible to water vapor because they react rapidly and intact particles are little affected. Thus, under water ingress conditions, fuel releases are dominated by exposed kernel releases (at least at modest temperatures). This behavior is dependent on the water partial pressure and Figure 2-38 shows the relationship between released stored fission gas and water partial pressure. Finally, it is not known if a catalysis could increase reaction rates under certain conditions.

##### **2.5.3.1.1 Kernel Water Ingress Accident Factors Identified By The PIRT Panel**

Table 2-35 lists the kernel water ingress accident factors identified by the PIRT panel along with the relevant general issues identified previously.



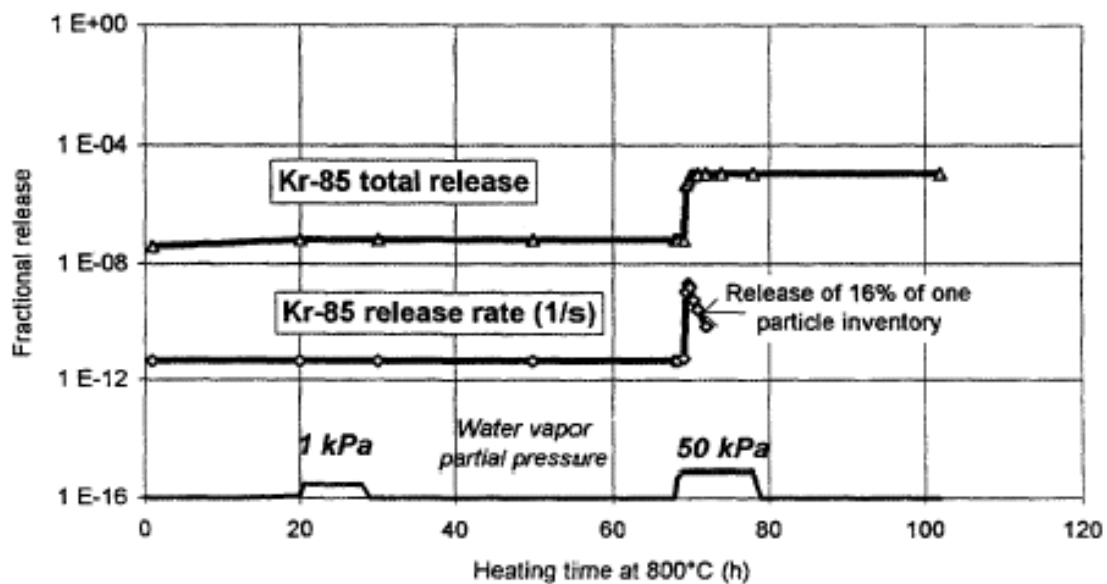


Figure 2-35  $^{85}\text{Kr}$  release from a fuel sphere exposed to water vapor (AVR 92/7, type GLE-3, 9.2% FIMA) at 800C (IAEA TECDOC-978)

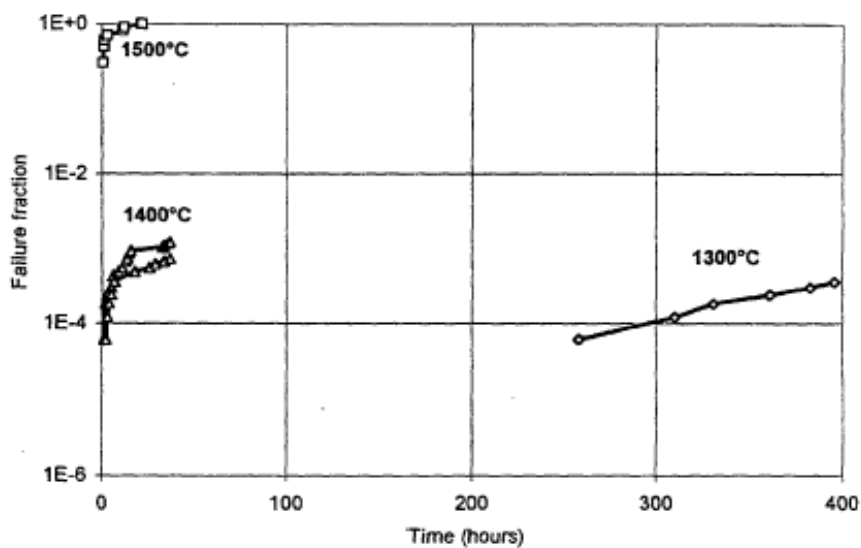


Figure 2-36 Sphere particle failure fractions and 10 unbonded particles (1500°C line) heated in air (IAEA TECDOC-978)

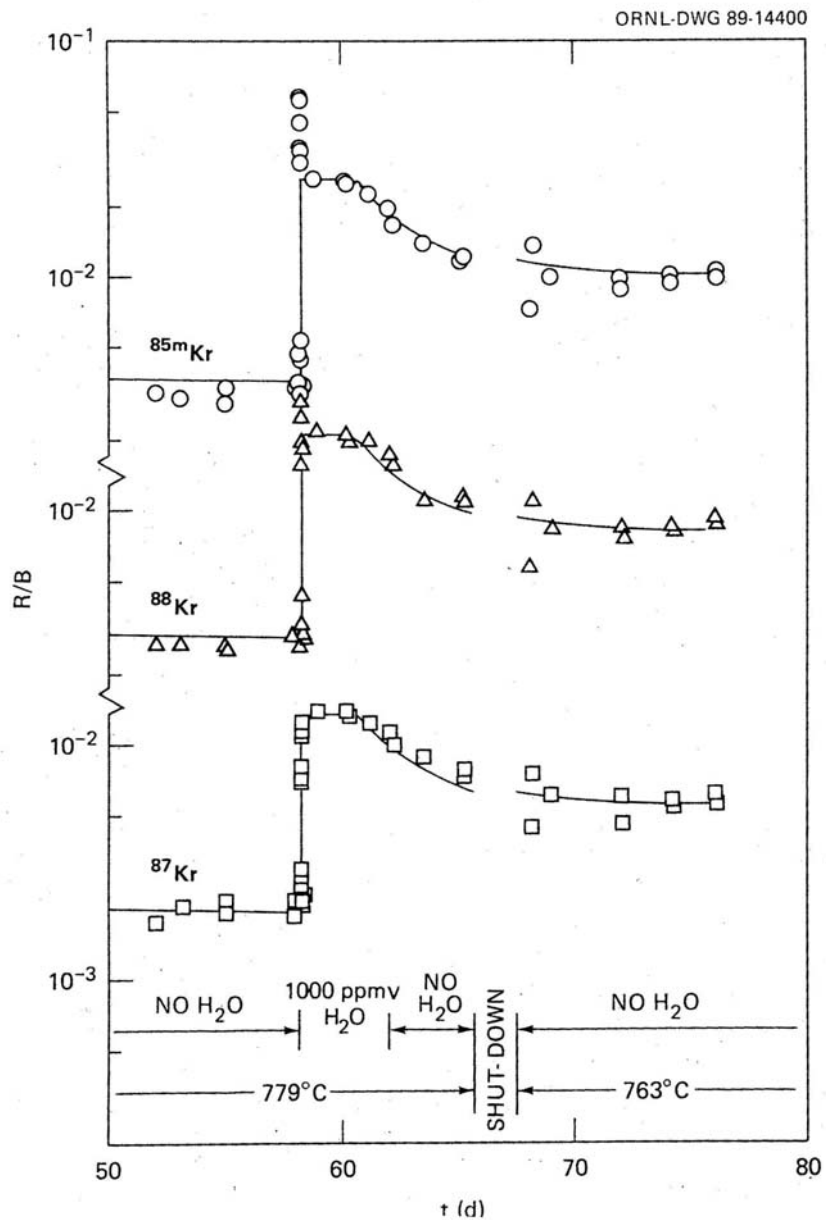


Figure 2-37 Expose to water vapor causes the kernel to restructure and release much of its stored inventory (From Meyers, DOE-HTGR-88486)

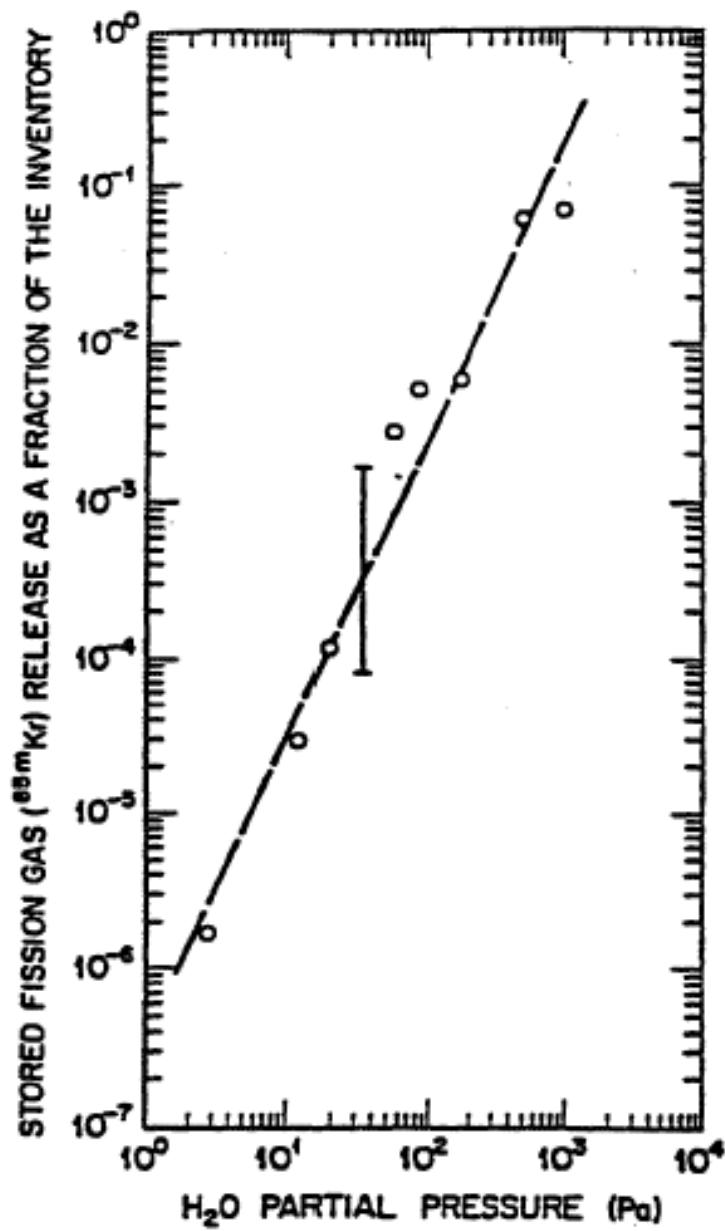


Figure 2-38 Stored fission gas release versus water partial pressure for exposed kernels at 770°C for experiments HFR-B1 and HRB-17 (IAEA TECDOC-978)

**Table 2-35 Kernel Water Ingress Accident Factors Identified By The PIRT Panel**

<b>Kernel Water Ingress Factor</b>	<b>Rationale</b>
Maximum fuel temperature	Chemical reactions and fission product diffusion are strongly influenced by temperature.
Temperature vs. time transient conditions	Time at temperature strongly influences the amount of material reacted or fission products released.
Energy Transport: Conduction within kernel	Determines kernel temperature and fission product diffusion from the kernel.
Thermodynamic state of fission products	The diffusivity of fission products is strongly influenced by their chemical form.
Oxygen flux	A significant redistribution of oxygen has the potential to change the oxygen potential and thus the fission product chemical species within the kernel.
Grain growth	Grain growth could release fission products from the grain to the grain boundary region, thus enhancing transport.
Buffer carbon-kernel interaction	The kernel periphery may react with the Buffer carbon.
Chemical attack by water - Kinetics	The reaction rates will determine the rate of fission product release.
Chemical attack by water - Catalysis	Some impurities or fission products may increase the reaction rate.
Chemical attack by water - Changes in chemical form of fission products	The changes in chemical form of the fission products may change transport properties.
Chemical attack by water – Changes in kernel properties	Structural and chemical changes to the kernel may release stored fission products.

### **2.5.3.2 Buffer Layer**

The buffer layer plays a role similar to that played during a heatup accident for an intact particle. Once exposed, it offers little resistance to fission product transport and may be slowly oxidized by exposure to water. It will also quickly transport water vapor to the kernel.

#### **2.5.3.2.1 Buffer Layer Water Ingress Accident Factors Identified By The PIRT Panel**

Table 2-36 lists the buffer layer water ingress accident factors identified by the PIRT panel. Many of the factors are the same as discussed in previous sections with similar behavior.

**Table 2-36 Buffer Layer Water Ingress Accident Factors Identified By The PIRT Panel**

<b>Buffer Water Accident Ingress Factor</b>	<b>Rationale</b>
Gas phase diffusion	This layer offers little impedance to the transport of fission products.
Condensed phase diffusion	
Response to kernel swelling	The buffer will have to accommodate any kernel distortion to protect the other layers.
Maximum fuel gaseous fission product uptake	The buffer layer must have sufficient free volume to withstand the pressure.
Layer oxidation	The buffer will be oxidized from the outside if the particle is cracked or broken.
Thermal gradient	The gradient may influence the transport of fission products to the other layers.
Irradiation and thermal shrinkage	The buffer is expected to isolate the kernel from the other layers. Damage to it may compromise this ability.
Chemical attack by water - Kinetics	The reaction rates will determine the rate of fission product release.
Chemical attack by water - Changes in chemical form of fission products	The changes in chemical form of the fission products may change their transport properties.
Chemical attack by water - Changes in graphite properties	Changes in the graphite (or PyC) properties may affect the transport of fission products or the release of trapped fission products.
Chemical attack by water - Holdup reversals	Stored fission products in the buffer may be released quickly if its structure is seriously attacked.
Chemical attack by water - Temperature distributions	The temperature of the buffer and surrounding materials determines reaction rates and transport.

### 2.5.3.3 IPyC Layer

The factors for the IPyC during air or water ingress are similar to those for heatup with the additional aspect of chemical attack. If the OPyC and SiC remain intact, the IPyC will not be exposed to a new environment. However, if the other layers become damaged or cracked, this layer would be exposed to water and be subject to a chemical reaction. Since this layer would be the final barrier for gases (metals will have begun diffusing through the layer), its failure would allow the release any stored gases and expose the kernel to the oxidizing environment with the subsequent kernel reaction.

### 2.5.2.3.1 IPyC Layer Water Ingress Accident Factors Identified By The PIRT Panel

Table 2-37 lists the IPyC layer water ingress accident factors identified by the PIRT panel.

**Table 2-37 IPyC Layer Water Ingress Accident Factors Identified By The PIRT Panel**

<b>IPyC Water Ingress Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	Gas diffusion is low, while metallic diffusion is high; chemical attack could increase both rates.
Condensed phase diffusion	
Pressure loading (fission products)	High pressures can challenge this layer. The IPyC can help keep the SiC in compression if the bonding between layers is strong.
Pressure loading (CO)	The same CO issues apply here. High CO production will result in high pressures.
Layer oxidation	If the IPyC cracks or otherwise allows internal CO to the SiC, corrosion may result (inside out). Cracks in the OPyC and SiC will allow oxidation of the IPyC (outside in).
Stress state (compression/tension)	See general discussions on coating stress distributions.
Cracking	Same situation as layer oxidation and stresses. If other layers are broken, cracking will allow the oxidizer to the kernel.
Intercalation	Chemical attack of the IPyC may allow release of trapped fission products.
Chemical attack by water - Kinetics	The reaction rates will determine the rate of fission product release.
Chemical attack by water - Catalysis	Some impurities or fission products may increase the reaction rate.
Chemical attack by water - Changes in chemical form of fission products	The changes in chemical form of the fission products may change their transport properties.
Chemical attack by water - Changes in graphite properties	Changes in the graphite (or PyC) properties may affect the transport of fission products or the release of trapped fission products.
Chemical attack by water - Holdup reversal	Serious damage to the IPyC will allow access to the Buffer and its relatively large inventory of fission products.
Chemical attack by water - Temperature distributions	The temperature of the IPyC and surrounding materials determines reaction rates and transport.

#### **2.5.3.4 SiC Layer**

Under water ingress conditions, the SiC layer performs in a manner similar to that of a heatup accident until the OPyC begins to be subject to chemical attack. As the OPyC becomes weakened and fails, the stress state of the SiC would change. This may lead to failure if the SiC has been weakened by other causes. The SiC would then be exposed to water and perhaps some generated CO [2-6, 2-31].

Thinning of this layer can cause the particle to fail by pressure vessel failure and penetration of the layer will cause the release of metallic fission products and expose the IPyC to oxidation.

##### **2.5.3.4.1 SiC Layer Water Ingress Accident Factors Identified By The PIRT Panel**

Table 2-38 lists the SiC layer water ingress accident factors identified by the PIRT panel.

#### **2.5.3.5 OPyC Layer**

Under ingress conditions, the OPyC first functions in the same way as in the heatup case; it then suffers erosion. Its loss due to chemical attack has a similar effect except that the SiC is now exposed to attack (covered previously).

##### **2.5.3.5.1 OPyC Layer Water Ingress Accident Factors Identified By The PIRT Panel**

Table 2-39 lists the OPyC layer water ingress accident factors identified by the PIRT panel.

#### **2.5.3.6 Fuel Element**

Under ingress conditions, the fuel element responds first and would be in a manner similar to that for a heatup condition. However, the matrix material of an element is more reactive than either the fuel particle pyrocarbon or core graphite blocks and is the first material to be damaged due to chemical attack (if exposed, in some cases a thick reflector region may first see the ingress and consume the water). For low levels of water this could be an advantage as there are large amounts of matrix material and some protection could be provided to the fuel particles. Some fuel forms have a thick outer layer that must be consumed before the fuelled region can be reached and other forms are imbedded into graphite blocks so specifics of the situation must be considered.

Loss of significant amounts of matrix material is not desirable, however. Increased porosity and cracks can expose the fuel particles to the water and consumption of the matrix material might structurally weaken the fuel element. Fission products sorbed in the matrix during normal operation would be released into the remaining coolant as the matrix material reacts with the water.

For the water-carbon reaction, the reaction is endothermic and is driven by the nuclear decay heat, so high temperatures and high material consumption rates driven by aggressive chemical reactions are not expected.

**Table 2-38 SiC Layer Water Ingress Accident Factors Identified By The PIRT Panel**

<b>SiC Water Ingress Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	The SiC layer is the major barrier to the transport of fission products. Significant changes in the transport properties can greatly increase release of fission products.
Condensed phase diffusion	
Thermal deterioration or decomposition	Above about 1600-1800°C the SiC begins to decompose and its ability to retain fission products is greatly reduced.
Fission product corrosion	Some fission products may migrate to the SiC and damage it. This process is a function of temperature and can begin during normal operation. Pd is the main concern
Heavy metal diffusion	If fissile material were to be transported across the SiC layer, a possible core configuration issue may arise (considered unlikely).
Layer oxidation	Attack of the SiC layer by CO could occur due to a failed OPyC at high temperatures (outside in). The SiC can also be damaged by CO exposure due to IPyC failure (inside out).
Fission product release through undetected defects	Undetected SiC defects could have poor fission product retention behavior not apparent during normal operations.
Fission product release through failures, e.g. cracking	Failure of the SiC will allow the release of metallic fission products even with intact PyCs.
Thermodynamics of the SiC fission product system	SiC is known to be attacked by some noble metals. At higher burnups and temperatures, other chemical concerns may arise.
Sintering	High temperatures could results in microstructural changes to the SiC that change transport behavior.
Chemical attack by water - Kinetics	The reaction rates will determine the rate of fission product release.
Chemical attack by water - Catalysis	Some impurities or fission products may increase the oxidation rate.
Chemical attack by water - Changes in chemical form of fission products	The changes in chemical form of the fission products may change their transport properties.
Chemical attack by water - Changes in SiC properties	Changes in the SiC properties may affect the transport of fission products or the release of trapped fission products.
Chemical attack by water - Holdup reversal	Extensive damage to the SiC would allow access to the IPyC and its relatively poor retention of metallic fission products.
Chemical attack by water - Temperature distributions	The temperature of the SiC and surrounding materials determines reaction rates and transport.



**Table 2-39 OPyC Layer Water Ingress Accident Factors Identified By The PIRT Panel**

<b>OPyC Water Ingress PIRT Factor</b>	<b>Rationale</b>
Gas phase diffusion	Gas diffusion is low, while metallic diffusion is high; chemical attack could increase both rates.
Condensed phase diffusion	
Layer oxidation	The OPyC is the most exposed layer. It would be the first layer subjected to oxidation (sound particle).
Stress state	See general discussions on coating stress distributions. Its failure will increase the likelihood of SiC failure.
Intercalation	Chemical attack of the OPyC may allow release of trapped fission products.
Trapping	Chemical attack of the OPyC may allow release of trapped fission products.
Cracking	Failure of the OPyC will change the stress state of the particle; if the other layers are breached, the gases will be released.
Chemical attack by water - Kinetics	The reaction rates will determine the rate of fission product release.
Chemical attack by water - Catalysis	Some impurities or fission products may increase the reaction rate.
Chemical attack by water - Changes in chemical form of fission products	The changes in chemical form of the fission products may change their transport properties.
Chemical attack by water - Changes in graphite properties	Changes in the graphite (or PyC) properties may affect the transport of fission products or the release of trapped fission products.
Chemical attack by water - Holdup reversal	Attack of the OPyC may result in the release of any stored fission products.
Chemical attack by water - Temperature distributions	The temperature of the OPyC and surrounding materials determines reaction rates and transport.

**2.5.3.6.1 Fuel Element Water Ingress Accident Factors Identified By The PIRT Panel**

Table 2-40 lists fuel element water ingress accident factors identified by the PIRT panel. Many of these factors are the same as were seen for the heatup conditions.

**Table 2-40 Fuel Element Water Ingress Accident Factors Identified By The PIRT Panel**

<b>Fuel Element Water Ingress Accident Factor</b>	<b>Rationale</b>
Irradiation history	Matrix behavior is strongly affected by fast fluence and temperature exposure, which may cause shrinkage of the matrix and change its chemical reactivity.
Condensed phase diffusion	Gas diffusion is high, while metallic diffusion is variable. The less volatile metals can be sorbed in the matrix material. Chemical attack could increase the transport.
Gas phase diffusion	
Gaseous transport - holdup	Gas transport through the matrix is generally high.
Transport of metallic FPs through fuel element - Chemical form	Changes in the chemical form of the fission products could increase their transport rate through the matrix.
Chemical attack by water - Kinetics	The reaction rates will determine the rate of fission product release.
Chemical attack by water - Catalysis	Some impurities or fission products may increase the reaction rate.
Chemical attack by water - Changes in chemical form of fission products	The changes in chemical form of the fission products may change their transport properties.
Chemical attack by water - Changes in graphite properties	Changes in the graphite (matrix) properties may affect the transport of fission products or the release of trapped fission products.
Chemical attack by water - Holdup reversal	Serious damage to the matrix may allow release of sorbed fission products.
Chemical attack by water - Temperature distributions	The temperature of the matrix and surrounding materials determines reaction rates and transport.

#### **2.5.4 Air Ingress Accident Phenomena**

The following sections detail the PIRT panel selected coated particle fuel factors, characteristics, and phenomena for air ingress. As part of the PIRT process, the panel members felt that calculations of the postulated air ingress event were needed to better inform the panel members about the potential conditions that fuel pebbles experience in a postulated air ingress event. A MELCOR model was available at the INEEL to address the important phenomena related to air ingress events in a pebble bed reactor. Appendix G contains the results of the MELCOR calculations performed for the air-intrusion accident scenario.

### 2.5.4.1 Kernel

If the fuel particle is intact during an ingress accident, the kernel behavior is much the same as during a heatup event (described previously). If the kernel is exposed, it can be oxidized by oxygen or CO (the oxygen is converted to CO by the large amount of carbon in the reactor system) [2-6, 2-22, 2-24]. During the oxidation process, the structure of the kernel changes and it releases much of its stored fission product inventory relatively quickly. After the kernel completes the process, the kernel releases return to approximately the pre-oxidation level.

Finally, it is not known if a catalysis could increase reaction rates under certain conditions.

#### 2.5.4.1.1 Kernel Air Ingress Accident Factors Identified By The PIRT Panel

Table 2-41 lists the kernel air ingress accident factors identified by the PIRT panel along with the relevant general rationales identified previously.

**Table 2-41 Kernel Air Ingress Accident Factors Identified By The PIRT Panel**

<b>Kernel Air Ingress Accident Factor</b>	<b>Rationale</b>
Maximum fuel temperature	Chemical reactions and fission product diffusion are strongly influenced by temperature.
Temperature vs. time transient conditions	Time at temperature strongly influences the amount of material reacted or fission products released.
Energy Transport: conduction within kernel	Determines kernel temperature and fission product diffusion from the kernel.
Thermodynamic state of fission products	The diffusivity of fission products is strongly influenced by their chemical form.
Oxygen flux	A significant redistribution of oxygen has the potential to change the oxygen potential and thus the fission product chemical species within the kernel.
Grain growth	Grain growth could release fission products from the grain to the grain boundary region, thus enhancing transport.
Buffer carbon-kernel interaction	The kernel periphery may react with the buffer carbon.
Chemical attack by air - Kinetics	The reaction rates will determine the rate of fission product release.
Chemical attack by air - Catalysis	Some impurities or fission products may increase the reaction rate.

**Table 2-41 Kernel Air Ingress Accident Factors Identified By The PIRT Panel (continued)**

Chemical attack by air - Changes in chemical form of fission products	The oxygen potential of the kernel may increase due to the available oxygen. The changes in chemical form of the fission products may change transport properties.
Chemical attack by air – Changes in graphite properties	Changes in the graphite (or PyC) properties may affect the transport of fission products or the release of trapped fission products.
Chemical attack by air - Holdup reversal	Structural and chemical changes to the kernel may release stored fission products.
Chemical attack by air – Temperature distributions	The temperature of the kernel and surrounding material will affect reaction rates and the transport of fission products.

### **2.5.4.2 Buffer Layer**

The buffer layer plays a role similar to that played during a heatup accident for an intact particle. Once exposed, it offers little resistance to fission product transport and will be oxidized by exposure to air. It will also quickly transport oxidizer to the kernel. Air attack is much more aggressive than water. It is possible a catalysis could increase reaction rates under certain conditions if present.

#### **2.5.4.2.1 Buffer Layer Air Ingress Accident Factors Identified By The PIRT Panel**

Table 2-42 lists the buffer layer air ingress accident factors identified by the PIRT panel. Many of the factors are the same as discussed in previous sections with similar rationales.

### **2.5.4.3 IPyC**

The factors for the IPyC during air ingress are similar to those for heatup with the additional aspect of chemical attack. If the OPyC and SiC remain intact, the IPyC will not be exposed to a new environment. However, if the other layers become damaged or cracked, this layer will be exposed to air and CO and be subject to a chemical reaction. Since this layer is now the final barrier for gases (metals will have begun diffusing through the layer), its failure will release any stored gases and expose the kernel to the oxidizing environment with the subsequent kernel reaction.

#### **2.5.4.3.1 IPyC Layer Air Ingress Accident Factors Identified By The PIRT Panel**

Table 2-43 lists the IPyC layer air ingress accident factors identified by the PIRT panel.

**Table 2-42 Buffer Layer Air Ingress Accident Factors Identified By The PIRT Panel**

<b>Buffer Air Ingress Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	This layer offers little impedance to the transport of fission products.
Condensed phase diffusion	
Response to kernel swelling	The buffer will have to accommodate any kernel distortion to protect the other layers.
Maximum fuel gaseous fission product uptake	The buffer layer must have sufficient free volume to accommodate the fission products at an acceptable pressure.
Layer oxidation	The buffer layer may react with oxide materials in the kernel (prior to air exposure).
Thermal gradient	The gradient may influence the transport of fission products to the other layers.
Irradiation and thermal shrinkage	The buffer is expected to isolate the kernel from the other layers. Damage to it may compromise this ability.
Chemical attack by air - Kinetics	The reaction rates will determine the rate of fission product release.
Chemical attack by air - Catalysis	Some impurities or fission products may increase the reaction rate.
Chemical attack by air - Changes in chemical form of fission products	The changes in chemical form of the fission products may change their transport properties.
Chemical attack by air - Changes in graphite properties	Changes in the graphite (or PyC) properties may affect the transport of fission products or the release of trapped fission products.
Chemical attack by air - Holdup reversal	Stored fission products in the buffer may be released quickly if its structure is seriously attacked.
Chemical attack by air - Temperature distributions	The temperature of the buffer and surrounding materials determines reaction rates and transport.

#### **2.5.4.4 SiC Layer**

Under air ingress conditions, the SiC layer performs in a manner similar to that of a heatup accident until the OPyC begins to be subject to chemical attack. If the OPyC becomes weakened and fails, the stress state of the SiC would change. This may lead to failure if the SiC has been weakened by other causes. After OPyC loss, the SiC would then be exposed to air and CO. Depending on the conditions, either SiO<sub>2</sub> or SiO (low oxygen) can be formed. SiO is volatile while SiO<sub>2</sub> is not. SiO<sub>2</sub> will impede the reaction rate as the reactant has to diffuse through it. Since SiO is gaseous, it does not offer this protection. Thus, the specific details of the reacting environment determine the damage and the rate of damage to the SiC layer [2-6, 2-31].

Thinning of this layer can cause the particle to fail by pressure vessel failure and penetration of the layer will cause the release of metallic fission products and expose the IPyC to oxidation.

**Table 2-43 IPyC Layer Air Ingress Accident Factors Identified By The PIRT Panel**

<b>IPyC Air Ingress Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	Gas diffusion is low, while metallic diffusion is high; chemical attack could increase both rates.
Condensed phase diffusion	
Pressure loading (fission products)	High pressures can challenge this layer. The IPyC can help keep the SiC in compression if the bonding between layers is strong.
Pressure loading (CO)	The same CO issues apply here. High CO production will result in high pressures.
Layer oxidation	If the IPyC cracks or otherwise allows internal CO to the SiC, corrosion may result (inside out). Cracks in the OPyC and SiC will allow oxidation of the IPyC (outside in).
Stress state (compression/tension)	See general discussions on coating stress distributions.
Cracking	Same situation as layer oxidation and stresses. If other layers are broken, cracking will allow the oxidizer to the kernel.
Intercalation	Chemical attack of the IPyC may allow release of trapped fission products.
Chemical attack by air - Kinetics	The reaction rates will determine the rate of fission product release
Chemical attack by air - Catalysis	Some impurities or fission products may increase the reaction rate.
Chemical attack by air - Changes in chemical form of fission products	The changes in chemical form of the fission products may change their transport properties.
Chemical attack by air - Changes in graphite properties	Changes in the graphite (or PyC) properties may affect the transport of fission products or the release of trapped fission products.
Chemical attack by air - Holdup reversal	Serious damage to the IPyC will allow access to the buffer and its relatively large inventory of fission products.
Chemical attack by air - Temperature distributions	The temperature of the IPyC and surrounding materials determines reaction rates and transport.

**2.5.4.4.1 SiC Layer Air Ingress Accident Factors Identified By The PIRT Panel**

Table 2-43 lists the SiC layer air ingress accident factors identified by the PIRT panel.

**Table 2-44 SiC Layer Air Ingress Accident Factors Identified By The PIRT Panel**

<b>SiC Air Ingress Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	The SiC layer is the major barrier to the transport of fission products. Significant changes in the transport properties can greatly increase release of fission products.
Condensed phase diffusion	
Thermal deterioration or decomposition	Above about 1600-1800°C the SiC begins to decompose and its ability to retain fission products is greatly reduced.
Fission product corrosion	Some fission products may migrate to the SiC and damage it. This process is a function of temperature and can begin during normal operation. Pd is the main concern
Heavy metal diffusion	If fissile material were to be transported across the SiC layer, a core configuration issue may arise.
Layer oxidation	Attack of the SiC layer by CO due to a failed OPyC at high temperatures problematic. The SiC can also be damaged by CO exposure due to IPyC failure (inside out). The formation of either SiO or SiO <sub>2</sub> can determine the rate at which the SiC is eroded.
Fission product release through undetected defects	Undetected defective SiC could have poor accident behavior not apparent during normal operations.
Fission product release through failures, e.g. cracking	Failure of the SiC will allow the release of metallic fission products even with intact PyCs.
Thermodynamics of the SiC fission product system	SiC is known to be attacked by some noble metals. At higher burnups and temperatures, other chemical concerns may arise.
Sintering	High temperatures could results in microstructural changes to the SiC that change transport behavior.
Chemical attack by air - Kinetics	The reaction rates will determine the rate of fission product release.
Chemical attack by air - Catalysis	Some impurities or fission products may increase the oxidation rate.
Chemical attack by air - Changes in chemical form of fission products	The changes in chemical form of the fission products may change their transport properties.
Chemical attack by air - Changes in SiC properties	SiC property changes may affect the transport of fission products or release of trapped fission gases.
Chemical attack by air - Holdup reversal	Serious damage to the SiC will allow access to the IPyC and its relatively poor retention of metallic fission products.
Chemical attack by air - Temperature distribution	The temperature of the SiC and surrounding materials determines reaction rates and transport.

### 2.5.4.5 OPyC

Under air ingress conditions, the OPyC first functions in the same way as in the heatup case; it then suffers erosion. Its loss due to chemical attack has a similar effect except that the SiC is now exposed to attack (covered previously).

#### 2.5.4.5.1 OPyC Layer Air Ingress Accident Factors Identified By The PIRT Panel

Table 2-44 lists the OPyC layer air ingress accident factors identified by the PIRT panel.

**Table 2-45 OPyC Layer Air Ingress Accident Factors Identified By The PIRT Panel**

<b>OPyC Air Ingress Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	Gas diffusion is low, while metallic diffusion is high; chemical attack could increase both rates.
Condensed phase diffusion	
Layer oxidation	The OPyC is the most exposed layer. It would be the first layer subjected to oxidation
Stress state	See general discussions on coating stress distributions. Failure will increase the likelihood of SiC failure.
Intercalation	Chemical attack of the OPyC may allow release of trapped fission products.
Trapping	Chemical attack of the OPyC may allow release of trapped fission products.
Cracking	OPyC failure will change the stress state of the particle; gases will be released if other layers breached.
Chemical attack by air - Kinetics	The reaction rates will determine the rate of fission product release
Chemical attack by air - Catalysis	Some impurities or fission products may increase the oxidation rate.
Chemical attack by air - Changes in chemical form of fission products	The changes in chemical form of the fission products may change their transport properties.
Chemical attack by air - Changes in graphite properties	Changes in the graphite (or PyC) properties may affect the transport of fission products or the release of trapped fission products.
Chemical attack by air - Holdup reversal	Attack of the OPyC may result in the release of any stored fission products.
Chemical attack by air - Temperature distribution	The temperature of the OPyC and surrounding materials determines reaction rates and transport.



#### **2.5.4.6 Fuel Element**

Under air ingress conditions, the fuel element responds first in a manner similar to that for a heatup. However, the matrix material of a fuel element is more reactive than either the fuel particle pyrocarbon or core graphite blocks and is the first material to suffer damage due to chemical attack (if exposed, in some cases a thick reflector region may first see the air ingress and buffer the damage). For small amounts of air this could be an advantage as there are large amounts of matrix material and some protection could be provided to the fuel particles. Some fuel forms have a thick outer layer that must be consumed before the fuelled region can be reached and other forms are imbedded into graphite blocks so specifics of the situation must be considered.

Loss of significant amounts of matrix material is not desirable, however and air ingress is more aggressive than water ingress. Increased porosity and cracks can expose the fuel particles to the air and consumption of the matrix material might structurally weaken the fuel element.

Fission products sorbed in the matrix would be released into the remaining coolant-air mix as the matrix material reacts with the oxidizer. The chemical forms of the released fission products may change (oxidize) if large amounts of air enter the core. A sudden release of this sorbed material might change the accident analysis if the chemical attack is extensive.

Generally, the heat generated by these reactions is considered to be small compared to the nuclear decay heat, but this must be examined on a case-by-case basis. Experiments in air have seen greater releases from fuel elements than from unbonded particles at similar apparatus temperatures, suggesting that the energy of combustion may have an important local effect.

Figure 2-39 shows the oxidation of two similar fuel spheres in air. Note that the matrix material was oxidized at 1100°C with no releases. Releases were seen from the remaining fuel particles when the temperature was raised to 1400°C. In an inert environment, no failures would be expected [2-6, 2-20 to 2-24].

##### **2.5.4.6.1 Fuel Element Air Ingress Accident Factors Identified By The PIRT Panel**

Table 2-46 lists the fuel element air ingress accident factors identified by the PIRT panel. Many of these issues are the same as were seen for the coated particle fuel ingress and heatup conditions.

**Table 2-46 Fuel Element Air Ingress Accident Factors Identified By The PIRT Panel**

<b>Fuel Element Air Ingress Accident Factor</b>	<b>Rationale</b>
Irradiation history	Matrix behavior is strongly affected by fast fluence and temperature exposure, which may cause shrinkage of the matrix and change its chemical reactivity.
Condensed phase diffusion	Gas diffusion is high, while metallic diffusion is variable. The less volatile metals can be sorbed in the matrix material. Chemical attack could increase the transport.
Gas phase diffusion	
Transport of metallic FPs through fuel element - Chemical form	Changes in the chemical form of the fission products could increase their transport rate through the matrix.
Chemical attack by air - Kinetics	The reaction rates will determine the rate of fission product release
Chemical attack by air - Catalysis	Some impurities or fission products may increase the oxidation rate.
Chemical attack by air - Changes in chemical form of fission products	The changes in chemical form of the fission products may change their transport properties.
Chemical attack by air - Changes in graphite properties	Changes in the graphite (or PyC) properties may affect the transport of fission products or the release of trapped fission products.
Chemical attack by air - Holdup reversal	Significant damage to the matrix material could allow the release of the sorbed fission products.
Chemical attack by air - Temperature distribution	The temperature of the matrix and surrounding materials determines reaction rates and transport.

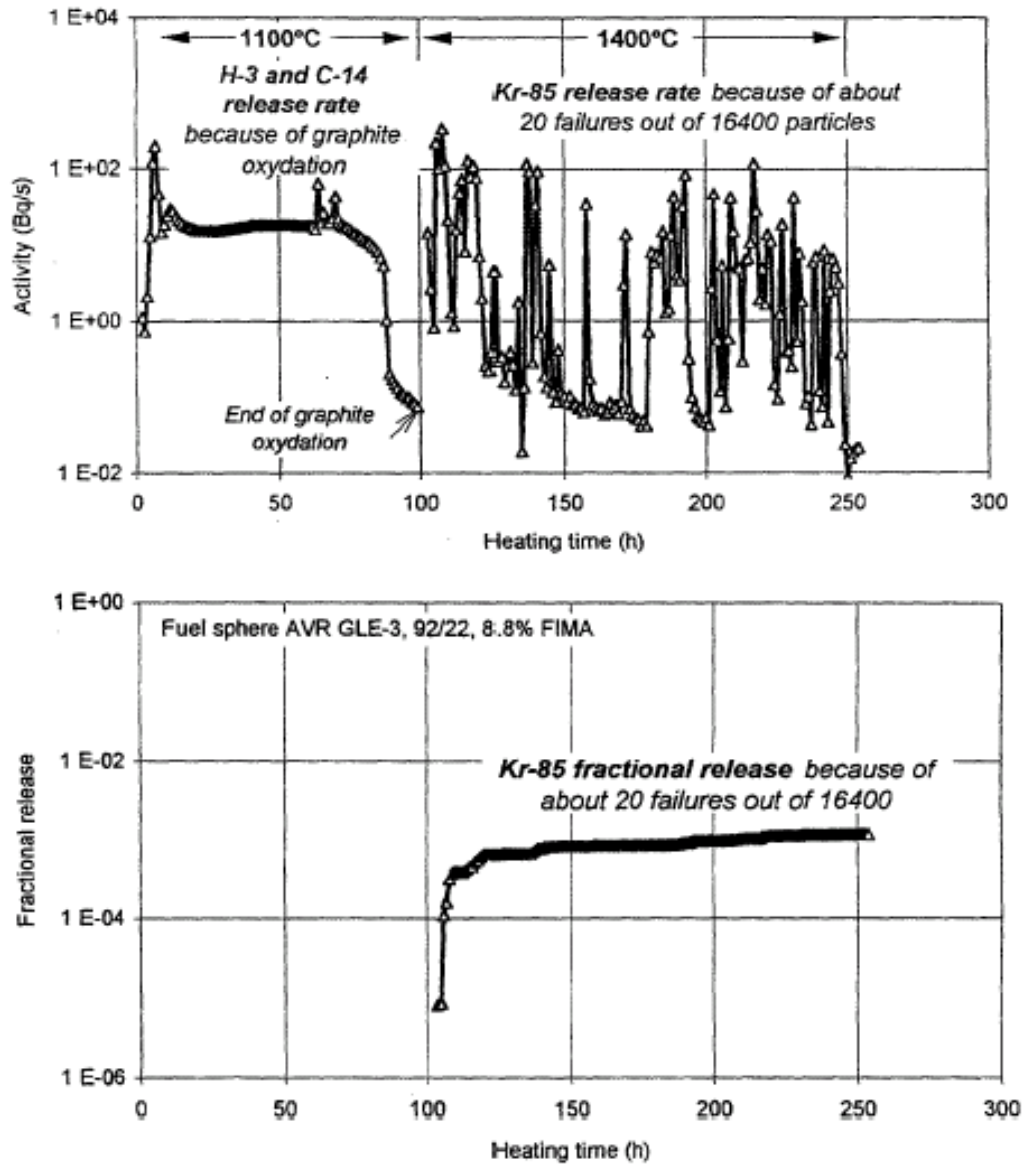


Fig. 2-39 Oxidation of two similar fuel spheres in air. Top is from AVR 92/8, 9% FIMA, bottom is from AVR 92/22, 8.8% FIMA (IAEA TECDOC-978)

### 2.5.5 Reactivity Accident

An important class of potential accidents for reactors in general is the sudden increase in reactor local or core-wide power due to a reactivity increase. This might be caused by a control system malfunction, control rod ejection, or a sudden change in the core internal arrangement such as pebble compaction in a pebble bed reactor.

Because of the constant cycling of the fuel, a pebble bed reactor has low excess reactivity. The prismatic core design is more like an LWR in terms of excess reactivity (and burnable poisons) at the start of life, which is reduced as the core nears end of life. The designers of each type strive to minimize excess reactivity and to limit control rod worth and accident movement, so the actual accident pulse could be modest.

A sudden pulse of power might damage the fuel elements and the fuel particles, leading to a large release of fission products from the fuel. If the pressure boundary has also been damaged, such as for a rod ejection accident, a driving force would be available to transport fission products outside of the reactor boundary. For a reactor like the HTTR design, a certain regulatory event could lead to a pulse length of 8 seconds with an energy deposition of  $1.26 \times 10^4$  J/g of  $\text{UO}_2$  (no mitigation) [2-6, 2-44]. However, for other postulated HTGR events, pulses could be much shorter with much higher energy deposition. While limited reactivity insertion accident (RIA) testing has been done and much of it is at difference energies and pulse lengths, an understanding has evolved [2-6].

The large amount of graphite in the core along with its high temperature capability eliminates concerns of core melting, but the kernel can still see high temperatures and the resulting pressures can fail coatings.

The examination of Japanese fuel compacts after pulse testing (10-30 ms) showed internal  $\text{UO}_2$  kernel melting and central vaporization for coated particles with an energy deposition of 2300 J/g  $\text{UO}_2$ . Microprobe analysis revealed that uranium vapor penetrated the cracks in the coating layers. Particles that saw lower energies appeared almost unchanged, but layer cracking was still present [2-6].

Russian pulse tests used both short pulses and long pulses. In a first set of tests done at the Impulse Graphite Reactor (IGR) reactor, a pulse of duration (half width peak) 0.7 seconds and energy of  $2.6 \times 10^4$  J/g  $\text{UO}_2$  was deposited into spherical fuel elements, which remained intact. A second set of tests with a pulse duration of 30 seconds and an energy of  $9 \times 10^4$  J/g  $\text{UO}_2$  resulted in the failure of the coated particles and fragmentation of the fuel element sphere [2-6].

A set of short pulses (1-2 ms) was also conducted by the Russians in the HYDRA reactor with energies of 100 to 1700 J/g  $\text{UO}_2$ . They observed coating cracking at  $<1050$  J/g  $\text{UO}_2$  and kernel restructuring at  $>1300$  J/g  $\text{UO}_2$ . Figure 2-40 outlines this behavior. Three particle types are shown. A normal density 490 micron diameter kernel ( $10.8 \text{ g/cm}^3$ ), a lower density 532 micron diameter kernel ( $7.9 \text{ g/cm}^3$ ), and a particle with a layer composed of both SiC and PyC on both sides of the SiC [2-6].

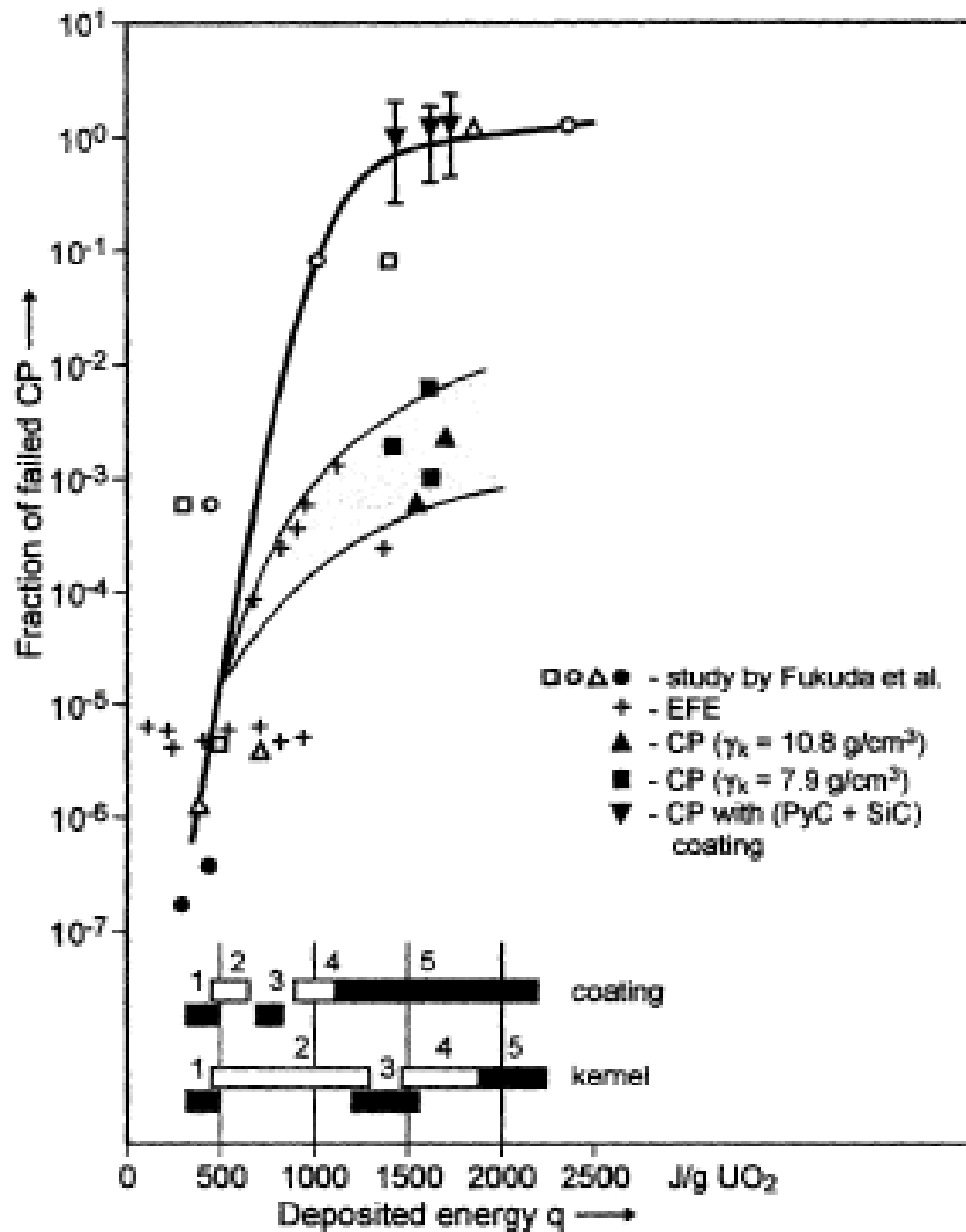


Fig. 2-40 Particle failure rate versus pulse energy deposition (IAEA TECDOC-978)

The Russian fuel is different than US or German fuel because different layer thickness are used, so the results may not be directly transferable.

### 2.5.5.1 Kernel

The energy is deposited in the kernel and a sudden deposition of energy will raise the temperature of the kernel causing the sudden release of fission products trapped in the grains and crystal structure and even kernel melting. A sudden pressure increase or pressure pulse may over pressurize and break the coating layers as well as increase the temperature of the coatings.

Past testing indicates that the energy deposition rate for fuel damage is in the range of 1000-2000 J/g [2-6].

#### 2.5.5.1.1 Kernel Reactivity Accident Factors Identified By The PIRT Panel

Table 2-47 lists the kernel reactivity accident factors identified by the PIRT panel as well as the environmental conditions of interest.

**Table 2-47 Kernel Reactivity Accident Factors Identified By The PIRT Panel**

<b>Kernel Reactivity Accident Factor</b>	<b>Rationale</b>
Maximum fuel temperature	Chemical reactions and fission product diffusion are strongly influenced by temperature.
Temperature vs. time transient conditions	Time at temperature strongly influences the amount of material reacted or fission products released.
Energy deposition (total)	Determines the fuel and core temperature
Energy deposition rate	Determines the likelihood of impulsive fuel damage and system response time.
Energy Transport: Conduction within kernel	Determines kernel temperature and fission product diffusion from the kernel.
Thermodynamic state of fission products	The diffusivity of fission products is strongly influenced by their chemical form.
Gas phase diffusion	The details of the event, such as melting of the kernel, can modify the diffusion of fission products.
Condensed phase diffusion	
Oxygen flux	A significant redistribution of oxygen has the potential to change the oxygen potential and thus the fission product chemical species within the kernel.
Grain growth	Grain growth could release fission products from the grain to the grain boundary region, thus enhancing transport.
Buffer carbon-kernel interaction	The kernel periphery may react with the buffer carbon.

### 2.5.5.2 Buffer Layer

The buffer layer affects the response of the coated fuel particle to a reactivity event because it provides the expansion space for the particle gases. Any gases suddenly released from the kernel during the accident would have to diffuse into the buffer layer. This process could result in a brief pressure pulse that could damage the coatings. Otherwise the buffer plays the same role as described previously.

#### 2.5.5.2.1 Buffer Layer Reactivity Accident Factors Identified By The PIRT Panel

Table 2-48 lists the buffer layer reactivity accident factors identified by the PIRT panel plus the relevant general issues.

**Table 2-48 Buffer Reactivity Accident Factors Identified By The PIRT Panel**

<b>Buffer Reactivity Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	The fission product transport through the buffer layer is expected to be high at accident temperatures.
Condensed phase diffusion	
Response to kernel swelling	The buffer layer must be weak enough that it will deform to accommodate the kernel.
Maximum fuel gaseous fission product uptake	The buffer layer must have sufficient free volume to accommodate the fission products at an acceptable pressure.
Layer oxidation	Any oxygen released from the kernel will oxidize a small portion of the buffer. This is of no consequence for the buffer, but may increase the particle pressure.
Thermal gradient	Gradients could be high for a high energy pulse.
Irradiation and thermal shrinkage	The buffer is designed to isolate the kernel from the IPyC, but small cracks could act to concentrate fission products.

### 2.5.5.3 IPyC Layer

During the pulse, the particle pressure may stress the IPyC. The main goal is to maintain particle integrity by accommodating the particle pressure. This is similar to normal operation, expect that the pulse may impose increased stresses in the layer.

#### 2.5.5.3.1 IPyC Layer Reactivity Accident Factors Identified By The PIRT Panel

Table 2-49 lists the IPyC layer reactivity accident factors identified by the PIRT panel.

**Table 2-49 IPyC Layer Reactivity Accident Factors Identified By The PIRT Panel**

<b>IPyC Reactivity Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	Gaseous fission products are generally retained by the IPyC, but metallics transport is high. High local accident temperatures could increase the diffusion rate.
Condensed phase diffusion	
Pressure loading (fission products)	High pressure could challenge this layer. The IPyC can help keep the SiC in tension if the bonding between layers is strong (same as normal operation).
Pressure loading (CO)	The same CO issues apply here. High CO production will result in high pressure (same as normal operation).
Layer oxidation	In some cases internal oxidation of the layer could be significant. IPyC cracks can allow CO to the SiC and corrosion may result.
Stress state	See general discussion on particle layers. An impulse load may be more demanding. If this layer cracks or debonds, the stress distribution of the particle could change.
Intercalation	Material trapped in the layer could be released.

#### **2.5.5.4 SiC Layer**

The main goal is for the SiC layer is to stay intact under the induced stresses in the layer. If the accident causes a significant internal pressure pulse, fracture toughness of the SiC could be important. One factor is the temperature of the layer during and after the event. High-pressure stresses and layer temperatures near or above 1600°C could result in failure.

The details of the accident need to be modeled to get a reasonable estimate of the evolution of the event so that proper analysis and testing can be done.

##### **2.5.5.4.1 SiC Layer Reactivity Accident Factors Identified By The PIRT Panel**

Table 2-50 lists the SiC layer reactivity accident factors identified by the PIRT panel. Generally, many of the factors are the same as those that were discussed before.



**Table 2-50 SiC Layer Reactivity Accident Factors Identified By The PIRT Panel**

<b>SiC Reactivity Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	Except for silver, fission product diffusion is low through the SiC even at accident temperatures.
Condensed phase diffusion	
Thermal deterioration or decomposition	Above about 1600-1800°C the SiC begins to decompose into Si and C and its ability to retain fission products is greatly reduced.
Fission product corrosion	Some fission products may migrate to the SiC and damage it. This process is a function of temperature and can begin during normal operation. Pd is believed to be the main source of chemical attack, but others are also possible.
Heavy metal diffusion	The diffusion of fissile material could change the core configuration. A concern is major particle cracking and the expulsion of the kernel material during the pulse.
Layer oxidation	Attack by CO due to a cracked IPyC at high temperatures may be possible.
Fission product release through undetected defects, e.g. cracking	In order to release gases, both PyCs must be cracked in addition to a failed SiC. The SiC can be attacked by fission products and fabrication impurities (see manufacturing). Also, the impulse loads of this event may be important.
Fission product release through failures, e.g. cracking	
Thermodynamics of the SiC fission product system	At the temperature of interest, SiC is just stable to oxidation in its intact particle environment. (See corrosion issues.)
Sintering	High temperatures could result in changes to the microstructure and changes in fission product transport.

### 2.5.5.5 OPyC Layer

The main goal of the OPyC is to support the SiC during the event to keep it from exceeding its ultimate stress. Many of the factors that have been discussed previously apply.

#### 2.5.5.5.1 OPyC Layer Reactivity Accident Factors Identified By The PIRT Panel

Table 2-51 lists the OPyC layer reactivity accident factors identified by the PIRT panel. Note that, like the other layers, they overlap with other events discussed previously.

**Table 2-51 OPyC Layer Reactivity Accident Factors Identified By The PIRT Panel**

<b>OPyC Reactivity Accident Factor</b>	<b>Rationale</b>
Gas phase diffusion	Gaseous diffusion is low, while metallic diffusion is high.
Condensed phase diffusion	
Layer oxidation	Coolant impurities could be a factor.
Stress state	See general discussion on the stress of the particle. OPyC loss is generally not as significant as IPyC loss.
Intercalation	Because the fission product inventory is low in this layer, these two items may be important in determining transport.
Trapping	
Cracking	Cracks can lead to a higher probability of SiC failure. Gases will be released if the other layers have failed.

**2.5.5.6 Fuel Element**

The purpose of the fuel element form during a reactivity accident event is to prevent relocation of the fuel during the accident should the particles fail and to sorb a portion of the released fission products. Cracks in the fuel element may occur, but fragmentation of the element is not expected.

**2.5.5.6.1 Fuel Element Reactivity Accident Factors Identified By The PIRT Panel**

Table 2-52 lists the fuel element reactivity accident factors identified by the PIRT panel. These concerns are similar to those previously identified.

**Table 2-52 Fuel Element Reactivity Insertion Factors Identified by the PIRT Panel**

<b>Fuel Element Reactivity Accident Factor</b>	<b>Rationale</b>
Irradiation history	Matrix behavior (shrinkage) is related to time temperature history, particularly temperatures and fast fluence.
Condensed phase diffusion	In the matrix gas diffusion is high, while metallic diffusion is variable. The less volatile metals can be sorbed in the matrix material to a high degree.
Gas phase diffusion	
Transport of metallic FPs through fuel element - Chemical form	The chemical form of the fission product determines its transport behavior. The chemical environment outside the fuel kernel may be significantly different than inside. Generally, significant sorbing of the metallics is observed.

## 2.6 Summary of Fuel Failure Mechanisms

The previous sections detailed the fuel behavior on a layer-by-layer basis so the PIRT process could be applied to each aspect of the fuel constitution. This section will briefly review the fuel particle damage/failure mechanisms on a broader scale so that the reader might better catalog the general overall picture.

A review of the irradiation and safety testing of coated particle fuel reveals a number of potential failure mechanisms. These failure mechanisms are functions of temperature, burnup, fluence, and macroscopic temperature gradient across the particle. Mechanisms that may result in particle failure, which ultimately leads to fission product release, are:

- 1) Pressure vessel failure caused by internal gas pressure
- 2) Pyrocarbon layer cracking and/or debonding due to irradiation induced shrinkage that ultimately leads to the failure of the SiC layer
- 3) Fuel kernel migration (amoeba effect), which leads to interactions with the coating layers
- 4) Fission product/coating layer chemical interactions
- 5) Matrix/OPyC interaction
- 6) As-manufactured defects produced during fabrication of fuel particles or during pressing of fuel compacts/spheres
- 7) Thermal decomposition of the SiC layer at very high temperatures
- 8) Enhanced SiC permeability and/or SiC degradation (high burnup considerations)
- 9) Chemical attack (ingress accidents)
- 10) Reactivity insertion (accident)

In this section, these mechanisms and the variables that control them are briefly described; for more detail refer to the previous section(s).

### 2.6.1 Pressure Vessel Failure

Under irradiation, coated particle fuel is subjected to a number of forces that put stress on the TRISO coating. One of the earliest recognized mechanisms is overpressure due to gas generation under irradiation. During irradiation, fission gases are released from the kernel to the porous buffer layer. The pressure that is generated exerts tensile forces on the layers of the particle. In addition to fission gas, in coated particle fuel with UO<sub>2</sub> kernels, there is excess oxygen released during fission. This excess oxygen will react with the buffer to form CO gas. Both the fission gas and CO production are functions primarily of burnup and temperature. In UCO fuels, CO is not produced provided sufficient uranium carbide is added to the kernel to buffer the oxygen over the burnup life of the fuel. The key variables that affect this mechanism are burnup and temperature. Fluence does not significantly affect these processes. Particles are generally sized with a large enough buffer to ensure that nominal particles do not fail by overpressure. Particle failure is postulated to occur in the event that during the coating process, particles are coated with an insufficient or missing buffer layer (i.e., insufficient void volume to accommodate the gases). Thus, fabrication specifications limit the number of particles produced with thin

or missing buffer layers and impose limits on the statistical variation in kernel diameter and buffer thickness. This is a much analyzed but seldom seen failure mechanism. No indications of pressure vessel failure were observed in the past German pebble fuel irradiations.

### **2.6.2 Irradiation-induced IPyC Cracking and Debonding**

Under irradiation, PyC shrinks in both the radial and tangential direction. At modest fluences ( $\sim 2 \times 10^{25}$  n/m<sup>2</sup>) depending on the density, temperature and anisotropy of the material, it begins to swell in the radial direction and continues to shrink in the tangential direction. This behavior puts the PyC layers into tension in the tangential direction. At longer irradiation times, irradiation induced creep works to relieve the tensile stress in the PyC layer. If the IPyC is strongly bonded to the SiC layer, the IPyC shrinkage provides a strong compressive stress in the SiC layer that offsets the tensile stresses generated by gas production. In fact, the particles are designed such that in intact particles, the SiC layer remains in compression throughout the irradiation.

The shrinkage, swelling and creep behavior of the pyrocarbons is complex and depends strongly on the fabrication details. If the shrinkage is much larger than anticipated the tangential stresses in the PyC can be high enough to cause cracking in the layer. These cracks can lead to stress concentrations in the SiC layer high enough to cause failure of that layer. This failure mechanism has been attributed to high anisotropy in the PyC layer.

Post irradiation examination of German pebble fuel did not reveal any shrinkage cracks in the IPyC layer as has been observed in many U.S. irradiations. Thus, the experimental evidence to date suggests that this mechanism is most likely not important for very isotropic IPyC, but may play a role in less isotropic IPyC. The issue is complicated because the PyC isotropic measurement is somewhat controversial and US fuels often saw much more aggressive irradiation conditions. Control of the fabrication process appears to be the most reliable current way to assure sound PyC properties.

In addition to irradiation-induced shrinkage, debonding at the IPyC/SiC interface has been observed in many U.S. irradiations. This debonding is believed to be related to the nature of the IPyC/SiC interface. Weakly bonded coating layers can partially detach because of the tensile stresses generated by the PyC shrinkage under irradiation. A particle for which partial debonding of the IPyC from the SiC has occurred can develop relatively large tensile stresses in the SiC (although significantly smaller than in the case of a cracked IPyC). Tensile stresses occur at the point of IPyC/SiC contact as the IPyC shrinks under irradiation. Irradiation induced creep relieves the stress at longer times. When these stresses are used in concert with Weibull statistics to calculate the SiC failure probability, it is found that the SiC fails at a low, but not insignificant, rate.

### **2.6.3 Kernel Migration**

Kernel migration is defined simply as movement of the kernel in the coated particle toward the TRISO coating. If the migration is excessive, the kernel will penetrate the TRISO coating leading to failure of the particle. Kernel migration is associated with carbon transport in the particle in the presence of a temperature gradient. In the fuel kernel equilibrium is established among C, UO<sub>2</sub> and CO. When there is a thermal

gradient across the particle, the equilibrium is different on each side of the particle. The different equilibrium conditions lead to mass transport of carbon down the temperature gradient. This movement of carbon appears in photomicrographs of fuel as a movement of the kernel up the temperature gradient and hence the name kernel migration. This phenomenon is strongly dependent on the temperature and the macroscopic temperature gradient in the fuel with secondary dependence on burnup. It can also occur as solid-state carbon transport through carbide kernels.

In prismatic cores with  $\text{UO}_2$  fuel, where power densities in the particles are greater, the potential for kernel migration is greater. In pebble bed cores, the power densities and hence the thermal gradients are much smaller. For prismatic cores, this phenomenon prompted the U.S. to change their kernel design from  $\text{UO}_2$  to UCO, an oxycarbide kernel, in which no CO is produced and thus the equilibrium and carbon transport phenomena mentioned above are not expected to occur.

#### **2.6.4 Fission Product/Coating Layer Chemical Interactions**

Past irradiation experiments indicate that fission products can be transported from the kernel to the inner surface of the SiC where they interact and can damage and potentially fail the SiC layer. In older uranium carbide kernels rare earth fission product migration was of concern. In  $\text{UO}_2$  kernels, palladium is very important, as are some other noble metal fission products. In UCO kernels, the oxycarbide form of the kernel generally ties up all fission products with the exception of a few metals (e.g., Ag, Cs, Pd) as either carbides or oxides, which tend to limit their mobility in the UCO system. However, Pd transport has still been observed in UCO coated particle fuel. In addition, although not a failure mechanism, the migration of silver in both  $\text{UO}_2$  and UCO has been observed. The silver can migrate through apparently intact particles and be released into the reactor coolant system where it will deposit on cold surfaces. For direct cycle gas reactors, this may be in the turbine, which has important maintenance (worker dose) implications. Studies have been conducted to understand the mechanism for the Ag migration through SiC and Pd attack of the SiC. The migration of the fission products is thought to be a function of temperature and burnup as well as temperature gradient. Although a complete understanding of the phenomena is not available, the role of temperature and temperature gradient are recognized as being critical. The degree of fission product attack is generally correlated with the temperature and temperature gradient in the fuel. Thus, these fission product attack mechanisms are expected to play a more important role in prismatic reactors where power densities in the particle are larger than corresponding particles in a pebble bed reactor (reference design for this PIRT).

Also of note here is the fact that the enrichment of the fuel is important in defining the magnitude of the Ag and Pd problem. The yields of Ag and Pd are 25 to 50 times greater for Pu than for U. Thus, in LEU fuels where at the end of life significant fission comes from Pu, the concentration of Ag and Pd can be much greater than in HEU fuel of similar burnups.

#### **2.6.5 Matrix/OPyC Interaction**

In early U.S. irradiations, high levels of OPyC failure due to cracking or debonding from the SiC layer were observed. These failures were attributed to intrusion of the low viscosity carbonaceous matrix material in the OPyC during compact fabrication followed

by shrinkage under irradiation. Specifications on this matrix material and its injection were developed based on the irradiation experiments to limit this failure mechanism. No similar behavior was observed in German pebble fuel because of the use of a high viscosity matrix/binder mix that does not tend to penetrate the OPyC and because of the higher isotropy of German OPyC.

#### **2.6.6 As-Manufactured Defects**

In the absence of any of the above failure mechanisms, fission gas and metal release during irradiation is attributed to heavy metal contamination outside of the SiC layer and to initially defective particles. Initially defective particles can be the result of undetected defective particles that have not been removed during fabrication, attack of the particles during fabrication or irradiation by impurity metals (e.g., Fe), or particles that have failed as a result of damage from fuel element fabrication. In pebble bed fuel, a soft overcoating is put on the particle after the OPyC layer to limit stresses induced by particle-to-particle contact during pebble manufacture. In prismatic fuel using the injection process, recent process development work has been carried out to reduce particle stresses and limit introduction of impurities during compact formation.

During the three decades of German particle fuel production, the fraction of as-manufactured defects has continuously dropped to very low levels ( $< 1 \times 10^{-6}$ ). This is evident by the low beginning of life Kr-85m R/B values (reaching a minimum value of  $2 \times 10^{-10}$  in the FRJ2-K15 experiment) from each of the German experiments. Even at these low defect levels, as-manufactured defects were the most common source of particle abnormalities reported. In all, one fuel kernel was reported to be without coating in the FRJ2-P27 experiment and two kernels were reported to be without coating in the R2-K12 experiment.

#### **2.6.7 SiC Thermal Decomposition**

At very high temperatures (above 1600-1800°C), thermodynamics and data from German high temperature heating tests show that the SiC layer undergoes thermal decomposition at a significant rate. This phenomenon is primarily a function of temperature and time and has not played a major role in fuel failure at lower accident temperatures (1600°C) where safety testing has been routinely performed. Fuel releases generally increase at temperatures above 1600°C, with releases at 1800°C being much greater, although SiC behavior in the 1600-1800°C range may be a combination of corrosion and decomposition.

#### **2.6.8 Enhanced SiC Permeability and/or SiC Degradation**

Although not formally a failure mechanism, there is some limited evidence that fast neutron fluence and/or burnup plays a role in the permeability or degradation of the SiC layer with respect to fission products during high temperature heating. Pebbles exposed to higher fluence ( $4.6 \times 10^{25} \text{ n/m}^2$ ) and higher burnup (14 % FIMA) have exhibited a greater release of fission products (e.g., cesium) in heating tests than similar pebbles exposed to less severe conditions. This phenomenon could become more important as coated particle fuel is pushed to high burnup. The reference German pebble has not been tested at more aggressive irradiation conditions (temperature and burnup).

### **2.6.9 Chemical Attack (Ingress Accidents)**

Under accident conditions, fuel may be exposed to air and/or water. Both will react with the carbon materials and the fuel kernels. Water ingress primarily affects exposed kernels, causing them to release a large fraction of their stored inventory. Reactions with the carbon materials are relatively modest at the temperatures of interest and the resulting fuel damage is not aggressive.

Air is much more aggressive than water and reacts not only with the exposed kernels, but also reacts with the carbon materials at a greater rate than water. The possibility of the establishment of a flow may result in significant fuel damage if allowed to continue unabated. In both cases, reactions with the carbon materials will release sorbed fission products.

### **2.6.10 Reactivity Insertion (Accident)**

The sudden generation of high energies within the coated particle fuel can cause it to overheat, overpressure, and break, thus releasing its fission products. This accident is not well defined, but results to date indicate that coated particle fuel will fail with energy depositions in the range of 1000-2000 J/gm. Much higher energies may cause the fuel element to be damaged.

Past estimates for the reactivity insertion duration were in the seconds with energies much larger than that required to damage fuel particles.

## **2.7 References**

Reference 2-6 is an excellent compilation of coated particle fuel information and is a good starting point for the interested reader. Reference 2-9 is a good starting point for understanding how coated particles are bonded into a fuel element.

- 2-1. Mehner, A. W., et.al., "Status of Qualification of High-Temperature Reactor Fuel Element Spheres, Nuclear Technology," 69 (1985), page 44.
- 2-2. "Spherical Fuel Elements for Advanced HTR Manufacture and Qualification by Irradiation Testing," Journal of Nuclear Materials, 171 (1990), pages 1-18.
- 2-3. Nickel, H., et. al., "Long Time Experience with the Development of HTR fuel Elements in Germany," Nuclear Engineering and Design, 217 (2002), pages 141-151.
- 2-4. Wolf, L., et. al., "Fuel Elements for the High Temperature Pebble Bed Reactor, Nuclear Engineering and Design," 34, (1975), pages 93-108.
- 2-5. Gontard, R., and H. Nabilek, "Performance Evaluation of Modern HTR TRISO Fuel," HTA-1B-05/90 (July 1990).
- 2-6. "Fuel Performance and Fission Product Behavior in Gas Cooled Reactors," IAEA-TECDOC-978 (1997).

- 2-7. Moormann, R., et. al., "Source Term Estimation for Small-Sized HTRs: Status and Further Needs, Extracted From German Safety Analysis," Nuclear Technology, 135, (2001), pages 183-193.
- 2-8. Nuclear Technology, Volume 35, Number 2, 1977 (entire issue is devoted to coated particle fuels).
- 2-9. "Fuel Compact Design Basis Report," DOE-GT-MHR-100212 (1994).
- 2-10. Proksch, E., et. al., "Production of Carbon Monoxide During Burn-up of UO<sub>2</sub> Kernal HTR Fuel Particles," Journal of Nuclear Materials, 107 (1982) pages 280-285.
- 2-11. Horsley, G. W., et. al., "Influence of Irradiation Temperature, Burnup, and Fuel Composition on Gas Pressure (Xe, Kr, CO, CO<sub>2</sub>) in Coated Particle Fuels," Journal of the American Ceramic Society, 59 (1976), Number 1-2, pages 1-4.
- 2-12. Stansfield, O. M., et. al., "Kernel Migration in Coated Carbide Fuel Particles," Nuclear Technology, 25 (1975), pages 517-530.
- 2-13. Martin, D. G., "Considerations Pertaining to the Achievement of High Burn-ups in HTR Fuel," Nuclear Engineering and Design, 213 (2002), pages 241-258.
- 2-14. Kaae, J. L., "A Mathematical Model for Calculating Stresses in a Pyrocarbon and Silicon Carbide Coated Fuel Particle," Journal of Nuclear Materials, 29 (1969), page 249.
- 2-15. Stevens, D. W., "An Explicit Solution for Stresses in Pyrocarbon-Coated Fuel Particles," Nuclear Technology, 10 (1971), page 301.
- 2-16. Kaae, J. L., "A Mathematical Model for Calculating Stresses in a Four-Layer Carbon-Silicon-Carbide-Coated Fuel Particle," Journal of Nuclear Materials, 32, (1969), page 322.
- 2-17. Gulden, T. D., et. al., "The Mechanical Design of TRISO-Coated Particle Fuels for the Large HTGR," Nuclear Technology, 16 (1972), pages 100-109.
- 2-18. TRISO Fuel Particle Coating Design Basis, DOE-GT-MHR-100225 (1994).
- 2-19. Miller, G. K., and D.C. Wadsworth, "Treating Asphericity in Fuel Particles Pressure Vessel Modeling," Journal of Nuclear Materials, 211 (1994), pages 57-69.
- 2-20. Verfondern, K., "Modeling of Fuel Performance and Metallic Fission Product Release During HTTR Normal Operating Conditions," Nuclear Engineering and Design, 210 (2001), pages 225-238.



- 2-21. Mitake, S., et. al., "An analytical Study of Volatile Metallic Fission Product Release From Very High Temperature Gas-Cooled Reactor Fuel and Core," Nuclear Technology, 81 (1988), pages 7-12.
- 2-22. Martin, R. C., "Compilation of Fuel Performance and Fission Product Transport Models and Database for MHTGR Design," ORNL/NPR-91/6 (1993).
- 2-23. Martin, R. C., "Revised MHTGR High-Temperature Fuel Performance Models," ORNL/NPR-92/16 (1993).
- 2-24. Verfondern, K., et. al., "Methods and Data for HTGR Fuel Performance and Radionuclide Release Modeling during Normal Operational and Accidents for Safety Analysis," Forschungszentrum Juelich GmbH, Juel-2721 (1992).
- 2-25. Godfrey, T. G., et. al., "Coated-Particle Fuels," ORNL-4324 (1968).
- 2-26. "TRISO Fuel Particle Coating Design Basis," DOE-GT-MHR-100225 (1994).
- 2-27. Petti, D. A., et. al., "Key Differences in the Fabrication, Irradiation and Safety Testing of U.S. and German TRISO-coated Particle Fuel and Their Implications on Fuel Performance," INEEL/EXT-02-00300, also Nuclear Engineering and Design, 222 (2003) 281-297.
- 2-28. "Fission Recoil Damage to Pyrolytic Carbon," Carbon, 7 (1969), pages 703-706.
- 2-29. "MHTGR TRISO-P Fuel Failure Evaluation Report," DOE-HTGR-90390 (1993).
- 2-30. Miller, G., et. al., "Consideration of the Effects on Fuel Particle Behavior from Shrinkage Cracks in the Inner Pyrocarbon Layer," Journal of Nuclear Materials, 295 (2001), pages 205-212.
- 2-31. Minato, K., et. al., "Carbon Monoxide-Silicon Carbide Interaction in HTGR Fuel Particles," Journal of Materials Science, 26 (1991), pages 2379-2388.
- 2-32. Forthmann, R., et. al., "Influence of Material Properties on the Retention of Fission Products by Silicon Carbide Coatings," High Temperature – High Pressure 1982, Volume 14 (1982) pages 477-485.
- 2-33. "An Assessment of the Methods for Determining Defect or Failure Fractions in HTGR Coated Particle Fuels and Their Relationship to Particle Microstructure," DOE-HTGR-88260 (1989).
- 2-34. Tokar, M., Evaluation of High Temperature Gas Cooled Reactor Fuel Particle Coating Failure Models and Data, NUREG-0111 (1976)
- 2-35. Tiegs, T. N., "Fission Product Pd-SiC Interaction in Irradiated Coated-Particle Fuels," Nuclear Technology, 57 (1982), pages 389-398. YEAR?

- 2-36. Lauf, R., et. al., "Out-of-Reactor Studies of Fission Product-Silicon Carbide Interactions in HTGR Fuel Particles," *Journal of Nuclear Materials*, 120 (1984), pages 6-30.
- 2-37. Federer, J. I., "Fluidized Bed Deposition and Evaluation of Silicon Carbide Coating on Microspheres," ORNL/TM-5152 (1977).
- 2-38. "MHTGR Fuel Manufacturing Quality Assurance Plan," DOE-HTGR-88091 (1988).
- 2-39. "Data Support Document: Operating Procedures for SiC Defect Detection," DOE-HTGR-88359 (1991).
- 2-40. Bullock, R. E., "Fission-Product Release During Postirradiation Annealing of Several Types of Coated Fuel Particles," *Journal of Nuclear Materials*, 125 (1984), pages 304-319.
- 2-41. "Fission Product Plateout and Liftoff in the MHTGR Primary System: A Review," NUREG/CR-5647 (1991).
- 2-42. Myers, B., "The effect of Water Vapor on the Release of Gaseous Fission Products from High-Temperature Gas-Cooled Reactor Fuel Compacts Containing Exposed Uranium Oxycarbide Fuel," DOE-HTGR-88486 (1991).
- 2-43. Moorman, R., "Chemical Behavior of Fission Products in Core Heatup Accidents in High-Temperature Gas-Cooled Reactors," *Nuclear Technology*, 94 (1991), pages 56-67.
- 2-44. "Design of High Temperature Engineering Test Reactor (HTTR)," Japan Atomic Energy Research Institute document JAERI 1332, Japan Atomic Energy Research Institute (September 1994).
- 2-45. Hishida, M., and T. Takeda, "Study on Air Ingress During an Early Stage of a Primary-Pipe Rupture Accident of a High-Temperature Gas-Cooled Reactor," *Nuclear Engineering and Design*, 126 (1991), pages 175-187.